From: "Thomas Harter" <thharter@ucdavis.edu>
To: "Marcus, Felicia@Waterboards" <Felicia.Marcus@waterboards.ca.gov>, "Dadamo, Dorene@Waterboards" <Dorene.Dadamo@waterboards.ca.gov>
Cc: "Polhemus, Darrin@Waterboards" <Darrin.Polhemus@waterboards.ca.gov>, "Doduc, Tam@Waterboards" <Tam.Doduc@waterboards.ca.gov>, "Spivy-Weber, Frances@Waterboards" <Frances.Spivy-Weber@waterboards.ca.gov>, "Moore, Steven@Waterboards" <Steven.Moore@waterboards.ca.gov>
Subject: groundwater nitrate trends in Europe

Hi Felicia, DeeDee,

At the May 17 meeting you had asked, whether there are scientific data from places outside of California that demonstrate the reversal of groundwater nitrate concentrations as a result of long-term changes in agricultural management practices. I have put this question to some of my European colleagues for help. I am attaching some examples:

Within the EU, Denmark and The Netherlands have perhaps the most significant and widespread groundwater nitrate pollution related to agriculture and have been very pro-active in setting policy, beginning in the early 1990s after the EU Nitrate Directive was put in place.

Three articles reporting on trend reversals in Denmark are attached. Hansen et al., 2011 in Figure 3 shows the long-term trends in nitrogen surplus (essentially the difference in nitrogen applied, "A", and nitrogen removed, "R") and national trends in groundwater nitrate. Spruill et al., 2011 critiqued the statistical methods used by Hansen et al (2011) and disputed that the data indeed show a relationship between improved practices and improved groundwater quality. Hansen et al 2012 takes a more thorough look at the data with some data through 2010. Dalgaard et al 2014 provides an excellent discussion of policies, agricultural practices and trends in Denmark.

Baumann et al (2012) reports on Dutch policy actions, agricultural practices, and resulting groundwater quality at various depth (pp, 69 - 112). Visser et al. (2007) also considers groundwater age in interpreting trend reversals in groundwater quality. Visser et al. (2009), in comparing four distinct methodologies to identify long-term groundwater nitrate trends, shows for examples from central European basins where this has been the case.

There are a number of studies that point out the long time lag between changes in management practices and changes in groundwater quality, depending on local hydrogeology. This is an issue we also investigated for the Tulare Lake Basin as part of the SBX2 1 nitrate study in 2012 (Boyle et al., 2012, Chapter 7: http://groundwaternitrate.ucdavis.edu/files/139106.pdf).

Let me know if I you have questions or would like to discuss further.

Best regards,
Thomas

Baumann et al - Nitrate Program in NL and Trend...

Thomas Harter
Dept. Land, Air, and Water Resources
University of California
Davis, CA 95616-8628
ph/ 530-400-1784
http://groundwater.ucdavis.edu
"Sustainable Groundwater in Agriculture
- International Conference Linking Science and Policy"
Burlingame/San Francisco, California, June 28-30, 2016;
http://ag-groundwater.org

<Hansen et al - Trend reversal in groundwater nitrate Denmark - EST 2011.pdf>
<Spruill - Critical comment on Hanson - EST 2011.pdf>
<Dalgaard et al - Nitrate policies and groundwater trends in Denmark - ERL 2014.pdf>
<Hansen et al - Groundwater nitrate trends in Denmark - Biogeosciences 2012.pdf>
<Visser - Trend reversel groundwater nitrate Netherlands - Env Poll 2007.pdf>
<Visser et al - Comparison of trend analysis methods - JEM 2009.pdf>

Trend Reversal of Nitrate in Danish Groundwater - a Reflection of Agricultural Practices and Nitrogen Surpluses since 1950

BIRGITTE HANSEN,*,† LÆRKE THORLING,† TOMMY DALGAARD,‡ AND MOGENS ERLANDSEN§

Geological Survey of Denmark and Greenland (GEUS), Denmark, Department of Agroecology, Aarhus University, Denmark, and Department of Biostatistics, Aarhus University, Denmark

Received August 11, 2010. Revised manuscript received November 18, 2010. Accepted November 18, 2010.

This paper assesses the long-term development in the oxic groundwater nitrate concentration and nitrogen (N) loss due to intensive farming in Denmark. First, up to 20-year time-series from the national groundwater monitoring network enable a statistically systematic analysis of distribution, trends, and trend reversals in the groundwater nitrate concentration. Second, knowledge about the N surplus in Danish agriculture since 1950 is used as an indicator of the potential loss of N. Third, groundwater recharge CFC (chlorofluorocarbon) age determination allows linking of the first two data sets. The development in the nitrate concentration of oxic groundwater clearly mirrors the development in the national agricultural N surplus, and a corresponding trend reversal is found in groundwater. Regulation and technical improvements in the intensive farming in Denmark have succeeded in decreasing the N surplus by 40% since the mid 1980s, while at the same time maintaining crop yields and increasing the animal production of especially pigs. Trend analyses prove that the youngest (0–15 years old) oxic groundwater shows more pronounced significant downward nitrate trends (44%) than the oldest (25–50 years old) oxic groundwater (9%). This amounts to clear evidence of the effect of reduced nitrate leaching on groundwater nitrate concentrations in Denmark.

Introduction

Danish drinking water supply relies on simple treated groundwater, and protection of groundwater is therefore a high priority. At the same time, Danish farming is among the most intensive in the world (*1*). Intensive livestock farming and extensive use of nitrogen (N) fertilization causes severe N losses to soil, water, and air. Numerous waterworks and wells have been closed due to nitrate pollution, and approximately 15% of Denmark has been classified as nitrate vulnerable groundwater abstraction areas (*2*). Commonly observed N-related environmental effects include a decline in biodiversity, eutrophication of ecosystems and surface waters (*3*), acidification (*4*), global warming via emission of N_2O (*5*), and diffuse nitrate pollution of groundwater. Both European policies (The Nitrates Directive, 1991/696/EC; The Water Framework Directive, 2000/60/EC; and The Groundwater Directive, 2006/118/EF) and national Danish legislation aim to protect groundwater resources and surface waters from the effect of N loss to the environment. Denmark has therefore introduced several political action plans since 1985 (*6*).

During the past 100 years, the agricultural development in Denmark has contributed significantly to the increase in societal prosperity owing to the growth in farming output and improved farming efficiency, especially after the Second World War. Like most countries in the industrialized world, Danish farming has experienced a significant structural development toward larger and more intensive farms, and Danish livestock farming in particular has grown significantly (*7*). Today, the Danish food production accounts for about 23% of the private sector's total turnover and investments, 21% of exports, and 16% of all employment (*8*).

The increase in the Danish agricultural production has been strongly spurred by the growing use of synthetic fertilizers and imported feed, which has resulted in more use of fertilizers and a higher nutrient turnover (*9*). The past 100 years have also seen the introduction of new crops in plant production and the emergence of new production branches in animal production. Crop yield per hectare has risen, and the yearly milk yield per cow has grown from approximately 2800 to 10,000 kg per cow from 1950 to 2007. Additionally, the production time of pigs and chickens is shorter today than ever before. The production of pigs has seen a dramatic rise from 3.2 million animals per year in 1950 to 11.2 million in 1967 and 20.9 million in 2007. Thus, pig production has been the largest Danish animal production branch since the mid 1970s (*8*).

Intensive farming causes leaching of N because of mineralization of the soil's N pools and because the amount of N used exceeds the nutrient demand of the crops (*10, 11*). This results in pollution of shallow groundwater that threatens drinking water resources and groundwater associated and dependent ecosystems (*12*). Nitrate leached to groundwater is expected to act as an inert tracer under the presence of oxygen and the generally low reactivity of organic matter below the root zone. Therefore, this study focuses on oxic groundwater, where the nitrate concentration represents the original nitrate leaching from the root zone.

Groundwater recharge age determination facilitates comparison of developments in the potential loss of N in agriculture and the measured groundwater nitrate concentrations (e.g. refs *13–18).*

This study aims to link nitrate trends in oxic Danish groundwater since 1950 to structural changes in agriculture related to the national, annual farming N surplus. The objectives is thus to present two separately statistical approaches: a) assessment of the general national trend of nitrate in oxic Danish groundwater and b) aggregation of the individual nitrate trends at the oxic groundwater monitoring points according to the age of the groundwater recharge.

2. Methods

2.1. Agricultural, Geological, and Hydrological Conditions. Denmark is a small country with a total area of about 43,000 km^2 and a population approaching 5.5 million. About 2/3 of the total land area is under agricultural use. Compared with European and global averages, Denmark has a high N

* Corresponding author phone: 0045 20555245; fax: 004538142050; e-mail: bgh@geus.dk. Corresponding author address: Geological Survey of Denmark and Greenland (GEUS), Department of Quaternary and Groundwater Mapping, Lyseng Allé 1, DK-8270 Højbjerg, Denmark.

† Geological Survey of Denmark and Greenland (GEUS).

‡ Department of Agroecology, Aarhus University.

§ Department of Biostatistics, Aarhus University.

10.1021/es102334u

Published on Web 12/07/2010

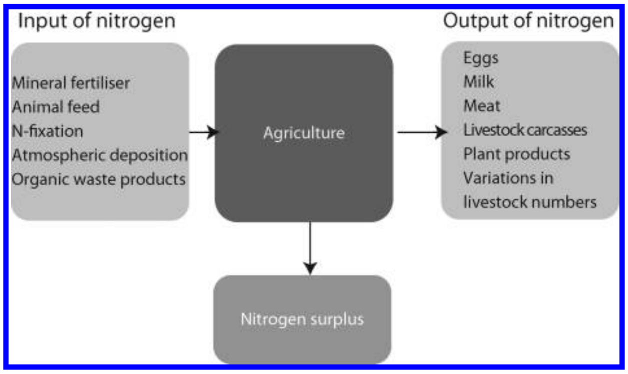


FIGURE 1. Input-output principle for nutrient balances in agriculture.

fertilization rate and a high livestock density (*1, 19*). The average livestock density in Denmark is about 0.8 animal units per hectare (*10*), and the average input of N to agricultural land is about 180 kg ha^{-1} yr^{-1} in 2008 (*8, 20*). The land surface has a modest topography with the highest point 170 m above sea level. Denmark has a coastal temperate climate where the precipitation varies from about 600 to 1000 mm/yr. The geological conditions in the upper layers are 50−200 m thick quaternary deposits which are underlain by tertiary deposits or Cretaceous limestone and chalk. Thus, the aquifers either consist of unconsolidated sands and gravels or fractured limestone and chalk.

2.2. N Surpluses in Agriculture. The surplus of nitrogen in agriculture is defined in terms of the balance between inputs (synthetic fertilizer, import of animal feed, organic waste products, net atmospheric deposition, and fixation) and outputs (export of plant and animal products) (Figure 1). The surplus of nitrogen, especially N, is regarded as the best overall environmental indicator of the changes in the agricultural impact on the environment over at certain time period (*19*). The surplus represents the amount of nitrogen pooled in the soil, or not being used up by the production system, and therefore at risk of being lost to the environment (*10, 11*).

2.3. The Danish Groundwater Monitoring Program. The principal aim of the approximately 20-year-old Danish Groundwater Monitoring Programme is to document the quantitative and qualitative status and trends of groundwater in order to evaluate the effect of the national action plans on the aquatic environment and thereby to meet the objectives of Danish legislation and the relevant EU directives.

The program covers 74 clustered catchments (Figure 2) covering areas of 5 to 50 km^2, each containing about 25 wells with short screens with a typical length of around 1 m capable of sampling a specific groundwater layer with minimal mixing of different water types. This gives a total of more than 1500 groundwater monitoring points with a median depth of 20 mbs (meter below surface) and a maximum depth of 164 mbs. The oxic subset of data used in this study has the monitoring points at a median depth of 17 mbs and a maximum depth of 68 mbs. The groundwater monitoring points are affected by different types of 1) agricultural land uses, 2) geological settings, 3) hydrological conditions, e.g. groundwater recharge rates, 4) depths, and 5) groundwater redox conditions.

The thickness of the unsaturated zone at the monitoring sites is up to 50 m with a median thickness of approximately 10 m. The oxic subset of data used in this study also has a median thickness of the unsaturated zone of 10 m but a maximum thickness of 36 m.

The wells are sampled annually for the main chemical components (nitrate, chloride, sulfate, ammonium, iron, etc.). Specific wells are analyzed for pesticides and their metabolites annually, while trace elements (arsenic, copper, nickel, etc.)

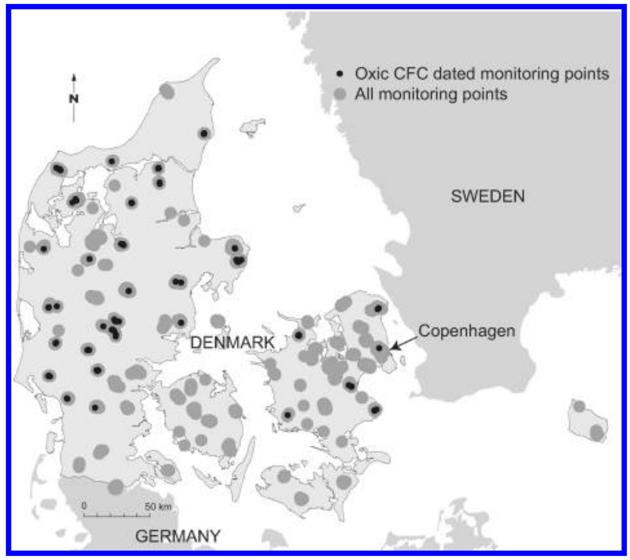


FIGURE 2. The groundwater monitoring points and the CFC dated oxic groundwater monitoring points in Denmark.

and organic pollutants are sampled with a lower frequency. More details on the Danish Groundwater Monitoring Programme can be found in ref *21*.

2.4. Groundwater Chemistry Data. The groundwater samples were analyzed in the field as well as in the laboratory. Online measurements of pH, redox potential, oxygen concentration, temperature, and conductivity were performed during sampling in order to ensure a high analytical quality and representative groundwater samples. The sampling and field analyses were performed according to Danish technical standards. The remaining chemical constituents of the groundwater were analyzed by professional, certified laboratories (*22*).

2.5. Groundwater Recharge Age Determination. Groundwater recharge age determination with the CFC method (*23*) makes it feasible to compare long-term changes in N surplus in agriculture with changes in oxic groundwater quality (*16, 24*). Groundwater recharge age was determined at the groundwater monitoring points, typically only once in the period from 1997−2006, using the CFC method (*23*).

Determination of the groundwater recharge ages followed the procedure of refs *13 and 23*, and the CFC analyses were performed at the laboratory of Geological Survey of Denmark and Greenland (GEUS). The method allows determination of the age of groundwater younger than 1940 with an uncertainty of ±2 years under optimal conditions (*13, 23*). Determination of the groundwater age follows the procedure of Laier (*13*). Samples of groundwater are collected in flame-sealed 60 mL boron silicate ampules which have been flushed with pure nitrogen prior to sampling. Approximately 30 mL of the sample is transferred to a purge and trap system, and finally the gases are swept into a gas chromatograph equipped with an electron capture detector (EDC) to quantify the amount of CFC gases dissolved in the water. The CFC concentration in the atmosphere at the time of recharge and thus the age of the recharge of groundwater is calculated using the constants in ref *25*.

Dating using the CFC method assumes that infiltration water maintains equilibrium with the unsaturated zone air during recharge (*23*). Previous studies of sandy aquifers in Denmark have shown that the residence time of water in the unsaturated zone using tritium dating (Andersen and Sevel, 1974) is similar to that of air diffusion through the unsaturated zone determined by CFC-gases on pore-space gases (*13, 26*). However, at some monitoring sites, depending on the hydrological conditions, there might be a difference between the resistance time of water and air diffusion in the unsaturated zone especially in areas with a deep unsaturated zone of up to 36 m. If the diffusion of CFC through the unsaturated zone is faster than the advection of water particles, then the groundwater recharge age determined with the CFC method will be overestimated.

The groundwater chemistry data as well as the CFC-data were downloaded from the Danish national geo-database JUPITER in October 2009 and thus includes all data uploaded until September 2009 (*27*).

2.6. Statistical Methods. Significant upward or downward trends can be determined either with statistical, linear methods (e.g., linear regression), or with nonparametric methods (e.g., based on Mann-Kendall) (*14, 28, 29*).

Data analyses including determination of trends and trend reversal of nitrate time-series in groundwater were done using the SAS software system (*30*). Trend analysis of nitrate time-series in groundwater at each monitoring point was performed as simple linear regression with PROC REG. A single analysis of trend reversal was performed on data from all monitoring points as a two-section linear regression with one unknown change point and fitted with PROC MCMC. Distribution of trends between monitoring points grouped according to the age of oxic groundwater were compared in a regression model with separate regression lines for each age group and fitted as a random coefficient model with PROC MIXED to allow for repeated measurements at each monitoring point. For each model, probability plots of the residuals were checked for normality.

2.7. Preprocessing of Data. The entire data set was filtered via this ordered, stepwise procedure:

1. Monitoring points with oxic CFC dated groundwater are included.

2. Monitoring points with more than 8 years of data are included.

3. Monitoring points with unstable redox conditions are excluded.

4. Outliers, e.g. effects of establishment, are found and excluded.

Only monitoring points with oxic conditions are included because both nitrate and CFC gases used for dating are degraded in anoxic groundwater (*31, 32*). The concentration levels of the redox sensitive parameters (NO_3^-, Fe^{2+}, and O_2) are used to sort out 194 samplings points with oxic conditions. Oxidized groundwater is defined as $[NO_3^-] > 1$ mg/L, $[Fe^{2+}] < 0.2$ mg/L, and $[O_2] > 1$ mg/L. This definition minimized the uncertainty in the determination of the groundwater redox state by giving third priority to the most uncertain parameter (i.e., oxygen). Iron in groundwater is thus used as an indication of complete nitrate reduction. The concentration levels of the other redox parameters (nitrite and manganese) show that an oxygen concentration higher than 1 mg/L is sufficient to ensure oxic conditions. This is due to a high quality of the oxygen field measurements of the groundwater.

During the past 20 years, the Danish groundwater monitoring program has been continuously adapted to scientific, environmental, and economic demands. The data from this program are therefore not entirely uniform, for example as regards the length of the time-series of chemical analyses. When performing trend analyses, the length of the time-series of nitrate analyses at each sampling point should be consistent (*33*). In this study, monitoring points with 8−20 years of data were accepted for trend analyses. Approximately 85% (165 monitoring points) of the oxic monitoring points have consistent time-series.

Some groundwater samplings points showed changing, unstable redox conditions due, among others, to i) mixing of groundwater with different redox conditions, ii) variable hydrological (precipitation, water table) conditions, and iii) influences by nearby groundwater abstraction. In this study, 92% (152 monitoring points) of the oxic monitoring points with more than 8-year time-series had stable redox conditions.

Both sampling and analytical errors may arise in groundwater sampling and chemical groundwater analyses. Drilling of groundwater wells can affect the chemical composition of the groundwater up to two years after the drilling, depending on the chemical substances used during drilling, the natural flow of groundwater, and the degree of flushing and pumping. Times-series from the 152 monitoring points were therefore examined for clear outliers and the effects on the water chemistry of the establishment of the wells; approximately 99% of the data was accepted.

The entire Danish monitoring data set with more than 8-year time-series counts 37,372 nitrate analyses from 1189 monitoring points sampled from 1973−2009. The subset of oxic CFC dated data used in this study for trend analyses comprises 5321 nitrate analyses from 152 sampling points sampled from 1988−2009.

3. Results

3.1. Trend Reversals of N at the National Level. Figure 3 illustrates the groundwater recharge age and the matching nitrate concentration at the year of the CFC measurement of the 194 oxic groundwater monitoring points found in step 1 described in the preceding section 2.7. In those monitoring points where the nitrate concentration is measured more than once a year the average nitrate concentration at the CFC sampling year is used.

The level of nitrate concentration at the year of groundwater recharge can be described by a piece-wise linear regression line with one unknown change point, as required by the EU Water Framework Directive, and shows a trend reversal at the year 1980 (±3.4 years) with a significant upward trend before and a significant downward trend after 1980 (see Figure 3). The slopes of the upward (c. 1.83 mg/L/yr) and downward (c. −1.61 mg/L/yr) curves are significantly different from zero at the 0.05 probability level.

The trend reversal in 1980 can also be illustrated by means of a simple 5-year moving averages curve relating nitrate concentration and recharge age. The time period under consideration saw a dramatic increase in N surplus in agriculture from approximately 60 to 180 kg N/ha/yr from 1950 to about 1981. After a 15-year stagnation period, the N surplus in agriculture started to decrease approximately in 1995, and in 2007 the average N surplus was 117 kg N/ha/yr. Thus, Figure 3 shows that around 1980 there is a trend reversal in the groundwater nitrate concentration and the increase of N surplus in agriculture in 1960s and 1970s levels out.

3.2. Distribution of Nitrate Concentrations at Each Monitoring Point. The choice of method used for trend analyses of the chemical composition at each monitoring point is determined by the distribution of the measurements (*29*). In this study, the distribution of nitrate was assessed for each of the 152 oxic monitoring points by inspection of standard normality plots. An example is given in Figure 4.

The residuals of the nitrate concentration at each of the 152 monitoring points are considered typical for normally distributed data. We therefore chose a simple linear regression for evaluation of trends (see Figure 4). Overall, the trend analyses at each point were found to meet the assumptions for linear regression based on scatter plots and normal probability plots of the residuals.

3.3. Trends in N Concentration in Oxic Groundwater of Different age. The results from linear regression analysis of the nitrate concentration versus time for each of the 152 oxic monitoring points are illustrated in Figure 5 as a quantile plot of the slope of each of the linear regression lines. The slopes represent the changes in the nitrate concentration per year, and the quantile plot of the slopes illustrates the overall variability between the monitoring points. Negative

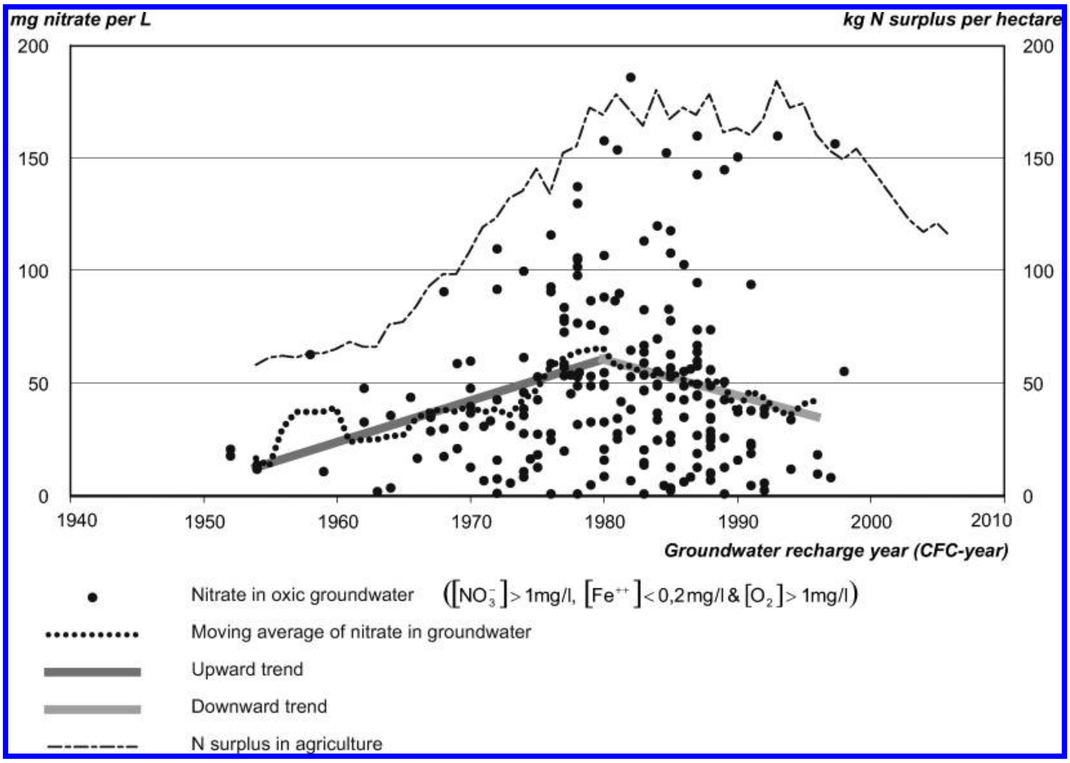


FIGURE 3. Time series of N surplus in agriculture and nitrate in oxic groundwater versus recharge age (CFC age) on an annual mean level.

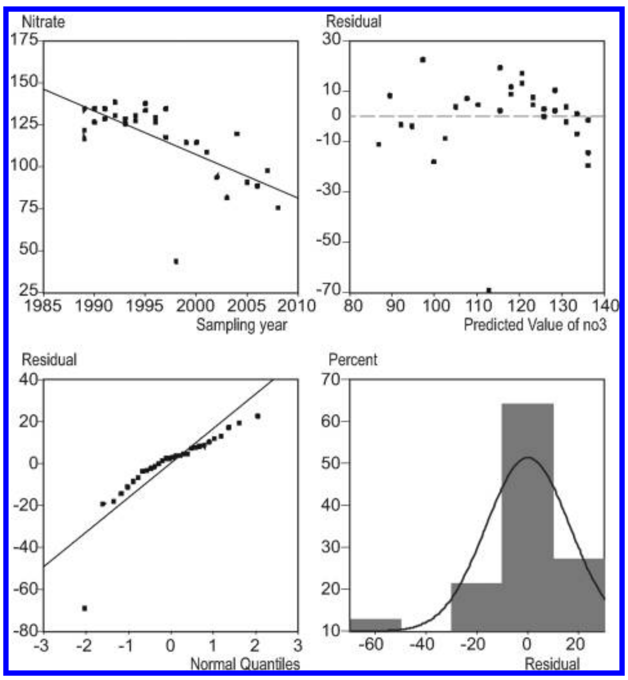


FIGURE 4. Linear regression and check for normality of an example with a downward statistically significant trend on a probability level of 0.05. Units are mg nitrate per L.

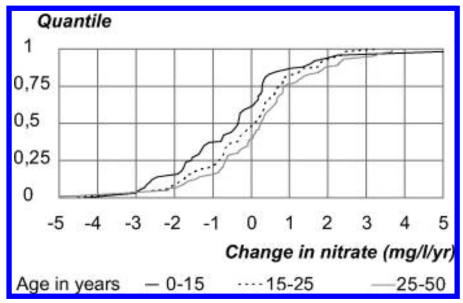


FIGURE 5. Quantile plot of changes in groundwater nitrate concentration (mg/L/yr) on a monitoring point level grouped by recharge age. The change in nitrate concentration is equivalent to the slope of the linear regression lines of nitrate versus sampling year seen in Figure 4.

TABLE 1. Amount (%) of Statistically Significant Nitrate Trends on a 95% Confidence Level of Oxic CFC Dated Groundwater According to the Age of the Groundwater Recharge

recharge age in years	upward	downward	no trends	total
0−50 (all)	50 (33%)	44 (38%)	58 (29%)	152 (100%)
0−15	10 (18%)	24 (44%)	21 (38%)	55 (100%)
15−25	19 (30%)	17 (27%)	28 (43%)	64 (100%)
25−50	21 (64%)	3 (9%)	9 (27%)	33 (100%)

x-values represent downward trends, while positive x-values represent upward trends. At a probability level of 0.05, 71% of the monitoring points show significant trends, with 38% downward and 33% upward trends. The remaining 29% show no significant trends at a probability level of 0.05 (Table 1).

The data shown in Figure 5 are divided into three groups according to the age of the groundwater, which was determined as the difference between the sampling year and the groundwater recharge year. Consistently with the data analysis in Figure 1, the youngest oxic groundwater has more monitoring points with downward trends than the older oxic groundwater. However comparison of the common trends in the three age groups demonstrated an only slight statistical significance ($p = 0.0394$).

Although the general trend (Figure 3) shows at trend reversal around 1980, the trends from the individual monitoring points show a more complex picture. According to Table 1, a significant downward trend was demonstrated in approximately 44% of the youngest oxic groundwater (0−15 years old), 27% of the medium old oxic groundwater (15−25 years old), and 9% of the oldest oxic groundwater (25−50 years old). In comparison, a significant upward trend was found in approximately 18% of the youngest oxic groundwater (0−15 years old), 30% of the medium old oxic groundwater (15−25 years old), and 64% of the oldest oxic groundwater (25−50 years old).

4. Discussion

Long-term groundwater quality monitoring with up to 20-year time-series provides optimal opportunities for investigating and understanding the impact of pressures and political action plans on groundwater quality. When addressing trends of pollutants in groundwater, it is fundamental to have long-term time-series of data, thereby being able to combine general trends and trends for individual monitoring points as shown in this study. This requires a consistent national monitoring program. Due to economic and political adjustments of the monitoring system, many time series are too short or abrupt, which reduces the payoff of monitoring investment.

This study demonstrates that groundwater recharge ages can be included as an essential component of groundwater trend investigations and that their inclusion may help to correlate changes in land use and management practices with changes in groundwater quality. Groundwater recharge age determination allows concentrations of nitrate to be related to the time of recharge instead of the time of sampling (e.g. refs *13−15 and 17)*, which, in turn, makes comparison between nitrate in groundwater and N loss from agriculture possible. In this study using the CFC method on oxic groundwater it is assumed that infiltration water maintains equilibrium with the unsaturated zone air during recharge and that degradation of CFC does not occur under the oxic redox conditions. Deep unsaturated zone of up to 36 m and small niches with anoxic redox conditions might increase the uncertainty in the determination of the groundwater recharge year using the CFC method. However, previous studies of sandy aquifers in Denmark have shown that the residence time of water in the unsaturated zone using tritium dating is similar to that of air diffusion through the unsaturated zone determined by CFC-gases on pore-space gases (*13, 26*).

In the Danish groundwater monitoring program it was decided to use the CFC method for dating groundwater due to successfully preliminary results under Danish hydrogeological conditions. Dating relative young (<20 years old) groundwater is a very important tool when evaluating the effect of agricultural action plans on the environment. However, atmospheric concentrations of CFCs were rapidly increasing before the 1990s, but as a result of the Montreal Protocol on Substances that Deplete the Ozone Layer adopted in 1987, atmospheric concentrations are now declining, which makes it impossible to use the CFC method for dating groundwater younger than 1990 (*34*). Therefore, there is a need for developing, testing, and implementing new methods suitable for dating young groundwater (e.g. refs *24, 34, and 35).*

A trend in groundwater quality is defined as a change over a specific period in time within a given region that is related to land use or water quality management (*36, 37*). Both temporal variations due to climatic and meteorological factors and spatial variability may complicate trend detection (*27*). Important in this context is the period of time under consideration. Periods of 8 to 30 years have been recommended, depending on the sampling frequency for water analysis (*28*). This study demonstrates that preprocessing of data (oxic redox condition, 8 years time-series, stabile redox conditions, and no outliers) and selection of a subgroup for trend analyses are very important for the interpretation of the data.

Significant upward or downward trends can be determined either with statistical, linear methods (e.g., linear regression), or with nonparametric methods (e.g., based on Mann-Kendall) (*28, 29*). This study deployed a standard linear regression model for trend analysis of the nitrate concentration at each monitoring point in which normality was established. More complex statistical models were avoided in order to make the analyses as simple as possible. This study shows that the simple statistical analyses of the Danish groundwater monitoring data as required by the EU Water Framework Directive (*28, 33*) are adequate.

The nitrate concentration of the monitoring points at each groundwater recharge year demonstrates much variation, and it exceeds 50 mg/L in groundwater recharged after 1950 at many monitoring points (approximately 40%). This variation in the nitrate content is due to local and regional variation mainly in land use, application of fertilizer, and complex geological settings of the surface layers and aquifers in Denmark.

Trend reversal with a single change point can be determined for instance by a two-section linear model (*14, 28*). In this study, such a model successfully described a statistically significant trend reversal in 1980 (±3.4 years) of the nitrate concentration of Danish oxic groundwater. A trends reversal around 1980 in the N concentrations and loads at a hydrological catchment scale has also recently been documented in the Odense river in Denmark (*38*).

The trend reversal around 1980 of the nitrate concentrations in oxic groundwater coincided with the clear leveling out of the N surplus in agriculture after a period of strong increase, which actually occurred before the initiation of the first Danish environmental action plan in 1985, and the following action plans in 1987, 1991, 1998, 2000, 2001, 2004, and forward (*20*). All these plans have focused on the reduction of N-pollution from agriculture, being far the most important source for nitrate leaching in Denmark (*20*). However, especially in the beginning of the action plan period, and around the overall nitrate leaching turning point in the 1970s and 1980s, reduction of N-losses from point sources in the form of better wastewater storage and treatment facilities in the rural areas, and reduced runoff from livestock houses, silage clamps and manure heaps had a significant effect.

Since the initialization of the first Danish environmental action plan in 1985, the turnover of N has declined owing to several initiatives, the most important one being restrictions imposed on the application of N in the form, for example, of maximum N norms for specific crops and minimum thresholds for utilization of the N content of animal manure (*20*). The N surplus in the agricultural sector has therefore deceased by 40% since the mid 1980s. In addition, the nitrate leaching from agriculture has decreased by on average 33% and N concentrations and loads in surface waters have fallen by an average 29−32% (*20*). At the same time, the utilization (output/input) of N rose from 20 to 38% from 1980 to 2005 to maintain crop yields. In spite of the better utilization of nitrate, the environmental impact from agriculture remains high given current environmental objectives due mainly to the relatively high nutrient surplus levels.

The Danish findings are comparable to the results from other countries as the US (*15, 24*), Belgium (*29*), and The Netherlands (*14*) although differences exist. Thus, the increasing nitrate concentrations in the US groundwater since 1950 are likely the results of increasing fertilizer use, and highest nitrate concentrations are found in shallow, oxic groundwater beneath areas with high N inputs (*15, 17, 39*). In The Netherlands, concentrations of conservative pollutants increased in groundwater recharged before 1985 and decreased after 1990 (*14*).

Thus, the Danish data strongly indicate that a reduction in the N surplus and the nitrate leaching from the Danish agriculture have a significant effect on the groundwater nitrate concentration. However, the decrease in the nitrate concentration of oxic groundwater may also be influenced by climatic and meteorological factors. The fact that the monitoring points with the youngest oxic groundwater have seen the more pronounced significant downward nitrate trends (44%) than the older oxic groundwater (9−27%) supports the existence of the anticipated effect on groundwater of a reduced nitrate leaching following a lowering of the N surplus in Danish agriculture.

Although this study shows clear evidence of an effect of reduced N leaching on the groundwater nitrate concentrations the last evaluation of the Danish action plans showed that the measures have not had the expected and required effect on all parts of the environment. For example, Danish coastal waters are among the coastal water most frequently experiencing hypoxia, globally (*40*). In addition, groundwater nitrate concentrations still have to be lowered significantly in order to ensure good ecological status of Danish estuaries and good chemical status of groundwater according to EU legislation (*12*).

Geologic and hydrological information about the groundwater wells and the chemical composition of the Danish groundwater is available free of charge via the Internet at http://geus.dk in the database JUPITER.

Acknowledgments

The presented groundwater data has been collected as part of the governmentally supported Danish National Environmental Monitoring Programme. Since 1988 many colleagues from the former Danish counties (abolished in 2006), The Environmental Centres (established in 2007) and GEUS have been playing central roles. Also, we thank Arne Kyllingsbæk for information regarding agricultural N surpluses. Especially thanks to the anonymous reviewers for constructive comments and suggestions to the first draft of the manuscript.

Literature Cited

(1) OECD. Environmental Performance in Agriculture in OECD Counties since 1990. Paris, France, 2008. www.oecd.org/tad/env/indicators (accessed November 26, 2010).

(2) Hansen, B.; Thorling, L. Use of geochemistry in groundwater vulnerability mapping in Denmark. *Geological Survey of Denmark and Greenland Bulletin* **2008**, *15*, 45–48. www.geus.dk/publications/bull(accessed November 26, 2010).

(3) Iversen, T. M.; Grant, R.; Nielsen, K. Nitrogen enrichment of European inland and marine waters with special attention to Danish policy measures. *Environ. Pollut.* **1998**, *102* (1), 771–780.

(4) Bak, J.; Tybirk, K. The EU Acidification Strategy: Sensitivity of calculated emission ceilings for nitrogen and sulphur for Denmark. *Environ. Pollut.* **1998**, *102* (1), 625–633.

(5) Dämmgen, U.; Hutchings, N. J. Emissions of gaseous nitrogen species from manure management: A new approach. *Environ. Pollut.* **2008**, *154* (3), 488–497.

(6) Fraters, B.; Kovar, K.; Willems, W. J.; Stockmar, J.; Grant, R. Monitoring effectiveness of the EU Nitrates Directive Action Programmes; RIVM report 500003007/2005; 2005; 290pp.

(7) Dalgaard, T.; Olesen, J. E.; Petersen, S. O.; Petersen, B. M.; Jørgensen, U.; Kristensen, T.; Hutchings, N. J.; Gyldenkærne, S.; Hermansen, J. E. Developments in greenhouse gas emissions and net energy use in Danish agriculture - How to achieve substantial CO_2 reductions. *Environ. Pollut.* 2010 (Accepted NitroEurope Special Issue, December 2010).

(8) Statistics Denmark. National Statistical Database 2010 (Statistikbanken). Danmarks Statistik, København. www.dst.dk (accessed November 26, 2010).

(9) Kyllingsbæk, A.; Hansen, J. F. Development in nutrient balances in Danish agriculture 1980−2004. *Nutr. Cycling Agroecosyst.* **2007**, *79* (3), 1573–0867.

(10) Dalgaard, T.; Hutchings, N.; Dragosits, U.; Olesen, J. E.; Kjeldsen, C.; Drouet, J. L.; Cellier, P. Effects of farm heterogeneity on modelling and upscaling of nitrogen losses and greenhouse gas emissions in agricultural landscapes. *Environ. Pollut.* 2010. NitroEurope Special Issue, December 2010.

(11) Hansen, B.; Kristensen, E. S.; Grant, R.; Høgh-Jensen, H.; Simmelsgaard, S. E.; Olesen, J. E. Nitrogen leaching from conventional versus organic farming systems - system modelling approach. *Eur. J. Agron.* **2000**, *13*, 65–82.

(12) Hinsby, K.; de Melo, M. T. C.; Dahl, M. European case studies supporting the derivation of national background levels and groundwater threshold values for the protection of dependent ecosystems and human health. *Sci. Total Environ.* **2008**, *401* (1−3), 1–20.

(13) Laier, T. Nitrate monitoring and CFC-age dating of shallow groundwaters - an attempt to check the effect of restricted use of fertilizers. In *Nitrates in Groundwater*; Razowska-Jaworek, L., Sadurski, A., Eds.; International Associations of hydrogeologists selected papers, A.A. Balkema Publishers: 2005; Chapter 24.

(14) Visser, A.; Broers, J.; van der Grift, B.; Bierkens, M. F. P. Demonstrating trend reversal of groundwater quality in relation to time of recharge determined by $^3H/^3He$. *Environ. Pollut.* **2007**, *148*, 797–807.

(15) Rupert, M. G. Decadal-Scale Changes of Nitrate in Groundwater of the United States, 1988−2004. *J. Environ. Qual.* **2008**, *37* September-October (supplement), 240–248.

(16) Böhlke, J. K.; Denver, J. M. Combined Use of Groundwater Dating, Chemical, and Isotopic Analyses to Resolve the History and Fate of Nitrate Contamination in 2 Agricultural Watersheds, Atlantic Coastal-Plain, Maryland. *Water Resour. Res.* **1995**, *31* (9), 2319–2339.

(17) Tesoriero, A. J.; Saad, D.; Burow, K. R.; Flick, E. A.; Puckett, L. L. Barbash. Linking ground-water age and chemistry data along flow paths: Implications for trends and transformations of nitrate and pesticides. *J. Contam. Hydrol.* **2007**, *94*, 139–155.

(18) Visser, A.; Broers, H. P.; Heerdink, R.; Bierkens, M. F. P. Trends of pollutants concentrations in relation to time of recharge and reactive transport at the groundwater body scale. *J. Hydrol.* **2009**, *369*, 427–439.

(19) European Environmental Agency. *Agriculture and environment in EU-15— the IRENA indicator report*; EEA Report No 6/2005 ISBN 92-9167-779-5; EEA: Copenhagen, 2005; ISSN 1725-9177.

(20) Kronvang, B.; Andersen, H. E.; Børgensen, C.; Dalgaard, T.; Larsen, S. E.; Bøgestrand, J.; Blicher-Mathiasen, G. Effects of policy measures implemented in Denmark on nitrogen pollution of the aquatic environment. *Environ. Sci. Policy* **2008**, *11* (2), 144–152.

(21) Jørgensen, L. F.; Stockmarr, J. Groundwater monitoring in Denmark: characteristics, perspectives and comparison with other countries. *Hydrogeol. J.* **2009**, *17*, 827–842.

(22) Geological Survey of Denmark and Greenland 2010. Information on the Danish groundwater monitoring programme can be found at http://www.geus.dk/ (accessed November 26, 2010).

(23) Busenberg, E.; Plummer, N. L. Use of chlorofluoromethanes (CCl_3F and CCl_2F_2) as hydrologic tracers and age-dating tolls: Example-The alluvium and terrace system of Central Oklahoma. *Water Resour. Res.* **1992**, *28*, 2257–2283.

(24) Hinsby, K.; Purtschert, R.; Edmunds, W. M. Groundwater age and Quality. In Quevauviller, Ph., Ed.; *Groundwater Science and Policy - An International Overview*; RSC Publishing, The Royal Society of Chemistry: Cambridge, 2008; pp 217−239.

(25) Warner, M. J.; Weiss, R. F. Solubilities of chlorofluorocarbons 11 and 12 in water and seawater. *Deep-Sea Res.* **1985**, *21*, 1485–1497.

(26) Engesgaard, P.; Højberg, A. L.; Hinsby, K.; Jensen, K. H.; Laier, T.; Larsen, F.; Busenberg, E.; Plummer, L. N. Transport and Timelag of Chlorofluorocarbon Gases in the Unsaturated Zone Rabis Creek, Denmark. *Vadose Zone J.* **2004**, *3*, 1249–1261.

(27) Geological Survey of Denmark and Greenland 2009. Download of data from the database JUPITER at http://www.geus.dk/.

(28) Grath, J.; Scheidleder, A.; Uhlig, S.; Weber, K.; Kralik, M.; Keimel, T.; Gruber, D. *The EU Water Framework Directive: Statistical aspects of the identification of groundwater pollution trends, and aggregation of monitoring results*; Final report. Austrian Federal Ministry of Agriculture and Forestry, Environment and Water Management (Ref.: 41.046/01-IV1/00 and GZ 16 2500/2-1/6/00), European Commission (Grant Agreement Ref.: Subv 99/130**794**). 2001.

(29) Aguilar, J. B.; Orban, P.; Dassargues, A.; Brouyère, S. Identification of groundwater quality trends in a chalk aquifer threatened by intensive agriculture in Belgium. *Hydrogeol. J.* **2007**, *15*, 1615–1627.

(30) SAS 2008. SAS/STAT 9.2; SAS Institute Inc.: Cary, NC, 2008.

(31) Postma, D.; Boesen, C.; Kristiansen, H.; Larsen, F. Nitrate Reduction in An Unconfined Sandy Aquifer - Water Chemistry, Reduction Processes, and Geochemical Modeling. *Water Resour. Res.* **1991**, *27* (8), 2027–2045.

(32) Hinsby, K.; Højberg, A. L.; Engesgaard, P.; Jensen, K. H.; Larsen, F.; Plummer, L. N.; Busenberg, E. Transport and degradation of chlorofluorocarbons (CFCs) in the pyritic Rabis Creek aquifer, Denmark. *Water Resour. Res.* **2007**, *43* (w10423), 1–15.
(33) EU. *Common implementation strategy for the Water Framework Directive (2000/60/EC)*. Guidance Document No. 18. Guidance of groundwater status and trend assessment. Technical Report - 2009−026; 2009.
(34) Busenberg, E.; Plummer, N. L. Dating groundwater with trifluoromethyl surfurpentafluoride (SF_5CF_3), sulfur hexafluoride (SF_6), CF_3Cl (CFC-13), and CF_2Cl_2 (CFC-12). *Water Resour. Res.* **2008**, *44*, W02431.
(35) Schlosser, P.; Stute, M.; Dorr, H.; Sonntag, C.; Munnich, K. O. Tritium/3He Dating of Shallow Groundwater. *Earth Planet. Sci. Lett.* **1988**, *89*, 353–362.
(36) Loftis, J. C. Trends of groundwater quality. *Hydrol. Processes* **1996**, *10*, 335–355.
(37) Broers, H. P.; van der Grift, B. Regional monitoring of temporal changes in groundwater quality. *J. Hydrol.* **2004**, *296*, 192–220.
(38) Larsen, M. A. D.; Soegaard, H.; Hinsby, K. Temporal trends in N & P concentrations and loads in relation to anthropogenic effects and discharge in Odense River 1964−2002. *Hydrol. Res.* **2008**, *39.1*, 41–54.
(39) Burow, K. R.; Nolan, B. T.; Rupert, M. G.; Dubrovsky, N. M. Nitrate in Groundwater of the United States, 1991−2003. *Environ. Sci. Technol.* **2010**, *44* (13), 4988–4997.
(40) Diaz, R. J.; Rosenberg, R. Spreading Dead Zones and Consequences for Marine Ecosystems. *Science* **2008**, *321*, 926–929.

ES102334U

Environmental Science & Technology

CORRESPONDENCE/REBUTTAL

pubs.acs.org/est

Comment on "Trend Reversal of Nitrate in Danish Groundwater—A Reflection of Agricultural Practices and Nitrogen Surpluses since 1950"

Hansen et al.[1] conclude in their paper "regulation and technical improvements in the intensive farming in Denmark have succeeded in decreasing the N surplus by 40% since the mid-1980s, while at the same time maintaining crop yields and increasing the animal production of especially pigs. Trend analyses prove that the youngest (0−15 years old) oxic groundwater shows more pronounced significant downward nitrate trends (44%) than the oldest (25−50 years old) oxic groundwater (9%). This amounts to clear evidence of the effect of reduced nitrate leaching on groundwater nitrate concentrations in Denmark."

I believe that there are at least two problems with these conclusions that make them doubtful. The first problem relates to the ultimate interpretation of results shown in Table 1 regarding "clear" evidence of reduced leaching and the second problem relates to methodology used to obtain results of areal trends shown in Figure 3 that show overall decreases in concentrations of nitrate after about 1980. These two problems are discussed in order below.

1. Context of interpretation for results shown in Table 1.
   A major conclusion of the paper relating to Table 1 is misleading. The interpretation implies that reduced applications of fertilizer and better management practices have resulted in decreasing nitrate concentrations in groundwater, observable since the 1980s. Compared to the oldest water (>25 years), this is statistically true, as shown in the subsequent explanation. The more important question for environmental managers is, however, are there more decreasing trends *currently and in the recent past (post mid-1980s)* than "increasing" or "no trends" that reflect reduced leaching of nitrate to groundwater? Although it is true, on the basis of a simple Chi-square analysis of downward and no trends, that water younger than 15 years old had significantly higher percentage occurrence of decreasing trends than water older than 25 years (44% vs 9%), there is no statistical difference at a significance level of 0.05 between the occurrence of upward, downward, and no trends in the 0−15 year old water (χ-square = 5.927, df = 2, p-value = 0.0516) or in the 15−25 year old water (χ-square = 3.219, df = 2, p-value = 0.1998). *Thus, based on the data shown, there are no more downward than upward or no trends in the data occurring in either of the younger waters, providing no quantitative basis to say concentrations are currently decreasing in response to any factor.*
   Therefore, although there are more downward trends in both of the younger waters compared to the oldest, basically this is what we already know and has been documented in many studies: that groundwater nitrate concentrations were increasing beginning in the 1950s and that increases may have peaked after the 1980s. What we do not know, and this paper has not demonstrated, is whether nitrate may still be increasing, staying the same, or decreasing. However, the answer to the question "Are there more decreasing trends in younger water that reflect impact of changes in management?" based on the data shown in Table 1, is "no".
2. Methodology and decreasing trends presented in Figure 3. This particular figure would be quite compelling were it not for critical deficiencies including the (a) unequal distribution of sampling points through the period and (b) inappropriate application of a moving average. In addition, the decrease in nitrate concentration before any implementation of remedial actions is not explained, and the phenomenon by itself should have been key to the authors re-examining the data set and how it was analyzed. There also was no discussion about possible changes in lab methods relating to accuracy and precision of determinations that could have been wrongly interpreted as trends due to other factors, and no justification for outlier removal was given.
   a. Unequal distribution of sampling points through time and space
      Most problematic is the use of unequal number of sampling points through time and space. The first decade (1950−1960) has only about 5 points. By around 1995, there are 3 or 4 points, considerably fewer (and different) than the area represented in 1980s, where there are at least 10 points or more for several years. Although it is not absolutely necessary that each year have exactly the same sampling points, it is necessary that the same areas are represented every year; clearly, this is not possible with one or two points some years and ten or twelve in others. In short, the area and recharge time representation is not consistent for the period of analysis. If the area toward the end of the record is different from the beginning and middle, any resulting statistics analyzed for trend would be meaningless.
   b. In addition to unequal space and time representation, the concentrations of nitrate do not appear to be normally distributed and skewed. A running mean, sensitive to outliers, was used instead of simpler and more reliable estimator such as the median, which can be analyzed by nonparametric methods. If the concentration values are not normally distributed, the probabilities of occurrence associated with the sample mean are incorrect and confidence intervals are likewise incorrect. A statistically legitimate way to analyze the data would be to simply group the data by recharge period (assuming again, that the areas represented are the same for all

Published: April 01, 2011

three periods), compute the basic statistics for the three periods, show the median and confidence intervals for the nitrate concentrations, and use a Kruskal–Wallis test[2] to answer the question "Are there significant differences between the time steps?" Another method would be to apply a LOWESS smooth curve to these data that would give a reasonable indication about trend (both upward and downward). A Spearman rho correlation analysis[2] of the data split before and after the approximate middle of the entire period, could also have legitimately be applied to determine upward, downward, or no trends.

With non-normally distributed nitrate concentration data, inappropriate use of an estimator such as the mean and uneven distribution through time and space of data points, the conclusions of the article are questionable in their present form.

Timothy B. Spruill
*Hydrologist**

U.S. Geological Survey North Carolina Water Science Center, 3916 Sunset Ridge Road, Raleigh, North Carolina 27607, United States

AUTHOR INFORMATION

Corresponding Author
*E-mail: tspruill@usgs.gov.

DISCLOSURE

The author's stated views are his own and do not reflect the views of the U.S. Geological Survey.

REFERENCES

(1) Hansen, B.; Thorling, L.; Dalgaard, T.; Erlandsen, M. Trend reversal of nitrate in Danish groundwater—A reflection of agricultural practices and nitrogen surpluses since 1950. *Environ. Sci. Technol.* **2011**, *45*, 228–23410.1021/es102334u.

(2) Conover, W. J. *Practical Nonparametric Statistics*; John Wiley and Sons: New York, 1980; 493 pp.

Biogeosciences, 9, 3277–3286, 2012
www.biogeosciences.net/9/3277/2012/
doi:10.5194/bg-9-3277-2012

Biogeosciences

Regional analysis of groundwater nitrate concentrations and trends in Denmark in regard to agricultural influence

B. Hansen[1], **T. Dalgaard**[2], **L. Thorling**[1], **B. Sørensen**[1], **and M. Erlandsen**[3]

[1]Department of Groundwater and Quaternary Geology Mapping, Geological Survey of Denmark and Greenland – GEUS, Lyseng Allé 1, 8270 Højbjerg, Denmark
[2]Department of Agroecology, Aarhus University, Denmark
[3]Department of Public Health, Aarhus University, Denmark

Correspondence to: B. Hansen (bgh@geus.dk)

Received: 3 April 2012 – Published in Biogeosciences Discuss.: 4 May 2012
Revised: 5 July 2012 – Accepted: 8 July 2012 – Published: 23 August 2012

Abstract. The act of balancing between an intensive agriculture with a high potential for nitrate pollution and a drinking water supply almost entirely based on groundwater is a challenge faced by Denmark and similar regions around the globe. Since the 1980s, regulations implemented by Danish farmers have succeeded in optimizing the N (nitrogen) management at farm level. As a result, the upward agricultural N surplus trend has been reversed, and the N surplus has reduced by 30–55 % from 1980 to 2007 depending on region. The reduction in the N surplus served to reduce the losses of N from agriculture, with documented positive effects on nature and the environment in Denmark. In groundwater, the upward trend in nitrate concentrations was reversed around 1980, and a larger number of downward nitrate trends were seen in the youngest groundwater compared with the oldest groundwater. However, on average, approximately 48 % of the oxic monitored groundwater has nitrate concentrations above the groundwater and drinking water standards of 50 mg l^{-1}. Furthermore, trend analyses show that 33 % of all the monitored groundwater has upward nitrate trends, while only 18 % of the youngest groundwater has upward nitrate trends according to data sampled from 1988–2009. A regional analysis shows a correlation between a high level of N surplus in agriculture, high concentrations of nitrate in groundwater and the largest number of downward nitrate trends in groundwater in the livestock-dense northern and western parts of Denmark compared with the southeastern regions with lower livestock densities. These results indicate that the livestock farms dominating in northern and western parts of Denmark have achieved the largest reductions in N surpluses. Groundwater recharge age determinations allow comparison of long-term changes in N surplus in agriculture with changes in oxic groundwater quality. The presented data analysis is based on groundwater recharged from 1952–2003, but sampled from 1988–2009. Repetition of the nitrate trend analyses at five-year intervals using dating of the groundwater recharged in the coming years and a longer time series of the nitrate analyses can reveal the evolution in nitrate leaching from Danish agriculture during the past 10 yr. Similar analyses can be carried out to compare with other regions internationally.

1 Introduction

Intensive agriculture is a major source of environmental N (nitrogen) pollution with severe N losses to soil, water and air. The environmental effects include a decline in biodiversity, eutrophication of ecosystems and surface waters, acidification, global warming, air pollution and diffuse nitrate pollution of groundwater. N pollution from intensive agriculture not only affects the environment, but may also affect human health due, for example, to the presence of N-containing particles in the atmosphere, which may give rise to respiratory health problems and diseases, or nitrate in drinking water, which may pose risks for some types of cancer, although no firm conclusions exist (van Grinsven et al., 2010; Erisman et al., 2011).

The manufacture of nitrogen-containing fertilizer for food production and the cultivation of leguminous crops convert

Published by Copernicus Publications on behalf of the European Geosciences Union.

atmospheric N_2 into reactive forms that significantly perturb the global nitrogen cycle and threaten the stability of the planet (Rockström et al., 2009). Globally, industrial N fixation has increased exponentially from near zero in the 1940s (Vitousek et al., 1997). The production of nitrogen fertilizers has been the main reason for the increase in world crop productivity, thus supporting the human population growth, but nitrogen fertilizers also cause N imbalances in agricultural development in all parts of the world (Vitousek et al., 2009).

A global challenge is to produce enough food for the ever-growing population and at the same time minimizing the loss of N to the environment. Since the 1980s, agriculture in Western Europe has managed to reduce its nitrogen surpluses, owing to stringent national and European community policies (Vitousek et al., 2009; Grizzetti et al., 2011; Hansen et al., 2011; Dalgaard et al., 2012). However, Vitousek et al. (2009) reckon that regions in Africa continue to extract the nutrient capital of what were once highly fertile soils with low yields, while in contrast intensive agricultural production in Northern China has a very high input of N to agricultural fields and high yields, but also a very high N loss to the environment.

Nutrients in the soil are leached when the supply exceeds the nutrient demand of the plant. Since 1980, agriculture in Denmark has been able to reduce its N surplus by approximately 40 % while maintaining crop yields. The result of the reduction in the agricultural N surplus is reflected in respective reductions in nitrate leaching of on average 33 % (Kronvang et al., 2008), the N load in surface waters of approx. 29–32 % and groundwater nitrate concentrations of approx. 40 % (Hansen et al., 2011). Also other countries such as The Netherlands (Visser et al., 2007), Belgium (Aguilar et al., 2007) and the US (Rupert, 2008; Burow et al., 2010) have observed effects on groundwater nitrate concentrations due to impact from fertilizer use in agriculture. Several Danish initiatives have been taken to reduce the N pollution from agriculture. Some of the most effective environmental measures have been a reduction in the statutory and crop-specific N fertilisation standards and N utilization requirements of manures which has raised the overall N use efficiency from 27 % in 1985 to 40 % in 2008 (Dalgaard et al., 2011a).

In Denmark, public drinking water supplies almost entirely originate from groundwater and approximately 15 % of the total area of Denmark has therefore been classified as nitrate-vulnerable abstraction areas (Hansen and Thorling, 2008) with many waterworks and wells having been turned off due to nitrate pollution. Groundwater protection is therefore a high priority, and since 1985 it has been one of the most important drivers of regulation of the Danish agricultural sector through national action plans (Kronvang et al., 2008) and EU policies (Uthes et al., 2011; Happe et al., 2011).

The present paper continues the analysis initially presented and published in Hansen et al. (2011). The focus is still on nitrate in the oxic zone of the Danish groundwater, because we are examining the effect of nitrate leaching on groundwater nitrate concentrations. In oxic Danish groundwater, it can been assumed that nitrate leached to groundwater acts as a conservative compound under the presence of oxygen and the generally low reactivity of organic matter below the root zone. The aims of the present study are to better understand the geographic distribution of nitrate in groundwater and the evolution of nitrate trends in Denmark at different groundwater recharge ages by examining the influence from (1) regionally calculated N losses from agriculture and (2) the local amount of groundwater recharge.

2 Methods

2.1 Agricultural, geological and hydrological conditions

Denmark has a total land area of about 43 000 km^2, and about two-thirds of this is under agricultural use. The fertilization rate in Denmark is high compared with other European countries (OECD, 2010; European Environmental Agency, 2005). The average livestock density is about 0.8 livestock units per hectare (Dalgaard et al., 2011b), and the average input of N to agricultural land was about 180 kg ha^{-1} yr^{-1} in 2008 (Statistics Denmark, 2010; Kronvang et al., 2008). The land surface has a modest topography where the highest point is 170 m above sea level. The climate in Denmark is coastal temperate, and the precipitation varies from about 600 to 1000 mm yr^{-1}. The upper geologic layers are mainly 50–200 m thick Quaternary glacial deposits underlain by Tertiary marine and fluvial deposits or Cretaceous limestone and chalk. The aquifers thus consist of either unconsolidated sands and gravels or fractured limestone and chalk.

2.2 Nitrate reduction in Danish groundwater

The Danish groundwater can be divided into an upper oxic zone and a, usually, deeper reduced zone (Fig. 1). Nitrate reduction takes place in an intervening zone, called the nitrate-containing anoxic zone, between the oxic and the reduced zones. The redox interface divides the upper nitrate containing zones from the reduced zone. Hydro-geological heterogeneity and variation in the reduction capacity of the sediments can locally result in a complex transition between the oxic and the reduced zones (Hansen and Thorling, 2008). These circumstances also give rise to variation in the thickness of the anoxic zone from a few mm to more than 15 m across the country. In Denmark, the oxic zone in the Quaternary deposits has developed after the latest glaciations. On exposed residues of Saalean landscapes in western Denmark, the oxidation processes have been active for more than 100 000 yr, whereas, in the rest of the country, they have only been active for about 12 000 yr due to differences in the extension of the latest glaciations.

Nitrate reduction in the soil and groundwater is often microbially controlled. In the unsaturated zone, the

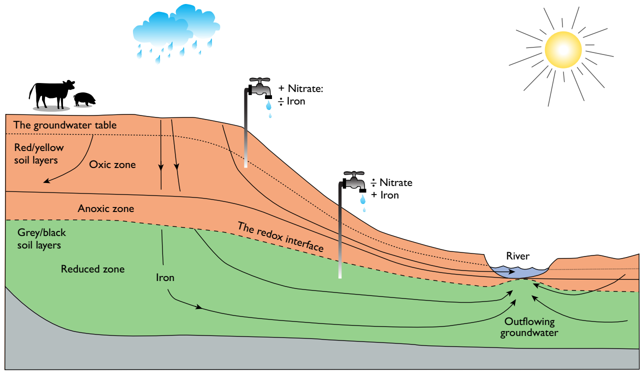


Fig. 1. Conceptual model of the typical groundwater redox environment in Denmark with oxic zone, anoxic zone with nitrate reduction and reduced zone. The interface between the anoxic zone containing nitrate and the reduced nitrate-free zone is called the nitrate interface and can also be determined based on the colours of the sediments. Above the nitrate interface, the soil layers have red and yellow colours, and below the nitrate interface the soil layers have grey and black colours.

denitrification processes take place in the reduced microenvironments due only to reduction of organic matter (Ernstsen, 1999). In the groundwater aquifers, organic matter, ferrous ions and pyrite (Postma et al., 1991) are the dominating nitrate-reducing agents causing the denitrification processes that take place in the nitrate-reducing anoxic zone.

2.3 Nitrate concentrations in oxic groundwater

Different type of nitrate data has been used in the statistical analyses of nitrate in Danish groundwater according to the purposes which are summarized in Table 1. Data on nitrate concentrations from all types of wells (monitoring, investigations, abstraction, etc.) have been integrated to obtain a national overview of the geographic distribution of the nitrate in oxic groundwater in Denmark set out in Fig. 2a. Like many other data used for chemical analyses of the Danish groundwater, nitrate concentrations are being reported to the national database JUPITER. Totally, there are 162 144 nitrate analyses sampled from 1890–2011. The data used in Fig. 2 were downloaded in January 2011 and consist of 3757 oxic monitoring points with average nitrate concentration based on 11 518 analyses sampled in the period 1967–2011. Data from such a long period are used in order to obtain as many nitrate analyses as possible from the oxic zone to create a national overview. Data stored in JUPITER have been analyzed by professionally certified laboratories.

Before nitrate concentrations are determined in the laboratories, the groundwater samples undergo normal analysis in the field which includes online measurements of pH, redox potential, oxygen concentration, temperature and conductivity. This approach ensures a high analytical quality and representative groundwater samples. Performing field analyses has been normal procedure over the last approximately 20 yr in the Danish Groundwater Monitoring Programme. The sampling and the field analyses are performed according to Danish technical standards.

Table 1. Data used in the statistical analyses of nitrate in Danish groundwater.

	All types of wells	Danish Groundwater Monitoring Programme	
Total amount of nitrate analyses	162 144	46 800	46 800
Sampling period	1890–2011	1973–2009	1973–2009
Total amount of nitrate analyses from oxic groundwater	11 518	194[1]	5321[2]
Sampling period of oxic analyses	1967–2011	1997–2006[1]	1988–2009[2]
Amount of oxic monitoring points	3757	194[1]	152[2]
Presentations	Fig. 2a	Fig. 3	Fig. 2b and c Figs. 4 and 5

[1] Nitrate analyses used for trend analyses on a national scale. CFC dated oxic monitoring points.
[2] Nitrate analyses used for trend analyses on an individual scale. CFC dated oxic monitoring points with stable oxic conditions and time series of more than 8 yr.

The redox condition of the groundwater is used to segregate the relevant subset of data used in this study where only data from the oxic zone are used. This subset of data from the oxic zone represents groundwater with a nitrate content mirroring the original nitrate leaching from the root zone. In oxic groundwater, nitrate is expected to act as an inert tracer due to the presence of oxygen and the generally low reactivity of organic matter below the root zone.

In this study the oxic zone is defined as follows:

– Nitrate > 1 mg l^{-1},

– Iron < 0.2 mg l^{-1}, and

– Oxygen > 1 mg l^{-1}

2.4 Nitrate concentrations used for trend analyses on a national scale

Only nitrate concentrations from the Danish Groundwater Monitoring Programme are used in the national trend analysis presented in Fig. 3. Details about the purpose, construction and hydro-geological conditions of the sites in the Danish Groundwater Monitoring Programme can be found in Jørgensen and Stockmarr (2009) and Hansen et al. (2011).

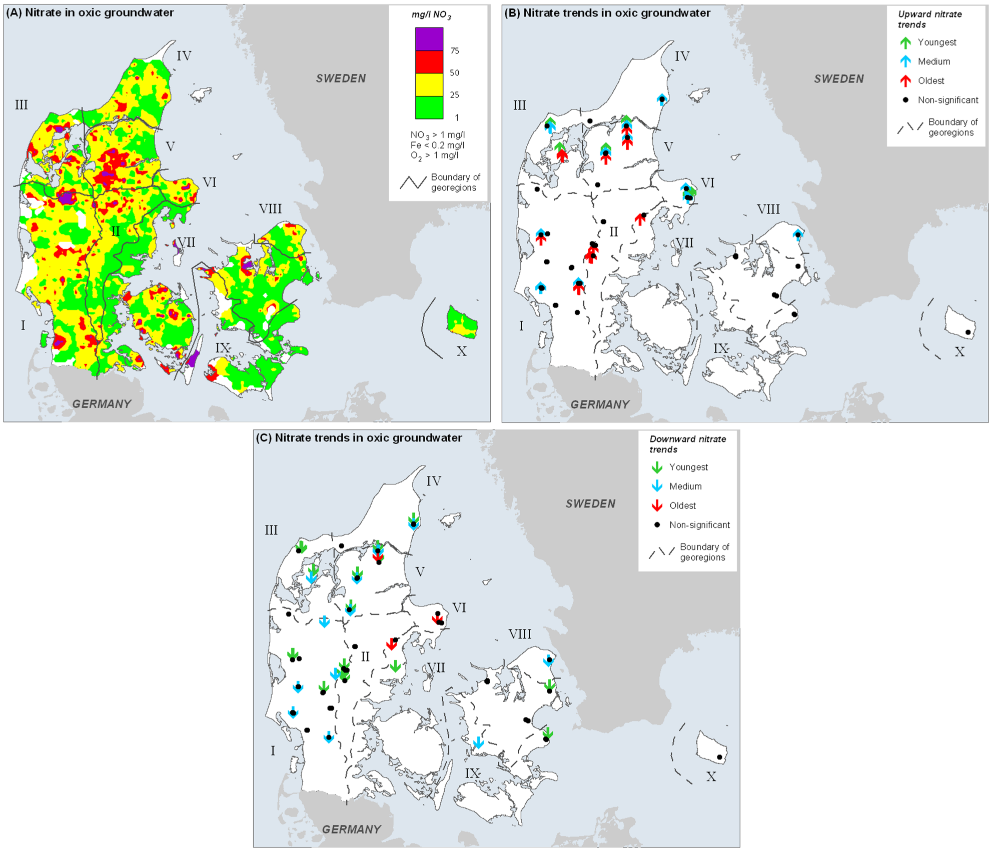


Fig. 2. (A) Interpolated geographic distribution of nitrate concentrations in oxic groundwater in Denmark based on 3757 analyses performed from 1890 to 2010. Average values from each measuring point are used in the interpolation. Shown are also 10 different geo-regions. **(B, C)** Geographic distribution of nitrate trends determined in 152 oxic CFC-dated groundwater monitoring points sampled from 1988 to 2009. A downward nitrate trend represents a negative slope of the linear regression line of nitrate versus sampling year for each groundwater monitoring point, while an upward trend shows a positive slope. The upward and downward nitrate trends are statistically significant with a 95 % confidence level. Statistically non-significant nitrate trends are also shown.

The programme covers 74 clustered catchments covering areas of 5 to 50 km^2, each containing about 25 wells. The groundwater monitoring sites are affected by different types of (1) agricultural land uses, (2) geological settings, (3) hydrological conditions, (4) depths, and (5) groundwater redox conditions. The entire Danish monitoring dataset includes approximately 46 800 nitrate analyses from 1500 groundwater monitoring points (Table 1). Only a subset of the complete dataset is used in the trend analyses in the present study.

The nitrate concentrations used for trend analyses on a national scale originate from 194 groundwater monitoring points with oxic groundwater where the groundwater recharge age has also been determined using the CFC (chlorofluorocarbon) method, typically once during the period 1997–2006. The groundwater recharge age determination allows the comparison of long-term changes in N surplus in agriculture with changes in oxic groundwater quality (Hinsby et al., 2008). The CFC analyses were performed according to the procedure of Laier (2005) as described in Hansen et al. (2011).

2.5 Nitrate concentrations used for trend analyses on an individual scale

Nitrate concentrations from the Danish Groundwater Monitoring Programme are also used in the trend analyses for individual point measurements depicted in Figs. 2b, c, 4 and 5.

The nitrate concentrations used for trend analyses for individual point measurements comprise 5321 nitrate analyses from 152 monitoring points sampled from 1988–2009 where the groundwater (1) had stable oxic conditions, (2) was CFC-dated and (3) had time series of between 8 and 20 yr with approximately one nitrate analysis per year (Table 1).

The groundwater chemistry data from the Danish Groundwater Monitoring Programme used in the trend analyses were downloaded from the Danish national geo-database (JUPITER) in October 2009 (www.geus.dk).

2.6 Nitrogen surpluses in agriculture

The annual national surplus of N in agriculture is estimated as the difference between inputs (synthetic fertilizer, import of animal feed, organic waste products, net atmospheric deposition and fixation) and outputs (export of plant and animal products). The annual national N surplus presented in this paper is estimated based on information from Statistics Denmark (2010) and Dalgaard and Kyllingsbæk (2003) on these entries in the budget.

The surplus of nutrients, and especially N, is regarded as the best overall environmental indicator for the changes in the agricultural impact on the environment over a certain time period (European Environmental Agency, 2005). The surplus represents the amount of N pooled in the soil, or not being used up by the production system, and which is therefore at risk of being lost to the environment (Dalgaard et al., 2011b; Hansen et al., 2000). The N surplus can be described as the sum of nitrate leaching ($N_{leaching}$), ammonia emission ($N_{emission}$), denitrification, and accumulation of N in the soil ($N_{accumulation}$) according to the definition of Dalgaard et al. (2011b), and formulated in Eq. (1):

$$N_{surplus} = N_{leaching} + N_{emission} + N_{denitrification} + N_{accumulation} \quad (1)$$

In this paper, the regional N surpluses for each of the ten Danish geo-regions shown in Fig. 2 are estimated from the annual, national N surplus as accounted for by Hansen et al. (2011). For each geo-region and for each year from 1950 to 2007, the livestock units are sourced from national county statistics and accord with the linear relationship between livestock units and N surplus identified by Dalgaard et al. (2011b). The annual, national N surplus values are apportioned according to the number of livestock units in each geo-region. In this way an approximate N surplus in each geo-region is found. However, this might differ from the "true" N surplus in the geo-region, for example, due to different distributions of livestock and individual farming practices in each region.

2.7 Water balances

Water balance components were estimated by the national water resources model called the DK model (Højberg et al., 2012), which is a coupled surface-groundwater model with a horizontal discretization of 500×500 m covering the entire country with the exception of minor islands. The model is set up in the MIKE SHE/MIKE11 model system, where the unsaturated zone is described by a water balance module, while the saturated zone is described by a comprehensive three-dimensional groundwater component to estimate recharge to and hydraulic heads in different geological layers. Stream-aquifer interaction and stream flow-routing are described by MIKE11. The model is constructed on the basis of comprehensive national databases on geology, soil, topography, river systems, climate and hydrology and has recently been updated to include hydrological interpretations from regional- and local-scale hydrological models.

Daily groundwater recharge values (mm day^{-1}) are extracted from the model simulations with MIKE SHE and represent 10-yr average values for the period 1998–2007. For each of the 152 groundwater monitoring points, a groundwater recharge value is found from the 500×500 m cell from the groundwater model where the well is situated.

2.8 Statistical methods

2.8.1 Gridding of nitrate concentrations

The data on the nitrate concentration map in Fig. 2 were interpolated using the kriging method of the Surfer programme (Surfer, 2002). A semi-variogram was fitted with an exponential function. The search radius of 10 km and a cell size of 2500 m were used.

2.8.2 Nitrate trend analyses

Determination of nitrate trends was achieved using the SAS software system (SAS, 2008) as described in Hansen et al. (2011).

3 Results

3.1 Geographic assessment of nitrate in oxic groundwater

Figure 2a shows that nitrate has been found in the oxic part of the groundwater throughout Denmark, with concentrations of up to 360 mg l^{-1} when using average nitrate concentrations based on all available data from oxic groundwater from 1967–2011. Nitrate concentrations of 25 mg l^{-1} are exceeded in 54 % of the datasets, corresponding to 55 % of the total area of Denmark, and nitrate concentrations of 50 mg l^{-1} are exceeded in 24 % of the dataset, equivalent to 10 % of the total area of Denmark (Fig. 2a). However, if we consider only data from the Danish Groundwater Monitoring Programme sampled from 1988–2009 and representing ages up to maximum 50 yr, then the nitrate concentrations of 25 mg l^{-1} and 50 mg l^{-1} are exceeded in 79 % and 48 % of the 152 oxic monitoring points, respectively. The nitrate concentrations in the oxic groundwater data from the Danish Groundwater Monitoring Programme are higher than the concentrations that appear from the data shown in Fig. 2a, because the oxic data from the Danish Groundwater Monitoring Programme on average represent younger and more nitrate-polluted groundwater.

However, Fig. 2a shows a regional pattern of nitrate concentrations where the oxic groundwater is most severely polluted with nitrate in northern and western Denmark (geo-regions I, II, III, IV, and V), while eastern Denmark (geo-regions VI, VII, VIII, IX, and X) is less polluted with nitrate.

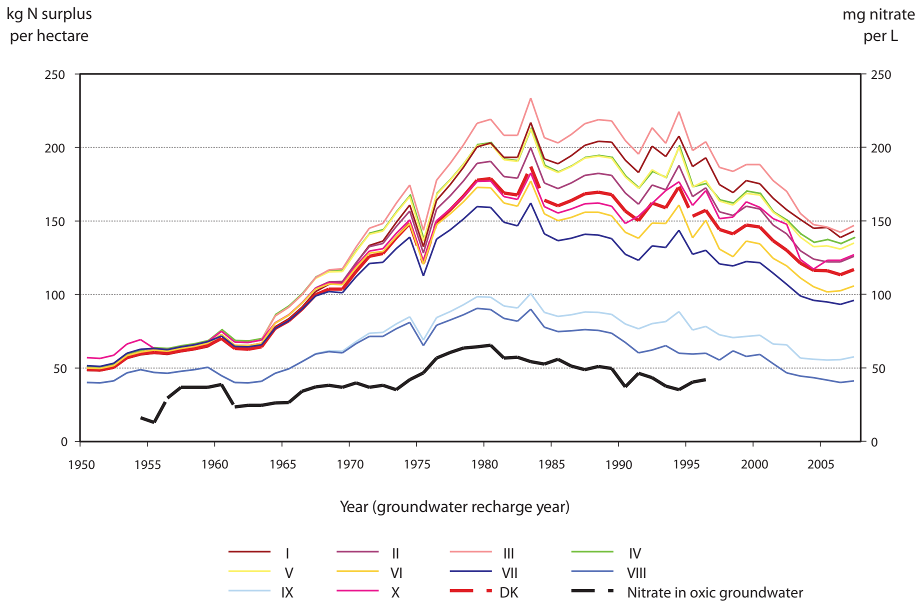


Fig. 3. Time series of agricultural N surplus in 10 different geo-regions of Denmark, and nitrate in oxic groundwater versus recharge age (CFC-age) at an annual mean level. The nitrate concentrations in oxic groundwater are shown as a 5-yr moving average curve. The location of the 10 geo-regions is seen in Fig. 2a.

An examination of the geographic distribution of the upward and downward nitrate trends and non-significant nitrate trends in the 152 oxic groundwater monitoring points sampled from 1989–2009 reveals no obvious regional pattern (see Fig. 2b and c). However, the most pronounced downward nitrate trends seem to be where the concentrations of nitrate in the groundwater are highest in northern and western Denmark (geo-regions I, II, III, IV, and V).

The occurrence of oxic groundwater with high concentrations of nitrate is most likely due to a combination of (1) insufficient protection of the aquifer from overlying clay layers, (2) a low nitrate reduction capacity of the sediments of the aquifer, e.g. low content of potential reduction agents like pyrite, Fe^{II} and organic matter, (3) a high groundwater recharge, and (4) high nitrate leaching from agricultural land. Widespread pollution of groundwater with nitrate is therefore likely to be found where the redox interface has penetrated deeply into the soil layers (see Fig. 1.).

3.2 Regional trends of N surpluses and nitrate in oxic groundwater

Denmark is divided into ten different geo-regions according to Kronvang et al.(2008), and regional N balances have been calculated for each geo-region as seen in Fig. 2a. The regional balances are shown together with the national N balance in Fig. 3 and Table 2. All the ten different regional N balances have the same overall temporal pattern as the national N balance with minimums and maximums occurring at the same time. However, the regional N balances are staggered so that northern and western Jutland (geo-regions I, II, III, IV and V) have a higher and eastern and southern Denmark (geo-regions VI, VII, VIII, IX, X) have a lower N surplus level than the national average N surplus level.

The higher N surplus level in northern and western Jutland and lower N-surplus level in eastern and southern Denmark are in accordance with the geographic pattern of the nitrate concentration levels in oxic groundwater seen in Fig. 2a. There are many reasons for the geographic distribution of nitrate in oxic groundwater where the N surplus might be one of them, but other reasons might be the geographic distribution of soil types, land use, nitrate reduction capacity of the sediments, precipitation and groundwater recharge in Denmark.

In almost every geo-region in Denmark, the N-surpluses were approximately 50 kg N ha^{-1} yr^{-1} in 1950, where the country as a whole was characterized by mixed farming and the livestock was distributed evenly across all regions (Dalgaard and Kyllingsbæk, 2003). Over the following 30 yr, N surpluses rose dramatically in every geo-region and they reached a maximum around 1980 where they ranged from 234 kg N ha^{-1} yr^{-1} in Thy (geo-region III) in northern Jutland to 91 kg N ha^{-1} yr^{-1} in North Zealand (geo-region VIII) in the eastern part of Denmark. Since 1980, the N surpluses have decreased by 30–55 % in all Danish geo-regions. The national average N surplus reduction since 1980 is about 37 %, and the national average N surplus was 117 kg N ha^{-1} yr^{-1} in 2007 (see Table 2).

The trends in the regional N surpluses and the national N surpluses both show the same fluctuations as the nitrate

Table 2. Distribution statistics of annual agricultural N surpluses from 1950 to 2007 in 10 different geo-regions of Denmark, and in Denmark as a whole.

Geo-region No.	Geo-region Name	1950	Max	2007	Mean 1950–2007	Increase 1950–2007	Reduction 1980–2007
I	West Jutland	48	217	143	139	95 (198 %)	74 (34 %)
II	Mid Jutland	50	200	126	130	76 (152 %)	74 (37 %)
III	Thy	50	234	147	148	97 (194 %)	87 (37 %)
IV	North Jutland	50	212	138	137	88 (176 %)	74 (34 %)
V	Himmerland	50	213	135	137	85 (170 %)	78 (37 %)
VI	Djursland	50	177	106	117	56 (112 %)	71 (40 %)
VII	East Denmark	51	162	96	117	45 (88 %)	66 (41 %)
VIII	North Zealand	40	91	41	60	1 (3 %)	50 (55 %)
IX	South Zealand	40	101	58	68	17 (43 %)	43 (42 %)
X	Bornholm	57	183	127	125	70 (123 %)	56 (30 %)
DK	Denmark (in total)	49	187	117	122	68 (139 %)	70 (37 %)

concentration trends in oxic groundwater at the national level (Fig. 3). The nitrate concentrations in oxic groundwater are shown as a 5-yr moving average curve based on groundwater measurements (nitrate analyses and CFC dating) performed from 1997–2006 representing groundwater recharged from 1954–1996 (Fig. 3). This phenomenon is elucidated in detail in Hansen et al. (2011), who report a statistically significant nitrate trend reversal in oxic groundwater around 1980. Figure 3 also shows that the increase in N surpluses in agriculture in the 1950s, 1960s and 1970s levelled out after 1980.

3.3 Nitrate trends in oxic groundwater

The nitrate trends of the time series of nitrate concentrations in the 152 oxic groundwater monitoring points were assessed by linear regression, as described in detail in Hansen et al. (2011). The slopes of each of these 152 linear regression lines represent the nitrate trend or the changes in nitrate shown in mg nitrate l^{-1} yr^{-1}. Thus, negative slopes represent downward trends, while positive slopes represent upward nitrate trends. In Fig. 4, the upward and downward nitrate trends are illustrated according to the age of the groundwater determined with the CFC method and the data are divided into three age groups.

The general national nitrate trends (Fig. 3) show a trend reversal around 1980; however, the nitrate trends from the individual monitoring points show a more complex picture as seen in Fig. 4 and Table 3.

Ninety-four of the 152 oxic groundwater monitoring points have statistically significant ($p < 0.05$) nitrate trends, of which 50 are upward and 44 are downward. The remaining 58 monitoring points have non-significant nitrate trends.

As far as the upward nitrate trends are concerned, 20 % are in the youngest groundwater (< 15 yr), while 42 % are in the oldest groundwater (25–50 yr). The reverse pattern is found for the downward nitrate trends where 54 % are in the youngest groundwater and only 7 % in the oldest. Furthermore, these findings are challenged; according to Fig. 4 for the oldest groundwater, upward nitrate trends can be as high as 4 mg nitrate per l per year. In addition, the youngest groundwater can have downward nitrate trends up to almost 5 mg nitrate per l per year.

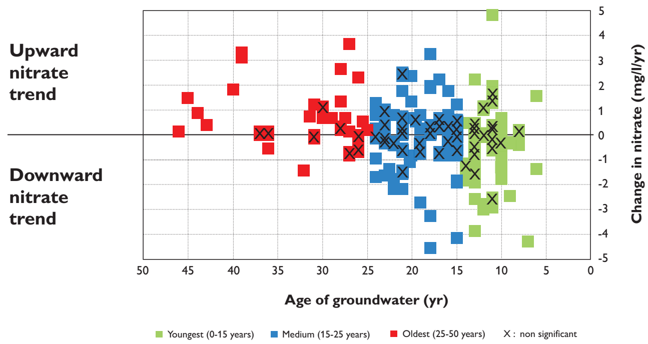


Fig. 4. Three age groups of upward and downward nitrate trends in oxic groundwater at a monitoring point level for 152 oxic CFC-dated groundwater measuring points sampled from 1988 to 2009. The change in nitrate (mg l^{-1} yr^{-1}) is equivalent to the slope of the linear regression lines of nitrate versus sampling year for each groundwater monitoring point. The age of the groundwater is determined with the CFC method.

The rate of changes in the nitrate concentrations (mg l^{-1} yr^{-1}) is highest in the groundwater monitoring points with the highest concentrations, which is both reflected in the significant upward and downward nitrate trends and in the non-significant nitrate trends (see Fig. 5a). Sixty-four per cent of the downward nitrate trends have mean nitrate concentrations above 50 mg l^{-1} as opposed to only 38 % of the upward nitrate trends. These findings are in line with the indications in Fig. 2 that the largest number of downward nitrate trends is found where the mean nitrate concentrations in groundwater are highest, i.e. in the northern and western parts (geo-regions I, II, III, IV, and V).

Table 3. Amount (%) of statistically significant upward, statistically significant downward and non-significant nitrate trends in 152 oxic CFC-dated groundwater monitoring points with a 95 % confidence level. The nitrate trends are grouped according to (1) groundwater recharge age, (2) the average nitrate concentrations in groundwater, (3) the annual groundwater recharge, and (4) the N load to groundwater.

		Upward	Downward	Non significant	Total
Total		50 (100 %)	44 (100 %)	58 (100 %)	152 (100 %)
Recharge age (yr)	< 15	10 (20 %)	24 (54 %)	21 (36 %)	55 (36 %)
	15–25	19 (38 %)	17 (39 %)	28 (48 %)	64 (42 %)
	25–50	21 (42 %)	3 (7 %)	9 (16 %)	33 (22 %)
Nitrate in groundwater ($mg\,l^{-1}$)	1–10	6 (12 %)	0 (0 %)	5 (9 %)	11 (7 %)
	10–50	25 (50 %)	16 (36 %)	27 (47 %)	68 (45 %)
	≥ 50	19 (38 %)	28 (64 %)	26 (44 %)	73 (48 %)
Groundwater recharge ($mm\,yr^{-1}$)	< 400	19 (38 %)	15 (34 %)	23 (40 %)	57 (38 %)
	400–600	24 (48 %)	23 (52 %)	21 (37 %)	68 (45 %)
	600–750	7 (14 %)	6 (14 %)	13 (23 %)	26 (17 %)
N load to groundwater ($kg\,N\,ha^{-1}\,yr^{-1}$)	< 25	18 (36 %)	15 (34 %)	21 (37 %)	54 (36 %)
	25–75	25 (50 %)	18 (41 %)	23 (40 %)	66 (44 %)
	≥ 75	7 (14 %)	11 (25 %)	13 (23 %)	31 (20 %)

As far as groundwater recharge is concerned, most of the upward and downward nitrate trends (approx. 50 %) are found at an annual mean recharge level of 400–600 $mm\,yr^{-1}$, and no obvious differences can be seen between the upward and downward nitrate trends (Fig. 5b).

The N load to groundwater is calculated by multiplying the nitrate concentrations and the groundwater recharge. Twenty-five per cent of the groundwater monitoring points with downward nitrate trends have a high N load (> 75 $kg\,N\,ha^{-1}\,yr^{-1}$) to the groundwater compared with only 11 % of the upward nitrate trends. This finding seems to be due to the nitrate concentration levels rather than to the local mean level of the groundwater recharge in Denmark.

4 Discussion and conclusions

Denmark has seen its farming sector develop a high livestock density due notably to the increase in pig production and the concentration of dairy farming in western Denmark over the last century (Dalgaard and Kyllingsbæk, 2003). Farms with a high livestock density have accomplished larger reductions in N surplus between the years 1990 and 2008 than farms with cash crop production using synthetic fertilizers and little livestock manure (Dalgaard et al., 2012). These circumstances can explain the findings in the present study where there seems to be consistency between a high N surplus in agriculture, high concentrations of nitrate in groundwater and the most pronounced downward nitrate trends in groundwater in northern and western parts of Denmark. In absolute values, the reduction in N surplus from 1980–2007 in these regions is also highest (74–87 $kg\,N\,ha^{-1}\,yr^{-1}$); however, the relative reduction in N surplus is lower (34–37 %) than in eastern and southern Denmark (40–55 %).

A clear indication of an effect of reduced N surplus in agriculture on groundwater nitrate concentrations in Denmark is seen in the age of the groundwater recharge relating to the upward and downward nitrate trends. Here 20 % of the youngest (0–15 yr old) and 42 % of the oldest (25–50 yr old) groundwater display upward nitrate trends, while the opposite pattern is seen for the downward nitrate trends where 54 % can be found in the youngest and 7 % are in the oldest groundwater.

Mean nitrate concentrations above 50 $mg\,l^{-1}$ are seen in 64 % of the downward nitrate trends in oxic groundwater, but only in 38 % of the upward nitrate trends. The N load in groundwater is the amount of nitrogen being transported by the groundwater and which might eventually flow out into groundwater-dependent ecosystems. The geographical variation in nitrate concentrations in groundwater rather than the amount of groundwater recharge influences the distribution of the groundwater N load, and 25 % of the groundwater monitoring points with downward nitrate trends have a high N load compared with only 11 % of the upward nitrate trends.

Together with the findings in Hansen et al. (2011), this study demonstrates a clear relationship between changes in the N surplus in agriculture, both at national and regional level, and changes in the nitrate concentrations in oxic groundwater with the same synchronic temporal pattern and trend reversals around 1980. The change and the development in Danish agricultural management of N have been driven mainly by politically enforced regulations since 1985, but also changes in the economic and technical conditions for farming with fluctuating product prices and revisions of the European agricultural policies have been instrumental in accomplishing these changes (Uthes et al., 2011; Happe et al., 2011). Regulations in agriculture have a significant impact

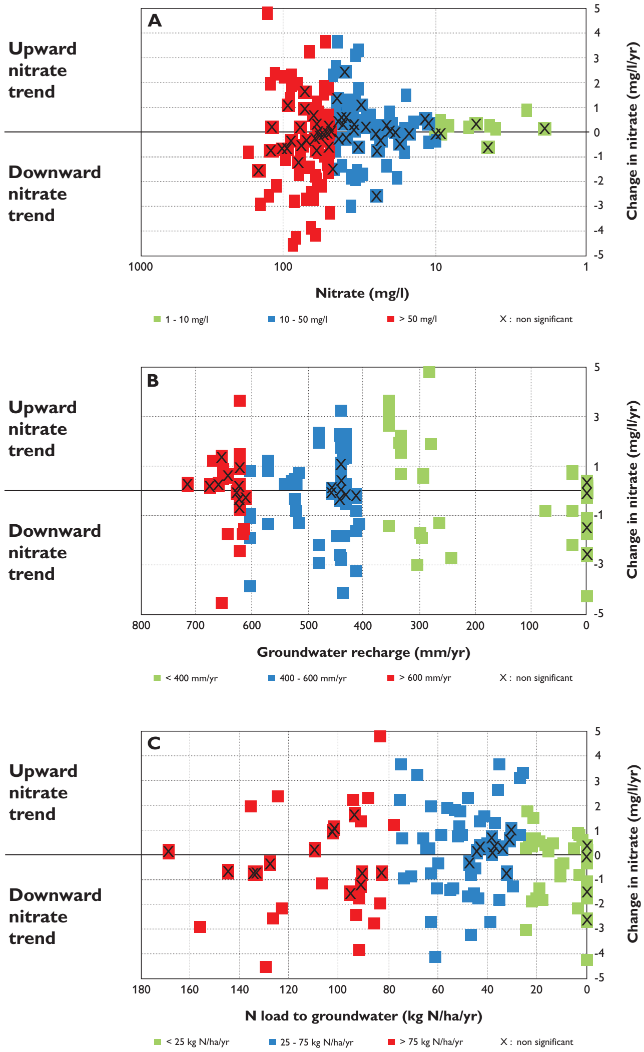


Fig. 5. Upward and downward nitrate trends in 152 oxic CFC-dated groundwater monitoring points sampled from 1988 to 2009. **(A)** The mean nitrate concentration (mg l^{-1}) in each groundwater monitoring point. **(B)** The annual groundwater recharge (mm yr^{-1}) in a circle of 1 km around each groundwater monitoring point. **(C)** the N load (kg N ha^{-1} yr^{-1}) to groundwater based on data shown in **(A)** and **(B)**.

on the environment, as strongly indicated by the simultaneousness of the reduction in N surplus in agriculture and the reduced nitrate contamination of oxic groundwater. The experience from Denmark may provide inspiration for other countries where control of agricultural N losses is needed, as for example pointed out by Vitousek et al. (2009).

Of all the coastal waters in the world, those in Denmark are some of the most frequently exposed to hypoxia (Diaz and Rosenberg, 2008). The EU Water Framework Directive stipulates that Denmark reverses the upward trends in groundwater nitrate concentrations and complies with the groundwater quality standards of 50 mg l^{-1} in certain areas. Thus, the environmental goals in both Danish legislation and EU directives (The Nitrate Directive, 1991/696/EC; The Water Framework Directive, 2000/60/EC; and the Groundwater Directive, 2006/118/EF) have not yet been fully met. There is a need for more holistic future solutions to protect both groundwater, nature and the wider environment and to meet legislative requirements for a good chemical status of groundwater, and a good ecological status of the Danish estuaries and oceans is just one of the important goals.

The latest report from the Danish agricultural monitoring sites (Grant et al., 2011) shows a small increase in modelled nitrate leaching from 2003 to 2010. The results from the individual trend analyses presented in this paper are based on groundwater data sampled from 1988–2009, and they represent groundwater recharged from 1952–2003 where the oldest monitored groundwater is about 46 yr old and the youngest monitored groundwater is about 6 yr old. Repetition of the nitrate trend analyses at five-year intervals based on dating of the groundwater recharge in the coming years and a longer time series of the nitrate analyses will shed more light on the groundwater effect of the evolution in nitrate leaching from Danish agriculture during the past 10 yr.

Acknowledgements. The presented groundwater data are collected as part of the governmentally supported Danish Environmental Monitoring Programme.

Edited by: H. van Grinsven

References

Aguilar, J. B., Orban, P., Dassargues, A., and Brouyère, S.: Identification of groundwater quality tends in a chalk aquifer threatened by intensive agriculture in Belgium, Hydrogeol. J., 15, 1615–1627, 2007.

Burow, K. R., Nolan, B. T., Rupert, M. G., and Dubrovsky, N. M.: Nitrate in Groundwater in the United States, 1991–2003, Environmental Science and Technology, 44, 4988–4997, 2010.

Dalgaard, T. and Kyllingsbæk, A.: Developments in the nitrogen surplus and the fossil energy use in Danish Agriculture during the 20th century, in: Advances in Ecological Sciences, edited by: Usó, J. L., Patten, B. C., and Brebbia, C. A., WIT Press Southampton, UK, 18, 669–678, 2003.

Dalgaard, T., Hutchings, N., Børgesen, C., Vinther, F. P., and Hansen, B.: How can nitrogen utilization in farming systems be optimized to minimize losses?, proceeding from ifs (International Fertiliser Society), 2011a.

Dalgaard, T., Hutchings, N., Dragosits, U., Olesen, J. E., Kjeldsen, C., Drouet, J.,L., and Cellier, P.: Effects of farm heterogeneity on modelling and upscaling of nitrogen losses and greenhouse gas emissions in agricultural landscapes, Environ. Pollut., 159, 3183–3192, 2011b.

Dalgaard, T., Bienkowski, J. F., Bleeker, A., Drouet, J. L., Durand, P., Dragosits, U., Frumau, A., Hutchings, N. J., Kedziora, A., Magliulo, V., Olesen, J. E., Theobald, M. R., Maury, O., Akkal, N., and Cellier, P.: Farm nitrogen balances in six European agricultural landscapes – a method for farming system assessment, emission hotspot identification, and mitigation measure evaluation, Biogeosciences Discuss., 9, 8859–8904, doi:10.5194/bgd-9-8859-2012, 2012.

Diaz, R. J. and Rosenberg, R.: Spreading Dead Zones and Consequences for Marine Ecosystems, Science, 321, 926–929, 2008.

Erisman, J.W., van Grinsven, H., Grizzetti, B., Bouraoui, F., Powlson, D., Sutton, M.A., Bleeker, A. Reis, S.: The European nitrogen problem in a global perspective, in: The European Nitrogen Assessment, edited by: Sutton, M., Howard, C. M., Erisman J. W., Billen, G., Bleeker, A., Grennfelt, P., Grinsven, H. V., and Grizzetti, B, Cambridge, 9–31, 2011.

Ernstsen, V.: Micro- and macro-geochemical environments of clayey till deposits.The Geological Society of America, 1999 Annual meeting and exposition, October 25–28 1999, Denver, USA, GSA Abstracts with programme, p. A450, 1999.

European Environmental Agency: Agriculture and environment in EU-15 – the IRENA indicator report, EEA Report No 6/2005 ISBN 92-9167-779-5, ISSN 1725-9177, EEA, Copenhagen, 2005.

Grant, R., Blicher-Mathiesen, G., Jensen, P. G., Hansen, B., and Thorling, L.: Landovervågningsoplande 2010, NOVANA, Aarhus Universitet, DCE – Nationalt Center for Miljø og Energi, – Videnskabelig rapport fra DCE – Nationalt Center for Miljø og Energi nr. 3, 130 pp., 2011.

Grizzetti, B., Bouraoui, F., Billen, G., van Grinsven, H., Cardoso, A. C., Thieu, V., Garnier, J., Curtis, C., Howarth, R., and Johnes, P.: Nitrogen as a threat to European water quality, in: The European Nitrogen Assessment, edited by: Sutton, M., Howard, C. M., Erisman, J. W., Billen, G., Bleeker, A., Grennfelt, P., Grinsven, H. V., and Grizzetti, B., Cambridge, 379–404, 2011.

Hansen, B. and Thorling, L.: Use of geochemistry in groundwater vulnerability mapping in Denmark, Geol. Surv. Den. Greenl., 15, 45–48, 2008.

Hansen, B., Kristensen, E. S., Grant, R., Høgh-Jensen, H., Simmelsgaard, S. E., and Olesen, J. E.: Nitrogen leaching from conventional versus organic farming systems – system modelling approach, Eur. J. Agron., 13, 65–82, 2000.

Hansen, B., Thorling, L., Dalgaard, T., and Erlandsen, M.: Trend Reversal of Nitrate in Danish Groundwater – a Reflection of Agricultural Practices and Nitrogen Surpluses since 1950, Environm. Sci. Technol., 45, 228–234, 2011.

Happe, K., Hutchings, N., Dalgaard, T., and Kellermann, K.: Modelling the interactions between regional farming structure, nitrogen losses and environmental regulation, Agr. Syst., 104, 282–291, 2011.

Højberg, A. L., Troldborg, L., Stisen, S., Christensen, B. S. B., and Henriksen, H. J.: Stakeholder driven update and improvement of a national water resources model, Environ. Modell. Softw., submitted, 2012.

Hinsby, K.; Purtschert, R.; Edmunds, W.M:. Groundwater age and Quality, in: Groundwater Science and Policy – An International Overview, RSC Publishing, edited by: Quevauviller, P., Roy. Soc. Chem., Cambridge, 217–239, 2008.

Jørgensen, L. F. and Stockmarr, J.: Groundwater monitoring in Denmark: characteristics, perspectives and comparison with other countries, Hydrogeol. J., 17, 827–842, 2009.

Kronvang, B., Andersen, H. E., Børgensen, C., Dalgaard, T., Larsen, S. E., Bøgestrand, J., and Blicher-Mathiasen, G.: Effects of policy measures implemented in Denmark on nitrogen pollution of the aquatic environment, Environ. Sci. Policy, 11, 144–152, 2008.

Laier, T.: Nitrate monitoring and CFC-age dating of shallow groundwaters – an attempt to check the effect of restricted use of fertilizers, in: Nitrates in Groundwater, edited by: Razowska-Jaworek, L. and Sadurski, A., International Associations of hydrogeologists selected papers, A. A. Balkema Publishers, Chapter 24, 2005.

OECD: Environmental Performance in Agriculture in OECD Counties since 1990, available at: www.oecd.org/tad/env/indicators (last access: November 2010), Paris, France, 2010.

Postma, D., Boesen, C., Kristiansen, H., and Larsen, F.: Nitrate Reduction in An Unconfined Sandy Aquifer – Water Chemistry, Reduction Processes, and Geochemical Modeling, Water Resour. Res., 27, 2027–2045, 1991.

Rockström, J., Steffen, W., Noone, K., Persson, Å., Chapin, F. S., Lambin, E. F., Lenton, T. M., Scheffer, M., Folke, C., Schellnhuber, H. J., Nykvist, B., de Wit, C. A., Hughes, T., van der Leeuw, S., Rodhe, H., Sörlin, S., Snyder, P. K., Costanza, R., Svedin, U., Falkenmark, M., Karlberg, L., Corell, R. W., Fabry, V. J., Hansen, J., Walker, B., Liverman, D., Richardsonm K., Crutzen, P., and Foley, J. A.: A safe operating space for humanity, Nature, 461, 472–475, 2009.

Rupert, M. G.: Decadal-Scale Changes of Nitrate in Groundwater of the United States, 1988–2004, September–October, J. Environ. Qual., 37, 240–248, 2008.

SAS: SAS/STAT 9.2; SAS Institute Inc., Cary, NC, 2008.

Statistics Denmark: National Statistical Database 2010 (Statistikbanken), Danmarks Statistik, København, 2010.

Surfer: Surfer 8, Counturing and 3D Surface Mapping for Scientists and Engineers, Golden Software, 2002.

Uthes S., Piorr, A., Zander, P., Kedziorac, A., Ungaro F., Dalgaard, T., Kjeldsen, C., Stolze, M., Moschitz, H., Schader, C., Happe, K., Sahrbacher, A., Damgaard, M., Sattler, C., Toussainta, V., Casini, L., and Müller, C.: Regional impacts of abolishing direct payments: and integrated analysis in four European regions, Agr. Syst., 104, 110–121, 2011.

van Grinsven, H. J. M., Rabl, A., and de Kok, T. M.: Estimation of incidence of social cost of colon cancer due to nitrate in drinking water in the EU: a tentative cost-benefit assessment, Environ. Health-Glob., 9, 58, doi:10.1186/1476-069X-9-58, 2010.

Visser, A., Broers, J., van der Grift, B., and Bierkens, M. F. P.: Demonstrating trend reversal of groundwater quality in relation to time of rechragae determined by $^{3}H/^{3}He$, Environ. Pollut., 148, 797–807, 2007.

Vitousek, P. M., Aber, J. D., Howarth, R. W., Likens, G. E., Matson, P. A., Schindler, D. W., Schlesinger, W. H., and Tilman, G.: Human alternation of the global nitrogen cycle: sources and consequences, Ecol. Appl., 7, 737–750, 1997.

Vitousek, P. M., Naylors, R., Crews, T., David, M. B., Drinkwater, L. E., Holland, E., Johnes, P. J., Katzenberger, J., Martinelli, L. A., Matson, P. A., Nziguheba, G., Ojima, D., Palm, C. A., Robertson, G. P., Sanchez, P. A., Townsend, A. R., and Zhang, F. S.: Nutrient imbalances in agricultural development, Science, 324, 1519–1520, 2009.

Policies for agricultural nitrogen management—trends, challenges and prospects for improved efficiency in Denmark

2014 Environ. Res. Lett. 9 115002

(http://iopscience.iop.org/1748-9326/9/11/115002)

OPEN ACCESS
IOP Publishing

Environmental Research Letters

Environ. Res. Lett. 9 (2014) 115002 (16pp)

doi:10.1088/1748-9326/9/11/115002

Policies for agricultural nitrogen management—trends, challenges and prospects for improved efficiency in Denmark

Tommy Dalgaard[1], Birgitte Hansen[2], Berit Hasler[3], Ole Hertel[3], Nicholas J Hutchings[1], Brian H Jacobsen[4], Lars Stoumann Jensen[5], Brian Kronvang[6], Jørgen E Olesen[1], Jan K Schjørring[5], Ib Sillebak Kristensen[1], Morten Graversgaard[1], Mette Termansen[3] and Henrik Vejre[7]

[1] Aarhus University, Department of Agroecology. Blichers Allé 20, DK-8830 Tjele, Denmark
[2] Geological Survey of Denmark & Greenland—GEUS. Lyseng Allé 1, DK-8270 Højbjerg, Denmark
[3] Aarhus University, Department of Environmental Sciences, Frederiksborgvej 399, DK-4000 Roskilde, Denmark
[4] University of Copenhagen, Department of Food and Resource Economics. Rolighedsvej 25, DK-1870 Frederiksberg C, Denmark
[5] University of Copenhagen, Department of Plant and Environmental Sciences. Thorvaldsensvej 40, DK-1871 Frederiksberg C, Denmark
[6] Aarhus University, Department of Bioscience.Vejlsøvej 25, DK-8600 Silkeborg, Denmark
[7] University of Copenhagen, Department of Geosciences and Natural Resource Management. Rolighedsvej 23, DK-1858 Frederiksberg C, Denmark

E-mail: tommy.dalgaard@agro.au.dk

Received 31 July 2014, revised 8 September 2014
Accepted for publication 24 September 2014
Published 3 November 2014

Abstract

With more than 60% of the land farmed, with vulnerable freshwater and marine environments, and with one of the most intensive, export-oriented livestock sectors in the world, the nitrogen (N) pollution pressure from Danish agriculture is severe. Consequently, a series of policy action plans have been implemented since the mid 1980s with significant effects on the surplus, efficiency and environmental loadings of N. This paper reviews the policies and actions taken and their ability to mitigate effects of reactive N (N_r) while maintaining agricultural production. In summary, the average N-surplus has been reduced from approximately 170 kg N ha^{-1} yr^{-1} to below 100 kg N ha^{-1} yr^{-1} during the past 30 yrs, while the overall N-efficiency for the agricultural sector (crop + livestock farming) has increased from around 20–30% to 40–45%, the N-leaching from the field root zone has been halved, and N losses to the aquatic and atmospheric environment have been significantly reduced. This has been achieved through a combination of approaches and measures (ranging from command and control legislation, over market-based regulation and governmental expenditure to information and voluntary action), with specific measures addressing the whole N cascade, in order to improve the quality of ground- and surface waters, and to reduce the deposition to terrestrial natural ecosystems. However, there is still a major challenge in complying with the EU Water Framework and Habitats Directives, calling for new approaches, measures and technologies to mitigate agricultural N losses and control N flows.

Keywords: Denmark, nitrogen management, nitrogen policy development, nitrogen surplus, nitrogen use efficiency, reactive nitrogen, regulation

1. Introduction

In contemporary political debates it is often argued that the development of a green economy, with high levels of environmental protection, and the promotion of low-emission production chains, is an efficient pathway to sustainable economic growth (Carter 2007, The Commission on Nature and Agriculture 2013). In line with this it is argued that environmental protection and pollution mitigation are compatible with the benefits of economic growth, and that the costs are less than the environmental and resource costs of not protecting the environment resources (Stern 2007, OECD 2011, Jacobs 2013). This perspective implies that green growth can be promoted through correcting for environmental market failures, not only by market based incentives, but also by regulation that requires minimum efficiency standards. This may be particularly relevant for agriculture and the related bio-based production sectors (Parajuli *et al* 2014), where OECD (2011) emphasizes that 'an understanding of how growth occurs (production methods) is at least as important as how much growth takes place'.

Sustainable production methods imply a focus on all aspects of the agricultural production (Gu *et al* 2011). This paper deals with nitrogen (N) as a critical nutrient in agriculture. With N being central both as an input factor affecting agricultural production and as a critical component for pollution of the aquatic, marine, terrestrial and atmospheric environments (Sutton *et al* 2011), N policy and regulation is crucial for a sustainable production.

Denmark is here taken as an example of how concerns for agricultural production can be balanced against environmental protection concerns. It is one of the most intensively farmed regions in the world, where more than 60% of its land surface is used for agriculture, it has a food export that is more than twice the national consumption (FAO 2014), and has one of the most well-developed environmental regulation systems in the world (van Grinsven *et al* 2012).

For centuries, Denmark has been a major food supplier to the neighbouring countries, first with live steers to Germany and surplus cereals to Norway, and subsequently with butter and bacon to Britain and other overseas countries, following the introduction of cooperative dairy and slaughterhouses in the late 1800s (Bjørn 191748-9326, Odgaard and Rømer 2009). Initially the food production surplus was driven by biologically fixed N inputs from grass-clover and pulses (Kjaergaard 1994, Dalgaard and Kyllingsbæk 2003) and a significant expansion of the proportion of land ploughed (Dam and Jakobsen 2008). However, the largest expansion in the food production took place after world war II, and was driven by synthetic N-fertilizer inputs, increasing from 15 kg N ha^{-1} agricultural land in 1945 to 143 kg N ha^{-1} when it peaked in 1983 (Dalgaard and Kyllingsbæk 2003, Dalgaard *et al* 2009), and falling to the 74 kg N ha^{-1} it is today (Statistics Denmark 2012).

The expansion of agricultural N inputs after world war II gradually led to a parallel increase in agricultural N surpluses, and significantly increased N leaching to groundwater (Hansen *et al* 2011). With more than 60% of the land farmed and a 7500 km long coastline with shallow estuaries and coastal waters, this has resulted in severe environmental problems, and according to the EU Nitrate Directive Denmark has designated the whole territory as nitrate vulnerable. Increasing groundwater nitrate concentrations exacerbated the problems since the drinking water supply in Denmark is almost 100% groundwater based and, consequently, approximately 15% of the land was designated as nitrate vulnerable abstraction areas in 2005 (Hansen and Thorling 2008).

From 1985 and onwards the following series of political action plans to mitigate losses of N and other nutrients were implemented (updated from Dalgaard *et al* 2005, Kronvang *et al* 2008, Mikkelsen *et al* 2010):

A. 1985 Action Plan on nitrogen, phosphorus and organic matter (NPo)
B. 1987 Action Plan for the Aquatic Environment I (AP-I)
C. 1991 Action Plan for Sustainable Agriculture
D. 1998, 2000 Action Plan for the Aquatic Environment II (AP-II)
E. 2001 Ammonia Action Plan
F. 2004 Action Plan for the Aquatic Environment III (AP-III)
G. 2009 Green Growth Plan
H. 2011 Draft Plans for River Basin Management Plans (RBMPs), implementing the EU Water Framework Directive (WFD)

As a result of the early attention to marine pollution, the first action plans were based on both national and international political initiatives. Already in 1972 Denmark, France, Iceland, Norway, Portugal, Spain, and Sweden signed the Oslo Convention, prohibiting the direct dumping of harmful substances at sea. After the inclusion of among others the United Kingdom, The Netherlands and Germany, this treaty was amended in 1981 and is today included in the OSPAR (1992) 'Convention for the Protection of the Marine Environment of the North–East Atlantic'. The Danish action plans have subsequently been used to implement the EU Nitrates Directive of 1991 (The Council of the European Communities 1991), and the WFD in year 2000 (The European Parliament and the Council of the European Union 2000). Moreover, at an international level the ambitions of reducing nutrient loads to the environment are also important parts of treaties in relation to the HELCOM Baltic Marine Environment Protection Commission (The Helsinki Commission 2008), The Marine Strategy Framework

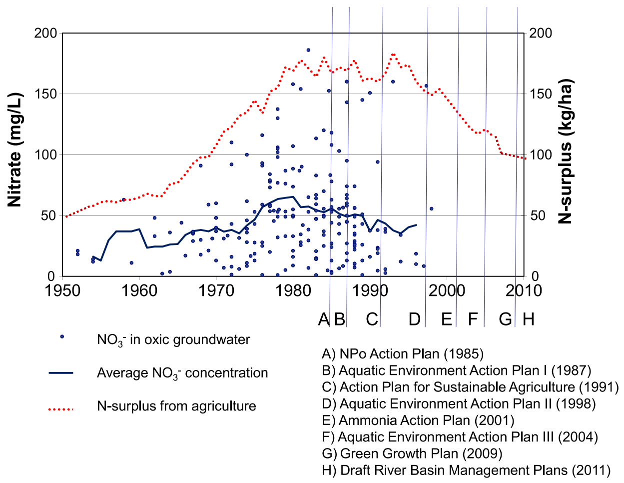


Figure 1. Relation between the nitrogen (N) surplus from Danish agriculture and the 5 yr moving average nitrate (NO_3^-) concentrations in Danish oxic groundwater samples (updated from Hansen *et al* 2011). The vertical lines labelled A–H indicate the timing of the series of major Danish action plans to reduce nitrogen pollution of the aquatic or atmospheric environment (for further details, see text and table 2).

Directive (The European Parliament and the Council 2008), and the from 1983 and onwards enforced UN Convention on Long-range Transboundary Air Pollution (CLRTAP 1979, Sutton *et al* 2014).

These initiatives and actions have led to significant reductions in N surplus from Danish agriculture (figure 1). The development in N surplus aligns well with the implementation of action plans and also with nitrate concentration in groundwater.

The objective of the present paper is to review and discuss listed developments in N management and environmental impacts in Denmark and the different types of policy action, pinpointing trends, challenges and prospects for improved future actions. We present a typology of the policy actions and the related effects on N surplus and N efficiency. Moreover, we assess the effects on the specific N loadings to the environment with special emphasis on loadings to the aquatic and atmospheric environments. Finally, we synthesize and discuss the relevant N flows cascading from agriculture to the environment, via stocks of N in soils, water, atmosphere and biomass, and through the various oxidation steps of reactive nitrogen (N_r); i.e., in the form of nitrate (NO_3^-), ammonia (NH_4), nitrous oxide (N_2O), nitrogen dioxide (NO_2), etc, with their impacts on nature and environment, production, consumption, economic costs, benefits and public health.

2. Materials and methods

2.1. Nitrogen data

Information on agricultural N inputs and outputs were collected, with focus on the period from 1990 until today, but also including longer time series back in time. In the analyses of N flows and balances, we delineated the system boundary to comprise the entire primary production of the Danish agricultural sector, including cropland, livestock and grazing lands, but excluding processing industry (slaughter houses, flour mills etc). This means that inputs include chemical fertilizers (all imported, since no national production), feedstuff (mainly imported concentrates) and urban and industrial waste products used as soil amendments, but not animal manures, which are entirely produced and recycled within the agricultural sector, as no national export or import of animal manures take place in Denmark (contrary to some other European countries, e.g. The Netherlands). Outputs accounted for include cash crops (cereals, seeds etc sold), milk, eggs, meat and live animals exported, but not fodder crops produced and fed to animals within the agricultural sector. National and regional data are collected from Statistics Denmark (1968, 1969 and 1961–2012) and supplemented by farm and field scale data available via the Danish Research Register for Agriculture, FRJOR (2014), including geo-coded information about all Danish livestock, manure and housing systems, fertilizer use, standard yields and the crops grown on all commercial farms.

Values for N fluxes into the environment are queried from the National Monitoring and Assessment Programme for the Aquatic and Terrestrial Environment (NOVANA), including the discharge at 178 near-coast gauging stations covering 57% of the entire land area of Denmark. Total nitrogen (TN) concentrations are measured at 118 monitoring stations based on discrete sampling at a frequency of 12–26 times per year (mean: 18 samples per year) (Windolf *et al* 2011). The runoff and loading of TN from the ungauged areas is then calculated utilizing a national model (DK-QN) (Windolf *et al* 2011), and annual loads of TN from point sources are provided from national databases under the NOVANA programme (Jensen *et al* 2012).

Simultaneously, N depositions were monitored in the form of wet depositions (bulk samplers) and dry depositions estimated from measured ambient air concentrations and dry deposition velocities determined on the basis of meteorological data derived by MM5 model calculations performed within the Danish THOR system (Ellermann *et al* 2010, 2012, Brandt *et al* 2000). Depositions to water systems were estimated as average values from the island based monitoring stations, and assumed representative for marine conditions, while terrestrial depositions are estimated as average values from the land based monitoring stations. In the Danish NOVANA programme, dry deposition of ammonia is furthermore computed using the Danish Ammonia Modelling System (DAMOS: Geels *et al* 2012, Hertel *et al* 2013) applied on a routine basis to 129 selected nature areas around the country and verified by comparisons to results from campaign measurements (Sommer *et al* 2009). The DAMOS model consists of a combination of the long-range transport model Danish Eulerian Hemispheric Model (DEHM: Christensen 1997, Frohn *et al* 2001) and the local-scale plume model OML-DEP (Hertel *et al* 2006, Sommer *et al* 2009). We did not take account of bi-directional ammonia fluxes in the calculations, because deposition of local ammonia represents less than a third of the nitrogen deposition on land (and less at sea) and including bi-directional fluxes would only change the results a few per cent, but would require much more detailed input data. The present ammonia emission inventory for Denmark is based on information at single farm level and accounts for local agricultural practice that plays a significant role for the temporal variation in emissions (Hertel *et al* 2012). The inventory is based on information from the above national databases (Nielsen *et al* 2011). A standardised ammonia emission for each livestock farm and associated fields is then estimated based on information on animal type/number and type of crops. The final inventory includes: the total annual ammonia emission from identified point sources (farms, storages etc) in Denmark, and from area sources, i.e., field emissions from the growing of crops as well as emissions resulting from the application of manure and mineral fertilizer. The area sources are distributed in a 100 × 100 m grid covering all of Denmark (Plejdrup and Gyldenkaerne 2011), and the temporal variation in emission is described by 15 additive functions reflecting different agricultural activities and a 16th function describing the contribution from traffic (Skjøth *et al* 2004). Examples of emission distributions are presented in Geels *et al* (2012).

Data on nitrate concentrations in groundwater, from certified professional laboratories, was queried from the national Danish geological and hydrological database JUPITER. A subgroup of 194 points in oxic groundwater, where the groundwater recharge age has moreover been determined using the chlorofluorocarbon (CFC) method, allows the comparison of long-term changes in N surplus in agriculture with changes in nitrate content of oxic groundwater (figure 1). In addition, nitrate trend analyses in 152 individual monitoring points were performed (figure 7, left), and 11 518 samples from 3757 oxic monitoring points, sampled from 1967–2011, were used to create a national overview (figure 7, right). The CFC analyses were performed according to the procedure of Laier (2005) and Hinsby *et al* (2008) as described in Hansen *et al* (2011).

The national emissions of nitrous oxide were calculated using the standard IPCC emission factor approach (Nielsen *et al* 2013). These calculations use national inventories of N in fertilizers and manures applied as well as N in various types of manure storages and estimates of ammonia volatilization and nitrate leaching.

Changes in soil organic matter content of agricultural mineral soils was monitored by sampling in 1986, 1997 and 2009 in a Danish nationwide square grid net (7 × 7 km) and analysed for soil organic carbon content (Taghizadeh-Toosi *et al* 2014). Soils were sampled in three layers, 0–25 cm, 25–50 cm and 50–100 cm. The measured changes in soil carbon content were converted to changes in soil N by assuming a carbon to N ratio of 10 for the labile soil organic matter (Thomsen *et al* 2008).

The costs of the measures implemented in previous action plans are based on ex-post and ex-ante analyses made from 2003 until today looking at the cost-efficiency of the measures both before and after implementation (Jacobsen 2004, 2012b, Jacobsen *et al* 2004, Børgesen *et al* 2009).

2.2. A typology for nitrogen management policies

To analyse the trend in N management policies over years, the major measures implemented in Denmark 1985–2014 are listed and classified according to the following matrix (table 1).

The first category, command and control (C&C), is the classic regulation type, where a certain action or pollution practice is forbidden by law, controlled by the authorities, and fined if the law is violated. In contrast, the second category marked-based regulation (MBR) includes all types of MBR and governmental expenditure that directly affect the market and thereby the economic optimum for production and hence pollution. This category covers both: (i) market-based instruments where the management and pollution behaviour are regulated via market incentives, typically via a green tax (for example N-taxation) under the polluter pays principle (Carter 2007), (ii) other types of MBR (for example N quotas combined with manure trading possibilities), and (iii) governmental expenditure, that in the form of subsidies affects the market in a similar way to taxes, but by encouragement rather than inhibition. Governmental expenditure is not necessarily under the polluter pays principle (for example, most of the EU agri-environmental policy measures are under this category, as they are designed to promote environmentally friendly production practices, but financed by the EU member states budgets and not via a specific farm tax; Buller *et al* 2000). Finally, the remaining types of policy measures are classified as information and voluntary action (IVA). This includes knowledge production and communication of information about more sustainable N-management practices and technologies via research and extension services (which may be subsidized), and actions by ‘individuals or organizations doing things to protect the environment that are neither required by law nor encouraged by financial incentive’, and which ‘the government can encourage through a range of communicative strategies’ (Carter 2007).

The typology further distinguishes between general versus geographically targeted regulation, which is especially relevant for N management policies, because both the risk of N losses, the pressure of N load to the environment, and the sensitivity of the environment depend strongly on local geology, soil, climate and recipient ecosystems (Blicher-Mathiesen *et al* 2014). ‘General regulation’ measures are implemented equally in all parts of Denmark, whereas ‘geographically targeted regulation’ measures are implemented differently (i.e. localized) depending on specific geographical conditions (for instance, with different N regulations for different watersheds, and different regulations depending on the sensitivity of the receiving environment to N losses, see Jacobsen *et al* 2007).

Finally, the regulation type can be either N input based (for example, regulation of fertilizer inputs) or N output based (for example, maximum ammonia emissions to a defined habitat area).

Table 1. Typology for the classification of N policy measures. Each of the combinations could be either N input based (for example regulation of mineral fertilizer inputs) or N output based (for example maximum ammonia emissions to a defined habitat area).

	General regulation		Geographically targeted regulation	
	Input based	Output based	Input based	Output based
Command and control (C&C)	X	X	X	X
Market-based regulation and governmental expenditure (MBR)	X	X	X	X
Information and voluntary action (IVA)	X	X	X	X

2.3. Nitrogen surplus and efficiency

In line with OECD (2000), the main indicators used in the present paper comprise N surplus and N use efficiency (NUE, or just N-efficiency). As mentioned above, system boundary comprises the entire primary production of the Danish agricultural sector, including cropland, livestock and grazing lands, and the N surplus and NUE. The agricultural N surplus is defined as N input minus N output in agricultural products from the agricultural sector (equation (1)), while N efficiency is defined as N output in agricultural products per unit N input (equation (2))

$$\begin{aligned} \text{N surplus} &= \text{N input} - \text{N output} \\ &\cong \text{N leaching} + \text{NH}_3\,\text{emission} \\ &\quad + \text{denitrification} - \text{soil N change}, \end{aligned} \tag{1}$$

$$\text{N}-\text{efficiency} = (\text{N output})/(\text{N input}). \tag{2}$$

Annual values for N inputs and N outputs are derived from national agricultural statistics (1968, 1969 and 1961–2012) according to Vinther and Olsen (2013) and Kyllingsbæk (2000). N inputs include N in commercial fertilizers and urban and industrial waste materials spread to the fields, N in imported concentrate feedstuffs like soybean cake, meat and bone meal (banned from year 2000), fodder urea, fish products, etc, and N derived from the atmosphere. The latter includes estimated values for net N deposition and biological N fixation via legumes and free-living microorganisms. N outputs include N in: (i) animal products, embracing eggs, milk, meat, live animals or livestock received by offal destruction plants, and (ii) plant products in the form of cereals for food products, straw for energy purposes etc, seeds for manufacturing and sowing, beets for sugar production, potatoes, and other fruits and vegetable products.

N surplus can be used as a proxy for N losses from farming, assuming no change in the soil N pool, and covers a number of N loss components (see equation (1)). In general, the largest N loss component is the leaching of nitrates (Dalgaard *et al* 2011a). N leaching is of special importance in relation to ground and surface water pollution. Other N loss components are gaseous N (ammonia, di-nitrogen, nitrous oxides etc) and particulate N (mainly organic matter). Some of the surpluses may temporarily accumulate in farm product stocks, as biomass or in soil organic matter, but over time and as the soil system approaches steady state, N surplus and N loss will converge, although this may take many decades or even centuries. It must be noted that in the present paper N surpluses and N efficiencies are calculated for the whole Danish agricultural system and cannot be directly compared with results calculated at other scales.

3. Results

3.1. Trends in nitrogen policies and management

The major N policy measures implemented in Denmark are presented in table 2. C&C regulation was implemented first, followed by a mix with MBRs including increased governmental expenditures along with the more IVA based measures.

From the beginning, all measures were implemented as ‘general regulation’, i.e., with equal norms and standards for all parts of the country. The first exception to this was in 1990 with the implementation of environmentally sensitive areas (ESAs) which was designated with the major aim to reduce N pollution (MAFF 2004), and with EU co-financed subsidies to reduce N-fertilization of grasslands (Buller *et al* 2000). Originally, regional authorities were responsible for the designation of these areas. In this way they were geographically targeted (but with the same fixed subsidy classes for the whole country, and with subvention opportunities for farmers living inside the designations), and for farmers it was voluntary whether they signed up to the scheme or not. Further geographical targeting was implemented in 1998 with the AP-II agri-environmental subsidy schemes for (voluntary) construction of wetlands and afforestation in designated areas. Another initiative in 1998 was the initiation of the detailed Danish groundwater mapping programme on approximately 40% of the land surface aiming at establishing site-specific groundwater protections zones based on assessments of the nitrate vulnerability of the aquifers (Thomsen *et al* 2004, Hansen and Thorling 2008).

Until the AP-III in 2004 all regulation was carried out on the input side. The first exception to this was the designation of buffer zones set around ammonia sensitive habitats, followed by special restrictions for emissions from new livestock buildings to designated nature areas. Even though the 1991 Nitrates Directive aimed to reduce the ‘end of pipe’ NO_3^- concentration in leached water to below 50 mg L^{-1}, the regulation was implemented on the input side in the form of

Table 2. The major N policy measures, implemented over the past 30 years with the Danish action plans (AP) in 1985 (NPo), 1987 (AP-I), 1991 (AP for a more sustainable agriculture), 1998 (AP-II), 2001 (ammonia AP), 2004 (AP-III) and 2009 (Green Growth AP), classified into the table 1 policy typology. Localized measures represent the geographically targeted types of table 1, and brackets indicate that the measures fit only partly to the typology class. Until more recently, nearly all regulations focused on the N input side, but from 2004 and onwards output-based regulation also appears.

Year	N measures imposed:	C&C: command and control	MBR: market-based regulation and governmental expenditure	IVA: information and voluntary action
1985	Max. stock density.	X	—	—
	Mandatory slurry tank floating barriers.	X	—	—
	No runoff from silage clamps and manure heaps.	X	—	—
	Min. slurry capacity and ban on winter spreading of slurry for spring crops (including subsidies to invest in slurry tanks etc).	X	(X)	—
1987	Mandatory fertilizer and crop rotation plans.	X	—	—
	Min. proportion of area with winter crops.	X	—	—
	Mandatory manure application within 12 h.	X	—	—
1991	Statutory norms for manure N utilization.	X	—	—
	Max N applied to crops equalling economic optimum.	X	(X)	—
	Subsidies to low-N grasslands in environmentally sensitive areas.	—	(X) (localized)	(X)
1998	Max. N applied 10% below economic optimum.	X	(X)	—
	6% obligatory catch crops.	X	—	—
	Subsidies to more organic farming, wetlands, extensification and afforestation.	—	X (localized)	(X)
2001	Promotion of low excretion livestock feeding.	—	—	X
2004	More catch crops.	X	—	—
	Tightened ammonia restriction (e.g. broadcasting banned), and special restrictions near sensitive nature areas.	X (localized) (output-based)	—	X
	Subsidies to promote better manure handling and animal housing (BAT).	—	X	X
2009	Buffer zones around streams, lakes and NH_4 sensitive habitats.	localized (output-based)	—	—
	Tax on mineral P in feed.	—	X	—
	Max. N applied ≈15% below economic optimum.	X	(X)	—
	Optimized feed practice promotion.	—	—	X

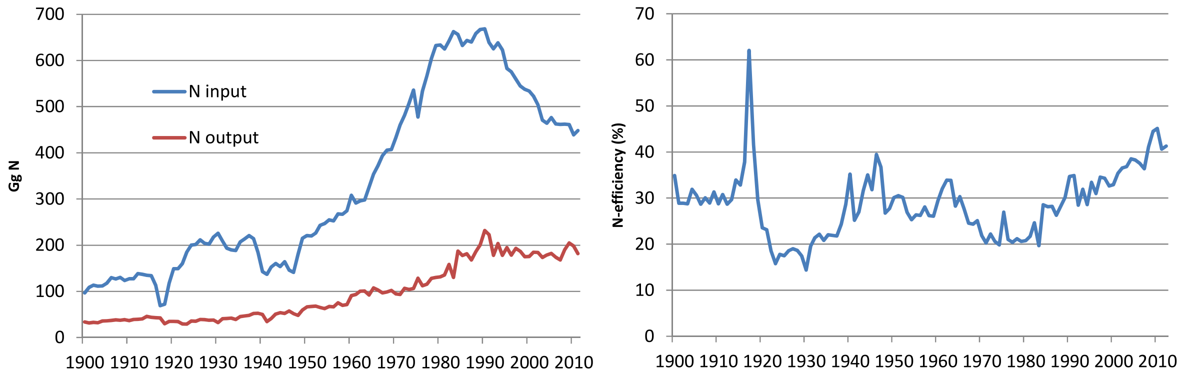


Figure 2. (a) Total sum of N inputs to and sum of N output in products from Danish agriculture, and (b) overall nitrogen use efficiency (N-efficiency) for Danish agriculture over the period 1900–2012. For definitions, see equations (1) and (2).

restrictions on the livestock density and manure applied to the fields (van Grinsven *et al* 2012). It was found that the direct N-effect of reduced livestock density was low, but it has helped to avoid very high livestock intensity resulting in national or regional nutrient surplus, as found in The Netherlands, Belgium (Flanders) and parts of France (Brittany). The key measures in Action Plan II were a higher required fertilizer equivalency of animal manure N, and reduced statutory fertilization N-norms (Jacobsen 2004).

In the future, and in particular with the further implementation of the EU WFD, this regime is expected to change dramatically, requiring both new types of regulation supported by considerable research and development to make the measures more geographically specific (The Commission on Nature and Agriculture 2013).

3.2. Trends in nitrogen surplus and efficiency

In this study, we analyse the overall N use efficiency of the entire agricultural sector *per se*, and have not intended to analyse NUE of agricultural subsystems, like crop or livestock production, separately. Overall agricultural N balances will be a key determinant for environmental impacts, and in that sense, temporal trends and dynamics in N surplus and NUE for the agricultural sector will be better linked to the environmental N indicators, like aquatic environment N levels, which we compare against in the paper. The drawback is, that the NUE becomes dependent on the specific Danish ratio of livestock to crop production, which makes it difficult to compare against other countries with a different ratio, but has the advantage that national registry data can be utilized to a much larger extent for the inventory.

When the development in total agricultural N inputs and N outputs in products is calculated (figure 2) it is clear that especially the input of N to the agricultural sector has been reduced since the implementation of the action plans (from 662 Gg N when it peaked in 1983 to 448 Gg N in 2012). Seen over the period from 1990 to 2010, the N input was reduced by 34%, mainly through reduction in the application of synthetic fertilizer N, while the N output in products was only reduced by 15% in the same period and, consequently, the total N surplus was reduced by 45%, from 437 Gg to 241 Gg.

This means that over the past 25 years the average N surplus has been reduced from approximately 170 kg N ha^{-1} yr^{-1} to below 100 kg N ha^{-1} yr^{-1} for agricultural, while the overall NUE for the agricultural sector has increased from around 20–30% to 43% (figure 2). However, it is worth noting that this increase in NUE appeared after a period where the NUE was reduced from 30% in 1950 to below 20% in 1983—its lowest ever level. However, it is fair to say that the present NUE of more than 40% is high, considering the high level of animal production in Danish agriculture (Denmark is one of the world's largest exporters of animal products, and the export of animal products grew by 39% from 33 kg N ha^{-1} in 1990 to 46 kg N ha^{-1} agricultural land in 2010). It is true that during the two world wars the NUE was even higher (peaking at around 60% during WW1 from 1914 to 1918, and 40% during WW2 from 1939 to 1945), but this represents an unsustainable situation caused by a severe shortage of fertilizer inputs (represented by the drop in N-inputs on figure 2, left), and a similar drop in the production of plant products (Dalgaard and Kyllingsbæk 2003).

N input to crop production in the form of imported fertilizers and atmospheric N from biological N-fixation and deposition has been reduced from the mid 1980s (figure 3), with an important turning point when the import of synthetic fertilizers peaked in 1983 with 143 kg N ha^{-1} and a total N input from fertilizers and the atmosphere of 162 kg N ha^{-1} agricultural land. However, the total N input from fertilizers and atmosphere did not peak until 1989 with 169 kg N ha^{-1} (142 kg N ha^{-1} from fertilizers and 27 kg N ha^{-1} from atmospheric fixation and deposition), but was reduced by 50% over the period 1990–2010. Over the same period the import of feed was sustained (equal figures of 71 kg N ha^{-1} in both 1990 and 2010), whereas the N output in the form of plant products was halved from 48 kg N ha^{-1} to 24 kg N ha^{-1}, and the animal production as mentioned above increased by 39%. It must be noted that there are a large annual variation, especially in the plant production figures, but overall it must be assumed that the decreased output of grain cereals and other plant products was primarily caused by an increased use of domestically harvested plant products for livestock feed, so that these do not appear as product outputs in the inventory. Simultaneously, a smaller decrease in domestically harvested

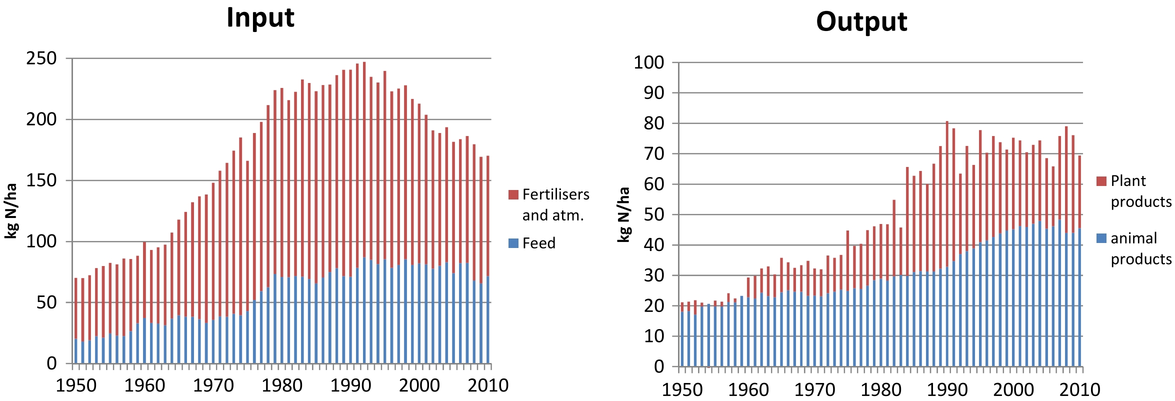


Figure 3. Developments in the form of net N inputs to and net product N outputs from Danish Agriculture, average per hectare agricultural land. Inputs include livestock feed (i.e. imported concentrates), plant fertilizers (imported since no Danish fertilizer production) and atmospheric N inputs (deposited + biologically fixed), and N outputs include animal products (i.e. milk, egg, meat) and cash crop products (sold from the agricultural sector, so not including fodder products utilized for livestock feed).

N was observed by Blicher-Mathiesen *et al* (2013), from 128 kg N ha^{-1} harvested in 1990 to 107 kg N ha^{-1} harvested in 2010, among other things because of a changed cropping pattern, crop use and the overall reduced N-fertilization level.

3.3. Nitrate leaching and emissions to the aquatic environment

The dominant source of TN loadings to coastal waters in Denmark is agriculture, which based on calculations for the period 2007–11 contributed 70% of the TN loadings, followed by background losses (21%), and emissions from point sources (9%) (Nielsen *et al* 2011, Windolf *et al* 2011).

The water runoff from Denmark to coastal waters showed large inter-annual variations during the period 1990–2012, with the driest year being 1996 and the wettest 1994 (figure 4, top). A similar pattern can be seen for the TN loadings to Danish coastal waters showing the lowest and highest values in the same two years (figure 4, bottom). The TN loading from all sources to coastal waters has decreased from an average of ca. 100 Gg N in the period 1990–94 to ca. 59 Gg N in 2012 (figure 4, centre). The export coefficient of TN from the entire Danish terrestrial area thus decreased from 23.3 kg N ha^{-1} to 13.7 kg N ha^{-1} during these two periods (41%). The point source contribution to TN loadings in Denmark has decreased significantly due to improved treatment of especially urban wastewater during the period 1990–2012 (figure 4, mid). The emission from all point sources amounted to an average of 19.6 Gg N during the period 1990–4 and had in 2012 been reduced to 5.7 Gg N, corresponding to a reduction of 71% (Wiberg-Larsen *et al* 2013). The flow weighted concentration of TN shows a decline from an average of 7.1 mg N L^{-1} in 1990–4 to 3.9 mg N L^{-1} in 2012 (46%). Most of the reduction in TN loadings to coastal waters shown in figure 4 is derived from a reduction of TN emissions from diffuse sources, mainly agricultural sources.

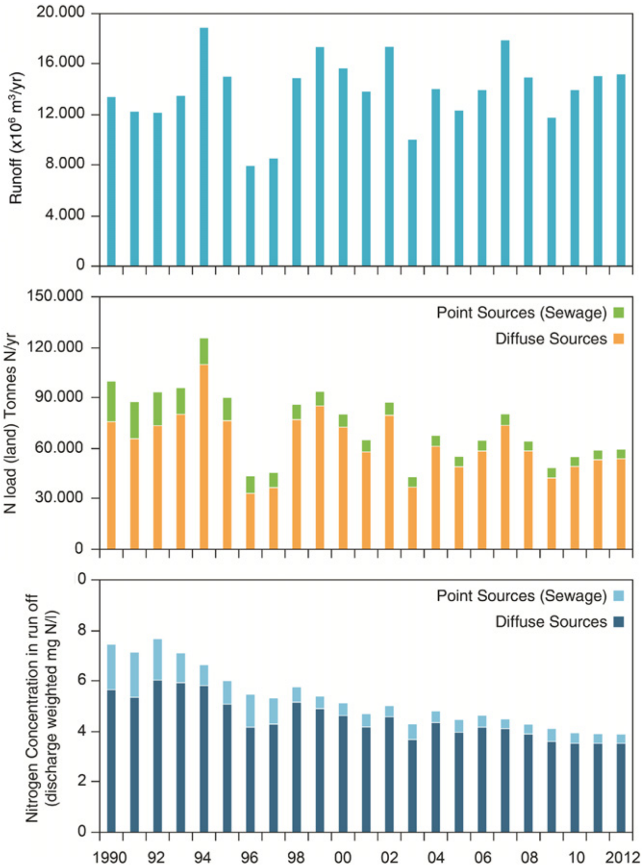


Figure 4. Annual total volume of runoff (top), annual total N loading (centre), and annual flow-weighted total N concentration in Danish surface water outflow to the sea (bottom). (Based on Wiberg-Larsen *et al* 2013).

3.4. Atmospheric ammonia volatilisation and deposition

Deposition of atmospheric N to Danish terrestrial areas varies significantly between different parts of the country (figure 5) but also very locally due to differences in local agricultural production and the type and roughness of the surface.

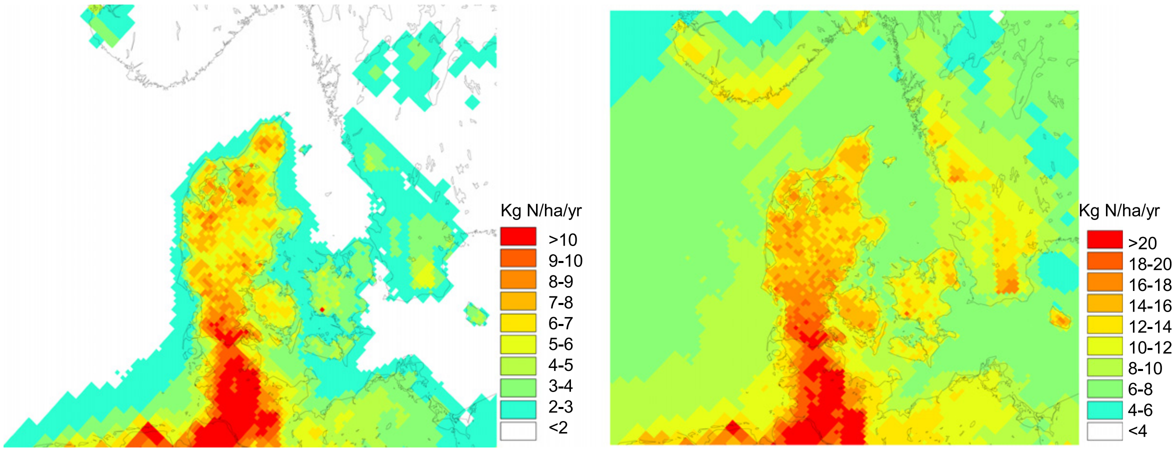


Figure 5. The deposition of atmospheric ammonia (left) and total atmospheric nitrogen (right) computed for 2012 in kg N ha^{-1} yr^{-1}. The displayed data represent the average deposition for the grid cells. Deposition rates vary for different surface types and in the displayed data this is handled by weighting depositions according to the fraction covered by each surface type. Grid cells are 6 km × 6 km, except for the outer part of the domain where the resolution is 17 × 17 km. (Based on results from JH Christensen, Aarhus University, and Ellermann *et al* 2013).

For 2012, the atmospheric deposition of ammonia was found to vary from 0.5 to 15 kg N ha^{-1} yr^{-1} (figure 5, left), and total N deposition from 8 to 23 kg N ha^{-1} yr^{-1} (figure 5, right). Total atmospheric N deposition to Danish terrestrial areas has been calculated at 60 Gg for 2012 (Ellermann *et al* 2013). This gives an average annual deposition of 14 kg N ha^{-1} yr^{-1}, which is an N input above the critical loads for many of the sensitive ecosystems in Denmark (Hertel *et al* 2013). The deposition varies over the country due to a general South–North gradient in concentration contributions from long-range transport from source areas in Central Europe, but also due to local differences in ammonia emissions over Denmark and to differences in precipitation. The largest depositions are thus found in Southwestern Denmark where the livestock production is high and so is precipitation. Uncertainties on the model calculations are estimated to be up to ±40% for the averages in the grid cells. This uncertainty has been derived from comparisons with measurements from the routine monitoring stations in Denmark. Generally an integrated monitoring approach is applied where models and measurements are used in combination (Hertel *et al* 2006).

In comparison, the total annual atmospheric N deposition to Danish marine waters has been calculated at 81 Gg N, which gives an average deposition of 7.7 kg N ha^{-1} yr^{-1} in 2012 (the area of Danish marine waters measures 105 000 km^2) (Ellermann *et al* 2013).

Both measurements and model calculations show a decrease in deposition to Danish land surfaces of about 25% over the time period from 1989 to 2009 (figure 6, top), and a decrease of about 20% in depositions to water surfaces (figure 6, bottom), with an apparent continuous downward trend in the measured depositions over the whole period 1989–2012. The displayed series of DEHM modelled depositions are on a 17 × 17 km resolution (Christensen 1997, Frohn *et al* 2001), and are about 20% overestimated for the land surfaces and about 10% overestimated for the water surfaces. Nevertheless, the modelled depositions reproduce well the development over the years. This indicates that the emission inventories reflect the actual development and that the model responds correctly to the development. Better agreement has been found when DAMOS has been applied

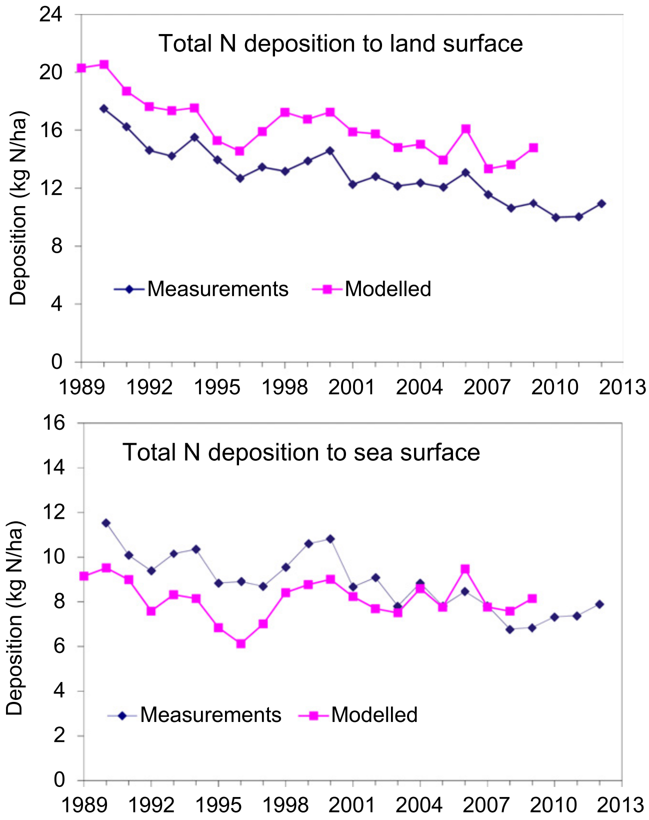


Figure 6. Development in measured and modelled atmospheric N depositions to land surfaces (top) and water surfaces (bottom) during the time period 1989–2012 (modelled only shown up to 2009). 'Measured' deposition is constructed from measured wet deposition (bulk samplers) and dry deposition velocities applied to measured ambient air concentrations of gaseous and particulate nitrogen compounds. Part of this data has previously been depicted in Ellermann *et al* (2010).

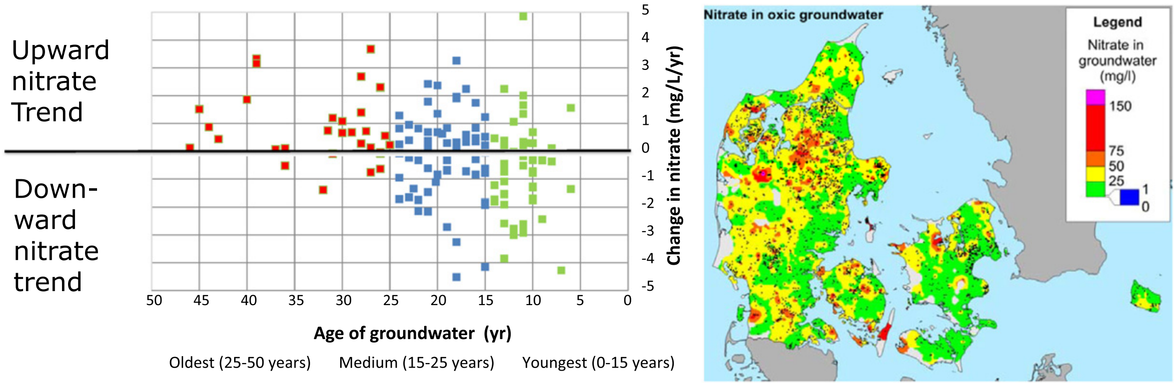


Figure 7. Left: three age groups of upward and downward nitrate trends in oxic groundwater at 152 CFC-dated groundwater measuring points. Right: interpolated map of the nitrate concentration in the Danish oxidized groundwater based on 3757 analyses performed from 1967–2010. Average values from each measuring point are used in the interpolation (based on results from Hansen *et al* 2012).

and the OML-DEP model has calculated depositions in a 400 × 400 m resolution (Sommer *et al* 2009, Geels *et al* 2012, Hertel *et al* 2012), but such simulations cannot be made for the entire 20 yr, national time series.

In total, about 97% of the present Danish NH_3 emissions originate directly from agriculture and only 3% from non-agricultural sources (with 80 of the 97% from manure management, 9% from other soil based activities, and 8% from other agricultural activities). In contrast, the Danish emissions of NO_x primarily originates from other industries, and only indirectly from agriculture (with about 47% from the transport sector, 21% from energy industries, 19% from non-industrial combustion and the remaining 13% from manufacturing industries, construction and other types of activities. The agricultural N policies shown in table 2 have primarily affected the ammonia-based emissions (and depositions), and thus only a minor part of the total depositions in figure 5. However, in the livestock-intensive country of Denmark these lower emissions from agriculture have contributed significantly to reducing total N depositions; and for local depositions to sensitive nature areas the effects of agricultural N mitigation measures have been particular decisive.

The ammonia emission from agriculture has been reduced from 97 Gg NH_3 in 1990 to around 66 Gg in NH_3 (more than 30%), when looking at the total NH_3 emission as included in the NEC directive reporting (Nielsen *et al* 2013b). This reduction has been possible both as a side effect of the measures to reduce N-losses to the aquatic environment and because of a trend to implement new technologies. The technology change has been widespread and this has also meant that the cost of increasing utillization and lower NH_3 emission has been relative low as the technologies were easy available (Jacobsen 2012c).

3.5. N in groundwater

Nitrate has been found in the oxic part of the groundwater throughout Denmark, with large geographical variations, and a tendency for higher concentrations on the sandy soils in Western Denmark, and in areas with a high livestock density (figure 7, right, Hansen *et al* 2012). Data from the Groundwater Monitoring Programme (191748-9326–2009) show that the nitrate concentration of 25 mg L^{-1} is exceeded at approximately 79% of the oxic monitoring points, and the groundwater and drinking water standard of 50 mg L^{-1} is exceeded in approximately 48%. Since the 1980s the overall national upward trend of the nitrate concentrations in oxic groundwater has been reversed. In addition there is a tendency for the frequency of very high nitrate concentrations in oxic groundwater to decline. Locally, nitrate trend analyses in monitoring wells have shown a complex pattern with both upward and downward nitrate trends depending on the age of the groundwater and local agro-hydro-geochemical conditions (figure 7, left; Hansen *et al* 2012). Therefore site-specific groundwater mapping and protection plans are being carried out in order to further protect drinking water resources from nitrate pollution.

3.6. Nitrous oxide emissions

According to the national inventory of greenhouse gas emissions, N_2O emissions were reduced from 17.1 Gg N in 1990 to 11.2 Gg N in 2011. About half of the estimated emissions originate from soil following application of fertilizers and manure, whereas the other emissions come from manure storages and indirect emissions from volatilized ammonia and leaching of nitrates. The reduction in fertilizer use and in losses of N therefore contributed to the reduction in estimated N_2O emissions. It should be noted that the emission factors applied generally is slightly higher than found in experimental studies in Denmark (Chirinda *et al* 2010, Rees *et al* 2013).

3.7. Changes in soil carbon

The average soil organic carbon stock in 0–100 cm of Danish agricultural mineral soils was 137 Mg C ha^{-1}, which assuming a C : N ratio of 10 corresponds to 13.7 Mg N ha^{-1} (Taghizadeh-Toosi *et al* 2014). The changes in soil organic matter over the period from 1986 to 2009 varied between soil types, most likely linked to different land use and management. Loamy soils dominated by cereal crops lost soil organic

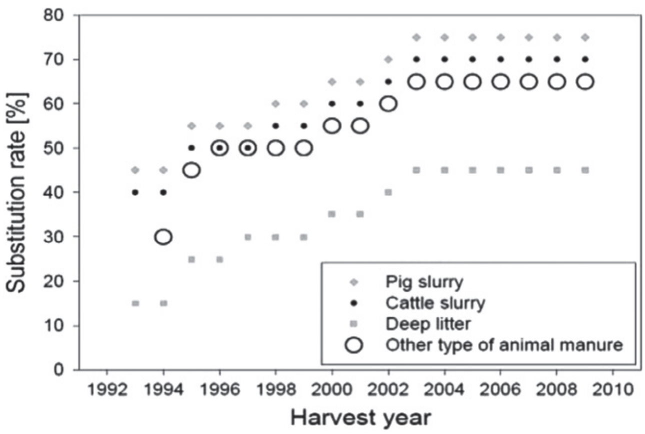


Figure 8. Development of statutory norms for the minimum fertilizer substitution rate for total-N in manure compared with N in synthetic mineral N fertilizers (mainly ammonium-nitrate) in Denmark (Petersen and Sørensen 2008).

matter from the entire soil profile to 100 cm depth corresponding to annual N loss of 124 kg N ha^{-1}, whereas sandy soils gained N at an average rate of 51 kg N ha^{-1}. As an average of all agricultural mineral soils, there was a small and non-significant annual reduction of 20 kg N ha^{-1}.

4. Discussion and conclusions

4.1. The development in N-measures and their effect

Some of the most effective Danish policy measures for N mitigation have focused on a better utilization of N in manures and lower fertilization N-norms (Grant and Waagepetersen 2003, Børgesen *et al* 2009, 2013). This has been driven by the implementation in Denmark of statutory norms for the minimum utilization of N in organic fertilizers (figure 8), combined with strict maximum limits for how much N (plant-available N in organic as well as mineral fertilizers) can be applied. This means that farmers must keep an account of their N-application based on a state-defined norm system, document their use of manure and mineral fertilizers in mandatory fertilizer and crop rotation plans, and adhere to prescribed minimum substitution rates of mineral fertilizers with manure (Petersen and Sørensen 2008, MAFF 2013).

A central question is; why the C&C related measures were implemented first—before the combination of more MBR and IVA types of measures, and why they were so effective? Especially for the manure related measures, a central answer to this seems to be that it was simply economically worthwhile for the farmers to comply with the new restrictions, as the new technology was not too costly (Jacobsen 2004). Already in the late 1970s the benefits of better manure utilization was documented (Højmark and Fogh 1977, Skriver 1978). Therefore the farmers' associations from the very start supported the development, test and implementation of new low-emission technologies for manure management for application of via the extension services (Skriver 1989). It can thus be argued that the C&C measures in the first action plans were effectively combined with farmers' voluntary actions. However, according to table 2 these IVAs were not directly a part of the official measures implemented, but may partly explain the reversing trend of N surplus and nitrate in oxic groundwater before the first action plan was implemented in 1985 (Hansen *et al* 2011). Such IVA schemes should therefore arguably be more widely supported (Petersen *et al* 2014). However, simultaneously it should be mentioned that around 1985–90 the inter-farm variation in manure N use efficiencies was very large (Grant *et al* 1995), and that the C&C measures had a significant effect, especially for farms with a low NUE and only little response to voluntary action. Moreover, in Danish agriculture the tax system and subsidies to restore housing and manure systems are important for the investments in new and more environmental friendly production systems (DLR 1990). This has provided strong incentives for farmers to increase the fertilizer value of manures, and development and implementation has continuously driven forward with the gradually increasing regulatory pressure (table 2).

From 1991 and onwards, an N quota system was implemented consisting of statutory norms for the N fertilization of defined combinations of crops, soil types, climate zones etc, and these norms were not to be exceeded for the farm as a whole. This quota was initially calculated to tally with the economically optimum N fertilization rate based on data from a large number of annual N fertilizer response experiments (Mikkelsen *et al* 2010). However, from 1998, the N quota was as part of a political agreement in parliament reduced below the economic optimum (initially 10% below the optimum, currently around 15% below the optimum crop N fertilization level in terms of production economy). This sub-optimal N application regime has meant that farmers have been very focused on utilizing their available N quota on the crops which gives the highest economic return. The sub-optimal N-quota means that higher utilization of N in e.g. manure has a value which is higher than the price of mineral N. This encourages the use of technologies that lead to a higher utilization than the minimum requirements (figure 8) e.g. through lower ammonia emissions. In this way, the regulations have affected both the production value and the market for manure and for land to receive the manure. The Danish legislation has prevented very high concentrations of livestock by coupling livestock production with requirements for agricultural land. In consequence, the N quota system has not lead to much 'on paper only' re-distribution of manure, in contrast to what has been a problem with for instance the corresponding Dutch regulations. The Netherlands have therefore found it necessary to monitor each truck load of manure with GPS and to install official weighing stations, which is fairly costly (Van der Straeten *et al* 2011), and also in other countries as for instance Italy, France and Scotland (Dalgaard *et al* 2012) high livestock concentration have led to considerable hot spots for N-losses.

In summary, the N-measures have from the mid 1980s until today (2012) helped reduce the N surplus by around 40% if measured per ha, or by more than 50% if measured by kg N (because the agricultural area declined during the

Table 3. Estimated costs of agricultural measures in the different N action plans (APs) and the first version of the River Basin Management Plans (RBMP) of the EU Water Framework Directive to reduce N losses from agriculture (Jacobsen *et al* 2004, Jacobsen 2009, 2012a, 2012b). The administrative costs are not included.

	Ex-ante costs (mill. € yr^{-1})	Ex-post costs (mill. € yr^{-1})
AP-I for the aquatic environment (1987)	84	Not calculated
AP for a more sustainable agriculture (1991)	134	Not calculated
AP-II (1998–2003)	92	70
AP-III (2004–2015)	22	21
Green Growth AP and RBMP 1.0 (2011)	41	48[a]
Total	340	NA

[a] Of the €48 million total costs the agricultural sector pays €18 million and the public sector including EU €30 million annually. The plan has been altered in 2014 and so a re-estimation of the costs has not been carried out yet. The costs for the agricultural sector of RBMP 1.0 are likely to be lower as some measures are dropped (e.g. targeted catch crops).

period). Over the same period, N-leaching from the root zone has also been approximately halved (Grant and Waagepetersen 2003, Dalgaard *et al* 2005, Børgesen *et al* 2009, 2013). Although the N surplus was reduced by more than 50% from 1990 to 2011, the total N loadings to the aquatic environment and the N-deposition from ammonia were reduced by less (roughly 42% and 25%, respectively). This shows the need to understand the relationships between the different N pools and flows, including the denitrification of N, and the buffers of N in biotic N pools (soils and biomass) as well as the retention in ground- and surface water aquifers before it enters the aquatic environment and the atmosphere. Modelling such effects is outside the scope of the present paper but is an integral part of the further work in the DNMARK (2014) research alliance, where time-series for all relevant N flows and stocks will be calculated for the period 1990–2010 (Hutchings *et al* 2014), and scenarios for selected N mitigations options will be modelled and demonstrated (Dalgaard *et al* 2014).

4.2. Socio-economy and public health

According to Jacobsen (2009, 2012a, 2012b) total annual costs of the major Danish N action plans to date are roughly €600 million. About €340 million of these annual costs relate to the agricultural measures from AP-I onwards (table 3). The rest primarily covers costs related to industry and sewage treatment plants outside agriculture.

In general, measures were chosen partly on their N mitigation cost-effectiveness, partly on other benefits which politicians wanted to promote. A significant reason for the success of the Danish policies is that, when designing the policies, efforts have been taken to reduce the costs to farmers. In summary, the most cost-effective measures in AP-II have been the requirements for catch crops (obligatory % of cropped area to be undersown by grass or other species, to reduce N-leaching after main crop) and constructed wetlands, increased utilization-efficiency for N in animal manures, and improved feeding practices (lowering excretion of N in manure). The least cost effective measures have been land set-aside and increased area under grass, as well as the requirement for reduced animal density (Jacobsen 2009). Other benefits besides N mitigation (such as biodiversity and climate protection) are not included in the calculation, and this is the main reason why area-related measures generally have the lowest cost efficiency.

In recent assessments of the costs and benefits of N regulation in agriculture, Andersen *et al* (2013) found that large economic benefits are related to drinking water health impacts, rather than to improvements in surface water quality as such. Nitrate in drinking water has been suspected of negatively affecting human health, for example by causing cancer (Schullehner and Hansen 2014) although no clear evidence has as yet been found (Jensen 1982, De Roos *et al* 2003, Ward *et al* 2005). An assessment of social costs of the health effect (in this case colon cancer) due to nitrate in drinking water in 11 EU member states, estimated that Denmark had the largest percentage of the population (16.2%) exposed to elevated nitrate concentrations (>25 mg L^{-1}) (van Grinsven *et al* 2010). The social costs associated with the loss of healthy life years were at 6.6 euro per capita—more than twice the average of the 11 EU member states assessed. However, a new assessment by Schullehner and Hansen (2014) reveals that only approximately 5.1% of the Danish population was exposed to nitrate concentrations >25 mg L^{-1} in 2012, and further investigations on the actual health effects are needed.

As expected, the N measures related to agriculture have, over time, become gradually more expensive per kg reduced N loss. The current costs are around €3–4 per kg N lost to the environment, and the lower crop yields caused by N fertilization below the economic optimum has over time led to higher costs than estimated in 2004. New solutions to meet the requirements in the EU Habitats Directive, the Marine Strategy Framework Directive and the WFD are therefore called for. The vision for future regulation is to be able to implement measures where the N mitigation benefit is the highest, and allow for increased production elsewhere, i.e. on arable land with higher N retention capacity. The Danish N-policies have so far been national and input based. However, the WFD requires local and output-based approaches with management at the river basin scale, analyses of cost-effectiveness, and measures targeted where the effect is the highest, calling for a new regulation regime. However, more targeted regulation requires more detailed data, there will be more uncertainty in the effects estimated, and some measures might be placed in the wrong place. Therefore, knowledge about the uncertainty related to the estimates is important (Refsgaard *et al* 2014), and the choice of modelling and monitoring framework will be critical for the identification of sensitive (high risk of N loss) and robust (high capacity for N retention) areas in regard to nitrate vulnerability of surface waters. Regardless of whether empirical or mechanistic models are applied for estimating the effect of specific measures, combination with output monitoring (e.g. drainage water monitoring of N concentrations) are likely to produce the most reliable guidance Furthermore, experiences on how to better integrate the different types of regulation in table 2, with a more localized and output based approach, will also be important.

Over the coming years (2014–7) pilot studies of the implementation of new, local RBMPs will be carried out in selected pilot areas in connection to the DNMARK (2014) research alliance. It is expected that these studies can help drive N-regulation towards more output based and localized regulation, facilitate better N-management, and overall improve the cost-effectiveness and mitigation effect. Over the past 30 yrs the table 2 mix of policy measures has helped to increase the overall NUE from 20–30% to around 40–45%. According to Dalgaard *et al* (2011b), it should be possible to continue this development in the coming decades, but this will require a continuous implementation of new technologies and management practices. The pilot areas can be used to demonstrate this and facilitate the locally-adapted actions needed for farmers to comply with policy requirements without incurring excessive costs. In this way we hope to develop a greener economy and develop solution scenarios for the further sustainable management of N.

4.3. Overall conclusion

Losses of N from Danish agriculture have been reduced significantly over the last three decades through regulation that obliges all farmers to consider manure and fertilizer as a valuable resource. However, further reductions are required, especially to comply with the EU WFD. Applications of N to crops are now well below the economic optimum and a further general reduction of N supply to crops would be very expensive. A change of paradigm is therefore planned, with severe restrictions placed on applications to land vulnerable to nitrate leaching to the aquatic environment and a potential easing of restrictions in other areas. The lesson for other countries is that general regulation can be usefully applied to control widespread excessive applications of N but that once this has been achieved, and if further reductions are necessary, a switch to more spatially targeted measures is required.

Acknowledgements

This work was supported by the www.dnmark.org Strategic Research Alliance (DNMARK: Danish Nitrogen Mitigation Assessment: Research and Know-how for a sustainable, low-Nitrogen food production (2013–2017) funded by The Danish Council for Strategic Research (Ref. 12-132421)/Innovationsfonden and the Aarhus University Research Foundation, and the Platform for Research in Biobased Production (BIOBASE) funded by the Danish Centre for Food and Agriculture.

References

Andersen M S, Nielsen H Ø, Pedersen A B, Thodsen H, Pizzol M, Wulff A F, Hansen L B and Senante M M 2013 Macroeconomic perspective on water quality and quantity issues of relevance to the System of Environmental-Economic Accounting for Water (SEEAW) *PART A—Water Quality (Odense River Basin, Denmark) EPI-Water Deliverable no: D 4.5* available at (www.feem-project.net/epiwater/pages/download-public-deliv.html), accessed 1 September 2014

Bjørn C 191748-9326 Det danske landbrugs historie 1810–1914 (The history of Danish agriculture 1810–1914) vol 3 (Odense, Denmark: Landbohistorisk Selskab) p 431

Blicher-Mathiesen G, Andersen H E, Carstensen J, Børgesen C D, Hasler B and Windolf J 2014 Mapping of nitrogen risk areas *Agric. Ecosyst. Environ.* **195** 149–60

Blicher-Mathiesen G, Rasmussen A, Grant R, Jensen P G, Hansen B and Thorling L 2013 *Landovervågningsoplande 2012 (Environmental Land Monitoring Program 2012)* NOVANA. Aarhus Universitet, DCE—Nationalt Center for Miljø og Energi, p 150—Videnskabelig rapport fra DCE nr 74 p 153 available at (http://dce2.au.dk/pub/SR74.pdf), accessed 1 September 2014

Børgesen C D, Nordemann Jensen P, Blicher-Mathiesen G and Schelde K (ed) 2013 Udviklingen i kvælstofudvaskning og næringsstofoverskud fra dansk landbrug for perioden 2007–2011 (Development of Nitrogen leaching and nutrient surplus in Danish agriculture for the period 2007–2011) *DCA Report no* 31 p 153, available at (http://pure.au.dk/portal/files/68362856/dcarapporten31.pdf), accessed 1 September 2014

Børgesen C D, Waagepetersen J, Iversen T M, Grant R, Jacobsen B and Elmholt S 2009 Midt-vejsevaluering af vandmiljøplan III—hoved og baggrundsnotater (mid-term evaluation of aquatic action plan III) *Det Jordbrugsvidenskabelige Fakultet og Danmarks Miljøundersøgelser DJF Report Markbrug* **142** 233 available at(http://pure.agrsci.dk:8080/fbspretrieve/2841678/djfma142.pdf.pdf) accessed 1 September 2014

Brandt J, Christensen J, Frohn L, Berkowicz R and Palmgren F 2000 *The DMU-ATMI THOR Air Pollution Forecast System System Description NERI Technical Report no* 321 Ministry of Environment and Energy National Environmental Research Institute, Roskilde, Denmark available at (www2.dmu.dk/1_viden/2_Publikationer/3_fagrapporter/rapporter/FR321.PDF) accessed 1 September 2014

Buller H, Wilson G and Höll A 2000 *Agri-Environmental Policy in the European Union* (Famham: Ashgate) p 291

Carter N 2007 *The Politics of the Environment. Ideas, Activism, Policies* 2nd edn (Cambridge: Cambridge University Press) p 410

Chirinda N, Carter M S, Albert K R, Ambus P, Olesen J E, Porter J R and Petersen S O 2010 Emissions of nitrous oxide from arable organic and conventional cropping systems on two soil types *Agric. Ecosyst. Environ.* **136** 199–208

Christensen J H 1997 The Danish Eulerian hemispheric model—a three-dimensional air pollution model used for the Arctic *Atmos. Environ.* **31** 4169–91

CLRTAP 1979 *1979 Convention on Long-range Transboundary Air Pollution* (Geneva: United Nations Economic Commission for Europe) p 7 available at (www.unece.org/env/lrtap/), accessed 1 September 2014

Dalgaard T *et al* 2012 Farm nitrogen balances in European Landscapes *Biogeosciences* **9** 5303–21

Dalgaard T *et al* 2014 DNMARK: Danish nitrogen mitigation assessment: research and know-how for a sustainable, low-nitrogen food production *Chapter Submitted to the Book Publication after the N2013 Conference* (Berlin: Springer) p 12

Dalgaard T, Børgesen C D, Hansen J F, Hutchings N J, Jørgensen U and Kyllingsbæk A 2005 How to half N-losses, improve N-efficiencies and maintain yields? The Danish Case *3rd Int. Nitrogen Conf. Contributed Papers* ed Z Zhu, K Minami and G Xing (Monmouth Junction, NJ: Science Press) pp 291–6

Dalgaard T, Guul-Simonsen F, Kjeldsen C and Liboriussen T 2009 Det moderne landbrug (Modern agriculture) ed O Høiris and T Ledet *Modernitetens Verden. Natur, Menneske, Samfund, Kunst og Kultur* (Aarhus, Denmark: Aarhus University Press) pp 423–42

Dalgaard T, Hutchings N, Dragosits U, Olesen J E, Kjeldsen C, Drouet J L and Cellier P 2011a Effects of farm heterogeneity and methods for upscaling on modelled nitrogen losses in agricultural landscapes *Environ. Pollut.* **159** 3183–92

Dalgaard T and Kyllingsbæk A 2003 Developments in the nitrogen surplus and the fossil energy use in Danish agriculture during the 20th century ed J L Usó, B C Patten and C A Brebbia *Advances in Ecological Sciences* vol 18 (Southampton: WIT Press) pp 669–78

Dalgaard T, Olesen J E, Petersen S O, Petersen B M, Jørgensen U, Kristensen T, Hutchings N J, Gyldenkærne S and Hermansen J E 2011b Developments in greenhouse gas emissions and net energy use in Danish agriculture—How to achieve substantial CO_2 reductions? *Environ. Pollut.* **159** 3193–203

Dam P and Jakobsen J G G 2008 *Historisk-Geografisk Atlas* ed J Brandt, R Guttesen, P Korsgaard, B Hermansen, T Dalgaard and A og Reenberg Atlas over Danmark odense: Det Kongelige Danske Geografiske Selskab og Geografforlaget p 179

De Roos A J, Ward M H, Lynch C F and Cantor K P 2003 Nitrate in public water supplies and the risk of colon and rectum cancers *Epidemiology* **14** 640–9

DLR 1990 *Landbrugshåndbog 1990* (Copenhagen: Dansk Landbrugs Realkreditfond) p 216

DNMARK 2014 *Danish Nitrogen Assessment, Strategic Research Alliance Innovative Solutions for a Sustainable Management of Nitrogen in Agriculture* available at (http://dnmark.org/wp-content/uploads/2013/12/DNMARK_UK_tryk.pdf) and (www.dNmark.org) (Aarhus: Aarhus University) p 8

Ellermann T, Andersen H V, Bossi T, Christensen J H, Løfstrøm P, Monies C, Grundahl L and Geels C 2010 *Atmosfærisk Deposition 2009, NOVANA Scientific Report no* 801 from National Environmental Research Institute Aarhus University p 95 available at (www2.dmu.dk/Pub/FR801.pdf), accessed 1 September 2014

Ellermann T, Andersen H V, Bossi R, Christensen J, Løfstrøm P, Monies C, Grundahl L and Geels C 2012 Atmosfærisk deposition 2011 (Atmospheric Deposition 2011) *NOVANA Scientific Report no* 30 from DCE National Center for Environment and Energy Aarhus University p 82 available at (www2.dmu.dk/pub/sr30.pdf), accessed 1 September 2014

Ellermann T, Andersen H V, Bossi R, Christensen J H, Løfstrøm P, Monies C, Grundahl L and Geels C 2013 Atmosfærisk deposition 2012 (atmospheric deposition 2012) *NOVANA Scientific Report no* 73 from DCE National Center for Environment and Energy Aarhus University p 85 available at (http://dce2.au.dk/pub/SR73.pdf), accessed 1 September 2014

FAO 2014 *FAOSTAT, Food and Agriculture Organization of the United Nations Statistics Country Profile for Denmark* (Rome: FAO) available at (www.fao.org/countryprofiles/index/en/?iso3=DNK), accessed 21 September 2014

FRJOR 2014 Research Register for Agriculture (Det Forskningsrelaterede Jordbrugsregister, FRJOR) Aarhus University, Department of Agroecology

Frohn L M, Christensen J H, Brandt J and Hertel O 2001 Development of a high resolution integrated nested model for studying air pollution in Denmark *Phys. Chem. Earth* B—*Hydrol. Oceans Atmos.* **26** 769–74

Geels C *et al* 2012 A coupled model system (DAMOS) improves the accuracy of simulated atmospheric ammonia levels over Denmark *Biogeosciences* **9** 2625–47

Grant R, Blicher Mathiesen G, Andersen H E, Berg P, Jensen P G and Laubel A R 1995 *Landovervågningsoplande 1994 (Environmental Land Monitoring Program 1994) Vandmiljøplanens Overvågningsprogram, Faglig rapport fra DMU. No* 141 p 140 available at (www2.dmu.dk/1_viden/2_Publikationer/3_fagrapporter/rapporter/FR141.pdf), accessed 1 September 2014

Grant R and Waagepetersen J 2003 *Vandmiljøplan II—slutevaluering (aquatic action plan II—final evaluation) Report* National Environmental Research Institute, Silkeborg December 2003 p 32 available at (www.vmp3.dk/Files/Filer/Slutrapporter/VMPII_Slutevaluering_web.pdf), accessed 1 September 2014

Gu B, Zhu Y, Chang J, Peng C, Liu D, Min Y, Luo W, Howarth R W and Ge Y 2011 The role of technology and policy in mitigating regional nitrogen pollution *Environ. Res. Lett.* **6** 014011

Hansen B, Dalgaard T, Thorling L, Sørensen B and Erlandsen M 2012 Regional analysis of groundwater nitrate concentrations and trends in Denmark in regard to agricultural influence *Biogeosciences* **9** 3277–86

Hansen B and Thorling L 2008 Use of geochemistry in groundwater vulnerability mapping in Denmark *Geol. Surv. Denmark Greenland Bull.* **15** 45–8

Hansen B, Thorling L, Dalgaard T and Erlandsen M 2011 Trend reversal of nitrate in Danish groundwater—a reflection of agricultural practices and nitrogen surpluses since 1950 *Environ. Sci. Technol.* **45** 228–34

Hertel O *et al* 2012 Governing processes for reactive nitrogen compounds in the European atmosphere *Biogeosciences* **9** 4921–54

Hertel O, Geels C, Frohn L M, Ellermann T, Skjøth C A, Løfstrøm P, Christensen J H, Andersen H V and Peel R G 2013 Assessing atmospheric nitrogen deposition to natural and semi-natural ecosystems—experience from Danish studies using the DAMOS *Atmos. Environ.* **66** 151–60

Hertel O, Skjøth C A, Løfstrøm P, Geels C, Frohn L M, Ellermann T and Madsen P V 2006 Modelling nitrogen deposition on a local scale—a review of the current state of the art *Environ. Chem.* **3** 317–37

Hinsby K, Purtschert R and Edmunds W M 2008 Groundwater age and quality ed P Quevauviller *Groundwater Science and Policy—An International Overview* (Cambridge: RSC Publishing) pp 217–39

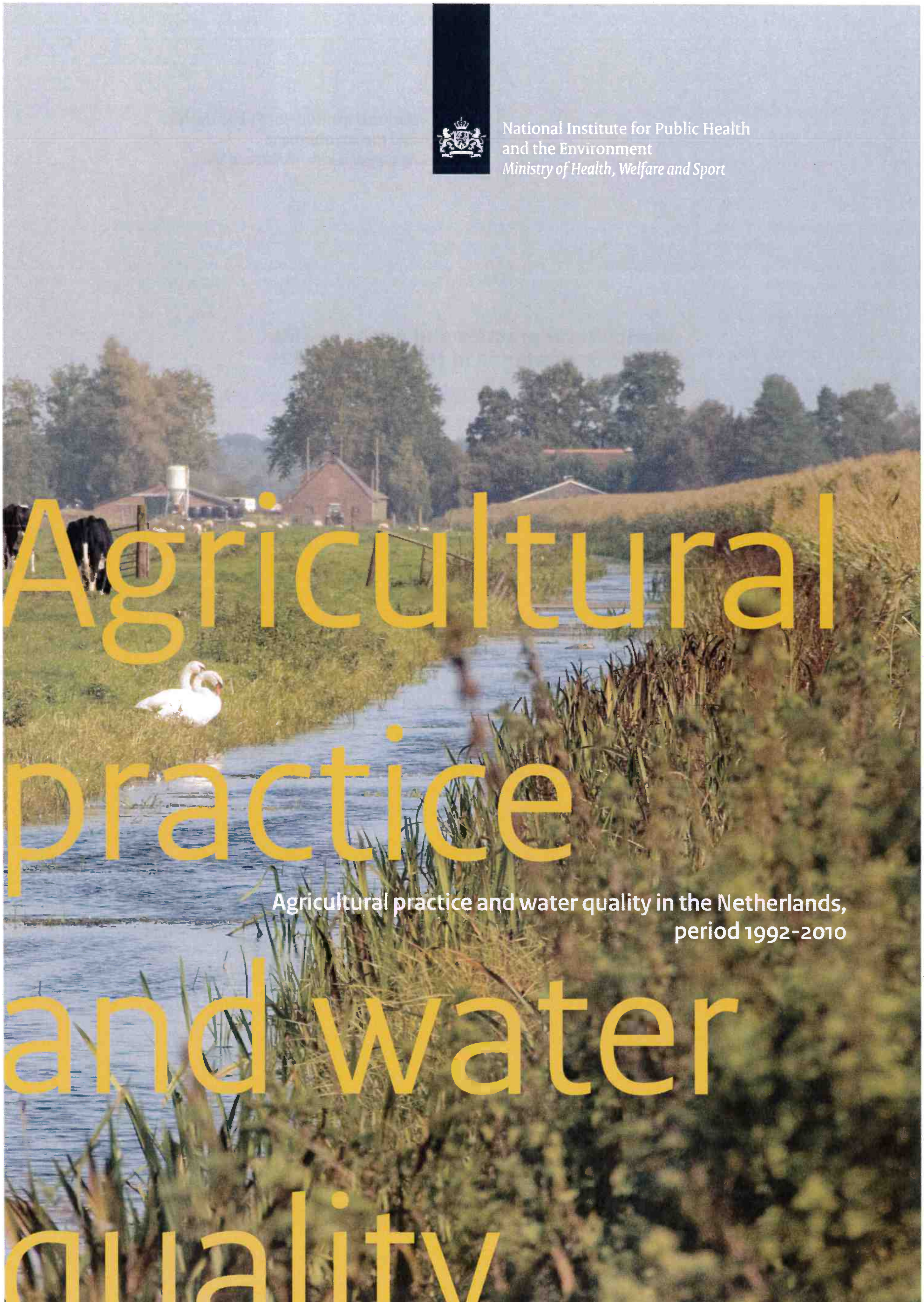
Højmark J V and Fogh H T 1977 Nedvaskning af kvælstof og eftergødskning af byg på sandjord 1977 *Statens Planteavlsforsøg, Medd.* **1382** 2

Hutchings N J, Nielsen O K, Dalgaard T, Mikkelsen M H, Børgesen C D, Thomsen M, Ellermann T, Højberg A L, Mogensen L and Winther M 2 014 A nitrogen budget for Denmark; developments between 1990 and 2010, and prospects for the future *Environ. Res. Lett.* (submitted to the present focus issue)

Jacobs M 2013 Green growth: economic theory and political discourse *Handbook of Global Climate and Environmental Policy* ed R Falkner (Chichester: Wiley Blackwell) p 546

Jacobsen B H 2004 Økonomisk slutevaluering of vandmiljøplan II (final economic evaluation of the Action Plan for the Aquatic Environment II) Report no. 169 (Copenhagen: Danish Institute of Food Economics)
Jacobsen B H 2009 Farm economic consequences of the water framework directive in Europe *J. Farm Manage.* **13** 687–96
Jacobsen B H 2012a *Analyse af Omkostningerne Ved En Yderligere Reduktion Af N-Tabet Fra Landbruget Med 10.000 tons N. Notat Til N-Udvalget (Analyses of Further Reduction Cost for Reducing N Loss From Danish Agriculture by 10 000 ton)* FOI Udredning nr 2012/26 (Copenhagen: Department of Food and Resource Economics) p 91 available at (http://curis.ku.dk/ws/files/45184163/FOI_Udredning_2012_26.pdf), accessed 1 September 2014
Jacobsen B H 2012b *Analyse Af Landbruget Omkostninger Ved Implementering Af Kvælstofreduktionen I Vandplanerne Fra 2011 (Analysis of Agricultural Costs of Implementing N Reduction in Danish Aquatic Action Plans 2011)* FOI Udredningsnotat 2012/6 (Copenhagen: Department of Food and Resource Economics) p 91 available at (http://curis.ku.dk/ws/files/40739929/FOI_udredning_2012_6.pdf accessed 1 September 2014)
Jacobsen B H 2012c Reducing ammonia emission in Europe—cost, regulation and targets with focus on Denmark *J. Agric. Sci. Technol.* A **2** 285–93
Jacobsen B H, Abildtrup J, Andersen J M, Christensen T, Hasler B, Hussain Z B, Huusom H, Jensen J D, Schou J S and Ørum J E 2004 Omkostninger ved reduktion af landbrugets næringsstoftab til vandmiljøet (Costs of reducing nutrient loss from agriculture to the aquatic environment) *Report no* 167, (Copenhagen: Department of Food and Resource Economics) 299 available at (www.vmp3.dk/Files/Filer/Rap_fra_t_grupper/Ny_rapport_167.pdf), accessed 1 September 2014
Jacobsen T, Refsgaard A, Jacobsen B H and Ørum J E 2007 Integrated modelling of nitrate loads to coastal waters and land rent applied to catchment scale water management *Water Sci. Technol.* **56** 39–47
Jensen O M 1982 Nitrate in drinking water and cancer in Northern Jutland, Denmark, with special reference to stomach cancer *Ecotox. Environ. Saf.* **6** 258–67
Jensen P N *et al* 2012 Vandmiljø og Natur 2011 NOVANA Tilstand og udvikling—faglig sammenfatning (Aquatic environment and nature: 2011 Status and development—summary) *DCE Report no* 36 Danish Center for Environment and Energy, Aarhus University, Denmark available at (www2.dmu.dk/pub/sr36.pdf), accessed 1 September 2014
Kjaergaard T 1994 *The Danish Revolution,* 1500-1800 *An Ecohistorical Interpretation* (Cambridge: Cambridge University Press) p 314
Kronvang B, Andersen HE, Børgesen CD, Dalgaard T, Larsen SE, Bøgestrand J and Blicher-Mathiasen G 2008 Effects of policy measures implemented in Denmark on nitrogen pollution of the aquatic environment *Environ. Sci. Policy* **11** 144–52
Kyllingsbæk 2000 Kvælstofbalancer og kvælstofoverskud i dansk landbrug 1979–1999 (Nitrogen balances and surplus in Danish agriculture 1979–1999) *DJF Markbrug Report nr* 36 Danish Institute of Agricultural Sciences, Foulum, Denmark available at (http://pure.agrsci.dk:8080/fbspretrieve/458542/djfm36.pdf), accessed 1 September 2014
Laier T 2005 Nitrate monitoring and CFC-age dating of shallow groundwaters—an attempt to check the effect of restricted use of fertilizers *Nitrates in Groundwater, International Associations of hydrogeologists selected papers* ed L Razowska-Jaworek and A Sadurski (London: A. A. Balkema Publishers) chapter 24
MAFF 2004 *Vejledning Om Særligt Følsomme Landbrugsområder (SFL-områder)—Udpegning Samt Registrering i MVJ-TAS (Guide on environmentally vulnerable agricultural areas)* (Copenhagen, Denmark: Ministry of Food, Agriculture and Fisheries)
MAFF 2013 *Vejledning Om gødnings- Og Harmoniregler (Guide on F and Harmonisation Regulations)* (Copenhagen, Denmark: The Danish Agrifish Agency, Ministry of Food, Agriculture and Fisheries) p 150
Mikkelsen S, Iversen T M, Jacobsen B H and Kjær S S 2010 Danmark—Europe Reducing nutrient losses from intensive livestock operations ed P Gerber, H Mooney and J Dijkman *Livestock in a Changing Landscape vol 2: Experiences and Regional Perspectives* Chapter 8 pp 140–53 (Washington, DC: Island Press) p 208
Nielsen O K *et al* 2011 Annual Danish informative inventory report to UNECE emission inventories from the base year of the protocols to year 2009 *Scientific Report no* 821 from NERI, Roskilde, Denmark: Aarhus University p 601 available at (www2.dmu.dk/pub/fr821.pdf), accessed 1 September 2014
Nielsen O K *et al* 2013 *Denmark's National Inventory Report 2013 Emission Inventories 1990–2011—Submitted under the United Nations Framework Convention on Climate Change and the Kyoto Protocol* Aarhus University, DCE—Danish Centre for Environment and Energy p 1202 Scientific Report no 56 from DCE—Danish Centre for Environment and Energy available at (www2.dmu.dk/pub/sr56.pdf), accessed 1 September 2014
Nielsen O K, Plejdrup M, Hjelgaard K, Nielsen M, Winther M, Mikkelsen M H, Albrektsen R, Fauser P, Hoffmann L and Gyldenkærne S 2013b Projection of SO_2, NO_x, NMVOC, NH_3 and particle emissions—2012–2035 Aarhus University, DCE—Danish Centre for Environment and Energy, 151 pp *Technical Report from DCE—Danish Centre for Environment and Energy No 81* Available at (www.dce2.au.dk/pub/SR81.pdf), accessed September 2014
Odgaard B V and Rømer J R (ed) 2009 *Danske Landbrugslandskaber Gennem 2000 år. Fra Digevoldinger Til Støtteordninger* pp 253–81 (Aarhus: Aarhus University Press) p 301
OECD 2000 *Environmental Indicators for Agriculture: Methods and Results, Executive Summary* (OECD) p 22
OECD 2011 *A Green Growth Strategy for Food and Agriculture Preliminary Report* OECD p 82 available at (www.oecd.org/greengrowth/sustainable-agriculture/48224529.pdf), accessed 1 September 2014
OSPAR 1992 *Convention for the Protection of the Marine Environment of the North–East Atlantic* (London: OSPAR Secretariat) available at (www.ospar.org) p 33
Parajuli R, Dalgaard T, Knudsen M T, Birkved M, Hermansen J E, Jørgensen U, Adamsen A P, Gylling M and Schjørring J K 2014 Biorefining in the prevailing energy and material crisis: a review on sustainable pathways for biorefinery value chains and sustainability assessment methodologies *Renew. Sust. Energy Rev.* (in press)
Petersen C A, Hansen B, Thorling L and Dalgaard T 2014 Hvorfor stoppede stigningen i nitrat i det nydannede grundvand før vandmiljøplanerne? *Vand og Jord* **4** (in press)
Petersen J and Sørensen P 2008 Fertilizer value of nitrogen in animal manures—basis for determination of a legal substitution rate *Report DJF Markbrug 138*
Plejdrup MS and Gyldenkaerne S 2011 Spatial distribution of emissions to air—the SPREAD model *NERITechnical Report no* 823, National Environmental Research Institute, Denmark: Aarhus University p 72 available at (www2.dmu.dk/Pub/FR823.pdf), accessed 1 September 2014
Rees R M *et al* 2013 Nitrous oxide emissions from European agriculture; an analysis of variability and drivers of emissions from field experiments *Biogeosciences* **10** 2671–82
Refsgaard J C *et al* 2014 Nitrate reduction in geologically heterogeneous catchments—a framework for assessing the

scale of predictive capability of hydrological models *Sci. Total Environ.* **468–469** 1278–88
Schullehner J and Hansen B 2014 Nitrate exposure from drinking water in Denmark over the last 35 years *Environ. Res. Lett.* **9** 095001
Skjøth C A, Hertel O, Gyldenkærne S and Ellermann T 2004 Implementing a dynamical ammonia emission parameterization in the large-scale air pollution model ACDEP *J. Geophys. Res.—Atmos.* **109** D06306
Skriver K 1978 Gødskning og kalkning (Fertilisatio and liming) *Oversigt over forsøg og undersøgelser i Landbo- og Husmandsforeningerne* ed J Olesen (Aarhus: Landskontoret for Planteavl) pp 100–24
Skriver K 1989 Aktion for bedre udnyttelse af husdyrgødning gennem en forstærket og koordineret rådgivningsindsats. Brev per 16/1 1989, KS/LL/4251P (Aarhus: Landskontoret for Planteavl)N. 3
Sommer S G, Østergård H S, Løfstrøm P, Andersen H V and Jensen L S 2009 Validation of model calculation of ammonia deposition in the neighbourhood of a poultry farm using measured NH_3 concentrations and N deposition *Atmos. Environ.* **43** 915–20
Statistics Denmark 1968 *Agricutural Statistics 1900–1965 vol I, Agricultural Area and Harvest and Utilization of Fertilizers* Statistiske Undersøgelser no 22 (Copenhagen: Statistics Denmark)
Statistics Denmark 1969 *Agricutural Statistics 1900–1965 vol II Livestock and Livestock Products and Consumption of Feeding Stuffs* Statistiske Undersøgelser no 25 (Copenhagen: Statistics Denmark)
Statistics Denmark 1961–2012 *Agricultural Statistics. Statistics on Agriculture, Gardening and Forestry* (Copenhagen: Annual publication publications from Statistics Denmark)
Stern N 2007 *The Economics of Climate Change: The Stern Review* (Cambridge: Cambridge University Press) p 712
Sutton M A, Oenema O and Dalgaard T 2014 Preface: Nitrogen on the table: the influence of food choices on nitrogen emissions and the European environment *ENA Special Report on Nitrogen and Food The Food Climate Research Network* ed H Westhoek, J P Lesschen, T Rood, S Wagner, A Leip, A De Marco, D Murphy-Bokern, M A Sutton and O Oenema (Oxford: University of Oxford)
Sutton M, Howard C M, Erisman J W, Billen G, Bleeker A, Grennfelt P, van Grinsven H and Grizetti B (ed) 2011 *The European Nitrogen Assessment Sources, Effects and Policy Perpectives* (Cambridge: Cambridge University Press) p 664 available at (www.nine-esf.org/ENA-Book), accessed 1 September 2014
Taghizadeh-Toosi A, Olesen J E, Kristensen K, Elsgaard L, Østergaard H S, Lægdsmand M, Greve M H and Christensen B T 2014 Changes in carbon stocks of Danish agricultural mineral soils during 1986–2009: effects of management *Eur. J. Soil Sci.* in press doi:10.1111/ejss.12169
The Commission on Nature and Agriculture 2013 Richer nature, new environmental regulation and new growth opportunities for agriculture Executive summary *The Danish Commission on Nature and Agriculture, Copenhagen* available at (www.naturoglandbrug.dk)
The Council of the European Communities 1991 Council Directive of 12 December 1991 Concerning the protection of waters against pollution caused by nitrates from agricultural sources (91 /676/EEC) *Off. J. Eur. Comm.* No L 375/1
The European Parliament and the Council of the European Union 2000 Directive 2000/60/EC of the European Parliament and of the Council of 23 October 2000 establishing a framework for Community action in the field of water policy *Off. J. Eur. Comm.* L 327/1
The European Parliament and the Council 2008 Directive 2008/56/EC of the European Parliament and the council establishing a framework for community action on the field of marine environmental policy (Marine Strategy Framework Directive) *Off. J. Eur. Comm.* L 164/19
The Helsinki Commission 2008 *Convention on the Protection of the Marine Environment of the Baltic Sea Area, 1992* Markets of concentration permits: the case of manure policy (Helsinki, Finland: HELCOM Secretariat) p 43
Thomsen R, Søndergaard V H and Sørensen K I 2004 Hydrogeological mapping as a basis for establishing site-specific groundwater protection zones in Denmark *Hydrogeology J.* **12** 550–62
Thomsen I K, Petersen B M, Bruun S, Jensen L S and Christensen B T 2008 Estimating soil C loss potentials from the C to N ratio *Soil Biol. Biochem.* **40** 849–52
Van der Straeten B, Buysse J, Nolte S, Lauwers L, Claeys D and Huylenbroeck G van 2011 Markets of concentration permits: the case of manure policy *Ecol. Econ.* **70** 2098–104
van Grinsven H J M *et al* 2012 Management, regulation and environmental impacts of nitrogen fertilization in northwestern Europe under the Nitrates Directive; a benchmark study *Biogeosciences* **9** 5143–60
van Grinsven H J M, Rabl A and de Kok TM 2010 Estimation of incidence and social cost of colon cancer due to nitrate in drinking water in the EU: a tentative cost-benefit assessment *Environ. Health* **9** 58
Vinther F P and Olsen P 2013 Næringsstofbalancer og næringsstofoverskud i landbruget 1991/92–2011/12 (Nutrient balances and surplus in Danish agriculture 1991/92–2011/12) *DCA—Report* 06/2013; Foulum: Aarhus University p 25 available at (http://pure.au.dk/portal/files/56329476/NPK_1991_92_2011_netfil.pdf), accessed 1 September 2014
Ward M J, de Kok T M Levallois P, Brender J, Gulis G, Nolan B T and VanDerslice J 2005 Workgroup report: drinking-water nitrate and health—recent findings and research needs *Environ. Health Perspect.* **113** 1607–14
Wiberg-Larsen P *et al* 2013 Vandløb 2012 NOVANA Aarhus Universitet, DCE—Nationalt Center for Miljø og Energi, 84 s —Videnskabelig rapport fra DCE—Nationalt Center for Miljø og Energi nr 75 (http://dce2.au.dk/pub/SR75.pdf), accessed 8 September 2014
Windolf J, Thodsen H, Troldborg L, Larsen S E, Bøgestrand J, Ovesen N B and Kronvang B 2011 A distributed modelling system for simulation of monthly runoff and nitrogen sources, loads and sinks for ungauged catchments in Denmark *J. Environ. Monit.* **13** 2645–58



National Institute for Public Health
and the Environment
Ministry of Health, Welfare and Sport

Agricultural practice and water quality

Agricultural practice and water quality in the Netherlands, period 1992-2010

National Institute for Public Health
and the Environment
Ministry of Health, Welfare and Sport

Agricultural practice and water quality in the Netherlands in the period 1992-2010

RIVM Report 680716008/2012

Colophon

R.A. Baumann, RIVM
A.E.J. Hooijboer, RIVM
A. Vrijhoef, RIVM
B. Fraters, RIVM
M. Kotte, Waterdienst
C.H.G. Daatselaar, LEI, part of Wageningen University and Research Centre (UR)
C.S.M. Olsthoorn, Statistics Netherlands
J.N. Bosma, Ministry of Economic Affairs, Agriculture and Innovation, Regulatory Service (Dienst Regelingen)

Contact:
Bert Baumann
Centre for Environmental Monitoring (CMM)
bert.baumann@rivm.nl

This investigation has been performed by order and for the account of Ministry of Infrastructure and the Environment and the Ministry of Economic Affairs, Agriculture and Innovation, within the framework of the Manure Policy Advisory Project (project no. M/680716).

Abstract

Agricultural practice and water quality in the Netherlands in the period 1992-2010

The nitrogen surplus in Dutch agriculture decreased by almost 50% between 1992 and 2010. This decrease is the result of measures applied to Dutch agriculture pursuant to the EU Nitrates Directive, such as using less manure during a shorter period of the year. This improvement is evident from an inventory of changes in groundwater and surface water quality and in agricultural practice. The report on the developments is a four-yearly EU obligation. RIVM (National Institute for Public Health and the Environment) carried out this inventory in cooperation with Statistics Netherlands, the Water Department of the Directorate-General for Public Works and Water Management (RWS), the Agricultural Economics Research Institute (LEI, part of Wageningen University and Research Centre, UR) and Dienst Regelingen (Regulatory Service) of the Ministry of Economic Affairs, Agriculture and Innovation.

Decreasing nitrate content
As a result of the implementation of the EU Nitrates Directive, the nitrate concentration in water that leaches from root zones in agricultural land to groundwater and surface water declined steeply between 1992 and 2010. This is especially true of in sandy areas, where the average concentration decreased from 140 mg/l to 60 mg/l. In areas containing clay, the average nitrate concentrations in leaching water also decreased, falling to 29 mg/l. Regarding peat regions, the leaching water there never contains much nitrate (less than 10 mg/l), as nitrate decomposes rapidly in such regions.

Fresh surface water
Since 2002, the average nitrate concentration in fresh surface waters has hovered around the same level (15 mg/l from 2008 to 2010). Despite this, between 2004 and 2010 the chlorophyll-a concentration in the summer (an indicator for eutrophication) in regional fresh surface waters affected by agriculture increased slightly.

Water quality continues improving
The quality of water in the Netherlands is expected to improve further over the next few years. To be precise, it will actually take some years before the measures implemented under the current action programme (2010-2013), such as more stringent application standards for manure, expressed as nitrogen content, lead to better water quality.

Keywords: nitrates directive, agricultural practice, groundwater and surface water quality

Rapport in het kort

Landbouwpraktijk en waterkwaliteit in Nederland, periode 1992-2010

Het stikstofoverschot in de Nederlandse landbouw is tussen 1992 en 2010 met bijna 50% afgenomen. Dit is een gevolg van maatregelen die vanwege de Europese Nitraatrichtlijn in de Nederlandse landbouw zijn genomen, zoals minder mest gebruiken gedurende een kortere tijd van het jaar. Dit blijkt uit een inventarisatie van de ontwikkelingen in de grond- en oppervlaktewaterkwaliteit en de landbouwpraktijk. De rapportage hiervan is een vierjaarlijkse Europese verplichting. Het RIVM heeft de inventarisatie uitgevoerd met het Centraal Bureau voor de Statistiek, de Waterdienst, LEI (onderdeel van Wageningen UR) en Dienst Regelingen van het ministerie van Economische Zaken, Landbouw en Innovatie.

Nitraatconcentratie daalt
Dankzij de uitvoering van de Europese Nitraatrichtlijn is ook de nitraatconcentratie in het water dat uitspoelt uit de 'wortelzone' van landbouwpercelen naar het grond- en oppervlaktewater sterk gedaald tussen 1992 en 2010. Vooral in de zandgebieden is dat het geval: in deze gebieden daalde de gemiddelde concentratie van 140 naar 60 milligram per liter. In de gebieden met kleigrond zijn de gemiddelde nitraatconcentraties in het uitspoelende water eveneens gedaald, naar 29 milligram per liter. In veengrond is altijd weinig nitraat in het uitspoelende water aanwezig (minder dan 10 milligram per liter). Dat komt doordat nitraat in veengronden snel afbreekt.

Zoet oppervlaktewater
In zoet oppervlaktewater schommelt de gemiddelde nitraatconcentratie sinds 2002 rond hetzelfde niveau (15 milligram per liter in 2008-2010). Desondanks is tussen 2004 en 2010 de chlorofyl-a-concentratie in de zomerperiode (een indicator voor eutrofiëring) in regionale zoete oppervlaktewateren die door de landbouw worden beïnvloed licht toegenomen.

Waterkwaliteit blijft zich verbeteren
Het is te verwachten dat de waterkwaliteit in Nederland in de komende jaren verder verbetert. Het duurt namelijk enkele jaren voordat de maatregelen uit het huidige actieprogramma (2010-2013), zoals aangescherpte gebruiksnormen voor mest, uitgedrukt in de hoeveelheid stikstof, zich vertalen naar een betere waterkwaliteit.

Trefwoorden: nitraatrichtlijn, landbouwpraktijk, grondwater- en oppervlaktewaterkwaliteit

Preface

This report was prepared by order and for the account of the Ministry of Infrastructure and the Environment (IenM) and the Ministry of Economic Affairs, Agriculture and Innovation (EL&I). Kaj Locher as project supervisor represented the Ministry of IenM, and Martin van Rietschoten represented the Ministry of EL&I. The authors of this report would like to thank both gentlemen for their discerning questions and comments. Our thanks also go to Jaap Willems of the Netherlands Environmental Assessment Agency and Gerard Velthof of Alterra, part of Wageningen UR, who checked the final draft of the report and helped improve the consistency of the conclusions presented here with those presented in other reports being prepared for the Evaluation of the Fertilisers Act 2012.

Bert Baumann, Arno Hooijboer, Astrid Vrijhoef, Dico Fraters, Marcel Kotte,
Co Daatselaar, Kees Olsthoorn, Annet Bosma

Contents

Summary and conclusions

Introduction
This report provides the background information for the Netherlands Member State report that is obligatory under the EU Nitrates Directive. It has to be submitted to the European Commission in mid-2012. The contents of this Member State report conform to the guidelines published in November 2011. The report covers the period 1992-2010, with data for 2011 also presented if available.

The report provides an overview of current agricultural practices, and groundwater and surface water quality in the Netherlands, outlines the trends in these waters and assesses the time scale for change in water quality due to changes in agricultural practice. The implementation and impact of the measures taken as part of the action programmes are described, including a forecast of the future evolution of water body quality.
The data presented in this background document are for the period preceding the First Action Programme (before December 1995) as well as the period of the First (1995-1999), Second (1999-2003), Third (2004-2009), and partly the Fourth Action Programme (2010-2013).

Agricultural policy measures and practice
Policy measures
The system of manure bookkeeping (introduced in 1987) was replaced in 1998 by a system of minerals accounting, known as MINAS. It was based on the mineral balance of nitrogen (N) and phosphorus (P) (farm gate balance). Under this system, limits were set for the permitted levels of the N and P surpluses on farms (MINAS loss standards). These loss standards have gradually been tightened. On 1 January 2002, the Manure Transfer Contracts (MAO) system came into force to ensure compliance with the application limits under the Nitrates Directive. Livestock farmers who produced too much manure were obliged to enter into manure transfer contracts with arable farmers, other less intensive livestock farmers or manure processors. The MAO system was abolished in early 2005. In January 2006, the Netherlands adopted a new manure policy based on application limits instead of loss standards. This manure policy, including application limits for nitrogen in manure and fertilisers as stipulated by the Nitrates Directive, also means a further tightening of the regulations governing the use of nitrogen and phosphorus.

Agriculture in the period 2008-2011
In the period 2008-2011, the area under cultivation in the Netherlands totalled 1.85 million ha, corresponding to 54.7% of the country's land surface. Of this area, 52% comprised grassland (81% permanent), 13% silage maize and 29% other arable crops. The rest (6.4%) was used for horticulture. There were about 72,700 farms, of which 52% represented farms for grazing animals, 17% arable farms, 17% horticultural farms, and 14% factory and mixed farms.

Livestock comprised 3.9 million head of cattle, 12.2 million pigs, 98 million head of poultry, and 1.5 million sheep and goats. The livestock produced manure containing about 487 million kg of nitrogen (N) and 77 million kg of phosphorus (P). Cattle manure was responsible for some 58% of the nitrogen and 51% of the phosphorus. Of the phosphorus in the manure, around 22% was exported or used for non-agricultural purposes; for nitrogen, the amount was about 11%. Nitrogen (N) input to agricultural soils was on average 354 kg/ha, of which

186 kg/ha was via manure, 120 kg/ha via artificial fertiliser and 47 kg/ha via atmospheric deposition and other sources. The nitrogen surplus on the soil surface balance was on average about 145 kg/ha. Phosphorus (P) input to agricultural soils was on average 38 kg/ha, of which 33 kg/ha was via manure, 4 kg/ha via artificial fertiliser, and 2 kg/ha via other sources. The phosphorus surplus on the soil surface balance was 9 kg/ha on average.

Trends in agricultural practice in the period 1992-2011
The land area used for agriculture in the period 1992-2011 shrank by 6.2% and the number of farms by 38%. The number of cattle decreased by 18% and the number of pigs by 16%. By contrast, the number of poultry increased by 4%.

Nitrogen and phosphorus in manure from livestock decreased by 30% and 23% respectively, due to the combined effect of a reduction in the number of livestock and in the amount of mineral excretion per animal. The latter was a consequence of lower nitrogen and phosphorus content in fodder and improved fodder conversion. As a result of this, as well as from a steep decline in the use of artificial fertiliser, the nitrogen and phosphorus surpluses in Dutch agriculture decreased by 48% and 75% respectively (Figure S1).

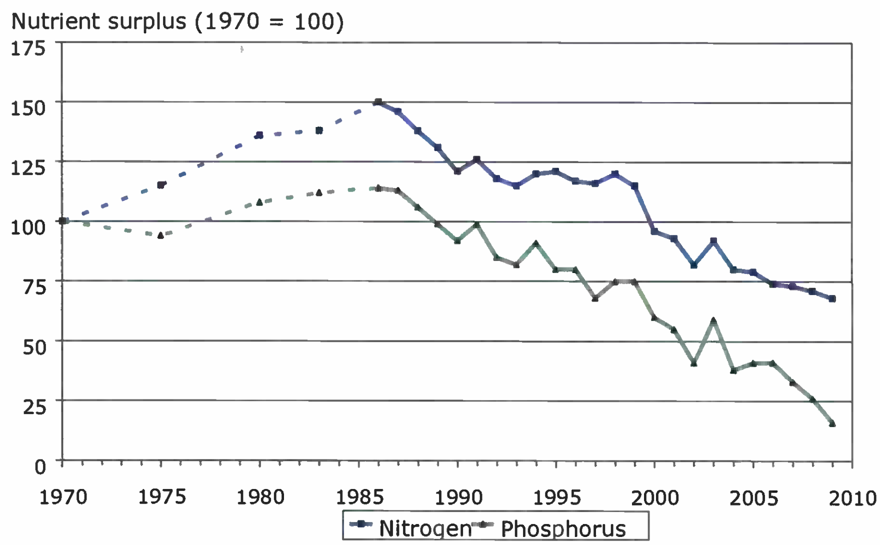


Figure S1. Trends in the nitrogen and phosphorus surpluses in Dutch agriculture in the period 1970-2009, with 1970 values defined as 100.

Compared with the previous reporting period (2004-2007), the net manure transport (difference between input and output) increased significantly in a number of areas. Exports (to other countries) increased threefold.

Ammonia emissions from agricultural sources into the atmosphere continue to decrease, for the latest reporting period (2008-2010) being 55% below the level for the period 1992-1995.

By comparison with the previous reporting period, the storage capacity for manure has expanded substantially. In 2010, 96% of dairy farms, 95% of pig farms and 87% of intensive veal calf farms had storage facilities sufficient for at least six months of manure production.

Quality of groundwater and surface waters

Nitrate concentrations in the period 2008-2010

Since changes in agricultural practice affect water leaching from the root zone (leach water) first, it was decided to monitor the effects of the action programmes in the top metre of the groundwater, in tile drain water or in the moisture of soil layers just beneath the root zone. This report also includes the data from nitrate measurements in deeper groundwater and surface waters.

Nitrate concentration decreases the further it is measured from the source, i.e. agricultural activities (see Table S1). This applies to groundwater as regards depth (of measurement) and to surface waters as regards distance. The nitrate concentration in groundwater decreases with respect to depth, which can clearly be seen in Table S1. In surface waters, the nitrate concentration is lower the further away the nitrogen source is. The overview below shows the different types of surface water according to nitrate concentration, from the highest to the lowest: regional waters strongly affected by agriculture > other regional waters > fresh national waters > coastal waters > open sea.

Two factors influence this falling concentration. The first is the conversion of nitrate into nitrogen molecules (denitrification) during transport; the second is the mixing with water originating from non-agricultural areas (dilution). In the case of groundwater, two additional factors play a role: time and the hydrological conditions. Water leaching from a root zone is young water (1 to 5 years old). In the Sand region, groundwater at a depth of 5 to 15 metres has a travel time of approximately 10 years, and groundwater at a depth of 15 to 30 metres, a travel time of approximately 40 years. Hence, groundwater at a depth of 15 to 30 metres reflects agricultural practice of at least 40 years ago. The groundwater at the above-mentioned depths in Clay and Peat regions is generally even older. Hydrological factors (channels) play a key role here, as the groundwater in aquifers in clay and peat areas to a depth of 5 to 15 metres, as well as 15 to 30 metres, is often wholly or partly confined by a weakly permeable clay aquifer. In such regions, the precipitation surplus drains away through the soil surface to the surface water. Wholly and partly confined aquifers also occur locally in the Sand region.

In the Peat region, nitrate concentrations in leach water and groundwater are lower than in the Clay region, the concentrations here being in their turn lower than in the Sand region (Table S1). This is caused by differences in denitrification rates. Soils in the Sand region have the lowest denitrification capacity, soils in the Clay region come next, and soil in the Peat region have the highest.

Table S1. Average nitrate concentrations measured (in mg/l) and exceedance of the EU standard of 50 mg/l (as a percentage of the number of monitoring wells) in groundwater and surface waters for the period 2008-2010[1].

Water type	Sand	Clay	Peat	Loess	All types
Leaching from root zone (agriculture)	60 (53%)	29 (21%)	7.5 (2%)	78 (66%)	48 (38 %)
Groundwater at a depth of 5-15 metres (agriculture)	32 (19%)	<1 (0%)	<1 (0%)	-	-
Groundwater at a depth of 15-30 metres (agriculture)	8 (4%)	< 1 (0%)	< 1 (0%)		-
Groundwater at a depth exceeding 30 metres (phreatic extraction)	7 (0%)	-	-		-
Fresh surface waters[2]					
Affected by agriculture					15 (3%)
Other regional water					14 (1%)
Marine surface waters[2]					
Coastal water					4 (0%)
Open sea					< 1 (0%)

[1] The percentages between brackets are the relative numbers of exceedances of the EU standard of 50 mg/l in the period 2008-2010. For water leaching from a root zone (under 5 metres in depth), the percentage is the relative number of farms with exceedance of the standard. For groundwater at a depth exceeding 5 metres, the percentage is the relative number of wells, and for surface water, the relative number of monitoring locations.

[2] Average nitrate concentrations in the winter, the season when the leaching has a substantial effect on the quality of the surface water.

Around 65% of the total amount of nitrogen found in the Netherlands' fresh surface waters originates from other countries. The remainder found in the Dutch water system originates from various other sources, with leaching and run-off from agriculture as the main sources of nitrogen in the Netherlands. The remainder originates from various other sources.

Eutrophication state of surface waters in the period 2008-2010

The eutrophication of surface waters can be assessed from the chlorophyll-a concentration, with the total nitrogen and phosphorus concentrations as state indicators. Just as for nitrates, eutrophication indicators have lower concentrations the further the nitrogen and phosphorus sources are (see Table S2). The overview below shows the different types of surface water according to concentration of eutrophication indicators, from the highest to the lowest: regional waters strongly affected by agriculture > other regional waters > fresh national waters > coastal waters > open sea. At 21% of the regional monitoring locations in waters strongly affected by agriculture and at 13% in other regional waters, the chlorophyll-a concentrations exceed 75 µg/l.

Table S2. Eutrophication parameters (chlorophyll-a in µg/l and total nitrogen and phosphorus in mg/l), average summer values[1] for different types of surface water in the period 2008-2010.

Water type	Chlorophyll-a	Total nitrogen	Total phosphorus
Regional waters affected by agriculture	46 (21%)[a]	3.7	0.55
All waters	34 (13%)[a]	3.1	0.25
Coastal water	9 (0%)[a]	0.3[b]	-
Open sea	1 (0%)[a]	< 0.5[b]	-

[1] The average summer values are presented here, as this season is the most significant one for eutrophication.
[a] The percentages in brackets express the proportion of locations with a concentration exceeding 75 µg/l.
[b] Total amount of dissolved inorganic nitrogen.

Trends in the quality of groundwater and surface water

Nitrate concentrations in the period 1992-2010

In the period 1992-2010, the nitrate concentrations in water leaching from root zones of farms (Figure S2) decreased. This was also the case with farms that exceeded the EU standard of 50 mg/l (Figure S3).

Especially in the Sand region, but also in the Clay region, the nitrate concentrations measured in the latest reporting period (2008-2010) are below those of the preceding reporting period (2004-2007). In the Sand regions, the average concentration decreased from 140 mg/l (1992-1995) to 60 mg/l (2008-2010). Regarding nitrate concentrations in peat regions, there has been no change between the previous and the latest reporting periods.

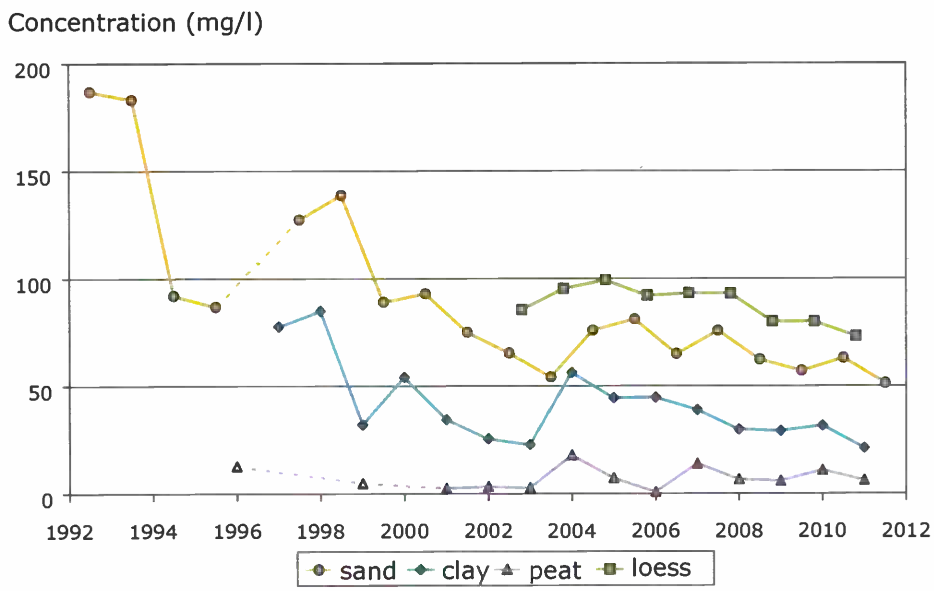


Figure S2. Nitrate concentrations in water leaching from root zones of farms by region in the period 1992-2011. Average annual measured concentrations.

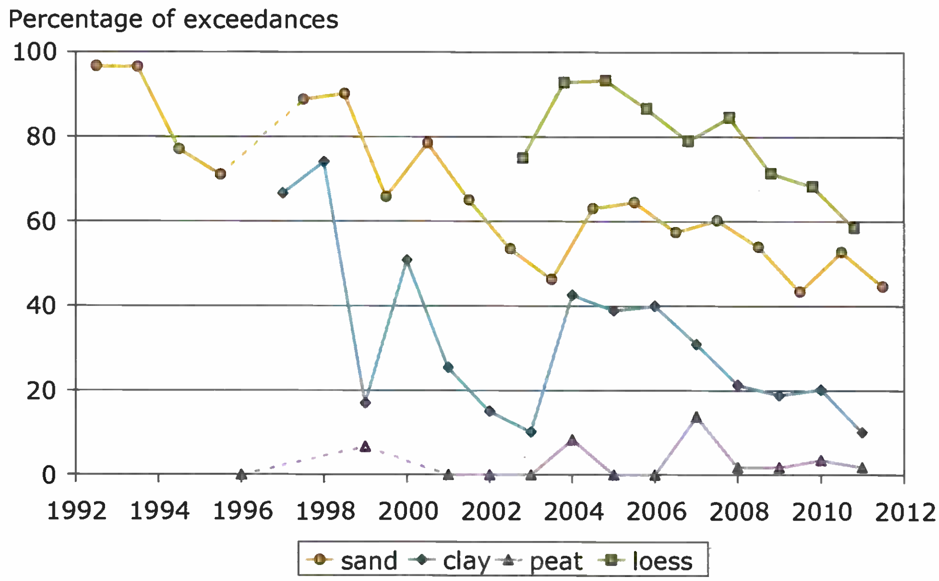


Figure S3. Percentage of exceedance of the EU standard of 50 mg/l nitrate in water leaching from root zones of farms by region in the period 1992-2011. Exceedance according to measured concentrations.

From 1984 to 2010, the average annual nitrate concentrations in groundwater at depths of 5 to 30 metres showed no clear trend, with the exception of groundwater at depths of 5 to 15 metres in the areas Sand Central and Sand South. Average annual nitrate concentrations and the exceedance of the EU standard at these depths in the period 2008-2010 were all below those in the period 2004-2007. The nitrate concentration in phreatic groundwater in areas for the extraction of drinking water (at a depth exceeding 30 metres in the Sand region) showed a slight increase between 1992 and 2004. From 2005 to 2010, this concentration was stable. The nitrate concentration in the groundwater of the Clay and Peat regions will probably not change, as the concentrations are low. Moreover, aquifers in these types of soil are often confined, and agricultural activities have little or no effect on the quality of groundwater in Clay or Peat regions.

In the period 1992-2002, the average nitrate concentration in fresh surface waters fell during the winter. In the period 2002-2010, the average nitrate concentration in the winter showed no trend. The winter maximum nitrate concentration in fresh surface waters fell between 1992 and 2010. However, the decrease in the latest reporting period (2008-2010) was negligible compared with the preceding reporting period (2004-2007). In the period 1991-2010, the average nitrate concentration in the winter showed no trend as regards marine and coastal waters. Adjusted for output via rivers (precipitation), the average inorganic nitrogen concentration in marine and coastal waters showed a decrease between 1991 and 2002. After 2002, the inorganic nitrogen concentration became stable.

Eutrophication in the period 1992-2010

In the period 1992-2004, the average summer chlorophyll-a concentration decreased in (fresh) regional waters strongly affected by agriculture. This trend did not continue in the period 2004-2010. The average summer total nitrogen concentration shows the same pattern for all (fresh) regional waters, whereas no trend is discernible in the average summer total phosphorus concentration for the period 1992-2010.

All Dutch marine waters are classified as eutrophication problem areas (OSPAR convention). Average summer chlorophyll concentrations in marine and coastal waters showed a modest reduction between 1992 and 2010.

Impact of action programmes and forecast of the future evolution of water body quality

It generally takes several years before all intended policy measures are fully implemented in agriculture. Moreover, changes in agricultural practice only have a discernible effect on water quality after some considerable time, particularly regarding the quality of the deeper groundwater and the larger bodies of surface water. This is due to processes in the soil and in the water, and to factors such as the variation in precipitation surplus from year to year. The nitrate concentration in groundwater and the exceedance of the EU target value of 50 mg/l are not solely dependent on human activities; they also depend on weather conditions, soil type and sampling depth. This last factor is related to the local hydrological and geochemical characteristics of the subsoil.

The quality of the water on farms (leaching from the root zone and ditch water) will exhibit the fastest and greatest response to measures that have been implemented as part of the action programmes. It is expected that measures of the fourth Action Programme (2010-2013) will produce noticeable changes between 2014 and 2019.

The effects on the quality of phreatic groundwater at a depth of 5 metres or more will not be manifest until after some decades have passed. Moreover, these effects will be difficult to detect owing to the mixing of groundwater of different ages and origins, as well as to the physical-chemical processes in the subsoil. The impact of the Fourth Action Programme in terms of the quality of surface waters strongly affected by agriculture will probably also be noticeable between 2014 and 2019. The effects will be difficult to demonstrate, and then only after a long time, especially in national and marine waters. This is the result of these waters mixing with water originating elsewhere (for example, water flowing in from other countries by the major rivers), and of chemical processes in the groundwater and surface waters.

Model calculations indicate that the tightening of the nitrogen application limits until 2013 will cause the nitrate concentration in upper groundwater after 2010 to decrease everywhere even further, mainly affecting a number of crops on sandy soil. For the entire sand region, the average nitrate concentration is falling to 50 milligrams per litre. The calculations also indicate that the nitrate objective for the Sand North and Sand Central areas will easily be reached. After adjusting for weather conditions, the groundwater quality is improving in both Sand South and the Loess region to an average of 70 and 60 mg/l respectively. The nitrate objective has not yet been achieved however.

From model calculations, the impact of the surface water due to the run-off and leaching of nutrients will reduce by 4% in the case of nitrogen and 2% in the case of phosphorus, compared with the levels for the 2010 application limits.

Forecasting the evolution of eutrophication due to agricultural activities is even more difficult than it is for nitrate concentrations, the main reasons being:
- the differences in surface waters with regard to their proneness to eutrophication;
- phosphorus concentrations and other factors such as hydromorphology, which also play an important part in the eutrophication process;
- contributions from other sources of nutrient input, such as urban wastewater and cross-border rivers;
- the extreme difficulty with predicting the response times of aquatic ecosystems to a substantial reduction in nutrient inputs and concentrations.

Conclusions
The years 1950 to 1987 saw the growth of the nitrogen and phosphorus surpluses in Dutch agriculture. Since 1987, however, the Netherlands has been successfully reducing them. Following the implementation of MINAS in 1998, the nitrogen surplus, which had remained stable for about seven years, underwent a further decrease.

During the reporting period (1992-2010), the water quality in terms of nitrate concentrations and eutrophication improved, thanks to the measures adopted since 1987. The nitrate concentrations in water on farms in the Sand and Clay regions were significantly lower in the period 2008 to 2010 than in preceding periods, which can be ascribed to the lower use of nitrogen after 1998. The nitrate concentrations in deeper groundwater (at depths of 5 to 30 metres) are more or less constant, apart from those in groundwater at a depth of 5 to 15 metres in Sand region. In this region, the nitrate concentration is decreasing.

The water quality should continue to improve between 2014 and 2019, owing to the measures that have been and are being taken during the Fourth Action Programme (2010-2013). Changes in deep groundwater are gradual and, therefore, no sharp changes in nitrate concentrations are to be expected. Concerning eutrophication, the quality of fresh and marine water is expected to stabilise or improve slightly in the near future.

1 Introduction

1.1 General

This report is part of the Netherlands Member State reporting under Article 10 of the Nitrates Directive and has to be submitted to the European Commission in mid-2012. The report provides an overview of current agricultural practices, and of groundwater and surface water quality in the Netherlands, describes the trends in these waters, and assesses the time scale for the change in water quality due to changes in the aforementioned practices. It deals with the implementation and impact of the measures taken as part of the action programmes, covering the period 1992-2010. If available, the data for 2011 are also presented.

This introductory chapter summarises the goal of the Nitrates Directive and the main obligations arising from it (section 1.2). The two obligations relevant for this report, i.e. reporting (section 1.3) and monitoring (section 1.4), are discussed in detail. The 2012 Member State report covers the fifth reporting phase. A review of the first four reports is given in section 1.5, with a detailed description of the fifth report's contents given in section 1.6. References (section 1.7) are included at the end of each chapter.

1.2 Nitrates Directive

The purpose of the European Nitrates Directive (EU, 1991) is to reduce water pollution attributable to nitrates from agricultural sources, and to prevent further pollution of this type. The Directive obliges Member States to take a number of measures to realise this objective.

First, Member States are obliged to designate vulnerable areas in their territory (Nitrate Vulnerable Zones, or NVZs). These zones drain into fresh surface waters and/or groundwater (Article 3, Annex 1) that contain more than 50 mg/l of nitrate, or might have this concentration if the measures described in the Directive are not taken. This applies to freshwater bodies, estuaries, sea and coastal waters that are now eutrophic or that might become eutrophic in the near future if the measures described in the Directive are not implemented. Second, the Directive obliges Member States to prepare action programmes for the designated NVZs so that the objective of the Directive can be realised (Article 5). Third, Member States are obliged to conduct suitable monitoring programmes to determine the extent of nitrate pollution in waters from agricultural sources and to assess the effectiveness of the action programmes (Article 5(6); see section 1.4 of this report for more information). Each Member State has to submit a report on the preventive measures taken, including the actual and expected results of the action programmes, to the European Commission (Article 10; see section 1.3 for more information).

The Netherlands has not designated any Nitrate Vulnerable Zones. However, it informed the European Commission in 1994 that it would prepare an action programme for the entire territory of the Netherlands, in conformity with the Nitrates Directive. According to a 1994 study (Working Group Designation NVZ, 1994), agriculture is a major source of nitrate emissions into groundwater and/or fresh surface waters and/or coastal waters. The working group therefore concluded that an action programme had to be carried out for the whole country. The research report published recently by Alterra in connection with the

Snijder motion (Schoumans et al., 2010) on the designation of zones susceptible to nitrate reaches a similar conclusion.

1.3 Reporting obligations

Annex 1 to the Nitrates Directive sets out the obligation of reporting to the Commission on preventive measures taken and their results, and on the expected results of the action programme measures. This Annex stipulates the information for inclusion in the reports, which have to be brought out every four years. In the Netherlands, this is the joint responsibility of the Ministry of Infrastructure and the Environment (I&M) and the Ministry of Economic Affairs, Agriculture and Innovation (EL&I).

Reporting obligations:

1) A statement of the preventive measures taken pursuant to Article 4. This article states that within two years following publication of the Directive, a code of Good Agricultural Practice (GAP) has to be drawn up, together with a programme for promoting the code.
2) A map showing the following:
   a) Waters identified as being affected or susceptible to being affected by pollution.
   b) The locations of the Nitrate Vulnerable Zones, distinguishing between zones already existing and zones designated since the previous report.
3) A summary of the monitoring results obtained for the purpose of designating NVZs, including a statement of the considerations that led to the designation of each zone and to the revision of the list of zones.
4) A summary of the action programmes drawn up, showing in particular:
   a) The measures required with respect to the application of artificial fertiliser, storage capacity for manure and other restrictions on the use of artificial fertilisers, as well as measures prescribed by the GAP code.
   b) The specifying of a maximum for the amount of nitrogen from manure that is allowed to be applied per ha, i.e. 170 kg/ha.
   c) Any additional or expanded measures taken to supplement measures inadequate for achieving the objective of the Directive.
   d) A summary of the results from the monitoring programmes for assessing the effectiveness of the action programmes.
   e) The assumptions made by the Member State for the likely time scale within which the measures in the action programmes are expected to have an effect, along with an indication of the degree of uncertainty inherent in these assumptions.

This report concentrates on items 4d and 4e of the reporting obligations, with the results being presented so that the effectiveness of the action programmes as a whole can be assessed. Reporting on the results of the monitoring relating to the exemption is done separately and, moreover, every year (Fraters et al., 2008; Zwart et al., 2011; Buis et al., 2012).

1.4 Monitoring obligation

Member States that have designated NVZs have different obligations from Member States who apply their action programmes to their entire territory.

Member States that had designated NVZs became obliged to monitor nitrate concentrations in fresh waters and groundwater for at least one year within two years of announcement of the Directive, i.e. before the end of 1993, and to repeat the monitoring programme at least every four years. This is necessary for designating vulnerable zones and revising the list of such zones. The monitoring for the designation of zones does not have to be conducted by the same agency that monitors the effectiveness. Effectiveness of an action programme is monitored for the purpose of studying the effect that the measures taken have on water quality.

Member States applying their action programme to their entire territory, the Netherlands for example, have to monitor the nitrate concentrations in fresh water and groundwater to determine the extent of nitrate pollution from agricultural activities. The Directive does not specify a time limit in this case. Given that the First Action Programme came into effect on 20 December 1995, monitoring needed to be performed before that date in order to establish the baseline.

The Nitrates Directive provides limited advice on how monitoring is to be conducted. In fact, only a few monitoring guidelines are given for the purpose of designating vulnerable zones (Article 6, Annex IV).

In 1998, the European Commission published draft guidelines for monitoring (EC/DG XI, 1998) in accordance with Article 7 of the Directive. Revisions were published in 1999 (EC/DG XI, 1999) and 2003 (EC/DG XI, 2003), but these are still only draft versions. The guidelines are not binding. Their purpose is to define all types of monitoring and suggest possible ways in which Member States might carry them out. In addition, the Commission aims to ensure that the monitoring regimes of the Member States are comparable.
An especially large effort has gone into the monitoring relating to the Water Framework Directive (KRW) and the Groundwater Directive (GR), for which guidance documents were published. A study is also underway into the harmonisation of the monitoring and reporting relating to the Water Framework Directive, Nitrates Directive (NiR) and State of the Environment (SoE).

1.5 The initial four Member State reports of the Netherlands

The first Member State Report of the Netherlands was submitted to the Commission in 1996 (LNV, 1996 (Ministry of Agriculture, Nature and Food Quality)). This report relates to the period between 20 December 1991 and 20 December 1995. It does not include any monitoring data to demonstrate the effectiveness of the First Action Programme, as this only came into effect on 20 December 1995. The report provides an overview of the operational monitoring programmes, with the following comments made about the results:

> "The effectiveness of the action programme cannot be properly assessed if only the results of monitoring groundwater and surface waters are considered. Measures aimed at a decrease of nutrient emissions will have a delayed effect on nitrate content, especially in surface waters. Accordingly, the estimate of the surplus on the national agricultural nitrogen balance is an appropriate tool to use when assessing the effectiveness of the

> measures. This tool makes it possible to follow the progress achieved due to reduction measures in agriculture in a more direct way."

This report also states that the effectiveness of the action programme will be reported on within four years.

The second Member State report of the Netherlands was submitted to the Commission in 2001 (LNV, 2001). This report relates to the period from 20 December 1995 to 20 December 1999. It contains the results of the monitoring programmes for assessing the effectiveness of the action programme, and is based on the report of the Working Group - Monitoring Nitrates Directive (Fraters et al., 2000). The following comments on the results of these programmes were included in the Member State report:

> "The report (of the Working Group - Monitoring Nitrates Directive) indicates that there is stabilisation, but not yet a substantial improvement of the environmental quality. This lack of improvement was foreseen because:
>
> 1. During the reporting period (1995-1999), only the use of manure was regulated, and not the use of artificial fertiliser. The decrease in the amount of nitrogen from manure was often negated by the use of artificial fertiliser. Since 1998, the Netherlands has had regulations that also restrict the use of artificial fertiliser containing nitrogen, i.e. the Mineral Accounting System (MINAS). Hence, the effects of MINAS fall outside this reporting period. Moreover, it is expected that the tightening of the Manure Policy (September 1999) will produce results in 2002 and 2003. This means that improvement of the environmental quality due to this policy will become apparent in the third reporting period.
> 2. Due to transport, decomposition and conversion processes in soil and groundwater, the effects of measures taken are not noticeable yet. It will also take some time before the monitoring results show a decrease in nitrate concentration, although just how long cannot be said. The monitoring results mainly reflect the stabilisation in agricultural practice in the 1980s and early 1990s, when the increasing pressure on the environment was brought to a halt."

The third Member State report of the Netherlands was submitted to the Commission in 2004 (VROM, 2004 (Ministry of Housing, Spatial Planning and the Environment)). This report relates to the period from 20 December 1999 to 20 December 2003. It contains the results of the monitoring programmes for assessing the effectiveness of the action programme, and is based on the report of the Working Group - Monitoring Nitrates Directive (Fraters et al., 2004). The following comments on the results of these programmes were included in the Member State report:

1. In Dutch agriculture, the nitrogen and phosphate surpluses stopped increasing in 1987, and since then, have been decreasing. After the introduction of the MINAS registration system in 1998, the nitrogen surplus, which had remained stable between 1990 and 1998, showed a further decline.
2. Due to policy measures taken since 1987, the water quality in the reporting period improved, as regards nitrate concentrations as well as eutrophication. The nitrate concentration in the upper groundwater on farms clearly decreased between 2000 and 2002, compared with previous periods. This is related to the decrease in nitrogen use since 1998. Nitrate concentrations in deep groundwater (> 30 meters) continue to increase, probably caused by the increase in nitrogen surpluses in the period before 1987.

3. The expectation is that water quality will continue to improve during the next reporting period, thanks to the measures taken during the second Action Program (1999-2003). It is expected to take a few more decades before the effects of these measures start to change the quality of deep groundwater. Despite the initial improvement in water quality, a noticeable improvement in the ecological quality of surface water is not expected in the following reporting period. In other words, the symptoms of eutrophication will not decrease rapidly.
4. The nitrate concentrations in groundwater and the exceedance of the EU target value of 50 mg/l are not solely dependent on human activities; they also depend on weather conditions, soil type and sampling depth. This last factor is related to the hydrological and geochemical characteristics of the subsoil.

The fourth Member State report of the Netherlands was submitted to the Commission in 2008 (Zwart et al., 2008). This report relates to the period from 20 December 2003 to 20 December 2007. It contains the results of the monitoring programmes for assessing the effectiveness of the action programme, and is based on the report of the Working Group - Monitoring Nitrates Directive (Zwart et al., 2008). The following comments on the results of these programmes were included in the Member State report:

1. Whereas the nitrogen and phosphorus surpluses in Dutch agriculture kept growing until 1987, since then the Netherlands has been successful in reducing them. After the introduction of the MINAS registration system in 1998, the nitrogen surplus, which had remained stable for approximately seven years, decreased again.
2. During the reporting period (1992-2006), the water quality in terms of nitrate concentrations and eutrophication improved, thanks to the measures adopted since 1987. The nitrate concentrations in water on farms were significantly lower in the period 2004 to 2006 than in preceding periods, which can be ascribed to less use of nitrogen after 1998. The nitrogen concentrations in deep groundwater (> 30 metres) continue to increase owing to the large emissions of nitrogen in the period before 1987.
3. The expectation is that water quality will continue to improve between 2010 and 2015, thanks to the measures taken during the third Action Program (2004-2007). It will probably be a few decades more until the nitrate concentration in deep groundwater stops increasing and starts to decrease. Concerning eutrophication, no clearly noticeable acceleration of the recovery process is expected.

The nitrate concentration in groundwater and the exceedance of the EU target value of 50 mg/l are not solely dependent on human activities; they also depend on weather conditions, soil type and sampling depth. This last factor is related to the local hydrological and geochemical characteristics of the subsoil.

1.6 The fifth Member State report and this report

1.6.1 Delineation and accountability

The Member States have to submit their Nitrates Directive Member State reports to the European Commission in mid-2012. The fifth Member State report covers the period 20 December 2007 to 20 December 2011. It also needs to contain the results of the monitoring programmes for assessing the effectiveness of the action programme (item 4d of section 1.3), as well as the assumptions made by the Member States about the likely time scale within which the designated waters are expected to respond to the measures in the action programme (item 4e of section 1.3).

The Ministries responsible for the Dutch reporting (see section 1.3) requested the Working Group - Monitoring Nitrates Directive to report on the two above-mentioned topics. This report represents the results of the Working Group's activities.

The starting point for preparing this report was a combination of the reporting guidelines published by the Commission in 2000 (EC/DGXI, 2000), together with the subsequently published supplements and revisions. In March 2008, the Commission published a supplement to the reporting guidelines (EC/DGXI, 2008). In November 2011, a revised version of the guidelines for the fifth Member State report was published (EC/DGXI, 2011). As far as possible, changes from the reporting guidelines from 2000 have been taken into account for the preparation of this report. The 2000 guidelines contain a request for the monitoring period results to be published on the basis of three years' monitoring for each period. Because the guidelines have not been revised in this respect, it is not clear whether results for only two monitoring periods have to be given or for all periods (in this case five). It is just as unclear as regards which periods have to be used for comparing results, as prescribed in the guidelines.

For the fourth Member State report (2008), the Working Group recommended (Fraters et al., 2007) that, in order to provide an informative overview of the status and trends of agricultural practice and the aquatic environment, the first and last two periods should be presented in the form of tables. This method is used again in this report for preparing the fifth Member State report. It means that the results of the 1992-1995, 2004-2007 and 2008-2010 monitoring periods are presented in tables. In addition, graphs are provided showing yearly averages for the 1992-2010 period. Moreover, if earlier data are available, often going as far back as the mid-1980s, these are presented as well. To limit the number of maps in the report, only those showing the water quality for the period 2008-2010 and the change in water quality between 2004 and 2010 (fourth and fifth periods) will be included.

1.6.2 *Structure of the report*

This report consists of an introduction and written account (sections 1 and 2), the results of the monitoring programmes for assessing the effectiveness of the action programmes (sections 3 to 7), a forecast of how the quality of water bodies will evolve in the future (section 8) and a summary of the results from the preceding sections, together with conclusions drawn from them. For the convenience of the reader, this summary is at the beginning of the report. To allow the sections containing the results of the monitoring programmes to be read independently, references are provided at the end of each section.

After the general introduction to the report in section 1, section 2 goes on to describe the national monitoring programme, and the purpose and design of the respective sub-programmes that provide results for this report.

The status of and trends in agricultural practice are described in section 3. The effect of both agricultural practice and changes in this practice on farm water quality is monitored by measuring the nitrate concentration in the upper metre of groundwater. Section 4 contains a description of the effect. In the remaining three sections, the status of and trends in the aquatic environment are described: deep groundwater in section 5, fresh surface waters in section 6, and marine surface waters in section 7.

Groundwater nitrate concentrations are given for four depths: upper metre of groundwater within 5 metres of the soil surface, and 5-15 metres, 15-30 metres, and more than 30 metres below the soil surface. Measurements are taken at different depths, because nitrate concentrations vary considerably with depth. Other important environmental factors considered when measuring nitrate concentrations in groundwater are land use, soil type and aquifer type. These factors are described in sections 4 and 5.

Nitrogen and phosphorus emissions are given for surface waters, along with a description of the water quality. Water quality is presented in terms of nitrate concentrations for the winter period and eutrophication parameters for the summer period. Four types of water are distinguished for fresh surface waters: regional waters affected by agriculture, other regional waters, national waters, and water at drinking water stations. They are given here in order of decreasing impact of agriculture on water quality. Other sources affecting water quality are, for example, effluent from wastewater and sewage treatment plants, sewage overflow during heavy rainstorms, and atmospheric deposits. Marine waters are divided into coastal waters and open sea, making clear the differences in nutrient emissions, which are mainly from rivers, rather than the result of direct discharge.

The forecast for the future evolution of water quality is set out in section 8. The estimates are based for the most part on recent data from the current monitoring programme. (For a detailed forecast, see PBL, 2012.)

The summary of results from the preceding chapters, as well as any corresponding conclusions, are in the section "Summary and conclusions", at the beginning of this report.

1.7 References

Buis, E., Van den Ham, A., Boumans, L.J.M., Daatselaar, C.H.G., Doornewaard, G.J. (2012). Landbouwpraktijk en waterkwaliteit op landbouwbedrijven aangemeld voor derogatie. Resultaten meetjaar 2010 in het derogatiemeetnet. Bilthoven, RIVM Report 680717028.

EC/DGXI (2011). NITRATES DIRECTIVE (91/676/CEE). Status and trends of aquatic environment and agricultural practice. Development guide for Member States' reports, November 2011.

EC/DGXI (2008). NITRATES DIRECTIVE (91/676/CEE). Status and trends of aquatic environment and agricultural practice. Development guide for Member States' reports, March 2008.

EC/DGXI (2003). Draft guidelines for the monitoring required under the Nitrates Directive (91/676/EEC). European Commission, DG XI, March 2003.

EC/DGXI (2000). Reporting guidelines for member states (art. 10) reports 'Nitrates Directive'. Status and trends of aquatic environment and agricultural practice. European Commission, DG XI, March 2000.

EC/DGXI (1999). Draft guidelines for the monitoring required under the Nitrates Directive (91/676/EEC). European Commission, DG XI, March 1999.

EC/DGXI (1998). Draft guidelines for the monitoring required under the Nitrates Directive (91/676/EEC). European Commission, DG XI, March 1998.

EU (1991). Council Directive 91/676/EEC of 12 December 1991 concerning the protection of waters against pollution caused by nitrates from agricultural sources. Official Journal of the European Communities, no. L375:1-8.

Fraters, B., Reijs, J.W., Van Leeuwen, T.C., Boumans, L.J.M. (2008). Landelijk Meetnet effecten Mestbeleid, Resultaten van de monitoring van waterkwaliteit en bemesting in meetjaar 2006 in het derogatiemeetnet, RIVM Report 680717004.

Fraters, B., Doze, J., Hotsma, P.H., Langenberg, V.T., Van Leeuwen, T.C., Olsthoorn, C.S.M., Willems, W.J., Zwart, M.H. (2007). Inventarisatie van de gegevens-, monitor- en modelbehoefte voor de EU-Nitraatrichtlijnrapportage 2008, RIVM Report 680717001.

Fraters, B., Hotsma, P., Langenberg, V., Van Leeuwen, T., Mol, A., Olsthoorn, C.S.M. (2004). Agricultural practice and water quality in the Netherlands in the 1992-2002 period, Bilthoven, RIVM Report 500003002.

Fraters, B. Van Eerdt, M.M., De Hoop, D.W., Latour, P., Olsthoorn, C.S.M., Swertz, O.C., Verstraten, F., Willems, W.J. (2000). Landbouwpraktijk en waterkwaliteit in Nederland. Achtergrondinformatie periode 1992-1997 voor de landenrapportage EU-Nitraatrichtlijn. Bilthoven, RIVM Report 718201003. (http://www.rivm.nl/bibliotheek/rapporten/718201003.pdf).

LNV (2001). Verslag als bedoeld in artikel 10 van de richtlijn 91/676/EEG inzake de bescherming van water tegen verontreinigingen door nitraten uit agrarische bronnen, over de periode van 18 December 1999 tot 18 December 1995. The Hague, Ministry of Agriculture, Nature and Food Quality.

LNV (1996). Verslag als bedoeld in artikel 10 van de richtlijn 91/676/EEG inzake de bescherming van water tegen verontreinigingen door nitraten uit agrarische bronnen, over de periode van 18 december 1995 tot 18 December 1995. The Hague, Ministry of Agriculture, Nature and Food Quality.

PBL (2012) Summary report on the Evaluation of the Fertilisers Act 2012. PBL report number 500252.

Schoumans, O.F., Keessen, A.M., Runhaar, H., Van Rijswick, H., Driessen, P., Oenema, O., Zwart, K. (2010) http://content.alterra.wur.nl/Webdocs/PDFFiles/Alterrarapporten/AlterraRapport2062.pdf

VROM (2004). Derde verslag van Nederland als bedoeld in artikel 10 van richtlijn 91/676/EEG inzake de bescherming van water tegen verontreiniging door nitraten uit agrarische bronnen. Rapportageperiode december 1999 tot december 2003. The Hague, Ministry of Housing, Spatial Planning and the Environment.

Werkgroep Aanwijzing (1994). De aanwijzing van kwetsbare zones in het kader van de EG nitraatrichtlijn: Milieukundige onderbouwing. Report of the Working Group Designation NVZ EC-ND. The Hague, Ministry of Housing, Spatial Planning and the Environment.

Zwart, M.H., Daatselaar, C.H.G., Boumans, L.J.M., Doornewaard, G.J. (2011) Landbouwpraktijk en waterkwaliteit op landbouwbedrijven aangemeld voor derogatie. Resultaten meetjaar 2009 in het derogatiemeetnet. Bilthoven, RIVM Report 680717022.

Zwart, M.H., Hooijboer, A.E.J., Fraters, B., Kotte, M., Duin, R.N.M., Daatselaar, C.H.G., Oltshoorn, C.S.M., Bosma, J.N. (2008). Agricultural practice and water quality in the Netherlands in the 1992-2006 period. Bilthoven, RIVM Report 680716003.

2 National monitoring programmes

2.1 Introduction

Several programmes exist in the Netherlands for monitoring agricultural practice and the aquatic environment. Each focuses on one of the following aspects: agricultural practice (section 2.2), effectiveness of the manure policy (section 2.3), groundwater (section 2.4), fresh and marine surface waters (section 2.5), and the water used in the production of drinking water (section 2.6). These programmes are carried out under the responsibility of different institutes and organisations.

This chapter provides a brief description of each programme. In addition to a general description of the collecting of data, information is provided about the processing of these data. They are used for the summaries that illustrate the status of and trends in agricultural practice and the aquatic environment. Details of the data collecting and processing are given in the publications in the list of references.

2.2 Monitoring agricultural practice

2.2.1 General

Agricultural practice is monitored in several ways in the Netherlands. The monitoring programmes themselves are discussed in the next sections, followed in 2.2.3 by an explanation of how a mineral balance, the production and excretion of livestock manure and nutrient excretion, and manure storage capacity are calculated.

2.2.2 Data collection

There are two agricultural monitoring programmes in the Netherlands: the Agricultural Census and the Farm Accountancy Data Network (FADN). Compliance with the regulations is also monitored.

Agricultural Census
Statistics Netherlands (CBS) collects general information about all farms, such as area of cultivated land and number of farm animals (CBS Statline, 2012). This annual collecting of data is referred to as the "Agricultural Census".

The lower limit for farms to be included in the census is Standard Revenue of € 3000. Until the end of 2009, the economic size of a farm was expressed in NGEs (Dutch Magnitude Units). As from the beginning of 2010, this was replaced by the concept of Standard Revenue (SO). As a result, the lower limit for farms appearing in Agricultural Census publications changed from 3 NGEs to € 3000 SO. For comparing different periods, the CBS data from 2000 to 2009 inclusive were recalculated on the basis of SO criteria and categories.
SO is a standardised measure of the economic size of a farm, based on the average annual revenue per crop or animal category. SO standards are set separately for each crop and animal category. They are based on five-year averages and updated every three years. The SO of a farm is the sum of all its SOs for crops and animals.

Farm Accountancy Data Network
The Agricultural Economics Research Institute (LEI, part of Wageningen University and Research Centre) collects information of a more specific nature about farm economics and technical management, via the Farm Accountancy Data Network (FADN) (Lodder and De Veer, 1985; Vrolijk, 2002; Poppe, 2004). This farm management information includes environmentally relevant data such as mineral balances (inputs and outputs of minerals), the use of pesticides, water and energy consumption, use of inorganic fertilisers, import and export of minerals (input and output of minerals, including changes in stock), and grazing frequency.

The FADN represents 1500 farms from the Agricultural Census, selected by stratified random sampling, thus forming a representative sample of Dutch agriculture. The FADN network is part of a larger EU network (EU Council Regulation 79/65/EEC). Farms included in the FADN were visited each year. Between 15% and 20% of the farms used to be replaced every year, so that the FADN network would remain representative of Dutch agriculture. Research showed that stopping the active replacement of companies after five or six years would not make the data less representative (Vrolijk et al, 2010). Replacement every five or six years was considered necessary in the past, otherwise participants would have had access to more information than non-participants would have. With the recent steep increase in computerisation (Internet and the like), this distinction hardly seems relevant. Accordingly, since 2006 the replacement has been limited to farms that stop operating, move to another region, or cease to participate for some other reason. As a result, the annual replacement of farms is no more than 3% to 5%.

The FADN represents about 75% of the total number of farms and about 91% (in NGEs) of registered agricultural production in the Netherlands. As the change from NGEs to SO units was recent, the NGE will continue to be used as the unit for economic measurement in reports utilising FADN data.

To ensure the representative nature of the FADN, farms smaller than 16 NGEs, where agriculture is generally not the main activity, are excluded from the network. Farms larger than 1200 NGEs (mostly greenhouse nurseries) are not completely suitable for data collection and are therefore also excluded. Currently, the FADN covers more than 90% of Dutch agricultural area (Vrolijk et al., 2010). Past years show similar results.

Monitoring compliance with the regulations
Compliance with regulations is largely monitored from the mineral production returns that each farmer has to complete and send back to Dienst Regelingen (DR, the government's regulatory service). Monitoring is not usually conducted in relation to an individual measure. From 2006, the policy has been in line with the EU directives, so that the emphasis is more on livestock manure and inorganic fertilisers, rather than on total mineral flows. Data collected by the General Inspection Service (AID/NVWA) shows the extent of compliance with the regulations governing legal obligations relating to matters such as manure application (quantity, timing and application method) and manure processing contracts.

Dienst Regelingen of the Ministry of Economic Affairs, Agriculture and Innovation (EL&I/DR) has prepared a summary of the activities relating to manure application guidelines and demonstration projects.

2.2.3 *Data processing*

Nitrogen and phosphorus balances
Each year, Statistics Netherlands calculates the nitrogen and phosphorus balances of the agricultural sector. All balance items are based on statistical data, except for atmospheric deposition, which is based on model calculations made by RIVM (Erisman et al., 1998; Van Jaarsveld, 1995) using statistical data on emissions into the air (Van Amstel et al., 2000). The surplus on the nutrient balance represents the difference between the input and output items. The destination of the balance surplus is not specified because leaching, output, denitrification and accumulation can only be estimated by model calculations. The method used for the calculation of the balance items was described by Statistics Netherlands in 1992 (CBS, 1992). Since 1992, minor changes have been made to the method. Until 2000, these were published in every fourth issue of the quarterly bulletin *Milieustatistieken* (statistics on the environment) of Statistics Netherlands, together with the final balances for the two years before the previous one, and the provisional balance for the previous year (see, for example, Fong, 2000 and earlier issues). Since 2000, this information has been published on the Internet (see, for example, CBS, 2012 and prior years).

Nutrient excretion and production
In the above-mentioned balance calculations, the mineral excretion from Dutch national livestock is calculated according to the difference between fodder consumption and animal products. Statistics Netherlands also calculates the manure and mineral production of the livestock based on a nutrient balance per animal in combination with the number of animals from the Agricultural Census. This method is based on the following:

1. Excretion factors calculated for each nutrient from the excretion balance, i.e. defined as the difference between the intake via feed and the retention in animal products.
2. Statistics and technical records of a particular year as source material for basic figures, supplementary to expert knowledge and feeding standards. This makes it possible from the calculations to follow not only year-to-year changes in feed composition, but also zootechnical developments affecting the efficiency of milk and meat production. Statistics are the preferred choice for source material, since they reflect continuity of method, outcome and time of publication. Basic information is used from animal feed statistics (compound feed and its nutritional value, use and production of roughage, quantity of feed per animal, in kg, et cetera) and from animal production statistics (milk production per cow, protein content of milk, egg production per hen, meat growth per animal, birth weight of piglets, et cetera).
3. Actual emission factors are calculated each year per animal category (as defined in the Agricultural Census). This means that the results derived from the technical records and statistics have to be harmonised in this respect. Care should be taken to check whether basic data refer to counted animals, housed animals or born animals.

The two calculations of the nitrogen content of manure are not completely independent of one another. Differences between nitrogen excretion (508 million kg N in Figure 3.2) and the sum of the nitrogen produced in manure and ammonia volatilisation (424 million plus 60 million kg N in Figure 3.2) are mainly due to the use of species-specific data on animal life cycles, animal production, et cetera in the calculation of manure production.

Manure storage capacity
Manure storage capacity on livestock farms has been included in the Agricultural Census for only a few years of the monitoring periods (1993, 2003, 2007 and 2010). Part of the questionnaire deals with the storage capacity for animal manure on the farm itself. Here the answers have to be in the form of the storage capacity in months for different types of manure. The data are presented in Table 3.11.

Data on the production and storage capacity of manure at each farm can also be obtained from the Farm Accountancy Data Network (FADN, see section 2.2.2), which is a representative sample of Dutch farms. In the FADN, the data only relate to liquid manure and not solid manure. These are the data used in this report (chapter 4).

2.3 Monitoring effectiveness of the manure policy

2.3.1 General

The effects of the action programme are monitored via standard programmes for groundwater and surface water, and a special programme known as Landelijk Meetnet effecten Mestbeleid (LMM), based on a national network for measuring the effects of the Manure Policy. LMM was developed to measure the effect of the Dutch Manure Policy on nutrient emissions, especially nitrate emissions, from agricultural sources into groundwater and surface water, and to monitor the effects of changes in agricultural practice on these emissions. Consequently, LMM can also identify the impact of the action programmes.

The LMM programme monitors both water quality and farm management, i.e. agricultural practice. LMM's policy measures have the aim of changing farm management in such a way that water quality improves. The quality of groundwater and surface waters is generally affected not only by agricultural practice, but also by other sources of pollution and by environmental factors such as the weather. To exclude other, diffuse sources of pollution as much as possible, the quality of water that leaches from root zones and ditchwater is monitored on farms. This type of water reveals the effects of recent agricultural activities (carried out less than four years ago). To distinguish between the effects of measures on water quality and the effects of interfering factors, such as weather, these factors are monitored as well (see Fraters et al., 2004). The next section (2.3.2) provides more details on LMM data collection, followed in section 2.3.3 by a discussion of the data processing.

2.3.2 Data collection

LMM and FADN
When the LMM monitoring programme commenced in the sandy regions in 1992, it was decided that linking LMM and FADN (see section 2.2.2) would have many advantages. Linking the two networks would make both farm management and water quality data available to all participating farms. In 1996, after the evaluation of the initial four-year period, it was decided to continue this collaboration. Because of the nature of Dutch agriculture and the high level of dynamics, the advantages of linking the FADN and the LMM to each other were obvious. The decision to use a group of farms with a changing composition for the FADN was taken in the mid-1960s. Monitoring a fixed group independent of the FADN would mean doubling the FADN's activities. The dynamic nature of the

Dutch farming sector ensures that even a fixed group of participants has a changing composition (Fraters et al., 2005). Account needs to be taken of the fact that both the FADN and LMM exclude some farms from participation. To keep the selection of participants representative, farms smaller than 16 NGEs or larger than 1200 NGEs are excluded from the FADN (see section 2.2.2). In addition to these FADN thresholds, the LMM uses a minimum participation criterion of 10 hectares of land per farm for inclusion in its network.

The monitoring network was expanded in 2006 owing to the EU granting exemption for the application per ha of 250 kg of nitrogen together with livestock manure. Not all farms in the exempt measuring network meet the conditions for inclusion in the standard monitoring programme. These farms are not suitable because they are not randomly selected. The monitoring group now has a fixed composition, except for changes arising from developments at individual farms.

Main soil type regions
The Netherlands applies the Nitrates Directive action programme to its entire territory. Even so, legislation distinguishes between main types of soil, with measures based on soil vulnerability to nitrate leaching. Accordingly, the monitoring programmes focus on the most important main soil-type regions in the Netherlands, i.e., sandy, loess, clay and peat regions. For the sandy and loess regions, consideration is given to differences in vulnerability, the result of dry or wet soil conditions (groundwater traps, or GTs) for example.

All these regions together can be regarded as a group of similar groundwater bodies. The status of the aquatic environment on farms is described for the four regions (each named according to the dominant soil type). Each region comprises one or more areas.

In all, 11 GTs have been distinguished, based on the average high and low groundwater levels (GHG and GLG respectively) in a hydrological year (from April to April). Averages are first calculated using the three highest values and the three lowest values in a year. Then the average for a number of successive years is calculated. GTs are mainly identified from estimates made in the field, using soil characteristics in combination with measurements. The effect of a GT on the nitrate concentration in the top metre of groundwater was studied by Boumans et al. (1989), who described the effect using the factor of relative nitrate concentration (RNC), with the concentration found in soils with GT VII* (lowest GHG and GLG) having the factor RNC 1.

Main farm types
Within each region, the LMM focuses on the main farm types with respect to acreage (i.e. arable farms and dairy farms). To some extent, the LMM also includes other farm types. These are factory farms (with mainly pigs and/or poultry) in sandy regions, and other livestock farms in sandy, clay and loess regions. The reason for the restriction on this group is to limit the variation in farm practice and water quality within the sample, and, hence, increase the ability to detect changes in farm practice and water quality.

Sampling and other data collection methods
The water quality on farms is monitored by sampling the water leaching from root zones and ditchwater (if present). Leaching water is measured by taking samples from different types of water: from soil water in the unsaturated zone

beneath the root zone, at a depth of between 1.5 and 3.0 metres below the surface if the groundwater is more than 5 metres below the surface; from the top metre of phreatic groundwater if the groundwater is less than 5 metres below the surface; and from drain water if the plots of land are drained by pipes. Supplementary data on environmental parameters, such as the quantity of precipitation and evapotranspiration, the percentage of land area for each soil type and GT, are collected and utilised, with the aid of models, to explain the effect of these parameters on the measurements (see section 2.3.3 and Fraters et al., 2004).

Sampling unit
The sampling unit used in the LMM is the farm. It was chosen because Dutch legislation regulates agricultural practices at farm level, farm management can be monitored more easily at farm level than on any other scale (e.g. plot level), and because farm management is already monitored at farm level in the FADN (section 2.2.2.).

Sampling frequency
Sampling frequency varies according to programme and region. The sampling frequency depends on the expected change in water quality over time, and on the variation in quality by time and space. For groundwater and surface waters, changes in nitrate concentrations over time need to be relatively large if the targets are to be reached. The current sampling frequency in the LMM is based on a statistical analysis of the results of research conducted in the period 1992-2002. This comprises research into the sandy regions in the period 1992-1995 (Fraters et al., 1998), and in the clay regions (Fraters et al., 2001) and peat regions (Fraters et al., 2002) in the period 1995-2002. In these periods, samples were taken from farms every year.

The above research revealed three major sources of variation in nitrate concentration (in decreasing order of significance):

1. Differences in nitrate concentration from farm to farm;
2. Differences in nitrate concentration from year to year on the same farm;
3. Differences in nitrate concentration from sampling point to sampling point on the same farm in any particular year.

A fourth source of this variation was differences in nitrate concentration according to farm type. The effect of this was relatively small, however. The results of the statistical analysis show that taking a limited number of samples from a large number of farms, and only taking samples a limited number of times from each farm for as long as it participates in the LMM, is more effective than frequently taking a large number of samples from a limited number of farms. A primary justification for such an approach is the fact that differences in nitrate concentration from farm to farm constitute the main source of variation.

Apart from statistical considerations, organisational and financial aspects also play a role in setting up a monitoring programme. For example, there is the effort needed to include a farm in the measuring network and maintain contact with the participant, the travelling time to go from one farm to another, and the number of samples that a sampling team can take from a farm each day. From this standpoint, it is less costly to take many samples from a farm, the number of samples being in line with the number that can be taken in one day.

Moreover, a limiting factor is the number of farms participating in the FADN that are suitable for joining the LMM.

Until 2006, the number of farms in the FADN that were potentially eligible for participation in the LMM programme was large. In the sandy, loess and peat regions, it appeared that the most productive and cost-effective method was to take samples from farms only in their first, fourth and seventh years of participation. In the clay regions, where most water drains away artificially through pipes, and samples are taken from the drain water, it appeared to be more productive and cost-effective to take the samples from farms each year.

There was a change in 2006 owing to the European Commission granting exemption for the application per ha of 250 kg of nitrogen together with livestock manure. Since that year, samples have been taken annually from every participating farm.

Since the start of the LMM, the information on agricultural practices from all participating farms has been recorded each year. Owing to circumstances, however, information is not always available from the year preceding the one in which the samples were taken.

The relationship over time between the information collected in the FADN and the actual sampling period per region is illustrated by Table 2.1.

Table 2.1. Relationship between year of information on agricultural practices and year of water sampling for all regions in the LMM.

Month	Jan-Sep	Oct	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan
Agricultural information																	
Soil water loess region																	
Groundwater sandy regions																	
Groundwater clay regions[1]																	
Groundwater peat regions[1]																	
Drain + ditch water all regions[2]																	

[1] Start of the sampling depends on the quantity of precipitation, as enough has to have fallen before leaching into groundwater occurs. Sampling begins as soon as the drain water in the area can be sampled, but no later than 1 December.

[2] Start of sampling from the drains depends on the quantity of precipitation, as enough has to have fallen in order to obtain output via drains. As soon as drains start to output water, the sampling of drain and ditch water begins.

Loess region
The loess region has been part of the LMM since 2001, the first year that data on agricultural practices were recorded in the FADN. The first data on the quality of groundwater dates from 2002. Water-quality data from the Provincial Soil Moisture Network for Limburg is added to the LMM data to obtain a picture of changes over a longer period. Instead of the farm as sampling unit, the Provincial Soil Monitoring Network uses the plot. As such, the design differs from that of the LMM (IWACO, 1999; Voortman et al., 1994). Apart from crop types, no information on agricultural practices is available for the plots in question.

Sample size
Between 1992 and 2006, the number of participating farms varied from year to year for all regions (see Table 2.2). Since 2007, the number of farms per region has been reasonably stable. Moreover, data on agricultural practices and water quality are available for nearly all farms. In total, approximately 3,574 samplings were conducted for evaluation purposes at representative farms. For the period 1992-2007, the numbers in brackets are somewhat less than those in earlier reports, because LEI had been using probability limits for the recorded data since 2009. With hindsight consequently, the results have been labelled as unreliable and are no longer being reported.

The number of distinct farms per reporting period and farm type where samples were obtained (Table 2.3) is larger than the number of farms in any single year (Table 2.2), especially in the period before 2006. This is because, in the period concerned, samples were obtained from a different group of farms each year. As a result, the average number of samples per annum in any four-year period is well below 4.

2.3.3 *Data processing*

Nutrient surpluses
The nitrogen and phosphate surpluses referred to in section 4 were calculated by a method derived from that used and described by Schröder et al. (2004, 2007). This means that, alongside the input quantities of nitrogen and phosphate in organic manure and inorganic fertilisers and the output quantities in crops, allowance is also made for other sources of input such as net mineralisation of organic substances in the soil, nitrogen fixation due to leguminous plants, and atmospheric deposition. When calculating nutrient surpluses for the soil balance, a state of equilibrium is assumed. It is expected that, in the long term, the input of organic nitrogen in the form of crop residues and organic manure is equal to the annual decomposition. An exception to this rule is made for peat soil and dal soil (a sandy subsoil covered with a layer of lumps of young peat). With these types of soil, an input due to mineralisation is taken into account: 160 kg N per hectare for grassland on peat, and 20 kg N per hectare for grassland or other crops on peat and dal soil. It is known that net mineralisation is a phenomenon of these soils, the result of groundwater-level management, which is necessary for using them for cultivation. Schröder et al. (2004, 2007) calculate the surplus for the soil balance by starting from the contribution of nutrients to the soil. In this study, a method was employed that uses farm data to calculate the surplus on the soil balance.

Nitrogen in livestock manure
For the calculation shown in chapter 4 to determine the nutrient usage via livestock manure, the production of manure on the farm concerned is calculated first. In the case of nitrogen, this concerns the net production, after deducting the nitrogen lost from stables and storage in the form of gas. The manure produced by grazing livestock is calculated by multiplying the average number of animals present by the appropriate legal standard of excretion (Dienst Regelingen, 2006). Exempt from this are farms that use special guidelines (Handreiking). To determine the manure produced by housed animals, the number of such animals is multiplied by the appropriate national standard of excretion as established by the Working Group Uniform Mineral and Manure Excretions (Van Bruggen, 2007). For more details, see Buis et al. (2012).

Table 2.2. Number of representative farms where water quality was measured in the period 1992-2011 (broken down by farm type and year) [1].

Year	Sandy regions Dairy farms	Arable farms	Other farms	Clay regions Dairy farms	Arable farms	Other farms	Peat regions Dairy farms	Loess region Dairy farms	Arable farms	Other farms
1992	67 (55)	18 (16)	7 (3)							
1993	64 (53)	19 (19)	5 (3)							
1994	32 (22)		3 (0)							
1995	62 (45)	18 (16)	3 (1)							
1996							16 (14)			
1997	14 (13)	10 (9)	3 (2)	2 (2)	4 (4)					
1998	18 (18)	11 (11)	12 (5)	15 (15)	11 (10)	1 (1)				
1999	17 (16)	8 (8)	16 (6)	23 (14)	26 (25)	4 (4)	15 (11)			
2000	23 (21)	8 (8)	11 (7)	26 (23)	27 (25)	4 (4)				
2001	30 (-)	8 (-)	5 (-)	26 (-)	25 (-)	4 (-)	8 (-)			
2002	31 (24)	10 (5)	15 (6)	25 (11)	22 (12)	6 (3)	20 (8)	7 (5)	5 (1)	4 (2)
2003	40 (31)	17 (14)	25 (13)	30 (13)	16 (6)	3 (1)	9 (8)	7 (6)	4 (3)	3 (2)
2004	68 (59)	15 (14)	20 (7)	28 (15)	36 (26)	4 (0)	12 (10)	6 (4)	7 (2)	2 (1)
2005	67 (62)	14 (10)	29 (14)	22 (20)	28 (25)	4 (2)	21 (21)	7 (5)	6 (3)	2 (1)
2006	128 (115)	15 (13)	31 (14)	21 (19)	27 (20)	7 (7)	20 (17)	22 (7)	13 (2)	8 (2)
2007	127 (123)	30 (29)	42 (22)	49 (48)	21 (20)	14 (12)	58 (58)	19 (19)	13 (10)	7 (7)
2008	117 (111)	33 (32)	48 (23)	50 (48)	23 (21)	16 (13)	57 (57)	18 (18)	12 (10)	12 (9)
2009	124 (119)	32 (31)	42 (23)	49 (48)	29 (27)	12 (7)	57 (56)	18 (18)	13 (12)	10 (9)
2010	119 (113)	30 (28)	44 (24)	50 (47)	26 (24)	13 (9)	57 (56)	18 (18)	12 (12)	11 (8)
2011	116 (107)	31 (31)	32 (20)	51 (50)	27 (26)	11 (7)	55 (54)	*	*	*

[1] The number in brackets is the number of farms for which data on agricultural practices were collected the year before. Since 2009, LEI has been using probability limits for fertilisation with livestock manure, inorganic fertiliser, other organic manure and total fertilisation. Farms that exceed these limits are considered sources of extreme observations and are therefore not included.

- No FADN data is available for 2000.

* Sampling data for 2011 from the loess region were not yet available when this report was prepared.

Table 2.3. Number of representative farms in the LMM where water quality was measured in the period 1992-2011 (broken down by farm type and year)[1].

	Sandy regions			Clay regions			Peat regions	Loess region		
Year	Dairy farms	Arable farms	Other farms	Dairy farms	Arable farms	Other farms	Dairy farms	Dairy farms	Arable farms	Other farms
1992-1995	71(3.2)	19(2.9)	7(2.6)							
1996-1999	48(1.0)	28(1.0)	31(1.0)	24(1.7)	29(1.4)	4(1.3)	16(1.9)			
2000-2003	89(1.4)	32(1.3)	42(1.3)	49(2.2)	38(2.4)	9(1.9)	24(1.5)	7(2.0)	6(1.5)	4(1.8)
2004-2007	168(2.3)	46(1.6)	80(1.5)	69(1.8)	44(2.5)	20(1.5)	62(1.8)	23(2.3)	18(2.2)	9(2.1)
2008-2011	129(3.7)	38(3.3)	62(2.7)	59(3.4)	32(3.3)	16(3.3)	60(3.8)	18(3.0)^	15(2.5)^	13(2.5)^

[1] The figure in brackets is the average number of years in which samples were obtained from a farm in the period shown in the "Year" column.

^ Data from the loess region for 2011 were not yet available. The figures shown are for the period 2008-2010.

In addition, the quantity of nutrients is recorded for all fertilisers input and output and for all fertiliser stocks (inorganic fertiliser, livestock manure and other organic fertilisers). In principle, the quantity of nitrogen and phosphate in fertilisers input and output is recorded, based on sampling. If no sampling has been conducted, standard content sizes for each type of fertiliser are used (Dienst Regelingen, 2006). Opening and closing stock levels are always calculated based on standard amounts (Dienst Regelingen, 2006).

The formula for the total quantity of fertiliser used at farm level is:

> Fertiliser used on farm = Production + Opening stock level – Closing stock level + Input – Output

The quantity of different fertilisers used on agricultural land is recorded directly in the information network.
Apart from type and quantity, time of application is also documented. The formula for the quantity of fertiliser used on grassland is:

> Fertiliser used on grassland = Fertiliser used on farm – Fertiliser used on arable land

The fertiliser used on grassland comprises fertiliser that is spread and manure excreted by grazing animals on grassland (grazing manure). The quantity of nutrients in grazing manure is calculated for each animal category by multiplying the percentage of a year that the animals spend grazing by the standard of excretion (Dienst Regelingen, 2006).

For more details, see Buis et al. (2012).

Calculating annual averages
Annual average concentrations and other parameters are taken as the average of the annual farm averages. The average value for a period is taken as the average of all farm averages for the period. An exception to this are the loess-region data from the Province of Limburg (BVM loess). These are based on average values per plot instead of farm, a consequence of the different design for this monitoring programme (section 2.3.2). Loess-region data from the LMM are, in common with the data for the other regions, based on farm averages.

Statistical analyses and observed effects
For the statistical analysis of the relationship between farm management and the nitrate concentration in water that leaches from root zones, the residual maximum likelihood (REML) method is used (Payne, 2000). A statistical method is used in order to distinguish the effect of the Manure Policy, and to filter out the effects of differences in weather patterns and sample sizes from year to year (Boumans et al., 2001; 1997). This method is currently available for the programmes in the sandy and clay regions. A detailed description of the method is given in Fraters et al. (2004). Making the correction for environmental parameters used in the method more accurate is described in Boumans and Fraters (2011).

2.4 Monitoring status and trends in groundwater

2.4.1 General

Monitoring deeper groundwater (more than 5 metres below the surface) in the Netherlands is carried out similarly to that in many other countries (Koreimann et al., 1996), by using permanent wells specially placed for the purpose of monitoring. These monitoring wells are located just outside fields to make sampling easier and to avoid hindering the work there. The first filter is at least one or two metres below the average of the lowest groundwater levels, but not more than a few metres below. This ensures (a) that the well screen is not in the unsaturated zone, and (b) that the groundwater sampled originated from the adjoining plot. The quality of the groundwater at this depth reflects the effect of agricultural practices of about ten years ago.

Preparation of this report utilised data from the National Groundwater Quality Monitoring Network (LMG).

2.4.2 *Data collection*

LMG design

The National Groundwater Quality Monitoring Network (LMG), established between 1979 and 1984, comprises about 360 monitoring sites spread throughout the Netherlands (Van Duijvenbooden, 1987). The main criteria for site selection were type of soil, land use and hydrogeological conditions. At each location, groundwater is sampled at depths of 5-15 metres and 15-30 metres below ground. Table 2.4a and b show the number of wells sampled for this study, broken down by soil type, land use and sampling depth.

Table 2.4a. Number of wells for which complete[1] data series are available for the period 1984-2010, broken down by soil type, land use and sampling depth.

Land use	Depth	Sand	Clay	Peat	Other
Agriculture	5-15	111	61	31	6
	15-30	110	60	31	5
Nature	5-15	55	4	4	3
	15-30	52	4	4	2
Other	5-15	36	18	2	6
	15-30	37	16	2	3

[1] Series were complete, or sufficient data were available to make estimates for locations that were missing data (see Fraters et al., 2004).

Table 2.4b. Number of wells for which complete[1] data series are available for the period 1984-2010 for the soil type sand, broken down by sand region and sampling depth.

	Sand North	Sand Central	Sand South	Outside sand regions
5-15 m depth	32	24	32	23
15-30 m depth	32	24	31	23

Sampling frequency
Locations were sampled annually between 1984 and 1998 (see results of Reijnders et al., 1998 and Pebesma and De Kwaadsteniet, 1997). After an evaluation in 1998 (Wever and Bronswijk, 1998), the frequency of sampling was reduced for certain combinations of soil type and depths. Shallow monitoring wells in sand regions are still sampled every year, whereas in other regions (clay and peat), they are sampled every two years. Deep wells are sampled every four years, as are shallow well screens at monitoring sites with a high chloride concentration (above 1000 mg/l due to marine effects). Finally, wells dominated by local conditions are eliminated (for example, wells near rivers and local sources of pollution). As a result, the number of wells sampled each year has been reduced from 756 to about 350. RIVM manages the network, and is responsible for both interpreting and reporting the data.

2.4.3 *Data processing*

Owing to the design of the LMG, there are locations (monitoring wells) that are not sampled each year. In order to avoid spurious trends that are a consequence of the network's design, an estimate is made of all missing data by interpolating from the data that are available. For data missing at the beginning and end of a series, the initial and final available values respectively are used to estimate them. Annual average concentrations are calculated by simply averaging the measured concentrations. Period average concentrations are calculated by averaging the period averages per location. More details on the data processing are given in Fraters et al. (2004).

The data presented in this report may deviate slightly from the data presented in the national Environmental Balance. In keeping with the previous report, a larger number of monitoring wells were used for analysis for this study. This approach resulted from the application of less strict criteria with respect to data missing for the period 1984-2010.

For the LMG wells in sandy soil, the average was determined separately for each of the areas Sand North, Sand Central and Sand South. The division into three areas was based on the 13 different areas that are used within the LMM. Moreover, this division is the same as that used for the Evaluation of the Fertilisers Act (EMW, Hooijboer et al., in preparation). Accordingly, LMG wells in sandy soil, but outside the LMM sand region are not part of this analysis.

Sand North	LMM areas: North Sand Area I, North Sand Area II and Peat Districts.
Sand Central	LMM areas: Central Sand Area plus East Sand Area.
Sand South	LMM area: only South Sand Area.

2.5 Monitoring status and trends in water used for drinking water production

2.5.1 *General*

Water production companies carry out monitoring programmes focusing on quality control of the water resource (both groundwater and surface waters), the production process and the end product. These companies have a legal obligation to report the results annually to the Netherlands' Human Environment and Transport Inspectorate, with the data management and reporting carried out by RIVM. The report utilises data on the quality of the groundwater used for the production of drinking water. Owing to the generally great depth from where groundwater is extracted, there is a substantial time lag between the measurement and the effect on the water used for production.

2.5.2 *Data collection*

Since July 2010, drinking water in the Netherlands has been supplied by 10 drinking water companies (Versteegh and Dik, 2011). About 65% of the drinking water originates from groundwater (Joosten et al., 1998). In 2010, there were 186 drinking water production sites utilising groundwater. Of this number, 108 were using phreatic (unconfined) groundwater and 78 artesian (confined) groundwater. There are about 25 sites where drinking water is produced from riverbank groundwater, dune infiltration water and surface water (see Table 2.5). The average depth of the groundwater from phreatic aquifers utilised for drinking water production is 45 metres; the average depth of the filters is between 30 and 65 metres. Of these sources, 70% are at an average depth exceeding 30 metres, and 30% are at a depth of less than 30 metres.

Concentration is measured per string of wells, and a monitoring site often comprises multiple strings. For each monitoring site, the minimum, maximum and average values of the strings are determined. Measurements are taken at each site several times a year (from three to six times, in some cases more often).

For this report, the annual average of the string averages was determined, as well as the annual maximum of the string maximums.

2.5.3 *Data processing*

For processing the data on drinking water, a supplementary database was created to tackle the issue of the changing number of drinking water production sites in the period 1992-2010. The database was constructed in two stages. First, minor information deficiencies were remedied. If no data were available for a specific site in a specific year, the average of the available values in the period from two years before to two years after the year concerned was used as an estimate. If no data were available for the aforementioned period, the production facility was classified as a site without data. Then, all sites that were still missing data were removed from the database, so that only sites with data (measured or estimated) remained.

The drinking water data are used in the section on groundwater for the production facilities that utilise phreatic and confined groundwater (see chapter 5, in particular section 5.4). These data are also used in chapter 6 (included in the surface water database) for production sites that directly or indirectly utilise surface water.

Table 2.5. Number of monitoring sites for drinking water production in the Netherlands in the period 1992-2010.

Year	Phreatic groundwater	Artesian groundwater	Surface water	Dune infiltration	River bank infiltration
1992	127	86	10	8	13
1993	126	85	11	9	14
1994	125	87	11	8	14
1995	123	86	12	8	15
1996	123	86	12	8	14
1997	121	87	11	7	14
1998	120	86	11	6	13
1999	117	86	11	7	13
2000	117	87	11	5	12
2001	113	82	9	5	12
2002	105	84	7	4	13
2003	108	82	7	4	13
2004	106	81	5	4	13
2005	102	78	3	5	12
2006	102	78	4	4	13
2007	101	78	4	4	12
2008	94	74	4	4	12
2009	98	74	4	4	11
2010	95	74	4	4	9

Figures representing annual averages and maxima for the 1992-2010 period are derived from the supplementary database. Each annual average and annual maximum is the average of averages and the average of maxima, respectively, of all drinking water production sites, for a particular year.

The tables and maps showing the status for each period and the trends between periods are derived from the original database. For each drinking water production site an average value is calculated per period, the value being based on between one and three annual averages or maxima. Only the sites monitored in both these periods are used for comparison purposes.

2.6 Monitoring status of and trends in surface water quality

2.6.1 General

The surface water monitoring networks comprise the monitoring networks for regional and large freshwater bodies, as well as those for coastal and marine waters. The large freshwater bodies, coastal waters and marine waters are collectively known as the main water system. Even a regional monitoring location covers an area larger than a farm, thus distinguishing it from the LMM (see section 2.3). As a result, the influence of other sources of pollution and the time between measurement and effect increase by steps in the order regional waters, large fresh water bodies, coastal waters and open sea. Details of the data collection are given in the next section, 2.6.2. Section 2.6.3 deals with the data processing.

2.6.2 *Data collection*

National as well as regional authorities monitor the quality of surface waters. The national authorities are responsible for monitoring the water status of the country as a whole (MWTL), and the local authorities for the regional monitoring networks (RWSNs).

Monitoring the water status of the country as a whole (MWTL)
The Directorate-General of Public Works and Water Management (RWS) of the Ministry of Infrastructure and the Environment (I&M) collects data from 39 monitoring locations at sea (including the Zeeland estuary) and from around 55 locations in large (national) fresh surface waters, such as larger rivers, canals and lakes. At sea, the frequency of sampling is once a month in winter and twice a month in summer. From the fresh surface waters, samples are generally taken every four weeks.

The RWS Water Department is responsible for the collection and presentation of marine water data and fresh surface water data.

Regional monitoring networks
The 26 water authorities have their own regional monitoring networks, comprising several thousands of monitoring sites in regional fresh water bodies. The frequency of sampling varies but is usually once every four weeks.

Every year, the RWS Water Department in collaboration with Informatiehuis Water (IHW) examines the water quality data obtained from these monitoring networks. In 2010, this survey covered data from around 450 monitoring sites in fresh water, which reflected the quality of the regional waters. In 1992, only some 250 sites were used. The water quality at these sites is affected not only by agriculture but also by other factors, in summer by the quality of water originating from the main water system.

The data presented in this report might deviate slightly from the data presented in the 2008 report (Zwart et al., 2008). For the current report, the same locations as in 2010 were used. For all these locations, the historical data were employed in such a way that the two previous periods, i.e. from 2004 to 2007 and 2008 to 2010, covered the same number of locations. These locations are sites in the KRW monitoring network of the water managers, supplemented by additional monitoring sites from outside the network.

For all monitoring sites, the water authorities have determined whether or not it is strongly affected by agriculture. In this report, consideration is given to two types of locations: locations strongly affected by agriculture, and main locations.

The data in the report here might also deviate slightly from those in the reports for the Evaluation of the Fertilisers Act 2012 (EMW, Klein et al., 2012b). Separate data were selected for the EMW by Deltares in consultation with the water authorities (Klein et al., 2012a, 2012b). Since these studies and reports were ongoing simultaneously, coupled with the uncertainty of whether the results of the EMW would be ready in time, it was decided in the case of the Nitrates Directive report to use the same procedure as in 2008.

2.6.3 *Data processing*

Nitrate concentration
The nitrate data that are obtained from measurements in fresh water relate to both nitrate and nitrite. From most monitoring sites, only combined data on nitrate and nitrite were available. There were just a few sites where separate data on nitrate had been kept up to date for a couple of years. Because nitrite concentrations in freshwater are very low compared with nitrate concentrations, the nitrate and nitrite ones are both presented here under the heading "Nitrate".

Annual average values
Figures showing winter and summer averages and maxima for the previous period are based on the data collected at different locations. The winter and summer averages are respectively the averages of the winter and summer averages of all monitoring sites in surface waters. The same approach is used to calculate the winter and summer maxima.

Definition of summer and winter
The six summer months are the most critical period with respect to eutrophication. The EU standard for nitrate is primarily aimed at assessing the effects of agriculture on surface water quality. In this context, the winter months, when leaching plays a significant role, are of particular importance. The winter period as regards fresh surface waters lasts from October to March inclusive.

In marine waters however, there is still considerable biological activity in October and November. These months are therefore not taken into account for the calculation of the winter average. The data from measurements at sea also indicate that by March biological growth is already underway and therefore nitrogen is then present in biomass. The March data are therefore not suitable for nutrient trend analyses. Accordingly, the winter period for analysing marine water is defined as December to February inclusive. To measure changes in water quality (eutrophication), the nitrogen concentrations in marine water are compared year by year. To avoid a distorted picture emerging from these comparisons, the data are analysed for the months when there is almost no biological activity.

Differences in salinity
During the winter period, the nutrient concentration in marine water is more or less constant, and shows a clear linear relationship with the salinity, the nutrient concentration becoming greater as the saline content decreases. In other words, the nutrient concentration increases the further the distance from a river-mouth. In order to compensate for differences in salinity at the various locations from year to year (due to differences in river outflows), nutrient concentrations are usually normalised for salinity (Bovelander and Langenberg, 2004).

For the present study of trends in nutrient concentration, no salinity correction has been made for the results presented in the context of the reporting guidelines. Consequently, the conclusions presented here that are based on years of in-depth studies of trends in nutrient concentration are affected by year-to-year differences in river outflows (because of differences in precipitation, et cetera), and therefore have to be treated with caution. Accordingly, additional figures are provided on inorganic nitrogen concentrations for which a correction has been made in respect of a number of monitoring sites in Dutch coastal waters. Dissolved inorganic nitrogen (DIN) is the sum of nitrite nitrogen (NO_2-

N), nitrate nitrogen (NO_3-N) and ammonium nitrogen (NH_4-N), with DIN standardised to a salinity of 30 PSU (Practical Salinity Units). The water in the Dutch part of the North Sea contains on average about 3.5% sodium chloride (NaCl), the equivalent of 35 PSU. This presentation of data is in accordance with the OSPAR Procedure, and shows the long-term trend in inorganic nitrogen concentrations corrected for the effects of precipitation.

2.7 References

Boumans, L.J.M., Fraters, B. and Van Drecht, G. (2001). Nitrate in the upper groundwater of 'De Marke' and other farms. Netherlands Journal of Agricultural Science, 49, (2-3), pp. 163-177.

Boumans, L.J.M., Van Drecht, G., Fraters, B., De Haan, T., De Hoop, D.W. (1997). Effect van neerslag op nitraat in het bovenste grondwater onder landbouwbedrijven in de zandgebieden; gevolgen voor de inrichting van het Monitoringnetwerk effecten mestbeleid op Landbouwbedrijven (MOL). Bilthoven, RIVM Report 714831002.

Boumans, L.J.M., Meinardi, C.R., Krajenbrink, G.W.J. (1989). Nitraatgehalten en kwaliteit van het grondwater onder grasland in de zandgebieden. Bilthoven, RIVM Report 728447013.

Bovelander, R.W. and Langenberg, V.T. (2004). National Evaluation Report of the joint and monitoring Programme of the Netherlands 2002. The Hague, RIKZ Report RIKZ/2004.006.

Buis, E., Van den Ham, A., Boumans, L.J.M., Daatselaar, C.H.G., Doornewaard, G.J. (2012). Landbouwpraktijk en waterkwaliteit op landbouwbedrijven aangemeld voor derogatie. Resultaten meetjaar 2010 in het derogatiemeetnet. Bilthoven, RIVM Report 680717028.

CBS Statline (2012) Statistics Netherlands, http://statline.cbs.nl/StatWeb/selection/?DM=SLNL&PA=81302NED&VW=T

CBS (1992). Mineralen in de landbouw, 1970-1990. Fosfor, stikstof, kalium. Statistics Netherlands, The Hague, Sdu/uitgeverij/cbs-publication.

Dienst Regelingen (2006). Brochure Mestbeleid 2006: het stelsel van gebruiksnormen, dated 14 March 2007 (brochure). Assen, Dienst Regelingen of the Ministry of Agriculture, Nature and Food Quality.

Erisman, J.W., Bleeker, A., Van Jaarsveld, J.A. (1998). Evaluation of ammonia emission abatement on the basis of measurement and model calculations. In: Van der Hoek, K.W., et al., Nitrogen the Conference, Proceedings of the First International Nitrogen Conference, 23-27 March 1998, Noordwijkerhout, pp. 269-274.

Fong, P.K.N., (2000). Mineralen in de Landbouw. Kwartaalbericht Milieustatistieken, 17 (4), 17-25 (and prior issues). Statistics Netherlands, Heerlen/Voorburg, 2000.

Fraters, B., Boumans, L.J.M., Van Leeuwen, T.C, Boers, P. (2005). Monitoring effectiveness of the EU Nitrates Directive action programmes: Approach by the Netherlands. In Fraters et al. (eds.), Workshop MonNO3, International workshop on monitoring the effectiveness of the EU Nitrates Directive action programmes on the environment, 11-12 June 2003, The Hague (Scheveningen).

Fraters, B., Hotsma, P., Langenberg, V., Van Leeuwen, T., Mol, A., Olsthoorn, C.S.M. (2004). Agricultural practice and water quality in the Netherlands in the 1992-2002 period, Bilthoven, RIVM Report 500003002.

Fraters, B., Boumans, L.J.M., Van Leeuwen, T.C., De Hoop, D.W. (2002. Monitoring nitrogen and phosphorus in shallow groundwater and ditch water on farms in the peat regions of the Netherlands. In: Proceedings of the 6th International Conference on Diffuse Pollution. Amsterdam, 30 September – 4 October 2002, pp. 575-576.

Fraters, B., Boumans, L.J.M., Van Leeuwen, T.C. and De Hoop, W.D. (2001). Monitoring nitrogen leaching for the evaluation of the Dutch minerals policy for agriculture in clay regions. TheScientificworld, 1 (S2), pp. 758-766.

Fraters, B., Boumans, L.J.M., Van Drecht, G., De Haan, T. and De Hoop, D.W. (1998). Nitrogen monitoring in groundwater in the sandy regions of the Netherlands. Environmental Pollution 102, S1: pp. 479-485.

IWACO (1999). Evaluatie Provinciaal meetnet grondwaterkwaliteit Limburg en Bodemvochtmeetnet Mergelland. Maastricht, IWACO report number 3361410.

Joosten, L.T.A., Buijze, S.T., Jansen, D.M. (1998). Nitrate sources of drinking water? Dutch drinking water companies aim at prevention. In: Van der Hoek, K.W. et al., Proceedings of the First International Nitrogen Conference, 23-27 March 1998, Noordwijkerhout, pp. 487-492.

Klein, J., Rozemeijer, J., Broers, H.P. (2012a). Meetnet Nutriënten Landbouw Specifiek Oppervlaktewater. Deelrapport A. Opzet meetnet. Bijdrage aan de Evaluatie Meststoffenwet 2012. Utrecht, Deltares, report 1202337-000-BGS-0007.

Klein, J., Rozemeijer, J., Broers, H.P., Van der Grift, B. (2012b). Meetnet Nutriënten Landbouw Specifiek Oppervlaktewater. Deelrapport B. Trend en Toestand. Bijdrage aan de Evaluatie Meststoffenwet 2012. Utrecht, Deltares, report 1202337-000-BGS-0008.

Koreimann, C., Grath, J., Winkler, G., Nagy, W., Vogel, W.R. (1996). Groundwater monitoring in Europe. Europees Milieuagentschap, European Topic Centre on Inland Waters, theme report 14/96.

Lodder, K. and De Veer, J. (1985). The statistical framework of the LEI-Farm Account Network. Paper presented at the "Seminar on Methodological Questions Relating to Farm bookkeeping Data", Voorburg, 21-25 October 1985; organised by FAO, the Statistical Commission, and the Economic Commission for Europe.

Payne, R.W. (ed.) (2000). The guide to GenStat. Part 2: Statistics. (Chapter 5, REML analysis of mixed models). United Kingdom, Lawes Agricultural Trust (Rothamsted Experimental Station).

Pebesma, E.J. and De Kwaadsteniet, J.W. (1997). Mapping groundwater quality in the Netherlands. Journal of Hydrology, 200: pp. 364-386.

Poppe, K.J. (ed.) (2004). Het Bedrijven-Informatienet van A tot Z. LEI, The Hague, report 1.03.06.

Reijnders, H.F.R., Van Drecht, G., Prins, H.F., Boumans, L.J.M. (1998). The quality of groundwater in the Netherlands. Journal of Hydrology, 207: pp. 179-188.

Schröder, J.J., Aarts, H.F.M., Van Middelkoop, J.C., Schils, R.L.M., Velthof, G.L., Fraters, B. and Willems, W.J. (2007). Permissible manure and fertilizer use in dairy farming systems on sandy soils in The Netherlands to comply with the Nitrates Directive target. European Journal of Agronomy 27(1): pp. 102-114.

Schröder, J.J., Aarts, H.F.M., De Bode,M.J.C., Van Dijk, W., Van Middelkoop, J.C., De Haan, M.H.A., Schils, R.L.M., Velthof, G.L. and Willems, W.L. (2004). Gebruiksnormen bij verschillende landbouwkundige en milieukundige uitgangspunten. Report no. 79. Wageningen, Plant Research International B.V.

Van Amstel, A.R., Olivier, J.G.J., Ruyssenaars, P.G. (eds.) (2000). Monitoring of greenhouse gases in the Netherlands; uncertainty and priorities for improvement. Minutes of the national workshop, 1 September 1999, Bilthoven, RIVM Report 773201002.

Van Bruggen, C. (2007). Dierlijke mest en mineralen 2002 en 2005. Voorburg/Heerlen, Statistics Netherlands.

Van Duijvenbooden, W. (1987). Groundwater quality monitoring networks: design and results. In: Van Duijvenbooden, W. and Van Wageningh, H.G. (eds.), Vulnerability of Soil and Groundwater to Pollutants. Minutes of the international conference, Noordwijk aan Zee, 30 March - 3 April 1987, pp. 179-191.

Van Jaarsveld, J.A. (1995). Modelling the long-term atmospheric behaviour of pollutants on various spatial scales. Thesis, Utrecht University, Utrecht.

Versteegh, J.F.M. and Dik, H.H.J., (2011). De kwaliteit van het drinkwater in Nederland in 2010. Publication no. VI-2011-119, RIVM report 703719081.

Voortman, A.G.W., Palsma, A.J., Ypenburg, C.G. (1994). Een studie naar de gewenste meetdichtheid ten behoeve van de monitoring van nitraatgehaltes in het bodemvocht van het Mergelland van de provincie Limburg. Delft, TNO Report TNO-MW-R94/325.

Vrolijk, H.C.J. (2002). Working procedures for the selection of farms in the FADN. In: Beers, G., et al. (eds.), Minutes of the ninth Pacioli Workshop in November 2001, Braunschweig, Germany. Published by the Agricultural Economics Research Institute (LEI), The Hague, pp. 190-199.

Vrolijk, H.C.J., Van der Veen, H.B. and Van Dijk, J.P.M. 2010. Sample of Dutch FADN 2008, LEI, The Hague, report 2010-096.

VROM (2003). De kwaliteit van het drinkwater in Nederland in 2001. VROM number 3134, Ministry of Housing, Spatial Planning and the Environment, The Hague.

Wever D. and Bronswijk J.J.B. (1998). Optimalisatie van het Landelijk Meetnet Grondwaterkwaliteit. Bilthoven, RIVM Report 714851002.

Zwart, M.H., Hooijboer, A.E.J., Fraters, B., Kotte, M., Duin, R.N.M., Daatselaar, C.H.G., Oltshoorn, C.S.M., Bosma, J.N. (2008). Agricultural practice and water quality in the Netherlands in the 1992-2006 period. Bilthoven, RIVM Report 680716003.

3 Agricultural practice

3.1 Introduction

This chapter deals with the development of agricultural practice in the Netherlands in general, and with use of nitrogen and phosphorus in Dutch agriculture in particular, for the period 1992-2011. The main topics discussed are the changes in land use, number of farms, livestock, et cetera resulting from policy measures as well as from autonomous developments (section 3.2). The nitrogen and phosphorus balances of agriculture are discussed in section 3.3, followed by a description of the other developments in agricultural practice in section 3.4.

To begin with, a summary is given of the Dutch policy measures taken in the framework of the first (1995-1999), second (1999-2003) and third (2004-2009) Action Programmes. In this context, three periods can be distinguished that do not exactly coincide with the action programme periods, i.e. 1996-1998, 1999-2005 and 2006-2010. More information is provided about this in the subsequent sections.
Between 1987 and 2000, measures were taken by means of fertiliser legislation to limit the maximum quantity of livestock manure that could be used. As part of this legislation, the application standards concerning the quantity of phosphate in fertiliser were tightened by setting a maximum for the use of livestock manure (see Table 3.1). In this way, the maximum quantity of nitrogen deposited on the land via livestock manure was also further limited. Moreover, rules were drawn up in this period governing the time of year when livestock manure could be applied as well as the method of application.

Table 3.1. Manure application standards in the period 1987-2000 in kg P_2O_5 per ha

Year	Grassland	Silage maize	Arable land
1987-1990	250	350	125
1991-1992	250	250	125
1993	200	200	125
1994	200	150	125
1995	150	110	110
1996-1997	135	110	110
1998-1999	120	100	100
2000	85	85	85

Source: LNV, 2001b, 1997, 1993b.

Between 1996 and 1998, the desired changes in agricultural practice occurred, in the form of a reduction in the quantity of manure being produced (manure production rights). In addition, a system of manure bookkeeping was introduced on livestock farms. During this period, all farms were subject to the following statutory regulations:

1. Maximum quantities for minerals that were allowed to be applied (application standards).
2. Designated time of the year when the application of manure was prohibited because of the risk of nitrogen leaching.
3. Prescribed method for applying manure to reduce ammonia emissions.
4. Covering of manure storage facilities to prevent ammonia emissions.

In 1998, the Dutch government introduced MINAS, the farm-level system for mineral accounting, based on the mineral balances of nitrogen (N) and phosphorus (P) (farm gate balance). Under this system, limits were set for the permitted levels of the N and P surpluses on farms (MINAS loss standards). MINAS did not regulate inorganic fertiliser and fixation separately, but performed accounting for the overall flow of minerals (including feed, livestock, animal products, et cetera). Farmers could therefore switch between the various components, provided they kept to the loss standards. In this way, the system regulated the N and P surplus of farms (farm gate balance). A certain N and P surplus was considered acceptable and was free of levy. The loss standards for nitrogen were tightened in the period 1998-2005 (Table 3.2). If a farmer had a surplus exceeding the loss standard, he had to pay a levy, with the levies increasing progressively between 1998 and 2003. The MINAS system was implemented in stages. On the introduction in 1998, it initially applied to livestock farms with a high animal density (above 2.5 LU/ha). In 2001, MINAS was extended to all farms. In addition, lower loss standards were set for cultivated land on sand and loess soils, which are vulnerable to nitrogen leaching.

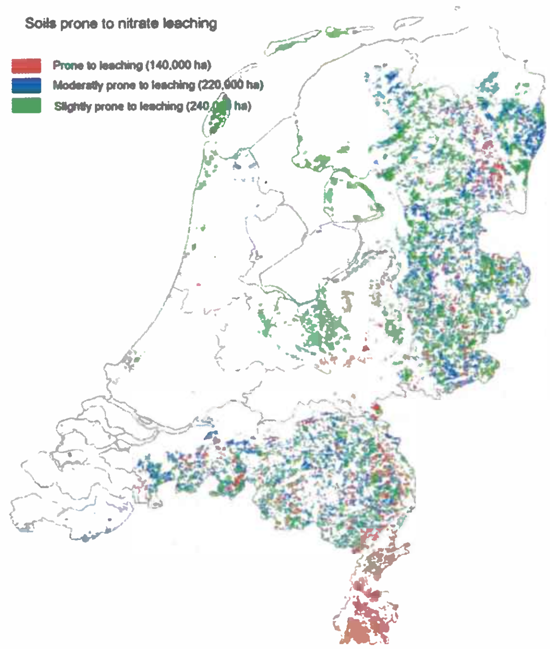
Table 3.2. Nitrogen loss standard for 1998-2005, in kg N/ha for arable land and grassland on clay, peat, sand and loess soils[1].

	Grassland		Arable land	
Year	All types	Sand[1]	All types	Sand/Loess
1998-1999	300	300	175	175
2000	275	275	150	150
2001	250	250	150	125
2002	220	190	150	110/100[1]
2003	220	190	150	110/100[1]
2004	180	160	135	100/80[1]
2005	180	160	125	100/80[1]

[1] Vulnerable soils are sand and loess soils prone to nitrate leaching, or soils with groundwater levels below average.

Source: LNV, 2001b, 1997; LEI, 2007.

The MINAS system had a greater impact than the previous system, which was based solely on application standards for livestock manure. The MINAS system also included the regulation of inorganic-fertiliser nitrogen and nitrogen fixation by legumes (arable land only). In 2002, special lower nitrogen loss standards were introduced for farms with soils prone to nitrate leaching. Overall, 140,000 ha of land were designated as having soil prone to nitrate leaching (see Map 3.1). The current system of application standards no longer includes this distinction, owing to the complexity of implementation and maintenance.



Map 3.1. Map of the Netherlands showing the areas with soil prone to nitrate leaching (red areas).
Source: LNV, 2001a.

On 1 January 2002, the Manure Transfer Contracts (MAO) system came into force to effect compliance with the application standards stipulated by the Nitrates Directive. Livestock farmers who produced too much manure were obliged to enter into manure transfer contracts with, for example, arable farms, less intensive livestock farms or manure processors. For calculating the exceedance of the allowable manure production level, the application limit was 170 kg N/ha (implemented in stages), with a higher limit of 250 kg N/ha for grassland. These limits were established in line with the Dutch notification of exemption at the time. Farmers unable to enter into manure transfer contracts for their excess manure had to reduce their livestock numbers. This change in policy was accompanied by extensive advisory campaigns and demonstration projects. In October 2003, the European Court of Justice rejected MINAS on the grounds of it being an improper implementation of the Nitrates Directive, following which the Dutch government decided to abandon MINAS and MAO. The MAO system was abolished early in 2005.

In January 2006, the Netherlands adopted a manure policy based on application standards instead of loss standards. Compared with MINAS, the new manure policy, including application standards for nitrogen in livestock manure and inorganic fertilisers as laid down by the Nitrates Directive, imposes more restrictions on the use of nitrogen and phosphorus.

The policy provides for different limits on the use of nitrogen from livestock manure, on the use of total nitrogen, and on the use of total phosphorus. The application standard for nitrogen from livestock manure is 170 kg N per ha. Farms with at least 70% grassland can adhere to a standard of 250 kg N per ha in the period 2006-2013, provided they follow a fertilising plan according to set rules. Application standards for the use of total active nitrogen differ by crop and soil type, and vary over time. After 2006, these nitrogen application standards were tightened on occasions. To show all the variations would require a table spanning several pages. Because of the size, the reader is referred to the website of the DR portal (Dienst Regelingen, 2012a and 2012b).

The Dutch manure policy in force since 2006 applies to all manure from animals kept for professional purposes or for profit. This policy has a wider scope of application than the pre-2006 policy, for example, horse manure is subject to the new legislation. There are also new regulations governing the application methods for manure and inorganic fertiliser, mainly concerning:
- the time of year when the application of manure is permitted;
- the ploughing up of grassland;
- the obligation to grow a catch crop after the cultivation of maize, to prevent nitrogen leaching.

3.2 Developments in agriculture

3.2.1 Land use

The Nitrates Directive action programme applies to the whole of the Netherlands. Land use is therefore reported at a national level (see Table 3.3). The Netherlands has a land surface area of 3.37 million ha, of which 1.85 million ha (55%) is cultivated (CBS, 2012). Land use in the first, fourth and fifth reporting periods is shown in the table below.

Table 3.3. Land use in the Netherlands (x 1,000 ha).

	1992-1995	2004-2007	2008-2011*
Grassland	1057	985	962
permanent	1021	779	778
temporary[1]	36	206	184
Silage maize	224	225	236
Other arable crops	576	572	529
Horticulture	110	114	118
Total cultivated area	1967	1897	1845
Fallow land	11	24	7
Nature and forest areas	452	484	485
Other land use	959	971	1035
Total land surface area[2]	3388	3376	3372

[1] Grassland less than five years in use by a farm.
[2] Data available for only 1993, 2006 and 2008.
**Provisional figures.*
Source: CBS (2012).

The total cultivated area steadily decreased between 1992 and 2011 by about 122,000 ha (6.2%) because of nature development, expansion of urban areas and construction of roads.

3.2.2 *Number of farms*

The total number of farms shrank by 38% during the period 1992-2011 (Table 3.4), the size of the decrease varying by farm type. The number of dairy farms contracted by 33%, horticultural farms by 45%, and pig and poultry farms by 40%. Regarding arable farms, their number fell the least (19%).

Table 3.4. Number of farms by main farm type.

	1992-1995	2004-2007	2008-2011
Arable farms	14718	12868	11858
Horticultural farms[1]	22408	14963	12381
Grazing livestock farms	56355	39874	37937
Factory farms	10997	7596	6582
Combination farms	12831	5130	3961
All farm types	117309	80430	72719

[1] Including farms under permanent cultivation.

Source: CBS (2012).

The growth of organic farming at the end of the 1990s stagnated between 2004 and 2007 (Figure 3.1). During that period, the number of farms and the land area in use for organic agriculture remained more or less stable (MNP 2007). After 2007, the number of farms increased a mere 3.5%, whereas the total acreage grew by over 15% in the same period. In 2010, 45,733 ha were in use as organic farmland. Of Dutch agriculture, 2.4% is organic (PBL, 2012a).

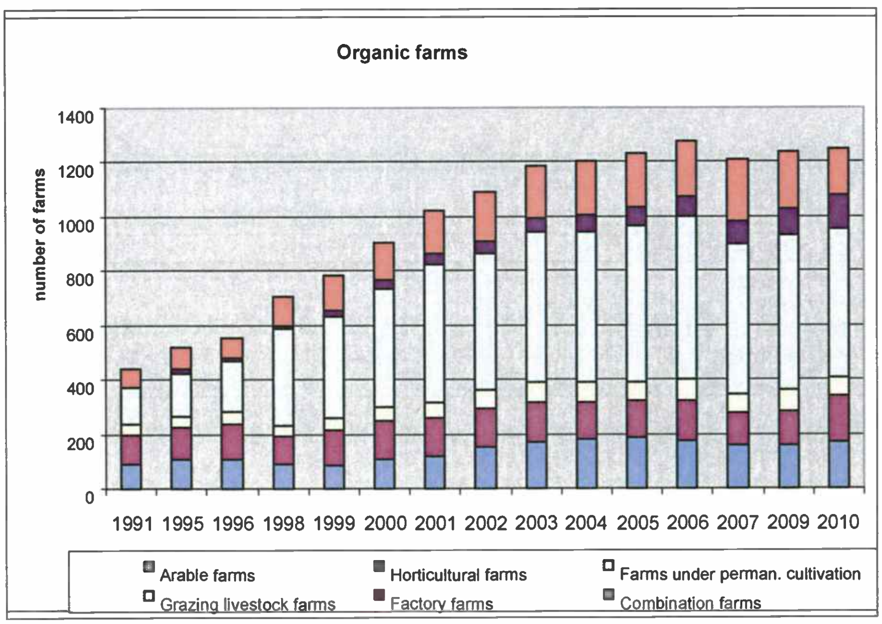


Figure 3.1. Number of organic farms in the Netherlands.

3.2.3 *Livestock*

Between 1992 and 2011, the number of cattle and pigs fell by 18% and 16% respectively. By contrast, poultry numbers rose by 4% (Table 3.5). There was a decrease in the period 1992-2007, however, with the numbers in the period 2008-2011 being slightly above those in the period 2004-2007. As the milk quota limit the number of dairy cows, an increase in milk production per cow led to a reduction in the number of cows needed to produce the permitted quantity of milk.

Table 3.5. Number of farm animals (in millions).

	1992-1995	2004-2007	2008-2011
Cattle	4.8	3.8	3.9
Pigs	14.5	11.4	12.2
Poultry	94.2	90.8	97.9
Sheep and goats	1.9	1.6	1.5

Source: CBS (2012).

3.2.4 *Nitrogen and phosphorus produced in livestock manure*

In the period 1992-2010, the nitrogen produced in livestock manure per animal decreased for all animal species (see Table 3.6). This was mainly due to a combination of lower nitrogen content in fodder and improved fodder conversion efficiency. The calculated amount of nitrogen produced per animal is greater than the amount of nitrogen in livestock manure applied to the soil as fertiliser (see Figure 3.2) because part of the nitrogen is lost via volatilisation during storage and application.

Table 3.6. Gross nitrogen excretion per animal per year (kg N per animal per year).

	1992-1995	2004-2007	2008-2010
Dairy cows	155.0	136.5	131.8
Young female livestock (1-2 years)	95.6	74.7	73.6
Meat-type pigs	14.6	12.6	12.6
Sows (with piglets)	31.3	31.5	30.4
Broilers	0.62	0.53	0.52
Laying hens	0.85	0.74	0.77

Source: CBS (2012).

From 2008 to 2011, the total annual quantity of nitrogen produced by livestock amounted to 487 million kg (see Table 3.7), which is about 30% below the annual production in the period 1992-1995. This decrease was caused mainly by a reduction in manure nitrogen produced by cattle (down 36%) and pigs (down 30%), a consequence of less nitrogen produced per animal and a smaller livestock number. Between 2008 and 2010, nitrogen production was slightly more (2.7%) than in the period before (2004-2007). Of the total nitrogen produced by Dutch livestock, 61% originates from cattle, approximately 22% from pigs, and about 13% from poultry.

Table 3.7. Nitrogen produced in livestock manure (kg N millions).

	1992-1995	2004-2007	2008-2011*
Cattle excl. veal calves	437	283	281
Veal calves	8	13	15
Pigs	153	101	107
Poultry	70	57	63
Horses and ponies	5	7	7
Other	24	14	14
All livestock	697	474	487

**Provisional figures.*
Source: CBS (2012).

The phosphorus produced in manure by Dutch livestock decreased by about 23% between the first and fifth reporting periods (see Table 3.8), mainly the result of a decrease in phosphorus production by pigs and cattle. Compared with the previous reporting period (2004-2007), the phosphorus produced in livestock manure rose slightly (5.4%).
Between 2008 and 2011, half the phosphorus produced came from cattle, a quarter from pigs and less than one sixth from poultry.

Table 3.8. Phosphorus produced in livestock manure (kg P millions).

	1992-1995	2004-2007	2008-2011*
Cattle excl. veal calves	52	38	39
Veal calves	1	2	2
Pigs	29	18	20
Poultry	13	11	12
Horses and ponies	1	1	1
Other	4	2	2
All livestock	100	73	77

**Provisional figures.*
Source: CBS (2012).

As from 2006, the Netherlands has had permission from the European Commission to use a higher application standard of 250 kg N per hectare from grazing livestock manure for farms with at least 70% grassland (exemption).
In the exemption is a condition agreed with the European Commission that the excretion of nitrogen and phosphorus by Dutch livestock be subject to an upper limit. Specifically, the excretion must not exceed the 2002 level, i.e., be no more than 75.5 million kg P (173 million kg phosphate) and 504 million kg N (including losses in the form of gas). Subsequent to the exceedance of the upper limit for phosphate from 2008 to 2010 inclusive, both targets were reached in 2011 (provisional figures; PBL, 2012b).

3.3 Nutrient balances

3.3.1 Nitrogen balance of agriculture

Figure 3.2 shows the N flows in Dutch agriculture for 2009. This flowchart combines the flows internal to the livestock production system with the nutrient flows to the soil.

Input of the flowchart comprises imported fodder, purchased inorganic fertilisers and a number of other inputs, including atmospheric nitrogen deposition from other sources in the Netherlands and abroad (mainly as NO_x). Output is a combination of the sale and export of agricultural products, the export and processing of manure, and the emission and transport of ammonia via the air. The figure illustrates the importance of the different flows. There are two major return flows: first, harvested crops used as fodder for livestock, and second, the atmospheric deposition of ammonia from livestock manure and inorganic fertilisers onto cultivated soil.

The difference between the input and output is the surplus on the national farm gate balance (shaded blue). In the figure is also shown the surplus on the national soil balance (shaded yellow). The difference between these two surpluses, due to a difference in the calculation of excretion and of manure production, is about 5% (see section 2.2.3).

Compared with 2005 (Figure 2 in the previous report), the use of inorganic fertiliser dropped sharply (down 19%). The excretion of nitrogen in livestock manure increased a little (up 5%). By contrast, the processing and export of manure together grew by such an extent (up 64%) that the surplus contracted by 19%.

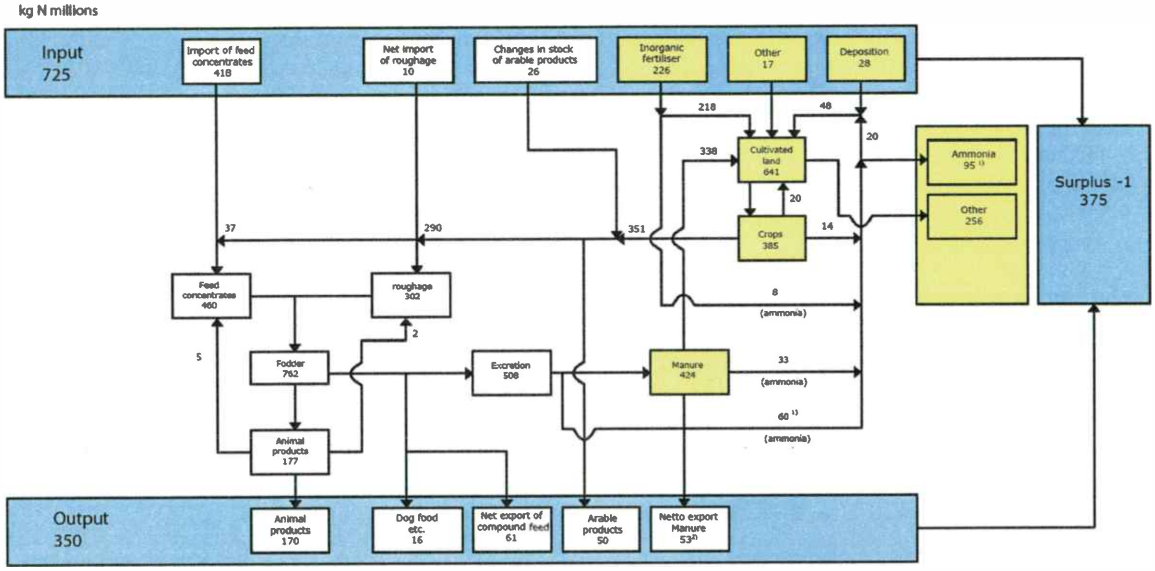
3.3.2 *Nitrogen and phosphorus soil balances*

The nitrogen surplus on the soil balance (net nitrogen input to the soil, calculated using the Statistics Netherlands' method described in section 2.2.3) was 267 million kg on average for the period 2008-2010 (Table 3.9), 18% less than for the period before. The surplus shown in Table 3.9 corresponds to the item "Other" on the national soil balance shown in Figure 3.2. The effect of this surplus on the environment, i.e. the destination of the N surplus, cannot be determined from statistical data. The surplus probably leaches partly into groundwater and/or surface water, and is partly reduced through denitrification.

The link connecting the calculation of the soil surface balance to that of the farm gate balance is the production of livestock manure. For the flowchart of Figure 3.2, excretion is calculated as the difference between the consumed fodder and the national agricultural product. Manure production is also computed per animal in a similar way and multiplied by the total number of animals.

Nitrogen in agriculture, 2009

kg N millions



1) Including volatilisation of other N-compounds (12 million kg N).
2) Including processing

Figure 3.2. Diagram of the nitrogen flows within Dutch agriculture, 2009. Source: CBS (2012).

Table 3.9. Nitrogen balance of cultivated land (kg N millions per annum).

	1992-1995	2004-2007	2008-2010*
Input[1] via:			
Livestock manure	495	349	344
Inorganic fertilisers	382	270	222
Other[2]	39	38	36
Atmospheric deposition	75	57	51
Total input	991	713	653
Total output (crops)	481	390	386
Surplus	510	324	267

[1] Excluding ammonia emissions from livestock manure and inorganic fertilisers.
[2] Including crop residues, seeds and suchlike, and other organic fertilisers (compost).
*Provisional figures.
Source: CBS (2012).

The largest inputs of nitrogen are from livestock manure and inorganic fertilisers, corrected for ammonia emissions during grazing and application. Total nitrogen input shows a decline of over 8% between the periods 2004-2007 and 2008-2010. The largest input item (livestock manure) shows a decrease exceeding 30% between the periods 1992-1995 and 2008-2010, whereas the inorganic fertiliser input is down 42%. All the nitrogen output is in harvested agricultural crops. Harvests differ from year to year because of variable weather conditions. It is likely that nitrogen uptake has decreased, although nothing indicates that harvests have become smaller due to the use of fertilisers with lower nitrogen content. The output of nitrogen fell by 20% between the periods 1992-1995 and 2004-2007.

The average phosphorus surplus on the soil balance was 16 million kg from 2008 to 2010 (Table 3.10). The main input items were livestock manure and, to a lesser extent, inorganic fertilisers. During the period 1992-2010, the input from livestock dropped by over 34%, from inorganic fertiliser by more than 65%. As the output in harvested crops fell by only 8%, the surplus decreased by 75%.

Table 3.10. Phosphorus balance of cultivated land (kg P millions per annum).

	1992-1995	2004-2007	2008-2010*
Input via:			
Livestock manure	93	65	61
Inorganic fertilisers	30	19	7
Atmospheric deposition	0	0	
Other[1]	5	4	3
Total input	128	88	71
Total output (crops)	60	55	55
Surplus	68	33	16

[1] Including crop residues, seeds and suchlike, and other organic fertilisers (compost).
Source: CBS (2012).

In order to place the effects of weather and other influences in a broader perspective, the trends in nitrogen and phosphorus surpluses since 1970 onwards are shown in Figure 3.3, with 1970 as reference year (1970 index = 100; first year for which nutrient balances were calculated).

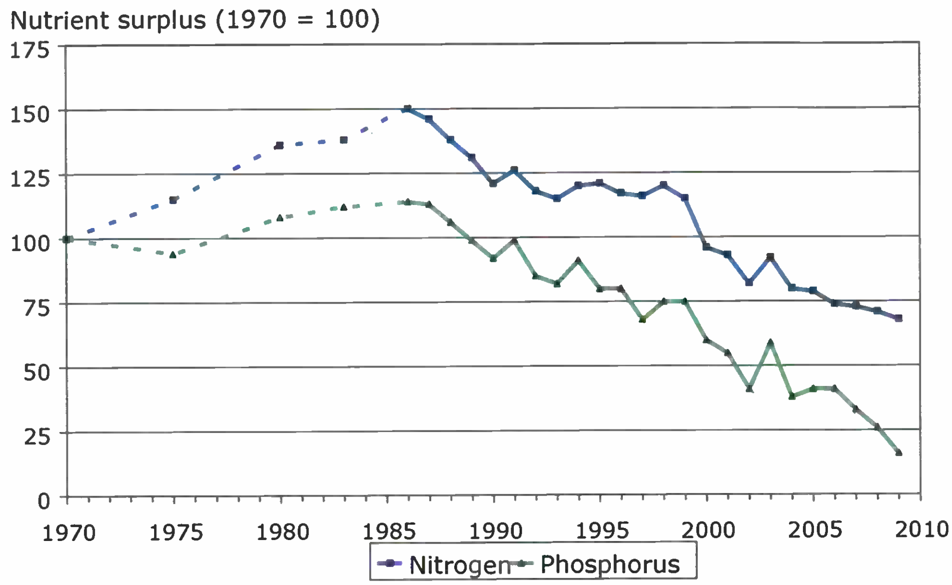


Figure 3.3. Trends in the nitrogen and phosphorus surpluses in Dutch agriculture in the period 1970-2009, with 1970 values defined as 100.
Source: CBS, 2012a.

The nitrogen surplus shows an almost constant decrease throughout the period 1986-1990. This trend is flat between 1991 and 1998. The year-to-year fluctuations shown in Figure 3.3 are mainly attributable to differences in harvest resulting from annually changing weather conditions. The nitrogen surplus shows a substantial decrease after 1998, largely attributable to the new statutory system based on the farm gate balance (MINAS) introduced in 1998. Especially affected were dairy farms, where the use of nitrogen fertiliser shrank by 40% to 50% (Fraters et al., 2004). The phosphorus surplus shows an almost constant decrease throughout the period 1986-2002, mainly the result of less manure produced because of reduced livestock numbers and feeding practices being more efficient (see Table 3.6). After 2002, the nitrogen and phosphorus surpluses stopped decreasing temporarily, but resumed falling from 2005 until 2009.

3.4 Developments in agricultural practice

3.4.1 Introduction

The previous section dealt with the use of nitrogen and phosphorus. This section considers other aspects of agricultural practice. First the changes are described regarding manure transport and processing, fertilising methods and timing, fertilisation close to waterways, green manure crops and irrigation (section 3.4.2). Figures are then given showing manure storage capacity in the Netherlands (section 3.4.3), followed by an explanation of fertilisation advice, demonstration projects and guidance (section 3.4.4). Other developments are also discussed, such as green manure crops, irrigation and limitation of ammonia emissions. The last part of 3.4 (section 3.4.6) considers compliance with the code of Good Agricultural Practice, the Mineral Accounting System, manure transfer contracts, and other aspects of agricultural regulations.

3.4.2 *Legislation governing manure application and nitrogen surplus*

3.4.2.1 Transport and processing of manure

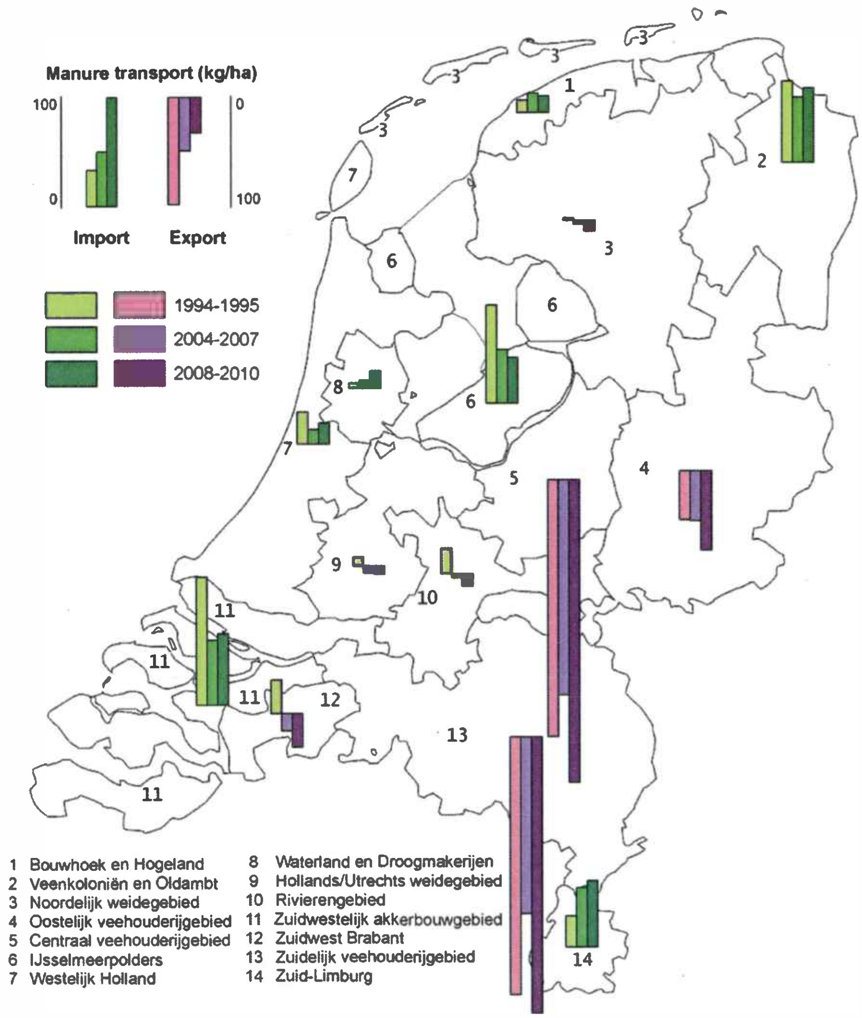
Due to the tightening of the application standards for livestock manure, increasing quantities of manure had to be transported from farms with a nitrogen and/or phosphate surplus to other farms that had space to accommodate it. Initially, as much of the excess manure as possible was transferred to nearby farms. However, manure increasingly needed to be transported over greater distances, mainly from areas where there were many farms with a surplus and where there was a surplus at the regional level. Map 3.2 shows the average import or export distance according to agricultural area for the years 1994-1995, 2004-2007 and 2008-2010, expressed as the quantity of nitrogen per hectare. Net import (green) means that on balance nitrogen was imported into an area in the form of manure, and net export (pink or lilac) means that on balance nitrogen was exported from the area concerned.

This map shows that manure transport was mainly from the central livestock area (5 on the map) and the southern livestock area (13), to the southwest arable area (11), the IJsselmeerpolders (6), and the Peat Districts and Oldambt (2). In almost all areas with substantial net transport (2, 4, 5, 6, 7, 10, 11, 12 and 13), this decreased between the periods 1994-1995 and 2004-2007. In the latest reporting period (2008-2010), a number of areas experienced an increase in net transport compared with the period before (2004-2007).

The processing and export of livestock manure to other countries have also increased significantly over the past few years. Between 2003 and 2007, processing and export accounted for the removal of 25 million kg N on average from agriculture. In the latest reporting period, the quantity was twice that amount, about 50 million kg.

3.4.2.2 Manure application, fertilisation method and timing

From 1993 to 1997, both the timing and method of manure application became subject to an increasing number of limitations. The rules for the method of application were specifically targeted at limiting the emission of ammonia into the atmosphere (see section 3.1). Since 1995, sand and loess soils (see Map 3.3) may only be fertilised between 1 February and 1 September using low-emission methods. At present, grassland and arable land on clay and peat soils may be fertilised between 1 February and 15 September.

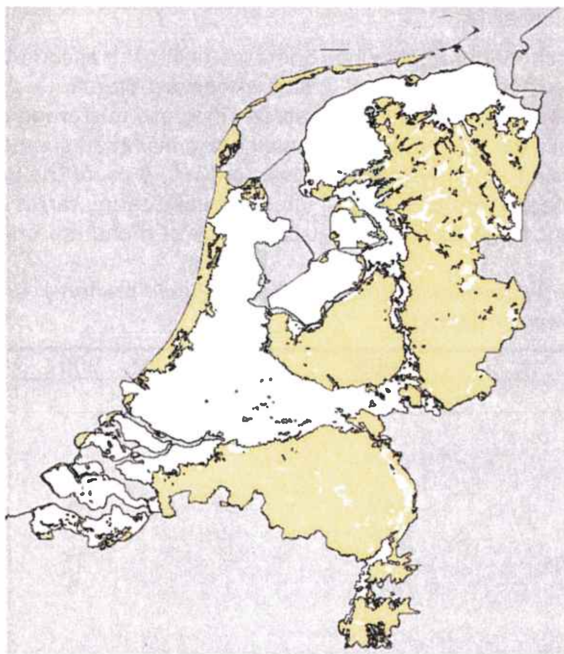


Map 3.2. Transport of nitrogen in livestock manure.
Source: CBS (2012).

In addition to the requirements for the timing of fertilisation as described above, the application of fertiliser to soil partially or completely covered with snow has been prohibited in the Netherlands since 1994. This ban was extended in 1998 to include the fertilising of completely or partially frozen soil (though this rarely occurred in practice due to the requirement to work the fertiliser into the soil, which is difficult if it is frozen).

It has also been prohibited since 1999 to use livestock manure or nitrogen fertiliser if the top layer of the soil is waterlogged. In practice, this already rarely occurred because the equipment needed for spreading fertiliser by a low-emission method is heavy and therefore causes considerable damage to the grass and soil structure in wet conditions.

Since 2006, manure spread on arable land has to be directly worked into the soil. Before then, the manure could be spread first and subsequently worked into the soil.



Map 3.3. Sand and loess areas in the Netherlands (yellow areas).
Source: LNV, 1991.

3.4.2.3 Fertilisation close to waterways

The requirement to spread manure by a low-emission method not only limits ammonia emissions and the associated nitrogen deposition, but also improves the quality of surface water. With the aid of techniques that limit ammonia emissions, manure is both spread and worked better into or under the sods, thus preventing the manure running off and directly entering watercourses.

In addition, the ban on fertilising sand and loess soils during the winter months prevents the spreading of manure during the wettest period of the year. As a result, the chance of nitrogen entering watercourses due to surface run-off is small.

Since 2000, surface water has also been protected against pollution by the Discharge Open Cultivation and Livestock Farming Decree (LOTV), which includes rules concerning the method (distance) of fertilising near watercourses. A strip of land next to a watercourse, known as a buffer strip, must not be fertilised. The width of this buffer strip varies from 0.25 to 6 metres (in special cases, as much as 14 metres wide) and corresponds to the width of the strip that must not be sprayed with pesticides. When spreading fertiliser alongside watercourses and/or their buffer strips, it is obligatory to use a limiter to prevent the fertiliser from entering the watercourse or its buffer strip. These rules are generally complied with. On about 91% of farms, the buffer strip has the required width (Vroomen and Van Veen, 2004). The Inspectorate concluded in 2006 that verifying compliance with the LOTV, especially concerning surprise checks, was substandard (Transport and Water Management Inspectorate, 2006). Projects are being employed to ascertain the effectiveness of buffer strips, as well as the factors that determine success or failure, both in practice and at policy level (STOWA, 2010a, 2010b).

3.4.3 *Manure storage capacity*

Since 2006, Dutch manure policy has operated under the assumption that farms are able to store their manure from 1 September to 1 March, i.e., for half a year. From 1993 to 2010, there was a steady increase in storage capacity (Table 3.11), with the percentage of farms having less than six months' storage capacity decreasing for all farm categories. In 2010, 96% of the dairy farms, 95% of the pig farms and 87% of the intensive calf rearing farms had storage facilities sufficient for at least six months' storage of manure production.

Table 3.11. Trend in available storage capacity (liquid manure) for various categories of livestock farms[1].

	1993	2003	2007	2010
		% of farms		
Dairy farms				
0-5 months	45	24	4	4
6-9 months	45	66	75	73
10-12 months	7	9	14	15
> 12 months	2	1	8	8
Calf rearing farms				
0-5 months	61	35	14	13
6-9 months	29	40	47	44
10-12 months	7	22	28	32
> 12 months	2	3	11	10
Pig farms				
0-5 months	30	11	5	5
6-9 months	41	43	37	36
10-12 months	23	37	36	46
> 12 months	6	9	22	13

[1] Percentage of farms with the period average capacity expressed in months for storing liquid manure. Due to the irregular collection of data, only one year (1993, 2003, 2007 and 2010) of data is available for each period.

Source: CBS, 2012a

3.4.4 *Fertilisation: recommendations, guidelines and demonstrations*

Fertilisation recommendations for arable crops, as well as for grassland and other feed crops, have scarcely changed in the past five years. Since 2006 however, policymaking has drawn many more distinctions between different crops and soil types concerning the standards for total nitrogen (see section 3.4.2). In prior years, all arable crops were treated alike within the MINAS system, without taking into account the differences in mineral needs between crops.

3.4.5 *Other developments*

3.4.5.1 Green manure crops

Cultivating winter grains on arable land is a suitable method in the Netherlands for preventing nitrate leaching. These grains are sown in the autumn and not fertilised until the following spring. The acreage with green manure crops is highly variable from year to year and dependent on the weather conditions in the autumn. There was a slight reduction in this acreage between 1992 and 2011 (Table 3.12), but this was in line with the reduction in agricultural land as a whole (Table 3.3).

Table 3.12. Cultivated land area (x 1,000 ha) in the Netherlands with crop cover in the winter (unfertilised)[1].

	1992-1995		2004-2007		2008-2011*	
Grassland[2]	1057	*54*	985	*52*	962	*52*
Winter wheat	110	*6*	120	*6*	129	*7*
Winter barley	4.0	*0.2*	3.2	*0.2*	4.6	*0.2*
Green manure crops	14.4	*0.7*	0.9	*0.0*	2.0	*0.1*
Total	1186	*60*	1109	*58*	1098	*60*

[1] The percentage of the total area fertilised with livestock manure and/or inorganic fertiliser is shown in italics in Table 3.3.

[2] Permanent as well as temporary grassland (see Table 3.3).

*Provisional figures for 2011.

Source: CBS, 2012, various tables.

Since 2006, it has been compulsory to sow sand soil with a winter crop after cultivating silage maize there. The Netherlands Food and Consumer Product Safety Authority (NVWA) conducts random inspections to check that this is done. A winter crop is not fertilised, its purpose being to absorb the nitrogen the silage maize did not. No systematically collected data were available on the area sown with winter crops after the cultivation of silage maize.

3.4.5.2 Irrigation

No cultivated land is irrigated in the Netherlands by allowing it to be temporarily submerged under water. If crops suffer from lack of water, they have to be irrigated using a sprinkler system. From 1992 to 1999, sprinkling was used several times a year to irrigate between 123,000 and 309,000 ha of land in the Netherlands (see Table 3.13), which represented 7% to 17% of cultivated land treated with fertiliser (Hoogeveen et al., 2003). The area irrigated by sprinkling is larger in dry years and smaller in wet years. In 1997, 60% of the sprinkling was on grassland, 13% on land where potatoes were being grown, and 7% on land where outdoor vegetables were being grown (Meeusen et al., 2000). Data for the years 2001 to 2009 originate from LEI (see Table 3.13). 2001, 2002 and 2005 were wet years, so that less irrigation by sprinkling was required.

Water used for irrigation is mostly groundwater (65%-80%). In normal and dry years, around 20% is surface water, while in wet years, the figure is about 15% (Hoogeveen et al., 2003).

Table 3.13. Cultivated land area (x 1,000 ha) in the Netherlands irrigated at least once a year by sprinkling between 1992 and 2009.

Year	1992	1996	1997	1998	1999
Weather		Dry		Wet	
Acreage (x 1000 ha)	265	309	198	123	161

Source: Hoogeveen et al., 2003: Meeusen et al., 2000.

Year	2001	2002	2003	2004	2005
Weather	Wet (?)		Dry		
Acreage (x 1000 ha)	22	69	278	105	82

Year	2006	2007	2008	2009
Weather	Dry			
Acreage (x 1000 ha)	180	110	86	121

Source: LEI, 2011, period 2001-2009.

3.4.5.3 Limiting ammonia emissions

Part of the nitrogen emission from agriculture is in the form of gas (ammonia for example). Most of these gas-type nitrogen compounds eventually end up in the soil and water via deposition from the atmosphere. A series of government measures has limited this type of emission, with the result that non-volatilised nitrogen remains behind in the fertiliser.

In the period 1992-2010, ammonia emissions declined by 55% (Table 3.14), the main causes being the reduction in the amount of manure produced by livestock and the obligation to apply manure using low-emission methods. After the prohibition on the spreading of manure above ground had come into force in the 1990s, it was no longer permitted from 2008 onwards to spread manure on agricultural land and work it into the soil in two passes. The effect of this ban was studied in 2010, from which it appeared that on agricultural land manure was mainly being injected into the soil. This is highly effective, and together with other modifications to fertilisation practices led to a decrease in emissions since 2005 of some 10 million kg.

Table 3.14. Ammonia emissions from agriculture (in kg NH_3 millions).

	1992-1995	2004-2007	2008-2010
Livestock manure	223	108	97
stable and storage	88	55	55
application	118	51	40
grazing	17	3	2
Inorganic fertilisers	13	14	10
Agriculture total	236	122	107

Source: Pollutant Release and Transfer Register, 2012.

3.4.6 Compliance with fertiliser legislation

For the implementation of the new manure policy, order enforcement is utilised. Current policy is based on the following standards and rules:

- Primary standards:
  - Nitrogen application standard;
  - Phosphate application standard;
  - Manure application standard.

- Secondary standards:
  - Duty of accountability for manure;
  - Timing for application of manure and organic fertilisers, and other regulations for manure and organic fertilisers;
  - Administrative obligations: determination of quantity, minimum storage of manure;
  - Animal rights regime for pigs and chickens.

- Tertiary standards:
  - Monitoring compliance with the administrative obligations that are important for the checks relating to the primary and secondary standards.

The information in Tables 3.15 and 3.16 originates from Dienst Regelingen (DR).

Dienst Regelingen administrative checks
Dienst Regelingen inspected the registered data of farms for 2009 to check compliance with the primary standards and the duty of accountability. This concerned two main target groups: farmers and manure transporters (intermediaries). If the information was incomplete, supplementary information was requested from the party concerned. The results from the sample survey are shown in Table 3.15.

Table 3.15. Overview of compliance at farm level, based on a random sample (sampling date 1 March 2011)

	Number of farms	Fines		Objections			**Collected**
Target group	investigated	Number of farms	% of farms	Number	%	Valid	
Grazing livestock	207	7	3%	2	29%	1	6
Arable	64	2	3%	-	-	-	2
Factory farm animals	45	3	7%	1	33%	1	3
Horticulture	33	-	-	-	-	-	-
Mixed	32	-	-	-	-	-	-
Intermediary	3	-	-	-	-	-	-
Total	384	12	3%	3	1%	0	**11**

Table 3.16 shows the number of violations broken down according to the three application standards and the duty of accountability. The 12 farms that received fines violated standards 14 times in total. There is no discernible trend in the types of standard violated simultaneously by a farm. The observed compliance levels are well above the desired expected levels.

Table 3.16. Overview of compliance at standard level, based on an administrative sample (sampling date 1 March 2011)

Standard	Number Investigated	Violations	Compliance percentage Observed	Target
Nitrogen application standard	380	3	99,2%	85%
Phosphate application standard	380	2	99.5%	75%
Livestock manure application standard	380	9	97,6%	85%
Duty of accountability	4	0	100%	85%

Besides the above-mentioned checks, preventive enforcement is also an important instrument. This is an instrument focussed on increasing the support for target-group policy measures such as communication, removal of grievances, and giving warnings to correct mistakes. Communication in the form of brochures, newsletters, advertisements, information meetings, etc. is itself an important instrument (Dienst Regelingen, 2010).

3.5 References

CBS (2012). Statistical database Statline at www.cbs.nl. http://statline.cbs.nl/StatWeb/dome/?LA=NL

CBS (2012a). Database containing articles from Statistics Netherlands at www.cbs.nl. http://www.cbs.nl/nl-NL/menu/themas/natuur-milieu/publicaties/default.htm.

Dienst Regelingen (2012a). Manure Policy 2006, tables, Table of Nitrogen Application Standards. www.minlnv.nl/cdlpub/servlet/CDLServlet?p_file_id=19214 (website visited on 26 April 2012)

Dienst Regelingen (2012b). Table 1, Nitrogen application standards 2008 – 2009. https://www.hetlnvloket.nl/actueel/document/fileitem/30907/tabel-1-stikstofgebruiksnormen-2008-2009 (website visited on 26 April 2012)

Dienst Regelingen (2010). Information originating from Dienst Regelingen.

Fraters, B., Hotsma, P., Langenberg, V., Van Leeuwen, T., Mol, A., Olsthoorn, C.S.M. 2004). Agricultural practice and water quality in the Netherlands in the 1992-2002 period, Bilthoven, RIVM Report 500003002.

Hoogeveen, M.W., Van Bommel, K.H.M., Cotteleer, G. (2003). Beregening in land- en tuinbouw. Rapport voor de Droogtestudie Nederland. The Hague, LEI, Report 3.03.02 (http://www.lei.dlo.nl/publicaties/PDF/2003/3_xxx/3_03_02.pdf).

LEI (2007). Bodemoverschotten op landbouwbedrijven; Deelrapportage in het kader van de Evaluatie Meststoffenwet 2007 (EMW 2007), Report 3.07.05.

LEI (2011). Farm Accountancy Data Network (FADN) 2001-2009.

LNV (2001a). Decree of 27 November 2001, containing designation of sand and loess soils and soils susceptible to leaching. Bulletin of Acts and Decrees 2001, no. 579: 1-11. The Hague, Sdu Uitgeverij.

LNV (2001b). Draft Decree containing adoption of special nitrogen loss standards for other soils susceptible to leaching for 2002. Bulletin of Acts and Decrees 2001, no. 238.

LNV (1997). Act of 2 May 1997, comprising an amendment to the Fertilisers Act. Bulletin of Acts and Decrees 360. The Hague, Sdu Uitgeverij.
LNV (1993a). Detailing of the Code of Good Agricultural practice, November 1993. The Hague, Ministry of Agriculture, Nature and Food Quality.
LNV (1993b). Notitie Derde fase Mest- en Ammoniakbeleid. Lower House, parliamentary year 1992-1993, 19 882, no. 34. The Hague, Sdu Uitgeverij.
Meeusen, M.J.G., Hoogeveen, M.H., Visee, H.C. (2000). Waterverbruik in de Nederlandse land- en tuinbouw in 1997. The Hague, LEI, report 2.00.02 (http://www.lei.dlo.nl/publicaties/PDF/2000/2_xxx/2_00_02.pdf.).
MNP (2007). Environmental balances from 2004 to 2007, report no. 500081004 (2007). Milieu en Natuurcompendium, 2008, http://www.milieuennatuurcompendium.nl/indicatoren/_nl0011-Biologische-landbouw.html?i=11-61.
PBL (2012a). Biologische landbouw: aantal bedrijven en areaal, 1991-2011. Compendium voor de leefomgeving. http://www.compendiumvoordeleefomgeving.nl/indicatoren/nl0011-Biologische-landbouw.html?i=39-178 (visited 1 September 2012).
PBL (2012b). Stikstof en fosfaat in dierlijke mest en kunstmest, 1990-2011. Compendium voor de leefomgeving. http://www.compendiumvoordeleefomgeving.nl/indicatoren/nl0106-Stikstof-en-fosfaat-in-mest.html?i=11-60 (visited 1 June 2012).
Pollutant Release and Transfer Register (2012). Emissions registration at www.emissieregistratie.nl (visited 3 July 2012)
STOWA (2010a) Bufferstroken in Nederland. Praktijk, ervaringen, onderzoek en kansen. Amersfoort, STOWA report 2010-39.
STOWA (2010b) Bijlagenrapport Bufferstroken in Nederland. Amersfoort, STOWA report 2010-39A.
Transport and Water Management Inspectorate (2006). Inspection report. Verification of compliance investigation concerning the Discharge Open Cultivation and Livestock Farming Degree (LOTV) 2005, by water authorities and the Ministry of Agriculture's General Inspectorate. http://www.ilent.nl/Images/Inspectierapport%20Verificatie%20van%20het%20nalevingsonderzoek%20LOTV_tcm334-325346.pdf (website visited on 26 April 2012)
Vroomen, H.J. and Van Veen H. (eds.) (2004). Final report on the national inspection plan relating to the Discharge Open Cultivation and Livestock Farming Decree 2003.

4 Effects of action programmes on agricultural practice and nitrate leaching

4.1 Introduction

The effect of the Dutch action programmes on nitrate emissions from agricultural sources into groundwater and surface water, and the effects of changes in agricultural practice on these emissions are assessed as part of LMM (the programme based on a national network for monitoring the effects of the Manure Policy). To this end, agricultural practice and the quality of the water that leaches from root zones are both monitored on farms (see chapter 2).

This chapter presents the results for the four main soil type regions in the Netherlands: the Sand, Clay, Peat and Loess regions. The Loess region was only added to the LMM programme in 2002, and in contrast to the other regions, no earlier LMM data is available on them. Each significant region comprises one or more areas. Approximately 46% of the Dutch agricultural area is in the Sand region, 1.5% in the Loess region, 40% in the Clay region and 12.5% in the Peat region.

Arable and dairy farming account for the largest share of land use in the Netherlands. (Together, they represent over 60% of the acreage in each region, as shown in Table 4.1.) Dairy farming accounts for the largest shares in the Peat, Sand and Loess regions. In the Clay region, arable and dairy farming both account for significant shares of land use. LMM covers 76% to 83% of the agricultural area in the different regions.

Table 4.1. Overview of the agricultural area represented by LMM, broken down by farm type and region (% of agricultural area).

	Arable	Dairy	Other[1]	Non-LMM[2]
Sand/Loess regions	14%	46%	23%	17%
Clay region	38%	31%	11%	20%
Peat region	-	76%	-	24%

[1] The category "Other" relates to other livestock farms, the composition varying by region (see section 2.3.2).

[2] "Non-LMM" includes farm types that are not covered by LMM, as well as farms that do not satisfy the LMM criteria for acreage and/or economic size. This report does not include these farms.

In the next section (4.2), the data on agricultural practice are presented for the periods 1991-1994, 2003-2006 and 2007-2010. (The first period only applies to Sand regions; for Clay and Peat regions, the period is 1995-1998.) Section 4.3 describes the nitrate concentrations in water that leaches from the root zones of a farm. There is a difference of one year between the reporting period for the farm data and the reporting period for the quality of the water that leaches from the root zone of a farm. Specifically, the farm data from 1991 to 1994 are compared with the on-farm water quality between 1992 and 1995 (see also section 2.3.2). It is assumed that the main factor affecting on-farm water quality in year x is farm practice in year x-1. The relationship between changes in agricultural practice and nitrate concentrations in on-farm water are discussed in section 4.4.

4.2 Agricultural practice

This section describes the general characteristics of agricultural practice followed by farms that are part of LMM (Table 4.2 arable farms, Table 4.3 dairy farms, and Table 4.4 other livestock farms). The average size of the LMM farms is slightly larger than the average for all Dutch farms, because farms smaller than 10 ha are excluded from LMM (see section 2.3.2). The purpose of presenting data here on LMM farms is to provide background information for identifying trends in the quality of water at these farms. Developments in agricultural practice for the Netherlands as a whole are described in chapter 3.

Arable LMM farms in the Sand and Clay regions were approximately the same size on average (about 90 ha) between 2007 and 2010 (Table 4.2). In the preceding periods, arable farms were clearly larger in the Clay region than in the Sand region. There are also some differences in the cropping plans. In the Sand region, an average of 56% of farm acreage is used for the cultivation of potatoes and sugar beet, whereas in the Clay regions the comparable figure is 32%.

Dairy farms in LMM cover a smaller area than arable farms. Currently, dairy farms in the Sand and Loess regions are smaller (about 47 ha on average) than those in the Clay and Peat regions (about 59 ha on average; Table 4.3). The percentage of grassland is highest for dairy farms in the Peat region (92%) and lowest for farms in the Loess region (73%). On the remaining land, dairy farms cultivate mainly maize, apart from the Loess region, where crops different from grass and maize are grown on 9% of the land.

The LMM group of other livestock farms in the Sand, Clay and Loess regions resemble dairy farms more than arable farms. With these farms, their characteristics fluctuate considerably more over time, showing less evidence of trends (Table 4.4).

Over the years, however, a general tendency has emerged for LMM farms to become larger, with livestock density increasing slightly in recent years, and the use of inorganic fertilisers decreasing, although not much more in the past few years.

The application of nitrogen from livestock manure on arable farms went up from 15 to 20 kg per ha (Table 4.2). During the same period, the application of nitrogen from livestock manure on dairy farms in Sand and Clay regions went down, particularly between 1995 and 2002 (Table 4.3). In the Peat region, the application of this form of nitrogen on dairy farms was smaller, and in the Loess region, it increased slightly. Over time, other livestock farms have started to apply less nitrogen from livestock manure (Table 4.4). This is true for the Sand and Clay regions, as well as the Loess region.

In general, the application of nitrogen from inorganic fertiliser showed a clear decrease. Only dairy farms in the Clay and Loess regions and the other livestock farms in the Sand and Clay regions showed an increase, mostly limited, in the application of inorganic fertiliser nitrogen during the period 2007-2010 (Table 4.3).

Table 4.2. Arable farms in the Netherlands that are part of LMM; leading characteristics of agricultural practice for farms in the Sand, Clay and Loess regions[1] for each reporting period.

Table 22	Sand region			Clay region			Loess region
Arable farms	91-94	03-06	07-10	95-98	03-06	07-10	07-10
Area (ha)	58	89	89	78	98	95	58
% potatoes	43	42	36	32	22	22	12
% sugar beets	21	19	20	13	13	10	22
% cereals	20	28	28	31	33	39	47
% other crops	16	12	16	24	33	30	19
Nitrogen from livestock manure (kg/ha)	128	124	143	79	90	102	122
Inorganic fertiliser nitrogen (kg/ha)	122	88	75	141	133	124	101
Nitrogen surplus in soil (kg/ha)	170	128	123	117	123	122	121

[1] Arable farming is rare in peat regions; Clay and Peat regions have been part of LMM since 1996, and the loess region since 2002.

Table 4.3. Dairy farms in the Netherlands that are part of LMM; leading characteristics of agricultural practice for farms in the Sand, Clay, Peat and Loess regions[1] for each reporting period.

Table 23	Sand region			Clay region			Peat region		Loess region	
Dairy farms	91-94	03-06	07-10	95-98	03-06	07-10	03-06	07-10	03-06	07-10
Area (ha)	31	45	49	36	51	57	60	62	46	44
% grassland	79	73	76	94	78	82	92	92	63	73
% maize	20	22	22	4	16	14	7	8	14	18
% other crops	1	5	2	2	6	3	2	0	23	9
Livestock (LU/ha)	3.1	2.3	2.4	2.6	2.2	2.3	2.0	2.1	1.8	2.1
Nitrogen from livestock manure (kg/ha)	357	266	248	321	267	241	244	237	216	230
Inorganic fertiliser nitrogen (kg/ha)	256	129	120	285	144	147	115	114	63	92
% manure storage [2]	106	136	146	154	137	157	139	156	107	142
Nitrogen surplus in soil (kg/ha)	325	187	178	397	178	195	209	236	89	154

[1] Clay and Peat regions have been part of LMM since 1996, and the Loess region since 2002.

[2] Percentage of total manure production that can be stored on a farm for six months.

The average storage capacity for livestock manure is sufficient to store it for six months, the longest period during which spreading is prohibited (September - January) plus one extra month. Over time, the storage capacity on dairy farms has increased (see Table 3.11).

Table 4.4. Other livestock farms in the Netherlands that are part of LMM; leading characteristics of agricultural practice for farms in the Sand and Clay regions[1] for each reporting period.

Other livestock farms	Sand region 03-06	Sand region 07-10	Clay region 07-10
Area (ha)	53	47	62
% grassland	58	66	66
% maize	16	16	9
% potatoes, sugar beets, cereals	17	13	20
% other crops	10	5	5
Livestock (LU/ha)	3.0	3.4	1.8
Nitrogen from livestock manure (kg/ha)	197	223	180
Inorganic fertiliser nitrogen (kg/ha)	87	97	104
% manure storage [2]	131	191	256
Nitrogen surplus in soil (kg/ha)	140	194	168

[1] The Clay region has been part of LMM since 1996, and the Loess region since 2002. Other types of livestock farms are rare in peat regions. There were very few livestock farms in the Sand region in the period 1991-1994, in the Clay region in the period 1995-1998, and in the Loess region in the period 2003-2006. Accordingly, the data must be used with care.

[2] Percentage of total manure production that can be stored on a farm for six months.

Calculated by the LEI method (section 2.3.3), the average nitrogen surpluses of farms monitored as part of the LMM programme differed between farm types and, to a lesser extent, between soil type areas (see Figure 4.1). The reduction in nitrogen surplus is comparable to that shown in Figure 3.3, and is due to the decreasing use of inorganic fertilisers and, to a lesser extent, of livestock manure. During the period 2007-2010, many farm types and regions saw their nitrogen surpluses increase, in most cases only to a limited extent.

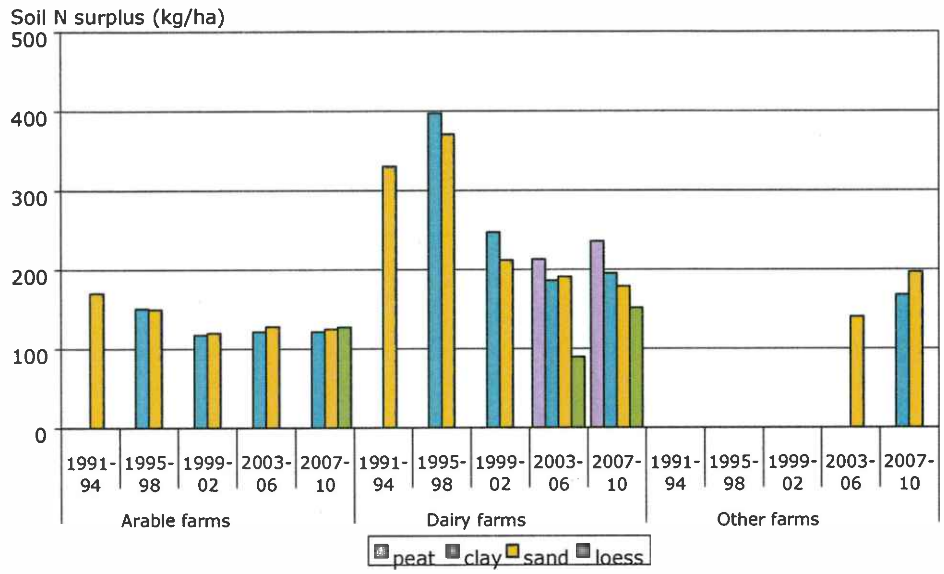


Figure 4.1. Average nitrogen surplus on the farm gate balance (calculated by the LEI method, see section 2.3.3) of arable, dairy and other types of farms in the Sand, Loess, Clay and Peat regions, from 1991 to 2010.

4.3 Nitrate in water that leaches from a root zone

4.3.1 Overview at national level

The average nitrate concentration in water that leaches from the root zones (leaching water) on farms in LMM varies from region to region. It is lowest in Peat region, higher in Clay region, and highest in Sand and Loess regions (see Figure 4.2). Nitrate is the main component of nitrogen in the leaching water on farms in the Sand, Loess and Clay regions (about 85%, over 95% and about 80%, respectively; see Figure 4.3). It is a smaller component of the nitrogen in leaching water and ditchwater in the Peat region (under 30%). In the Peat region, ammonia is the main form of nitrogen in leaching water (30%-60%) and of organic nitrogen in ditchwater (25%-50%). The ammonium concentration in the groundwater of the Peat region increases with the depth at which the water is located (Van der Grift, 2003), the cause being attributed to the mineralisation of organic material (Meinardi, 2005).

The status and trend of the nitrate concentration differs between regions, as well as between farm types within a region (see Figure 4.2).

During the first three periods, the nitrate concentration in the Sand region decreased for all farm types. In the fourth period, the average nitrate concentration at arable farms and at other livestock farms was up, whereas the concentration at dairy farms was the same as in the period before. In the fifth and latest period, the average nitrate concentration decreased for all farm types. For arable and other livestock farms, their average concentrations reverted to the levels in the period 2000-2003. The highest nitrate concentrations measured in the Sand region were at other livestock farms. In the first two reporting periods, nitrate concentration at dairy farms was higher than at arable farms. By contrast, in the next three reporting periods, the average concentration at dairy farms was actually lower than at arable farms.

In the Clay region, there was a clear reduction in the nitrate concentration at dairy farms between the second and third periods. By contrast, the concentration at arable and other livestock farms changed only marginally (see Figure 4.2). The number of other livestock farms was relatively small during the initial periods (see Table 2.2). Measurements taken in the fourth period show an increase in average nitrate concentration for all farm types, followed by a decrease during the period 2008-2011, the same as in the Sand region.

In the Peat region, the low average nitrate concentrations at dairy farms are exhibiting a trend comparable to that in the clay regions. There was a decrease between 2000 and 2003, followed by an increase between 2004 and 2007 and a decrease in the latest period (see Figure 4.2).

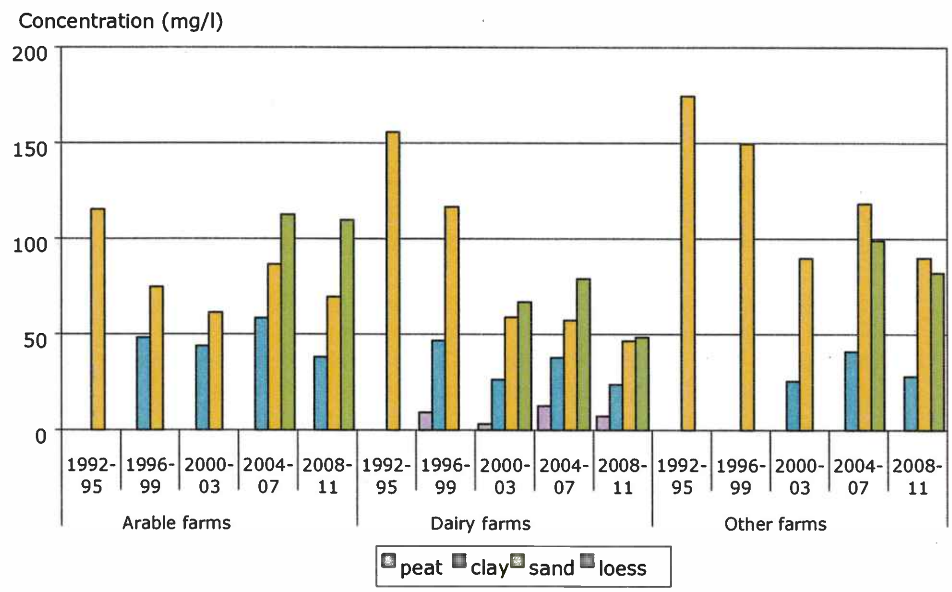


Figure 4.2. Average nitrate concentration in the water leaching from root zones of arable, dairy and other livestock farms, by region in the period 1992-2011 (2010 in the case of the Loess region).

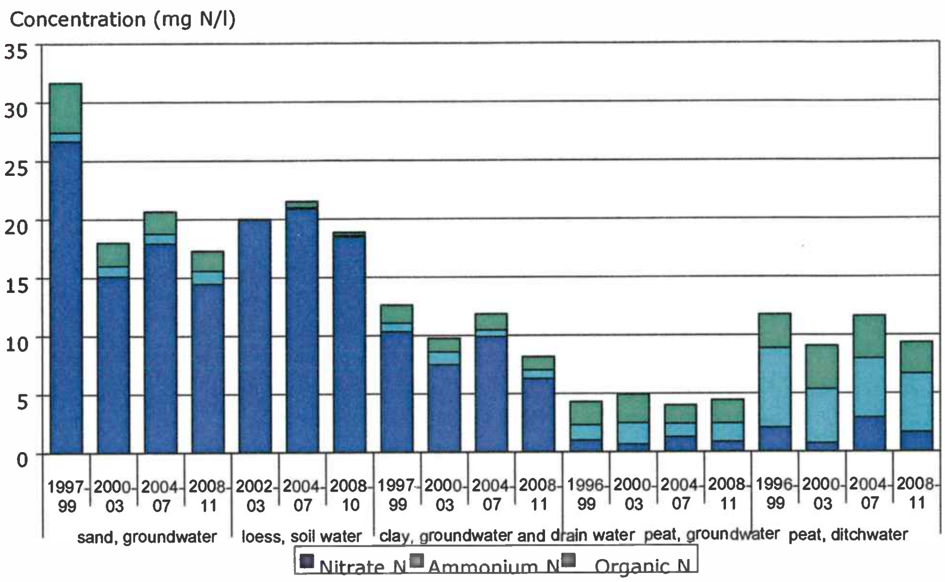


Figure 4.3. Nitrogen concentration (mg/l) in the water leaching from the root zones of farms in the Sand, Loess, Clay and Peat regions of the Netherlands, in the periods 1996-1999, 2000-2003, 2004-2007 and 2008-2011.

From 1992 to 2003, the nitrate concentrations measured in the leaching water on farms in the Sand regions showed a clear decrease, followed by a period of stabilisation. After 2008, there could be a further reduction (seeFigure 4.4). Following a time of stable, high nitrate concentration between 2002 and 2007, the Loess region showed a slight decline in the years 2008 to 2010. For the entire period, the Clay regions exhibited a downward trend, apart from a few years, 2004 being one of them. The explanation for the high concentration might be the relatively dry 2003. No discernible trend can be seen in the leaching water on farms in the Peat region. The higher concentrations in 2004 and 2007 are attributable to the greater proportion of farms that exceeded the EU standard of 50 mg/l (Figure 4.5).

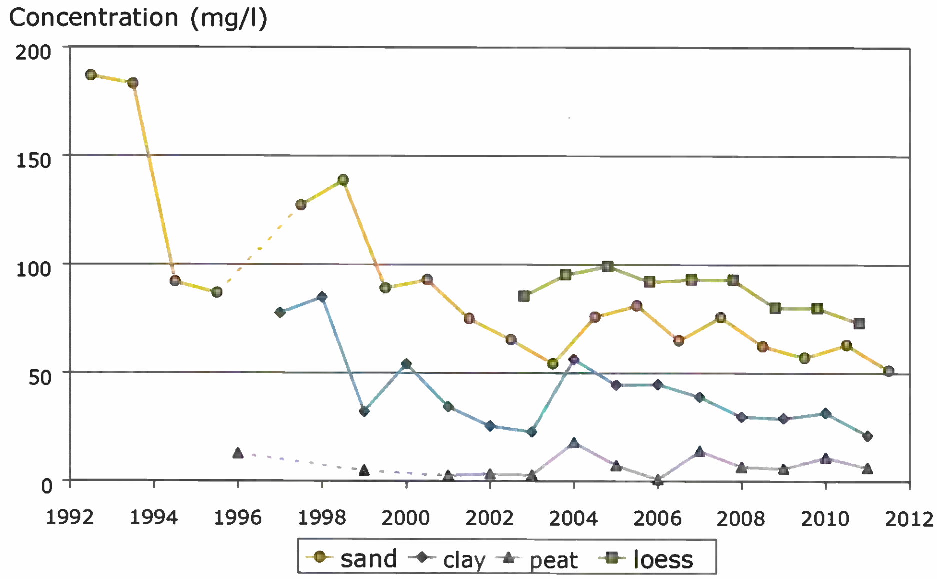


Figure 4.4. Nitrate concentrations in water leaching from root zones of farms by region in the period 1992-2011. Annual average of measured concentrations.

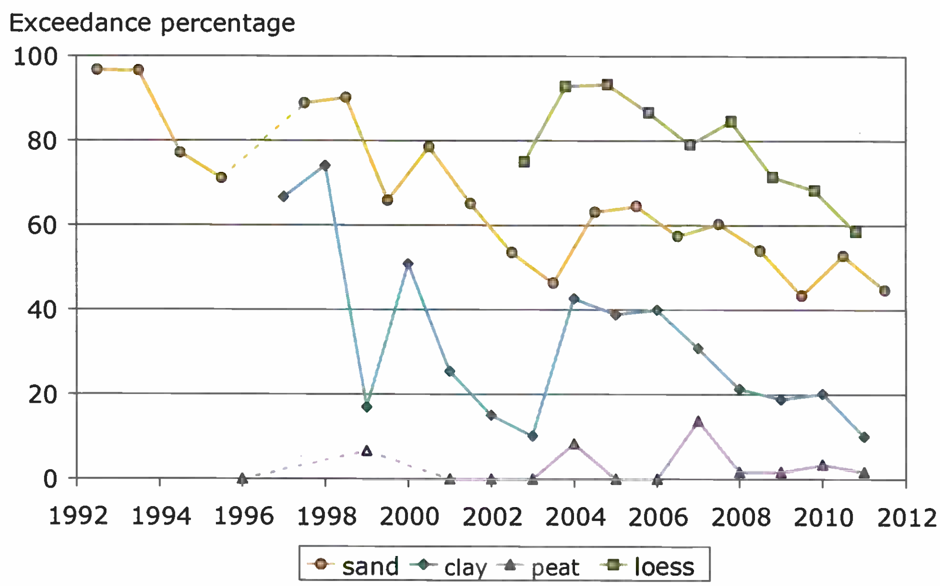


Figure 4.5. Percentage of exceedances of the EU standard of 50 mg/l nitrate in water leaching from root zones of farms by region in the period 1992-2011. Exceedance according to measured concentrations.

The average nitrate concentration varied sharply from year to year, the main cause of the fluctuations being differences in precipitation surplus. This leads to differences in the degree of dilution and the depth of the groundwater table (Boumans et al., 2001; 1997). Moreover, a rise by the groundwater table results in more denitrification. There were also changes in the composition of the group of farms being monitored. From 1996 to 2006, LMM was a variable monitoring network (Table 2.3 and section 2.3.2), causing year-to-year differences to be greater than those in the period after 2006. Since 2006, LMM has been a fixed monitoring network, although some farms cease operating and are then replaced. Apart from this, some farms buy and/or sell land, or are parties to land exchange transactions. Such changes led to differences in the proportions of soil types within LMM from year to year. In this way, any future increase in the percentage of peat soil on farms in the Sand region will in time result in lower measured nitrate concentrations, even if the nitrogen surplus does not change. A statistical model has been developed to determine the effects of the Manure Policy. It was designed so that the effect of such interfering factors is calculated, enabling a nitrate concentration curve to be estimated, with these interfering factors filtered out. This curve represents the standardised nitrate concentration (Fraters et al., 2004, Boumans and Fraters, 2011).

There was a marked reduction in the standardised nitrate concentration in the leaching water on farms in the Sand region from about 135 mg/l in the period 1992-1995 to about 70 mg/l in the period 2004-2007 (Figure 4.6). The standardised nitrate concentration also fell in the Clay region, although the data series was still relatively small (see section 2.3.2).

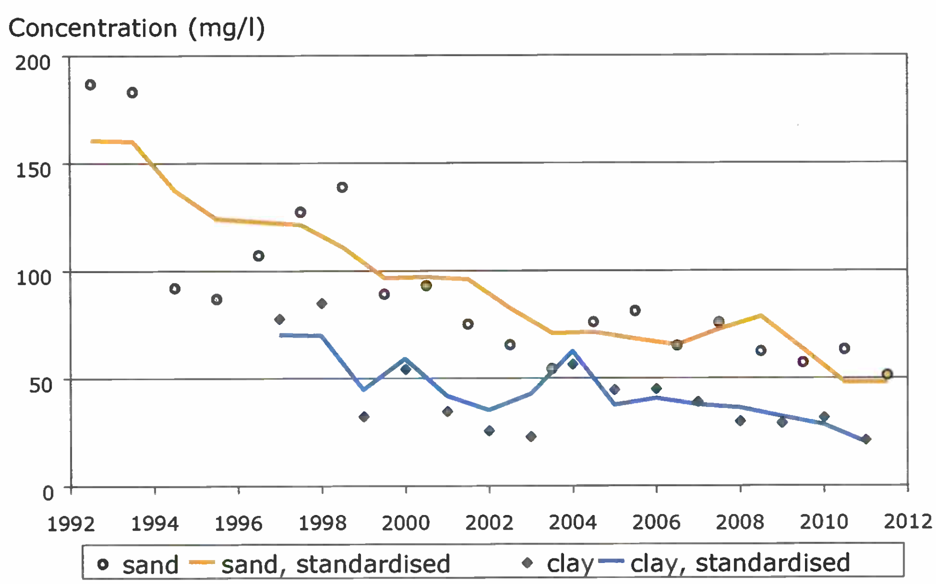


Figure 4.6. Nitrate concentrations in water leaching from root zones of farms in the Sand and Clay regions in the period 1992-2011. Average annual measured and standardised concentrations.

The percentage of farms with a nitrate concentration above the EU standard of 50 mg/l (see Figure 4.5) shows a falling trend similar to that in the nitrate concentration (Figure 4.4). The EU standard was most often exceeded in the Loess region. In the Sand region, there were more exceedances of the standard than in the Clay and Peat regions. The concentration in the Peat region is rarely above 50 mg/l. Despite the modest fall in average nitrate concentration on the Loess region (from 100 mg/l to 80 mg/l), the exceedance percentage has dropped by 30 percentage points since 2005. The exceedance percentage has fallen substantially since monitoring began. Variation from year to year is attributable to interfering factors. Even if allowance is made for them, the percentage of exceedances of the EU standard still shows a decline (Figure 4.7). The standardised exceedance in the Sand regions fell from about 95% in the period 1992-1995 to about 50% in the period 2008-2011.

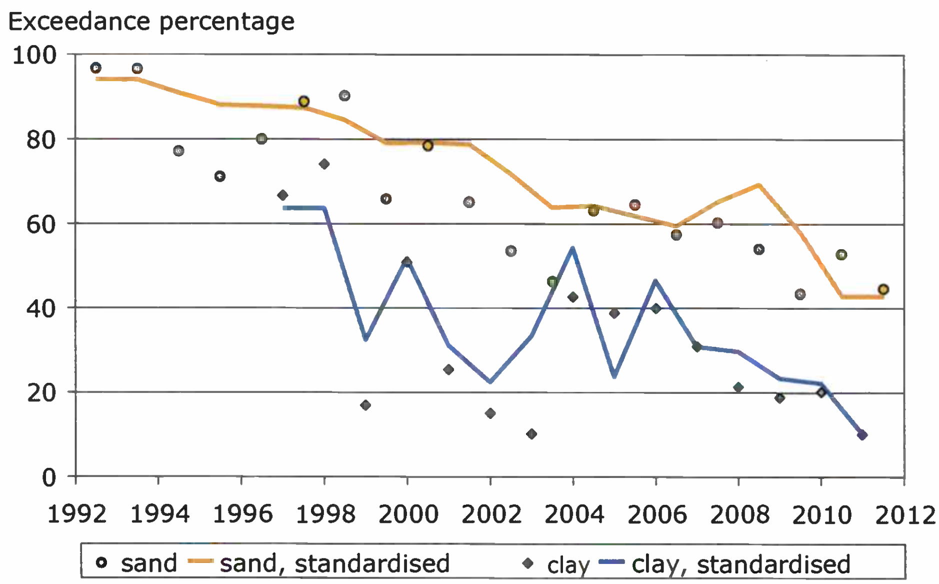


Figure 4.7. Percentage of exceedances of the EU standard of 50 mg/l nitrate in water leaching from root zones onto farms in the Sand and Clay regions in the period 1992-2011. Exceedance based on measured and standardised concentrations.

The following sections contain information by region, in the form of cumulative frequency diagrams and other representations. Although these types of diagram are highly informative, they require some explanation. With the aid of Figure 4.8, this paragraph explains how such a diagram has to be interpreted. It can be determined from the diagram that in the period 2008-2011 some 30% of the monitored arable farms had an average nitrate concentration below the EU standard of 50 mg/l, while 20% of these farms had a concentration above 100 mg/l. Follow the horizontal 50 mg/l line (EU standard, red line) from the y-axis until it intersects the cumulative frequency curve for the period 2008-2011 (squares). Then trace a vertical line downwards from the point of intersection of the 50 mg/l line to the x axis. Where it meets the x-axis is the percentage of farms that had a measured nitrate concentration below 50 mg/l. It is also possible to see from the curve that in this period some 80% of arable farms had an average concentration lower than 100 mg/l, and hence that 20% had a higher concentration. Trace a (vertical) line from the 80% point on the x axis until it intersects the cumulative frequency curve for the period 2008-2011 (squares). Then from the point of intersection, trace a horizontal line to the y-axis. Where it meets the y-axis is the concentration not exceeded by (80% of) farms, 100 mg/l in this example.

4.3.2 *Sand and Loess regions*

Between the first and second monitoring periods, there was a reduction in the nitrate concentrations in the upper groundwater of arable farms. Between the second and subsequent periods, no clear trends are discernible; the nitrate concentrations just hover around the level of the second period (see Figure 4.8). The percentage of arable farms with a period average nitrate concentration below the EU standard rose from 5% in the first period to roughly 30% in the fifth period.

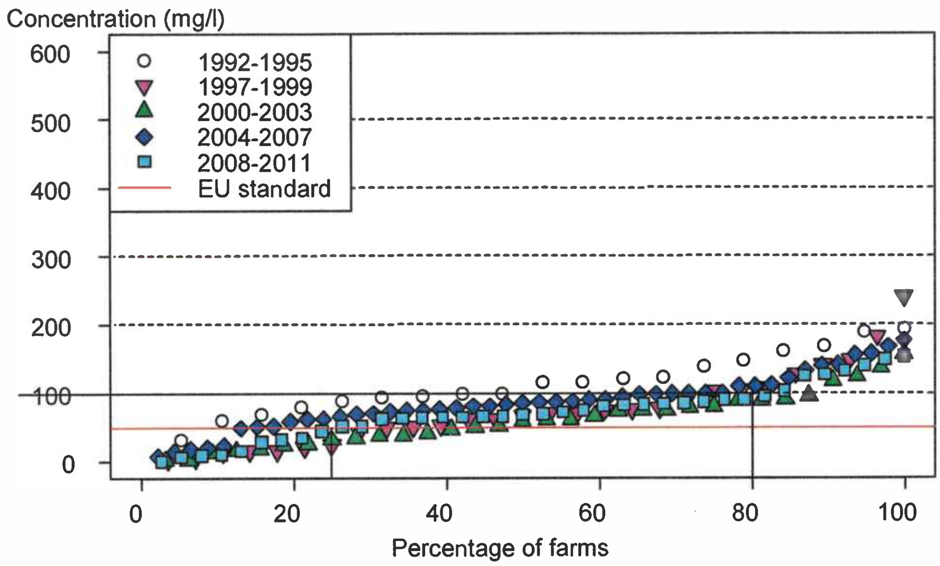


Figure 4.8. Nitrate concentration in water leaching from the root zones of arable farms in the Sand region, shown as a cumulative frequency diagram of the farm average per period.

At dairy farms, nitrate concentrations diminished gradually during the first three periods. They became stable in the fourth period, and then resumed their fall in the fifth period (Figure 4.9). The percentage of dairy farms with a concentration less than the EU standard grew from approximately 5% in the period 1992-1995 to over 60% in the period 2008-2011.

At other livestock farms in the Sand regions, nitrate concentrations decreased between the second and third monitoring periods. Between the third and fourth periods, the concentrations went up (Figure 4.10), but during the fifth period, they dropped to a level comparable to that of the third period. The percentage of other livestock farms with a period average concentration below the EU standard rose from about 5% in the period 1997-1999 to around 35% in the period 2008-2011.

Nitrate concentrations differ between the three areas of the Sand regions: they are higher in Sand South, and lower in Sand Central and Sand North (Figure 4.11). After 1992, the nitrate concentrations fell in all three sand areas, and stabilised after 2002, the same as the fall and stabilisation observed in the sand regions as a whole (Figure 4.4). Sand Central witnessed the largest decline, from 200 mg/l in the period 1992-1993 to 50 mg/l in the period 2008-2011. After monitoring commenced, the average nitrate concentration in Sand South went down from 240 mg/l to 100 mg/l in the period 2004-2011. The nitrate concentrations in Sand North decreased from 150 mg/l to less than 50 mg/l during the period 2008-2011.

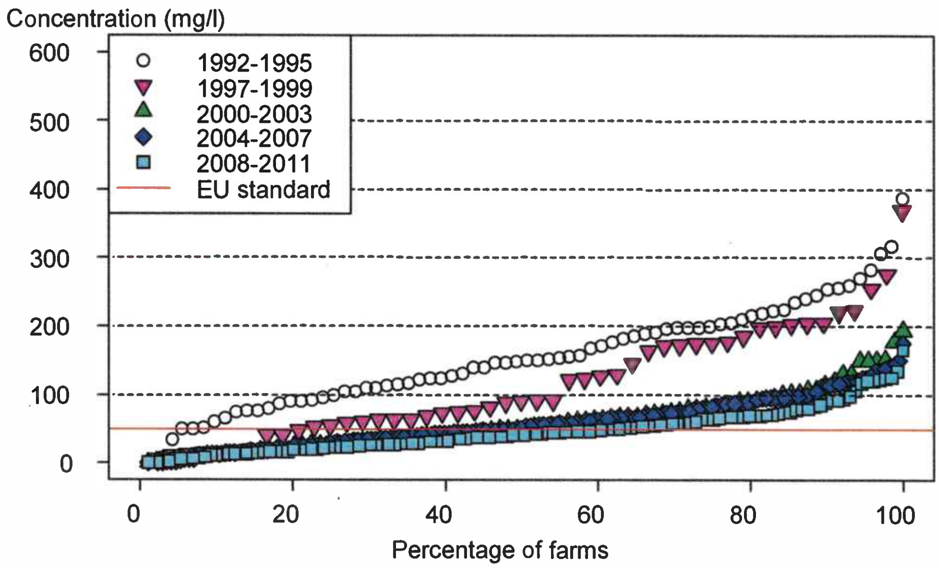


Figure 4.9. Nitrate concentration in water leaching from the root zones of dairy farms in the Sand regions, shown as a cumulative frequency diagram of the farm average per period.

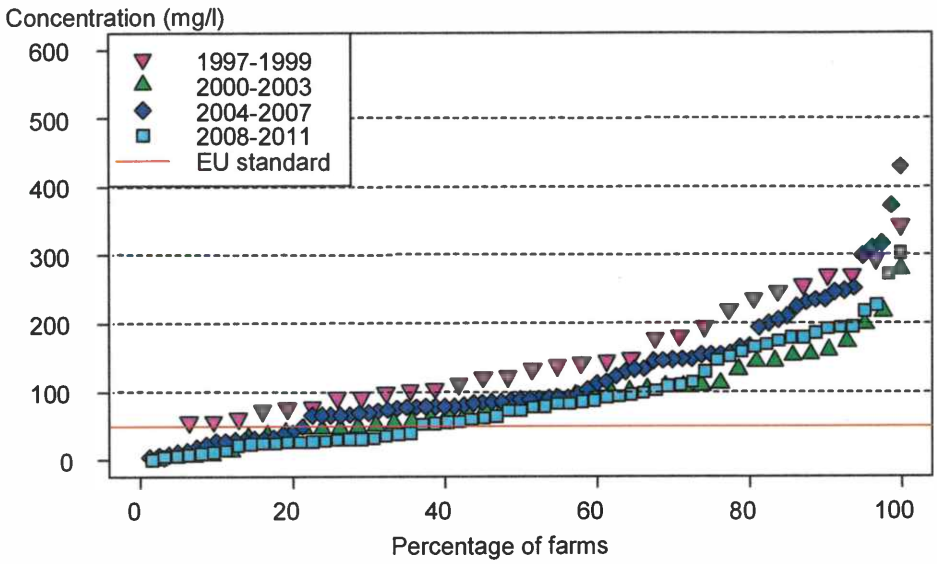


Figure 4.10. Nitrate concentration in water leaching from the root zones of other livestock farms in the Sand regions, shown as a cumulative frequency diagram of the farm average per period.

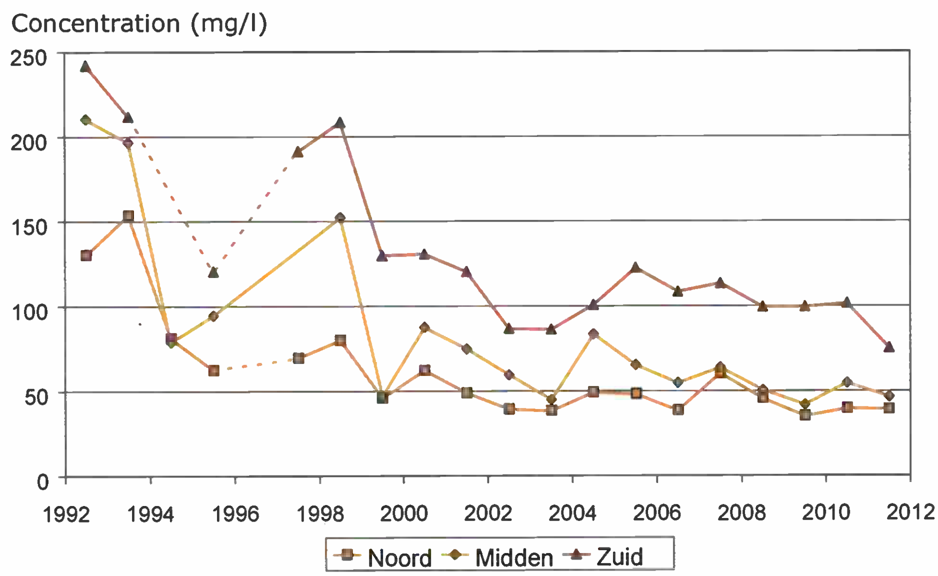


Figure 4.11. Nitrate concentrations (annual average of measured concentrations) in water leaching from root zones onto farms in the Sand North, Sand Central and Sand South areas in the period 1992-2011.

The trend of the nitrate concentrations in the Loess region, measured as part of the Soil Moisture Monitoring Network (BVM) of the province of Limburg, is comparable to that shown by the LMM farms in the sand regions (see Figure 4.12). The concentrations at the LMM farms in the Loess region are certainly higher than those measured in the BVM. The cause of the discrepancy between the BVM data and the LMM data could be a combination of unit-scale differences (farm versus plot) and different proportions of the acreage for the various crop types in both monitoring networks. Willems and Fraters (1995) showed that the scale used for presenting the monitoring results affects the percentage exceedance of the standard, even if the total average nitrate concentration is the same.

The percentage of agricultural plots in the BVM with a nitrate concentration below the EU standard increased from about 10% in the period 1996-1999 to about 40% in the period 2004-2006 (see Figure 4.13). The percentage of LMM farms with a concentration below the EU standard was in the range 10%-20% in the third and fourth periods (Figure 4.14). During the fifth period, the percentage climbed to 35%.

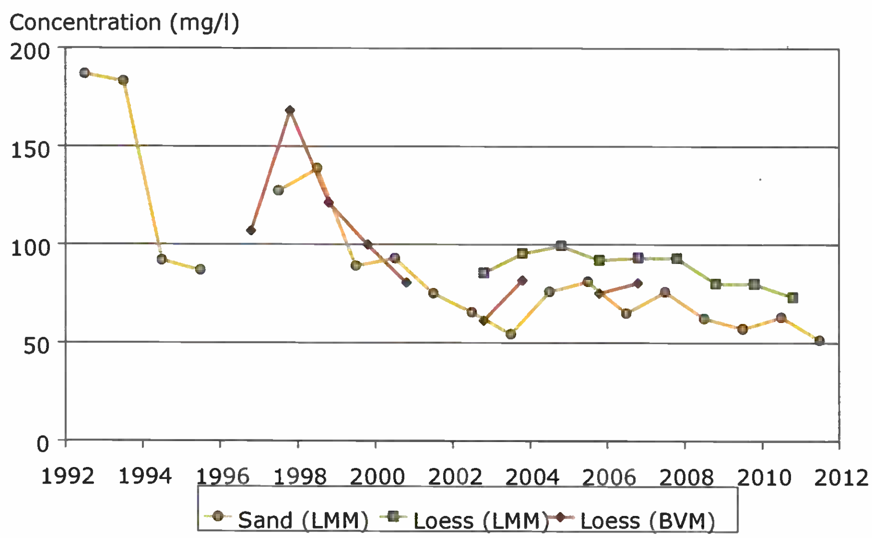


Figure 4.12. Nitrate concentration in water leaching from root zones in the Sand regions (LMM) and Loess region (BVM plots and LMM farms) in the period 1992-2011.
Source: RIVM (sand / LMM loess); Province of Limburg (BVM loess).

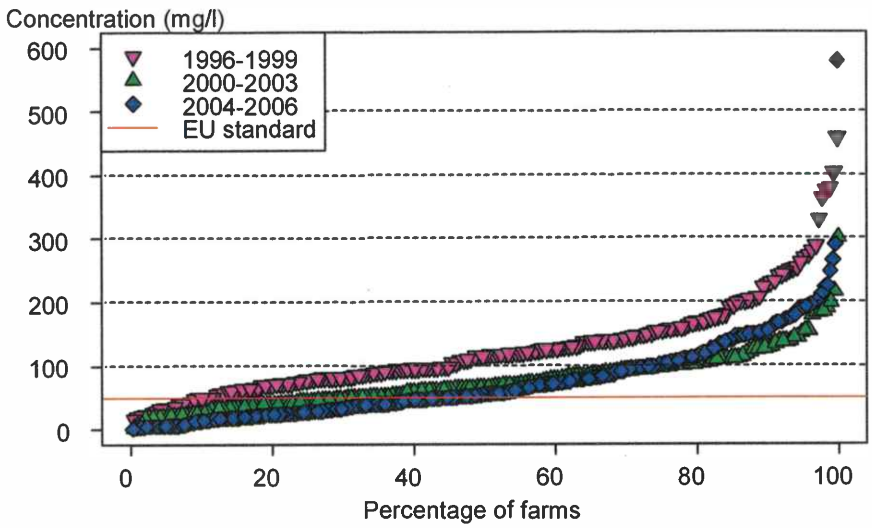


Figure 4.13. Nitrate concentration in water leaching from the root zones of BVM plots used for farming in the Loess region, shown as a cumulative frequency diagram of the plot average per period.
Source: Province of Limburg

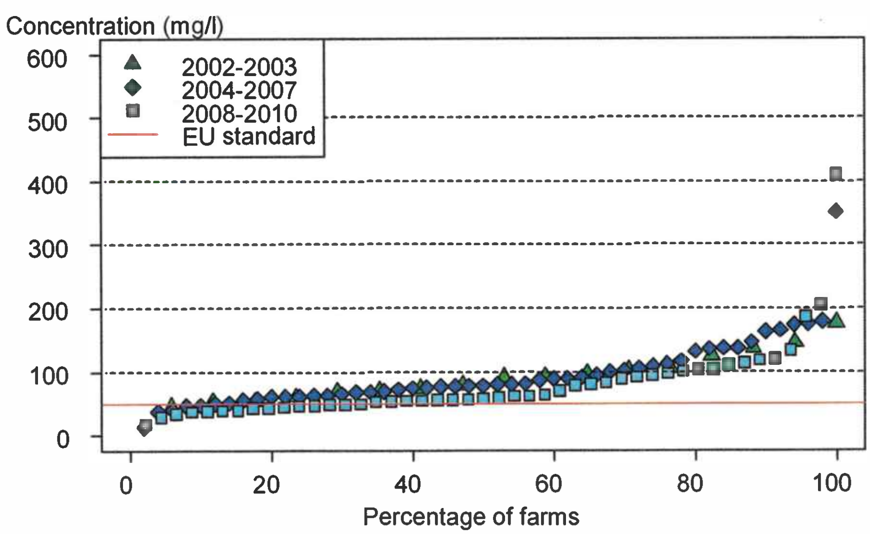


Figure 4.14. Nitrate concentration in water leaching from the root zones of LMM farms in the Loess region, shown as a cumulative frequency diagram of the farm average per period.
Source: RIVM.

4.3.3 *Clay regions*

On arable farms in Clay regions, the nitrate concentrations in water leaching from root zones did not change between 1997-2003, but did increase in the period 2004-2007, only to return in the period 2008-2011 to the previous level (Figure 4.15). The percentages of arable farms with a nitrate concentration less than the EU standard were 55% and 70% respectively in the second and third periods. This percentage dropped to around 40% in period four, but climbed in period five to almost 80% (see Figure 4.15). Nitrate concentrations at dairy farms did not change as much from 1997 to 2011, the percentage of them that did not exceed the EU standard being in the range 70% to 90% (see Figure 4.16).

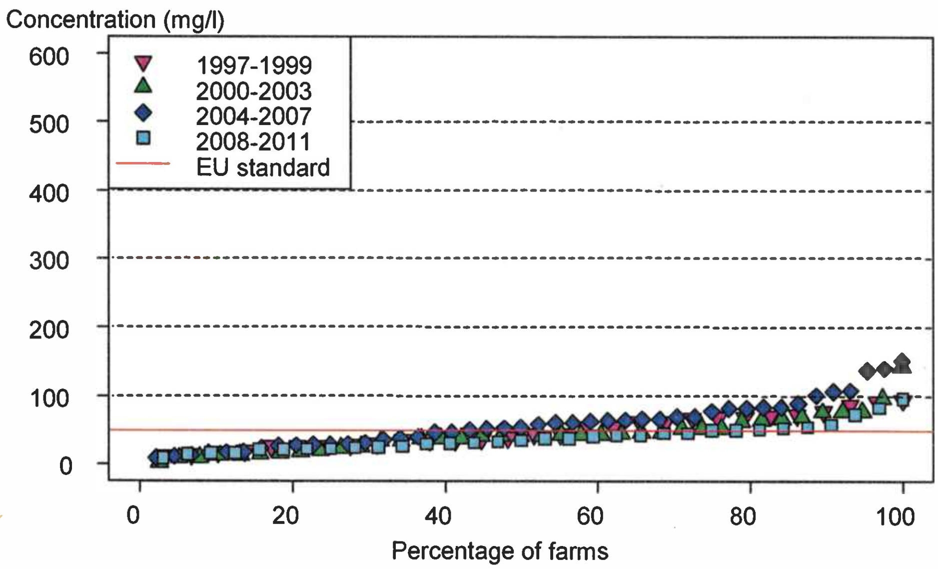


Figure 4.15. Nitrate concentration in water leaching from the root zones of arable farms in the Clay regions, shown as a cumulative frequency diagram of the farm average per period.

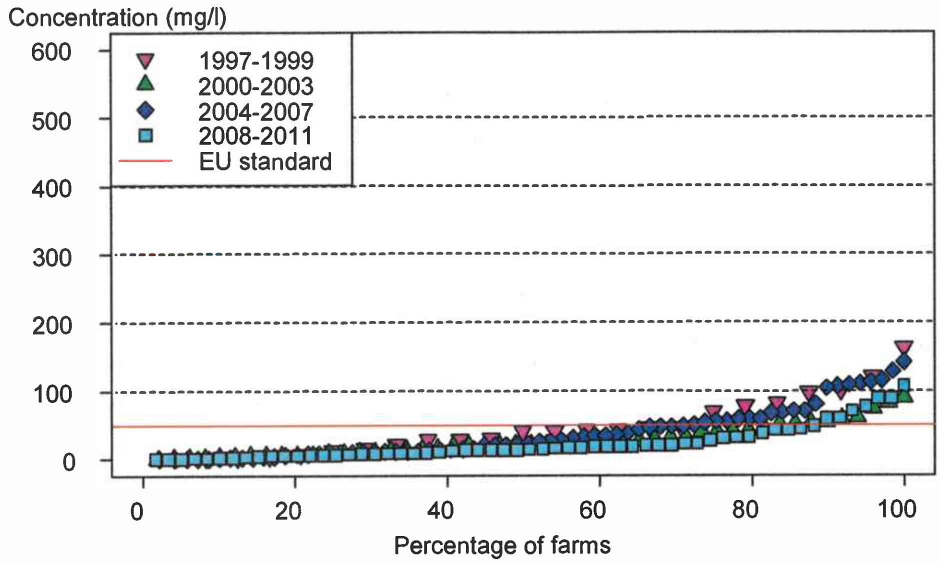


Figure 4.16. Nitrate concentration in water leaching from the root zones of specialised dairy farms in the Clay regions, shown as a cumulative frequency diagram of the farm average per period.

4.3.4 *Peat regions*

The average nitrate concentrations in water that leached from root zones were usually below 25 mg/l for dairy farms in the Peat regions (Figure 4.17). There were only isolated cases of the EU standard of 50 mg/l being exceeded in the period 2004-2007, with 90% to 100% of the farms meeting the standard. The average nitrate concentrations in ditchwater were mostly below 10 mg/l (see Figure 4.18), and no exceedances of the EU standard of 50 mg/l occurred during any of the monitoring periods.

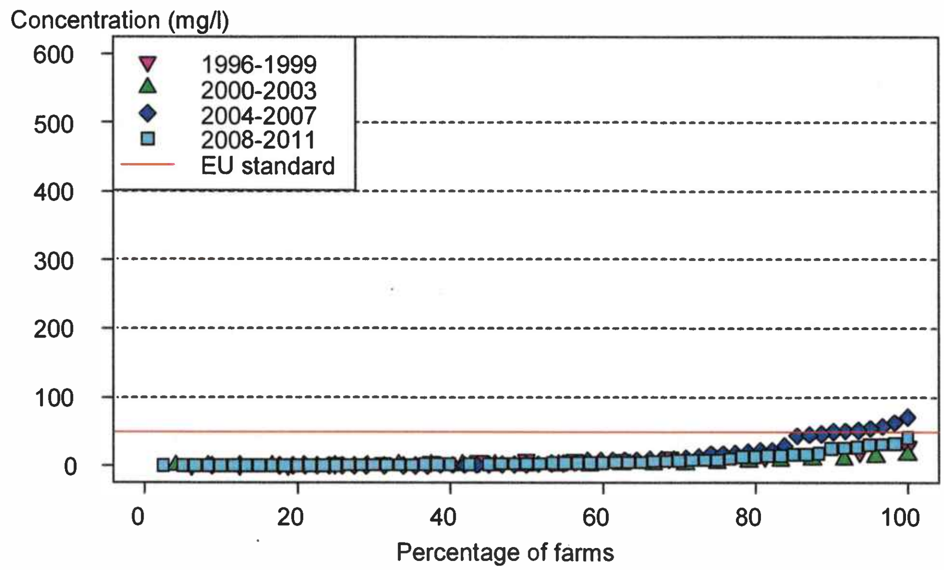


Figure 4.17. Nitrate concentration in water leaching from the root zones of dairy farms in the Peat regions, shown as a cumulative frequency diagram of the farm average per period.

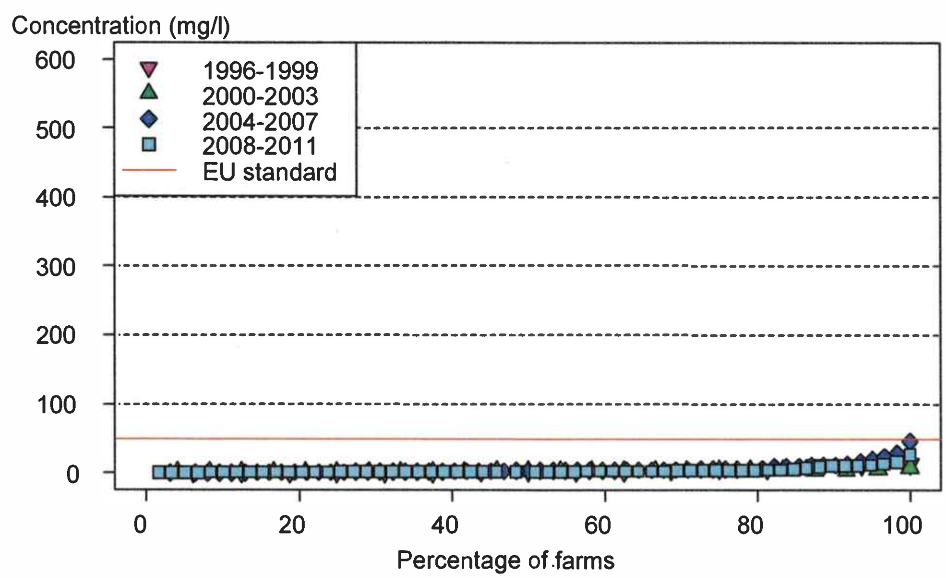


Figure 4.18. Nitrate concentration in ditchwater of dairy farms in the Peat regions in winter, shown as a cumulative frequency diagram of farm average per period.

4.4 Relationship between trends in agricultural practice and nitrate concentration

The nitrogen surplus decreased in the period 1991-2010. At arable farms and other livestock farms, however, there was in most cases a nitrogen surplus in the latest period (2007-2010) that was slightly higher than in the period before (2003-2006). In general, there was a decrease in the application of inorganic fertiliser at LMM farms between 1991 and 2010. There was also a reduction in the quantity of livestock manure at most dairy farms. At arable farms and other livestock farms, there was mostly an increase in the application of livestock manure.

The effects of the above are clearly visible in the decline of nitrate concentrations during the period 1992-2011, especially at dairy farms. Although the nitrate concentration at arable farms and other livestock farms has certainly fallen slightly compared with the initial period, it seems to have stabilised in the past few years.

Depending on the hydrogeological conditions, the impact of changes in agricultural practice on the leaching from root zones becomes detectible in three to five years.

In addition to the effects of measures adopted for agricultural practice, factors such as variation in weather conditions, climate and hydrogeological processes also influence the increase or decrease in nitrate leaching. Such factors can hide the effects of measures on water quality.

4.5 References

Boumans, L.J.M., Fraters, B. and Van Drecht, G. (2001). Nitrate in the upper groundwater of 'De Marke' and other farms. Netherlands Journal of Agricultural Science, 49, (2-3), pp. 163-177.

Boumans, L.J.M., Van Drecht, G., Fraters, B., De Haan, T., De Hoop, D.W. (1997). Effect van neerslag op nitraat in het bovenste grondwater onder landbouwbedrijven in de zandgebieden; gevolgen voor de inrichting van het Monitoringnetwerk effecten mestbeleid op Landbouwbedrijven (MOL). Bilthoven, RIVM Report 714831002.

Fraters, B., Hotsma, P., Langenberg, V., Van Leeuwen, T., Mol, A., Olsthoorn, C.S.M. (2004). Agricultural practice and water quality in the Netherlands in the 1992-2002 period. Bilthoven, RIVM Report 500003002.

Meinardi, C.R. (2005). Stromen van water en stoffen door de bodem en naar sloten in de Vlietpolder. Bilthoven, RIVM Report 500003004.

Van der Grift, B. (2003) Samenstelling grondwater Vlietpolder. Utrecht, NITG-TNO Report 005.63034.

Willems, W.J. and Fraters B. (1995). Naar afgestemde kwaliteitsdoelstellingen voor nutriënten in grondwater en oppervlaktewater, discussienotitie. Bilthoven, RIVM Report 714901003.

5 Groundwater quality

5.1 Introduction

The nitrate concentration in Netherlands' groundwater shows a wide variation spatially as well as in terms of depth. Spatial variation is only partly accounted for by changes in land use and nitrogen emissions. Other causes are the year-to-year changes in net precipitation, soil type, and the geohydrological characteristics of aquifers (see also chapter 4).

In general, the nitrate concentration in groundwater is low under peat soil, relatively high under sand soil, and average under clay soil (Van Vliet et al., 2010, Reijnders et al., 2004). Nitrate concentration usually decreases with an increase in the depth of the groundwater. This is caused by the reduction in nitrate concentration during transport (denitrification), the mixing of waters of different ages, and the lateral transport due to the presence of resistant layers that partially or completely inhibit downward movement.

This chapter comprises three parts, each dealing with one of the three depths at which groundwater is monitored in the Netherlands: 5-15 metres, 15-30 metres and more than 30 metres. Chapter 4 deals with the top metre of the groundwater.

5.2 Nitrate in groundwater at a depth of 5–15 metres

In the period 1992-2010, the nitrate concentration in groundwater at a depth of 5-15 metres below the surface for Dutch farm soil was on average 24 mg/l, with a range of 20-28 mg/l (Figure 5.1). The highest concentration was measured in 1996, about ten years after the peak in nitrogen surplus on the national nitrogen balance (Figure 3.3). In 2008, there was a strikingly low average nitrate concentration for farm soil. This is attributable to two wells that had high nitrate concentrations (around 150 mg/l) for almost the entire monitoring period, but where precisely in 2008 virtually no nitrate was detected. Validation indicates no question of extreme values (a low value had been measured before), and there is no evidence of a measuring mistake.

For nature reserves and areas used for other purposes (including orchards and urban areas), the concentration had an average value of about 13 mg/l and fluctuated between 10 and 21 mg/l (Figure 5.1). From 2001 to 2009, it was especially changeable regarding other land use. The sudden rising and falling is almost entirely due to one well where, between 2001 and 2009, the nitrate concentration climbed from less than 30 mg/l to almost 500.

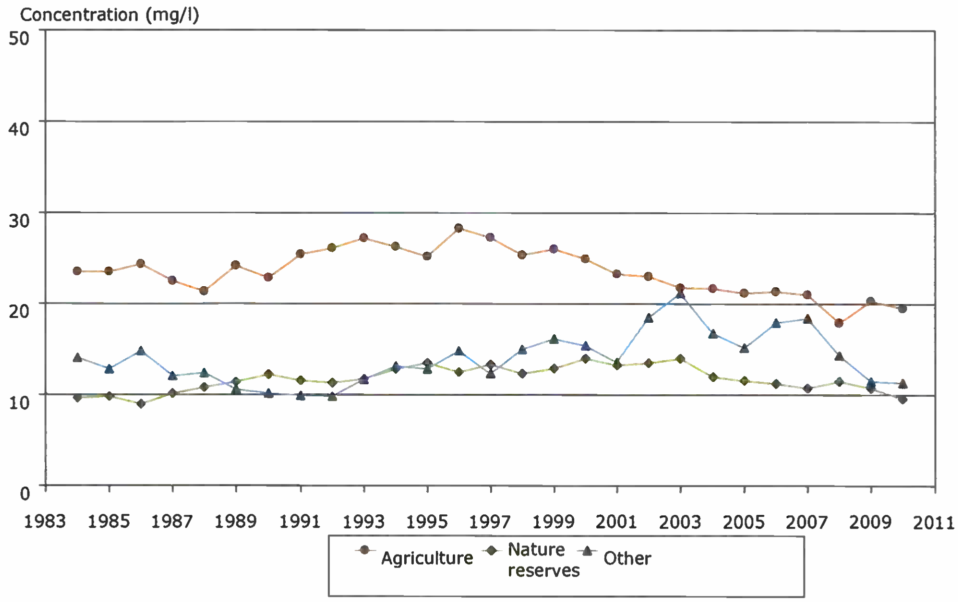


Figure 5.1. Average annual nitrate concentration (mg/l) in groundwater at a depth of 5-15 metres below the surface in the Netherlands by land use type for the period 1984-2010.

The nitrate concentration in groundwater originating from farming on sand soil (30 to 45 mg/l) was higher than in clay soil and peat soil (< 10 mg/l and < 5 mg/l respectively; Figure 5.2). Prior to 1992, concentrations were mostly below 40 mg/l, whereas in the period 1992-2000, concentrations hovered between 42 and 47 mg/l. After 2001, the average nitrate concentration remained below 40 mg/l, gradually falling to 33 mg/l in 2010.

Between 2008 and 2010, the EU standard of 50 mg/l for nitrate was exceeded at 10% of the groundwater monitoring wells at a depth of 5 to 15 metres. For agricultural areas, the figure was 12%; for nature reserves, about 5%; and for other areas about 9% (Figure 5.3 and Table 5.1). There were slight variations from year to year.

The EU standard was exceeded at 19% of the monitoring sites in farming areas on sand soil, whereas there were no more exceedances in the clay and peat regions (Figure 5.4).

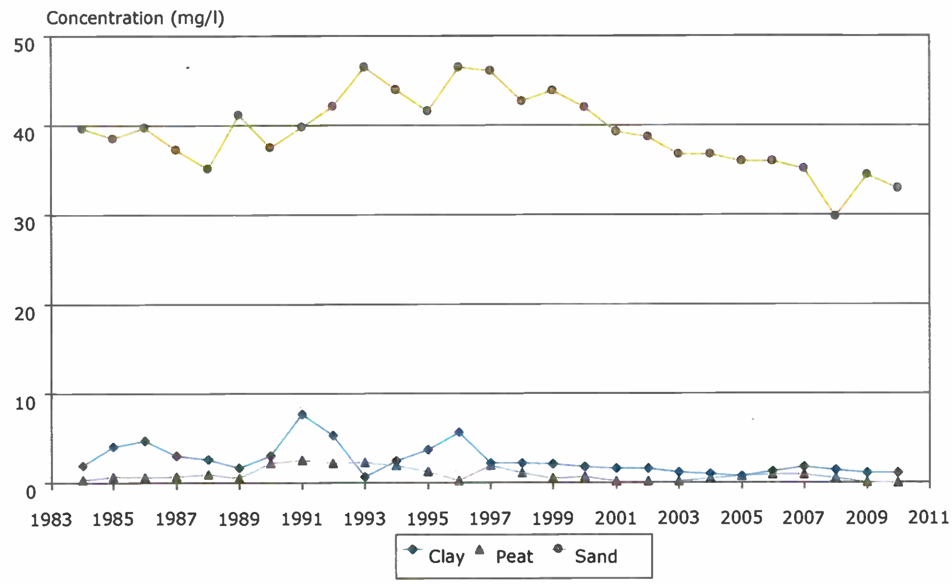


Figure 5.2. Average annual nitrate concentration (mg/l) in groundwater at a depth of 5-15 metres below the surface in agricultural areas by soil type for the period 1984-2010.

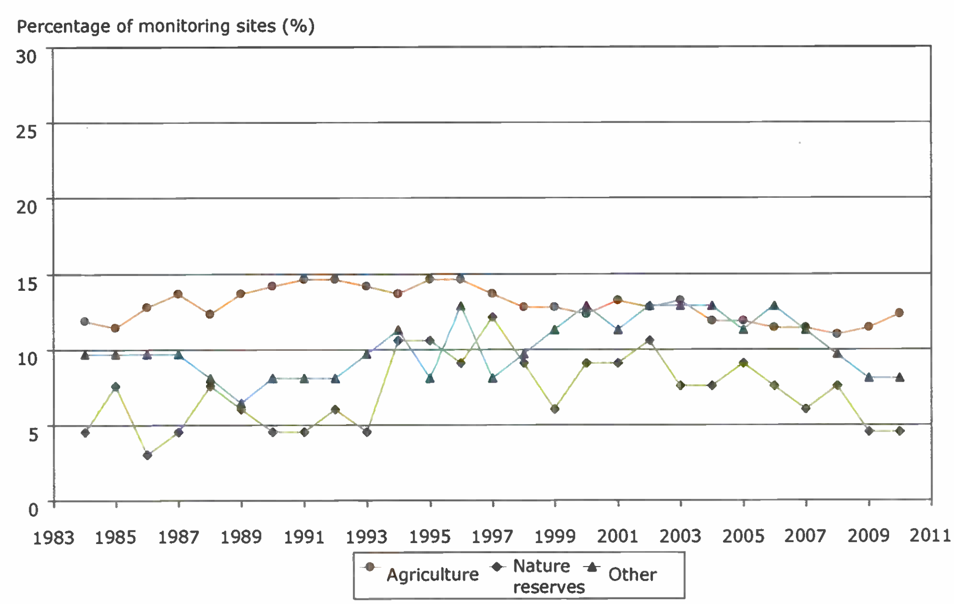


Figure 5.3. Exceedance of the EU standard of 50 mg/l for nitrate in groundwater at a depth of 5-15 metres below the surface by land use type for the period 1984-2010.

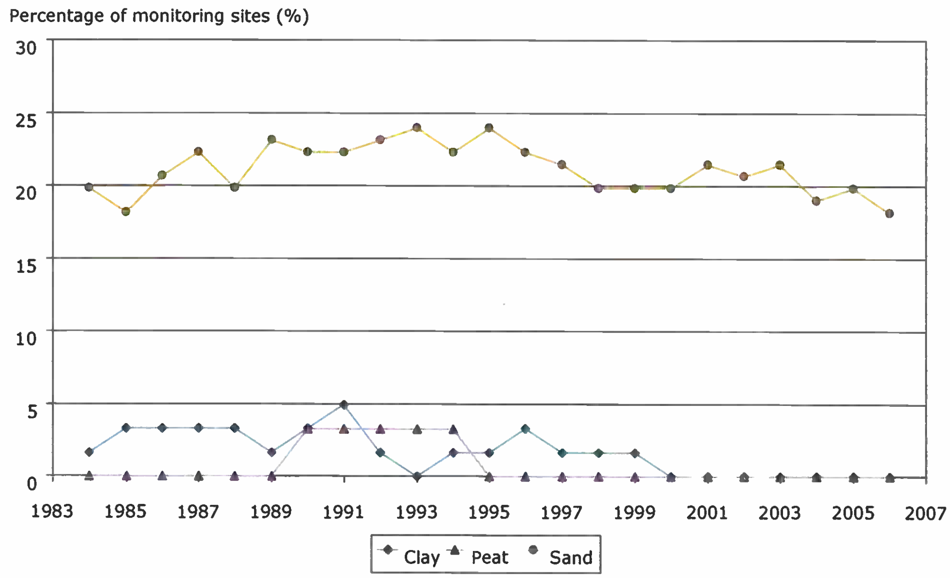


Figure 5.4. Exceedance of the EU standard of 50 mg/l for nitrate in groundwater in agricultural areas at a depth of 5-15 metres below the surface for the period 1984-2010.

Table 5.1. Nitrate in groundwater at a depth of 5-15 metres for the period 1992-2010 (%)[1].

	All monitoring sites			Monitoring sites in agricultural areas		
Concentration	1992-1995	2004-2007	2008-2010	1992-1995	2004-2007	2008-2010
0 - 15 mg/l	79.0	81.8	83.0	80.4	82.6	84.0
15 - 25 mg/l	3.7	2.9	2.3	1.8	1.8	1.8
25 - 40 mg/l	2.0	2.6	3.5	0.5	2.7	1.4
40 - 50 mg/l	2.6	1.7	1.2	1.8	0.5	0.9
> 50 mg/l	12.7	11.0	10.1	15.5	12.3	11.9
Number of monitoring sites	347	347	347	219	219	219

[1] Percentage of monitoring sites with a period average within a given concentration range for all monitoring sites and for monitoring sites with water mainly affected by agriculture. The total percentage could exceed 100 because of rounding.

Most monitoring sites (about 70%) showed no change in nitrate concentration between reporting periods (1992-1995, 2004-2007 and 2008-2010;Table 5.2). The number of sites where a change occurred decreased slightly, mainly because of a reduction in the number of sites showing a fall. The pattern of differences between the first and fourth periods is comparable to that between the fourth and fifth, in that the percentage of sites showing a fall is greater than the percentage showing a rise.

Table 5.2. Change in nitrate concentration in groundwater at a depth of 5-15 metres for the period 1992-2010 (%)[1]

Concentration	All monitoring sites		Monitoring sites in agricultural areas	
	1992-1995/ 2004-2007	2004-2007/ 2008-2010	1992-1995/ 2004-2007	2004-2007/ 2008-2010
Large increase (% > 5 mg/l)	8.9	4.9	7.8	5.5
Small increase (% 1-5 mg/l)	3.5	3.7	3.2	3.2
Stable (% ± 1 mg/l)	66.3	70.9	72.1	72.6
Small decrease (% > 1-5 mg/l)	4.3	6.9	1.8	6.4
Large decrease (% > 5 mg/l)	17.0	13.5	15.1	12.3
Number of monitoring sites	347	347	219	219

[1] Percentage of monitoring sites with given size of change in concentration between the first and fourth, and the fourth and fifth reporting periods. The table shows the data of all monitoring sites, as well of monitoring sites with water mainly affected by agriculture. The total percentage could exceed 100 because of rounding.

Of the three sand areas, North, Central and South (Figure 5.5, unbroken lines), the nitrate concentration is clearly the highest in Sand South (around 65 mg/l), lower in Sand Central (about 25 mg/l) and the lowest in Sand North (slightly over 10 mg/l). In these areas, other soil types also occur. If only the monitoring sites on sandy soil are considered (Figure 5.5, broken lines), the nitrate concentrations are slightly higher. Moreover, Sand South has the most wells where the EU standard is exceeded (Figure 5.6).

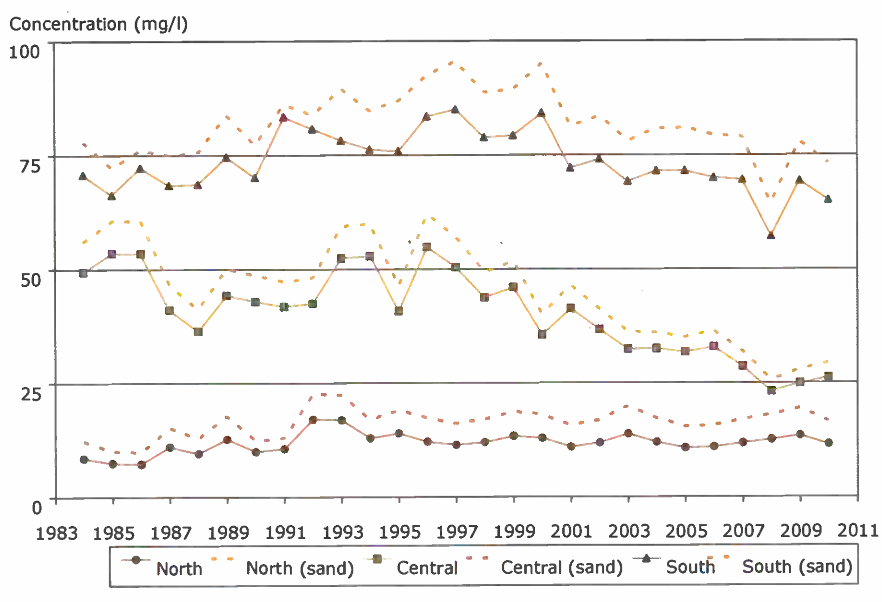


Figure 5.5. Nitrate in groundwater at a depth of 5-15 metres below the surface in agricultural areas in Sand North, Sand Central and Sand South (unbroken lines) and from farming on sandy soil within these areas (dotted lines).

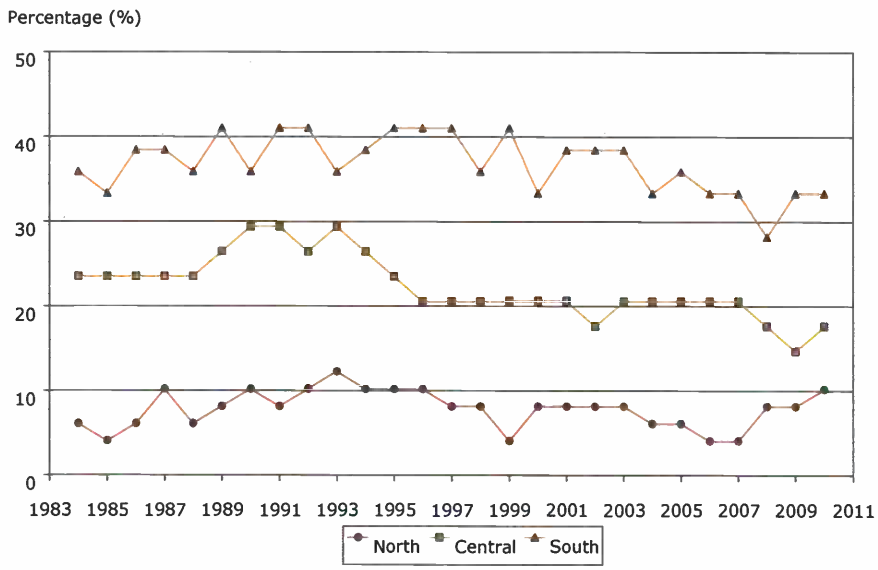
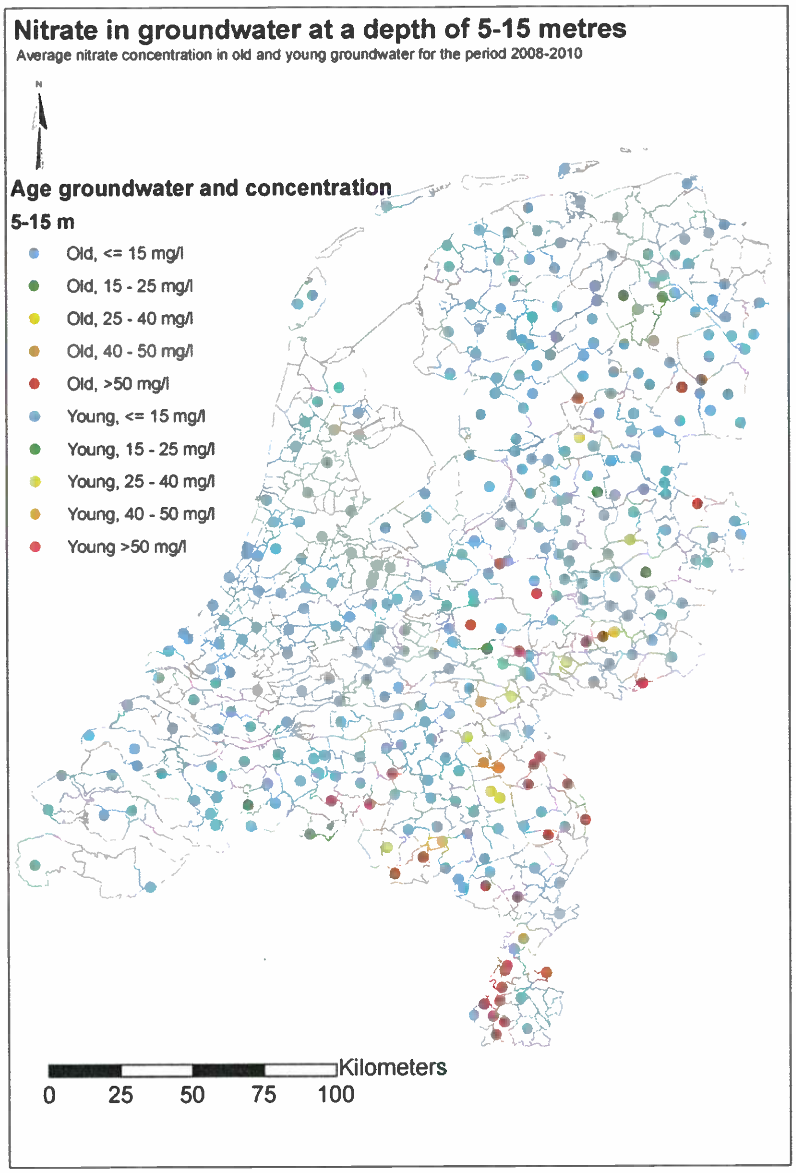


Figure 5.6. Exceedance of the EU standard of 50 mg/l for nitrate in groundwater at a depth of 5-15 metres below the surface in the areas Sand North, Sand Central and Sand South for the period 1984-2010.

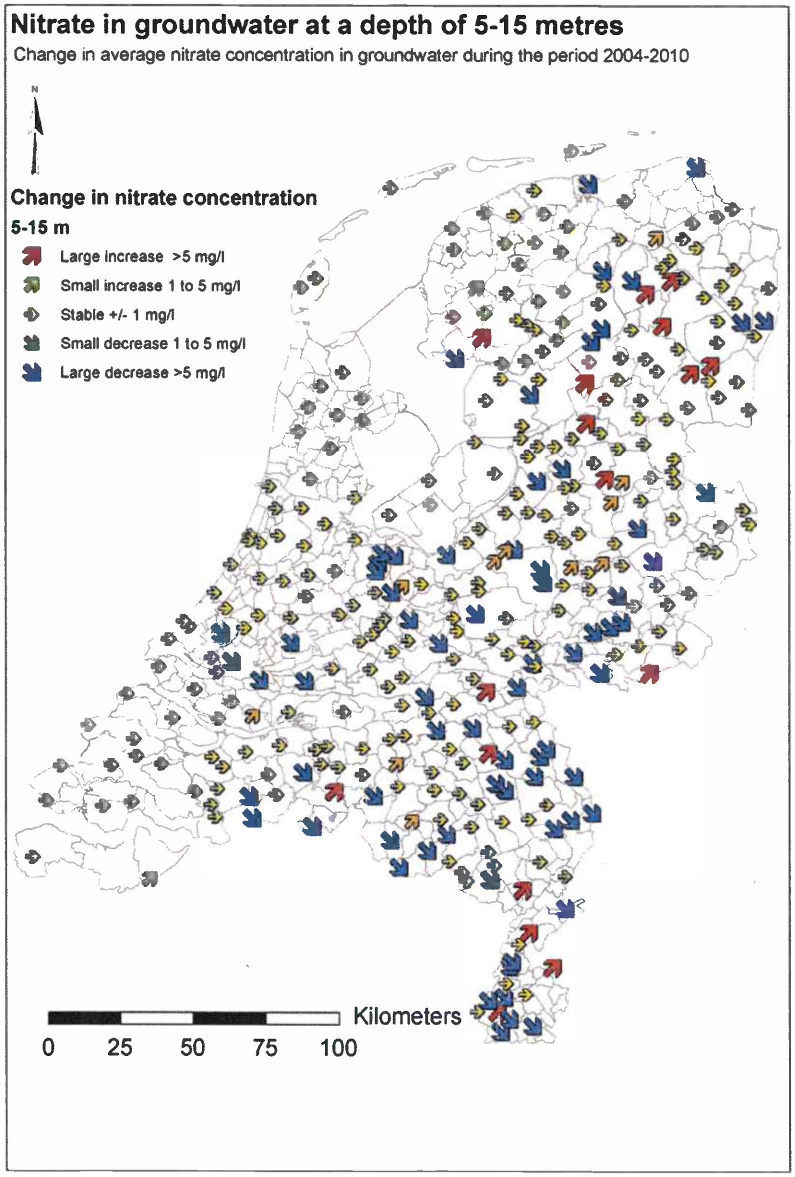
Starting in the mid-1990s, the nitrate concentration in groundwater between 5 and 15 metres below the surface in Sand South fell from about 85 mg/l to 65 mg/l. In Sand Central, the nitrate concentration fell from 55 mg/l to 25 mg/l. This decline also continued in the latest monitoring years, particularly in Sand South. Similar to the trend in nitrate in farming areas on sandy soil (Figure 5.2), nitrate concentration shows the same distinct dip in 2008. In Sand North, the nitrate concentration was reasonably stable during the monitoring period.

Map 5.1 shows the average nitrate concentration of all monitoring sites with monitoring depths of 5 to 15 m, for the period 2008-2010. The monitoring sites are divided into those with wells containing old groundwater (> 25 years) and young groundwater (< 25 years). In the wells containing old groundwater, this water is usually from confined or semi-confined aquifers, whereas in those containing young groundwater, it is from phreatic aquifers. High nitrate concentrations (> 50 mg/l) were found in young groundwater in the sand and loess regions (the eastern and southern parts of the Netherlands).

The change in nitrate concentration between the 2004-2007 and 2008-2010 periods is shown on Map 5.2. Most changes occurred in the sand and loess regions, with increases as well as decreases in nitrate concentrations being found.



Map 5.1. Average nitrate concentration in groundwater at a depth of 5-15 metres for the period 2008-2010. "Young" refers to groundwater younger than 25 years; "Old" means older than 25 years.



Map 5.2. Change in average nitrate concentration in groundwater at a depth of 5-15 metres for the period 2004-2010. Change shown here is the difference between the averages for the 2004-2007 and 2008-2010 periods.

5.3 Nitrate in groundwater at a depth of 15-30 metres

Until 1998, nitrate concentration at a depth of 15-30 metres was highest under agricultural land, followed by land used for other purposes and nature reserves (Figure 5.7). After 1998, the nitrate concentration in land used for other purposes rose substantially, so that it became more than in agricultural areas. This increase cannot be explained. Following a dip after 2002, the concentration in land used for other purposes seems to have stabilised in recent years, the average settling at 9 mg/l. In the case of agricultural areas, the average nitrate concentration is roughly 5 mg/l; for nature reserves, 3 mg/l.

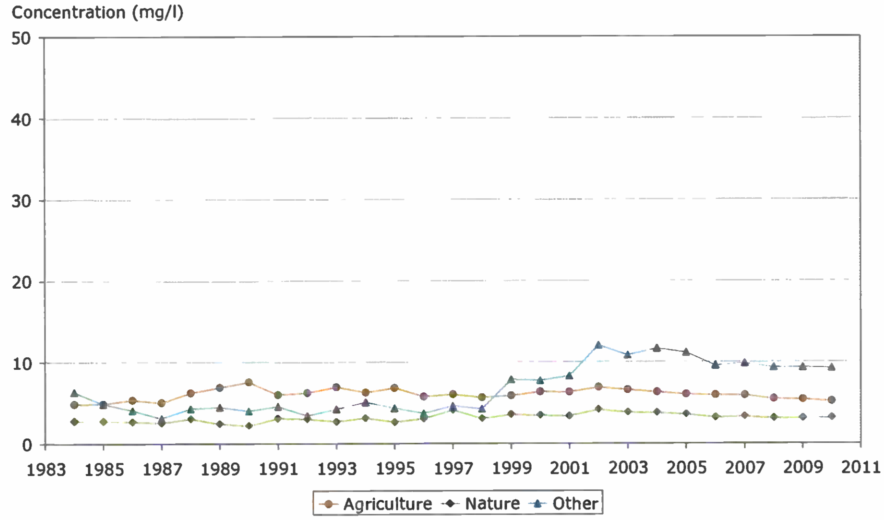


Figure 5.7. Average annual nitrate concentration (mg/l) in groundwater at a depth of 15–30 metres by land use type for the period 1984-2010.

The nitrate concentration in groundwater originating from farming on sandy soil is higher than the concentration under clay or peat soil, where virtually no nitrate is detected at a depth of 15-30 metres (Figure 5.8). After 2002, the nitrate concentration relating to agriculture on sandy soil fell from 11 mg/l to approximately 7 mg/l in 2010.

Between 2008 and 2010, the EU standard of 50 mg/l for nitrate was exceeded at 3% of the groundwater monitoring wells at a depth of 15 to 30 m. For agricultural areas, the figure was 3%; for nature reserves, 2%; and for other areas, 5% (Figure 5.9 and Table 5.3). There were slight variations from year to year.

The percentage of monitoring sites in agricultural areas on sandy soil where the EU standard for nitrate was exceeded shrank from 7% to 3%. During the same monitoring period, the percentage in the clay and peat regions was only 1% (Figure 5.10).

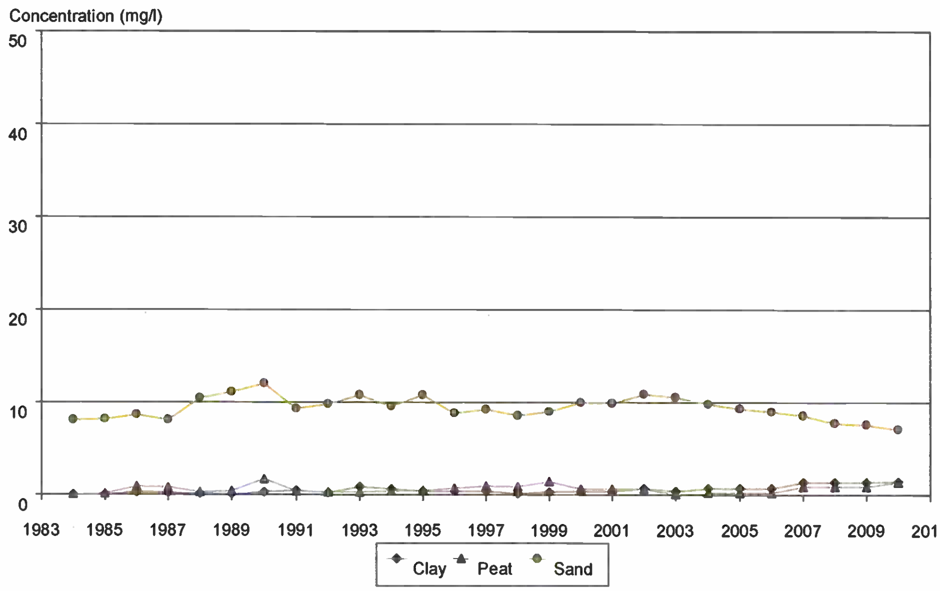


Figure 5.8. Average annual nitrate concentration (mg/l) in groundwater at a depth of 15-30 metres in agricultural areas by soil type for the period 1984-2010.

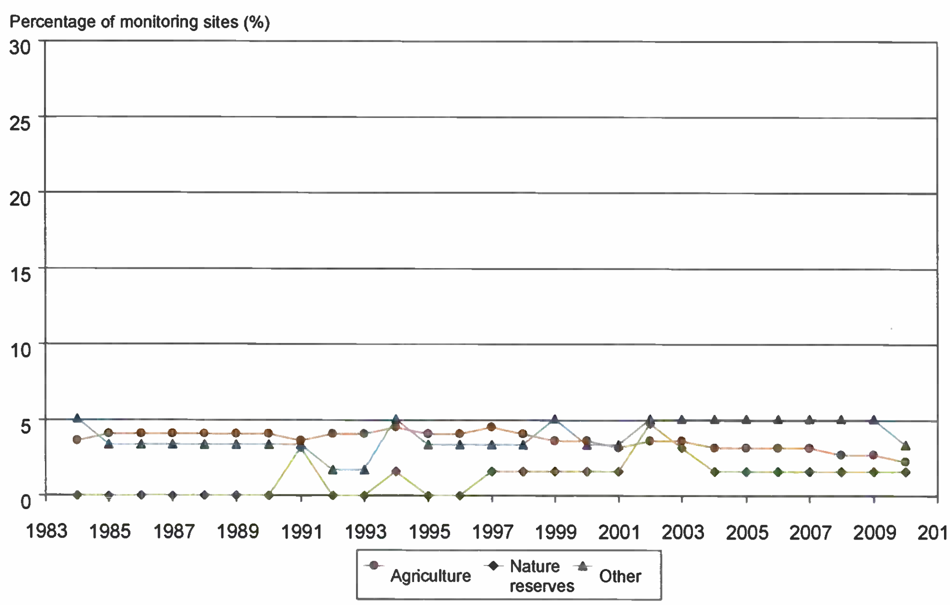


Figure 5.9. Exceedance of the EU standard of 50 mg/l for nitrate in groundwater at a depth of 15-30 metres by land use type for the period 1984-2010.

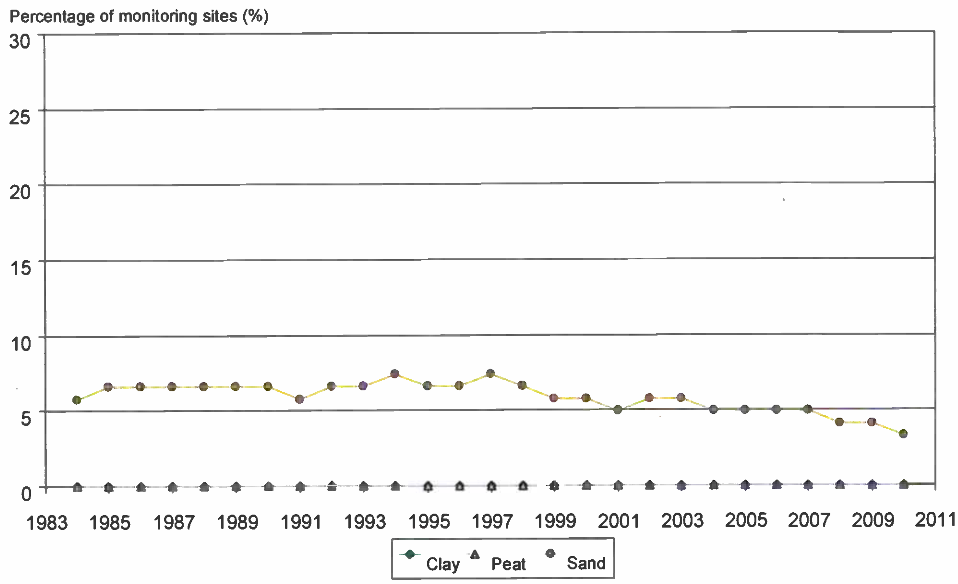


Figure 5.10. Exceedance of the EU standard of 50 mg/l for nitrate in groundwater at a depth of 15-30 metres below farming areas in the period 1984-2010.

Table 5.3. Nitrate in groundwater at a depth of 15-30 metres for the period 1992-2010 (%)[1].

Concentration	All monitoring sites			Monitoring sites in agricultural areas		
	1992-1995	2004-2007	2008-2010	1992-1995	2004-2007	2008-2010
0 - 15 mg/l	94.3	93.5	92.9	93.8	93.8	92.8
15 - 25 mg/l	0.9	1.2	2.1	-	1.0	1.9
25 - 40 mg/l	0.9	1.5	1.8	1.4	1.4	1.9
40 - 50 mg/l	0.6	0.9	0.9	0.5	1.0	1.4
> 50 mg/l	3.3	3.0	2.4	4.3	2.9	1.9
Number of monitoring sites	336	336	336	209	209	209

[1] Percentage of monitoring sites with a period average within a given concentration range for all monitoring sites and for monitoring sites with water mainly affected by agriculture. The total percentage could exceed 100 because of rounding.

Most monitoring sites (over 80%) showed no change in nitrate concentration between reporting periods (1992-1995, 2004-2008 and 2008-2010;Table 5.4). The number of sites with an increase was slightly higher than the number showing a decrease. The number of stable sites grew between periods, as the number showing an increase and the number showing a decrease both declined between periods.

Table 5.4. Change in nitrate concentration in groundwater at a depth of 15-30 metres in the period 1992-2010 (%)[1].

Concentration	All monitoring sites		Monitoring sites in agricultural areas	
	1992-1995/ 2004-2007	2004-2007/ 2008-2010	1992-1995/ 2004-2007	2004-2007/ 2008-2010
Large increase (% > 5 mg/l)	6.3	2.4	4.8	2.9
Small increase (% 1-5 mg/l)	3.3	6.0	3.8	5.7
Stable (% ± 1 mg/l)	83.0	86.0	83.3	86.6
Small decrease (% > 1-5 mg/l)	4.8	1.2	5.3	1.0
Large decrease (% > 5 mg/l)	2.7	4.5	2.9	3.8
Number of monitoring sites	336	336	209	209

[1] Percentage of monitoring sites with given rate of change in concentration between the first and fourth, and between the fourth and fifth reporting periods. The table shows the data of all monitoring sites, as well of monitoring sites with water mainly affected by agriculture. The total percentage could exceed 100 because of rounding.

Nitrate concentrations were determined separately for each of the Sand North, Sand Central and Sand South sand areas (Figure 5.11). Something striking is that, in contrast to the measuring results for groundwater 5-15 m below the surface, the nitrate concentration in deeper water is highest in Sand Central. The average nitrate concentration at 15-30 metres in the sand areas is entirely determined by a limited number of wells with a high nitrate concentration (Table 5.5).

Table 5.5. Number of wells at a depth of 15-30 metres below the surface in agricultural areas with sandy soil by nitrate concentration class.

Nitrate concentration class (nitrate in mg/l)	Sand North	Sand Central	Sand South
<1 mg/l	41	25	32
1 to 10 mg/l	3	4	5
>10 mg/l	4	5	1
Total number of wells	48	34	38

For the above reason, the average concentration can give a distorted impression, because it is largely dependent on chance. However, it certainly is the case that high nitrate concentrations were recorded at more locations in Sand Central than in Sand North or Sand South, as shown by Map 5.3. This map depicts all the deep screens of LMG, including those in areas designated as nature reserves or for other purposes, and for all soil types.

Map 5.3 shows the average nitrate concentration of all monitoring sites with monitoring depths of 15 to 30 m, for the period 2008-2010. The monitoring sites are divided into those with wells containing old groundwater (> 25 years) and young groundwater (< 25 years). In the wells containing old groundwater, the water is usually from confined or semi-confined aquifers, whereas in those containing young groundwater, it is from phreatic aquifers. High nitrate concentrations (> 50 mg/l) were found in young groundwater in the sand and loess regions (the eastern and southern parts of the Netherlands). The change in nitrate concentration between the 2004-2007 and 2008-2010 periods is shown in Map 5.4. Most changes occurred in the sand and loess regions, with increases as well as decreases in nitrate concentrations being found.

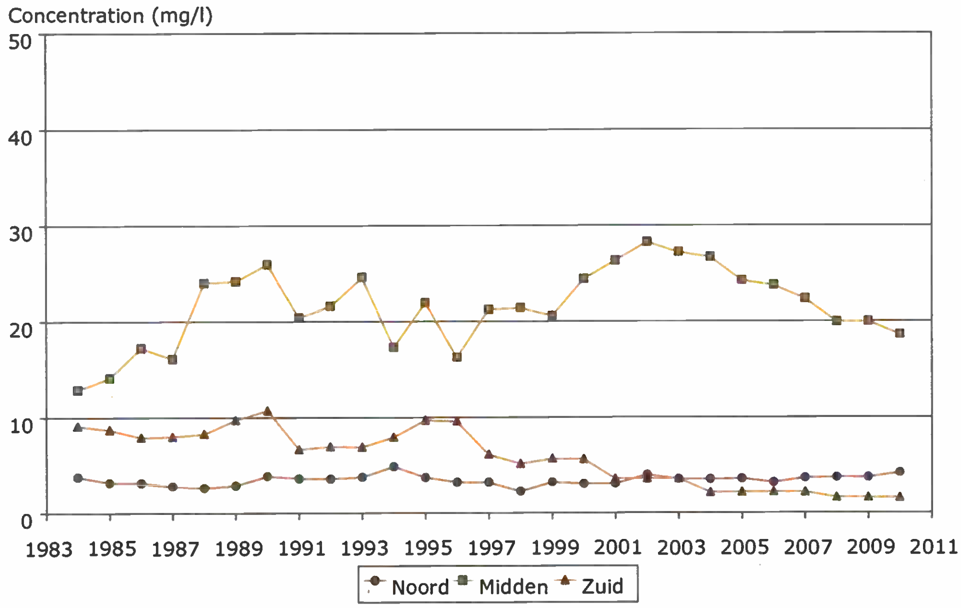


Figure 5.11. Nitrate in groundwater at a depth of 15-30 metres under farming areas by sand area.

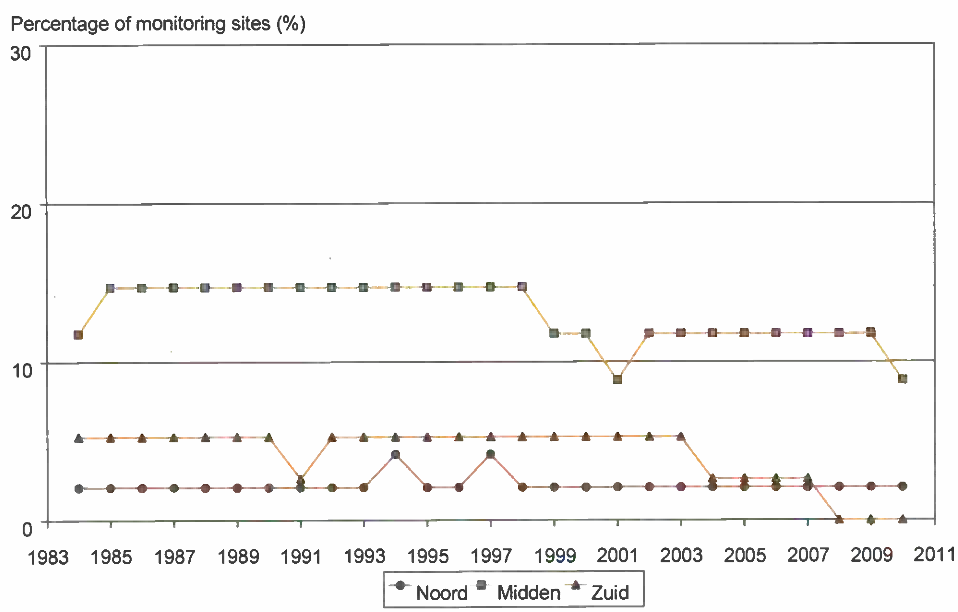
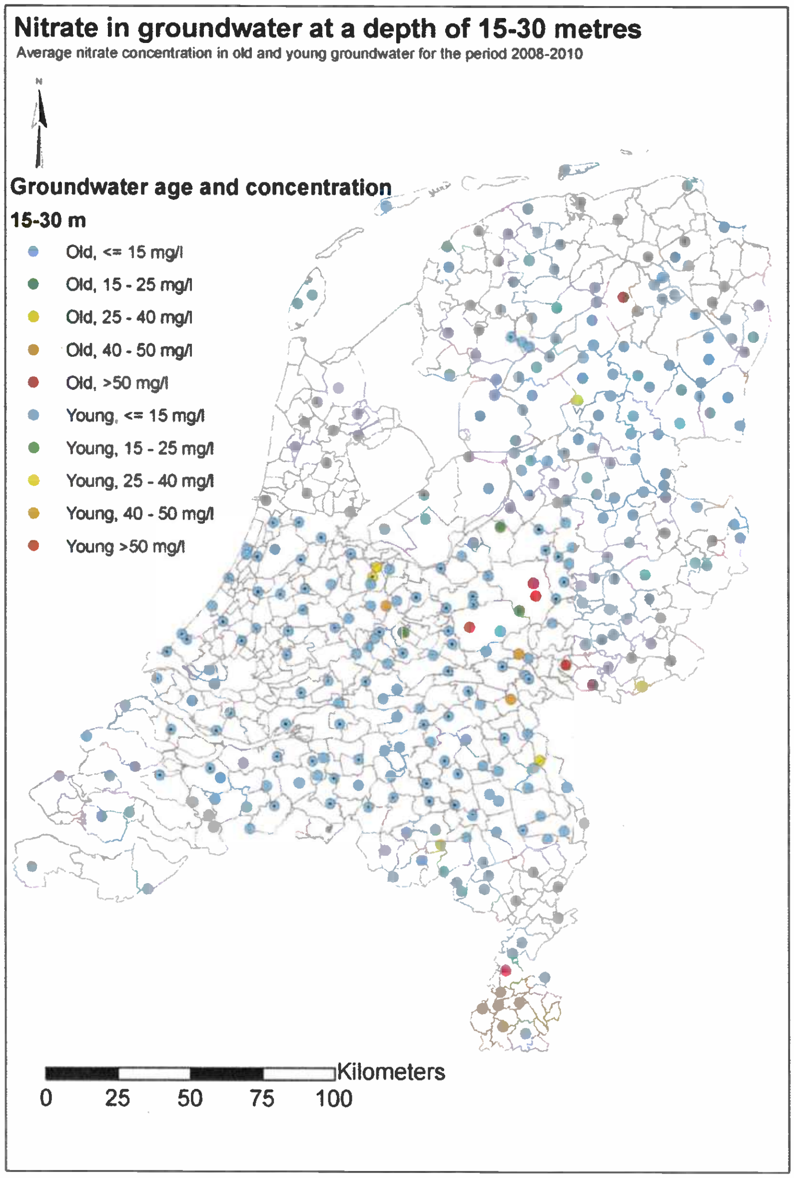
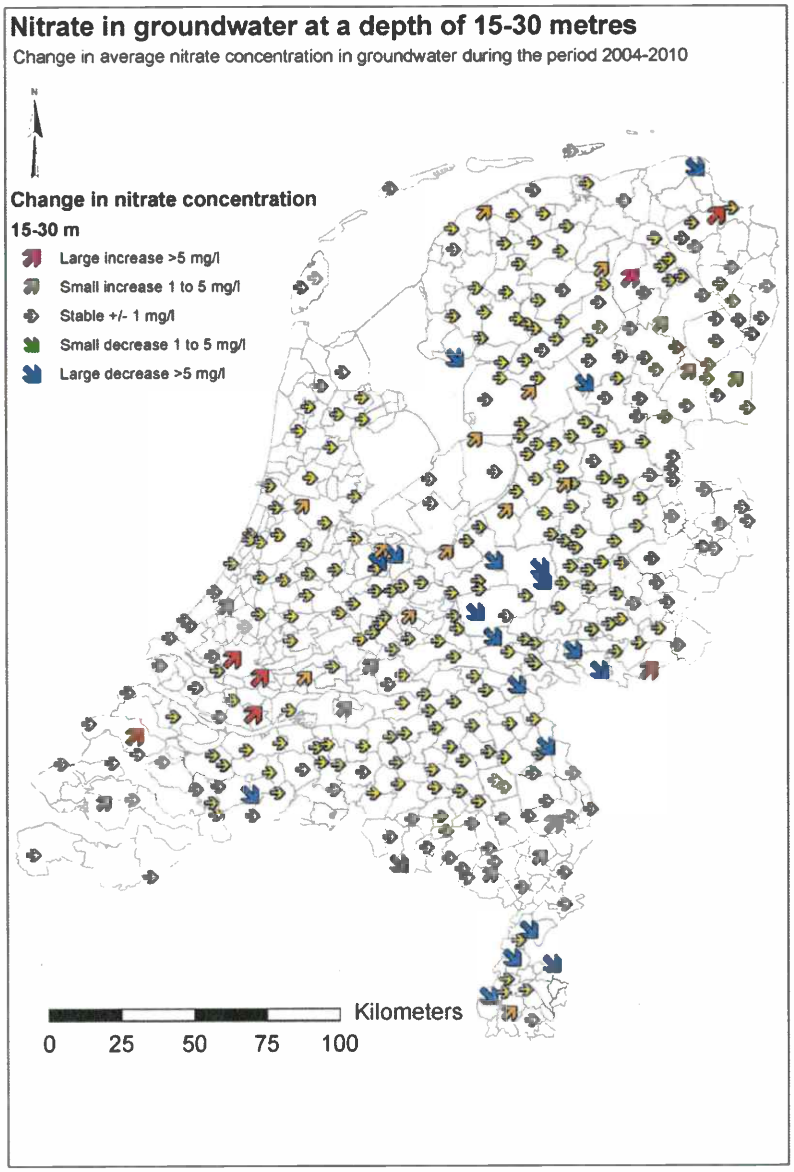


Figure 5.12. Exceedance of the EU standard of 50 mg/l for nitrate in groundwater at a depth of 15-30 metres by sand area for the period 1984-2010.



Map 5.3. Average nitrate concentration in groundwater at a depth of 15-30 metres in the Netherlands for the period 2008-2010. "Young" refers to groundwater younger than 25 years; "Old" means older than 25 years.



Map 5.4. Change in average nitrate concentration in groundwater at a depth of 15-30 metres for the period 2008-2010.
Change shown here is the difference between averages for the 2004-2007 and 2008-2010 periods.

5.4 Nitrate in groundwater below 30 metres

In the period 2008-2010, the average nitrate concentration in groundwater used for drinking water production (raw water) was about 6.5 mg/l for phreatic aquifers and less than 1 mg/l for confined aquifers. The nitrate concentration in raw water from phreatic aquifers increased slightly until 2003, and then decreased until 2006. After 2006, the nitrate concentration remained stable (Figure 5.13).

The percentage of drinking water production sites with an average nitrate concentration in raw water above 50 mg/l was less than 2% (Figure 5.14 and Table 5.6).

The fairly constant nitrate concentration in raw water is also apparent fromTable 5.7. The 40-50 mg/l class increased slightly, whereas other classes decreased slightly again. The number of sites with an increase went down, whereas the number showing a large increase went up.

The EU standard of 50 mg/l was not exceeded in distributed drinking water. In 2010, none of the 227 drinking water production sites had a nitrate concentration exceeding 50 mg/l.

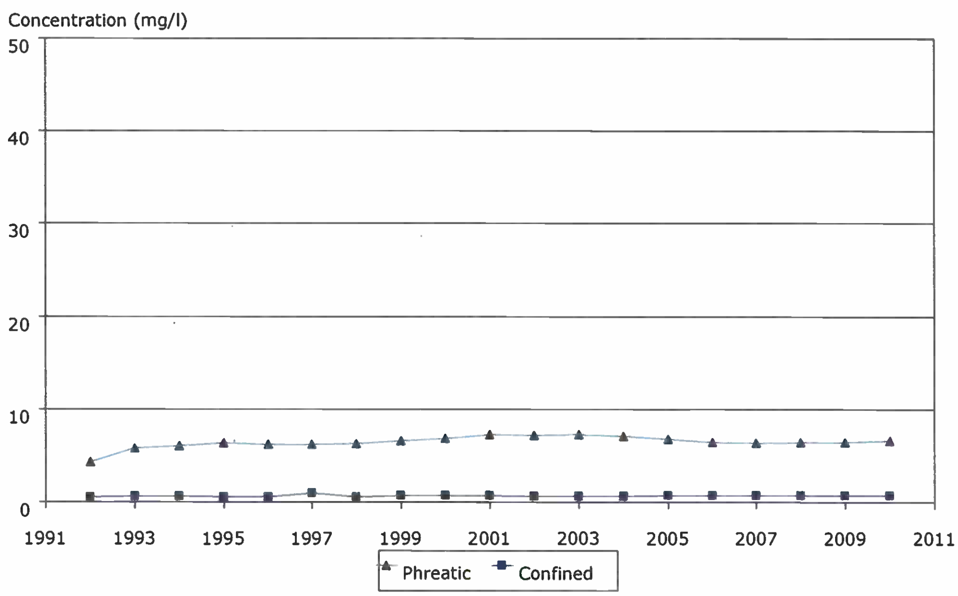


Figure 5.13. Average annual nitrate concentration (mg/l) in groundwater in phreatic and confined aquifers at drinking water production sites in the period 1992–2010.

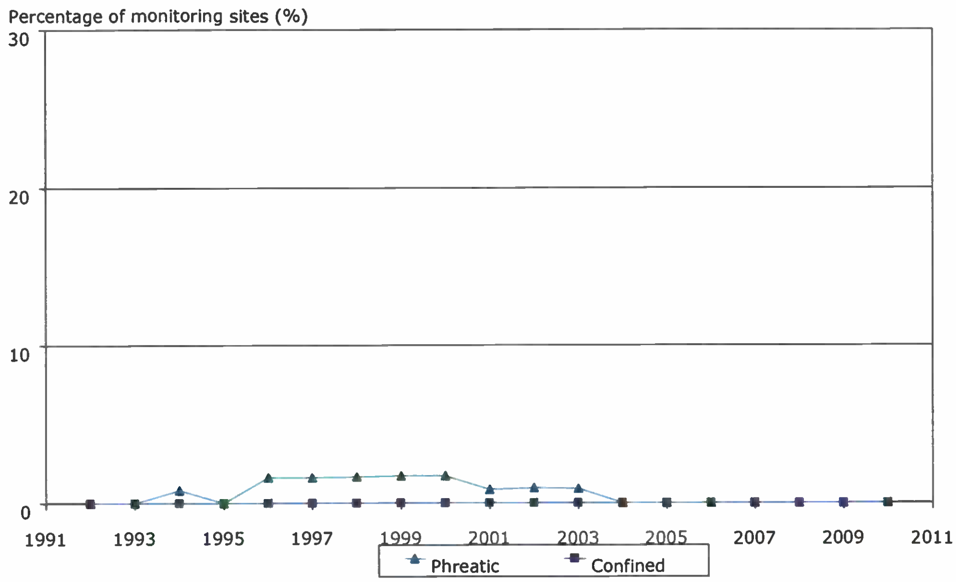


Figure 5.14. Exceedance of the EU standard of 50 mg/l for average nitrate concentration in groundwater at drinking water production sites for phreatic groundwater and confined groundwater in the period 1992-2010. Exceedance is expressed as a percentage of all production sites.

Table 5.6. Average nitrate concentration in groundwater at a depth of more than 30 metres in the period 1992-2010 (%)[1].

	All production sites			Phreatic sites		
Concentration	1992-1995	2004-2007	2008-2010	1992-1995	2004-2007	2008-2010
0-15 mg/l	91.2	90.0	90.9	85.3	82.4	83.8
15-25 mg/l	5.1	6.3	4.5	8.5	11.1	8.1
25-40 mg/l	3.2	3.2	2.8	5.4	5.6	5.1
40-50 mg/l	0.5	0.5	1.7	0.8	0.9	3.0
> 50 mg/l	0.0	0.0	0.0	0.0	0.0	0.0
Number of sites	217	190	176	129	108	99

[1] Percentage of drinking water production sites using groundwater with a period average within a given concentration range for all production sites and for sites with only phreatic groundwater. The total percentage could exceed 100 because of rounding.

In the period 2008-2010, the average maximum nitrate concentration in groundwater used for drinking water production was about 10 mg/l for phreatic aquifers and less than 1 mg/l for confined aquifers (Figure 5.15). The maximum nitrate concentration in raw water from phreatic aquifers remained constant over the last three years of the period. On average, 1% of the sites had a maximum nitrate concentration higher than the EU standard (Figure 5.16 and Table 5.8).

Table 5.7. Change in average nitrate concentration in groundwater at a depth of more than 30 metres in the period 1992-2010 (%)[1].

	All production sites		Phreatic sites	
Change	1992-1995/ 2004-2007	2004-2007/ 2008-2010	1992-1995/ 2004-2007	2004-2007/ 2008-2010
Large increase (% > 5 mg/l)	4.2	1.2	6.7	2.2
Small increase (% 1-5 mg/l)	7.3	3.6	13.3	6.7
Stable (% ± 1 mg/l)	86.7	86.7	65.6	76.7
Small decrease (% > 1-5 mg/l)	7.3	6.1	10.0	10.0
Large decrease (% > 5 mg/l)	1.8	2.4	2.2	4.4
Number of sites	165	165	90	90

[1] Percentage of drinking water production sites using groundwater with given size of change in concentration between the first and fourth, and the fourth and fifth reporting periods. The table shows the data of all production sites, as well as of sites with only phreatic groundwater. The total percentage could exceed 100 because of rounding.

Concerning the maximum nitrate concentration in groundwater used for drinking water extraction, the percentage of sources with a large increase went down, and the percentage of sources with a large decrease went up (Table 5.9).

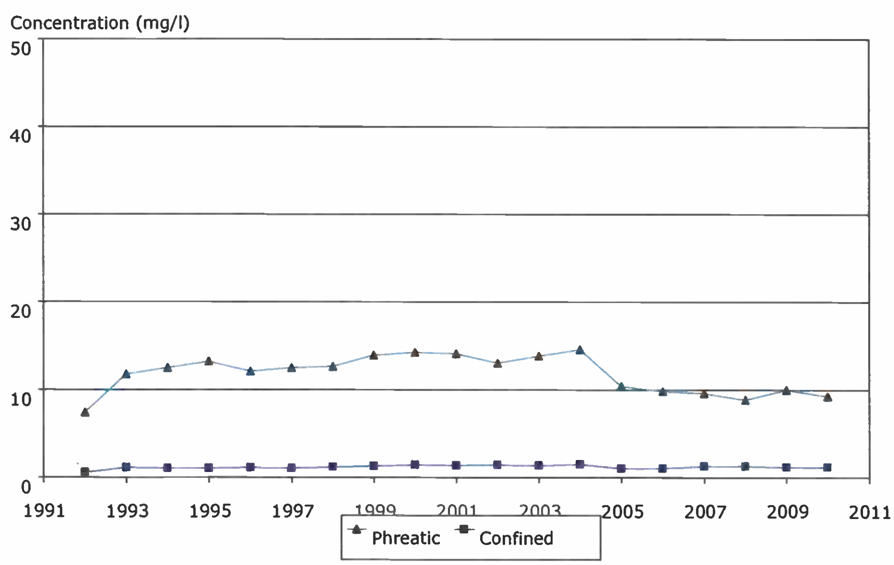


Figure 5.15. Maximum nitrate concentration (mg/l) in groundwater at drinking water production sites for phreatic groundwater and confined groundwater in the period 1992–2010.

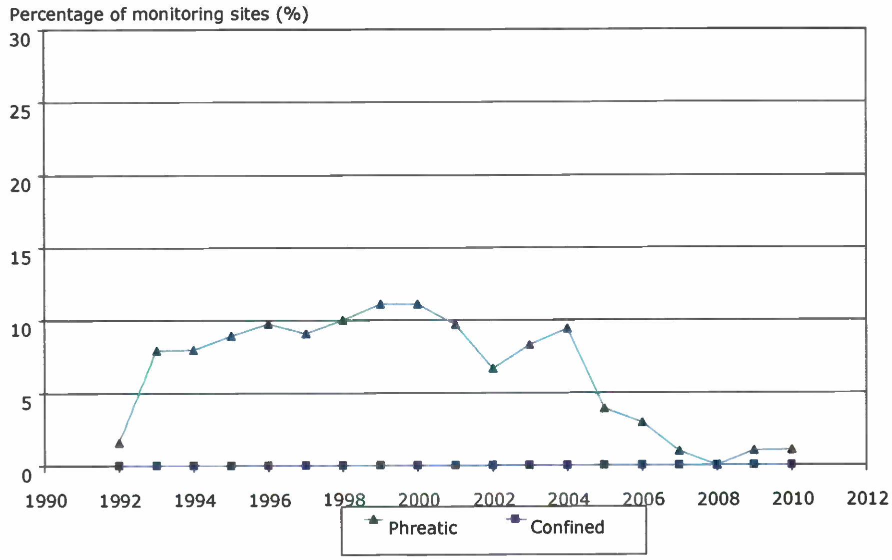


Figure 5.16. Exceedance of the EU standard of 50 mg/l for average nitrate concentration in groundwater at drinking water production sites for phreatic groundwater and confined groundwater in the period 1992-2010. Exceedance is expressed as a percentage of all production sites.

Table 5.8. Maximum nitrate concentration in groundwater at a depth of more than 30 metres for the period 1992-2010 (%)[1].

	All production sites			Phreatic sites		
Concentration	1992-1995	2004-2007	2008-2010	1992-1995	2004-2007	2008-2010
0-15 mg/l	84.3	83.2	85.8	75.2	71.3	74.7
15-25 mg/l	5.5	6.3	4.5	7.8	10.2	8.1
25-40 mg/l	5.1	3.7	4.5	8.5	6.5	8.1
40-50 mg/l	0.5	3.7	4.5	0.8	6.5	8.1
> 50 mg/l	4.6	3.2	0.6	7.8	5.6	1.0
Number of sites	217	190	176	129	108	99

[1] Percentage of drinking water production sites using groundwater with a period average within a given concentration range for all production sites and for sites with only phreatic groundwater. The total percentage could exceed 100 because of rounding.

Table 5.9. Change in maximum nitrate concentration in groundwater at a depth of more than 30 metres for the period 1992-2010 (%)[1].

	All production sites		Phreatic sites	
Change	1992-1995/ 2004-2007	2004-2007/ 2008-2010	1992-1995/ 2004-2007	2004-2007/ 2008-2010
Large increase (% > 5 mg/l)	4.2	0.6	7.8	1.1
Small increase (% 1-5 mg/l)	6.7	6.1	10.0	10.0
Stable (% ± 1 mg/l)	80.0	80.0	56.7	66.7
Small decrease (% > 1-5 mg/l)	10.3	4.8	6.7	6.7
Large decrease (% > 5 mg/l)	6.7	8.5	11.1	15.6
Number of sites	165	165	90	90

[1] Percentage of drinking water production sites using groundwater with given rate of change in concentration between the first and fourth, and between the fourth and fifth reporting periods. The table shows the data of all production sites, as well as of sites with only phreatic groundwater. The total percentage could exceed 100 because of rounding.

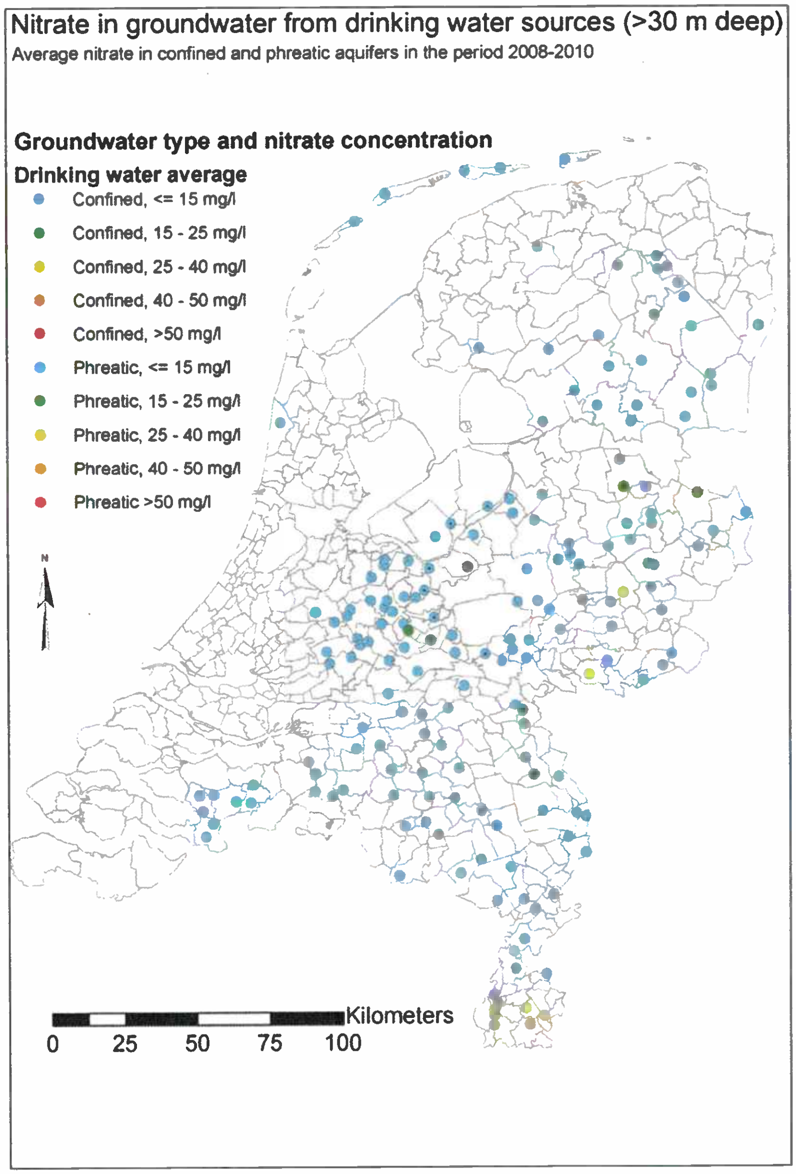
Map 5.5 shows the average concentration per drinking water production site in the period 2007-2009, while Map 5.6 shows the change between the 2004-2007 and 2007-2009 periods. The highest nitrate concentrations occur in the southern (mainly in the loess region) and eastern parts of the Netherlands near the German border (sand regions). These areas in particular showed decreasing trends.

Map 5.7 shows the maximum concentration per drinking water production site in the period 2007-2009, while Map 5.8 shows the change in maxima between the 2004-2007 and 2008-2010 periods. The highest maximum nitrate concentrations also occur in the southern and the eastern parts of the Netherlands.

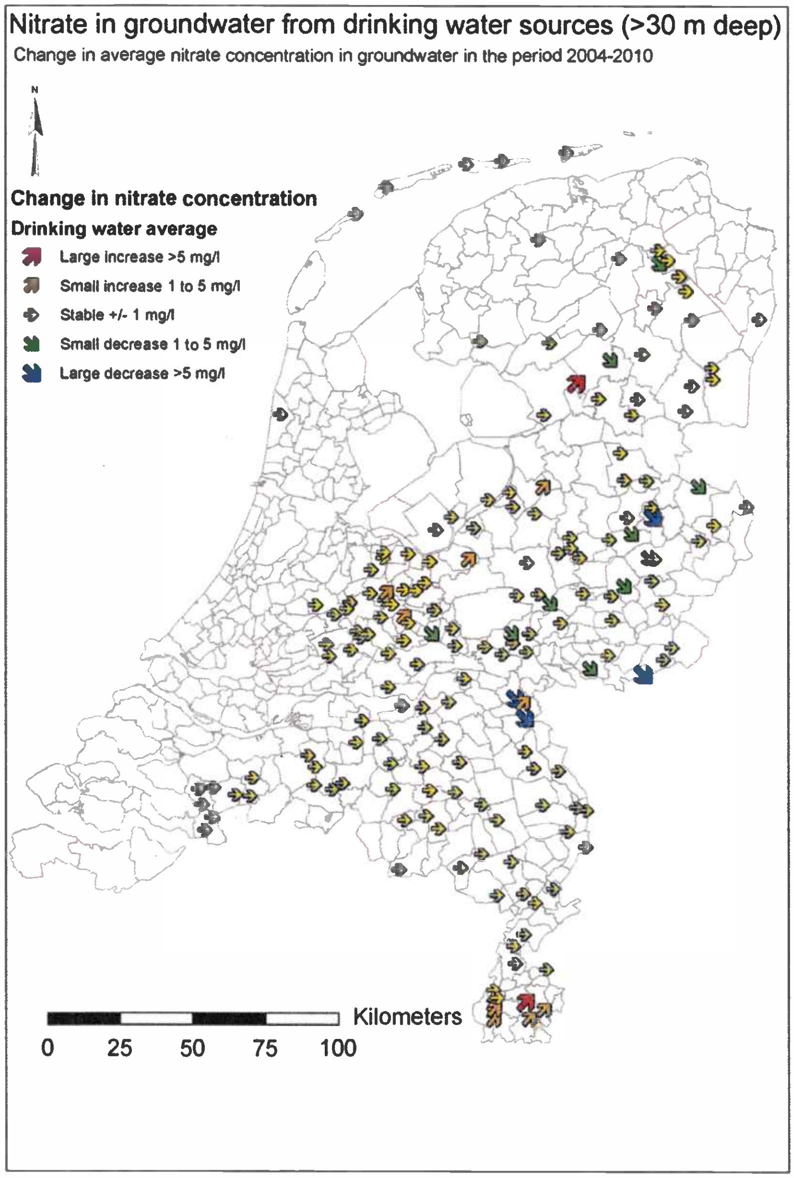
5.5 References

Reijnders H.F.R., Van Drecht G., Prins H.F., Bronswijk J.J.B., Boumans L.J.B. (2004). De kwaliteit van het ondiepe en middeldiepe grondwater in Nederland in het jaar 2000 en de verandering daarvan in de periode 1984-2000. RIVM Report 714801030.

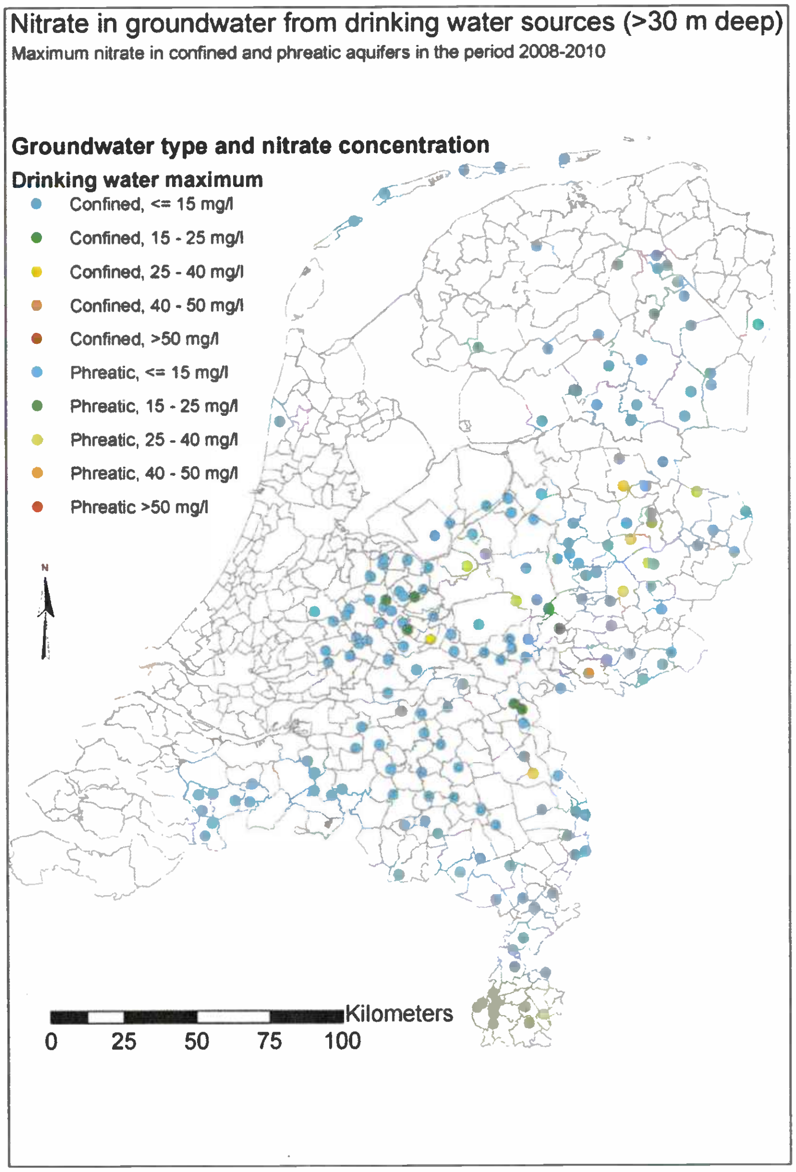
Van Vliet, M.E., Vrijhoef, A., Boumans, L.J.M., Wattel-Koekkoek, E.J.W. (2010). De kwaliteit van ondiep en middeldiep grondwater in Nederland in het jaar 2008 en de verandering daarvan in 1984-2008. RIVM Report 680721005.



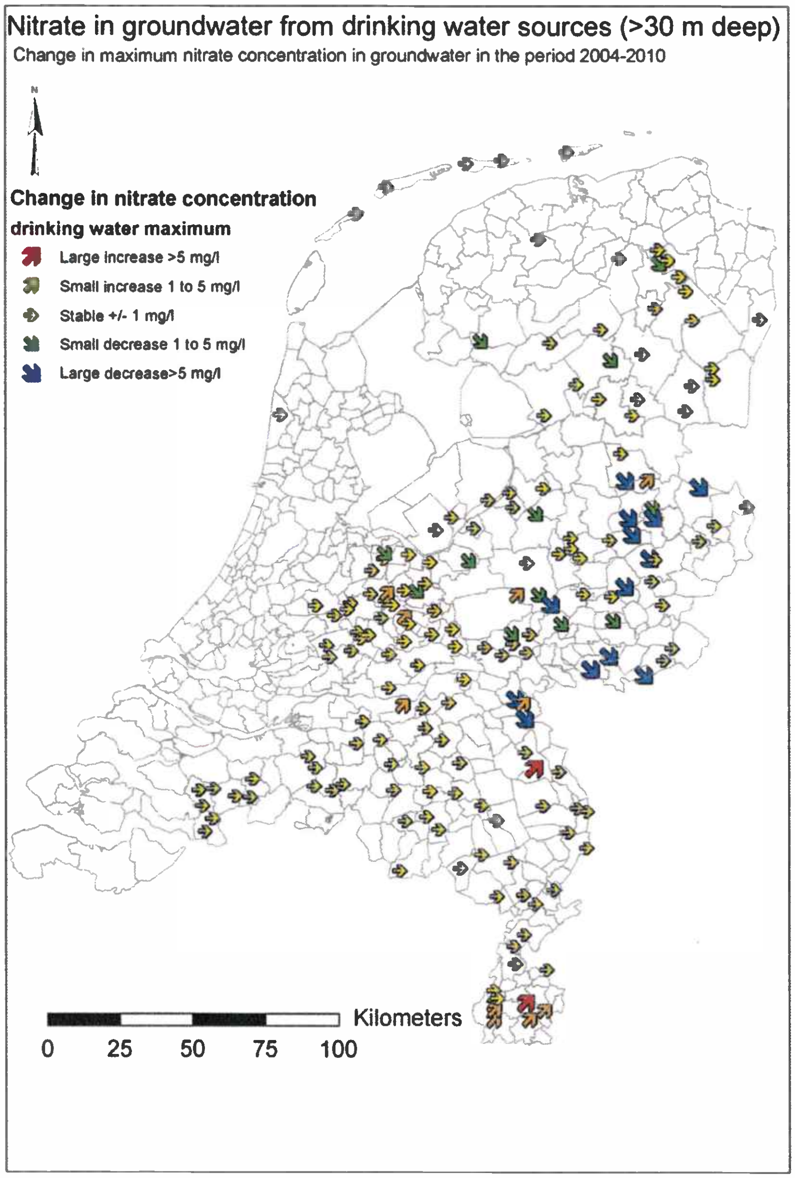
Map 5.5. Average nitrate concentration in groundwater used for drinking water production in the period 2008-2010.



Map 5.6. Change in maximum nitrate concentration in groundwater used for drinking water production in the period 2008-2010. Change shown here is the difference between averages for the 2004-2007 and 2008-2010 periods.



Map 5.7. Maximum nitrate concentration in groundwater used for drinking water production in the period 2008-2010.



Map 5.8. Change in maximum nitrate concentration in groundwater used for drinking water production in the period 2004-2010. Change shown here is the difference between the averages for the 2004-2007 and 2008-2010 periods.

6 Freshwater quality

6.1 Introduction

The first part (6.2) of this chapter deals briefly with the nutrient load on fresh surface waters. Section 6.3 presents an overview of the nutrient concentrations in the various fresh surface waters of the Netherlands, as well as the trend in nitrate concentration during three different periods. Section 6.4 deals with the various parameters that define the eutrophication status of freshwater. Nitrogen and phosphorus both affect the degree of eutrophication.

The data presented in this chapter originate from measurements of all the fresh surface waters, in particular, of those strongly affected by agriculture. In addition to these waters, main locations are also discussed in this chapter. It can be concluded in general that main locations are in the main water system, which comprises the large rivers and lakes. The remaining sites are those in regional waters.

Main locations are singled out as a separate category for two reasons. The first is that they show the impact on water quality of sources from outside the Netherlands. The second is that the effects of inland (non-agricultural) sources on water quality in the coastal zone are easy to identify by measuring the nitrate concentrations at main locations.

In conformity with EU standards, this report considers nitrate-nitrogen to be the most important variable for reflecting the effects of agriculture on surface water quality. In watercourses prone to eutrophication, nitrates disappear in varying degrees because algae absorb them in the summer months, which can result in the monitoring results presenting a distorted picture. The more eutrophicated a water body is, the lower the nitrate concentration during the summer. The winter average (October to March) is therefore more representative than the annual average. The winter period is also the time when leaching processes play a key role. In this report, the winter, summer and annual averages for nitrate are presented.

In the Netherlands, the degree of eutrophication is assessed from the chlorophyll-a concentration (algal biomass), total nitrogen concentration and total phosphorus concentration. Chlorophyll-a concentrations are at their highest during the summer months (April to October). For this reason, the summer average is used to determine the degree of eutrophication of the various water bodies during the designated periods. In accordance with Dutch standards, eutrophication is presented not only using the summer average of chlorophyll-a concentration, but also using the summer average of total phosphorus concentration and total nitrogen concentration (STOWA, 2007). Nitrogen concentration is an indicator of the quantity of nutrients present, as well as of the algal biomass.

6.2 Nutrient load on fresh surface waters

Water does not respect borders. As the Netherlands lies on the delta of several rivers, a large part of its water comes from other countries.

The greater part of the total quantity of nitrogen in the Dutch freshwater system originates from outside the country. Around 65% found in the Netherlands' freshwaters originates from abroad (Stroomgebiedbeheerplannen, 2009). The remainder in the Dutch water system originates from various other sources (Table 6.1).

Leaching and run-off are the leading domestic sources of both nitrogen (61%; Figure 6.1) and phosphorus (56%; Table 6.2). Their relative contributions have increased over time, mainly because the contributions from other sources, such as directly from agriculture, have decreased more steeply (Tables 6.1 and 6.2). In the case of nitrogen, between 89% and 94% of leaching and run-off amounts originate from farmlands. For phosphorus, the figure is between 92% and 94%. The ratio of nitrogen leaching to nitrogen run-off per ha from agriculture and natural sources varied between 1.6 and 2.8 in the period 1986-2000 (Schoumans et al., 2008). In the case of phosphorus, the ratio bracket was higher (1.8-7.8). The variations between soil types and between groups of groundwater steps were also significantly greater, with the reservation that the ratio for natural wetlands might be too low (Schoumans et al., 2008).

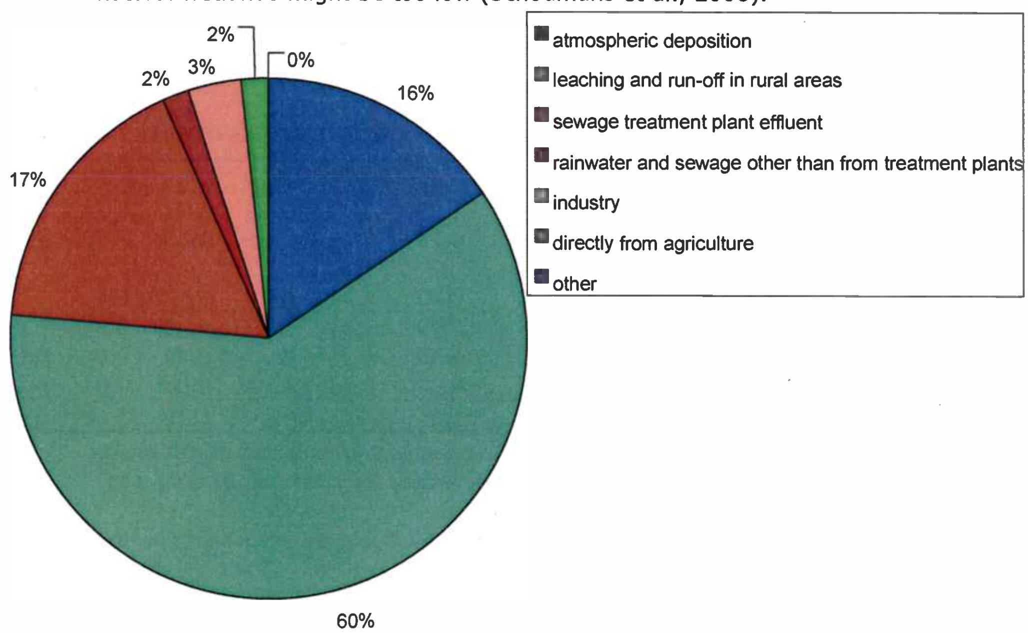


Figure 6.1. Percentages of different sources of the nitrogen load on surface water in 2009.
Source: Pollutant Release and Transfer Register, 2012.

For a detailed description of the different sources and an inventory of the quantities originating from abroad, refer to the various river basin management plans (SGBP, 2009).

Table 6.1. Surface water nitrogen load from domestic sources (kg million).

Origin	1990	1995	2000	2005	2008	2009
Atmospheric deposition[1]	24	20	17	16	15	14
Leaching and run-off in rural areas	55	83	83	43	54	54
Sewage treatment plant effluent	39	36	29	22	17	15
Rainwater and sewage other than from treatment plants[2]	5	4	3	2	2	1
Industry	13	7	5	4	3	3
Directly from agriculture[3]	6	5	2	2	1	1
Other	0.06	0.06	0.05	0.06	0.05	0.05
Total	143	154	138	89	92	89

1 Atmospheric deposition relates to fresh water and marine surface up to one mile from the coastal zone.

2 Sewage other than from treatment plants relates to overflowing, rainwater gullies, discharge from private wastewater treatment systems, and households not connected to sewer systems.

3 Directly from agriculture relates to greenhouse nurseries and unintended fertilisation of ditches.

Source: Pollutant Release and Transfer Register, 2012 (new data not available until after 1 June).

Table 6.2. Surface water phosphorus load from domestic sources (kg million).

Origin	1990	1995	2000	2005	2008	2009
Leaching and run-off in rural areas	2.75	3.78	4.26	3.02	3.51	3.51
Sewage treatment plant effluent	6.24	3.53	2.85	2.65	2.55	2.30
Rainwater and sewage other than from treatment plants[1]	0.63	0.42	0.27	0.19	0.11	0.11
Industry	10.99	3.47	1.87	0.38	0.36	0.24
Directly from agriculture[2]	0.50	0.33	0.15	0.12	0.06	0.06
Other	0.01	0.01	0.01	0.01	0.01	0.01
Total	21.11	11.54	9.40	6.36	6.60	6.23

1 Rainwater and sewage other than from treatment plants relates to overflowing, rainwater gullies, discharge from private wastewater treatment systems, and households not connected to sewer systems.

2 Directly from agriculture relates to greenhouse nurseries and unintended fertilisation of ditches.

Source: Pollutant Release and Transfer Register, 2012 (new data not available until after 1 June).

For smaller water bodies, such as regional waters, the nitrogen content originates almost entirely from domestic sources. For larger water bodies, such as the major rivers, the greater part of the nitrogen content originates from foreign sources. The reduction in nitrogen emissions in other countries will mainly affect the nitrogen concentration in these water bodies.

It is necessary to conclude agreements with neighbouring countries to reduce the foreign emissions in the Dutch water system. Such agreements are already part of the river basin management plans, developed in accordance with the Water Framework Directive (first planning period), and will be updated in the second planning period (SGBP, 2009 and Note in Reply, 2009). The progress with measures within the Netherlands to reduce the nitrogen concentration is also set out in Water in Beeld, 2011.

6.3 Nitrate concentration in fresh water

6.3.1 Nitrate concentration – winter average

In winter, the majority of freshwater sites (all waters) had an average nitrate concentration below the EU standard of 50 mg/l (Table 6.3 andMap 6.1). Comparing the first period with subsequent ones shows that the number of sites with an average greater than 50 mg/l was decreasing (Table 6.3). A comparison of the two latest periods shows that the nitrate concentration had stopped changing.

Water bodies where the EU standard of 50 mg/l was exceeded in winter during the latest period (2008-2010) are in Oost-Brabant, the southern part of Limburg, Westland, and the eastern part of the Netherlands (Map 6.1). A similar situation occurred in the preceding period (2004-2007; Zwart et al., 2008). During that period, however, water bodies in Oost-Brabant did not exceed the EU standard, unlike in West-Brabant, where this was the case. In the period 2008-2010, all sites in West-Brabant and Zeeland had a concentration less than 50 mg/l. Low concentrations were found during the latest period, in the area south of Friesland for example.

Table 6.3. Nitrate concentration (winter average) in fresh surface waters in different periods (%).

	All waters			Waters strongly affected by agriculture		
Concentration	1992-1995	2004-2007	2008-2010	1992-1995	2004-2007	2008-2010
0-2 mg/l	2	5	6	0	6	8
2-10 mg/l	20	40	46	28	42	45
10-25 mg/l	50	39	36	44	37	33
25-40 mg/l	17	12	9	13	8	10
40-50 mg/l	5	2	1	5	3	1
>50 mg/l	7	2	1	11	3	3
Number of sites	373	507	468	130	177	144

The winter average nitrate concentration remained stable or decreased at approximately 92% of sites (Table 6.4 and Map 6.2). At about 8% of the sites, there was an increase in nitrate concentration. If the first (1992-1995) and fourth (2004-2007) reporting periods are compared, then the winter average nitrate concentration remained stable or decreased at over 90% of sites (Table 6.4). In other words, the situation in the Netherlands was worse in the first period than in subsequent periods. The situation improved significantly in the fourth and fifth reporting periods. This was especially true in West-Brabant, which saw a reduction in the average nitrate concentration in the winter (Map 6.2). According to the previous report (Zwart et al., 2008), there had actually been an increase in average nitrate concentration in this region.

In the two latest periods, no appreciable difference existed between, on the one hand, sites strongly affected by agriculture and, on the other, main locations (Figure 6.2). The average nitrate concentrations in winter were almost halved between the first and fourth periods. Between the two latest periods, no significant change occurred. Something striking, though, is the higher winter average in 2007.

Table 6.4. Change in nitrate concentration (winter average) in fresh waters in different periods (%).

Change	All waters		Waters strongly affected by agriculture	
	1992/1995-2004/2007	2004/2007-2008/2010	1992/1995-2004/2007	2004/2007-2008/2010
Large increase (> 5 mg/l)	4	2	6	4
Small increase (1-5 mg/l)	4	6	5	6
Stable (± 1 mg/l)	8	28	11	24
Small decrease (1-5 mg/l)	36	48	36	47
Large decrease (> 5 mg/l)	48	15	42	19
Number of sites	326	431	118	125

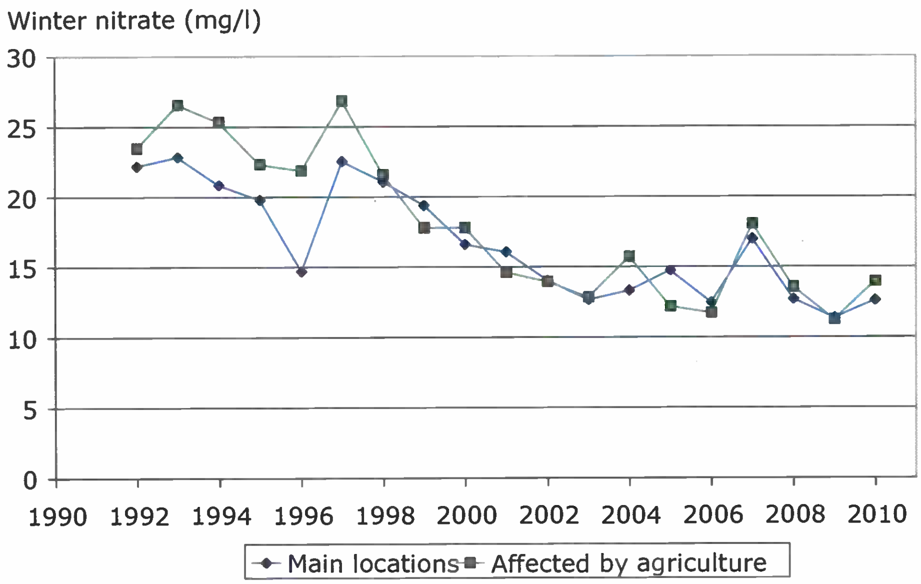


Figure 6.2. Nitrate concentration (winter average) in fresh surface waters in the period 1992-2010.

6.3.2 *Nitrate concentration – winter maximum*

For the vast majority of sites, the winter maximum nitrate concentration was below the EU standard of 50 mg/l, as shown in Table 6.5 and Map 6.3.

Table 6.5. Nitrate concentration (winter maximum) in fresh surface waters for different periods (%).

	All waters			Waters strongly affected by agriculture		
Concentration	1992-1995	2004-2007	2008-2010	1992-1995	2004-2007	2008-2010
0-2 mg/l	1	3	3	0	2	1
2-10 mg/l	10	22	28	13	25	30
10-25 mg/l	39	48	43	38	43	42
25-40 mg/l	26	13	15	24	15	13
40-50 mg/l	7	5	5	5	2	6
>50 mg/l	17	10	5	20	12	8
Number of sites	373	507	468	130	177	144

As with winter average concentrations, winter maximum concentrations did not change much between the two latest periods. The largest change occurred between the first and fourth reporting periods, as shown in Table 6.6. The increase between the fourth (2004-2007) and fifth (2007-2010) periods was mainly in the eastern half of the country (Map 6.4).

Table 6.6. Change in nitrate concentration (winter maximum) in fresh waters for different periods (%).

	All waters		Waters strongly affected by agriculture	
Change	1992/1995-2004/2007	2004/2007-2008/2010	1992/1995-2004/2007	2004/2007-2008/2010
Large increase (> 5 mg/l)	9	5	18	6
Small increase (1-5 mg/l)	6	10	7	10
Stable (± 1 mg/l)	12	23	9	15
Small decrease (1-5 mg/l)	24	36	20	34
Large decrease (> 5 mg/l)	49	26	46	35
Number of sites	336	431	120	125

6.3.3 *Nitrate concentration – annual average*

As stated in the introduction, the annual average and the winter average nitrate concentration are both presented in this section.

It is explained in section 6.1 that nitrate sometimes disappears in the summer months owing to it being absorbed by algae. The average concentration in winter is therefore a better indicator of the nitrate concentration at different sites, a standpoint confirmed by comparing the results in Table 6.7 with those in Table 6.3.

Table 6.7. Nitrate concentration (annual average) in fresh waters for different periods (%).

	All waters			Waters strongly affected by agriculture		
Concentration	1992-1995	2004-2007	2008-2010	1992-1995	2004-2007	2008-2010
0-2 mg/l	3	9	11	3	8	14
2-10 mg/l	33	52	55	44	59	54
10-25 mg/l	46	31	29	34	23	24
25-40 mg/l	9	5	4	5	4	5
40-50 mg/l	4	1	0	4	1	1
>50 mg/l	5	2	1	10	4	2
Number of sites	374	511	472	128	178	147

Most sites had an annual average nitrate concentration of between 2-10 mg/l, which differs from the winter average concentrations. For all waters, 55% had an annual average nitrate concentration between 2 and 10 mg/l, as opposed to 46% if winter average concentrations are considered. For sites strongly affected by agriculture, the percentages are 54% and 45%, respectively.

6.4 Eutrophication of freshwater

6.4.1 Chlorophyll-a

Eutrophication is defined in this section using the average chlorophyll-a concentration in the summer. However, the occurrence of eutrophication in the form of chlorophyll-a is not determined solely by nitrate concentrations in surface waters. Other nutrients, such as phosphorus, and physical and meteorological conditions, also play a role. Chlorophyll-a is related to the quantity of algae present in surface water and therefore the chlorophyll-a concentration is also an indicator of the degree of surface water eutrophication. According to proposed nutrient standards for ecological status (STOWA, 2007), the strictest standard will apply to moderately deep buffered lakes (water type M20), with a concentration of 10 µg chlorophyll-a/l as the line between the scores "adequate" and "good". Higher cut-off scores apply to other water types (STOWA, 2007).

The chlorophyll-a concentration decreased between the first and fourth reporting periods (Tables 6.8 and 6.9). At sites affected by agriculture, the reduction was less pronounced. After that, the situation was virtually stable as regards the fourth and fifth reporting periods.

Table 6.8. Chlorophyll-a concentration (summer average) in fresh waters in different periods (%).

	All waters			Waters strongly affected by agriculture		
Concentration	1992-1995	2004-2007	2008-2010	1992-1995	2004-2007	2008-2010
0-2.5 µg/l	2	1	0	3	0	0
2.5-8 µg/l	6	10	9	9	11	3
8.0-25 µg/l	32	41	47	28	39	46
25-75 µg/l	36	37	32	27	34	30
>75 µg/l	24	11	13	32	16	21
Number of sites	235	356	351	74	110	102

Table 6.9. Change in chlorophyll-a concentration (summer average) in fresh waters in different periods (%).

	All waters		Waters strongly affected by agriculture	
Change	1992/1995-2004/2007	2004/2007-2008/2010	1992/1995-2004/2007	2004/2007-2008/2010
Large increase (> 10 µg/l)	11	18	19	26
Small increase (5-10 µg/l)	5	10	8	13
Stable (± 5 µg/l)	25	37	31	34
Small decrease (5-10 µg/l)	12	13	4	5
Large decrease (> 10 µg/l)	47	22	38	22
Number of sites	193	294	52	82

The largest decrease in chlorophyll-a concentration occurred between the first and fourth periods (59%), after which the largest group of sites (37%) was stable. At the same time, the number of sites that showed an increase was approximately the same as those that showed a decrease in chlorophyll-a concentration (about 28% and 35% respectively; Table 6.9).

A decline in chlorophyll-a concentration occurred over the years (Figure 6.3). Sites strongly affected by agriculture show the same trend as other sites, although the concentrations at the former are slightly higher and fluctuate more strongly from year to year.

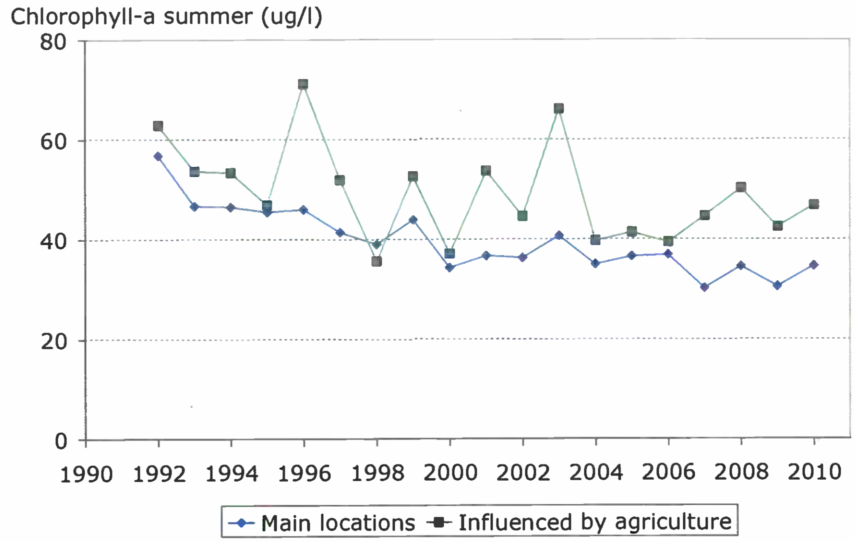


Figure 6.3. Chlorophyll-a concentration (summer average) in fresh waters in the Netherlands in the period 1992-2010.

6.4.2 *Other parameters indicating eutrophication*

Similar to chlorophyll-a in surface waters, the average of total nitrogen concentrations (Tables 6.10a and b, and Figure 6.4) in the summer and that of total phosphorus concentrations (Tables 6.11a and b, and Figure 6.5) show a falling trend over the years. In the Netherlands, separate standards for different water types have been derived for the summer average of total nitrogen (STOWA, 2007). Generally speaking, some 40% of Netherlands' waters met the relevant standard (State budget, 2011).

Table 6.10a. Total nitrogen concentration (summer average) in different periods (%).

	All waters			Waters strongly affected by agriculture		
Concentration	1992-1995	2004-2007	2008-2010	1992-1995	2004-2007	2008-2010
0-2 mg/l	5	23	27	4	23	29
2-5 mg/l	64	61	59	65	59	52
5-7 mg/l	12	9	9	10	7	10
>7 mg/l	19	8	5	21	11	9
Number of sites	350	510	422	113	177	157

For total nitrogen, the same situation existed as for chlorophyll-a (previous section), though less pronounced. Total nitrogen concentration in the summer showed a steep decline between the first and fourth periods, in common with all the other parameters discussed in this section.

Table 6.10b. Change in total nitrogen concentration (summer average) in fresh waters between different periods (%).

Change	All waters		Waters strongly affected by agriculture	
	1992/1995-2004/2007	2004/2007-2008/2010	1992/1995-2004/2007	2004/2007-2008/2010
Large increase (> 0.5 mg/l)	9	6	13	7
Small increase (0.25-0.50 mg/l)	3	7	6	11
Stable (± 0.25 mg/l)	8	39	5	34
Small decrease (0.25-0.50 mg/l)	6	18	1	18
Large decrease (> 0.5 mg/l)	74	30	74	31
Number of sites	350	370	113	131

Table 6.11a. Total phosphorus concentration (summer average) in different periods (%).

Concentration	All waters			Waters strongly affected by agriculture		
	1992-1995	2004-2007	2008-2010	1992-1995	2004-2007	2008-2010
<0.05 mg/l	0	4	2	0	2	2
0.05-0.10 mg/l	8	15	21	15	14	20
0.10-0.20 mg/l	38	38	33	31	33	25
0.20-0.50 mg/l	31	24	23	18	14	20
>0.50 mg/l	23	20	20	36	37	34
Number of sites	331	507	503	102	175	179

Table 6.11b. Change in total phosphorus concentration (summer average) between different periods (%).

Change	All waters		Waters strongly affected by agriculture	
	1992/1995-2004/2007	2004/2007-2008/2010	1992/1995-2004/2007	2004/2007-2008/2010
Large increase (> 0.10 mg/l)	5	6	11	11
Small increase (0.05-0.10 mg/l)	5	6	3	7
Stable (± 0.05 mg/l)	35	65	38	50
Small decrease (0.05-0.10 mg/l)	19	10	14	8
Large decrease (> 0.10 mg/l)	37	13	34	24
Number of sites	328	459	100	153

Of sites strongly affected by agriculture, there was an increase in average summer total phosphorus concentration at approximately 20% between 2004 and 2010, and a decrease at approximately 32% (Table 6.11b). For all waters, 12% showed an increase and 23 a decrease. In other words, the summer average concentration of total phosphorus decreases more slowly at sites strongly influenced by agriculture than at other sites (Figure 6.5). In the case of sites where the water is strongly affected by agriculture, the summer average concentration is above 0.50 mg/l at 34% of them, whereas for all sites together, the figure is 20% (Table 6.11a). The total phosphorus concentrations given in this report are some 0.2 mg/l higher than those reported in EMW 2012 (Klein et al., 2012b), the cause being a different selection of sites (see section 2.6.2).

Klein et al., (2012a) found there was a significant falling trend in median concentrations (50th percentile) in the period 1990-2010.

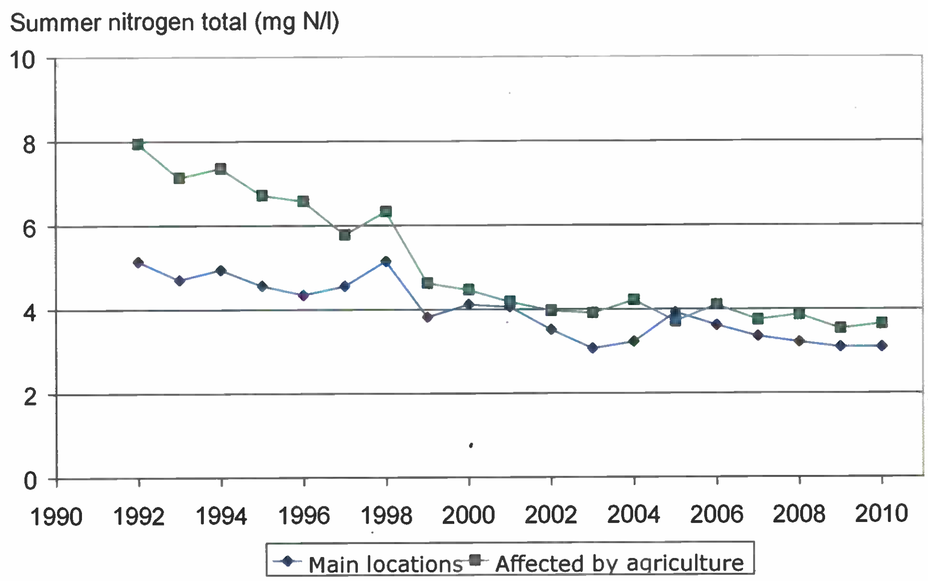


Figure 6.4. Total nitrogen concentration (summer average) in fresh waters in the period 1992-2010.

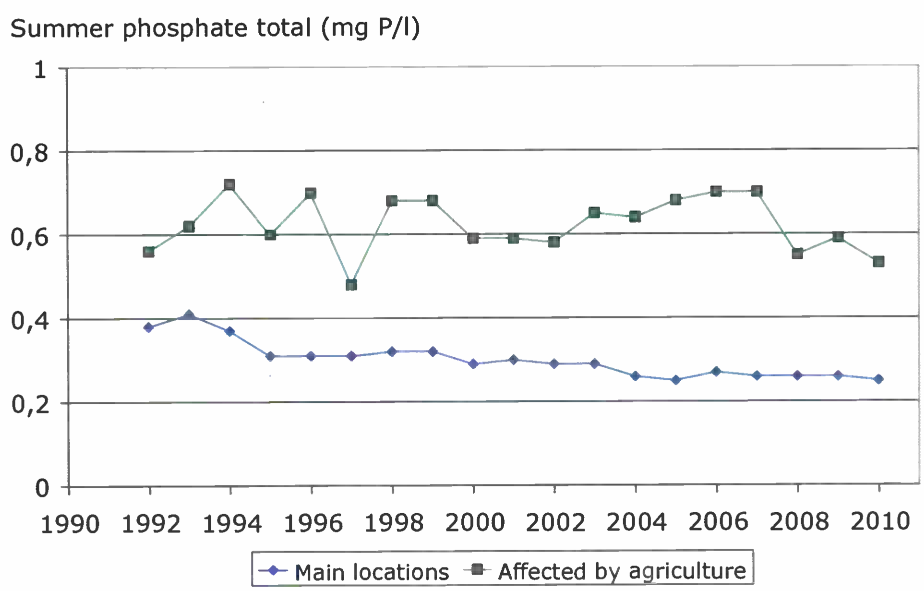


Figure 6.5. Total phosphorus concentration (summer average) in fresh waters in the period 1992-2010.

6.5 Trends

In general, more than 90% of all sites (main locations and sites strongly affected by agriculture) show a steady decrease in winter average nitrate concentration or a stable pattern for this concentration. This is clear from a comparison of the initial period with subsequent periods. In other words, the situation regarding nitrate in the water system has improved since 1992. The same applies to total phosphorus and chlorophyll-a, although the improvement in the case of the chlorophyll-a concentration is less pronounced.

6.6 References

EC/DGXI (2000). Nitrates Directive (91/676/EEC). Status and trends of aquatic environment and agricultural practice. Reporting Guidelines. European Commission, DGXI (Environment), April 2000.

Klein, J., Rozemeijer, J., Broers, H.P. (2012a). Meetnet Nutriënten Landbouw Specifiek Oppervlaktewater. Deelrapport A. Opzet meetnet. Bijdrage aan de Evaluatie Meststoffenwet 2012. Utrecht, Deltares, report 1202337-000-BGS-0007.

Klein, J., Rozemeijer, J., Broers, H.P., Van der Grift, B. (2012b). Meetnet Nutriënten Landbouw Specifiek Oppervlaktewater. Deelrapport B. Trend en Toestand. Bijdrage aan de Evaluatie Meststoffenwet 2012. Utrecht, Deltares, report 1202337-000-BGS-0008.

Note in Reply (2009). Inspraak ontwerp-stroomgebiedbeheerplannen, 2009; www.helpdeskwater.nl.

Pollutant Release and Transfer Register (2012) http://www.emissieregistratie.nl/ (14 May, 2012)

Schoumans, O.F., Groenendijk, P., Renaud, L., Van der Bolt, F.J.E. (2008). Nutriëntenbelasting van het oppervlaktewater. Vergelijking tussen landbouw en natuurgebieden. Wageningen, Alterra, Alterra Report 1700.

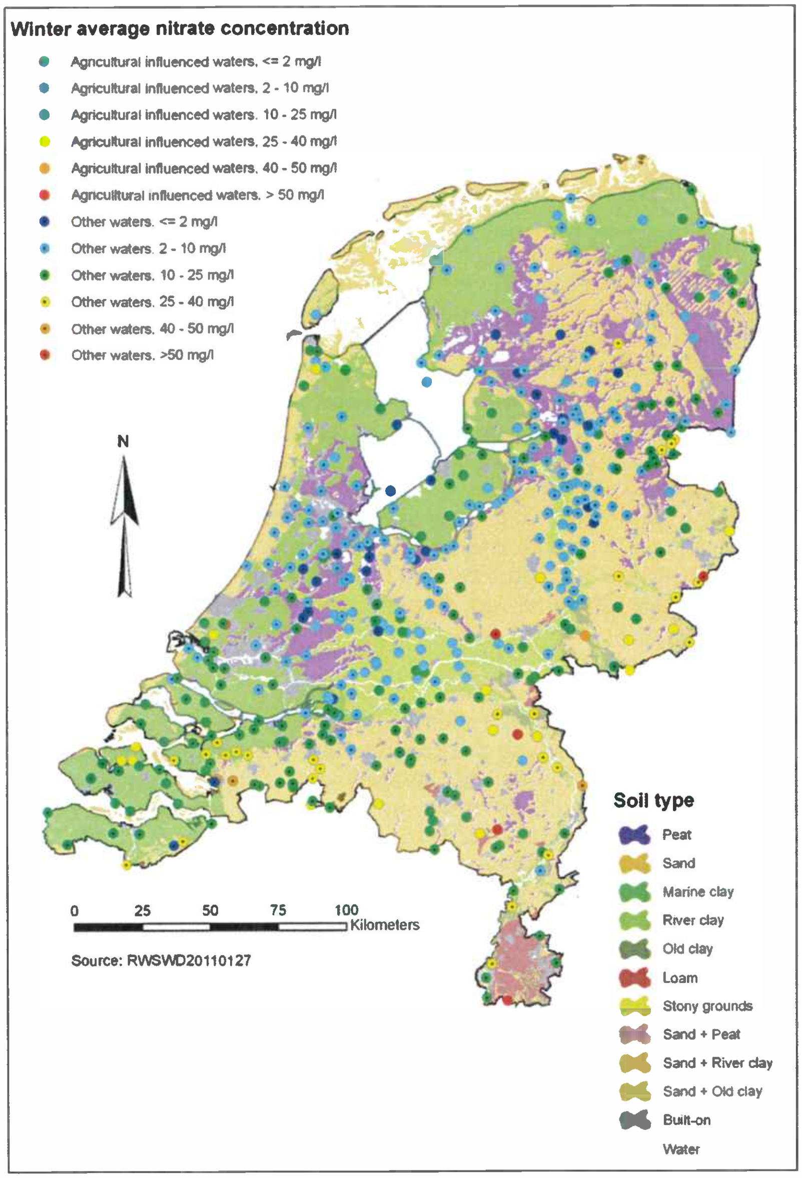
State budget (2011). Onderdeel 'Integraal waterbeleid', effectindicator waterkwaliteit (data for 2009), www.rijksbegroting.nl.

STOWA (2007). Toelichting op ecologische doelen voor nutriënten in oppervlaktewateren, report number: STOWA 2007.18 and RIZA 2007.29.

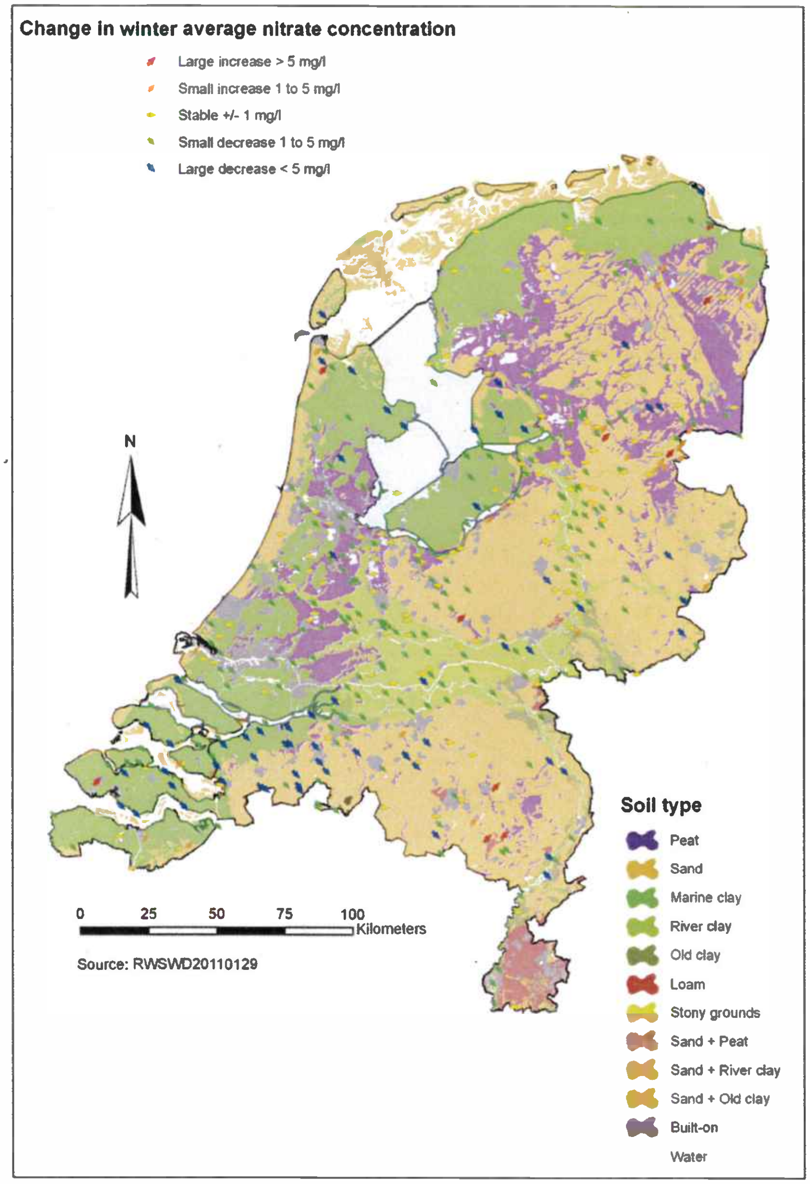
Stroomgebiedbeheerplannen (2009). Joint publication from the Ministry of Transport, Public Works and Water Management, the Ministry of Housing, Spatial Planning and the Environment, and the Ministry of Agriculture, Nature and Food Quality; www.helpdeskwater.nl.

Water in Beeld (2011). Ministry of Infrastructure and the Environment, in collaboration with its partners on the Nationaal Water Overleg (national water management platform, comprising the Netherlands' central government, Association of the Provinces of the Netherlands, Association of Water Boards, and the Association of Netherlands Municipalities). Report on the progress with the national water management plan during 2010. ISSN number: 1388-6622

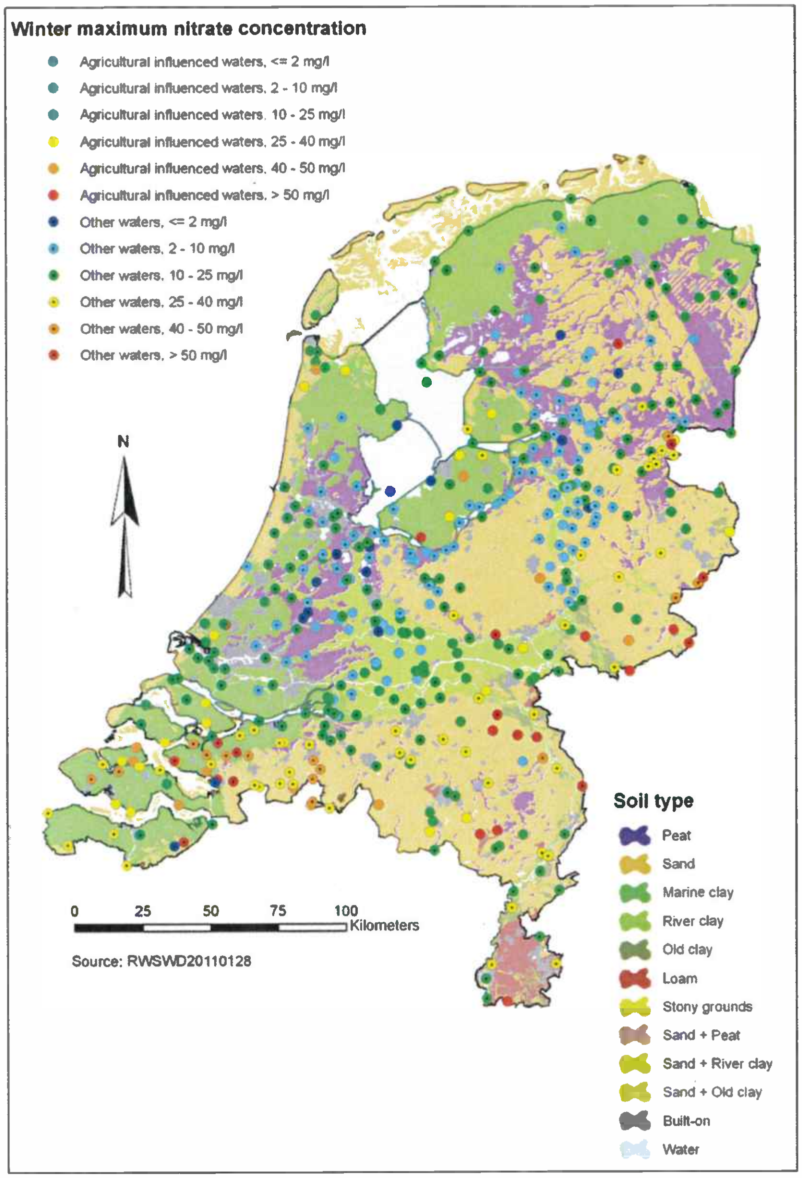
Zwart, M.H., Hooijboer, A.E.J., Fraters, B., Kotte, M., Duin, R.N.M., Daatselaar, C.H.G., Oltshoorn, C.S.M., Bosma, J.N. (2008). Agricultural practice and water quality in the Netherlands in the 1992-2006 period. Bilthoven, RIVM Report 680716003.



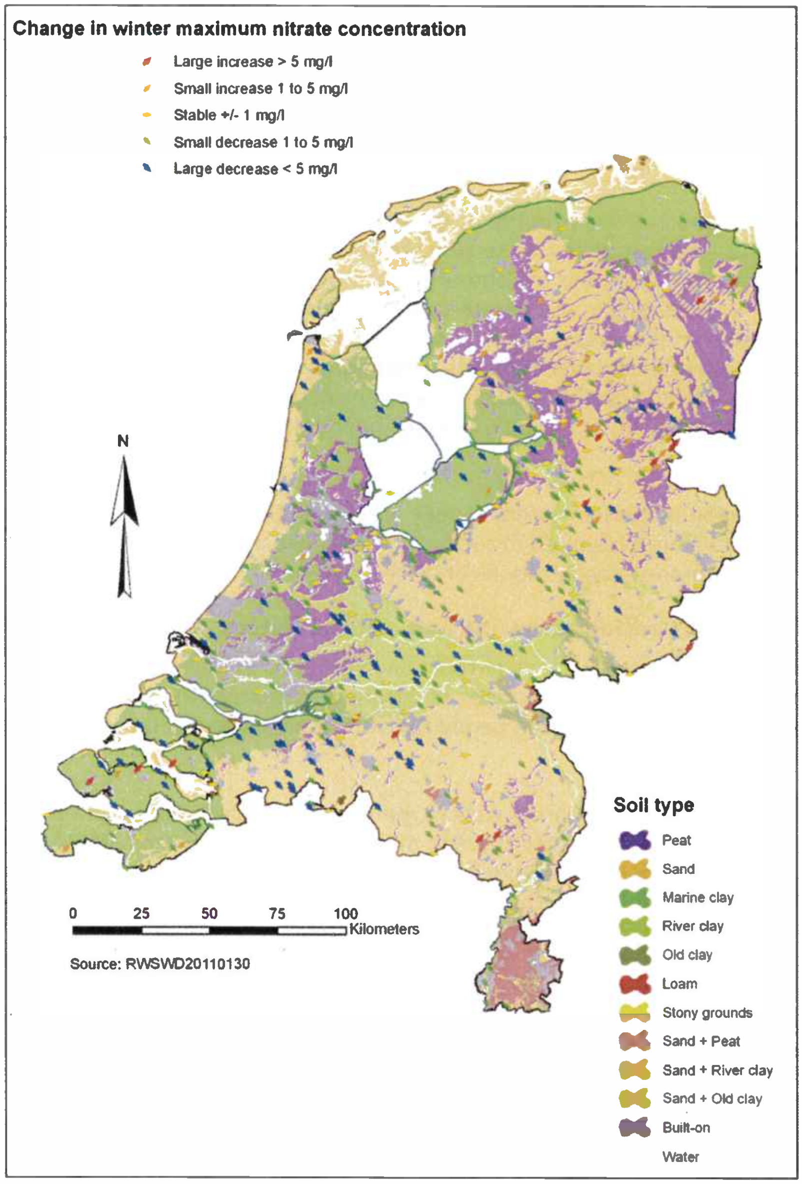
Map 6.1. Winter average nitrate concentration by monitoring site in the period 2008-2010 (fifth period).



Map 6.2. Change in winter average nitrate concentration by monitoring site between the fourth (2004-2007) and fifth (2008-2010) periods.
Change shown here is the difference between the averages for the 2004-2007 and 2008-2010 periods.



Map 6.3. Winter maximum nitrate concentration by monitoring site in the period 2008-2010 (fifth period).



Map 6.4. Change in winter maximum nitrate concentration by monitoring site between the fourth (2004-2007) and fifth (2008-2010) periods.
Change shown here is the difference between the averages for the 2004-2007 and 2008-2010 periods.

7 Marine and coastal water quality

7.1 Introduction

Chapter 7 discusses the results of the monitoring of nitrogen and phosphorus concentrations in marine surface waters.

The nitrate concentrations in the open sea and in coastal areas are described in section 7.2. The results in this section are based only on winter average concentrations (December to February), as the least biological activity occurs in this period. Accordingly, the nitrate concentrations measured in winter are a better indicator of changes in nutrient emissions than those measured in summer. Extensive studies of trends in inorganic nitrogen concentrations, corrected for saline content (salinity) are considered as well, meaning that the annual differences in output by rivers has been taken into account (section 2.6.3).

Eutrophication of marine waters is dealt with in section 7.3. Trends in eutrophication are presented in the form of changes in summer average and maximum average concentrations of chlorophyll-a.

7.2 Nitrate concentration in marine and coastal waters

In this section, winter average nitrate concentrations in marine waters are presented, expressed as milligrams of nitrate per litre. For the graphs, the winter period is defined as the period from 1 December to the last day of February (section 2.6.3).

At over 90% of the monitoring sites in marine and coastal waters during the period 2008-2010, the winter average nitrate concentration was below 10 mg/l, at none of the sites it was above 25 mg/l (Table 7.1). Nitrate concentrations remained stable during the three monitoring periods at all sites in the open sea (Table 7.2). Specifically, at none of the open sea sites was an absolute change measurement of more than 1 mg nitrate/l found. For coastal waters, changes during the two latest monitoring periods were found, however (Table 7.2). The decrease between the fourth and fifth periods is not as significant as that between the first and fourth periods. The nitrate concentration in coastal water is generally stable.

Table 7.1. Winter average nitrate concentration in marine waters in the period 1992-2010 (%)[1].

Concentration	1992-1995	2004-2007	2008-2010
0-10 mg/l	87	95	92
10-25 mg/l	13	5	8
25-40 mg/l	0	0	0
40-50 mg/l	0	0	0
>50 mg/l	0	0	0
Number of sites	39	40	39

[1] Percentage of monitoring sites with a winter average in a given concentration range.

Table 7.2. Change in winter average nitrate concentration in marine waters in the period 1992-2010 (%)[1].

Change	Open sea		Coastal water	
	1992/1995-2004/2007	2004/2007-2008/2010	1992/1995-2004/2007	2004/2007-2008/2010
Large increase (> 5 mg/l)	0	0	0	0
Small increase (1-5 mg/l)	0	0	3	6
Stable (± 1 mg/l)	100	100	72	88
Small decrease (1-5 mg/l)	0	0	25	6
Large decrease (> 5 mg/l)	0	0	0	0
Number of sites	7	7	32	32

[1] Percentage of sites with a given change in concentration between the stated periods.

The winter average nitrate concentrations in coastal zones have decreased slightly over the past seven years, apart from the steep drop in the period 1995-1996 (Figure 7.1). In recent years, the average has fluctuated between 3 and 6 mg nitrate/l. Concentrations in open sea remained virtually stable at far lower concentrations (< 0.5 mg/l). The lower nitrate concentration in coastal water in 1996 was a result of limited precipitation in preceding years. Apart from the fall in 1996, the temporary increases in 2007 and 2008 also stood out.

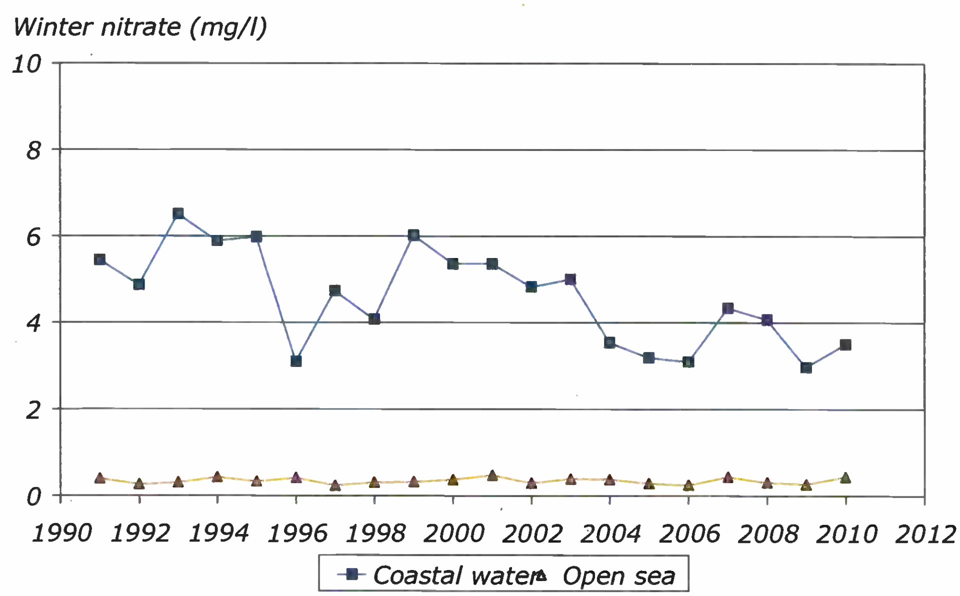


Figure 7.1. Winter average nitrate concentration (mg/l) in open sea and Dutch coastal waters in the period 1991-2010.

In the period 2008-2010, the winter average nitrate concentrations in open sea and Dutch coastal waters were almost everywhere below 10 mg/l (Map 7.1). They were only above this level in the Western Scheldt and the Eems-Dollard estuary. In open sea, the concentrations were less than 2 mg/l.

Compared with the period 2004-2007, from 2008 to 2010 there was a clearly discernible increase in winter average nitrate concentration in the Eems-Dollard estuary, and a decrease in the Western Scheldt (Map 7.2). At all other sites, concentrations remained essentially stable.

For the vast majority of sites, the maximum nitrate concentrations found ranged from 0 to 10 mg/l (Table 7.3). It seems that the number of sites with the lowest nitrate concentrations showed an increase between the first and second half of the 1990s.

Table 7.3. Winter maximum nitrate concentration in marine waters for the period 1992-2010 (%)[1].

Concentration	1992-1995	2004-2007	2008-2010
0-10 mg/l	81	90	90
10-25 mg/l	17	10	10
25-40 mg/l	2	0	0
40-50 mg/l	0	0	0
> 50 mg/l	0	0	0
Number of sites	39	40	39

[1] Percentage of monitoring sites with a maximum concentration in a given range. The total percentage could exceed 100 because of rounding.

Table 7.4 shows the percentages of monitored sites where an increasing, decreasing or stable winter maximum nitrate concentration was found. As in the previous section, only monitoring sites showing an absolute change of at least 1 mg of nitrate per litre were classified as having either an increasing or a decreasing concentration. Similar to the winter average nitrate concentrations (Table 7.2), all monitoring sites in open sea showed nitrate concentrations that remained stable during the reporting periods. The maximum nitrate concentration in coastal waters remained generally stable, with 16% of sites showing a decrease, and 6% an increase.

Figure 7.2 gives the trend in average winter maximum nitrate concentrations in open sea and coastal waters between 1991 and 2010. Apart from the drop in concentrations during the period 1996-1997, the graph shows that the winter maximum average in coastal zones fluctuated between 4 and 8 mg nitrate per litre, whereas concentrations in open sea remained fairly stable at significantly lower concentrations (< 0.5 mg/l). The text accompanying Figure 7.1 above explains why the maximum nitrate concentration was lower in 1996. For the maximum concentration, too, the increases in 2007 and 2008 are striking.

Map 7.3 shows the differences in winter maximum nitrate concentrations between open sea and Dutch coastal waters for the period 2008-2010. In the Western Scheldt and the Eems-Dollard estuary, the winter maximum nitrate concentrations exceeded 10 mg/l. At other sites in coastal areas, concentrations were generally less than 10 mg/l, whereas in open sea, they were below 2 mg/l.

Table 7.4. Change in winter maximum nitrate concentration in marine waters in the period 1992-2010 (%)[1]*.*

Change	Open sea		Coastal water	
	1992/1995-2004/2007	2004/2007-2008/2010	1992/1995-2004/2007	2004/2007-2008/2010
Large increase (> 5 mg/l)	0	0	0	0
Small increase (1-5 mg/l)	0	0	0	6
Stable (± 1 mg/l)	100	100	25	78
Small decrease (1-5 mg/l)	0	0	56	16
Large decrease (> 5 mg/l)	0	0	19	0
Number of sites	7	7	32	32

[1] Percentage of sites with given change in concentration between the reporting periods. The total percentage could exceed 100 because of rounding.

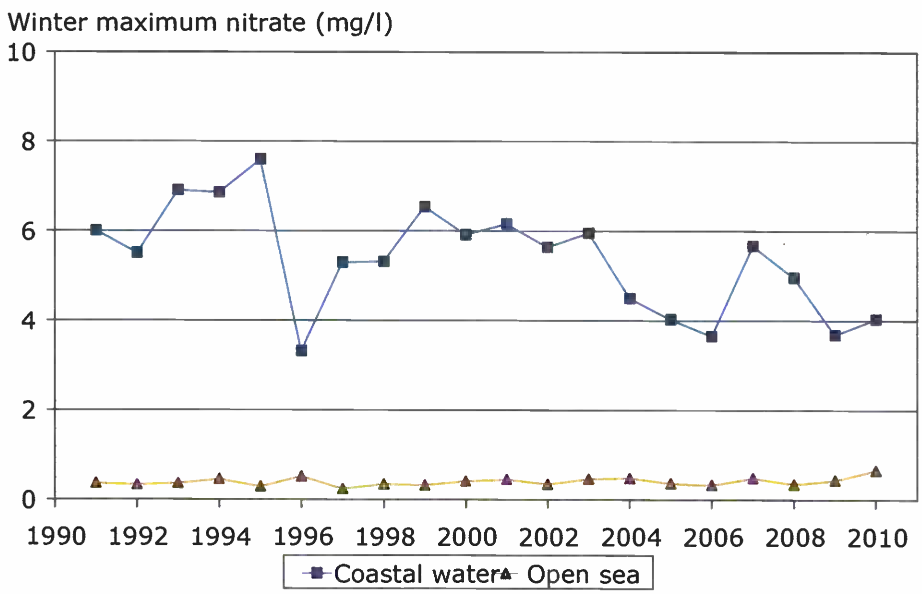


Figure 7.2. Winter maximum nitrate concentration (mg/l) in open sea and Dutch coastal waters in the period 1991-2010.

The trends in winter maximum nitrate concentrations at the different sites in the period 2008-2010 are the same as for winter average concentrations. The concentration in open sea remained fairly stable, while lower concentrations were found in estuaries (see Map 7.4).

The concentrations of nutrients in coastal waters are determined by natural background concentrations, direct discharge, and output from rivers. During winter, biological activity is low and inorganic nutrient concentrations remain fairly stable, showing negative linear correlation with salinity. For a long-term analysis of the relationship between changes in nutrient concentration and changes in nutrient emissions, the measured winter nutrient concentrations have to be corrected for changes in salinity at the selected monitoring sites (see section 2.6.3). Salinity is expressed in practical salinity units (PSUs), an international standard based on saline content in grams per litre.

Since 1991, the dissolved inorganic nitrogen concentrations (DINs) have been slowly but surely declining (Figure 7.3). The concentration in 2010 was approximately 30% lower than in 1991 (Figure 7.4). It seems that concentrations have remained stable since 2003.

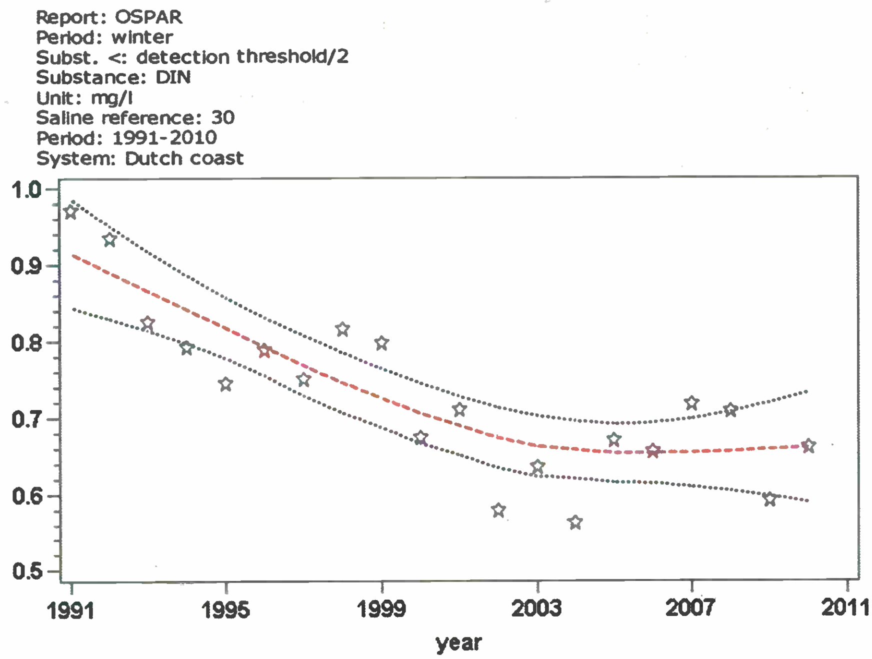


Figure 7.3. Winter average dissolved inorganic nitrogen concentrations (DIN, mg N/l), standardised to a salinity of 30 PSU, in Dutch coastal waters at Noordwijk in the period 1991-2010. The red line is the smoothed trend line and the dashed lines show the 95% confidence interval for the trend line.

7.3 Eutrophication of marine and coastal waters

Eutrophication is a major topic within OSPAR (Convention for the Protection of the Marine Environment of the Northeast Atlantic). A study of Dutch seawater in 2010 revealed that the entire Netherlands coastal zone and the open sea off the coast formed a problem area in terms of eutrophication (OSPAR, 2010). OSPAR specifically requires that DIN concentrations should be 50% below the 1985 level. This is not the case (OSPAR, 2010), so that action needs to be taken to reduce the nutrient load.

The percentage of sites in marine waters with a summer average chlorophyll-a concentration above 25 µg/l went up slightly in the latest period. Seemingly, this was at the expense of the number of sites with concentrations between 8.0 and 25 µg/l, which had apparently decreased (see Table 7.5). In general, though, conditions in marine waters remained fairly stable. It seems that, for open sea and coastal waters, the summer average chlorophyll concentrations were more or less stable (Table 7.6).

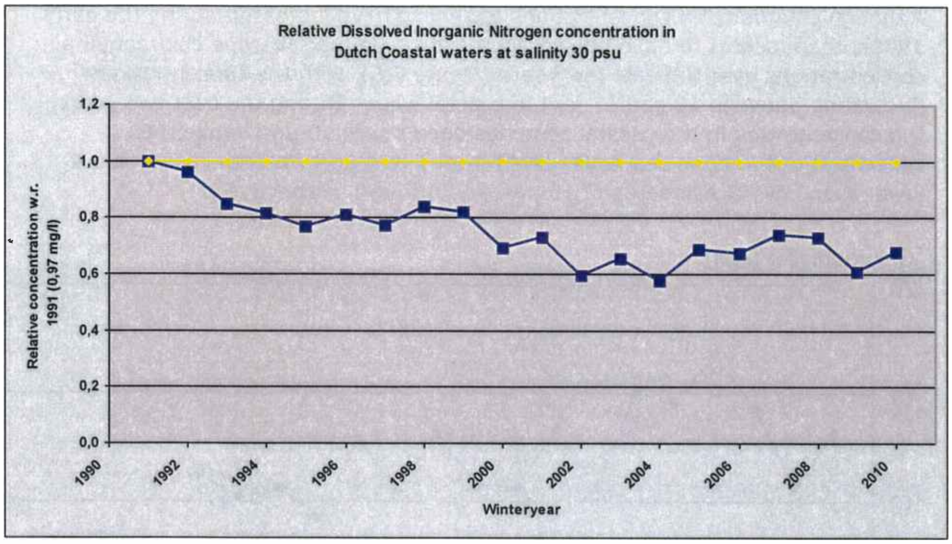


Figure 7.4. Relative winter dissolved inorganic nitrogen (DIN) concentrations, standardised to a salinity of 30 PSU, in Dutch coastal waters at Noordwijk in the period 1991-2010.
DIN concentrations compared to concentration in 1991 (0.97 mg/l is set at 1).

Table 7.5. Summer average chlorophyll-a concentration in marine waters in the period 1992-2010 (%)[1].

Concentration	1992-1995	2004-2007	2008-2010
0-2.5 µg/l	15	18	21
2.5-8.0 µg/l	17	32	29
8.0-25 µg/l	62	50	44
25-75 µg/l	6	0	6
> 75 µg/l	0	0	0
Number of sites	40	34	34

[1] Percentage of monitoring sites with a period average in a given concentration range. The total percentage could exceed 100 because of rounding.

Table 7.6. Change in summer average chlorophyll-a concentration in marine waters in the period 1992-2010 (%)[1].

Change	Open sea		Coastal water	
	1992/1995-2004/2007	2004/2007-2008/2010	1992/1995-2004/2007	2004/2007-2008/2010
Large increase (> 10 µg/l)	0	0	0	0
Small increase (5-10 µg/l)	0	0	0	7
Stable (± 5 mg/l)	100	100	81	85
Small decrease (5-10 µg/l)	0	0	19	7
Large decrease (> 10 µg/l)	0	0	0	0
Number of sites	7	6	27	27

[1] Percentage of sites with a given change in concentration between reporting periods (1992-1995 and 2004-2007, and 2004-2007 and 2008-2010).

Although chlorophyll-a concentrations appear to have increased during the early 1990s, there seems to have been a decrease in summer average chlorophyll-a concentrations over the past few years (Figure 7.5), with the concentrations fluctuating between 10 and 17 µg/l in coastal water. During the past two years, the concentration in the coastal zone has been below 10 µg/l, while the concentrations in open sea have varied from 1 to 4 µg/l. In both cases, this is lower than OSPAR standards of 15 µg/l and 4.5 µg/l, respectively.

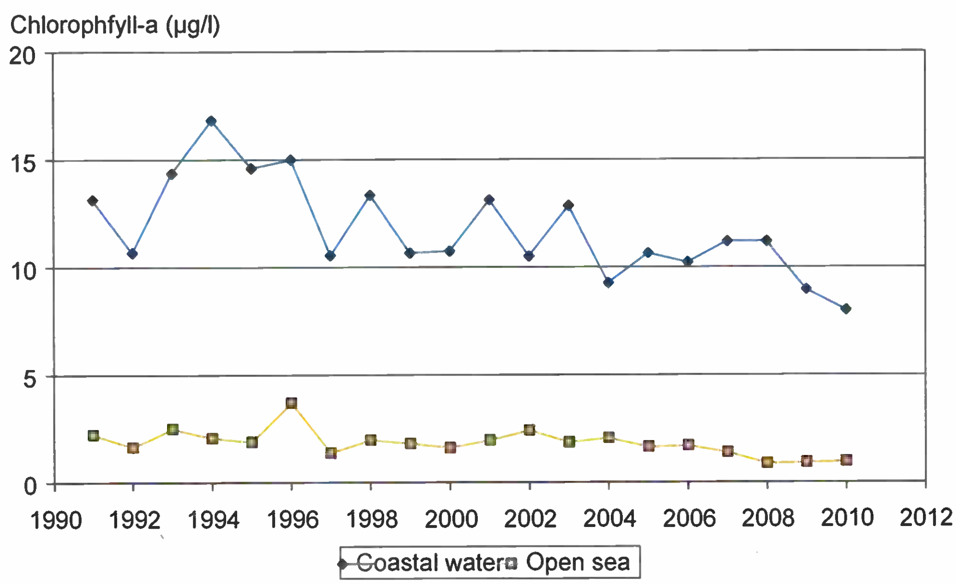


Figure 7.5. Summer average chlorophyll-a concentration (µg/l) in open sea and Dutch coastal waters in the period 1991-2010.

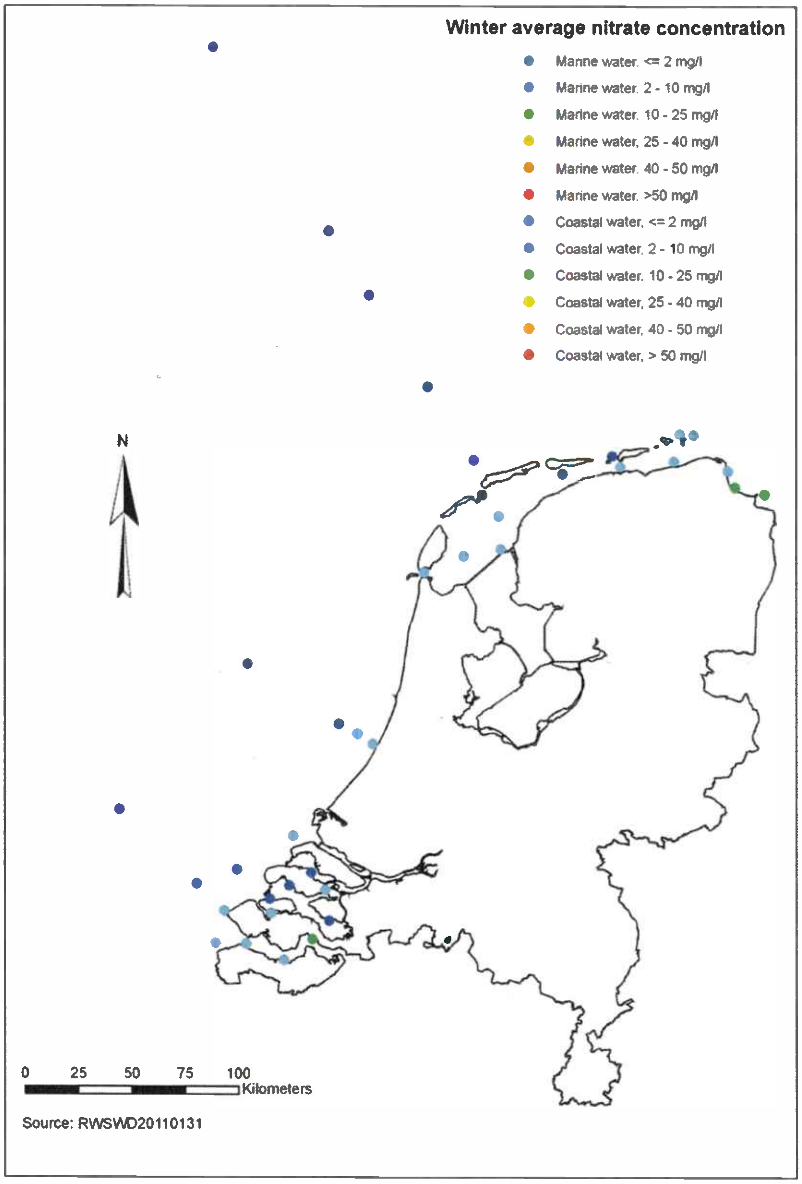
7.4 Conclusion

The marine waters of the Netherlands are characterised by elevated concentrations of nitrogen and chlorophyll-a. There is a slow but gradual decrease in dissolved inorganic nitrogen concentrations, the concentrations in 2010 being approximately 27% lower than those in 1991. Chlorophyll-a concentrations show a slight decreasing trend in coastal waters and remain stable in open sea.

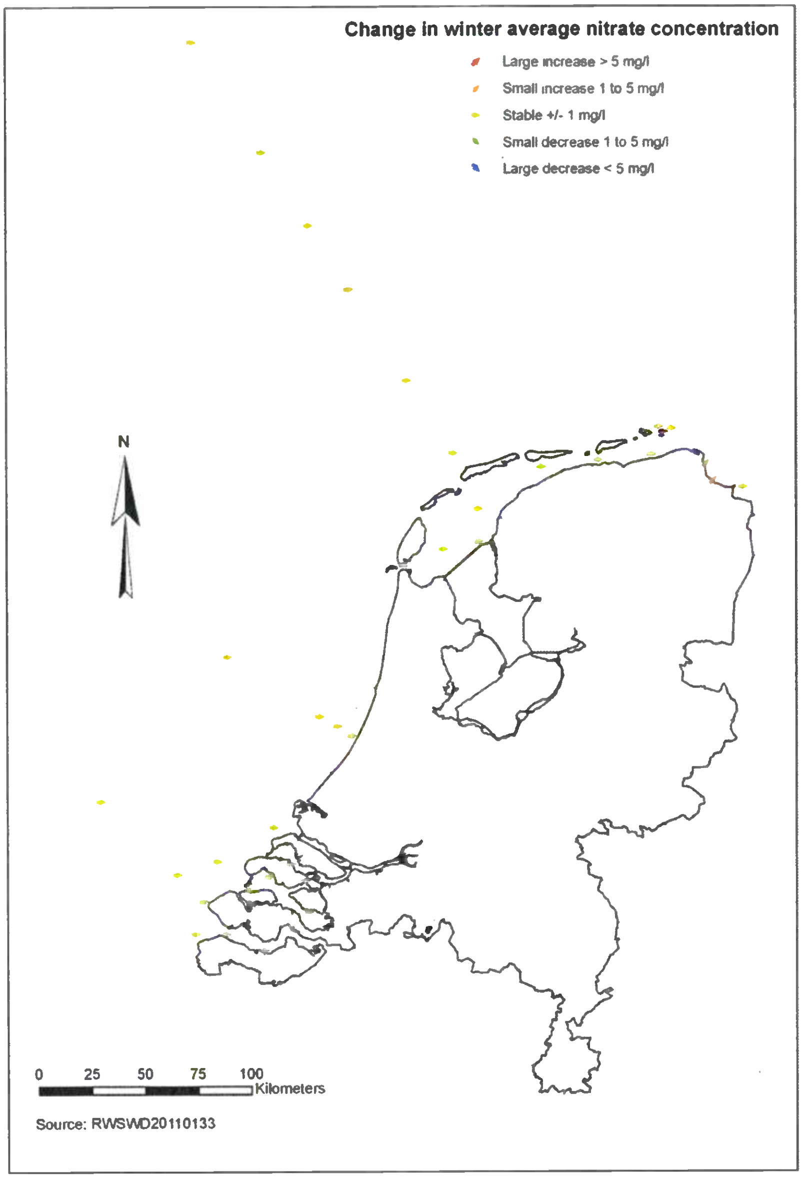
It is necessary to reduce indirect and direct nutrient emissions further, that is, in order to achieve a healthy marine environment free of eutrophication.

7.5 References

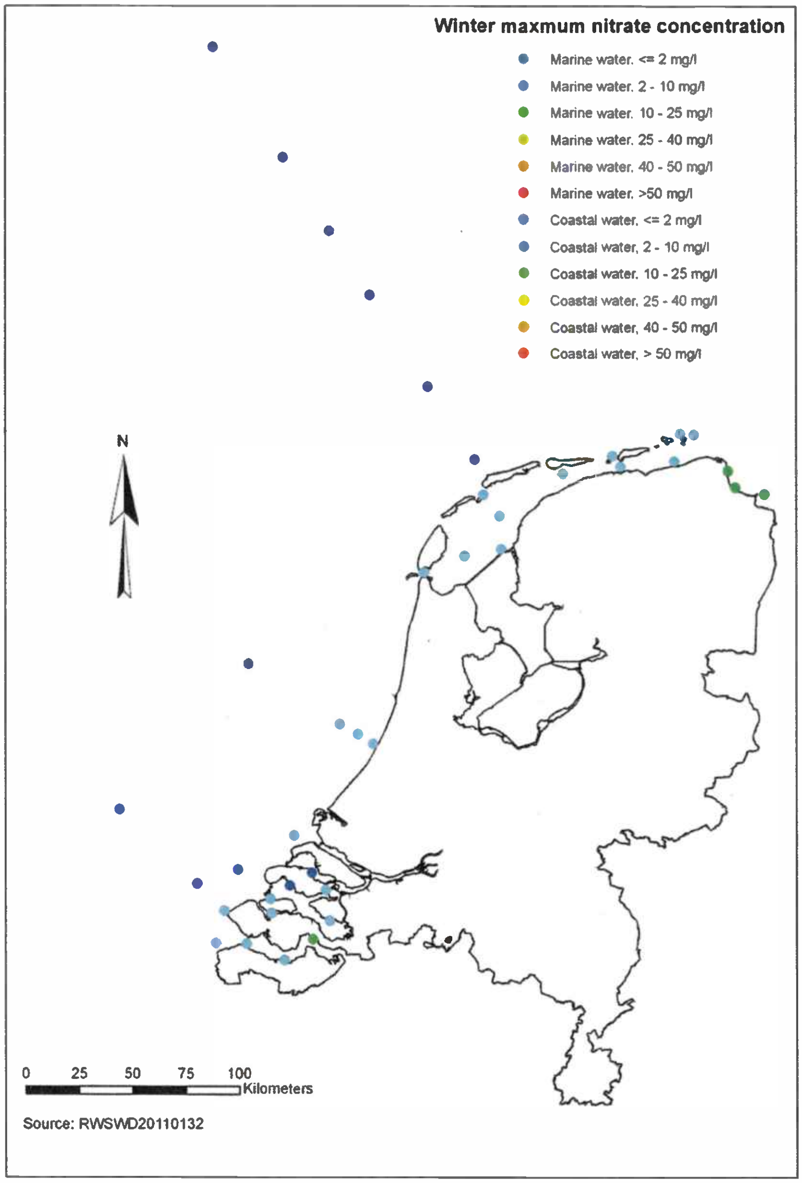
OSPAR (2010). Quality Status Report 2010. OSPAR Commission, London. 176 pp. (http://qsr2010.ospar.org/en/media/chapter_pdf/QSR_complete_EN.pdf)



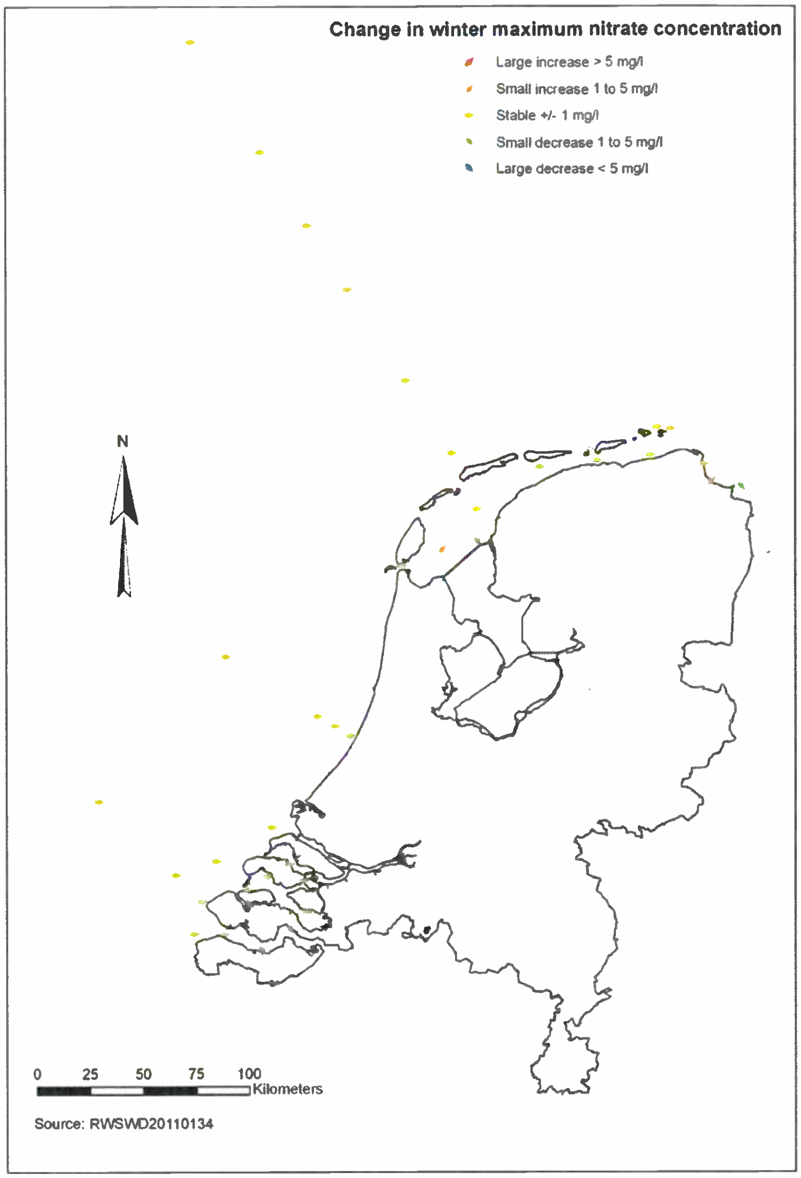
Map 7.1. Winter average nitrate concentration in Dutch marine and coastal waters in the period 2008-2010.



Map 7.2. Change in winter average nitrate concentration in Dutch marine and coastal waters between the fourth (2004-2007) and fifth (2008-2010) periods. Change shown here is the difference between the averages for the 2004-2007 and 2008-2010 periods.



Map 7.3. Winter maximum nitrate concentration in Dutch marine and coastal waters in the period 2008-2010.



Map 7.4. Change in winter maximum nitrate concentration in Dutch marine and coastal waters between the fourth (2004-2007) and fifth (2008-2010) periods. Change shown here is the difference between the averages for the 2004-2007 and 2008-2010 periods.

8 Future water quality development

8.1 Assessment of forecasting possibilities

It is exceptionally difficult to determine the time scale for changes in agricultural practice to result in changes in water quality. Groundwater travel times increase with the depth of the water, and as from a certain depth, these times exhibit enormous variation. Moreover, biological processes (e.g. denitrification and ammonification) and physical processes (e.g. dispersion, diffusion and dilution) lead to differences in water quality over time and from place to place, owing to the wide variety of physico-chemical characteristics of the saturated zones, aquifers and impermeable layers. Regional surface waters receive groundwater from different origins (agriculture, natural and urban areas) and of various ages. They are also fed by rainwater and sometimes waste water from, for example, farms, sewage treatment facilities or even industrial plants.

Travel times of water that leaches from root zones and that was studied under the LMM programmes are estimated to be less than five years (Meinardi and Schotten, 1999; Meinardi et al., 1998a, 1998b). It is therefore assumed that the effects of the fourth Action Programme (2010-2013) on the quality of the upper groundwater will become apparent between 2014 and 2019.

Travel times of groundwater at a depth of 5-15 metres in sand regions are on average 12 years, but range from less than 5 years to over 30 years (Meinardi, 1994). Travel times of groundwater at a depth of 15-30 metres are on average 36 years, but range from less than 25 years to over 80 years (Meinardi, 1994). In clay and peat regions, travel times are usually much longer, as the permeability of clay and peat aquifers is much lower than other types.

It will therefore be at least ten years before the effects of measures on nitrate concentrations in groundwater at a depth of 5-15 metres become apparent. Due to the large differences in travel times at any particular depth, nitrate concentrations will decrease slowly. In areas with confined aquifers and/or aquifers with a high denitrification capacity, nitrate concentrations are already low and will not change.

It will be at least several decades before the effects of measures to combat nitrate leaching at depths lower than 15 metres become apparent. This is certainly true concerning depths lower than 30 metres. Nitrate concentrations will change slowly due to the large variation in travel times at greater depths.

The effects of measures on nitrate concentrations in fresh surface waters strongly affected by agriculture will be discernible fairly quickly compared with their impact on nitrate concentrations in groundwater at a depth of more than 5 metres. Change in quality will probably be comparable to the effects on the upper groundwater of farms. Improvement in the quality of surface water in clay and peat regions will be comparable to that in the water that leaches from the root zones of farms, with the same improvement response as produced by the fourth Action Programme. The contribution of young groundwater (1-5 years old) to surface water in sand regions varies from less than 10% to more than 70%. It is therefore assumed that the effects of measures from the fourth Action Programme (2010-2013) on nitrate concentrations in fresh surface water will become apparent between 2014 and 2019. Because of mixing, it will probably be hard to distinguish the effects of the measures on nitrate concentrations from the effects of natural conditions on them. Examples of the latter are factors such as differences in precipitation.

Forecasting the future development of eutrophication due to agriculture is even more difficult than for nitrate concentrations, the main reasons being:
- the differences in surface waters with regard to their proneness to eutrophication;
- phosphorus concentrations and other factors such as hydromorphology, which also play an important part in the eutrophication process;
- contributions from other sources of nutrient input, such as urban wastewater and cross-border rivers;
- the extreme difficulty with predicting the response times of aquatic ecosystems to a substantial reduction in nutrient inputs and concentrations.

As well as source-oriented measures, regional effect-oriented measures, such as fish stock management, have been taken in situations offering good prospects for restoration and will be implemented further in the future. In some cases, the ecological restoration process was accelerated substantially (for example, the Veluwe border lakes). However, as the chlorophyll measurements reported in the previous sections reveal (Figures 6.3 and 7.5), the ecological restoration process in Dutch surface waters is generally progressing only slowly. An overall, clearly visible acceleration of this process is not expected in the short term.

8.2 Future water quality development

The report Evaluation of the Fertilisers Act 2012 (PBL, 2012) and the sub-report with the prognosis for the environment (Groenendijk et al., 2012) use simulation models to assess future development. Figure 8.1 presents the prognosis for the nitrate concentration in the upper metre of groundwater under farmland in sand regions (the most vulnerable). The prognosis includes corrections for variations in weather conditions.

PBL (2012) concludes that, for upper groundwater, tightening the nitrogen application standard for a number of crops, mainly on sandy soil, will lead to an overall reduction in average nitrate concentration from 2010 until the end of 2013. For the sand regions as a whole, the average nitrate concentration is predicted to fall to 50 mg/l (Figure 8.1; corrected results). The calculations also indicate that the nitrate targets for Sand North and Sand Central will easily be reached on average. After correcting for weather conditions, the groundwater quality should improve in Sand South and the loess region to an average of 70 and 60 mg/l respectively. Achieving the nitrate target is not yet in sight, however.

PBL (2012) states that the results from the model are uncertain, however, as they depend heavily on the assumptions for the further input of nitrate, as well as on the impact of weather. Concentrations calculated for the sand regions are slightly higher than the actual measurements. A reliable comparison between measurements and calculations from a model is difficult to make, though. First, not all farm types are monitored in all parts of the Netherlands. Second, land used for highly intensive or extensive farming is not included separately in the model, only the average being factored into the calculations.

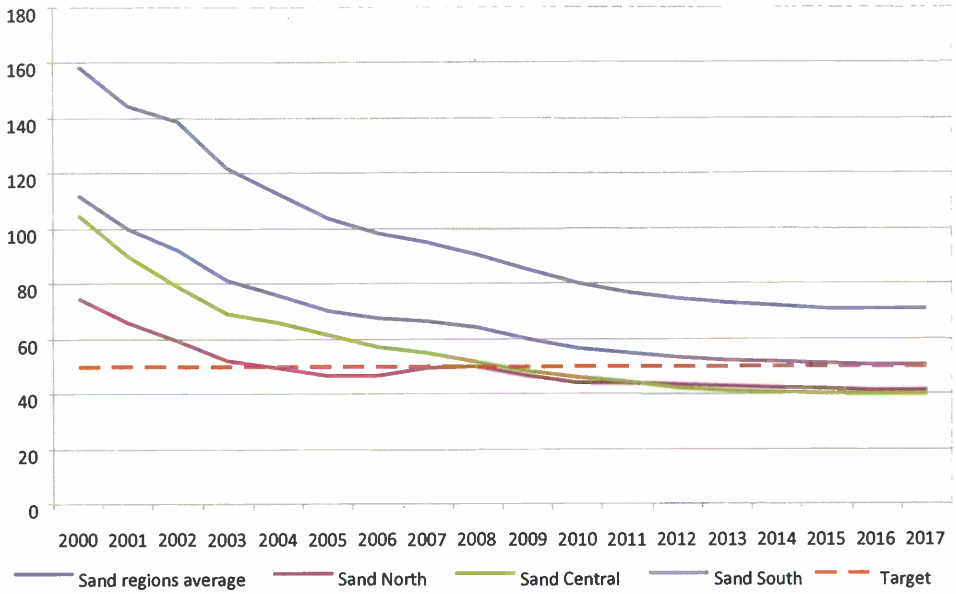


Figure 8.1. Calculated nitrate concentration in upper groundwater in sand regions, taking into account measures of the fourth Action Programme. Source: PBL (2012) / Alterra (2012).

Based on the calculated results from the model, the expectation for surface-water load from the run-off and leaching of nutrients is that it will reduce by 4% in the case of nitrogen and 2% in the case of phosphorus, compared with the levels for the 2010 application standards (PBL, 2012).

8.3 References

Groenendijk, P., Renaud, L.V., Schoumans, O.F., Luesink, H.H., De Koeijer T.J. and Kruseman, G. (2012). MAMBO en STONE-resultaten van rekenvarianten van gebruiksnormen. Evaluation of the Fertilisers Act 2012: final prognosis report. Wageningen, Alterra, Alterra Report 2317.

LNV (2003). Third Dutch Action Programme (2004-2007) for the Nitrates Directive; 91/676/EEC. The Hague, Ministry of Agriculture, Nature and Food Quality.

Meinardi, C.R. and Schotten C.G.J. (1999). Grondwateraanvulling en oppervlakkige afstroming in Nederland. Deel 3: De afwatering van veengebieden. Stromingen, 5 (1): 5-18.

Meinardi, C.R., Van den Eertwegh, G.A.P.H. and Schotten, C.G.J. (1998a). Grondwateraanvulling en oppervlakkige afstroming in Nederland. Deel 2: De afwatering van kleigronden. Stromingen, 4 (4): 5-19.

Meinardi, C.R., Schotten, C.G.J., De Vries, J.J. (1998b). Grondwateraanvulling en oppervlakkige afstroming in Nederland. Langjarig gemiddelde voor de zand- en leemgebieden. Stromingen, 4 (3): 27-41.

Meinardi, C.R. (1994). Groundwater recharge and travel times in the sandy regions of the Netherlands. Bilthoven, National Institute for Public Health and the Environment, RIVM Report 715501004.

PBL (2012) Summary report on the Evaluation of the Fertilisers Act 2012. PBL report number 500252.

Appendix 1. Agricultural areas by sector and region

Table B1. Agricultural areas (in 1,000 ha) for clay and sand regions by farmland category in LMM for the period 1992-2011.

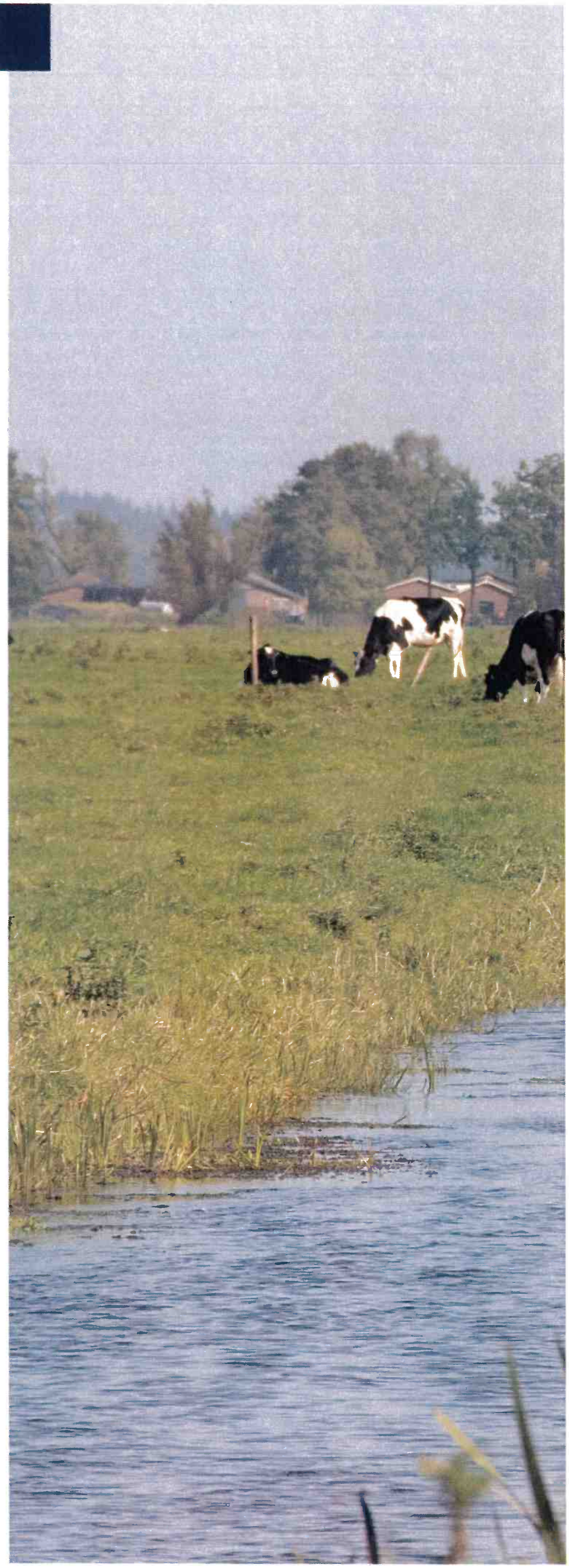
Clay regions	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011
Dairy	236	237	235	235	233	233	232	233	227	226	229	226	227	227	226	227	234	234	231	242
Arable	328	320	312	315	315	312	306	295	297	290	296	292	292	293	284	288	288	291	286	302
Other	64	71	71	73	73	72	76	81	86	87	94	93	94	94	99	103	102	96	83	85

Sandy regions	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011
Dairy	484	484	479	477	470	470	469	457	445	443	450	447	442	434	427	415	420	425	407	408
Arable	130	125	121	126	128	128	129	124	125	117	121	127	131	127	123	120	119	119	116	124
Factory farm animals	44	46	45	45	46	49	48	49	50	49	50	41	43	47	46	50	50	50	65	71
Other	139	146	147	149	150	156	158	155	167	158	167	157	157	161	166	169	168	157	140	125

Source: LEI, May census data processed by Statistics Netherlands.

R.A. Baumann et al.

RIVM report 680716008/2012



This is a publication of:

National Institute for Public Health and the Environment

P.O. Box 1 | 3720 BA Bilthoven
The Netherlands
www.rivm.nl

September 2012

004397

Available online at www.sciencedirect.com
ScienceDirect
Environmental Pollution 148 (2007) 797–807
ENVIRONMENTAL POLLUTION
www.elsevier.com/locate/envpol

Demonstrating trend reversal of groundwater quality in relation to time of recharge determined by $^3H/^3He$

A. Visser [a,*], H.P. Broers [b], B. van der Grift [b], M.F.P. Bierkens [a,b]

[a] Department of Physical Geography, Utrecht University, P.O. Box 80115, 3508 TC Utrecht, The Netherlands
[b] TNO Built Environment and Geosciences, Geological Survey of the Netherlands, P.O. Box 80015, 3508 TA Utrecht, The Netherlands

Received 25 January 2007; accepted 27 January 2007

Groundwater age dating reveals trends and trend reversal in groundwater quality.

Abstract

Recent EU legislation is directed to reverse the upward trends in the concentrations of agricultural pollutants in groundwater. However, uncertainty of the groundwater travel time towards the screens of the groundwater quality monitoring networks complicates the demonstration of trend reversal. We investigated whether trend reversal can be demonstrated by relating concentrations of pollutants in groundwater to the time of recharge, instead of the time of sampling. To do so, we used the travel time to monitoring screens in sandy agricultural areas in the Netherlands, determined by $^3H/^3He$ groundwater dating. We observed that concentrations of conservative pollutants increased in groundwater recharged before 1985 and decreased after 1990. Thereby, we demonstrated trend reversal of groundwater quality. From this research we concluded that $^3H/^3He$ dating can be used to facilitate (re)interpretation of existing groundwater quality data. The presented approach is widely applicable in areas with unconsolidated granular aquifers and large agricultural pressures on groundwater resources.

Keywords: Groundwater quality; Nitrate; Monitoring; Trends; Groundwater age; $^3H/^3He$

1. Introduction

Over the last 40 years, the development of intensive livestock farming in sandy areas in Noord-Brabant (in the south of the Netherlands) has greatly increased the use of agricultural contaminants (e.g. nitrate). These areas are vulnerable to agricultural contamination, because groundwater and contaminants can easily reach deeper parts of the aquifer.

Following the awareness of diffuse groundwater contamination, legislation was put into effect in the Netherlands to reverse upward trends of contaminant concentrations in shallow groundwater and bring concentrations down below quality standards. Both the EU Nitrates Directive (EU, 1991) and the Dutch Manure Law (1986) aim at reducing agricultural inputs to the groundwater system. It has also lead to the establishment of national and regional groundwater quality monitoring networks in the Netherlands, with the purpose of quantifying the magnitude of the contamination, to identify significant upward trends in concentrations and to guide the implementation of manure reduction legislation (Broers, 2002; Fraters et al., 2004; Van Duijvenbooden, 1993). The time series of these Dutch monitoring networks are now long enough to detect trends and trend reversals in concentrations of contaminants in recent groundwater, caused by increasing or decreasing agricultural pollution. In fact, the detection of trends in groundwater quality and the demonstration of trend reversal are key aspects of the new EU Groundwater Directive,

* Corresponding author. Tel.: +31 30 253 2779; fax: +31 30 253 2746.
E-mail addresses: a.visser@geo.uu.nl (A. Visser), h.broers@tno.nl (H.P. Broers), bas.vandergrift@tno.nl (B. van der Grift), m.bierkens@geo.uu.nl (M.F.P. Bierkens).

0269-7491/$ - see front matter © 2007 Elsevier Ltd. All rights reserved.
doi:10.1016/j.envpol.2007.01.027

which is supposed to become enacted in 2006 (EU, 2005). In line with the concepts used in the Groundwater Directive, we embraced the trend definition given by Loftis (1996; Loftis et al., 1991) who consider a temporal trend as a change in groundwater quality over a specific period in time, over a given region, which is related to land use or water quality management.

It is difficult to detect trends in the regional groundwater quality for a number of reasons (Broers and Van der Grift, 2004): (1) the uncertainty of the travel time of groundwater and contaminants to the monitoring screens; (2) attenuating processes retarding the arrival of contaminants at the monitoring screen, such as sorption or chemical reactions with the subsurface; (3) noise caused by short term temporal variation of concentrations, for example as a result of crop rotation; and (4) variation of concentrations in separate monitoring screens as a result of large scale spatial variation in inputs. The Dutch monitoring network consists of standardized multi-level monitoring wells with fixed screens at specific depths. Other monitoring networks may have less uniform well completion with unknown depth of borehole penetration, which is an additional complicating factor for trend analysis. To detect trends in regional groundwater quality, we need to find a way to aggregate the data from individual monitoring screens to a regional scale and, at the same time, reduce the effects of the aforementioned sources of error.

Earlier work on trend analysis for agricultural pollution problems used different ways of aggregating monitoring data. Foster et al. (1982) aggregated data from UK aquifers and related the observed concentrations to tritium levels and land use change. Frapporti (1994) aggregated data using a water type approach and was able to detect upward trends in nitrate concentrations using data of the Dutch National Monitoring Network. Broers and Van der Grift (2004) and Broers et al. (2004) aggregated time series data using data from fixed depth, assuming a relatively homogeneous travel time distribution with groundwater age increasing with depth in the aquifer. Bronswijk and Prins (2001) directly related groundwater concentrations in the Netherlands to the time of recharge. However, all of these studies were based on groundwater age determinations based on tritium measurements (Meinardi, 1994; Robertson and Cherry, 1989). Laier (2004) successfully used CFC groundwater dating (Busenberg and Plummer, 1992) to identify trends in individual monitoring wells.

In this study, we applied an alternative approach to trend analysis that is promising for the aggregation of time series in individual monitoring points to trends that represent larger areas or groundwater bodies. The approach is based on relating observed concentrations of contaminants in groundwater to the time of recharge of the sampled groundwater. As a check for consistency, we also reconstructed the historical concentrations of contaminants in recharging groundwater in agricultural recharge areas of the province of Noord-Brabant based on statistical data of atmospheric deposition, manure and fertilizer use and crop harvesting. This reconstruction of historical concentrations was compared with the recharge concentrations obtained from groundwater dating, leading to two independent assessments of possible trend reversal.

We analyzed the obtained relation between concentration and recharge year to: (1) demonstrate a trend reversal in

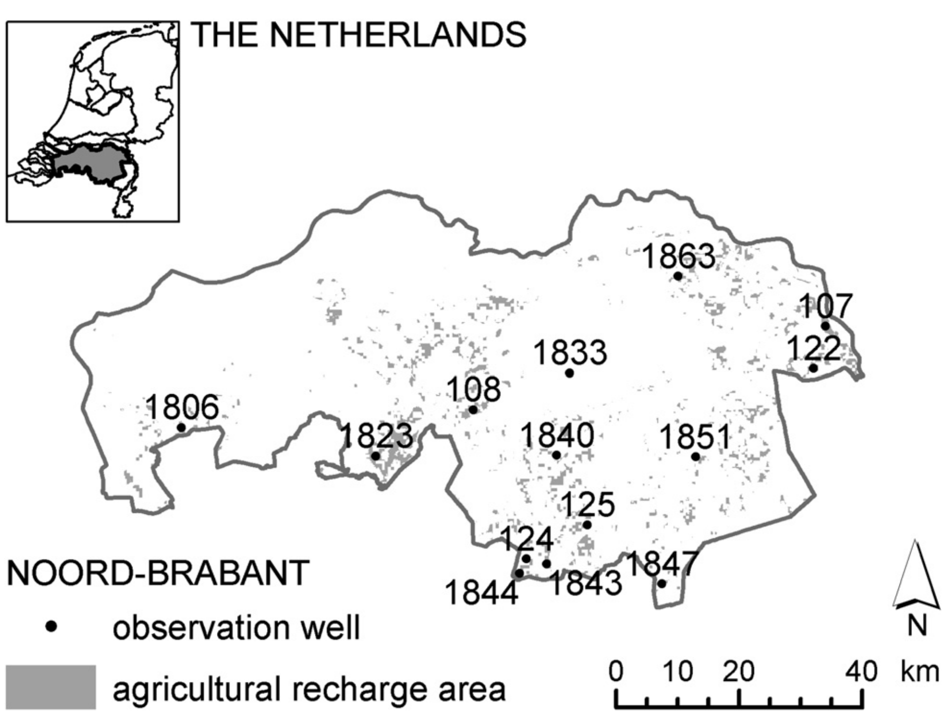


Fig. 1. Locations of 14 selected wells in agricultural recharge areas.

groundwater quality; and (2) to correlate the concentration – recharge year relation to the historical concentrations of agricultural contamination in recharging groundwater.

2. Methods

2.1. Study area and data

Noord-Brabant is one of the areas in Europe which is most affected by agricultural pollution, because intensive livestock farming in the area produces a large surplus of manure. Agricultural land covers 62% of the province of Noord-Brabant, which lies in the south of the Netherlands and has a total area of 5100 km^2. The agricultural area of the province of Noord-Brabant is vulnerable to diffuse groundwater pollution with nitrate because groundwater and contaminants can easily reach deeper parts of the aquifer. The subsurface consists of fluvial unconsolidated sand and gravel deposits from the Meuse River, overlain by a 2–30 m thick cover of Middle- and Upper-Pleistocene fluvio-periglacial and aeolian deposits consisting of fine sands and loam. In some areas pyrite occurs in the subsurface (Broers, 2004). Nitrate reduction by pyrite oxidation is described by the following reaction (Postma et al., 1991):

$$5FeS_2 + 14NO_3^- + 4H^+ \rightarrow 5Fe^{2+} + 10SO_4^{2-} + 2H_2O + 7N_2 \quad (1)$$

Noord-Brabant is a relatively flat area with altitudes ranging from 0 m above mean sea level (MSL) in the north and west to 30 m above MSL in the southeast. Groundwater tables are generally shallow, usually within 1–5 m below the surface. A natural network of brooks, which developed in equilibrium with a shallow groundwater table, drained the area before 1900. To allow for agricultural use of the poorly drained areas, the natural drainage was artificially extended during the 20th century, resulting in a dense network of ditches, drains and small watercourses.

The geohydrological situation was mapped to identify homogeneous recharge areas for the design of the groundwater quality network of Noord-Brabant (Broers, 2002). Homogeneous recharge areas lack a superficial drainage network; have relatively deep groundwater levels, relatively permeable soils and a relatively high topographical position. Monitoring wells were placed within the largest of the homogeneous areas, using stratified sampling (Broers and Van der Grift, 2004).

We selected the 14 monitoring wells in recharge areas (Fig. 1), such that the land use in the catchment area of each selected monitoring well was intensive livestock farming: mixed land use of mainly grassland and maize land. The wells consist of purpose built nested piezometers with a diameter of 2 inch and a screen length of 2 m at a depth of about 8 and 25 m below surface (Broers, 2002; Van Duijvenbooden, 1993). Three wells had an additional screen at about 4 meters below surface. The locations of the 31 screens we used are listed in Table 1.

Measurements of field parameters (pH, EC and dissolved oxygen), concentrations of major cations (Na, K, Ca, Mg, Fe, Al and NH_4), anions (Cl, NO_3, SO_4, HCO_3 and PO_4) and trace metals (i.e. Cd, Cu, Ni, and Zn) are collected annually as part of the national or provincial monitoring effort. For this paper, we have applied our method for trend demonstration to *nitrate* and *potassium*, and two additional chemical indicators: *sum of cations* and *oxidation capacity*. As mentioned before, nitrate can be reduced by pyrite in the subsurface, and downward transport of agricultural potassium is retarded compared to conservative chloride due to cation-exchange (Griffioen, 2001). The latter two composite chemical indicators are transported conservatively under most circumstances (Broers and Van der Grift, 2004).

The *sum of cations* is useful as a conditionally conservative indicator when cation-exchange processes dominate the transport of the cations and mineral dissolution does not occur. Moreover, the sum of cations is an indicator of the total load of solutes in the groundwater.

The *oxidation capacity* is defined as the weighted sum of molar concentrations of NO_3^- and SO_4 after Postma et al. (1991):

$$OXC = 5 * [NO_3^-] + 7 * [SO_4^{2-}] \quad (2)$$

OXC behaves conservatively during the process of nitrate reduction by pyrite oxidation assuming that no denitrification by organic matter occurs below the groundwater table. We assumed that sulfate reduction does not occur at relevant depths in these aquifers, based on Broers and Buijs (1997) who only found significant amounts of H_2S which indicate an ongoing process of sulfate reduction in the deepest parts of the aquifer, deep below the Fe/NO_3 redox cline.

Table 1
Well locations, screen depths and $^3H/^3He$ travel times

Location					^3H	$^3He^*$	Travel time
Well	Depth below surface	Screen	North	East			
#	m	#	deg°mm'ss″	deg°mm'ss″	TU	TU	years
107	7	1	51°37′52″	5°57′38″	7.9	5.7	10
107	24	3	″	″	11.2	36.5	26
108	8	1	51°30′38″	5°08′02″	10.1	4.8	7
108	24	3	″	″	14.3	66.2	31
122	9	1	51°34′10″	5°55′54″	9.2	3.7	6
122	24	3	″	″	8.5	22.4	23
124	8	1	51°17′33″	5°15′32″	8.6	4.8	8
124	26	3	″	″	8.4	7.6	11
125	12	1	51°20′33″	5°24′01″	9.7	10.5	13
125	23	3	″	″	10.6	32.0	25
1806	5	1	51°28′54″	4°26′59″	9.1	0.9	2
1806	23	3	″	″	12.4	28.9	21
1823	8	1	51°26′32″	4°54′22″	8.8	3.9	6
1823	18	3	″	″	10.4	10.3	12
1833	8	1	51°33′52″	5°21′31″	9.6	13.7	16
1833	19	3	″	″	14.2	118.9	40
1840	10	2	51°26′39″	5°19′44″	9.6	4.9	7
1840	24	4	″	″	8.9	37.2	29
1843	3.5	1	51°17′06″	5°18′21″	8.6	0.7	1
1843	8	2	″	″	9.8	2.3	4
1843	23	4	″	″	10.9	68.5	35
1844	4.5	1	51°16′17″	5°14′35″	9.3	1.3	2
1844	8	2	″	″	9.2	2.1	4
1844	23	4	″	″	12.6	36.4	24
1847	8	1	51°15′22″	5°34′27″	8.3	1.8	4
1847	24	3	″	″	15.4	64.1	29
1851	9	1	51°26′31″	5°39′14″	10.0	32.7	26
1851	23	3	″	″	21.6	129.3	35
1863	3.5	1	51°42′20″	5°36′49″	10.2	2.2	3
1863	10	2	″	″	15.7	67.1	30
1863	22	4	″	″	11.3	72.4	36

2.2. Groundwater age dating

In this study, we used $^3H/^3He$ groundwater dating (Schlosser et al., 1988; Tolstikhin and Kamenski, 1969) to determine the travel time of groundwater to the monitoring screen. $^3H/^3He$ groundwater dating is based on the radioactive decay of tritium and the containment of the decay product ^3He in groundwater. $^3H/^3He$ directly yields a travel time and can be applied to a single sample, whereas ^3H alone requires a depth profile to locate the ^3H-bomb peak (Schlosser et al., 1988). Groundwater travel times were determined from $^3H/^3He$ samples that we collected in 2001 and 2005 (Visser et al., 2007) and measured by the Bremen Mass Spectrometric Facility for the Measurement of Helium Isotopes, Neon, and Tritium in Water (Sültenfuß et al., 2004). The groundwater travel times were calculated from the ratio between tritiogenic helium and tritium (Table 1) as (Tolstikhin and Kamenski, 1969):

$$\tau = \lambda^{-1} \ln\left(\frac{^3He^*}{^3H} + 1\right) \quad (3)$$

with groundwater travel time (τ), tritium decay constant (λ) and measured tritium (^3H) and tritiogenic helium ($^3He^*$) concentrations in Tritium Units (TU). (1 TU represents a ratio of 1 atom of tritium per 10^{18} atoms of hydrogen and

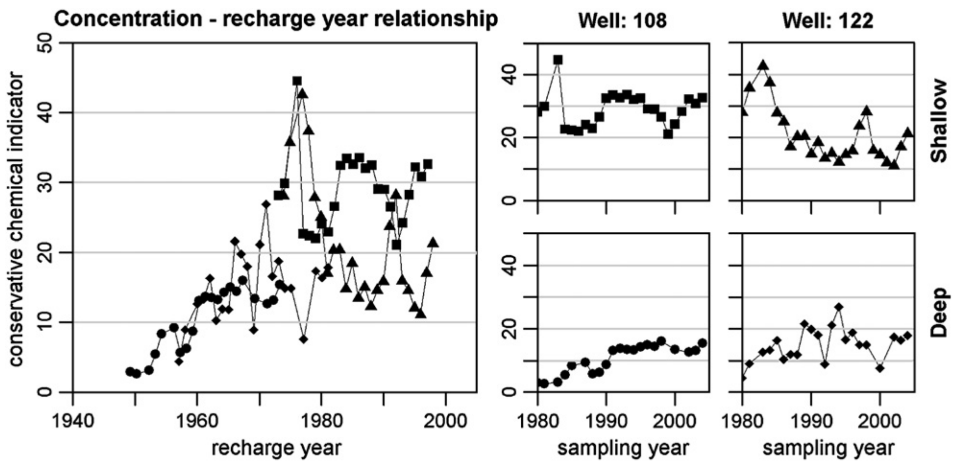


Fig. 2. The concentrations of a conservative chemical indicator (OXC) sampled from the shallow (8 m) and deep (24 m) screens of observation wells 108 and 122 (right) plotted at the recharge year of the sampled groundwater (left). The result is the *concentration − recharge year relationship*, from which a clear trend can be observed that was not visible in the individual time series.

corresponds to 2.488×10^{-12} cm^3 STP ^3He or 1.11×10^{-16} moles of ^3He per kg water: 1 TU of ^3H will decay to 1 TU of ^3He.)

2.3. Transposition of time series to time of recharge: constructing the concentration − recharge year relationship

We used the $^3H/^3He$ travel times to relate the observed concentration time series in the monitoring screens directly to the time of recharge. Thus we obtained a direct relation between all observed concentrations and a single independent variable: the time of recharge. With this relation, we aggregated the time series of concentrations from the national and regional monitoring network in the province of Noord-Brabant, the Netherlands.

As an example of how this works, we consider the time series of a conservative chemical indicator (OXC), collected from wells 108 and 122 of the national monitoring network (Fig. 2, right). Large variation exists in the individual time series and no clear trends can be observed. However, knowing the travel time to each screen, we can relate the observed concentration to the recharge year of the groundwater sample (Fig. 2, left). In this figure, an overall upward trend can be observed in groundwater that recharged before 1980. As such, groundwater travel time is a key variable to aggregate observed concentrations in individual monitoring screens.

We analyzed the resulting *concentration − recharge year* relationship in three ways. Firstly, we used a LOWESS smooth (Cleveland, 1979) to aggregate the median trend in all observations. The span parameter of the LOWESS smooth was 0.5 in all figures in this paper. Two extra LOWESS smooths were calculated through the residuals above and below the first LOWESS smooth. These are the upper and lower quartile of the conditional distribution of the concentrations as a function of the recharge year (Helsel and Hirsch, 1995). They indicate the spread and symmetry of the distribution of concentrations around the middle LOWESS smooth; fifty percent of the measured data falls between the upper and lower smooth.

Secondly, we correlated the three LOWESS smooths through all observations to the reconstructed historical concentrations in recharging groundwater, which were based on atmospheric deposition and manure statistics. A good correlation between reconstructed historical concentrations and the observed concentrations would indicate that future concentrations are well predictable.

Thirdly, to significantly determine trend reversal, two simple linear regression lines were fitted, one through all concentration data with a recharge time of before 1980 (''old water''), and one through all concentration data with a recharge time of after 1990 (''young water''). Trend reversal was considered significant if the regression line changed from significantly upward in ''old water'' to significantly downward in ''young water''.

2.4. Reconstruction of historical concentrations in recharging groundwater

To quantify the historical inputs, we constructed an annual mineral balance for the root zone of agricultural recharge areas in the province of Noord-Brabant. We considered four sources (*atmospheric deposition*, *animal manure*, *artificial fertilizer* and *lime*); and two sinks (*crop uptake* and *denitrification in the unsaturated zone*). We assumed that the surplus of this balance leaches to the groundwater. For a detailed description of the mineral balance we refer to the Appendix A.

A catchment to basin scale approach demands an averaged input curve for all agricultural land in a specific area, instead of the detailed information for individual land use types. In this case, we have constructed the aggregated

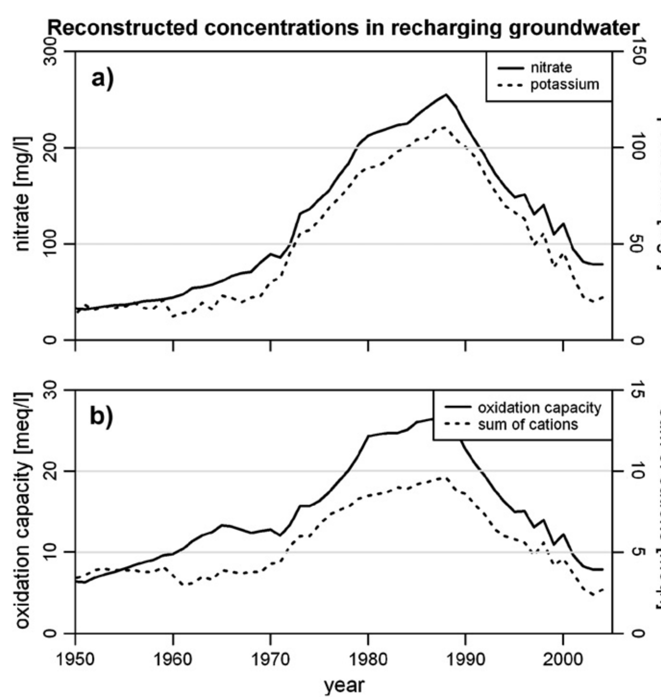


Fig. 3. Reconstructed historical concentrations in recharging groundwater based on the annual mineral balance (Appendix A) of nitrogen and potassium (a); and conservative chemical indicators (b). All indicators show a peak in the late 1980's when the use of manure was at it's maximum.

input curve for the recharge areas with intensive livestock farming – where more maize is present than on average in the province – to compare it to the measurements of groundwater quality in these vulnerable areas. To do so, we weighted the net deposited loads for each land use type by the relative area covered by that land use type in agricultural recharge areas.

We calculated the concentrations in recharging groundwater by dividing the annual mineral surplus by the average precipitation surplus since 1970. We did not consider using annual precipitation surpluses to calculate annual concentrations, because the reaction of shallow groundwater concentration on varying precipitation surplus is very non-linear. The counteracting effects of dilution and increased leaching at increased surplus depend heavily on processes in the unsaturated zone in previous years.

3. Results

3.1. Reconstructed historical concentrations in recharging groundwater

The most important change in land use was the introduction of maize in 1970 in the Netherlands. Maize is the only crop on which an unlimited amount of manure and fertilizer can be applied. The introduction of maize also incited the development of intensive livestock farming in Noord-Brabant. We plotted

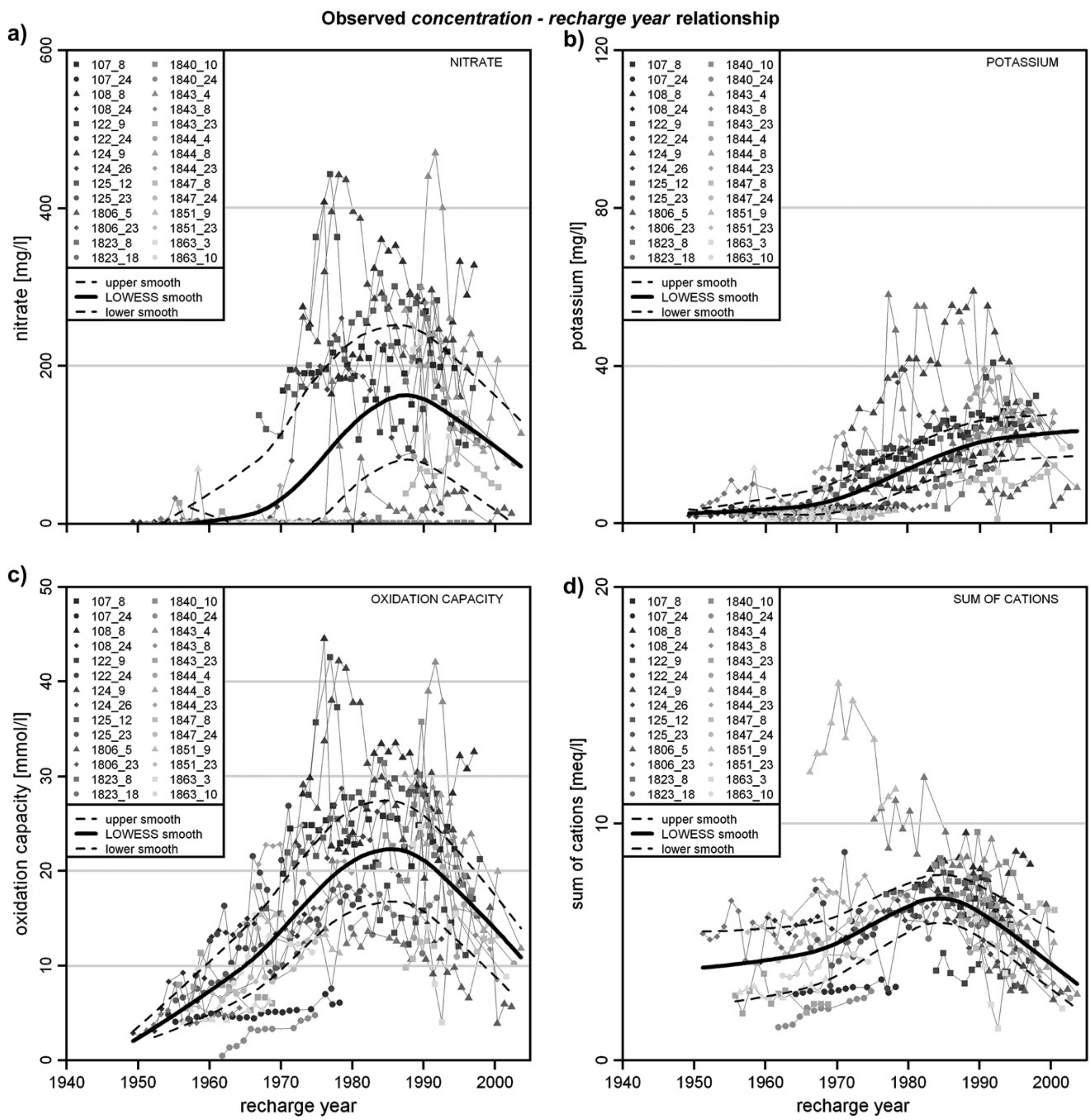


Fig. 4. The observed *concentration – recharge year relationship* of nitrate (a); potassium (b); oxidation capacity (c); and sum of cations (d). Measured concentrations are plotted in the year in which the sampled groundwater recharged, considering the groundwater travel time determined with $^3H/^3He$. A LOWESS smooth (black line) is used to show the overall trend in these data. Dashed black lines indicate the upper and lower LOWESS smooths, which indicate the spread of concentration data around the middle LOWESS smooth.

the reconstructed historical concentrations in recharging groundwater in Fig. 3. We found that the historical concentrations in recharging groundwater of nitrate and potassium (Fig. 3a) and oxidation capacity and sum of cations (Fig. 3b) increased between 1950 and the mid '80s as a result of increased application of manure and fertilizer. The highest reconstructed average nitrate concentration in recharging groundwater was over 250 mg/l in 1988. Concentrations decreased after 1990 as the result of the measures proscribed by the Dutch Manure Law (1986) and the EU Nitrates Directive (EU, 1991) aimed at reducing the application of manure, causing a reversal of the upward trend in historical concentrations in recharging groundwater. This reconstruction was aimed at capturing the trends in concentrations, rather than yielding accurate predictions of concentrations at specific years. The consistent regional approach, rather than on the plot-scale, makes this reconstruction useful for comparison with regionally aggregated data of concentrations in groundwater.

3.2. Trends in observed aggregated concentrations

Fig. 4 shows the concentration − recharge year plots for nitrate (a); potassium (b); oxidation capacity (c); and sum of cations (d). These figures include the data of 31 time series sampled from 2 or 3 depths in the 14 observation wells, plotted at the time of recharge determined by $^3H/^3He$. Individual time series show a large amount of temporal variation and a range of concentrations is present at every recharge year. We did not remove outliers, to show that this approach is robust and rather insensitive to outliers. For example, two time series of sum of cations are clearly affected by calcite dissolution and could be considered outliers, because one of the underlying assumptions

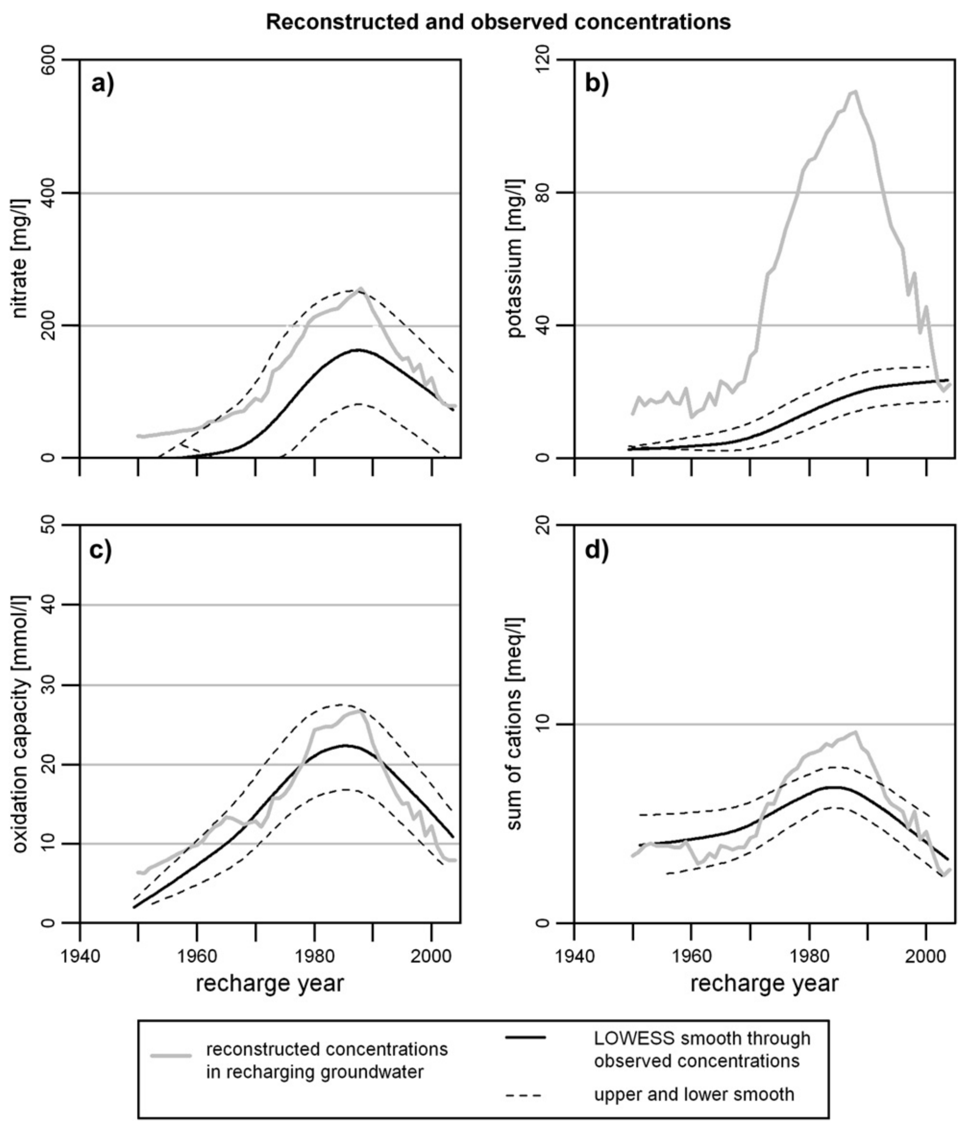


Fig. 5. Comparison of the LOWESS smooth through the observed *concentration − recharge year relationship* (solid black line with dashed upper and lower smooths) with reconstructed historical concentrations in recharging groundwater (grey line). Reactive substances (top) show large discrepancies with reconstructed concentrations, whereas conservative indicators (bottom) are consistent with the predicted trends.

for sum of cations – no mineral dissolution – is not valid. Nevertheless, the LOWESS smooth is hardly affected by the two time series. So despite the large amount of variation, aggregating the trends using upper, middle and lower LOWESS smooths revealed distinct trends of the concentrations with time.

We observed that the aggregated concentrations of *oxidation capacity* and *sum of cations* increased towards a peak in the mid 1980s, and decreased in younger groundwater (Fig. 4c,d). The individual measurements however showed a large variation around the LOWESS smooth. The upper and lower smooths indicate that 50% of the monitoring data is within a maximum bandwidth around the middle smooth of 10 mmol/l for oxidation capacity and 2 meq/l for sum of cations. Nevertheless, the upper and lower LOWESS smooths showed the same upward and downward trends as the middle LOWESS smooth.

Nitrate showed a similar pattern with a pronounced peak in 1985 and decreasing concentrations afterwards (Fig. 4a). However, the nitrate trends were probably affected by the reactivity of nitrate with the subsurface. Many time series of nitrate concentration in older water showed consistently low values, probably caused by complete denitrification by pyrite oxidation or organic matter in the saturated zone. Again the middle LOWESS smooth showed an upward trend before 1980, but the spread was much wider, caused by the complete denitrification in some of the time series. A downward trend was also present in young groundwater.

Potassium concentrations showed completely different trends (Fig. 4b). The upper, middle and lower LOWESS smooth of potassium were constantly increasing, even in young groundwater, but this trend was not yet reversed. Downward transport of agricultural potassium is retarded (compared to conservative chemicals) due to cation-exchange (Griffioen, 2001). Concentrations slowly increased with recharge year, and the maximum has not been reached. This is in agreement with reactive transport modeling of potassium by Broers and Van der Grift (2004) who showed that the potassium front slowly moves downward and that the maximum concentrations have not yet reached the uppermost monitoring screens.

To compare the reconstructed historical concentrations with the LOWESS smooths through the observed concentration – recharge year relationship, we plotted both in Fig. 5. The trends in the LOWESS smooth through the observed concentration recharge year relationship agreed well with the trends in the reconstructed historical concentrations in recharging groundwater of oxidation capacity and sum of cations (Fig. 5): upward before 1980, downward after 1990. In both cases, the reconstruction did overestimate the peak in observed concentrations, but this did not alter the agreement between the trends.

The reconstructed historical concentrations of nitrate in recharging groundwater structurally overestimated the LOWESS smooth through observed concentrations. This was caused by the removal of nitrate by denitrification in the saturated zone, which affected some of the time series. However, the median aggregated nitrate trend showed trend reversal around 1985, similar to conservative chemical indicators. The close resemblance between the reconstructed and observed trends in OXC concentration indicates that OXC acts as a conservative chemical indicator in our situation. From that we derive that our assumption (that the main mechanism of nitrate removal is the oxidation of pyrite) is valid here. Earlier studies showed the presence of abundant pyrite in the Noord-Brabant subsoil (Broers, 2002, 2004; Broers and Van der Grift, 2004) and the removal of nitrate by pyrite (Postma et al., 1991).

The reconstructed potassium concentrations largely overestimated the observed concentrations. Although the reconstructed concentrations showed a trend reversal in the '80s,

Table 2
Statistics of the correlation between reconstructed historical concentrations and LOWESS smooth through observed concentrations of nitrate, potassium, oxidation capacity and sum of cations

	Nitrate			Potassium		
	Lower LOWESS smooth	Middle LOWESS smooth	Upper LOWESS smooth	Lower LOWESS smooth	Middle LOWESS smooth	Upper LOWESS smooth
Correlation coefficient r	0.89	0.88	0.98	−0.01	−0.02	0.38
Significance	***	***	***	–	–	–
Intercept	−42.95	16.72	86.37	12.30	17.85	15.25
Slope	0.49	0.59	0.70	0.00	0.00	0.09
R^2	0.80	0.78	0.96	0.00	0.00	0.15
	Oxidation capacity			**Sum of cations**		
	Lower LOWESS smooth	Middle LOWESS smooth	Upper LOWESS smooth	Lower LOWESS smooth	Middle LOWESS smooth	Upper LOWESS smooth
Correlation coefficient r	0.97	0.97	0.95	0.97	0.96	0.95
Significance	***	***	***	***	***	***
Intercept	4.05	8.01	11.96	1.07	2.25	4.07
Slope	0.50	0.56	0.61	0.51	0.49	0.40
R^2	0.93	0.95	0.90	0.95	0.92	0.91

(Significance of t-test that Pearson's product-moment correlation coefficient = 0 ($P > |t|$); <0.001: ***; <0.005: **; <0.01: *; –.)

the aggregated median trend was increasing slowly into the 1990s. These results are in agreement with the results of Broers and Van der Grift (2004) who modeled the retarding effect of cation exchange between potassium, and calcium and magnesium described by Griffioen (2001).

We found an excellent correlation between the reconstructed historical concentrations in recharging groundwater and the middle LOWESS smooth through the *observed concentration − recharge year relationship*. Pearson's product-moment correlation coefficients (r) were as high as 0.97, and 0.96 for *oxidation capacity* and *sum of cations* respectively. These correlations were all significant at the 0.001-level. However, the relationships were not proportional. The regression lines had a considerable positive intercept and a slope angle of less than 1 (Table 2). This indicated that the reconstructed concentrations overestimated high concentrations and underestimated lower concentrations, as seen in Fig. 5. The high and significant correlation coefficients for oxidation capacity and sum of cations show that aggregation of multiple time series indeed reduces spatial and temporal variations.

3.3. Trend reversal

By fitting linear regression lines through the concentration − recharge year data (Fig. 6), we found significant upward trends in the concentrations of *nitrate*, *oxidation capacity* and *sum of cations* in "old water". These trends were reversed to significant downward trends in "young water" (Table 3) and therefore trend reversal is significant for conditionally conservative chemical indicators (*oxidation capacity* and *sum of cations*). Trend reversal is also significantly demonstrated for nitrate, despite the fact that nitrate removal by pyrite oxidation affected the absolute concentrations of nitrate. We found no significant trend in the concentrations of potassium in "young water", so trend reversal was not demonstrated

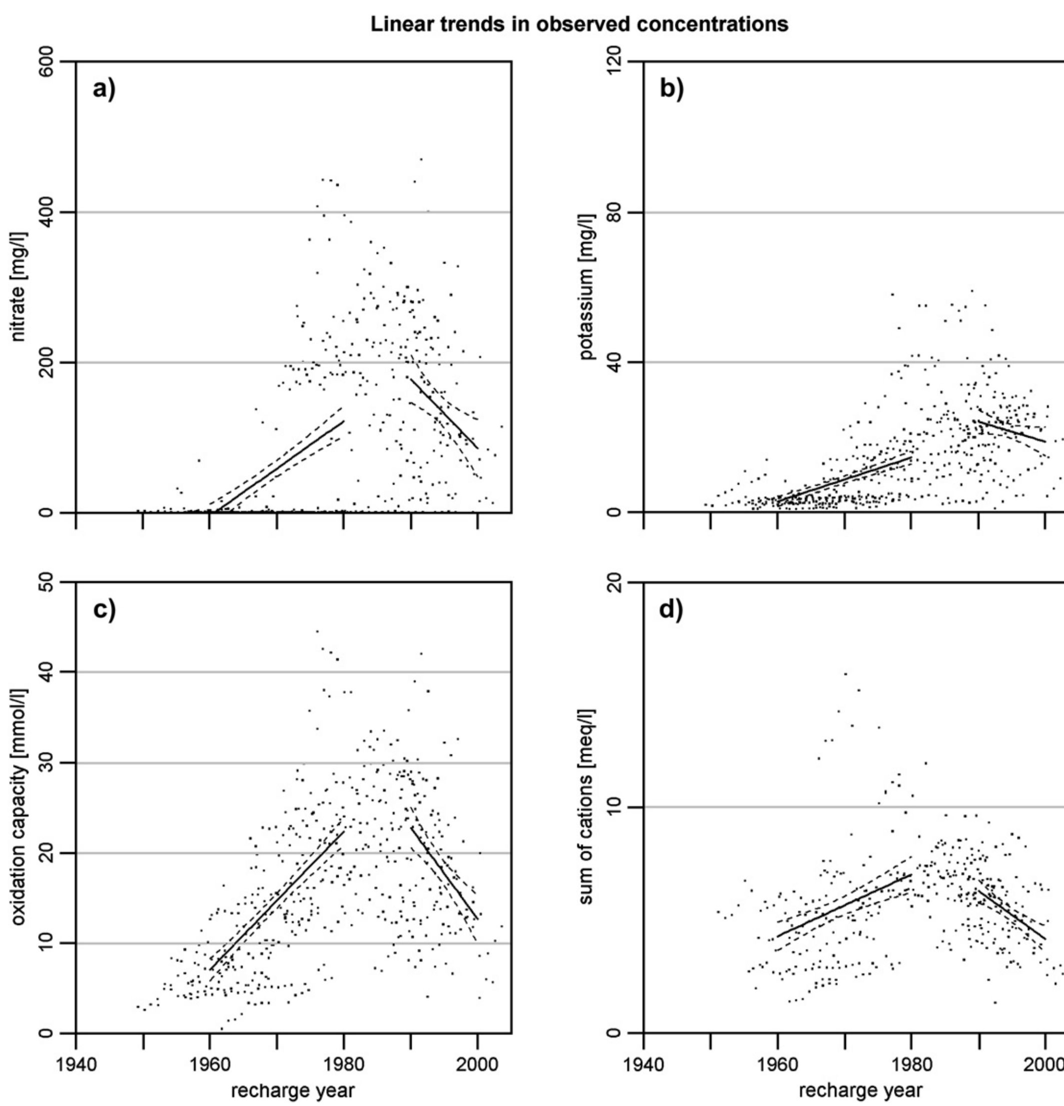


Fig. 6. Linear trends through *concentration − recharge year* data show significant trend reversal between 1980 and 1990 for nitrate (a); oxidation capacity (b); showed no significant trend reversal (c); and sum of cations (d). Potassium.

Table 3
Statistics of linear regression lines through concentration – recharge year data show significant trend reversal for nitrate, oxidation capacity and sum of cations

	Nitrate		Potassium	
Period	<1980	>1990	<1980	>1990
Slope	6.32	−9.17	0.59	−0.53
Significance	***	**	***	–
	Oxidation capacity		Sum of cations	
Period	<1980	>1990	<1980	>1990
Slope	0.77	−1.01	0.14	−0.21
Significance	***	***	***	***

(Significance of t-test that slope = 0 ($P > |t|$); <0.001: ***; <0.005: **; <0.01: *; –.)

for potassium. This is caused by its sorption to the subsurface, illustrating the behavior of reactive substances.

4. Discussion

We demonstrated trend reversal by relating the observed concentrations in groundwater to the time of recharge determined by $^3H/^3He$ dating. In this study, we aggregated all time series in agricultural recharge areas in the province of Noord-Brabant. With this approach we removed three of the four aforementioned difficulties with trend detection: (1) groundwater dating removes the variation in travel time towards the monitoring screen, and aggregation averages out both (3) short-term variations and (4) spatial variations. Attenuating and retarding processes that affect the arrival of contaminants at the monitoring screen (2) were partly removed by choosing two chemically conservative indicators, such as oxidation capacity and sum of cations to demonstrate trend reversal.

The advantages of relating time-series of groundwater quality to their time of recharge are 2-fold: (a) every time series provides information over a range of recharge years; and (b) data of multiple time series is available for each recharge year. Existing time series can thus be aggregated efficiently and trends are more easily observed from a limited number of time series, because spatial and temporal noise is reduced by aggregation. It can be applied to other (existing) datasets of groundwater quality, if the following conditions are plausible: (1) the travel times to the screens are constant. This is valid when no large-scale changes in the hydrological system take place; (2) the monitoring screen should be sufficiently short to be able to sample groundwater with distinct travel times. The screen should not sample a large range of travel times, which is usually the case with pumping wells or spring waters; (3) there exists a long term trend in input concentrations which spans many years, which is caused by diffuse agricultural pollution; and (4) dispersion or mixing along the groundwater flow lines is limited. The character of the aquifer should not cause mixing of older and younger groundwater, which might be the case in aquifers with dual porosity. All these conditions are fulfilled in the sandy agricultural areas of Noord-Brabant, and probably in many unconsolidated granular aquifers, which are typically threatened by agricultural pollution.

5. Conclusions

We showed that groundwater dating is useful to demonstrate trends in groundwater quality. Our method – relating observed concentrations to the time of recharge – can be used to demonstrate trend reversal, to correlate observed concentrations to the historical inputs and to improve future predictions of groundwater quality. By aggregating a large number of time-series, the method is robust and insensitive to outliers, subsurface heterogeneity, and spatial and temporal variation in inputs.

Acknowledgements

This work was supported by the European Union FP6 Integrated Project AquaTerra (Project no. GOCE 505428) under the thematic priority ‘‘sustainable development, global change and ecosystems’’. Alex Suckow (GGA-Hannover) is thanked for the instruction of the $^3H/^3He$ groundwater sampling and Jürgen Sültenfuss (Bremen University) for the discussions on $^3H/^3He$ data interpretation. Monique van der Aa (RIVM) and Alice Buijs (TNO) are thanked for their contributions to the 2001 sampling campaign. We thank two anonymous reviewers whose comments helped us improve the quality of the manuscript.

Appendix A. Mineral balance

We obtained estimates of *atmospheric deposition* from research done to quantify acidifying deposition by the Dutch National Institute for Public Health and the Environment (RIVM) and the Netherlands National Precipitation Chemistry Network (Buijsman, 1989). RIVM publishes annual model estimates of average national wet and dry deposition of nitrogen (NO_x and NH_y) and sulphur (SO_x) since 1980 (Bleeker and Erisman, 1996) and estimates of deposition in the province of Noord-Brabant for selected years. For the period before 1980, we used estimates by Stuyfzand (1991), who used a linear model to estimate atmospheric nitrogen and sulphur deposition from emission data for the period 1920–1980, calibrated on the period 1983–1987.

We based estimates of cation deposition on measurements of station 231 of the Dutch National Precipitation Chemistry Network located at the centre of Noord-Brabant. Data are available since 1978 (RIVM, 1979–1981; RIVM, 1982–1988; RIVM, 1989–1995), and on the website of RIVM since 1992 (RIVM, 2005). For the period before 1978, we used the average of the first five available years.

Because the application of manure and fertilizer depends on land use, we distinguished three land use categories: grass, arable land and maize. We obtained annual estimates of land use surface area for each category from the online database of Statistics Netherlands (CBS, 2006).

Animal manure is the main contributor of most minerals leaching to groundwater in Noord-Brabant. Because the concentrations of minerals in manure are not the same for all kinds of livestock, we constructed a detailed *manure* mineral balance for the province of Noord-Brabant, differentiating between types of livestock. CBS published figures on animal manure production since 1950 (CBS, 1976–1992). We obtained data from Statline (CBS, 2006) for the period 1994 to present. The figures on manure production were specified for dairy cows, calves, pigs (for meat production), sows (with piglets) and poultry.

We used data from Van der Grift and Van Beek (1996) who assembled data of the concentrations of minerals in manure, published by Evers (2000). The composition of animal manure was assumed to be constant in time, except for concentrations of N, and K which are published annually by CBS since 1980 (N) or 1995 (K) (CBS, 2006; Van Eerdt, 1999).

We considered the manure requirements and limitations of grass, maize and crops on arable land, given by the Commission Fertilizing Grassland and Crops (Hoeks, 2002). We assumed all animal manure is applied to agricultural land in Noord-Brabant in four steps. The first step is the application of 4/10 of the total dairy cow manure production on grassland, representing the deposition during the grazing season. Secondly, additional manure is applied during the rest of the year up to an equivalent of 180 kg K_2O/ha/yr. The amount of K_2O in applied manure may not exceed 180 kg K_2O/ha/yr because higher loads can cause diseases in cattle. Thirdly, the remainder of the manure is applied to arable land to a maximum of 250 kg N/ha/yr, from the different sources in the following order: remaining dairy cow manure, calves manure, pigs manure, sows manure and poultry manure. The application of manure to arable land is limited to a maximum load of 250 kg N/ha/yr from animal manure because most crops – except for maize, which is treated separately – do not tolerate higher loads. Finally, the manure surplus left after applying manure to the grassland and arable land is applied to maize land. No limit applies to the amount of manure used on maize.

The nationally applied amounts of artificial fertilizers (since 1950 reported by the Agricultural Economical Institute LEI (2006)) were distributed over the total national area of grassland, arable land and maize following the distribution key table (Table A1) published by CBS (1987). This national average of fertilizer application per unit area was converted to the amount of fertilizer applied in Noord-Brabant, by comparison with a study by Menke (1992), reporting fertilizer application for agricultural lands in Noord-Brabant for the years 1985 and 1990.

Table A1
Distribution of artificial fertilizers over national area of grassland, arable land and maize

Fertilizer type:	Distribution of artificial fertilizers over national area of:		
	Grassland	Arable land	Maize
Nitrogen	75%	25%	
Phosphor	30%	70%	
Potassium	20%	80%	

Table A2
Concentrations of minerals in crops

Crop	N	K	Ca	S
Grass	3.2%	2.1%	0.5%	0.3%
Potatoes	0.3%	0.02%	0.02%	0.03%
Sugar beets	0.5%	0.03%	0.1%	0.05%
Wheat	2.3%	0.2%	0.2%	0.3%
Maize	1.2%	1.0%	0.1%	0.1%

Van der Grift and Van Beek (1996) estimated the use of *lime fertilizers* since 1950. The Agricultural Economical Institute (LEI, 2006) published the national use of lime fertilizers since 1975 online. We used the regional estimates of lime fertilizer application by Menke (1992) to calculate the lime fertilizer application in agricultural recharge areas in Noord-Brabant.

Summing the aforementioned contributions of atmospheric deposition, manure, fertilizer and lime yielded the gross deposited loads. We then subtracted *crop uptake* from deposited loads, to calculate the composition of the water leaching from the root zone. We obtained historical crop yield data from CBS and from Van der Grift and Van Beek (1996) and converted these to crop mineral uptake using average mineral concentrations in crops published by Evers (2000) (Table A2). We assumed that on arable land the following three crops are used in equal and fixed proportions: potatoes, sugar beets and wheat.

We calculated *nitrogen uptake by crops* and *denitrification losses in the unsaturated zone* according to the solute prediction from regional distributed agricultural deposition (SPREAD) model, developed by Kiwa (Beekman, 1998).

The *annual surplus* of nutrients, cations and heavy metals deposited on the land surface since 1950, is calculated for each land use category (grassland, arable land and maize) as the sum of atmospheric deposition, manure, fertilizer and lime application minus the crop uptake and denitrification.

References

Beekman, W., 1998. Manual SPREAD: Prediction of Nitrate, Hardness, Chloride and Sulfate in Shallow Groundwater (in Dutch). Kiwa Research and Consultancy, Nieuwegein, the Netherlands. SWE 98.012.

Bleeker, A., Erisman, J.W., 1996. Deposition of Acidifying Components in the Netherlands in the Period 1980–1995 (in Dutch). National Institute for Public Health and Environment. RIVM, Bilthoven, the Netherlands. 722108018.

Broers, H.P., 2002. Strategies for Regional Groundwater Quality Monitoring. Ph.D thesis, Utrecht University, Utrecht, 231 pp.

Broers, H.P., 2004. Nitrate reduction and pyrite oxidation in the Netherlands. In: Razowska-Jaworek, L., Sadurski, A. (Eds.), Nitrate in Groundwaters, IAH Selected Papers on Hydrogeology. A.A. Balkema, Leiden, the Netherlands, pp. 141–149.

Broers, H.P., Buijs, E.A., 1997. Origin of Trace Metals and Arsenic at the Oostrum Well Field. Report NITG 97-189-A (in Dutch). Netherlands Institute of Applied Geosciences TNO, The Netherlands.

Broers, H.P., Van der Grift, B., 2004. Regional monitoring of temporal changes in groundwater quality. Journal of Hydrology 296 (1–4), 192–220.

Broers, H.P., Rozemeijer, J., van der Aa, M., van der Grift, B., Buijs, E.A., 2004. Groundwater Quality Trend Detection at the Regional Scale: Effects of Spatial and Temporal Variability, GQ2004, the 4th International

Groundwater Quality Conference. Bringing Groundwater Quality Research to the Watershed Scale. IAHS, Waterloo, Canada, pp. 50–62.

Bronswijk, H., Prins, H., 2001. Nitrogen fertilization and nitrate concentrations in deeper groundwater of the Netherlands (in Dutch). H_2O 34, 27–29.

Buijsman, E., 1989. Extensive Information About The Netherlands Air Quality Monitoring Network. I. The Netherlands National Precipitation Chemistry Network (in Dutch). National Institute for Public Health and the Environment, RIVM, Bilthoven, the Netherlands. 228703006.

Busenberg, E., Plummer, L.N., 1992. Use of chlorofluorocarbons (CCl_3F and CCl_2F_2) as hydrologic tracers and age-dating tools – the alluvium and terrace system of central Oklahoma. Water Resources Research 28 (9), 2257–2283.

CBS, 1976–1992. Manure Production: 1950/1974 – 1992. Statistics Netherlands (CBS), The Hague, the Netherlands.

CBS, 1987. Manure Production 1986. Staatsuitgeverij, The Hague, the Netherlands.

CBS, 2006. Statline Electronic Database (www.statline.nl). Statistics Netherlands (CBS), The Hague, the Netherlands.

Cleveland, W.S., 1979. Robust locally weighted regression and smoothing scatterplots. Journal of the American Statistical Association 74, 829–836.

Dutch Manure Law, 1986. Meststoffenwet (Manure law). Staatsblad 133, The Hague, the Netherlands.

EU, 1991. Nitrates Directive nr. 91/676/EEC.

EU, 2005. Common position Adopted by the Council with a View to the Adoption of a Directive of the European Parliament and of the Council on the Protection of Groundwater Against Pollution. Interinstitutional file 2003/0210 (COD), n. 12062/05, 9 November 2005.

Evers, M.A.A., 2000. Nutrient Manual (in Dutch). Nutrient Management Institute, Wageningen, the Netherlands, 296 pp.

Foster, S.S.D., Cripps, A.C., Smith-Carington, A., 1982. Nitrate leaching to groundwater. Philosophical Transactions of the Royal Society of London, Series B, Biological Sciences 296 (1082 The Nitrogen Cycle.), 477–489.

Frapporti, G., 1994. Geochemical and Statistical Interpretation of the Dutch National Groundwater Quality Monitoring Network. Ph.D. thesis, Geologica Ultraiectina no. 115, University of Utrecht, the Netherlands.

Fraters, B., et al., 2004. Agricultural Practice and Water Quality in the Netherlands in the 1992–2002 Period: Background Information for the Third EU Nitrates Directive Member States report. National Institute for Public Health and the Environment, Bilthoven, the Netherlands. 500003002.

Griffioen, J., 2001. Potassium adsorption ratios as an indicator for the fate of agricultural potassium in groundwater. Journal of Hydrology 254 (1–4), 244–254.

Helsel, D.R., Hirsch, R.M., 1995. Statistical Methods in Water Resources. In: Studies in Environmental Science, 49. Elsevier, Amsterdam, the Netherlands, 529 pp.

Hoeks, P.A., 2002. Advice on Fertilizing Grassland and Crops (in Dutch). Praktijkonderzoek Veehouderij, Lelystad, the Netherlands, 157 pp.

Laier, T., 2004. Nitrate monitoring and CFC-age dating of shallow groundwaters – an attempt to check the effect of restricted use of fertilizers. In: Razowska-Jaworek, L., Sadurski, A. (Eds.), Nitrate in Groundwaters, IAH Selected Papers on Hydrogeology. A.A. Balkema, Leiden, the Netherlands, pp. 247–258.

LEI, 2006. Agricultural Statistics (http://www3.lei.wur.nl/ltc/). Agricultural Economical Institute (LEI), Wageningen, the Netherlands.

Loftis, J.C., 1996. Trends in groundwater quality. Hydrological Processes 10, 335–355.

Loftis, J.C., McBride, C.B., Ellis, J.C., 1991. Considerations of scale in water quality monitoring and data analysis. Water Resources Bulletin 27, 255–264.

Meinardi, C.R., 1994. Groundwater Recharge and Travel Times in the Sandy Regions of The Netherlands. Ph.D. thesis, Free University, Amsterdam, the Netherlands, 211 pp.

Menke, M.A., 1992. Pressures of Fertilizers on Soils in the Netherlands (in Dutch). Delft Hydraulics, Delft, the Netherlands.

Postma, D., Boesen, C., Kristiansen, H., Larsen, F., 1991. Nitrate reduction in an unconfined sandy aquifer – water chemistry, reduction processes, and geochemical modeling. Water Resources Research 27 (8), 2027–2045.

RIVM, 1979–1981. Monitoring Network Assessing the Chemical Composition of Precipitation in the Netherlands: Annual Report (1978–1980). National Institute for Public Health and the Environment, RIVM, Bilthoven, the Netherlands.

RIVM, 1982–1988. Chemical Composition of Precipitation Over the Netherlands: Annual Report (1981–1987). National Institute for Public Health and the Environment, RIVM, Bilthoven, the Netherlands.

RIVM, 1989–1995. Netherlands Precipitation Chemistry Network: Monitoring Results (1988–1991). National Institute for Public Health and the Environment, RIVM, Bilthoven, the Netherlands.

RIVM, 2005. Chemical composition of precipitation over the Netherlands. Validated Data 1992-2004 (http://www.lml.rivm.nl/data_val/data/lmre_1992-2004.xls). National Institute for Public Health and the Environment, RIVM, Bilthoven, the Netherlands.

Robertson, W.D., Cherry, J.A., 1989. Tritium as an indicator of recharge and dispersion in a groundwater system in Central Ontario. Water Resources Research 25, 1079–1109.

Schlosser, P., Stute, M., Dorr, H., Sonntag, C., Munnich, K.O., 1988. Tritium He-3 dating of shallow groundwater. Earth and Planetary Science Letters 89 (3–4), 353–362.

Stuyfzand, P.J., 1991. The Composition of Rain Water Along the Coast of Holland (in Dutch). KIWA, Nieuwegein, the Netherlands. SWE 91.010.

Sültenfuß, J., Roether, W., Rhein, M., 2004. The Bremen Mass Spectrometric Facility for the Measurement of Helium Isotopes, Neon, and Tritium in Water. IAEA, Vienna. IAEA-CN-119/7. IAEA-CN-119/7.

Tolstikhin, I.N., Kamenski, I.L., 1969. Determination of groundwater age by the T-^{3}He method. Geochemistry International 6, 810–811.

Van der Grift, B., Van Beek, C.G.E.M., 1996. Hardness of Abstracted Groundwater: Indicative Predictions (in Dutch). Kiwa, Nieuwegein, the Netherlands.

Van Duijvenbooden, W., 1993. Groundwater quality monitoring in the Netherlands. In: Alley, W.M. (Ed.), Regional Groundwater Quality. Van Nostrand Reinhold, New York, pp. 515–535.

Van Eerdt, M.M., 1999. Manure Production and Mineral Secretion (in Dutch), Environmental Quarterly. Staatsuitgeverij/CBS-Publikaties, 's-Gravenhage, pp. 27–31.

Visser, A., Broers, H.P., Bierkens, M.F.P., 2007. ^{3}H/^{3}He dating of groundwater degassed by denitrification. Water Resources Research, submitted for publication.

Comparison of methods for the detection and extrapolation of trends in groundwater quality

Ate Visser,*[ab] Igor Dubus,†[c] Hans Peter Broers,[a] Serge Brouyère,[d] Marek Korcz,[e] Philippe Orban,[d] Pascal Goderniaux,[d] Jordi Batlle-Aguilar,‡[d] Nicolas Surdyk,[c] Nadia Amraoui,[c] Hélène Job,[c] Jean Louis Pinault[c] and Marc Bierkens[ab]

Received 31st March 2009, Accepted 3rd August 2009
First published as an Advance Article on the web 7th September 2009
DOI: 10.1039/b905926a

Land use changes and the intensification of agriculture since the 1950s have resulted in a deterioration of groundwater quality in many European countries. For the protection of groundwater quality, it is necessary to (1) assess the current groundwater quality status, (2) detect changes or trends in groundwater quality, (3) assess the threat of deterioration and (4) predict future changes in groundwater quality. A variety of approaches and tools can be used to detect and extrapolate trends in groundwater quality, ranging from simple linear statistics to distributed 3D groundwater contaminant transport models. In this paper we report on a comparison of four methods for the detection and extrapolation of trends in groundwater quality: (1) statistical methods, (2) groundwater dating, (3) transfer functions, and (4) deterministic modeling. Our work shows that the selection of the method should firstly be made on the basis of the specific goals of the study (only trend detection or also extrapolation), the system under study, and the available resources. For trend detection in groundwater quality in relation to diffuse agricultural contamination, a very important aspect is whether the nature of the monitoring network and groundwater body allows the collection of samples with a distinct age or produces samples with a mixture of young and old groundwater. We conclude that there is no single optimal method to detect trends in groundwater quality across widely differing catchments.

1. Introduction

Land use changes and the intensification of agriculture since the 1950s have resulted in increased pressures on natural systems. For example, the diffuse pollution of groundwater with agricultural contaminants such as nitrate and pesticides has resulted in a deterioration of groundwater quality.[1–3] The surplus of nitrogen applied to agricultural land, and possibly leaching to groundwater, shows similar trends for most European countries and the United States (Fig. 1): upward until the late 1980s and stabilization or decreasing trends afterward. The transfer of these contaminants to deeper groundwater and surface water represents a major threat to the long-term sustainability of water resources across the Europe and elsewhere.[4]

For the protection of groundwater quality in the European Union (EU), the Water Framework Directive [WFD, ref. 5] and the Groundwater Directive [GWD, ref. 6] require member states

[a]*Deltares/TNO—Geological Survey of the Netherlands, Division of Soil and Groundwater systems, P.O. Box 85467, 3508 AL Utrecht, The Netherlands. E-mail: ate.visser@deltares.nl; Fax: +31 30 256 4855; Tel: +31 30 256 4816*

[b]*Utrecht University, Department of Physical Geography, P.O. Box 80115, 3508 TC Utrecht, The Netherlands*

[c]*BRGM, Water Division, BP 6009, 45060 Orléans Cedex 2, France*

[d]*HG-ULg, Aquapôle, University of Liege, Building B52/3, 4000 Sart Tilman, Belgium*

[e]*Institute for Ecology of Industrial Areas, ul. Kossutha 6, 40-844 Katowice, Poland*

† *Present address*: FOOTWAYS, 10 avenue Buffon, 45071 Orleans Cedex 2, France.

‡ *Present address*: Ecological Engineering Laboratory, École Polytechnique Fédérale de Lausanne (EPFL), Station 2, Building GR, CH-1015, Lausanne, Switzerland.

Environmental impact

The transfer of diffuse agricultural contaminants to deeper groundwater and surface water represents a major threat to the long-term sustainability of water resources.

To identify possible threats to future water quality, it is essential to detect sustained upward trends in the concentrations of pollutants in an early stage. Trend detection is often complicated by limited monitoring data, variations in the transport of contaminants toward monitoring locations, variations in application of contaminants in space and time, and (partial) degradation of contaminants in the subsurface. This work provides the scientific basis for choosing the method for trend detection in the framework of environmental legislation by assessing the capabilities and efficiency of various tools to detect and extrapolate trends in groundwater quality.

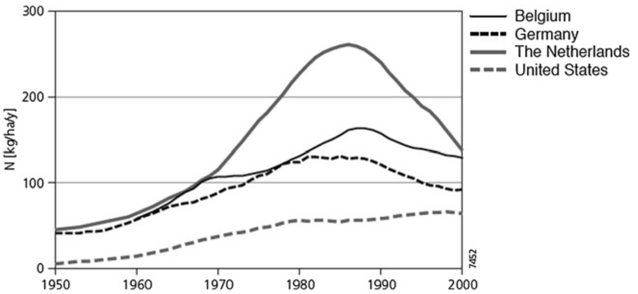


Fig. 1 Estimates of nitrogen deposition on agricultural land in Belgium (data courtesy of Gembloux Agricultural University), Schleswig-Holstein, Germany (data from ref. 89), Noord-Brabant, the Netherlands (data from ref. 32), and the United States (including pasture land, data courtesy of USGS).

to achieve good chemical status of their groundwater bodies by the year 2015. To achieve this, these directives ask member states to delineate groundwater bodies and assess the present chemical status of these groundwater bodies. To detect possible threats to future groundwater quality, the GWD asks for the detection of sustained upward trends in the concentrations of pollutants. If upward trends are found, these should be reversed when the concentration of the pollutant reaches 75% of the threshold value. The GWD also lays down requirements on the implementation of measures necessary to reverse any significant and sustained upward trend. In the context of the WFD and GWD, the scientific community will be asked the following with respect to groundwater quality: (1) assess the current status, (2) detect changes or trends, (3) assess the threat of deterioration by relating these trends to historical changes in land use, and (4) predict future changes by extrapolating present day trends and possibly predict trend reversal in response to legislation.

While the WFD requires groundwater quality monitoring networks to be operational by the year 2007, the awareness of the threat to groundwater quality has already led to the installation of monitoring networks in many countries such as Korea,[7,8] Denmark,[9] the Netherlands,[10] New Zealand,[11] Palestine,[12] the US,[13,14] and the UK.[15] These networks have since produced time series of monitoring data which have been used to detect and quantify changes in groundwater quality.[16–23]

These studies showed that trends in groundwater quality are difficult to detect. Most often the period of interest is longer than the period of record[24] and available time series are typically rather short and sparse because of the high costs of sampling and analysis. The lack of substantial data usually limits the application of statistical methods to simple approaches rather than more complex time series analysis tools. Other factors complicating trend detection are: variations in the duration and pathways of the transport of contaminants toward monitoring location by groundwater flow, variations in application of contaminants at the ground surface, in space and time, and (partial) degradation of contaminants in the subsurface.[20] Additionally, the travel time of sampled groundwater may be uncertain, in particular because the groundwater sample may represent a range of travel times. Whether the character of the groundwater flow system causes mixing of groundwater, for example in dual-porosity systems, and whether the groundwater sample contains a mixture of groundwater, for example from springs or production wells, are important factors for the success of trend detection in groundwater quality. This difference can influence the sampled concentration of nitrate, because the mixture sampled from a supply well contains a portion of old, pre-agricultural groundwater with no nitrate.[4] Data from supply wells should therefore be regarded as a sampling of a different sub-population.[25] In situations where samples contain groundwater with a distinct age, several studies have used groundwater age tracers to enhance the interpretation of measured concentrations of pollutants.[26–32]

To assess whether an upward trend in the concentration of a contaminant will threaten groundwater quality, a variety of tools can be used to detect and extrapolate trends in groundwater quality. These range from linear statistics[18,20,22] relating trends to changing land use patterns[33–36] and predict future trends based on land use scenarios;[37,38] trends in groundwater quality can also be predicted using empirical, functional or deterministic models of varying complexity.[39–41] The efficiency of these tools depends on several factors like the availability of groundwater quality data, the character of the groundwater flow system, and the available resources for trend assessment.

To address the challenges of the WFD, a number of working groups were asked to design a common implementation strategy (CIS) and produce "guidance documents" for the implementation of the WFD. The mandate of the Groundwater Working Group (WGC) required the development of practical guidance and technical specifications for the derivation of threshold values, the assessment of status compliance (both quantitative and chemical) and the assessment of groundwater trends and trend reversal. The focus of the guidance document was on "statistical aspects of the identification of groundwater pollution trends, and aggregation of monitoring results".[42] Scientific and technological advances in the meanwhile are expected to improve the success and efficiency of monitoring programs by the development and testing of novel techniques for trend detection.[43]

The aim of our work was to provide the scientific basis of science-policy integration needs [*e.g.* ref. 44], by assessing the capabilities and efficiency of various tools to detect and extrapolate trends in groundwater quality. We compared several novel techniques to detect trends in groundwater quality, that are not included in the EU guidance documents to a statistical approach for trend detection, as discussed in the EU guidance.[45] The following four approaches for the detection and extrapolation of trends in groundwater quality were included in the analysis: (1) statistical methods, (2) groundwater dating, (3) transfer functions, and (4) deterministic modeling. The comparison was based on the analysis of monitoring datasets at four different locations in a variety of different groundwater systems, ranging from unconsolidated unconfined aquifers to fissured dual-porosity systems, representing different types of hydrogeological settings.

2. Four approaches to trend analysis

2.1 Statistical trend detection and estimation

The success of a statistical trend analysis largely depends on selecting the right statistical tools[46] considering various aspects of the available data: whether the data are normally distributed or can be described by an alternative distribution function, whether the data contain seasonality,[47] whether the trend is monotonic or abrupt,[48] and whether the trends are expected to be univariate or multivariate.[49] Loftis[24] suggested that a clear definition of "trend" should be adopted before analyzing the data. Here we define a temporal "trend" as *a significant change in groundwater quality over a specific period of time, over a given region, which is related to land use or water quality management*.

The aim of the statistical methods deployed on three of the test sites was to detect and estimate statistically significant changes in the concentrations of contaminants over time. The methods had to be robust and applicable to typical groundwater quality time series, with a limited amount of data, a rather short observation period with possibly missing data, often non-normally distributed, either annually sampled or containing seasonal trends. To meet these requirements, a three-step procedure was adopted[22] following Hirsch *et al.*[48] First, time series were tested for normality; second, the presence of a trend was assessed; and third, the slope of the trend was estimated. The procedure (Fig. 2) was applied to various time series from different study sites.

To test the data for normality, the Shapiro–Wilks test[50] was applied to datasets with less than 50 records, while the Shapiro–Francia test[51] was applied to datasets containing 50 or more records. On time series which were normally distributed, a linear regression was performed. The correlation coefficient was used to assess the robustness of the trend.[52] On time series with a non-normal distribution, the non-parametric Mann–Kendall test,[53,54] was performed. This test is commonly used in hydrological sciences since its appearance in the paper by Hirsch *et al.*[47] as it is rather insensitive to outliers[55] and has recently been proven as powerful as the Spearman's $\bar{\rho}$ test.[56] If a significant trend was detected (based on a 95% significance level), the slope of the trend was determined as the slope of the linear regression equation for normally distributed time series, or using Kendall's slope for non-normal time series.[48] To aggregate the trend analysis over the entire groundwater body, the number of significant trends was expressed as a percentage. Additional analyses could include the determination of the median trend, or the spatial distribution of trends across groundwater bodies. This approach was applied using various commercial or open-source statistical software packages.

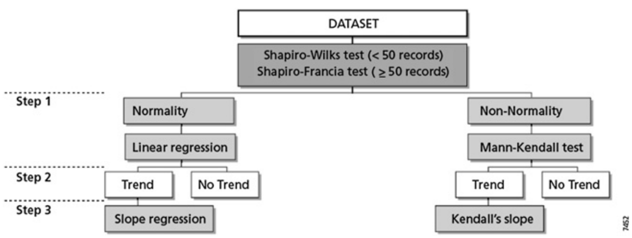


Fig. 2 A three-step procedure is adopted for statistical trend analysis of contaminant concentrations in the selected groundwater bodies: (1) normal/non-normal distribution data; (2) trend detection; (3) trend estimation (after ref. 22).

2.2 Groundwater dating

Groundwater dating as a tool to aid trend detection was applied on all four datasets. Groundwater dating requires the possibility to accurately sample a range of groundwater age tracers, preferably ^{3}H/^{3}He,[57] or CFCs[58] and/or SF_6.[59] If these gaseous tracers are impractical, a qualitative approach based on ^{3}H measurements alone can be applied to distinguish between old (recharged prior to 1950) and young (recharged after 1950) groundwater. The aim of groundwater dating is to remove the travel time of groundwater as a complicating factor for trend analysis and aggregate monitoring data from wells across groundwater bodies. This was obtained by relating measured concentrations directly to the time of recharge (Fig. 3).

2.3 Transfer functions to predict future trends

The aim of the transfer function approach deployed in the dataset from the Brévilles catchment in France was to detect and extrapolate trends in the concentrations of agricultural contaminants in macro-porous or dual-porosity systems where concentrations are strongly correlated to other hydrological

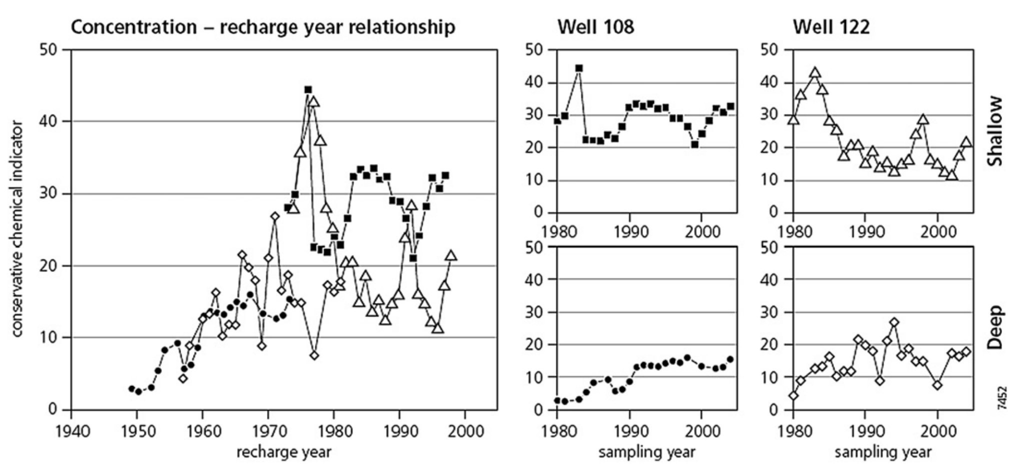


Fig. 3 Example of the use of groundwater dating as a tool to aid trend detection. The concentrations of a conservative chemical indicator (OXC) sampled from the shallow (8 m) and deep (24 m) screens of two observation wells. Concentrations are plotted at the time of sampling (right) and plotted at the recharge year of the sampled groundwater (left). The result is the concentration–recharge year relationship, from which a clear trend can be observed that was not visible in the individual time series related to sampling year (after ref. 32).

parameters, such as precipitation or stream flow. Following the approach described by Pinault *et al.*,[60] transfer functions were identified and applied using the TEMPO tool[61] which is capable of modeling time series through iterative calibrations of combinations of transfer functions. The effect of transformation processes on the contaminant concentrations was implicitly incorporated in the transfer functions.

2.4 Physical-deterministic modeling

The aim of using physical-deterministic models was to predict future trends in groundwater quality in systems of varying complexity considering changes in the pressure applied to the groundwater system. Distributed groundwater flow and transport models were developed separately and specifically for three sites. Transport models included advective transport, hydrodynamic dispersion and, where necessary, dual-porosity effects, sorption and degradation of contaminants. Predictions of future concentrations were based on scenarios of land use and agricultural application of fertilizer and pesticides, as well as climate scenarios.

3. Test sites and datasets

Groundwater bodies were selected at four distinctly different locations to evaluate trend detection methods: the Dutch part of the Meuse basin, a number of catchments in the Belgian part of the Meuse basin, the Brévilles catchment in France and the German Bille-Krückau watershed in the Elbe basin (Fig. 4). The characteristics of each of the test sites and available data are summarized in Table 1 and described in the following sections. The test sites vary strongly in geohydrological characteristics and were studied in different contamination settings (nitrate, pesticides). The contaminants under study were the ions of nitrate

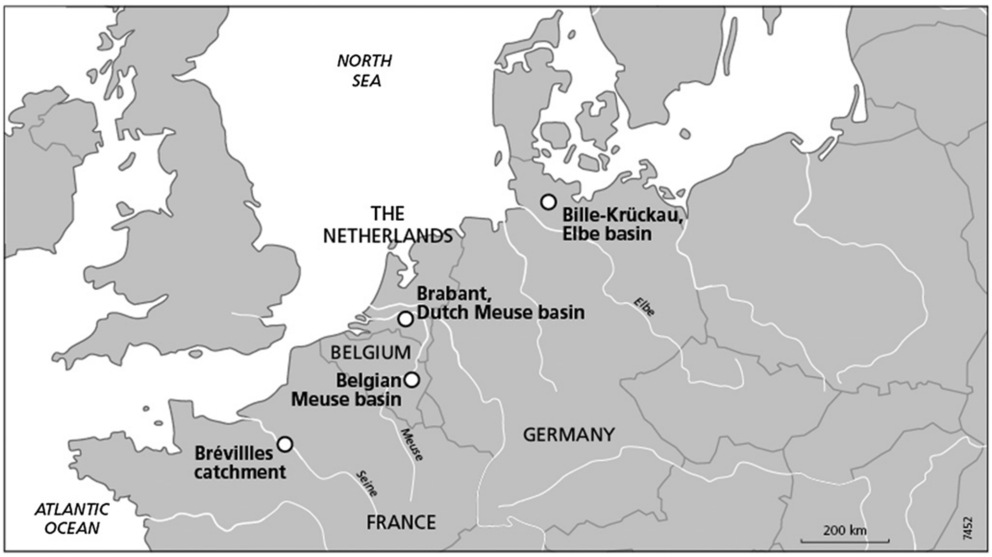


Fig. 4 Location of test sites within Europe.

Table 1 Selected characteristics of the four test sites

Sub-basin	Hydrogeological characteristics	Spatial scale	Contaminants	Methods used
Dutch part of Meuse basin (Brabant/Kempen)	Unconsolidated Pleistocene deposits; fine to medium coarse sands, loam	5000–500 km^2	Nitrate, potassium, oxidation capacity, sum of cations	Statistical, groundwater dating and deterministic modeling
Belgian part of Meuse basin				
Hesbaye	Cretaceous chalk, fissured, dual-porosity aquifer	440 km^2	Nitrate	Statistical, groundwater dating and deterministic modeling
Pays de Herve	Cretaceous chalk and sands, fissured	285 km^2	Nitrate	Statistical
Néblon catchment	Carboniferous limestone, folded karstified	65 km^2	Nitrate	Statistical
Meuse alluvial plain	Unconsolidated deposits; gravels, sands and clays	125 km^2	Nitrate	Statistical
Brévilles	Lutecian limestone over Cuise sands, limestone fissured	2.5 km^2	Pesticides (atrazine and de-ethylatrazine (DEA))	Groundwater dating, transfer functions and deterministic modeling
Elbe basin	Unconsolidated glacial deposits of sand and gravel	1300 km^2	Nitrate, potassium, aluminium, chloride, oxidation capacity, sum of cations	Statistical and groundwater dating

(NO_3), sulfate (SO_4), potassium (K), aluminium (Al), and chloride (Cl), as well as combined chemical indicators oxidation capacity (OXC) and charge-weighted molar sum of cations (SUMCAT). OXC is the weighted sum of molar concentrations of NO_3 and SO_4 which acts as a conservative solute if denitrification by pyrite oxidation takes place.[20] Furthermore, the concentrations of atrazine and its degradation product de-ethylatrazine (DEA) have been studied. To avoid the detection of artificial trends due to changes in the detection limit over the span of time series, all data were treated as follows before trend analysis. The largest detection limit of a dataset was accepted as the lowest detectable value, and all measurements below this value, including the measurements actually below the detection limit, were set to one half of the detection limit. For details on the sampling and measurement procedures we have referred to other papers if possible.

3.1 Dutch Meuse basin

The Dutch part of the Meuse basin almost entirely belongs to the groundwater body Sand Meuse, which covers most of the province of Noord-Brabant and part of Limburg (5000 km^2 in total). The groundwater body consists of fluvial unconsolidated Pleistocene sands, covered by 2–30 m of fluvio-periglacial and aeolian deposits of fine sands and loam. The history of intensive livestock farming on 62% of the area has produced a large surplus of manure contributing to widespread agricultural pollution (Appendix of ref. 32). The relatively flat area (0–30 m above mean sea level) is drained by a natural system of brooks, extended in the 20th century with drains and ditches to allow agricultural practices in the poorly drained areas. Groundwater tables are 1–5 m below surface as a result.[62] Net groundwater recharge is around 300 mm per year resulting in a downward groundwater flow velocity of about 1 m per year in recharge areas.[63]

Time series of major cations, anions and trace metals are available since 1992 from the dedicated national and provincial monitoring network sampled annually from 2 m long screens in multilevel wells at depths of 8 m ("shallow") and 25 m ("deep") below the surface.[20] Thanks to the dedicated monitoring wells with short screens and the character of the aquifer, little mixing occurs between recharge and sampling and a groundwater sample is estimated to contain a mixture of water recharged within a period of less than 5 years.

3.2 Belgian Meuse basin

Four groundwater bodies were selected as test cases in the Walloon part of the Meuse basin in Belgium, which represent various hydrogeological settings: the cretaceous chalk of Hesbaye, the cretaceous chalk of Pays de Herve, the Néblon basin in the carboniferous limestone of the Dinant synclinorium, and the alluvial plain of the Meuse River.

The cretaceous chalk groundwater body of Hesbaye also referred as the Geer basin covers an area of 440 km^2 located north-west from Liège.[64] The groundwater body is drained by the Geer River, a tributary of the Meuse River. Twenty-five million m^3 of groundwater are pumped annually from the fissured dual-porosity chalk aquifer to supply the city of Liège and surrounds. 85% of the area of the Hesbaye groundwater body is covered intensively by agriculture, mostly crops leading to an important and increasing nitrate contamination of the groundwater resource. Time series of nitrate are available from 32 monitoring points in the groundwater body, varying from dedicated monitoring wells to pumping wells, traditional wells, springs and galleries.[22]

The chalk groundwater body of Pays de Herve covers an area of 285 km^2, about 80% of which is covered by grassland. Groundwater is pumped at a rate of 12 million m^3 per year from the chalk aquifer. Nitrate concentrations of over 100 mg l^{-1} are observed in some of the 59 monitoring points which are distributed throughout the groundwater body.

The Néblon basin covers an area of 65 km^2 in the "Entre Sambre et Meuse" groundwater body, built of 500 m thick folded and karstified carboniferous limestone and sandstone. Land use is mainly divided between pastures (42%), crops (31%), and forests (24%). Around 10 million m^3 of groundwater are pumped annually from the limestone aquifers. Nitrate concentrations have been monitored since 1979 at two of the six monitoring locations in the basin.

The alluvial plain of the Meuse groundwater body (125 km^2 along 80 km of the Meuse) consists of gravel bodies embedded in old meandering channels filled with clay, silt and sandy sediments. Land use is 40% residential or industrial, and 60% natural land. Time series of groundwater quality data are available from 47 monitoring points.

3.3 Brévilles catchment

The Brévilles catchment (2.8 km^2), 75 km to the north-west of Paris, France, is built up out of a thick unsaturated zone (0–35 m) of dual-porosity Lutecian limestone, overlying the Cuise sands, 8–20 m thick and outcropping in the west of the catchment.[65] There is no superficial drainage and the catchment is drained by the Brévilles spring, an outcrop of the Cuise sands. Land use is largely agricultural, with predominantly peas, wheat and maize.[66] Monthly time series of concentrations of atrazine and its degradation product de-ethylatrazine (DEA) are available from seven piezometers and the Brévilles spring since 2001.[67] The application of atrazine on the catchment was halted in 2000.

3.4 Elbe basin

The groundwater bodies in the Bille-Krückau watershed (1300 km^2), located in Schleswig-Holstein, northern Germany, consist of unconsolidated glacial deposits of sand and gravel.[68] The sediments were deposited during the last and previous glaciations and subsequently denudated to a plateau-like landscape approximately 40 m above mean sea level. The area is drained by a dense network of natural streams, of which the Bille River is the largest draining 335 km^2. Groundwater is abstracted for drinking water purposes from the sandy and gravely deposits.

Two groundwater quality monitoring networks are in place, aimed at describing the natural conditions (baseline) and detecting trends in groundwater quality (trend). From these networks composed of 27 observation screens in total we selected 19 time series, sampled bi-annually from 8 shallow (<12.5 m below surface) and 11 deep (>12.5 m below surface) monitoring wells. The time series contain the concentrations of major cations and anions, from which we selected K, NO_3, Al and Cl, and constructed OXC and SUMCAT, for trend analysis.

4. Results

4.1 Application of statistical trend detection and estimation

Statistical trend analysis was applied to the dataset of 34 time series of concentrations from the Dutch part of the Meuse basin. Investigated solutes were NO_3 and K, OXC and SUMCAT. The time series from shallow screens (~8 m below surface) and deep screens (~25 m below surface) were analyzed separately. Non-parametric statistical trend analysis demonstrated significant trends for OXC and SUMCAT concentrations: increasing in deep screens and decreasing in shallow screens. No significant trends for NO_3 were detected.

Statistical trend analysis was applied to 97 nitrate time series from the Belgian part of the Meuse basin (Table 2). Significant trends were detected in 60% of the time series. Most of the detected trends were increasing, except for the Meuse alluvial plain, where both increasing and downward trends were detected. For 36 time series in the Geer basin, the estimated slope was used to predict the year in which the concentration of nitrate would exceed the drinking water limit (50 mg l^{-1}). For most of the points, the drinking water limit is estimated to be exceeded within 10–70 years.[22] This estimate represents a worst-case scenario, as it does not assume changes in land use and agricultural practices to protect groundwater quality.

Finally, statistical trend analysis was applied to the time series of NO_3, K, Al, OXC, Cl and SUMCAT concentrations from the Bille-Krückau watershed in the Elbe basin (Table 3). Time series from shallow and deep screens were analyzed separately. For the conservative solutes Cl, OXC and SUMCAT, significant upward trends were detected in time series from deep monitoring screens (>12.5 m below surface), whereas significant decreasing concentrations were detected in time series from the shallow screens (>12.5 m below surface). A further analysis of spatially weighted means indicated significant downward trend of potassium in shallow screens and significant upward trends of chloride and sum of negative ions. Significant trends were not detected in deep screens.

Table 2 Summary of results for trend tests for each groundwater body in the Belgian part of the Meuse basin

Groundwater body	Number of nitrate sampling sites	Number of downward trends	Number of upward trends	Percentage of significant trends
Geer basin	26	0	15	57.7
Pays of Herve	12	2	6	66.6
Néblon basin	6	1	4	83.3
Alluvial plain	38	15	11	68.4

Table 3 Percentage of the individual time series of the Bille-Krückau dataset showing a significant trend (⇑: significant upward trend, ⇓: significant downward trend)

	NO_3	K	Al	OXC	Cl	SUMCAT
Shallow	—	40% ⇓	0%	20% ⇓	20% ⇓	20% ⇓
Deep	—	11% ⇓	—	33% ⇑, 11% ⇓	44% ⇑	11% ⇑

4.2 Application of groundwater dating

$^3H/^3He$, CFC and SF_6 groundwater ages were available from 34 screens of 14 wells in agricultural recharge areas in the Dutch part of the Meuse basin.[69,70] CFC samples showed irregularities attributed to degassing caused by denitrification and contamination.[70] $^3H/^3He$ ages were considered more reliable following internal checks on degassing or contamination. The $^3H/^3He$ ages were used to interpret the time series of concentrations, by relating concentrations to the estimated time of recharge and aggregating all data available for the entire groundwater body.[32] The aggregated data were analyzed using a LOWESS smooth[55] with a smoother span of 0.5 to indicate the general pattern of change and compare that to contamination history and trends in the aggregated data were detected using simple linear regression to detect trends in concentrations in groundwater recharged between 1960 and 1980, or between 1990 and 2000 (Fig. 5). Significant upward trends were found in the concentrations of NO_3, K, OXC and SUMCAT in "old" groundwater (recharged between 1960 and 1980), but also significant downward trends were found in the concentrations of NO_3, OXC and SUMCAT in young groundwater (recharged between 1990 and 2000). Trends detected in this way could directly be related to changes in land use or contamination history. These results demonstrated trend reversal in groundwater quality[32] on the relevant scale of a groundwater body, as required by the EU Groundwater Directive.[6]

Tritium samples were taken from 33 monitoring points in the Geer basin and analyzed by the Lab of radioactive isotopes of the UFZ, Leipzig, Germany. The distribution of tritium concentrations only shows a distinction between "old" (>50 years) and "young" groundwater (Fig. 6), because travel times cannot be estimated accurately and unequivocally based on the tritium concentration only. High concentrations of tritium were observed in a large southwestern portion of the basin, where recharge is assumed to take place. Toward the downstream end of the basin, tritium concentrations decrease, indicating mixing of younger and older groundwater. No tritium is found in the northern confined part of the basin, indicating old (<1950) groundwater. The presence of old groundwater explains the absence of nitrate in this part of the aquifer.

The interpretation of groundwater age tracers (^3H and CFCs) is not straightforward in hydrogeological complex systems like the Brévilles catchment. An experimental sampling campaign was performed to assess whether an extensive dataset of groundwater age tracers would provide additional knowledge on the functioning of the system. Tritium and CFCs were analyzed in samples taken from 8 piezometers and the Brévilles spring.[71,72] The estimated ages showed a high variability within the small catchment with both old (<1960) and young (>1980) water in close proximity. The individual CFC ages (CFC-11, CFC-12, and CFC-113) were generally in good agreement, but some samples showed signs of degradation or contamination. Qualitative tritium groundwater age estimates were generally younger than the CFC age due to the dual-porosity nature of the system.

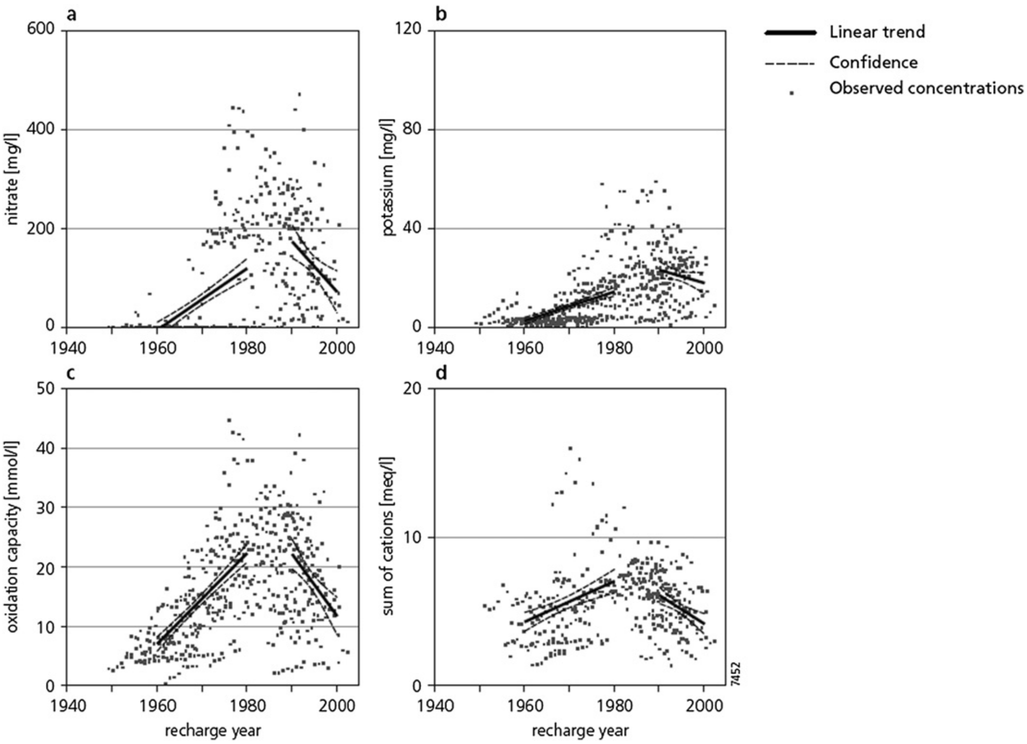


Fig. 5 Linear trends through concentration–recharge year data from the Dutch part of the Meuse basin show significant trend reversal between 1980 and 1990 for nitrate (a), oxidation capacity (c) and sum of cations (d) (after ref. 32). Potassium (b) showed no significant trend reversal.

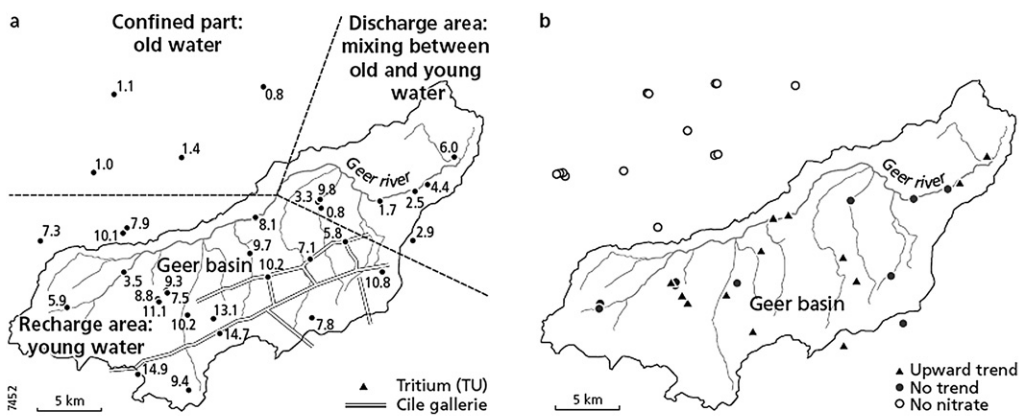


Fig. 6 Spatial distribution of tritium (a) and trends in nitrate concentrations (b) in the Geer basin (after ref. 22).

The tracers confirmed the complex hydrogeology of the system, but could not be used for reliable age dating and trend interpretation because of the likely mixing co-occurring in the thick unsaturated zone.

For the Elbe dataset, an empirical exponential relationship between depth and groundwater age was assumed, instead of dating groundwater using tracers. Such an exponential increase in groundwater age with depth may be expected in unconsolidated unconfined aquifers like the Bille-Krückau catchment.[73] Using the empirical relationship, the time series of Cl, OXC and SUMCAT were related to the approximate time of recharge and analyzed again for trends using a LOWESS smooth with a smoother span of 0.5 and a linear trend detection with a 95% confidence interval (Fig. 7). The LOWESS smooth approach showed that the overall pattern in the measured concentration–recharge time relationship is similar to the historical surplus of N applied at the surface. Similar results were found in the Dutch part of the Meuse basin, probably due to the similarities in land use history and hydrogeology.

4.3 Application of transfer functions

The transfer function approach was applied to time series of head, flux, and nitrate, atrazine and DEA concentrations from the piezometers and spring in the Brévilles catchment. The following section describes the followed approach, which has been described in detail by Pinault and Dubus.[66] Hydraulic heads were modeled as a function of effective rainfall using combined

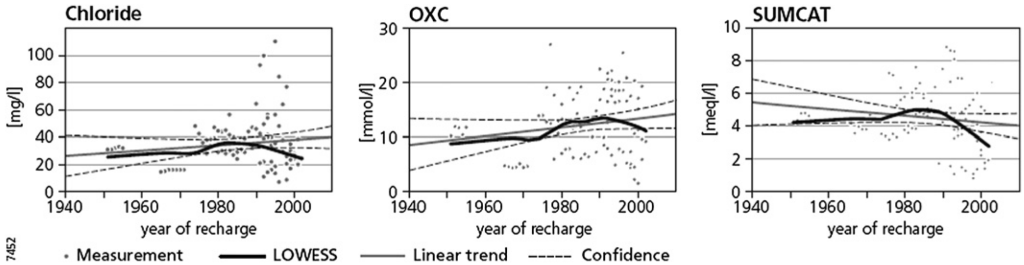


Fig. 7 Linear trends (with 95% confidence interval) and LOWESS smooth lines through concentrations of Cl, OXC, and SUMCAT in relation to time of recharge from the Elbe basin.

convolution functions for transport and dispersion, while effective rainfall was modeled as a function of the actual rainfall and of a threshold value representing the water storage in the soil. The threshold value for soil water storage was related to the rainfall and potential evapotranspiration with trapezoid impulse response functions with four degrees of freedom. Concentrations of contaminants were modeled in a similar fashion, using the effective flux of the contaminant from the unsaturated soil instead of the effective rainfall, to predict the flux or concentrations in the Brévilles spring. To predict spring fluxes and concentrations, the impulse response functions were extended to include the contribution of various pathways of contaminants to the spring. Future concentrations were calculated based on 5 year long generated meteorological time series based on the median annual precipitation and the 5, 10 and 20 year extreme wet and dry years.

The transfer function model was capable of reproducing the general trends in the time series, both in the monitoring wells and in the spring. We think the good fit is remarkable given the short monitoring period and the long travel times in the groundwater system, as indicated by impulse response functions of over 10 years long. Because of these long transfer times, it was possible to reconstruct the concentrations of the contaminants in the vadose zone. Interestingly, the reconstructed inputs were in agreement with the historical application of atrazine in the catchment.

Future concentrations of atrazine and DEA at the Brévilles spring were predicted using the calibrated transfer function model and rainfall data generated by the TEMPO tool (Fig. 8). The generated rainfall series contained either only wet or dry years, with historical recurrence intervals of 5, 10 or 20 years, to illustrate the response of atrazine concentrations to different future climates. Atrazine release was predicted to occur more during wet years than in dry years. While atrazine concentrations in the spring are predicted to decrease dramatically over the next 5 years, concentrations of its metabolite DEA are expected to remain constant over the next decade.[66]

4.4 Application of deterministic modeling

For the Kempen region in the Dutch part of the Meuse basin, the physical-deterministic groundwater flow and transport models were a steady-state MODFLOW[74] model for groundwater flow and MT3DMS[75] for solute transport. Historical concentrations of contaminants at the land surface were reconstructed based on statistical records of atmospheric depositions and manure applications.[76] Leaching of heavy metals from the unsaturated zone, sensitive to sorption and fluctuating water tables, was modeled with Hydrus-1D.[77] The coupled transport model, described in detail by Van der Grift and Griffioen,[41] was used to predict concentrations of nitrate, potassium and heavy metals in groundwater at the monitoring locations within the model area.

The model predicted significant trends in the concentrations of nitrate and OXC for the period 1995–2005: upward in deep groundwater and downward in shallow groundwater. Due to variations in groundwater travel times and the constant recharge concentrations from 2005 onward, few significant trends are predicted for the future, except a decrease in OXC between 2010 and 2020. Between 2010 and 2020, the model also predicted a significant upward trend in the concentration of zinc in shallow groundwater. This trend is caused by a slow release of zinc accumulated in the unsaturated zone and the retarded transport of zinc through the groundwater system due to cation exchange.[20,41]

For the Geer basin in the Walloon part of the Meuse basin, a physically based, spatially distributed, deterministic model was constructed using the control volume finite element SUFT3D code.[78] This model[79] uses a new approach, the hybrid finite element mixing cell,[80] combining mixing cells to model the solute transport with a conventional finite element model for groundwater flow based on Darcy's law both in the saturated and in the partially saturated zones. Transport processes considered with the mixing cells are advection, degradation and dual-porosity related to the presence of immobile water. The model was calibrated on groundwater levels, as well as measured tritium concentrations. The model was used to reproduce and to extrapolate observed nitrate concentrations in the Geer basin at the monitoring points.

The model was capable of reproducing both the groundwater levels and the distribution of tritium in the aquifer. The model also accurately reproduced the upward trends in nitrate concentrations in the Geer basin. The future evolution of nitrate trends in groundwater was computed for different scenarios of nitrate concentrations in the leaching water. The time before trend reversal is a function of the location of the monitoring points in the basin. In the Southern part of the basin, time before reversal is a function of the thickness of the unsaturated zone where the nitrates move slowly. In the Eastern part of the basin, due to the mixing between old and young water, trend reversal would not occur in the next 50 years and nitrate concentrations would still increase.

The physical-deterministic model developed for the Brévilles catchment, in France, consisted of the combination of a series of

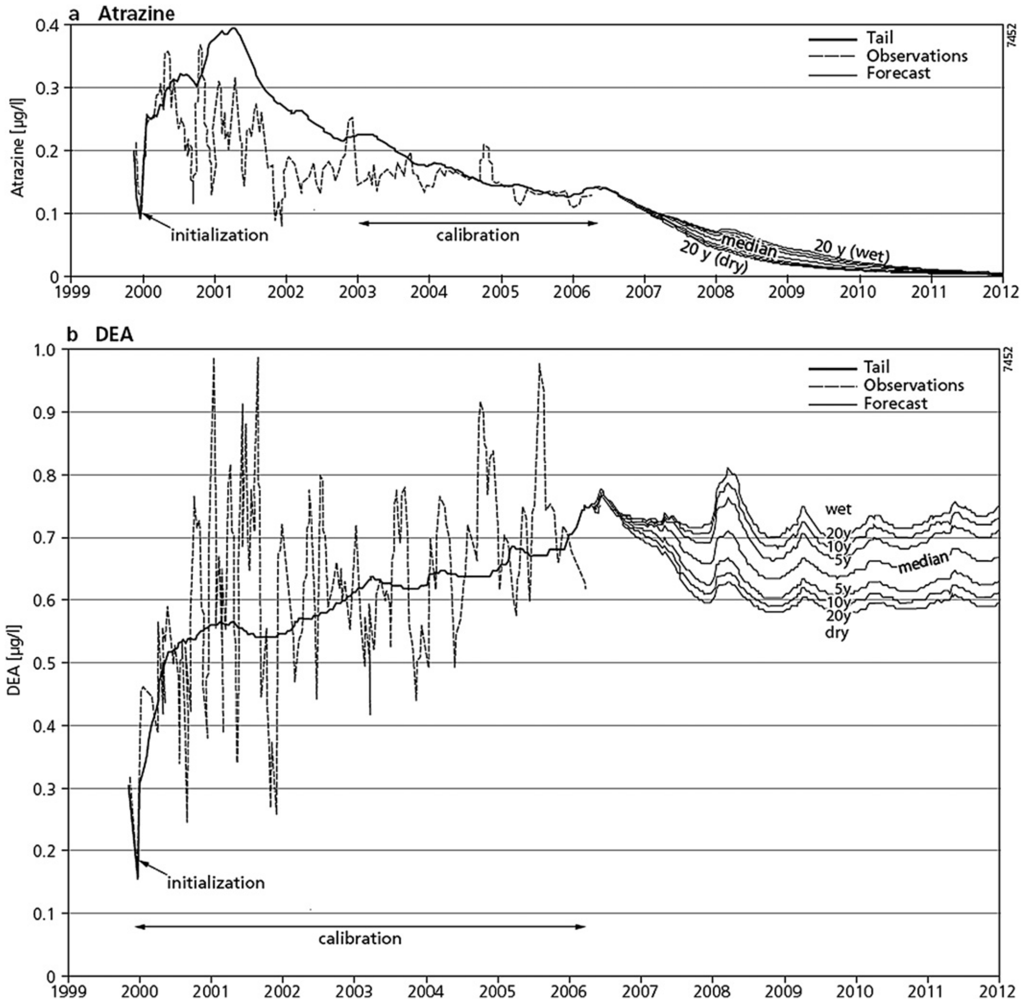


Fig. 8 Predictions of the TEMPO software for concentrations of atrazine and the atrazine metabolite DEA (de-ethylatrazine) at the Brévilles spring (after ref. 66).

a 1D unsaturated zone models to simulate water flow and contaminant transport through the fissured dual-porosity chalk, and a 2D groundwater flow and transport model for the Cuise sands. The 1D pesticide fate model MACRO[81] was used to simulate transport through the root zone, taking into account preferential flow phenomena, while transport through the unsaturated zone was modeled using MARTHE.[82] The combined model was used to reproduce observed groundwater levels, as well as nitrate, atrazine and DEA concentrations in piezometers and in the Brévilles spring. Thirteen regional climate model scenarios were used for predicting future trends in concentrations.

This combined model accurately reproduced the observed groundwater levels at the piezometers and also the discharge from the Brévilles spring. Predicted atrazine concentrations at the piezometers were in the same order of magnitude as the measurements, but predicted concentrations underestimated observations in the spring, probably due to the lack of accurate data on the application of atrazine on individual fields in the catchment. Similar to the predictions made using the transfer function approach, concentrations of atrazine in piezometers were predicted to decrease exponentially over the next 14 years. A slower decline in the concentrations was predicted for the spring.

5. Discussion of trend detection approaches

In this section, we discuss each of the methods applied in this study in terms of data requirement, additional monitoring costs, applicability in different geohydrological systems, and their power to extrapolate. Prerequisites, costs, and overall usefulness of all methods are summarized in Table 4.

The statistical three-step approach to detect trends in groundwater quality was applied at three test sites and proved to be a simple and easily applicable technique to existing time series of contaminant concentrations, having a normal distribution or not, making it universally applicable (Table 4). Statistical trend detection requires time series that span several years to decades to detect a significant trend, depending on the hydrogeological system and monitoring network. If the quality of the available datasets is sufficient, it requires no additional costs for sampling. The approach provides an objective detection of trends.

Statistical trend analysis may be of limited operational use because no link to the driving forces (*e.g.* meteorological data,

Table 4 Summary table comparing the strengths and weaknesses of each of the trend analysis methodologies based on the experiences of their application to the four test sites and datasets

	Purely statistical approaches	Transfer function approaches	Age dating	Deterministic modeling with poor fit to the data	Deterministic modeling with good fit to the data
Data requirements	Existing monitoring data or collection in the field	Existing monitoring data or collection in the field + collection of information on the input flux (rainfall, and either inputs or land use)	Existing monitoring data or collection in the field + information on the evolution of the input function + analysis of tracers in samples	Existing monitoring data or collection in the field + large effort in collection of additional information (other piezometers, pumping and tracer tests, geophysics, soil mapping)	Existing monitoring data or collection in the field + large effort in collection of additional information (other piezometers, pumping and tracer tests, geophysics, soil mapping)
Associated cost magnitude (on top of the data collection effort)	0	1 (surveys if not already available—purchasing of meteo data)	10	100 (geophysics, additional piezometers, soil mapping)	100 (geophysics, additional piezometers, soil mapping)
Requirements on the understanding of the groundwater system and contaminant properties	No understanding of the system	Functional understanding of the system (identification of the key factors and understanding of their influence)	Functional understanding of the system	Detailed data on the system, but lack of overall understanding of the functioning of the system (exemplified by the lack of fit of the deterministic model)	Detailed understanding of the system under study
Extrapolation potential	Poor	Good	Good	Poor	Very good
Potential universality to all systems	Potentially	Potentially	Limited, only applies to homogeneous systems	Potentially	Potentially
Potential for operational use	–	+	+	–	+
Source of new knowledge about the functioning of the system	–	+	+	+	++

groundwater flow and travel times or historical agricultural practices) is incorporated in the analysis, and no knowledge about the functioning of the system will be gained by its application. Therefore the trends that are found in individual time series may be extrapolated over short periods of time only. Statistical trends cannot sensibly be extrapolated over longer periods of time, because they are incapable of dealing with changes in agricultural practices, meteorological conditions or the groundwater flow system. Statistical trend analysis is not capable of predicting trend reversal, which is a major disadvantage.

Trend detection as required by the GWD is dedicated to detect trends in the concentrations of pollutants resulting from anthropogenic sources and distinguish these from natural variation with an adequate level of confidence and precision (GWD, Annex V, art 2(a)(i)). While the WFD Guidance[42] left little room to include conceptual understanding of the factors determining groundwater quality, in recent years, it was generally realized that a conceptual understanding of the groundwater systems is essential for the characterization of chemical status and the detection of trends.[45,83] It is our conclusion that only for an initial survey to detect changes in groundwater quality, a classical statistical approach is most suitable, but more elaborate approaches to trend detection including more information about the groundwater body and contaminant transport may have a higher chance of determining and understanding significant and sustained upward trends or trend reversal.

For the detection of trends in groundwater quality it is crucial to know whether the character of the subsurface or the monitoring system causes mixing of groundwater with different travel times. In single porous systems, groundwater at a specific location typically has a distinct groundwater age. In practice, the possibility of sampling groundwater with a distinct age also requires a monitoring network with short (<5 m) monitoring screens or the use of packers in long screened wells to prevent mixing during sampling. In simple single-porosity groundwater bodies with access to monitoring wells with short screens, groundwater dating has been applied by various studies as a tool for aggregating groundwater quality data and analyzing trends therein by relating the measured concentrations of pollutants to the time of recharge of the sampled groundwater.[23,26,27,29,32,84] The resulting concentration–travel time relationship can be linked directly to historical records of land use or agricultural practices. Recent publications presenting studies from the U.S. Geological Survey National Water-Quality Assessment (NAWQA) program in which concentrations are directly related to recharge time of groundwater conclude that groundwater travel times are invaluable to the detection of trends in existing groundwater quality data.[14,85–88]

Groundwater dating can thus be used to reinterpret groundwater quality time series and demonstrate trend reversal in groundwater quality, which represents a step further in comparison with statistical analysis. Knowledge about the travel times in the groundwater system enhances the understanding of the flow system and may also explain the slow improvement of groundwater quality (Table 4). It must be considered that groundwater dating requires a substantial financial investment for sampling and sample analysis, even if a proper monitoring network is in place. The benefit is that the existing groundwater quality data become more valuable as the re-analysis of this data may reveal trends that could not be demonstrated without knowledge of the recharge times of the groundwater samples. This makes groundwater dating suitable for operational use, where applicable.

In hydrogeologically complex systems such dual-porosity aquifers, or under a variably or thick unsaturated zone, groundwater age tracers are difficult to interpret and may only confirm the complexity of the system and proper application is limited to more simple groundwater systems. Qualitative groundwater dating using tritium can be applied to detect the presence of "old" groundwater, which may for example explain the presence of nitrate at low concentrations due to old age rather than denitrification. In such complex systems, possibly with seasonal influences, a transfer function approach is likely to be better suited for trend detection.

The transfer function approach is an intermediate approach between statistical and deterministic models. If the available data are sufficient to calibrate a transfer function that expresses the delay in transfers of water and pollutants in the systems considered, transfer functions require no additional financial investment (Table 4). The main advantages of transfer functions are that they require little information about the physical functioning of the system, but rather rely on the available data, which make them suitable for application in a wide variety of systems.

Transfer functions provide a good agreement with measured time series in the complex aquifer of the Brévilles catchment showing that they are capable of reproducing the non-linear behavior of dual-porosity systems,[66] where other approaches failed. This makes them suitable for operational use in these systems, if the transfer functions are capable of providing a representative description of the system and its variability. Transfer functions may be used for trend extrapolation, but only with great care to ensure that the predicted trends are within the range of the observations. In these systems, groundwater dating may serve to confirm the hydrological functioning and transfer times of the system.

Because of the geohydrological diversity among test sites, site-specific physical-deterministic models need to be built. One of the main issues associated with deterministic modeling is the need to have a detailed characterization of the system under study available in terms of meteorology, soils, subsoils, hydro(geo)logy and use of pollutants on the catchment. Furthermore, the relevant characteristics of water flow, transport and interactions between the contaminants and the system itself need to be known at the effective scale of the model, while many of these parameters are often derived from laboratory studies. This typically requires very significant financial and time investments in the field, which usually spans over several years. On the other hand, one of the great advantages of undertaking deterministic modeling activities is that it brings together various sources of information collected in the field that allow the expansion of the understanding of the catchment functioning using both measured and predicted information. Although there is no dispute as to the usefulness of intensive catchment modeling activities from a research point of view, the 'return on investment' from an operational perspective is mainly dependent on whether a model can be successfully fitted to the data and whether the model has shown potential for supporting extrapolation and management activities. A distinction was therefore made in Table 4 between modeling efforts which provide a successful fit to measured data and trends, and those where the fit to the data is considered to be below the standard for operational activities.

The very large financial, human resources and time investments associated with the collection of data and their integration into an overarching modeling exercise means that the deployment of deterministic models for operational analysis of trends across the EU is beyond reach, even if a good fit to the data would be obtained. Such modeling activities should concentrate on areas of high ecological, sustainability or economical importance within the context of the Water Framework Directive.

The main advantage of physically deterministic models is their capability to predict trends in the future that are not yet observed in the monitoring data, for example due to the slow release of zinc from the unsaturated zone. They can provide estimates of the time scales at which trend reversal should be expected as a result of protective legislation, which may be several decades because of the long travel times of groundwater.[41] While large-scale 3D models are suitable to extrapolate long-term trends, they are less capable of predicting short-term variations due in part to their coarse resolution and simplified aquifer characteristics. The quality of predicted future trends relies on the appropriate incorporation of important processes into the model, the fit of the model to the existing data, and accuracy of future land use, agricultural practices, and contamination scenarios. Such scenario analyses are useful to aid policy makers to decide on the effectiveness of proposed regulations.

6. Conclusions

There is no unique solution to detect trends in groundwater quality across widely differing catchments and monitoring systems. The choice of the method for trend detection and extrapolation should firstly be made on the basis of the specific goals of the study (only trend detection or also extrapolation), the available resources, and the system under study (Table 5). Among the aspects of trend detection investigated in this study, the most important difference between groundwater bodies is whether the character of the subsurface or the monitoring system causes mixing of groundwater with different travel times. While statistical trend detection may be suitable for preliminary surveys of trends in groundwater quality in any type of groundwater

Table 5 Recommended preliminary and elaborate methods for trend detection and extrapolation in simple and complex groundwater systems

		Groundwater system	
		Simple	Complex
Trend detection	Preliminary	Statistics	Statistics
	Elaborate	Groundwater dating	Transfer functions
Trend extrapolation	Preliminary (short-term)	Statistical methods	Transfer functions
	Elaborate (long-term)	Deterministic model	Deterministic model

body, more elaborate studies aimed at detecting trends should apply groundwater dating in unconsolidated aquifers or transfer functions in complex aquifers. Statistical approaches or transfer functions are suitable only for short-term extrapolation of trends, because no direct link with the driving forces of the trends is included in these methods. Long-term extrapolation requires the use of deterministic models with a good fit to the available data, which are capable of predicting the effects of land use changes and various management scenarios.

Regardless of the complexity of the model used, being transfer functions or deterministic models, trend detection and extrapolation are always associated with uncertainty. This means that groundwater quality monitoring should remain a priority. Additional data will improve the detection of trends and increase the knowledge of the functioning of the groundwater system. Better understanding of the system, possibly derived from deterministic modeling, can in turn provide feedback for the optimization of the groundwater quality monitoring networks.

Acknowledgements

This work was supported by the European Union FP6 Integrated Project AquaTerra (Project no. GOCE 505428) under the thematic priority "Sustainable development, global change and ecosystems". European Union FP5 Project PEGASE (FP5-EVK1-CT1999-00028), Seine River Basin Water Authority (AESN) and the BRGM research project TRANSPHYTO also supported the collection of data in the Brévilles catchment. The Belgian data were acquired thanks to the help of the administration of the Walloon Region (F. Delloye and his team) and the team of the Hydrology & Agricultural Hydraulics Group of the University of Gembloux, Belgium. Conceptual and numerical developments of the HFEMC method have also been performed in the framework of The Interuniversity Attraction Pole TIMOTHY (IAP Research Project P6/13) funded by the Belgian Federal Science Policy Office (BELSPO). The monitoring data used for the Bille-Krückau catchment were provided by Environmental Office of Schleswig-Holstein State, thanks to the assistance of Dr Henning Holthusen and his colleagues. We would like to thank Sylvia Dautrebande (Gembloux Agricultural University, Belgium) and Tom Nolan and Larry Puckett (USGS) for providing data for Fig. 1. We would like to thank two anonymous reviewers for their comments which helped us improve the manuscript.

References

1 R. F. Spalding and M. E. Exner, Occurrence of nitrate in groundwater—a review, *J. Environ. Qual.*, 1993, **22**(3), 392–402.
2 S. S. D. Foster, A. C. Cripps and A. Smith-Carington, Nitrate leaching to groundwater, *Philos. Trans. R. Soc. London, Ser. B*, 1982, **296**, 477–489.
3 O. Strebel, W. H. M. Duynisveld and J. Böttcher, Nitrate pollution of groundwater in western Europe, *Agric., Ecosyst. Environ.*, 1989, **26**(3–4), 189–214.
4 B. T. Nolan and et al., Risk of nitrate in groundwaters of the United States—a national perspective, *Environ. Sci. Technol.*, 1997, **31**(8), 2229.
5 EU, *Directive of the European Parliament and of the Council 2000/60/EC Establishing a Framework for Community Action in the Field of Water Policy*, 2000.
6 EU, *Directive 2006/118/EC on the Protection of Groundwater against Pollution and Deterioration*, 2006.
7 J. Y. Lee and et al., A review of the national groundwater monitoring network in Korea, *Hydrol. Process.*, 2007, **21**(7), 907–919.
8 N. J. Kim, M. J. Cho and N. C. Woo, Developing a national groundwater-monitoring network in Korea, *Hydrogeol. J.*, 1995, **3**(4), 89.
9 R. K. Juhler and G. Felding, Monitoring methyl tertiary butyl ether (MTBE) and other organic micropollutants in groundwater: results from the Danish national monitoring program, *Water, Air, Soil Pollut.*, 2003, **149**(1–4), 145.
10 W. Van Duijvenbooden, Groundwater Quality Monitoring in the Netherlands, in *Regional Groundwater Quality*, ed. W. M. Alley, Van Nostrand Reinhold, New York, 1993, pp. 515–535.
11 C. J. Daughney and R. R. Reeves, Definition of hydrochemical facies in the New Zealand national groundwater monitoring programme, *J. Hydrol. (New Zealand)*, 2005, **44**(2), 105–130.
12 M. N. Almasri and S. M. S. Ghabayen, Analysis of nitrate contamination of Gaza Coastal Aquifer, Palestine, *Journal of Hydrologic Engineering*, 2008, **13**(3), 132–140.
13 P. P. Leahy, B. Ryan and I. Johnson, An introduction to the U.S. geological surveys national water-quality assessment program, *Water Resources Bulletin*, 1993, **29**(4), 529.
14 M. R. Rosen and W. W. Lapham, Introduction to the U.S. Geological Survey National Water-Quality Assessment (NAWQA) of groundwater quality trends and comparison to other national programs, *J. Environ. Qual.*, 2008, **37**(S5), 190–198.
15 R. S. Ward and et al., A framework for monitoring regional groundwater quality, *Q. J. Eng. Geol. Hydrogeol.*, 2004, **37**(4), 271–281.
16 C. J. Daughney and R. R. Reeves, Analysis of temporal trends in New Zealand's groundwater quality based on data from the national groundwater monitoring programme, *J. Hydrol. (New Zealand)*, 2006, **45**(1), 41–62.
17 Y. Xu, L. A. Baker and P. C. Johnson, Trends in ground water nitrate contamination in the Phoenix, Arizona Region, *Ground Water Monit. Rem.*, 2007, **27**(2), 49–56.
18 M. E. Stuart and et al., Screening for long-term trends in groundwater nitrate monitoring data, *Q. J. Eng. Geol. Hydrogeol.*, 2007, **40**(4), 361–376.
19 H. P. Broers and and et al., Groundwater quality trend detection at the regional scale: effects of spatial and temporal variability, IAHS Publ., 2005, **vol. 297**, pp. 50–60.
20 H. P. Broers and B. Van der Grift, Regional monitoring of temporal changes in groundwater quality, *J. Hydrol. (Amsterdam, Neth.)*, 2004, **296**(1–4), 192–220.
21 J. Reynolds-Vargas, J. Fraile-Merino and R. Hirata, Trends in nitrate concentrations and determination of its origin using stable isotopes (^{18}O and ^{15}N) in groundwater of the western Central Valley, Costa Rica, *Ambio*, 2006, **35**(5), 229–236.
22 J. Batlle-Aguilar and et al., Identification of groundwater quality trends in a chalk aquifer threatened by intensive agriculture in Belgium, *Hydrogeol. J.*, 2007, **15**(8), 1615.
23 K. Burow, N. Dubrovsky and J. Shelton, Temporal trends in concentrations of DBCP and nitrate in groundwater in the eastern San Joaquin Valley, California, USA, *Hydrogeol. J.*, 2007, **15**(5), 991–1007.
24 J. C. Loftis, Trends in groundwater quality, *Hydrol. Process.*, 1996, **10**, 335–355.
25 *Regional Ground-Water Quality*, ed. W. M. Alley, Van Nostrand Reinhold, New York, 1993.
26 J. K. Böhlke and J. M. Denver, Combined use of groundwater dating, chemical, and isotopic analyses to resolve the history and fate of nitrate contamination in two agricultural watersheds, Atlantic coastal plain, Maryland, *Water Resour. Res.*, 1995, **31**(9), 2319–2339.
27 A. M. MacDonald and et al., Identifying trends in groundwater quality using residence time indicators: an example from the Permian aquifer of Dumfries, Scotland, *Hydrogeol. J.*, 2003, **11**(4), 504–517.
28 J. K. Böhlke and et al., Denitrification in the recharge area and discharge area of a transient agricultural nitrate plume in a glacial outwash sand aquifer, Minnesota, *Water Resour. Res.*, 2002, **38**(7), 1–26.
29 L. I. Wassenaar, M. J. Hendry and N. Harrington, Decadal geochemical and isotopic trends for nitrate in a transboundary aquifer and implications for agricultural beneficial management practices, *Environ. Sci. Technol.*, 2006, **40**(15), 4626–4632.

30 A. J. Tesoriero and et al., Nitrogen transport and transformations in a coastal plain watershed: influence of geomorphology on flow paths and residence times, *Water Resour. Res.*, 2005, **41**(2), W02008.
31 T. Laier, Nitrate Monitoring and CFC-Age Dating of Shallow Groundwaters—An Attempt to Check the Effect of Restricted Use of Fertilizers, in *Nitrate in Groundwaters, IAH Selected Papers on Hydrogeology*, ed. L. Razowska-Jaworek and A. Sadurski, A.A. Balkema, Leiden, The Netherlands, 2004, pp. 247–258.
32 A. Visser and et al., Demonstrating trend reversal of groundwater quality in relation to time of recharge determined by $^{3}H/^{3}He$, *Environ. Pollut.*, 2007, **148**(3), 797–807.
33 K. K. Gardner and R. M. Vogel, Predicting ground water nitrate concentration from land use, *Ground Water*, 2005, **43**(3), 343–352.
34 Y. Jiang and et al., Groundwater quality and land use change in a typical karst agricultural region: a case study of Xiaojiang watershed, Yunnan, *Journal of Geographical Sciences*, 2006, **16**(4), 405–414.
35 A. Ritter and et al., Agricultural land use and hydrology affect variability of shallow groundwater nitrate concentration in South Florida, *Hydrol. Process.*, 2007, **21**(18), 2464–2473.
36 D. J. Lapworth and et al., Pesticides in groundwater: some observations on temporal and spatial trends, *Water and Environment Journal*, 2006, **20**(2), 55–64.
37 H. J. Di and et al., A pilot regional scale model of land use impacts on groundwater quality, *Management of Environmental Quality*, 2005, **16**(3), 220–234.
38 M. N. Almasri and J. J. Kaluarachchi, Modular neural networks to predict the nitrate distribution in ground water using the on-ground nitrogen loading and recharge data, *Environmental Modelling and Software*, 2005, **20**(7), 851–871.
39 M. N. Almasri and J. J. Kaluarachchi, Modeling nitrate contamination of groundwater in agricultural watersheds, *J. Hydrol. (Amsterdam, Neth.)*, 2007, **343**(3–4), 211.
40 J. C. Refsgaard and et al., Large scale modelling of groundwater contamination from nitrate leaching, *J. Hydrol (Amsterdam, Neth.)*, 1999, **221**(3–4), 117–140.
41 B. Van der Grift and J. Griffioen, Modelling assessment of regional groundwater contamination due to historic smelter emissions of heavy metals, *J. Contam. Hydrol.*, 2008, **96**(1–4), 48–68.
42 J. Grath, et al., *The EU Water Framework Directive: Statistical aspects of the identification of groundwater pollution trends, and aggregation of monitoring results. Final Report, 41.046/01-IV1/00 and GZ 16 2500/2-I/6/00*, Austrian Federal Ministry of Agriculture and Forestry, Environment and Water Management and European Commission, Vienna, 2001.
43 P. Quevauviller, Groundwater monitoring in the context of EU legislation: reality and integration needs, *J. Environ. Monit.*, 2005, **7**(2), 89.
44 P. Quevauviller and et al., Science-policy integration needs in support of the implementation of the EU Water Framework Directive, *Environ. Sci. Policy*, 2005, **8**(3), 203.
45 J. Grath and et al., Report on EU guidance on groundwater monitoring developed under the common implementation strategy of the water framework directive, *J. Environ. Monit.*, 2007, **9**(11), 1162–1175.
46 J. Harris, J. C. Loftis and R. H. Montgomery, Statistical methods for characterizing ground-water quality, *Ground Water*, 1987, **25**(2), 185–193.
47 R. M. Hirsch, J. R. Slack and R. A. Smith, Techniques of trend analysis for monthly water quality data, *Water Resour. Res.*, 1982, **18**(1), 107–121.
48 R. M. Hirsch, R. B. Alexander and R. A. Smith, Selection of methods for the detection and estimation of trends in water quality, *Water Resour. Res.*, 1991, **27**(5), 803–813.
49 J. C. Loftis, C. H. Taylor and P. L. Chapman, Multivariate tests for trend in water quality, *Water Resour. Res.*, 1991, **27**(7), 1419–1429.
50 S. S. Shapiro and M. B. Wilk, An analysis of variance test for normality (complete samples), *Biometrika*, 1965, **52**, 591–611.
51 S. S. Shapiro and R. S. Francia, An approximate analysis of variance test for normality, *J. Am. Stat. Assoc.*, 1972, **67**(337), 215–216.
52 J. R. Carr, *Numerical Analysis for the Geological Sciences*, Prentice Hall, Englewood Cliffs, NJ, 1995, p. 592.
53 M. G. Kendall, *Rank Correlation Methods*, Charles Griffin & Company, London, 1948.
54 H. B. Mann, Nonparametric tests against trend, *Econimetrica*, 1945, **13**, 245–259.
55 D. R. Helsel and R. M. Hirsch, Statistical methods in water resources, *Studies in Environmental Science*, Elsevier, Amsterdam, The Netherlands, 3rd edn, 1995, vol. 49, p. 529.
56 S. Yue, P. Pilon and G. Cavadias, Power of the Mann–Kendall and Spearman's rho tests for detecting monotonic trends in hydrological series, *J. Hydrol. (Amsterdam, Neth.)*, 2002, **259**(1–4), 254–271.
57 P. Schlosser and et al., Tritium/^{3}He Dating of Shallow Groundwater, *Earth Planet. Sci. Lett.*, 1988, **89**(3–4), 353–362.
58 E. Busenberg and L. N. Plummer, Use of chlorofluorocarbons (CCl_3F and CCl_2F_2) as hydrologic tracers and age-dating tools: the alluvium and terrace system of Central Oklahoma, *Water Resour. Res.*, 1992, **28**(9), 2257–2283.
59 E. Busenberg and L. N. Plummer, Dating young groundwater with sulfur hexafluoride: natural and anthropogenic sources of sulfur hexafluoride, *Water Resour. Res.*, 2000, **36**(10), 3011–3030.
60 J. L. Pinault, H. Pauwels and C. Cann, Inverse modeling of the hydrological and the hydrochemical behavior of hydrosystems: application to nitrate transport and denitrification, *Water Resour. Res.*, 2001, **37**(8), 2179.
61 J. L. Pinault, *Manuel utilisateur de TEMPO: logiciel de traitement et de modélisation des séries temporelles en hydrogéologie et en hydrogéochemie, Projet Modhydro. Rap. BRGM/RP-51459*, BRGM, Orleans, 2001, p. 221.
62 H. P. Broers, *Strategies for regional groundwater quality monitoring*, in PhD thesis. Fysische geografie, Utrecht University, Utrecht, 2002, p. 231.
63 H. P. Broers, The spatial distribution of groundwater age for different geohydrological situations in the Netherlands: implications for groundwater quality monitoring at the regional scale, *J. Hydrol (Amsterdam, Neth.)*, 2004, **299**(1–2), 84–106.
64 A. Dassargues and A. Monjoie, The Chalk in Belgium, in *The Hydrogeology of the Chalk of North-West Europe*, ed. R. A. Downing, M. Price and G. P. Jones, Oxford Science Publ., Oxford, 1993, pp. 153–269.
65 C. Mouvet, et al., PEGASE. Pesticides in European Groundwaters: detailed study of representative aquifers and simulation of possible evolution scenarios, in *Final Report of the European Project #EVK1-CT1990-00028. BRGM/RP-52897-FR*, ed. I. G. Dubus and C. Mouvet, 2004.
66 J. L. Pinault and I. G. Dubus, Stationary and non-stationary autoregressive processes with external inputs for predicting trends in water quality, *J. Contam. Hydrol.*, 2008, **100**(1–2), 22–29.
67 N. Baran, C. Mouvet and P. Négrel, Hydrodynamic and geochemical constraints on pesticide concentrations in the groundwater of an agricultural catchment (Brévilles, France), *Environ. Pollut.*, 2007, **148**(3), 729–738.
68 F. Steinman, *Beobachtung der Grundwasserbeschaffenheit in Schleswig-Holstein: Trendmessnetz 1995-2000*, Landesamt für Natur und Umwelt des Landes Schleswig-Holstein, Flintbek, Germany, 2002, p. 54.
69 A. Visser, H. P. Broers and M. F. P. Bierkens, Dating degassed groundwater with $^{3}H/^{3}He$, *Water Resour. Res.*, 2007, **43**(10), WR10434.
70 A. Visser and et al., Degassing of $^{3}H/^{3}He$, CFCs and SF_6 by denitrification: measurements and two-phase transport simulations, *J. Contam. Hydrol.*, 2009, **103**(3–4), 206–218.
71 S. Roulier and et al., Controls on atrazine leaching through a soil-unsaturated fractured limestone sequence at Brevilles, France, *J. Contam. Hydrol.*, 2006, **84**(1–2), 81–105.
72 A. Gutierrez and N. Baran, Long-term transfer of diffuse pollution at catchment scale: respective roles of soil, and the unsaturated and saturated zones (Brévilles, France), *J. Hydrol. (Amsterdam, Neth.)*, 2009, **369**(3–4), 381–391.
73 J. C. Vogel, Investigation of Groundwater Flow with Radiocarbon, in *IAEA Symposium on Isotopes in Hydrology, 14–18 November 1966*, IAEA, Vienna, Austria, 1967.
74 A. W. Harbaugh, et al., *MODFLOW-2000, the U.S. Geological Survey Modular Ground-Water Model—User Guide to Modularization Concepts and the Ground-Water Flow Process. USGS Open-File Report 00-92*, 2000, p. 121.
75 C. Zheng and P. P. Wang, *MT3DMS: A Modular Three-Dimensional Multispecies Transport Model for Simulation of Advection, Dispersion, and Chemical Reactions of Contaminants in Groundwater Systems. Documentation and User's Guide*, Department of Geological Sciences, University of Alabama, Alabama, 1999, p. 220.

76 B. Van der Grift and C. G. E. M. Van Beek, *Hardness of Abstracted Groundwater: Indicative Predictions (in Dutch)*, Kiwa, Nieuwegein, the Netherlands, 1996.
77 J. Šimůnek, M. Šejna and M. T. Van Genuchten, *The Hydrus-1D Software Package for Simulating the One-Dimensional Movement of Water, Heat and Multiple Solutes in Variable-Saturated Media, Version 2*, U.S. Salinity Laboratory, Riverside, CA, USA, 1998.
78 G. Carabin and A. Dassargues, Modeling groundwater with ocean and river Interaction, *Water Resour. Res.*, 1999, **35**(8), 2347–2358.
79 P. Orban, *Solute Transport Modelling at the Groundwater Body Scale: Nitrate Trends Assessment in the Geer basin (Belgium)*, PhD Thesis, 2009, Faculty of Applied Sciences, University of Liège, Liège, p. 219.
80 S. Brouyère and et al., The hybrid finite element mixing cell method: a new flexible method for modelling mine ground water problems, *Mine Water and the Environment*, 2009, **28**(2), 102–114.
81 N. J. Jarvis, The MACRO Model (version 3.1). Technical Description and Sample Simulations, *Reports and Dissertations 19*, Department of Soil Sciences, Swedish University of Agricultural Sciences, Uppsala, Sweden, 1994.
82 D. Thiéry, *Logiciel MARTHE—Modélisation d'Aquifére avec maillage Rectangulaire, Transport et HydrodynamiquE, version 4.3. BRGM report R 38210*, BRGM, Orleans, France, 1990, p. 330.
83 EU, *Common Implementation Strategy for the Water Framework Directive (2000/60/EC), Guidance Document No. 15, Guidance on Groundwater Monitoring. Technical Report—002-2007*, Office for Official Publications of the European Communities, Luxembourg, 2007, p. 54.
84 A. J. Tesoriero and et al., Linking ground-water age and chemistry data along flow paths: implications for trends and transformations of nitrate and pesticides, *J. Contam. Hydrol.*, 2007, **94**(1–2), 139.
85 K. R. Burow, J. L. Shelton and N. M. Dubrovsky, Regional nitrate and pesticide trends in ground water in the eastern San Joaquin Valley, California, *J. Environ. Qual.*, 2008, **37**(S5), 249–263.
86 D. A. Saad, Agriculture-related trends in groundwater quality of the glacial deposits aquifer, central Wisconsin, *J. Environ. Qual.*, 2008, **37**(S5), 209–225.
87 M. G. Rupert, Decadal-scale changes of nitrate in ground water of the United States, 1988-2004, *J. Environ. Qual.*, 2008, **37**(S5), 240–248.
88 L. M. Debrewer, S. W. Ator and J. M. Denver, Temporal trends in nitrate and selected pesticides in mid-atlantic ground water, *J Environ. Qual.*, 2008, **37**(S5), 296–308.
89 H. Behrendt, et al., *Nutrient Emissions into River Basins of Germany on the Basis of a Harmonised Procedure*, Umweltbundesamt, Berlin, Germany, 2003.