



## Review

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**Abstract**

Sewage sludges are residues resulting from the treatment of wastewater released from various sources including homes, industries, medical facilities, street runoff and businesses. Sewage sludges contain nutrients and organic matter that can provide soil benefits and are widely used as soil amendments. They also, however, contain contaminants including metals, pathogens, and organic pollutants. Although current regulations require pathogen reduction and periodic monitoring for some metals prior to land application, there is no requirement to test sewage sludges for the presence of organic chemicals in the U. S. To help fill the gaps in knowledge regarding the presence and concentration of organic chemicals in sewage sludges, the peer-reviewed literature and official governmental reports were examined. Data were found for 516 organic compounds which were grouped into 15 classes. Concentrations were compared to EPA risk-based soil screening limits (SSLs) where available. For 6 of the 15 classes of chemicals identified, there were no SSLs. For the 79 reported chemicals which had SSLs, the maximum reported concentration of 86% exceeded at least one SSL. Eighty-three percent of the 516 chemicals were not on the EPA established list of priority pollutants and 80% were not on the EPA's list of target compounds. Thus analyses targeting these lists will detect only a small fraction of the organic chemicals in sludges. Analysis of the reported data shows that more data has been collected for certain chemical classes such as pesticides, PAHs and PCBs than for others that may pose greater risk such as nitrosamines. The concentration in soil resulting from land application of sludge will be a function of initial concentration in the sludge and soil, the rate of application, management practices and losses. Even for chemicals that degrade readily, if present in high concentrations and applied repeatedly, the soil concentrations may be significantly elevated. The results of this work reinforce the need for a survey of organic chemical contaminants in sewage sludges and for further assessment of the risks they pose.

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*Keywords:* Sludge; Biosolids; Land application**Contents**

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## 1. Introduction

Sewage sludges are residues generated at centralized wastewater treatment plants (WWTPs) as a result of the treatment of wastes released from a variety of sources including homes, industries, medical facilities, street runoff and businesses. The use of these sludges as soil amendments is widely practiced in the U.S., where more than 60% of the 6.2 million dry metric tons (MT) of sludge produced annually are applied to land (U.S. Environmental Protection Agency, 1999). Since 1991 when ocean dumping was banned, both the quantity produced and the percentage land-applied have increased (U.S. Environmental Protection Agency, 1999).

Sewage sludges contain nutrients and organic matter that can provide soil benefits, but they also contain contaminants including metals, pathogens, and organic pollutants. The fate of chemical contaminants entering a WWTP depends on both the nature of the chemical and the treatment processes (Zitomer and Speece, 1993). Organic chemicals may be volatilized, degraded (through biotic and/or abiotic processes), sorbed to sludge, or discharged in the aqueous effluent. Degradation results in the creation of breakdown products that can be either more or less toxic than the original compound.

For many hydrophobic organic chemicals, sorption to the sewage sludge solids is the primary pathway for their removal from wastewater. This is especially true of persistent, bioaccumulative toxics that may enter the waste stream (Petrasek et al., 1983). Even volatile chemicals, such as benzene, are commonly found in sewage sludges as a result of sorption to organic substances in the sludge matrix (Wild et al., 1992). After they have been separated from wastewater, land-applied sludges must be treated to reduce pathogens through one of a number of processes including anaerobic digestion, lime stabilization, or composting. Each of these processes has effects on the fate of both pathogens and the organic contaminants in the sludge (Rogers, 1996).

The information available on the concentration of organic chemicals in sewage sludges arises largely from academic reports or from the national sewage sludge survey (NSSS) which was conducted by the U.S. Environmental Protection Agency (EPA) in 1988 (U.S. Environmental Protection Agency, 1990). The NSSS was performed by analyzing samples of the final sludge product collected from approximately 180 wastewater plants for the presence of 411 chemicals. This survey was used in the development of the U.S. regulations (U.S. Environmental Protection Agency, 1996).

Very few countries have rules limiting the concentration of any organic chemicals in sewage sludges (Beck et al., 1995). The European Union is considering establishing limits for a handful of organic chemicals. Under the Clean Water Act, (CFR Section 405 (d)), the rules regarding the concentration of pollutants permitted in land-applied sewage sludges in the U.S. are mandated to be protective of human health and the environment. A biennial review is called for to determine if there are additional chemicals that might pose a risk and should thus be subject to regulatory review.

To date, EPA has not established regulations for any organic chemicals and there is no federal requirement to monitor the type or concentration of organic chemicals in sludges. When promulgating the original rules in 1993 (CFR 40 Part 503), the EPA declined to include any organic contaminants. There were three criteria that led to the elimination of all of those considered: 1. the chemical was no longer in use in the U.S.; 2. the chemical was detected in 5% or fewer of the sludges tested in the NSSS; or 3. a hazard screening showed the chemical to have a hazard index of one or greater (Beck et al., 1995). Where sufficient data were lacking to evaluate the hazard, for example the lack of fate and transport data, that chemical and pathway were also eliminated from further consideration (U.S. Environmental Protection Agency, 1996).

Concerns with this process include the persistence of some chemicals in the environment despite their elimination in commerce, the high detection limits for some chemicals, and the potential risks posed by chemicals that were eliminated from consideration merely due to a lack of data (National Research Council, 2002). In a court-ordered review of additional contaminants, the EPA reconsidered regulation of some organic chemicals. In that review, it eliminated chemicals that were detected in 10% or fewer of the sludges in the NSSS. Of the 411 analytes in the NSSS 269 were not detected and 69 were detected in fewer than 10% of the sludges. Fifteen of the 73 remaining chemicals were eliminated due to lack of toxicity data (U.S. Environmental Protection Agency, 1996). Hazard screening analysis was conducted on the remaining chemicals. Dioxins, furans and co-planar PCBs were the only organic chemicals that remained and a risk assessment was then conducted (U.S. Environmental Protection Agency, 2002). Based on the assessment, EPA decided not to extend regulation to dioxins or any other organic pollutant (U.S. Environmental Protection Agency, 2003a). The Round 2 review conducted by the EPA in 2003 was not limited to the chemicals analyzed in the

NSSS. It considered 803 chemicals and resulted in the selection of 15 chemicals as candidates for regulation based on available human health or ecological risk end points but not on concentration data from sludges. Among those were 9 organic chemicals (U.S. Environmental Protection Agency, 2003b).

The National Research Council of the U.S. Academy of Sciences (NRC) conducted two reviews of the land application of biosolids (National Research Council, 1996; 2002). Their 2002 report included a comparison of the limits of detection for samples analyzed in the NSSS to EPA soil screening limits (SSLs) and pointed out that high limits of detection for many chemicals in the NSSS were a concern. The SSLs are conservative risk-based soil concentrations of selected industrial pollutants (93 organic and 16 inorganic compounds) that are used in determining whether a site specific risk assessment is required at a Superfund site (U.S. Environmental Protection Agency Superfund, 1996).

The SSLs were used by the NRC as an indicator of concentrations that might pose a risk requiring remediation. For 5 of 8 organic chemicals examined in the NRC report, most sludge samples analyzed in the NSSS had limits of detection that were higher than the EPA-established SSLs. Thus the NSSS results were not sensitive enough to detect pollutant concentrations that, if present in soil at a Superfund site, would have triggered a risk assessment. For example, in the case of hexachlorobenzene (HCB), the NSSS did not detect HCB in any of the 176 samples tested, thus prompting EPA to exclude it from regulatory consideration. The NSSS limits of detection exceeded 5 mg/kg for the majority of samples and was greater than 100 mg/kg for 4 samples (National Research Council, 2002). Depending on the pathway of exposure being considered, the SSLs for HCB range from 0.1 to 2 mg/kg. Only one of the NSSS samples reached a limit of detection of 0.1 mg/kg. Analysis of the data compiled in this paper revealed that 9 of the 13 reports of HCB concentrations in sewage sludges exceeded 0.1 mg/kg and 3 exceeded 2 mg/kg. Thus the majority of samples exceeded an SSL for HCB.

In addition to concerns regarding analytical limitations, the introduction of new chemicals into commerce, suggests that there is a need for a new survey in order to better characterize sludges with respect to the presence and concentration of contemporary organic chemicals. Flame retardants, surfactants, chlorinated paraffins, nitro and polycyclic musks, pharmaceuticals, odorants, as well as chemicals used in treating sludges (such as dewatering agents) are among the chemical categories suggested by the NRC as compounds requiring

additional data collection and consideration in future risk assessments (National Research Council, 2002).

Although the EPA conducted a limited survey of sludges in 2001 to determine the concentration of dioxins, furans and co-planar PCBs, and plans to conduct a survey of sludges to test for the 9 organic chemicals being considered for regulation, it is not proposing a broader survey of organic chemicals in sludges (U.S. Environmental Protection Agency, 2003b).

## 2. Methods

To help fill the gaps in knowledge regarding the presence and concentration of organic chemicals in sewage sludges, we examined the peer-reviewed literature and official governmental reports to compile available data on the concentration of organic chemicals reported in sludges. In some cases sources did not contain sufficient information to permit comparison of chemical concentrations as a function of sludge dry weight and were therefore not included. One hundred and thirteen usable data sets were obtained. Reports were inconsistent in providing individual versus average or median values so we have reported the ranges detected and are not able to offer averages. Where available, average values from a specific report are noted (supporting information 1). There are several important aspects of wastewater and sludge treatment that can affect the fate of organic chemicals. Unfortunately many reports do not include such information. Where available, the type of treatment is noted (supporting information 1). Similarly, most reports did not include information on the type of catchment area or on significant non-domestic inputs that might contribute particular chemicals.

The chemicals were grouped into 15 classes and the range of concentrations reported for each chemical was recorded. Data were found for 516 chemicals and the range of concentrations detected in each of the sources was recorded (supporting information 1). For ease of presentation, this list was reduced to 267 chemicals through the grouping of congeners and isomeric compounds. The range of concentrations for compounds that have been reported in sewage sludges and the sources from which these data were obtained are shown in Table 1.

To provide a context for the sludge concentration data, we sought soil pollutant concentration standards with which to compare the sludge concentrations. We found that the U.S. SSLs, soil clean-up standards in Ontario and Dutch Intervention values were supported

Table 1

Concentrations of organic chemicals reported in sewage sludges and sources of those data

	Range mg/kg dry wgt	Data sources <sup>a</sup>
<i>Aliphatics—short chained and chlorinated</i>		
Acrylonitrile	0.0363–82.3	[1]
<b>Butadiene</b> (hexachloro-1,3-) <sup>SSL</sup>	ND–8	[1–4]
Butane (1,2,3,4-diepoxy)	ND–73.9	[5]
Butanol (iso)	ND–0.165	[5]
Butanone (2-)	ND–1540	[5]
<b>Carbon disulfide</b> <sup>SSL</sup>	ND–23.5	[5]
Crotonaldehyde	ND–0.358	[5]
Cyclopentadiene (hexachloro) <sup>SSL</sup>	<0.005	[2]
<b>Ethane (hexachloro)</b> <sup>SSL</sup>	0.00036–61.5	[3]
Ethane (monochloro)	ND–24	[3]
Ethane (pentachloro)	0.0003–9.2 g	[3]
Ethane (tetrachloro)	<0.1–5.0	[6]
<b>Ethane (trichloro)</b> isomers <sup>SSL</sup>	ND–33	[7]
<b>Ethylene (dichloro)</b> <sup>SSL</sup>	<0.01–865	[3,8]
<b>Ethylene (monochloro)</b>	<0.025–110	[2,3]
<b>Ethylene (tetrachloro)</b> <sup>SSL</sup>	ND–50	[1–3,5,7,8]
<b>Ethylene (trichloro)</b> <sup>SSL</sup>	ND–125	[2,3,5,7]
Hexanoic acid	ND–1960	[5]
Hexanone (2-)	ND–12.7	[5]
<b>Methane (dichloro)</b> <sup>SSL</sup>	ND–262	[3,5,8,9]
Methane (monochloro)	ND–30	[5]
<b>Methane (tetrachloro)</b> <sup>SSL</sup>	ND–60	[2,3,5–7]
<b>Methane (trichloro)</b> <sup>SSL</sup>	ND–60	[2,5–7]
Methane (trichlorofluoro)	ND–3.97	[5]
<i>N</i> -alkanes (polychlorinated)	1.8–93.1	[10]
<i>N</i> -alkanes	ND–758	[5]
Organic halides absorbable (AOX) and extractable (EOX)	1–7600	[7,11–13]
Pentanone (methyl)	ND–0.567	[5]
Polyorganosiloxanes	8.31–5155	[14–18]
<b>Propane (dichloro)</b> isomers <sup>SSL</sup>	ND–1230	[1,3,5]
Propane (trichloro)	0.00459–19.5	[1,3]
Propanenitrile (ethyl cyanide)	ND–64.7	[5]
Propanone (2-)	ND–2430	[5]
Propen-1-ol (2-)	ND–0.0312	[5]
Propene (trichloro)	<0.0010–167	[1]
<b>Propene chlorinated</b> isomers <sup>SSL</sup>	0.002–1230	[3,5]
Propenenitrile (methyl)	ND–218	[5]
Squalene	ND–16.7	[5]
Sulfone (dimethyl)	ND–0.784	[5]
<i>Chlorobenzenes</i>		
<b>Benzene (dichloro)</b> isomers <sup>SSL</sup>	ND–1650	[2,3,5,8, 19,20]
<b>Benzene (hexachloro)</b> <sup>SSL</sup>	ND–65	[1,2,4,7,11, 20–22]
<b>Benzene (monochloro)</b> <sup>SSL</sup>	ND–846	[3,5,19]

Table 1 (continued)

	Range mg/kg dry wgt	Data sources <sup>a</sup>
<i>Chlorobenzenes</i>		
Benzene (pentachloro)	<0.005–<0.01	[2,20]
Benzene (tetrachloro)	<0.001–0.22	[2,20]
<b>Benzene (trichloro)</b> isomers <sup>SSL</sup>	ND–184	[2,3,5,19,20]
<i>Flame retardants</i>		
Brominated diphenyl ether congeners (BDEs)	<0.008–4.89	[23–30]
Cyclododecane (hexabromo) isomers	<0.0006–9.120	[31]
Tetrabromobisphenol A	<0.0024–3322	[32]
Tetrabromobisphenol A (dimethyl)	<0.0019	[32]
<i>Monocyclic hydrocarbons and heterocycles</i>		
Acetophenone	ND–6.92	[5]
Aniline (2,4,5-trimethyl)	ND–0.220	[5]
<b>Benzene</b> <sup>SSL</sup>	ND–11.3	[3,5,33]
Benzene (1,4-dinitro)	ND–4.4	[5]
<b>Benzene (ethyl)</b> <sup>SSL</sup>	ND–65.5	[3,5]
<b>Benzene (mononitro)</b> <sup>SSL</sup>	ND–1.55	[2,5]
Benzene (trinitro)	12	[34]
Benzenethiazole (2-methylthio)	ND–64.4	[5]
Benzenethiol	ND–3.25	[5]
<b>Benzoic acid</b> <sup>SSL</sup>	ND–835	[5]
Benzyl alcohol	ND–156	[5]
<b>Aniline (chloro)</b> ( <i>P</i> -) <sup>SSL</sup>	ND–40.2	[5]
Cymene ( <i>P</i> -)	ND–84.3	[5]
Dioxane (1,4-)	ND–35.3	[5]
Picoline (2-)	ND–365	[5]
<b>Styrene</b> <sup>SSL</sup>	ND–5850	[3,5]
Terpeniol (alpha)	ND–2.56	[5]
Thioxanthone-9-one	ND–19.6	[5]
<b>Toluene</b> <sup>SSL</sup>	ND–1180	[3,5,6,8,9, 34,35]
Toluene (chloro)	1.13–324	[5]
<b>Toluene (2,4-dinitro)</b> <sup>SSL</sup>	ND–10	[2,5,34]
Toluene (para nitro)	100	[34]
Toluene (trinitro)	12	[34]
Xylene isomers <sup>SSL</sup>	ND–6.91	[5,8,33, 35–37]
<i>Nitrosamines</i>		
<b><i>N</i>-nitrosodiphenylamine</b> <sup>SSL</sup>	ND-19.7	[5]
<i>N</i> -nitrosodiethylamine	ND–0.0038	[38]
<i>N</i> -nitrosodimethylamine	0.0006–0.053	[38]
<i>N</i> -nitrosodi- <i>n</i> -butylamine	ND	[38]
<i>N</i> -nitrosomorpholine	ND–0.0092	[38]
<i>N</i> -nitrosopiperidine	ND–trace	[38]
<i>N</i> -nitrosopyrrolidine	ND–0.0042	[38]
<i>Organotin</i>		
Butyltin (di)	0.41–8.557	[39–44]
Butyltin (mono)	0.016–43.564	[39–44]

Table 1 (continued)

	Range mg/kg dry wgt	Data sources <sup>a</sup>
<i>Organotins</i>		
Butyltin (tri)	0.005–237.923	[9,39–44]
Phenyltin (di)	0.1–0.4	[42,43]
Phenyltin (mono)	0.1	[42,43]
Phenyltin (tri)	0.3–3.4	[42,43]
<i>Personal care products and pharmaceuticals</i>		
Acetaminophen	0.0000006–4.535	[45]
Gemfibrozil	ND–1.192	[45]
Ibuprofen	0.000006–3.988	[45]
Naproxen	0.000001–1.022	[45]
Salicylic acid	0.000002–13.743	[45]
<i>Antibiotics</i>		
Ciprofloxacin	0.05–4.8	[46,47]
Doxycycline	<1.2–1.5	[47]
Norfloxacin	0.01–4.2	[46,47]
Ofloxacin	<0.01–2	[47]
Triclosan (4-chloro-2-(2,4-dichlorophenoxy)-phenol and related compounds)	ND–15.6	[25,48–50]
<i>Fluorescent whitening agents</i>		
BLS (4,4'-bis(4-chloro-3-sulfoxy)phenyl)-biphenyl)	5.4–5.5	[51]
DAS 1 (4,4'-bis[4-anilino-6-morpholino-1,3,5-triazin-2-yl-amino]stilbene-2,2'-disulfonate)		
DSBP (4,4'-bis(2-sulfoxy)phenyl)biphenyl)	86–112	[51]
<i>Fragrance material</i>		
Acetyl Cedrene	9.0–31.1	[52]
Amino Musk Ketone	ND–0.362	[37]
Amino Musk Xylene (AMX)	ND–0.0315	[37]
Cashmeran (DPMI) (6,7-dihydro-1,1,2,3,3-pentamethyl-4(5H)-indanone)	ND–0.332	[34,37]
Celestolide (1-[6-(1,1-Dimethylethyl)-2,3-dihydro-1,1-methyl-1H-inden-4-yl]-ethanone)	0.010–1.1	[34,37,53,54]
Diphenyl Ether	ND–99.6	[5,52]
Galaxolide (HHCB) (1,3,4,6,7,8-Hexahydro-4,6,6,7,8,8-hexamethylcyclopenta[g]-benzopyran)	ND–81	[25,34,37,52–56]
Galaxolide lactone (1,3,4,6,7,8-Hexahydro-4,6,6,7,8,8-hexamethylcyclopenta[g]-2-benzopyran-1-one)	0.6–3.5	[54]
Hexyl salicylate	Trace–1.5	[52]

Table 1 (continued)

	Range mg/kg dry wgt	Data sources <sup>a</sup>
<i>Fragrance material</i>		
Hexylcinnamic Aldehyde (Alpha)	4.1	[52]
Methyl ionone (gamma)	1.1–3.8	[52]
Musk Ketone (MK) (4-tertbutyl-3,5-dinitro-2,6-dimethylacetophenone)	ND–1.3	[37,52,57]
Musk Xylene (1-tert-butyl-3,5-dimethyl-2,4,6-trinitrobenzene)	ND–0.0325	[57]
OTNE (1-(1,2,3,4,5,6,7,8-octahydro-2,3,8,8-tetramethyl-2-naphthalenyl))	7.3–30.7	[52]
Phantolide (1-[2,3-Dihydro-1,1,2,3,3,6-hexamethyl-1H-inden-5-yl]-ethanone)	0.032–1.8	[34,37,53,54]
Tonalide (1-[5,6,7,8-Tetrahydro-3,5,5,6,8,8-hexamethyl-2-naphthalenyl]-ethanone)	ND–51	[25,37,52–55]
Traseolide (ATII) (1-[2,3-Dihydro-1,1,2,6-tetramethyl-3-(1-methyl-ethyl)-1H-inden-5-yl] ethanone)	0.044–1.1	[53,54]
<i>Pesticides</i>		
<b>Aldrin</b> <sup>SSL</sup>	ND–16.2	[1–5,21,22,33,58,59]
Azinphos Methyl	ND–0.279	[5]
Benzene (pentachloronitro)	ND–8.83	[5]
Captan	ND–0.968	[5]
<b>Chlordane</b> <sup>SSL</sup>	ND–16.04	[1,3,5]
Chlorobenzilate	ND–0.104	[2,5]
Chloropyrifos	ND–0.529	[5]
Ciodrin	ND–0.093	[5]
<b>Cyclohexane isomers (lindane and others)</b> <sup>SSL</sup>	ND–70	[1–7,9,11,21,22,59–62]
<b>DDT and related congeners</b> <sup>SSL</sup>	ND–564	[1–5,7,9,11,21,22,33,58,60–62]
Diallate	ND–0.394	[2,5]
Diazinon	ND–0.151	[5]
Dicrotophos (Bidrin)	ND–0.550	[5]
<b>Dieldrin</b> <sup>SSL</sup>	ND–64.7	[1–7,21,22,33,60,61]
Dimethoate	ND–0.340	[2,5]
Disulfotone	<0.0050	[2]
Endosulfans	ND–0.280	[2,4,5,21]
<b>Endrin</b> <sup>SSL</sup>	ND–1.17	[1,2,4,5,21,22,59]
Famphur	<0.0050–0.400	[2]

(continued on next page)

Table 1 (continued)

	Range	Data sources <sup>a</sup>
	mg/kg dry wgt	
<i>Pesticides</i>		
<b>Heptachlor epoxides</b> <sup>SSL</sup>	ND–0.780	[1,2,5,21]
<b>Heptachlor</b> <sup>SSL</sup>	ND–16	[2,3,5,21,22]
Isobenzan	ND–0.130	[4]
Isodrin	ND	[4]
<b>Isophorone</b> <sup>SSL</sup>	<0.0050–0.08294	[2]
Leptophos	ND–0.319	[5]
Methoxychlor <sup>SSL</sup>	<0.015–0.330	[2]
Mevinphos (phosdrin)	ND–0.148	[5]
Naled (Dibrom)	ND–0.484	[5]
Naphthoquinone (1,4-)	<0.0050	[2]
Nitrofen	ND–0.195	[5]
Parathion (ethyl)	<0.0050–0.380	[2]
Parathion (methyl)	<0.0050–0.070	[2]
Permethrin isomers	<0.15–163	[20,63]
<b>Phenoxy herbicides</b> <sup>SSL</sup>	ND–7.34	[1,2,5]
Phenoxypropanoic acid (trichloro)	ND–0.121	[5]
Phorate ( <i>O,O</i> -diethyl <i>S</i> -[(ethylthio) methyl] phosphorodithioate)	<0.0050–0.200	[2]
Phosphamidon	ND–0.232	[5]
Pronamide (dichloro (3,5-)- <i>N</i> -(1,1-dimethylpropynyl) benzamide)	<0.0050–0.008	[2]
Pyrophosphate (tetraethyl)	ND–20	[5]
Quintozene	ND–0.100	[4]
Safrol (iso)	<0.0050–0.750	[2]
Safrole (EPN)	ND–0.545	[2]
<b>Toxaphene</b> <sup>SSL</sup>	51	[3]
Trichlorofon	ND–2.53	[5]
Trifluralin (Treflan)	ND–0.235	[5]
<i>Phenols</i>		
Bisphenol-A (BPA)	0.00010–32,100	[18,49,64,65]
Hexachlorophene (HCP)	0.0226–1.190	[49]
Hydroquinone	0.14–223	[3]
Hydroxybiphenyls	ND–0.172	[64]
<b>Phenol</b> <sup>SSL</sup>	ND–920	[2,3,5,7,8,36,66]
<b>Phenol chloro congeners</b> <sup>SSL</sup>	<0.003–8490	[1–3,5–9,33,35,49,61,66–68]
Phenol chloro methyl congeners	ND–136	[2,3,5,8,9,61,64]
<b>Phenol methyl congeners</b> <sup>SSL</sup>	ND–1160	[2,3,5,7–9,34,66]
Phenol nitro methyl congeners	0.2–187	[5]
<b>Phenols nitro congeners</b> <sup>SSL</sup>	<0.003–500	[2,3,8]

Table 1 (continued)

	Range	Data sources <sup>a</sup>
	mg/kg dry wgt	
<i>Phthalate acid esters/plasticizers</i>		
<b>Bis(2-chloroethyl) ether</b> <sup>SSL</sup>	<0.020–0.130	[2]
Bis(2-chloroisopropyl) ether	<0.150–5.700	[2]
Bis(2-chloroethoxy) methane	<0.020–0.240	[2]
Di(2-ethylhexyl) adipate	<0.100–0.450	[2]
<b>Phthalates</b> <sup>SSL</sup>	ND–58,300	[2,3,5–9,28,33,36,58,69–73]
<i>Polychlorinated biphenyls, naphthalenes, dioxins and furans</i>		
Aroclor 1016	0.2–75	[6,74]
Aroclor 1248	ND–5.2	[5,6,33,58]
Aroclor 1254	0.0667–1960	[1,5]
Aroclor 1260	ND–433	[1,5,6,58,60]
Biphenyl (decachloro)	0.11–2.9	[1]
Biphenyls (polybrominated)	431	[3]
Dibenzofuran	ND–59.3	[5]
Dioxins and furans (polychlorinated dibenzo)	ND–1.7	[5,8,72,75–81]
PCB congeners	ND–765	[2–5,7,11,13,21,22,28,35,53,59,61,71,72,79,81–87]
Phenylether (chloro)	<0.020	[2]
Terphenyls and naphthalenes (polychlorinated)	ND–11.1	[2,3,5,9,28,53]
<i>Polynuclear aromatic hydrocarbons</i>		
Acenaphthene <sup>SSL</sup>	ND–6.6	[2,5,8,21,53,82,88]
Acenaphthylene	0.00360–0.3	[2,8,21,53]
Anthracene <sup>SSL</sup>	ND–44	[2,3,5,8,21,28,31,53,74,88,89]
Benzidine	12.7	[3]
<b>Benzo(a)anthracene</b> <sup>SSL</sup>	ND–99	[2,3,5,8,21,53,82,88–90]
Benzo[ghi]perylene	ND–12.9	[1,2,5–8,21,22,28,53,88–91]
<b>Benzo(a)fluoranthene congeners</b> <sup>SSL</sup>	0.006–34.2	[3,89]
Benzo(a)fluorene congeners	ND–8.1	[62,89]
<b>Benzo(b)fluorene congeners</b> <sup>SSL</sup>	ND–24.7	[1–3,5–8,11,21,22,28,33,53,62,82,88–91]

Table 1 (continued)

	Range mg/kg dry wgt	Data sources <sup>a</sup>
<i>Polynuclear aromatic hydrocarbons</i>		
Biphenyl	ND–15,300	[3,5,53]
<b>Chrysene<sup>SSL</sup></b>	ND–32.4	[3,5,8,21,53, 82,88,90]
Chrysene+triphenylene	0.01–14.7	[2,89]
<b>Dibenzoanthracene congeners<sup>SSL</sup></b>	ND–13	[2,3,8,21,53, 88,89,91]
Dibenzothiophene	ND–1.47	[5]
Diphenyl amine	ND–32.6	[5]
Fluoranthene <sup>SSL</sup>	ND–60	[1–3,5–8,21, 22,28,33,53,62, 82,88–90]
Fluorene <sup>SSL</sup>	<0.01–8.1	[2,8,21,53, 82,88]
Fluorene (nitro)	0.941	[28]
<b>Indeno(1,2,3-c,d) pyrene<sup>SSL</sup></b>	ND–9.5	[2,7,8,21,22, 28,53,88–91]
<b>Naphthalene<sup>SSL</sup></b>	ND–6610	[2,3,5,6,8,21, 36,53,62,88]
Naphthalene methyl isomers	ND–136	[2,5,28,53]
Naphthalene methyl congeners		
Naphthalene nitro congeners	ND–0.0798	[28]
Perylene	ND–69.3	[3,5,53,89,91]
Phenanthrene	<0.01–44	[2,3,5,6, 8,21,28,53, 62,82,88–90]
Phenanthrene methyl isomers	ND–37.4	[5,53]
Pyrene <sup>SSL</sup>	0.01–37.1	[2,3,5,6, 8,21,53, 82,88–90]
Pyrene (phenyl)	0.06–6.86	[1]
Retene (7-isopropyl- 1-methylphenanthrene)	0.260	[28]
Total PAH	ND–199	[9,11,28, 72,86]
Triphenylene	ND–15.4	[5]
<i>Sterols, stanols and estrogens</i>		
Campestanol (5a+5b)	3.0–14	[55]
Campesterol	6.3	[55]
Cholestanol (5a-)	22.7	[49,87]
Cholesterol	57.4	[55]
Coprostanol	216.9	[55]
Estradiol (17b)	0.0049–0.049	[92,93]
Estrone	0.016–0.0278	[92,93]
Ethinylestradiol (17a)	<0.0015–0.017	[92,93]
Sitostanol (5a-b+5b-b-)	14.1–93.9	[55]
Sitosterol (b-)	29.6–31.1	[55]
Stigmastanol (5a+5b)	1.9–12.9	[55]
Stigmasterol	6.7	[55]

Table 1 (continued)

	Range mg/kg dry wgt	Data sources <sup>a</sup>
<i>Surfactants</i>		
Alcohol ethoxylates	ND–141	[70,94,95]
Alkylbenzene sulfonates	<1–30,200	[6,7,9, 70–72,74, 85,94,96–98]
Alkylphenolcarboxylates	10–14	[92]
Alkylphenolethoxylates	ND–7214	[2,7,25,28, 49,69,71,72, 85,90,92, 94,99–101]
Alkylphenols (nonyl and octylphenol)	ND–559,300	[2,6,9,18,25, 28,36,49,64, 69,74,92, 95,99–107],
Coconut diethanol amides	0.3–10.5	[70]
Poly(ethylene glycol)s	1.7–17.6	[70]
<i>Triaryl/alkyl phosphate esters</i>		
Cresyldiphenyl phosphate	0.61–179	[3]
Tricresyl phosphate	0.069–1650	[3]
Tricresyl phosphate	<0.020–12.000	[2]
Tri- <i>n</i> -butylphosphate	<0.020–2.400	[2]
Triphenylphosphate	<0.020–1.900	[2]
Trixylyl phosphate	0.027–2420	[3]

See Supporting Information 1 for further detail.

**Boldfaced**= one or more reported concentrations exceed an SSL. SSLs may be established only for a particular congener. Table 1 groups congeners and where any one of the congener concentration exceeds an SSL for that congener, the group of congeners is shown in bold. Available data for specific congeners is shown in supporting information 2.

<sup>SSL</sup> indicates that SSLs have been established for one or more congener in this group.

ND indicates not detected where the lower limit of detection is not specified. >XX indicates not detected at the specified (XX) limit of detection.

<sup>a</sup> The data sources for this table are identified by number and cited below as a part of this table.

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by risk-based analyses. The Ontario regulatory maximum soil concentration limits address several different land uses and pathways of exposure for 118 chemicals (Ontario Ministry of the Environment, 2004). The Dutch system includes target values that seek to prevent harm to human and ecological systems as well as intervention values where predicted harm requires clean-up to be implemented. The Ontario and Dutch values are generally comparable to the U.S. SSLs, but values for specific chemicals are not identical, presumably due to differences in assumptions (Netherlands Ministry of Housing Spatial Planning and Environment, 2000).

For the purposes of this paper, we compared the reported sludge concentrations to the SSL values for those compounds for which EPA has established an SSL. The SSLs are not regulatory standards, but are guidelines used by EPA relative to cleaning up industrially-contaminated sites. Sites with soil concentrations lower than the SSLs are considered “clean,” while sites with higher concentrations require site-specific risk analysis. Using default values for a residential exposure scenario, the EPA risk-based SSLs address exposure pathways including direct ingestion of contaminated soil, inhalation, dermal exposure, drinking of groundwater contaminated by migration of chemicals through soil, and ingestion of homegrown produce contaminated via plant uptake (U.S. Environmental Protection Agency Superfund, 1996). The groundwater pathway includes two values, one assuming no dilution or attenuation (1 DAF) and the other assuming a 20-fold dilution/attenuation (20 DAF). SSLs do not include risks posed by ingesting animal products grown on contaminated soils, nor do they address environmental and ecologic risks. These human health SSLs are based on a  $10^{-6}$  risk for carcinogens or a hazard quotient of 1 for non-carcinogens and separate SSL concentrations are listed for four different exposure pathways (ingestion, inhalation, groundwater 20 DAF, groundwater 1 DAF). For most organic contaminants, the groundwater SSL that assumes no attenuation or dilution is the most restrictive concentration (supporting information 2).

It is likely that the concentration of a chemical in a soil to which sludge has been applied would be lower than the concentration in the sludge itself due to mixing and subsequent dilution with soil as well as through degradation, volatilization and leaching processes. A single application of sludge tilled into the soil would be diluted approximately 100-fold, but concentrations would increase with repeated applications when losses are not as great as application rates and would also be higher in surface soils if sludge is not tilled into the soil

such as in pasture application. Despite the differences between contaminated soils and sludges, the NRC (National Research Council, 2002) used SSLs as an EPA-established metric to suggest whether further evaluation might be warranted. We thus report sludge concentrations of organic contaminants that exceed an SSL (Table 1; supporting information 2).

Two other EPA-generated lists of chemicals were also used to evaluate the organic chemicals reported in sludges. The first is a list of chemicals generated in 1979 and modified in 1981 for which technology-based water effluent limitations were required (Keith and Telliard, 1979). These 126 chemicals, known as priority pollutants, reflect the knowledge of contaminants in industrial wastewater effluents during the 1970s. One hundred and eleven of these are organic chemicals. Although there are no federal requirements for monitoring these compounds in sewage sludges, some states, including New York (New York State Department of Environmental Conservation, 2003), require screening of land-applied sludges for these priority pollutants. The second list includes chemicals that laboratories performing analyses on Superfund site soils must be able to detect and quantify. These 143 chemicals are known as target compounds (U.S. Environmental Protection Agency, 2004). Table 2 provides a summary, by class, of the number of chemicals reported in sludges that fall into these groups.

### 3. Results and discussion

Tens of thousands of organic chemicals are currently in use, however sludge concentration data could only be found for 516 organic chemicals in the peer reviewed literature and official government reports (supporting information 1). Table 2 shows the number of compounds in each of the 15 classes for which concentration data were found, and the number of studies from which these data were obtained.

Ninety of the 111 organic priority pollutants and 101 of the 143 target compounds were reported in sludges (Table 2). No data were found for the other 21 organic priority pollutants or 42 target compounds. Eighty-three percent of the reported chemicals were not on the priority pollutant list and 80% were not on the target compounds list. Thus monitoring sludges for priority pollutants will not capture the vast majority of chemicals that may be present.

Six of the 15 chemical classes for which data were found did not contain compounds included among the priority pollutants, target compounds, or those compounds with SSLs (Table 2). This may be due in part to

Table 2

Number of chemicals reported in sludges in each class, number of studies from which data were obtained, number that are priority pollutants, target compounds or for which there are SSLs, and number for which maximum reported concentrations in sludges exceed an SSL

	# chem	# of studies	# PP chem	# TC chem	# chem with SSLs	# chem that exceed an SSL
Aliphatics	58	19	16	17	16	15
Chlorobenzenes	11	13	6	7	5	5
Flame retardants	29	11	0	0	0	
Monocyclic HC	34	12	7	12	11	10
Nitrosamines	7	1	2	1	1	1
Organotins	6	7	0	0	0	
PCPs	36	17	0	0	0	
Pesticides	71	20	18	19	18	15
Phenols	40	20	10	14	9	8
Phthalate	19	16	9	8	6	6
PCBs	108	38	5	6	0	
PAHs	52	25	18	18	13	8
Sterols and stanols	16	3	0	0	0	
Surfactants	23	33	0	0	0	
Triaryl/alkyl phosphate.esters	6	2	0	0	0	
Total	516	113 <sup>a</sup>	91	102	79	68

<sup>a</sup> Note: # of studies is not a sum of the list above because some studies include data for more than one class.

the fact that all three of these lists arose out of a response to a concern over the fate of industrial contaminants. Thus some chemicals, such as personal care products, that are present in sludges primarily as a result of non-industrial sources, do not appear on those lists. In addition, the priority pollutant list is 25 years old, so industrial chemicals of current and emerging concern, such as polybrominated diphenyl ethers, which were not in wide use at that time, were not included.

There are SSLs for 15% of the 516 organic chemicals reported in sludges. The reported maximum sludge concentration exceeded an SSL for 86% of the chemicals for which there are SSLs (Table 2, supporting information 2). This high proportion is observed in most classes, with PAHs as an exception.

The proportion of individual reports that exceed an SSL for a particular chemical was examined to determine whether such exceedances were the result of single high-concentration reports or whether most

reported values exceeded an SSL. The data show that for chemicals in some classes such as aliphatics and monocyclic hydrocarbons, most reported concentrations for chemicals within that class exceed an SSL while for other classes including phthalates and polyaromatic hydrocarbons, a much smaller percentage of the reported concentrations were high enough to exceed an SSL (Table 3). However, even within these classes, there are some chemicals for which a high percentage of reports exceed an SSL (Fig. 1).

As a result of an evaluation of additional sludgeborne chemicals for which regulation should be considered, the EPA has suggested that it will conduct limited additional sludge testing including efforts to monitor the presence of 9 organic chemicals (acetone, anthracene, carbon disulfide, 4-chloroaniline, diazinon, fluoranthene, methyl ethyl ketone, phenol, and pyrene) (U.S. Environmental Protection Agency, 2003b). In the present work, no data were found for two of the 9

Table 3

The percentage of reported concentrations that exceed an SSL for chemicals within a class for which there are SSLs

	% for which 100% reports exceed SSL	% for which 75–99% reports exceed	% for which 50–74% reports exceed	% for which 25–50% reports exceed	% for which 0–25% reports exceed
Aliphatics	75	6	19	0	0
Chlorobenzenes	20	20	60	0	0
Monocyclic	75	8	0	0	17
Nitrosamines	100				
Pesticides	31	13	25	6	19
Phenols	22	22	33	11	11
Phthalate	17	0	17	17	50
PAHs	0	23	8	15	54

See Supporting Information 2 for the specific chemicals and SSLs.

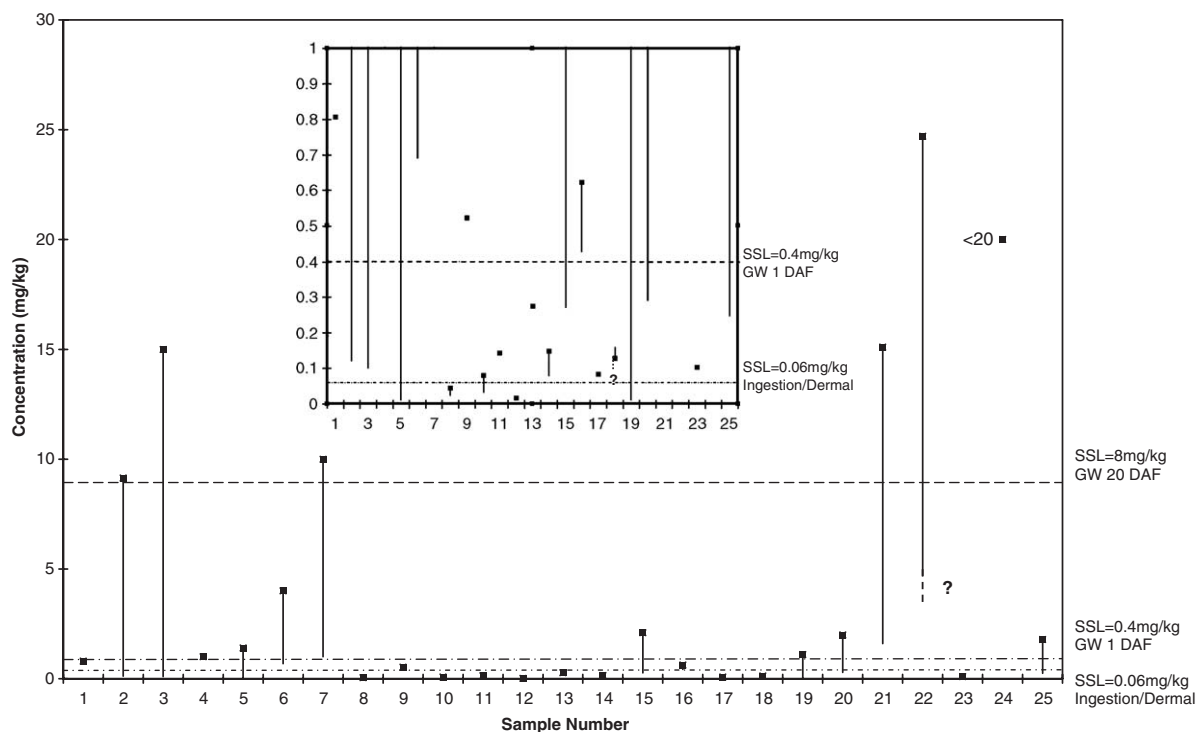


Fig. 1. Concentration (dry wgt) of benzo[a]pyrene in sewage sludges compared to soil screening levels. Note: ? means the report did not specify the concentration of values reported as non-detects.

compounds (acetone and methyl ethyl ketone). Data were found for the other 7 compounds (Table 1; supporting information 1; supporting information 2).

Anthracene was reported in 12 studies with a range from 0.0088 to 44 mg/kg. Six studies detected more than 1 mg/kg, but none exceeded an SSL. Only the NSSS reported concentrations for carbon disulfide, *p*-chloroaniline and diazinon, with maximum concentrations of 23.5, 40.2 and 0.15 mg/kg respectively. The carbon disulfide value exceeded the lower groundwater SSL and the *p*-chloroaniline value greatly exceeded both groundwater SSLs. There are no SSLs for diazinon. Fluoranthene was reported in 17 studies with concentrations ranging from 0.01 to 60 mg/kg, but none exceeded any SSL. Seven studies reported phenol ranging from 0.002 to 920 mg/kg, with concentrations of over 100 mg/kg reported in four studies, suggesting that these high concentrations were not a result of a particular source of contamination or analytic error. Six studies reported concentrations exceeding the lower groundwater SSL and four exceeded both groundwater SSLs. Eleven studies reported pyrene concentrations ranging from 0.1 to 36.8 mg/kg, but none exceeded any SSL. These data suggest that several of the contaminants that EPA proposes to study are not likely to be of concern since data on their concentration in sludges

exist and demonstrate concentrations below SSLs indicating they are unlikely to be present in concentrations high enough to be of significant risk.

Benzo(a)pyrene and hexachlorobenzene were suggested as pollutants requiring further analysis by the NRC in a 1996 report (National Research Council, 1996). In the present work, 19 sources reported benzo(a)pyrene in sludges at concentrations from <0.01 to 25 mg/kg, with 24 of 27 reported concentrations exceeding one or more SSL (Fig. 1; supporting information 2). Hexachlorobenzene was reported by 9 sources. Nine of 13 reported concentrations exceed an SSL (Fig. 2; supporting information 2). These data suggest the value of assessing the risks posed by these chemicals in sludges.

Another group of compounds suggested as a possible concern is nitrosamines. Given the toxicity of nitrosamines and the potential for their formation during the wastewater treatment process, it is surprising that only two sources from the 1980s report nitrosamine concentration in sludges. Of the 7 compounds reported, there are SSLs for only one and the reported concentrations for that compound (*N*-nitrosodiphenylamine) exceed the groundwater and ingestion/dermal SSLs. The NSSS detected *N*-Nitrosodiphenylamine in 1% of the sludges tested and hence it was eliminated from regulatory consideration by EPA. The maximum concentration

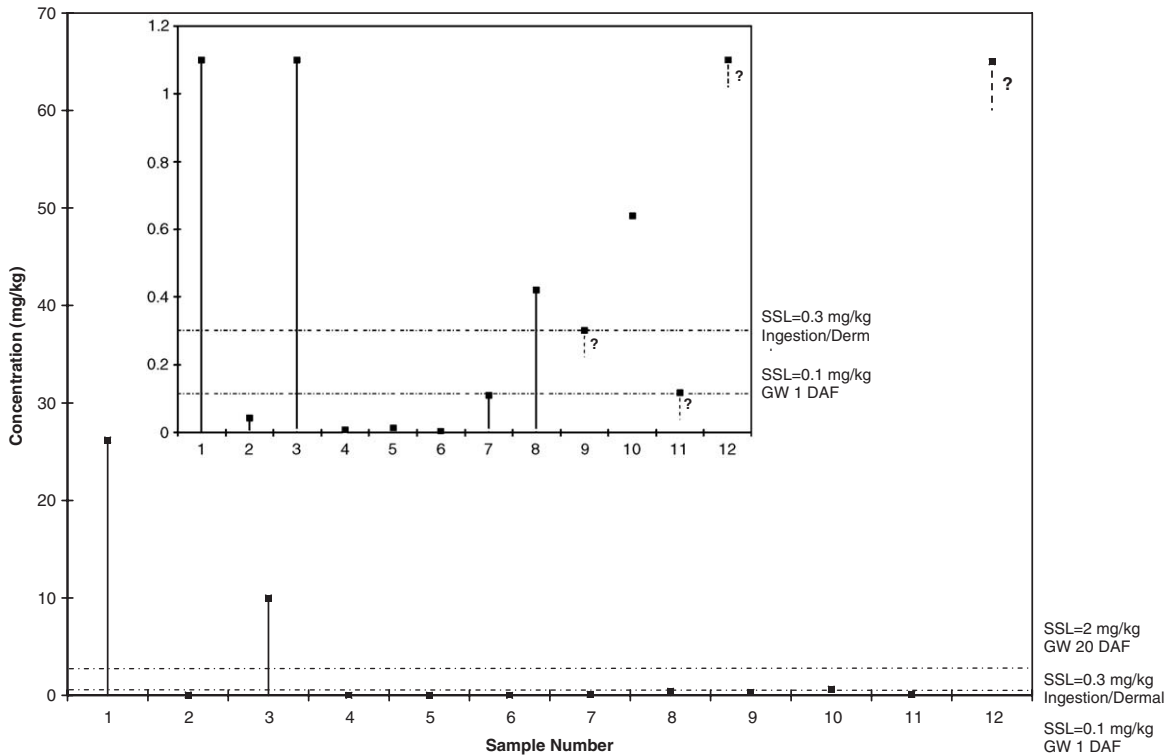


Fig. 2. Concentration (dry wgt) of hexachlorobenzene in sewage sludges compared to soil screening levels. Note: ? means the report did not specify the concentration of values reported as non-detects.

detected was 19.7 mg/kg. Most samples had a limit of detection exceeding 1 mg/kg although detection limits as high as 800 mg/kg were also reported. The high limits of detection in many cases helped prompt the NRC to speculate that *N*-Nitrosodimethylamine may be present in some sludges at concentrations of concern (National Research Council, 1996).

Reported concentrations exceeding an SSL should not be interpreted to indicate a significant risk, but rather indicate that the concentration of those chemicals would be sufficient to require further assessment if present in soil at the same level. While sludge management and environmental processes may alter the concentrations of these chemicals in field situations through mixing with soil, leaching, degradation and other processes, the number of SSL exceedences suggests that assessment of the potential risks may be warranted.

The use of SSLs as a screening tool, does not address some potential routes of human exposure that may represent significant risk (Wild and Jones, 1992), including food chain transfer through the consumption of animal products. For organic contaminants in land applied sludges, this has been suggested as one of the two exposure pathways representing the highest risk, the other being direct ingestion of soil and sludge by humans

(Chaney et al., 1996). Application of sludge products to lawns, athletic fields and home gardens could provide a route for direct ingestion. The lipophilic nature of many organic chemicals found in sludges causes them to accumulate in the fat of exposed animals. Livestock may be exposed to sludge contaminants through sludge adhering to plant materials as well as through the ingestion of soil when sludges are applied to pasture (Fries, 1996).

Much of the work evaluating the potential risks posed by organic chemicals in sludges addresses human health risks. However, in addition to potential human impacts, organic chemicals in land applied sludges may pose environmental or ecological risks. The use of SSLs as a trigger does not account for these risks as most SSLs are currently based only on human health criteria. A number of the chemicals detected in sludges have been shown to function as endocrine disrupters. For example, nonylphenols which are present in sludges at relatively high concentrations (concentrations greater than 1000 mg/kg are not unusual), may be of concern because of their potential impact on wildlife (Environment Canada, 2004), even though they are unlikely to represent a major direct human health risk. Soil processes may also be impacted by organic chemical

contaminants in land applied sludges as suggested by observed fungitoxic effects (Schnaak et al., 1997).

Specifying organic chemicals that should be monitored in sludges is not a simple task because it necessitates a degree of analytical competence that may not be widespread. The EPA has addressed this issue with respect to Superfund sites by developing a list of target compounds which includes priority pollutants in addition to other compounds. Certified laboratories performing analyses of Superfund samples are required to be able to test for these target compounds. As mentioned above, 80% of the organic chemicals reported in this paper, however, were not target compounds and could go undetected even in certified laboratories unless expensive mass spectral analyses were also performed. While the use of standardized methods that have been validated for individual chemicals is essential to ensure data quality, ongoing screening and validation efforts using generalized methods and robust detection technologies are required in order to identify chemicals of emerging concern.

For many compounds, there was wide variation in the reported concentrations found in sewage sludges. There are a number of potential sources of this variation. Discrepancies in analytical methods may account for some of the differences in the range of concentrations reported in this paper (Pryor et al., 2002). For most of the chemicals, no standard methods have been established for either sample extraction or analyte detection. The importance of methodological variation was clearly demonstrated in one report examining extraction efficiency, where a nearly five-fold difference was found in the concentration of several organic chemicals in sludge samples simply as a result of using different solvents (Bolz et al., 2001) and in another report where drying methods resulted in similarly large differences (Scrimshaw et al., 2004).

For some contaminants, differences in the source inputs to the WWTP may explain the range (Bodzek and Janoszka, 1999). For example, the high concentrations reported for some of the polynuclear aromatic hydrocarbons (PAHs) in one study (Constable et al., 1986) were likely due to inputs from local industry including two steel mills. Due to the large number of sludges sampled in the NSSS, that survey included a wide range of concentrations and yielded the highest reported concentrations for a number of contaminants (supporting information 1).

Another source of variability in chemical concentrations may be the type of treatment to which the sludges were subjected. The impact of this variable was difficult to gauge, however, as many reports did not provide information about wastewater and sludge processing methods. Where such information was available, it was noted (supporting information 1). Since pollutant con-

centrations have been found to vary significantly with different types of processing (Wild and Jones, 1989), some of the variation in concentrations may have been a result of the different treatments to which the sludges were subjected (Constable et al., 1986; Wild and Jones, 1989; Zitomer and Speece, 1993; Rogers, 1996) or to differences in sludge retention time (Ternes et al., 2004).

Changes in chemical use over time is another potential source of the large range in reported concentrations. The references from which data were obtained go back as far as 1976, though most were from the 1980s or later. Because of changes in chemical usage, including bans on some chemicals, the introduction of new chemicals and the increasing use of others, the use of old data can be problematic. A new survey of organic chemicals in sludges is needed since the NSSS dates back to 1988 (National Research Council, 2002). Due to the paucity of data, however, even older studies were included in this paper and the date of sampling was included when available (supporting information 1).

The vast majority of the data found were for sludges from the U.S. or Western Europe where chemical use and wastewater treatment are relatively similar, resulting in similar pollutant concentrations. There were, however, some noteworthy differences. In several European countries, for example, bans or the voluntary elimination of compounds such as penta-brominated diphenyl ethers and nonylphenol have been enacted. As a result, concentrations of these chemicals in sludges from those countries have decreased in recent years (Jobst, 1998).

There are also important differences between the European and U.S. approaches to the management of land application of sludges that would likely result in lower soil loadings of contaminants in most European countries. The soil concentration of a sludge-borne pollutant after land application is not only a function of the concentration of the chemical in the sludge, but also the amount of sludge applied. A number of European countries limit application rates either through direct limits on the number of dry MT/ha/yr or by limiting application to *P*-based agronomic rates, which are far more restrictive than the *N*-based rates used in the U.S. In Denmark, for example, no more than 30 kg/ha/yr of P can be applied (Ministry of Environment and Energy, 1997). This equates to an application rate of approximately 1 dry MT/ha/yr. While quantitative limits vary among the European countries, most limit application to a maximum of 1–4 dry MT/ha/yr (Schowanek et al., 2004). In conducting risk assessments, the European Commission assumes an application rate of 5 dry MT/ha/yr (European Commission Joint Research Centre, 2003). This compares to 10 dry MT/ha/yr which was the assumed high-end application rate used by EPA in developing the

regulations for land application (U.S. Environmental Protection Agency, 1995). Another critical management strategy pertains to the prohibition of pasture-application in some countries, which could reduce the potential contamination of animal products.

Other management practices such as depth of mixing into the soil and losses through various environmental processes will also affect chemical concentrations in soils after land application. Degradation is an important component of loss, but may be incomplete or slow, even for relatively easily degraded chemicals such as linear alkyl benzene sulfonates (LAS). LAS is present at such high concentrations in sludges (up to 3% by weight) that incomplete degradation coupled with repeated applications could result in consistently elevated LAS concentrations in soils. This was demonstrated in one study that detected over 10 mg/kg six years after land application of sludge. Importantly, no further decrease was found after two more years, indicating that the residual LAS was resistant to degradation (Carlsen et al., 2002).

#### 4. Conclusion

More data are needed on the chemicals that are in sludges today and on the temporal trends for those chemicals. Relying on existing lists of chemicals such as priority pollutants will not identify many chemicals of current concern.

To make more informed assessments about the impact of sludge processing on chemical concentrations, more information on the type of treatment (both of the wastewater and the sludge) and the sludge residence time as well as the nature of significant non-domestic inputs is needed. Detection methods and limits of detection need to be reported. Where multiple samples are analyzed, individual data points as well as median and means should be reported since averaging values among several sludges may obscure information relating to the differences due to inputs or treatment.

This paper demonstrates that there are groups of chemicals for which there are relatively abundant sludge concentration data (such as PCBs, pesticides and PAHs), while there are others for which few data have been collected (such as nitrosamines). Certain classes of chemicals also are shown to have high percentage of reported concentrations that exceed SSLs, suggesting that analysis of additional chemicals in those classes may be warranted. Few data exist on the fate of sludge-borne chemicals in field soils and such research is critical to assessing the risks posed by sludge application.

Evaluating the risks posed by individual chemicals, let alone mixtures requires multiple assumptions that can lead to unacceptably high levels of uncertainty. Current limitations in our knowledge base regarding the amount and type of chemicals in sludges exacerbate this problem, as does the limited availability of fate and toxicity data, for both human and non-human receptors. As sludge application occurs on farms, forests, and mines, as well as residential and recreational land, humans, wildlife and soil organisms may all be exposed to the organic contaminants present in sludges. Filling the gaps in knowledge regarding the concentration, fate and toxicity of sludge-borne contaminants is critical if the risks associated with land application are to be adequately characterized.

#### Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at [doi:10.1016/j.scitotenv.2006.04.002](https://doi.org/10.1016/j.scitotenv.2006.04.002).

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