



Review

Review of 'emerging' organic contaminants in biosolids and assessment of international research priorities for the agricultural use of biosolids

Bradley O. Clarke, Stephen R. Smith*

Department of Civil and Environmental Engineering, South Kensington Campus, Imperial College London, London, SW7 2AZ, United Kingdom

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Polydimethylsiloxanes

Perfluorochemicals

Phthalate acid esters

Quaternary ammonium compounds

Steroids

Hormones

Synthetics musks

Triclosan

Triclocarban

ABSTRACT

A broad spectrum of organic chemicals is essential to modern society. Once discharged from industrial, domestic and urban sources into the urban wastewater collection system they may transfer to the residual solids during wastewater treatment and assessment of their significance and implications for beneficial recycling of the treated sewage sludge biosolids is required. Research on organic contaminants (OCs) in biosolids has been undertaken for over thirty years and the increasing body of evidence demonstrates that the majority of compounds studied do not place human health at risk when biosolids are recycled to farmland. However, there are 143,000 chemicals registered in the European Union for industrial use and all could be potentially found in biosolids. Therefore, a literature review of 'emerging' OCs in biosolids has been conducted for a selection of chemicals of potential concern for land application based upon human toxicity, evidence of adverse effects on the environment and endocrine disruption.

To identify monitoring and research priorities the selected chemicals were ranked using an assessment matrix approach. Compounds were evaluated based upon environmental persistence, human toxicity, evidence of bioaccumulation in humans and the environment, evidence of ecotoxicity and the number and quality of studies focussed on the contaminant internationally. The identified chemicals of concern were ranked in decreasing order of priority: perfluorinated chemicals (PFOS, PFOA); polychlorinated alkanes (PCAs), polychlorinated naphthalenes (PCNs); organotins (OTs), polybrominated diphenyl ethers (PBDEs), triclosan (TCS), triclocarban (TCC); benzothiazoles; antibiotics and pharmaceuticals; synthetic musks; bisphenol A, quaternary ammonium compounds (QACs), steroids; phthalate acid esters (PAEs) and polydimethylsiloxanes (PDMSS).

A number of issues were identified and recommendations for the prioritisation of further research and monitoring of 'emerging' OCs for the agricultural use of biosolids are provided. In particular, a number of 'emerging' OCs (PFOS, PFOA and PCAs) were identified for priority attention that are environmentally persistent and potentially toxic with unique chemical properties, or are present in large concentrations in sludge, that make it theoretically possible for them to enter human and ecological food-chains from biosolids-amended soil.

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Abbreviations: <dl, Less than detection limit; ADBI, Synthetic polycyclic musk – Celestolide™; AHMI, Synthetic polycyclic musk – Phantolide™; AHTN, Synthetic polycyclic musk – Tonalide™; ATII, Synthetic polycyclic musks – Traseolide™; BFR, Brominated flame retardant; CAS, Chemical abstract service; CP, Chlorinated paraffin; DBT, Dibutyltin; DEHP, Di2-(ethylhexyl) phthalate; DPPI, Synthetic polycyclic musk – Cashmeran™; dw, Dry weight; GC, Gas chromatography; HBCB, Hexabromocyclododecane; HHCb, Synthetic polycyclic musk – Galaxolide™; HRGC, High resolution gas chromatography; IARC, International Agency for Research on Cancer; IPCS, International Programme on Chemical Safety; K_{ow} , Octanol–water partition coefficient; LCCP, Long-chain chlorinated paraffin; IPCA, Long-chain chlorinated alkane; MA, Musk ambrette; Max, Maximum concentration; MBT_{tin}, Monobutyltin; MBT_{thiazole}, Mercaptobenzothiazole; MCCP, Medium-chain chlorinated paraffin; Min, Minimum concentration; MK, Musk ketone; MM, Musk moskene; mPCA, Medium-chain chlorinated paraffin; MS, Mass spectrometer; MT, Musk tibetene; MX, Musk xylene; n, Number of samples; OBT, 2-hydroxybenzothiazole; OC, Organic contaminant; OT, Organotin; PAE, Phthalate acid ester; PBB, Polybrominated biphenyl; PBDE, Polybrominated diphenyl ether; PCA, Polychlorinated alkane; PCB, Polychlorinated biphenyl; PCDD, Polychlorinated dibenzo-p-dioxin; PCDF, Polychlorinated dibenzofuran; PCN, Polychlorinated naphthalene; PDMS, Polydimethylsiloxane; PEC, Predicted environmental concentration; PFAC, Perfluoroalkyl carboxylate; PFAS, Perfluoroalkyl sulfonate; PFDA, Perfluorodecanoic acid; PFDoDA, Perfluorododecanoic acid; PFHxS, Perfluorohexane sulfonate; PFNA, Perfluorononanoic acid; PFOA, Perfluorooctanoic acid; PFOS, Perfluorooctane sulfonate; PFOSA, Perfluorooctane sulfonamide; PFUnDA, Perfluoroundecanoic acid; PHS, Priority hazardous substance; PNEC, Predicted no-effect environmental concentration; POP, Persistent organic pollutant; PVC, Polyvinyl chloride; QAC, Quaternary ammonium compound; SCCP, Short-chain chlorinated paraffin; sPCA, Short-chain chlorinated paraffin; TBT, Tributyltin; TCC, Triclocarban; TCDD, Tetrachloro dibenzo-p-dioxin; TCS, Triclosan; TEF, Toxicity equivalency factors; TEQ, Toxicity equivalency – TCDD equivalence; TPhT, Triphenyltin; UNEP, United Nations Environment Program; US EPA, United States Environmental Protection Agency; Vp, Vapour pressure; WFD, European Water Framework Directive; WHO, World Health Organization; WWTP, Wastewater treatment plant.

* Corresponding author. Tel.: +44 20 75946051; fax: +44 20 75941511.

E-mail address: s.r.smith@imperial.ac.uk (S.R. Smith).

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1. Introduction

The land application of biosolids (treated sewage sludge) is the option favoured internationally for sludge management as it contributes positively to recycling nutrients, soil properties and fertility (CEC, 1986; CEC, 1991; US EPA, 1993; European Commission, 2010). Land application of biosolids is also likely to become an increasingly essential aspect of sustainable nutrient management as phosphorus resources become depleted (Steen, 1998).

Modern society depends on a large range of organic chemicals and these may ultimately enter urban wastewater. Degradation and attenuation during wastewater and sludge treatment remove significant amounts of organic contaminants (OCs). However, many OCs have lipophilic properties and hence transfer to sewage sludge and may be present in residual concentrations ranging from $< \text{ng kg}^{-1}$ to % values in the dry solids depending on the initial amounts present, their lipophilicity and the extent of destruction during wastewater and sludge treatment.

Over the past thirty years a significant volume of research has been completed on this topic. Particular attention has been given to selected priority groups of persistent organic pollutants (POPs) such as chlorinated dioxins/furans (PCDD/Fs), polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) (Wild et al., 1991; Alcock et al., 1996; Stevens et al., 2001). The body of published work on OCs in sludge covers: organochlorine pesticides (McIntyre and Lester, 1984; Clarke et al., 2010), PCBs (Alcock and Jones, 1993; Wilson et al., 1997), dioxin-like compounds (Sewart et al., 1995; Stevens et al., 2001; Clarke et al., 2008a); and more recently: chlorinated naphthalenes (PCNs), PAHs, polychlorinated alkanes (PCAs), synthetic musks (Stevens et al., 2003), oestrogens (Gomes et al., 2009), organotin compounds (Voulvoulis et al., 2004; Voulvoulis and Lester, 2006) and nonyl phenol (NP) (Sjöström et al., 2008). The concentrations of 'traditional' POPs (eg PAHs, PCBs, PCDD/Fs) in sludge have declined substantially due to effective source control (Wild et al., 1990; Clarke et al., 2008a, 2010).

The 'traditional' POPs were considered as a risk to human health and the environment from biosolids land application due to their persistence, potential to bioaccumulate up foodwebs and toxicity (Chaney et al., 1996). However, hydrophobic non-ionic OCs are tightly sorbed to sludge and soil organic matter, thus reducing their bioavailability to

microorganisms and for plant uptake, but this characteristic also increases their persistence in soil (Alexander, 2000). Risk assessments also found that entry into the human foodchain resulting from biosolids land application was negligible because plant uptake is minimal (Briggs et al., 1982; O'Connor et al., 1990; Hundal et al., 2008), the strong sorption to the soil matrix prevents groundwater contamination (Wilson et al., 1996), while restrictions on surface application to pasture for grazing and the promotion of biosolids incorporation reduces the likelihood of OC accumulation by grazing animals. The assimilation of the available international research through risk assessment in the majority of studies concludes that the OCs examined do not pose a risk to human health when land applying biosolids (Dean and Suess, 1985; Jackson and Eduljee, 1994; Wild et al., 1994; Chaney et al., 1996; Schowanek et al., 2004; Eriksen et al., 2009). Nevertheless, it is recognised that continued vigilance is required to monitor and determine the significance and implications of 'emerging' OCs for the land application of biosolids.

The aim of this review is to identify research and monitoring priorities for 'emerging' OCs in biosolids, and particularly identify chemicals that may be potentially significant for agricultural use of biosolids, requiring further assessment and investigation. A list of compounds was selected and the available published information for these chemicals was critically examined and reviewed. The OCs were selected for evaluation on the basis that they exhibited one or more of the following properties: environmental persistence, bioaccumulation, toxicity or endocrine disruption. Research and monitoring priorities for the 'emerging' OCs considered have been evaluated using a matrix assessment approach to rank the chemicals of concern. The assessment of OCs was based on the following criteria:

- persistence of the OC in soil;
- potential risks to the human foodchain from biosolids land application;
- evidence of bioaccumulation in ecological receptors;
- evidence of ecotoxicity;
- the extent, quality and consistency of the research conducted.

The OCs selected for review were:

- antibiotics and pharmaceuticals
- benzothiazoles

- bisphenol A
- organotins (OTs)
- polybrominated diphenyl ethers (PBDEs)
- polychlorinated alkanes (PCAs)
- polychlorinated naphthalenes (PCNs)
- polydimethylsiloxanes (PDMs)
- perfluorochemicals (PFCs)
- phthalate acid esters (PAEs)
- quaternary ammonium compounds (QACs)
- steroids
- synthetic musks
- triclosan (TCS) and triclocarban (TCC).

Chemical properties and structure of the selected compounds or class of compound are described in Table 1.

2. Review of emerging organic contaminants

2.1. Antibiotics and pharmaceuticals

The main transfer pathway for antibiotics and pharmaceuticals used in human medicine to enter the environment is via wastewater treatment plants (WWTPs). Antibiotics have been detected in sewage effluents (Golet et al., 2002), ground and river water (Hirsch et al., 1999; Golet et al., 2001; Kolpin et al., 2002), sewage sludge (Gobel et al., 2005), as well as soil and manure (due to veterinary use) (Golet et al., 2003). Studies on the fate of antibiotics and pharmaceuticals in wastewater, surface water and biosolids are primarily motivated by the question of whether antibiotics in the environment may contribute to the spread of antibiotic resistant bacterial pathogens (McArdell et al., 2003). However, concern regarding antibiotics and pharmaceuticals is also driven by understanding the ecological consequences of widespread environmental contamination and the possible entry of pharmaceuticals into the human foodchain (Daughton and Ternes, 1999; Thiele-Bruhn, 2003).

The chemical properties of antibiotics and pharmaceuticals can vary widely, however many contain a non-polar core with a polar functional moiety (Thiele-Bruhn, 2003). The varying chemical properties will influence the behaviour of the chemical through wastewater treatment as well as the mobility, persistence and bioavailability in the soil matrix. Antibiotics can be categorised into the following groups:

- fluoroquinolone (FQ)
- sulfonamide (SA)
- penicillin (PE)
- cephalosporin (CE)
- nitroimidazole (NI)
- tetracycline (TC)
- macrolide (MA)

In a study of antibiotics in Swedish WWTP, the most commonly detected antibiotics were: norfloxacin, ofloxacin, ciprofloxacin, trimethoprim, sulfamethoxazole and doxycycline. Norfloxacin, ofloxacin, ciprofloxacin and doxycycline were the main antibiotics detected in sludge at the low mg kg⁻¹ dry weight (dw) range (Lindberg et al., 2005). A mass balance study indicated that these chemicals passed unchanged through the WWTP and concentrations could be predicted based upon consumption and use data (Lindberg et al., 2005). Similar concentrations (low mg kg⁻¹ dw) were reported in a Swiss study that detected ciprofloxacin and norfloxacin in sewage sludge (Golet et al., 2002). These compounds were also measured in biosolids-amended soil 21 months after application in the µg kg⁻¹ dw range (Golet et al., 2003). The longer environmental persistence of antibiotics in biosolids-amended soil is apparently in contrast to aquatic environments, where degradation occurs in a matter of days (Andreozzi et al., 2003), and could be explained by increased sorption

to the sludge/soil matrix reducing bioavailability for microbial biodegradation (Alexander, 2000; Drillia et al., 2005; Williams and Adamsen, 2006). Greenhouse plant uptake experiments have demonstrated that certain compounds, such as carbamazepine and sulfamethazine, can be translocated from the soil matrix and into the aerial plant components (Dolliver et al., 2007; Winker et al., 2010). However, the risk of human exposure via this pathway is considered low and unlikely to exceed acceptable daily intakes (Thiele-Bruhn, 2003; Boxall et al., 2006).

A recent Norwegian risk assessment (Eriksen et al., 2009) screened pharmaceutical compounds in sludge against consumption, estimated mass entering WWTP, human metabolism, biodegradation and behaviour in WWTP. Of the 1400 pharmaceutical compounds currently prescribed in Norway and screened in the risk assessment, only 14 were identified for further detailed investigation. The predicted environmental concentration (PEC) and the predicted no-effect environmental concentration (PNEC) of these compounds are presented in Table 2. The concentrations of drug substances in agricultural soils amended with biosolids were estimated to be <1 mg kg⁻¹ dw and were significantly below the estimated soil PNEC values. The overall conclusion was that drug substances in sewage sludge constitute a low risk to the soil compartment (Eriksen et al., 2009).

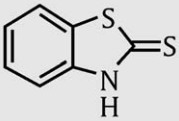
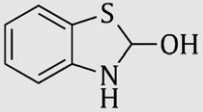
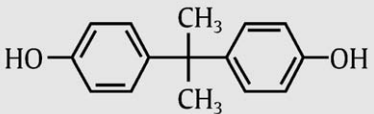
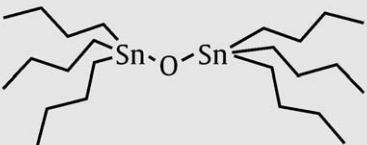
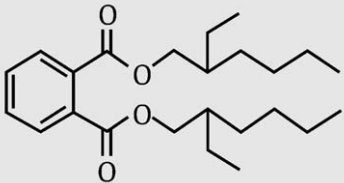
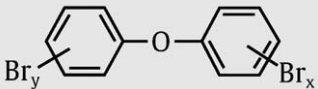
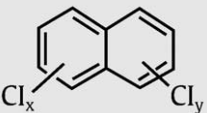
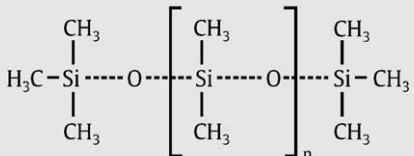
A survey of US biosolids found that, for 72 pharmaceuticals, two (viz. ciprofloxacin, diphenhydramine) were found in all samples (n = 84) and eight were found in at least 80 of the biosolids samples analysed. However, 15 pharmaceuticals were not found in any sample and 29 were present in fewer than three samples (US EPA, 2009). Many of the compounds identified as priorities in the Norwegian biosolids risk assessment were not included for analysis in the US biosolids survey. However, maximum concentrations of tetracycline (range: 0.04–5.3 and mean: 1.3 mg kg⁻¹ dw) and ciprofloxacin (range: 0.08–41.0 and mean: 10.5 mg kg⁻¹ dw) measured in the US survey (Table 3) were 12 and 24 times larger, respectively, than the amounts estimated in sludge for risk assessment by Eriksen et al. (2009). Nevertheless, applying these factors to the Norwegian soil_{PEC} for the drug compounds (Eriksen et al., 2009) still results in a value that is well below the estimated soil_{PNEC}.

The potential implications for human health of increased antibiotic resistance in soil bacteria are clearly a matter of concern (Nwosu, 2001). A large variety of soil-borne saprophytes including actinomycetes, fungi and bacteria are capable of synthesizing antibiotics and resistance develops in soil microbial communities to overcome the effects of natural microbial antibiotics released into the soil. However, antibiotic resistance levels may rise due to inputs of anthropogenic sources (eg antibiotics in wastes from intensively reared livestock for instance). These are apparently short-lived and return to the background level once the selection pressure has been removed, through biodegradation of the antibiotic, as there is no competitive advantage in maintaining this characteristic, which is subsequently lost from the soil microbial community (Sengeløv et al., 2003; Rysz and Alvarez, 2004). The Panel on Contaminants in the Norwegian Scientific Committee for Food Safety (Eriksen et al., 2009) noted the fluoroquinolone antibiotic drug, ciprofloxacin, could potentially lead to the development of antibacterial resistance, due to its persistence and limited mobility in soil. However, the risk assessment of biosolids application to agricultural land indicated that it was unlikely that antibacterial resistance would be promoted in treated effluent from WWTP, sewage sludge or amended soil (Eriksen et al., 2009).

2.2. Benzothiazoles

The German Government has proposed limit values in biosolids for two rubber vulcanising agents: 2-mercaptobenzothiazole (MBT_{thiazole}) and 2-hydroxybenzothiazole (OBT) (BMU, 2007). These chemicals, referred to as accelerators, are used for the polymerisation of sulphur with rubber (vulcanisation). They have been detected in wastewater in

Table 1
Chemical structure and properties of 'emerging' organic contaminants.

Compound	Chemical structure	Chemical properties
Benzothiazoles 2-Mercaptobenzothiazole (MBT _{thiazole})		CAS No: 149-30-4 Formula: C ₇ H ₅ NS ₂ Log K _{OW} : 2.42 Vp: <0.0014 Pa (25 °C) Half-life (soil): unknown (US EPA, 1984)
2-Hydroxybenzothiazole (OBT)		CAS No: 934-34-9 Formula: C ₇ H ₅ NOS Log K _{OW} : 1.76 Vp: 466 (25 °C) Half life (soil): unknown (Reddy and Quinn, 1997)
Bisphenol A		CAS No: 56-35-9 Formula: C ₁₅ H ₁₆ O ₂ Log K _{OW} : 3.40 Vp: 1.15 × 10 ⁻⁵ –0.005 Pa (20 °C) Half life (soil): days Staples et al. (1998); Oehlmann et al. (2008)
Organotin compounds Tributyltin (TBT) oxide		CAS No: 56-35-9 Formula: C ₂₄ H ₅₄ O ₂ Sn ₂ Log K _{OW} : 3.19–3.84 Vp: 1 × 10 ⁻³ Pa (20 °C) Half life (soil): 70 days IPCS (1990)
Phthalate acid esters Di (2-ethylhexyl) phthalate		CAS No: 84-74-2 Formula: C ₁₆ H ₂₂ O ₄ Log K _{OW} : 4.31–4.79 Vp: 0.01 Pa (25 °C) Half life (soil): 23–100 days IPCS (1992)
Polychlorinated diphenyl ethers (PBDEs)		CAS No: varied Formula: C ₁₂ H _(10-x-y) Br _x yO Log K _{OW} : 4.28–9.9 Vp: 3.85–13.3 Pa (20–25 °C) Half life (soil): 4–20 years IPCS (1994); Sellstrom et al. (2005); Eljarrat et al. (2008)
Polychlorinated alkanes (PCAs)	Numerous isomers with varying degrees of chlorine substitution. Short-chain PCAs–C _{10–13} Medium-chain PCAs–C _{14–17} Long-chain PCAs–C _{18–30}	CAS No: varied Formula: C _x H _{(2x+2)-y} Cl _y Log K _{OW} : varied Vp: varied Half life (soil): unknown
Polychlorinated naphthalenes (PCNs)		CAS No: varied Formula: C ₁₂ H _(8-x-y) Cl _x y Log K _{OW} : 6.42–10.11 Vp: 3 × 10 ⁻¹¹ –4.2 × 10 ⁻⁹ Pa Half life (soil): unknown/persistent IPCS (2001)
Polydimethylsiloxanes (PDMSs)		CAS No: varied Formula: Me ₃ SiO(SiMe ₂ O) _n SiMe ₃ ; n varies between 100 and >10,000 Log K _{OW} : non-polar Vp: variable Half life (soil): 2–28 days Griessbach and Lehmann (1999)

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Table 1 (continued)

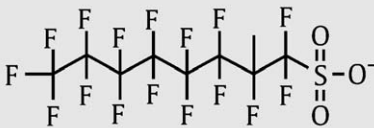
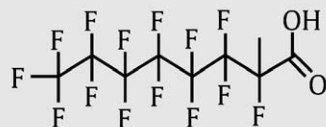
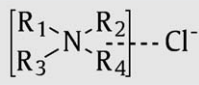
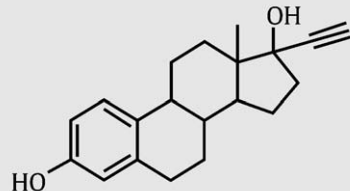
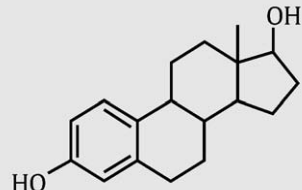
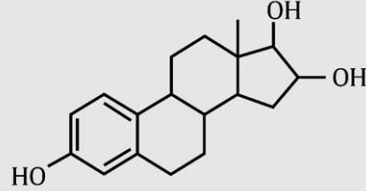
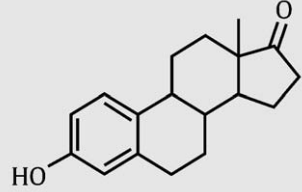
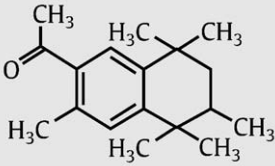
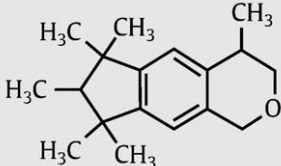
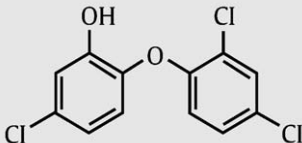
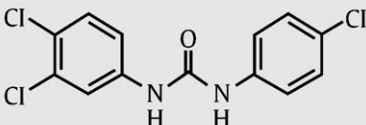
Compound	Chemical structure	Chemical properties
Perfluorochemicals Perfluorooctane sulphonate (PFOS)		CAS No: no specific number Formula: C ₈ F ₁₇ SO ₃ ⁻ Log K _{OW} : cannot be determined Vp: 3.31 × 10 ⁻⁴ Pa (20 °C) Half life (soil): unknown/persistent OECD (2002)
Perfluorooctanoic acid (PFOA)		CAS No: 335-67-1 Formula: C ₇ F ₁₅ COOH Log K _{OW} : unknown Vp: unknown Half life (soil): unknown/persistent
Quaternary ammonium compounds (QACs)	 <p>R1-4 represent alkyl or aryl substituents</p>	Generalised structure of QACs. High variation in substitution and therefore, chemical properties IPCS (1999)
Steroids 17 α-ethinyloestradiol		CAS No: 57-63-6 Formula: C ₂₀ H ₂₄ O ₂ Log K _{OW} : 3.67 Vp: 6.0 × 10 ⁻⁹ Pa Half life (soil): a few days Ternes et al. (2002); Lai et al. (2002)
17 β-Oestradiol		CAS No: 50-28-2 Formula: C ₁₈ H ₂₄ O ₂ Log K _{OW} : 4.01 Vp: 3.0 × 10 ⁻⁸ Pa Half life (soil): a few days Ternes et al. (2002); Lai et al. (2002)
Oestriol		CAS No: 5864-38-0 Formula: C ₁₈ H ₂₄ O ₃ Log K _{OW} : 2.81 Vp: 9.0 × 10 ⁻¹³ Pa Half life (soil): a few days Ternes et al. (2002); Lai et al. (2002)
Oestrone		CAS No: 53-16-7 Formula: C ₁₈ H ₂₂ O ₂ Log K _{OW} : 3.13 Vp: 3.0 × 10 ⁻⁸ Pa Half life (soil): a few days Ternes et al. (2002); Lai et al. (2002)

Table 1 (continued)

Compound	Chemical structure	Chemical properties
Synthetic musks AHTN (Tonalide™)		CAS No: 1506-02-1 Formula: C ₁₈ H ₂₆ O Log K _{ow} : 5.7 Vp: 0.0682 Pa Half life (soil): 180 days Balk and Ford (1999a)
HHCB (Galaxolide™)		CAS No: 1222-05-5 Formula: C ₁₈ H ₂₆ O Log K _{ow} : 5.9 Vp: 0.0727 Pa Half life (soil): 180 days Balk and Ford (1999a)
Triclosan (TCS)		CAS No: 3380-34-5 Formula: C ₁₂ H ₇ Cl ₃ O ₂ Log K _{ow} : 4.8 Vp: 0.00069 Pa (25 °C) Half life (soil): 266 days NICNAS (2008); Topp et al. (2008)
Triclocarban (TCC)		CAS No: 101-20-2 Formula: C ₁₃ H ₉ Cl ₃ N ₂ O Log K _{ow} : 3.5–4.2 (22.6 °C) Vp: <100 Pa Half life (soil): unknown EC (2005); Snyder et al. (2010)

the low ng L⁻¹ range (Kloepfer et al., 2004). Degradation rates are reported in days and there are contradictory studies claiming that the dominant removal mechanisms involved are biological (de Wever and Verachert, 1997) or chemical (Gaja and Knapp, 1998). These compounds have a degree of aquatic toxicity and have also been employed as fungicide, herbicide and anti-algal agents (de Wever and Verachert, 1997). Not only are there no studies reporting the concentrations of MBT_{thiazole} or OBT in sludge, but there also few studies reporting their environmental distribution (Spies et al., 1987). Only once empirical

measurements of MBT_{thiazole} and OBT in sewage sludge have been completed will it be possible to assess whether these chemicals pose a risk to human health and/or the environment when land applying biosolids.

2.3. Bisphenol A

Bisphenol A (2,2-Bis-(4-hydroxyphenyl)propane) is a plasticiser manufactured in high quantities and is used as a monomer for the production of polycarbonate and epoxy resins, unsaturated polyester-

Table 2

Risk assessment evaluation concentrations (mg kg⁻¹ dw) of selected pharmaceutical compounds in sludge-amended soil (Eriksen et al., 2009).

Therapeutical group	Drug substance	Predicted environmental concentration (PEC)		Predicted no-effect concentration (PNEC)
		Agricultural soil (60 t ha ⁻¹)	Park areas	
Alimentary tract and metabolism	Mesalazin	0.98	6.70	12
	Ranitidin	0.04	0.30	5277
Blood and blood forming organs Cardiovascular system	Dipyridamole	0.03	0.17	–
	Sotalol	0.02	0.15	4095
	Metoprolol	0.02	0.13	589
	Losartan	0.03	0.23	–
Antibacterial drugs	Atorvastatin	0.05	0.34	11
	Tetracycline	0.01	0.08	8.8
	Ciprofloxacin	0.04	0.29	26
Muscular–skeletal system	Carisoprodol	0.10	0.68	24368
Nervous system	Gabapentin	0.06	0.39	20460
	Levetiracetam	0.02	0.12	–
	Chlorprothixene	0.02	0.16	–
Respiratory organs	Fexofenadine	0.03	0.17	–

Table 3
Concentrations (mg kg⁻¹ dw) of 'emerging' organic contaminants in sewage sludge/biosolids.

Contaminant	Country	Year	n	Mean	Min	Max	Reference
<i>Antibiotics and pharmaceuticals</i>							
4-Epitetracycline	USA	2009	84	1.14	0.04	4.38	US EPA (2009)
Azithromycin	USA	2009	84	0.83	0.008	5.21	US EPA (2009)
Carbamazepine	USA	2009	84	0.14	0.009	6.03	US EPA (2009)
Cimetidine	USA	2009	84	1.33	0.004	8.33	US EPA (2009)
Ciprofloxacin	Germany	2002	2	2.35	2.27	2.42	Golet et al. (2002)
	Sweden	2005	10	2.5	0.5	4.8	Lindberg et al. (2005)
	USA	2009	84	10.5	0.075	40.8	US EPA (2009)
Diphenhydramine	USA	2009	84	0.871	0.037	5.73	US EPA (2009)
Doxycycline	Sweden	2005	10	1.4	<dl	1.5	Lindberg et al. (2005)
	USA	2009	84	0.877	0.034	5.09	US EPA (2009)
Erythromycin (Total)	USA	2009	84	0.036	0.002	0.18	US EPA (2009)
Fluoxetine	USA	2009	84	0.245	0.010	3.13	US EPA (2009)
Miconazole	USA	2009	84	1.239	0.007	9.21	US EPA (2009)
Norfloxacin	Germany	2002	2	2.25	2.13	2.37	Golet et al. (2002)
	Sweden	2005	10	1.51	0.1	4.2	Lindberg et al. (2005)
Ofloxacin	Sweden	2005	10	0.73	<0.1	2.0	Lindberg et al. (2005)
	USA	2009	84	8.573	0.025	58.10	US EPA (2009)
Tetracycline	USA	2009	84	1.278	0.038	5.27	US EPA (2009)
<i>Bisphenol A</i>							
	Germany	2002	38	*	0.004	1.363	Fromme et al. (2002)
	Germany	2002	18	*	~40	~325	Meesters and Schroder (2002)
	Australia	2007	4	0.089	0.004	0.158	Tan et al. (2007)
	Greece	2007	1	0.62	*	*	Gatidou et al. (2007)
	Greece	2008	1	0.03	*	*	Pothitou and Voutsas (2008)
	Greece	2008	27	0.53	<0.56	1.75	Stasinakis et al. (2008)
	China	2009	2	0.11	0.10	0.13	Nie et al. (2009)
	Overall		91	0.28	0.004	~325	
<i>Organotins</i>							
Monobutyltin (MBT _{tin})	Switzerland	1987	4	3.3	0.2	6	Mueller (1987)
	Switzerland	1991	3	0.78	0.10	0.97	Fent et al. (1991)
	Canada	1992	36	0.02	*	*	Chau et al. (1992)
	Switzerland	1996	25	0.5	*	*	Fent (1996b)
	France	2000	1	0.24	*	*	Bancon-Montigny et al. (2000)
	UK	2004	40	0.71	*	*	Voulvoulis et al. (2004)
	Overall		109	0.93	0.10	6	
Dibutyltin (DBT)	Switzerland	1987	4	5	0.7	7.5	Mueller (1987)
	Switzerland	1987	4	0.98	0.41	1.24	Fent et al. (1991)
	Canada	1991	3	0.04	*	*	Chau et al. (1992)
	Switzerland	1992	36	1.5	*	*	Fent (1996b)
	France	1996	25	0.08	*	*	Bancon-Montigny et al. (2000)
	UK	2004	40	0.06	*	*	Voulvoulis et al. (2004)
	Overall		112	1.28	0.41	7.5	
Tributyltin (TBT)	Switzerland	1987	4	3.5	0.3	6	Mueller (1987)
	Switzerland	1991	3	0.99	0.28	1.51	Fent et al. (1991)
	Canada	1992	36	0.1	*	*	Chau et al. (1992)
	Switzerland	1996	25	1.1	*	*	Fent (1996b)
	France	2000	1	0.05	*	*	Bancon-Montigny et al. (2000)
	Switzerland	2004	24	0.15	0.02	0.65	Plagellat et al. (2004)
	UK	2004	40	0.13	*	*	Voulvoulis et al. (2004)
	Overall		133	0.86	0.02	6	
Triphenyltin (TPhT)	Switzerland	1987	4	2.3	<0.02	9	Mueller (1987)
	Canada	1991	36	0.3	*	*	Chau et al. (1992)
	Switzerland	1992	25	0.5	*	*	Fent (1996b)
	France	1996	1	0.01	*	*	Bancon-Montigny et al. (2000)
	Switzerland	2000	24	0.02	<dl	0.28	Plagellat et al. (2004)
	Overall		90	0.63	<0.02	9	
<i>Phthalate acid esters (PAEs)</i>							
DEHP	Canada	1989	6	65	3	176	Webber and Lesage (1989)
	Germany	2002	15	~67	~28	~154	Fromme et al. (2002)
	Canada	2003	20	2.7	<0.02	11	Bright and Healey (2003)
	Finland	2003	13	95	28	122	Marttinen et al. (2003a)
	Spain	2005	134	67	2	3514	Abad et al. (2005)
	UK	2005	1	62	*	*	Gibson et al. (2005)
	UK	2005	*	30	*	*	Oliver et al. (2005)
	Denmark	2007	*	67	61	78	Roslev et al. (2007)
	Australia	2007	5	11.2	0.26	45.1	Tan et al. (2007)
	Australia	2008	14	17	2	44	Clarke et al. (2008b)
	France	2009	10	72.1	*	*	Dargnat et al. (2009)
	Spain	2009	4	159	13	345	Aparicio et al. (2009)
	USA	2009	84	53	<dl	310	US EPA (2009)
	Overall		306	58	<0.02	3514	

Table 3 (continued)

Contaminant	Country	Year	n	Mean	Min	Max	Reference	
<i>Polybrominated diphenyl ethers (PBDEs)</i>								
BDE47	Sweden	1992	2	0.015	0.015	0.015	Nylund et al. (1992)	
	Sweden	1999	3	0.065	0.036	0.080	Sellstrom et al. (1999)	
	USA	2001	11	0.568	0.359	0.754	Hale et al. (2001)	
	Netherlands	2003	3	0.020	0.010	0.040	de Boer et al. (2003)	
	Spain	2004	6	0.037	0.002	0.050	Fabrellas et al. (2004)	
	Germany	2004	8	0.047	0.025	0.088	Hamm (2004)	
	USA	2004	1	0.757	*	*	(North, 2004)	
	Sweden	2006	50	0.049	0.007	0.100	Law et al. (2006)	
	Germany	2007	19	0.052	0.020	0.115	Knoth et al. (2007)	
	China	2007	31	0.005	0.0004	0.059	Wang et al. (2007)	
	Australia	2008	16	0.126	<0.001	0.410	Clarke et al. (2008c)	
	Kuwait	2008	21	0.002	0.0002	0.008	Gevao et al. (2008)	
	Antarctica	2008	2	0.776	0.132	1.420	Hale et al. (2008)	
	USA	2009	84	0.709	0.073	5.000	US EPA (2009)	
	USA	2010	15	0.161	0.128	0.238	Andrade et al. (2010)	
	Overall		272	0.226	0.0002	5.000		
	BDE99	Sweden	1992	2	0.019	0.019	0.019	Nylund et al. (1992)
		Sweden	1999	3	0.085	0.056	0.100	Sellstrom et al. (1999)
		USA	2001	11	0.661	0.391	1.157	Hale et al. (2001)
		Netherlands	2003	3	0.021	0.011	0.038	de Boer et al. (2003)
Spain		2004	6	0.037	0.023	0.064	Fabrellas et al. (2004)	
Germany		2004	8	0.070	0.037	0.127	Hamm (2004)	
USA		2004	1	0.940	*	*	North (2004)	
Sweden		2006	50	0.060	0.008	0.150	Law et al. (2006)	
Germany		2007	39	0.057	0.024	0.124	Knoth et al. (2007)	
China		2007	31	0.005	0.003	0.068	Wang et al. (2007)	
Australia		2008	14	0.141	0.0004	0.400	Clarke et al. (2008c)	
Kuwait		2008	21	0.005	0.0004	0.015	Gevao et al. (2008)	
Antarctica		2008	2	0.735	0.200	1.270	Hale et al. (2008)	
USA		2009	84	0.716	0.064	4.000	US EPA (2009)	
USA		2010	15	0.169	0.128	0.245	Andrade et al. (2010)	
Overall			290	0.248	0.0004	4.000		
BDE209		Sweden	1999	3	0.220	0.170	0.270	Sellstrom et al. (1999)
		USA	2001	11	1.370	0.085	4.890	Hale et al. (2001)
		Netherlands	2003	3	0.096	0.009	0.190	de Boer et al. (2003)
		Spain	2004	6	5.968	0.756	18.632	Fabrellas et al. (2004)
	Germany	2004	8	0.326	0.100	0.639	Hamm (2004)	
	USA	2004	1	1.183	*	*	North (2004)	
	Sweden	2006	50	0.120	0.006	1.000	Law et al. (2006)	
	Germany	2007	39	0.442	0.113	1.339	Knoth et al. (2007)	
	China	2007	31	0.069	<0.001	1.109	Wang et al. (2007)	
	Australia	2008	14	0.705	0.003	3.780	Clarke et al. (2008c)	
	Kuwait	2008	21	0.182	0.005	1.596	Gevao et al. (2008)	
	Antarctica	2008	2	0.770	0.219	1.320	Hale et al. (2008)	
	USA	2009	84	2.180	0.150	17.000	US EPA (2009)	
	USA	2010	15	0.920	0.792	1.220	Andrade et al. (2010)	
	Overall		288	1.039	0.003	18.632		
	ΣPBDEs	USA	2004	1	3.381	*	*	North (2004)
		Germany	2007	39	0.555	0.142	2.491	Knoth et al. (2007)
		China	2007	31	0.094	0.005	1.115	Wang et al. (2007)
		Australia	2008	14	1.137	0.005	4.230	Clarke et al. (2008c)
		Kuwait	2008	21	0.191	0.006	1.600	Gevao et al. (2008)
Antarctica		2008	2	2.664	0.637	4.690	Hale et al. (2008)	
USA		2010	15	1.496	1.330	1.820	Andrade et al. (2010)	
Overall			123	1.360	0.005	4.690		
<i>Polychlorinated alkanes (PCAs)</i>								
sPCA	Germany	1995	2	56	47	65	Rieger and Ballschmiter (1995)	
	UK	2003	14	42	7	200	Stevens et al. (2003)	
	Overall		16	49	7	200		
mPCA	UK	2001	9	19.6	1.8	93	Nicholls et al. (2001)	
	UK	2003	14	1800	30	9700	Stevens et al. (2003)	
	Overall		23	910	1.8	9700		
<i>Polychlorinated naphthalenes (PCNs)</i>								
	Sweden	1992	2	0.005	0.003	0.006	Nylund et al. (1992)	
	UK	2003	14	0.083	0.050	0.190	Stevens et al. (2003)	
	China	2008	8	*	0.001	0.028	Guo et al. (2008)	
	Overall		24	0.044	0.001	0.190		
<i>Polydimethylsiloxanes (PDMSs)</i>								
	Japan	1984	1	144	*	*	Watanabe et al. (1984)	
	USA	1997	12	1120	122	5155	Fendinger et al. (1997)	
	Overall		13	632	122	5155		

(continued on next page)

Table 3 (continued)

Contaminant	Country	Year	n	Mean	Min	Max	Reference
<i>Perfluorochemicals</i>							
PFOS	USA	2001	12	0.58	0.06	3.12	3M Environmental Laboratory (2001)
	USA	2006	*	0.100	0.081	0.160	Schultz et al. (2006)
	USA	2006	10	0.031	<0.010	0.065	Sinclair and Kannan (2006)
	USA	2007	8	0.073	0.008	0.110	Loganathan et al. (2007)
	Denmark	2008	7	*	0.005	0.074	Bossi et al. (2008)
	Overall		37	0.196	0.005	3.12	
	PFOA	USA	2001	5	0.049	0.002	0.244
	USA	2006	*	<0.003	*	*	Schultz et al. (2006)
	USA	2006	10	0.107	0.018	0.241	Sinclair and Kannan (2006)
	USA	2007	8	0.068	0.0083	0.219	Loganathan et al. (2007)
	Denmark	2008	7	*	0.001	0.020	Bossi et al. (2008)
	Overall		30	0.075	0.001	0.244	
<i>Quaternary ammonium compounds (QACs)</i>							
DTDMAC	Switzerland	1991	5	3670	2570	5870	Fernandez et al. (1996)
	Switzerland	1992	5	960	730	1510	Fernandez et al. (1996)
	Switzerland	1993	5	470	300	570	Fernandez et al. (1996)
	Switzerland	1994	5	210	150	300	Fernandez et al. (1996)
Total QACs	Austria	2007	6	*	22	103	Martínez-Carballo et al. (2007)
	Overall		26	1328	22	5870	
<i>Steroids</i>							
Beta stigmastanol	USA	2009	84	168	3.44	1330	US EPA (2009)
Campesterol	USA	2009	84	101	2.84	524	US EPA (2009)
Cholestanol	USA	2009	84	680	3.86	4590	US EPA (2009)
Cholesterol	USA	2009	84	1129	2.34	5390	US EPA (2009)
Coprostanol	USA	2009	84	4367	7.72	43700	US EPA (2009)
Epicoprostanol	USA	2009	84	1703	0.87	6030	US EPA (2009)
Stigmasterol	USA	2009	84	321	0.46	569	US EPA (2009)
17 α -Ethinylloestradiol	Germany	2002	4	0.005	<0.004	0.017	Ternes et al. (2002)
	China	2009	2	*	<dl	<dl	Nie et al. (2009)
	USA	2009	84	*	<dl	0.355	US EPA (2009)
17 β -Oestradiol	Germany	2002	4	0.020	0.005	0.049	Ternes et al. (2002)
	China	2009	2	*	<dl	<dl	Nie et al. (2009)
	USA	2009	84	*	<dl	0.355	US EPA (2009)
Oestriol	China	2009	2	0.010	0.010	0.011	Nie et al. (2009)
	USA	2009	84	*	<dl	0.232	US EPA (2009)
Oestrone	Germany	2002	4	0.027	<0.002	0.037	Ternes et al. (2002)
	China	2009	2	0.016	0.011	0.022	Nie et al. (2009)
	USA	2009	84	*	<dl	0.965	US EPA (2009)
<i>Synthetic musks</i>							
AHTN (tonalid)	Switzerland	2000	12	1.54	0.74	4.16	Herren and Berset (2000)
	Germany	2002	4	3.56	2.52	5.07	Heberer (2002)
	Spain	2003	1	0.052			Llompart et al. (2003)
	UK	2003	14	4.7	0.12	16	Stevens et al. (2003)
	Switzerland	2004	16	7.3	2.5	11.2	Kupper et al. (2004)
	China	2005	3	2.56	0.72	6.20	Zeng et al. (2005)
	Hong Kong	2008	30	5.85	0.475	13.9	Shek et al. (2008)
	Overall		80	3.65	0.12	16	
	HHCB (galaxolide)	Switzerland	2000	12	4.85	2.29	12.16
Germany		2002	4	8.26	6.03	11.45	Heberer (2002)
Spain		2003	1	0.162			Llompart et al. (2003)
UK		2003	14	27	1.9	81	Stevens et al. (2003)
Switzerland		2004	16	20.3	7.4	36.0	Kupper et al. (2004)
China		2005	3	10.76	5.42	21.21	Zeng et al. (2005)
Hong Kong		2008	30	27.1	3.58	78.6	Shek et al. (2008)
Overall			80	14.06	1.9	81	
<i>Triclosan</i>							
Triclosan	USA	2002	10	4.55	0.53	15.6	McAvoy et al. (2002)
	Germany	2003	20	*	0.40	8.80	Bester (2003)
	Spain	2005	7	2.83	0.42	5.40	Morales et al. (2005)
	Canada	2007	12	3.21	0.62	11.55	Chu and Metcalfe (2007)
	Australia	2007	19	5.58	0.09	16.79	Ying and Kookana (2007)
	Greece	2007	1	1.84	*	*	Gatidou et al. (2007)
	Greece	2008	5	0.46	*	*	Pothitou and Voutsas (2008)
	Greece	2008	27	3.21	0.19	9.85	Stasinakis et al. (2008)
	USA	2009	4	1.87	0.09	7.06	Cha and Cupples (2009)
	USA	2009	84	16.10	0.33	133	US EPA (2009)
	Overall		189	4.41	0.09	133	
<i>Triclocarban</i>							
Triclocarban	USA	2006	3	51	*	*	Heidler et al. (2006)
	Canada	2007	12	4.17	2.17	5.97	Chu and Metcalfe (2007)

Table 3 (continued)

Contaminant	Country	Year	n	Mean	Min	Max	Reference
Triclocarban	USA	2007	5	19.3	7.5	25.9	Sapkota et al. (2007)
	USA	2009	4	7.19	4.89	9.28	Cha and Cupples (2009)
	USA	2009	84	39.43	0.19	441	US EPA (2009)
	Overall		108	24.2	0.19	441	

*No data; <dl – less than detection limit.

styrene resins and flame retardants (Staples et al., 1998). The final products are used as coatings on cans, as powder paints, as additives in thermal paper, in dental fillings and as antioxidants in plastics (Staples et al., 1998). Release into the environment is possible during manufacturing processes and by leaching from final products (Fromme et al., 2002). Bisphenol A has been shown to be weakly oestrogenic and to possess some anti-androgenic activity (Sohoni and Sumpter, 1998). However, the relative potency ranges are approximately 1×10^{-6} to 5×10^{-7} times less than 17β -oestradiol (Harris et al., 1997). Based on *in vitro* receptor-interaction studies, the oestrogenic activity was estimated to be 2×10^{-3} fold lower than for oestradiol. Bisphenol A has been implicated as an endocrine disrupting chemical and laboratory studies (using mice) indicate that development problems can be associated with environmentally relevant exposure (Newbold et al., 2009). The chemical structure of bisphenol A is given in Table 1.

Bisphenol A is regularly detected in surface waters primarily because it is continuously released into the environment (Heemken et al., 2001; Fromme et al., 2002; Oehlmann et al., 2008) and not because it is environmentally persistent (Dorn et al., 1987). While there is no direct evidence to confirm a detrimental causal link from exposure to bisphenol A (human or environmental) concern exists about this compound due to reports of adverse reproductive and developmental effects in wildlife that are possibly mediated *via* endocrine disruptive pathways (Fürhacker et al., 2000; Vandenberg et al., 2007; Oehlmann et al., 2008; Newbold et al., 2009).

Bisphenol A is widely used in households and industry, therefore, it can be expected to be present in raw sewage (Fürhacker et al., 2000). A German study identified that the paper industry was the major contributor of bisphenol A in wastewater (Fürhacker et al., 2000). WWTP mass balance studies have detected bisphenol A in raw water, sewage sludge and effluents (Meesters and Schroder, 2002). Significant reductions (up to 99%) during wastewater treatment have been reported (Fürhacker et al., 2000; Tan et al., 2007) and biodegradation is thought to be the principal removal mechanism (Pothitou and Voutsas, 2008).

There is a large variation in reported bisphenol A concentrations in sludges internationally, with values ranging from low $\mu\text{g kg}^{-1}$ dw (Fromme et al., 2002; Gatidou et al., 2007; Tan et al., 2007; Pothitou and Voutsas, 2008; Nie et al., 2009) to mid mg kg^{-1} dw (Meesters and Schroder, 2002). Bisphenol A is a bulk chemical manufactured in similar quantities to phthalates acid esters (PAEs), however, concentrations of bisphenol A are two orders of magnitude smaller in sludge than PAEs. This suggests that bisphenol A is considerably more degradable during wastewater and sludge treatment compared to phthalates. Similarly, bisphenol A is reported to rapidly dissipate in soil and has an estimated half-life of <3 days (Fent et al., 2003). A study of the toxic effects of bisphenol A to soil isopods has been reported, however, the concentrations used (10 – 300 mg kg^{-1} dw) were far higher than environmentally relevant values and the toxicity was associated with the delivery solvent rather than bisphenol A solely (Lemos et al., 2009). Few studies have examined the ecotoxicological effects of bisphenol A in soil and this requires further attention. Studies are also required to more accurately determine the concentrations of bisphenol A in sludge. Nevertheless, the concentrations of bisphenol A reported in sludge are not high and, coupled with its rapid biodegradation during wastewater treatment, this compound is unlikely to pose

an issue when land applying biosolids. Bisphenol A is under review for possible identification as a European Water Framework Directive (WFD) Priority Substance or Priority Hazardous Substance (PHS) to control emission sources (EPCEU, 2008). While evidence is indicating that bisphenol A is an endocrine disrupting chemical (Li et al., 2009; Newbold et al., 2009) that can be found in the human body (Lee et al., 2008), exposure primarily occurs in the domestic environment and via direct ingestion (Vandenberg et al., 2007). Transfer to humans from biosolids-amended soil, by contrast, is extremely unlikely since there is little evidence that bisphenol A is environmentally persistent or bioaccumulates via food-chain mediated pathways.

2.4. Organotin (OTs)

Organotin compounds (OTs) have been used since the 1960s for industrial and agricultural purposes viz., polyvinyl chloride (PVC) stabilisers, fungicides, bactericides, insecticides, industrial catalysts and wood preservatives (Hoch, 2001). This includes the use of monobutyltin (MBT_{tin}) and dibutyltin (DBT) as heat and light stabilisers in PVC processing, the use of tributyltin (TBT) in antifouling formulations and as a general-purpose wood preservative, as well as the use of triphenyltins (TPhT) in agriculture. The use of TBT as an antifouling agent for ship hulls and as a general wood preservative has ceased in the UK and internationally because of high toxicity in aquatic ecosystems (Alzieu, 1991; Fent, 1996a; Voulvoulis et al., 2004).

Mass balances for OTs in WWTP demonstrate that they are effectively removed during wastewater treatment and are concentrated in the sludge. Reported removal rates of MBT_{tin}, DBT and TBT are 95%, 84% and 86%, respectively (Voulvoulis et al., 2004). OT concentrations in wastewater show diurnal fluctuations, however there is no explanation for this apparent behaviour (Voulvoulis et al., 2004).

There is contradictory evidence from laboratory studies concerning the biodegradation of TBT and TPhT by the activated sludge process. For example, Stasinakis et al. (2005) showed these compounds were degraded by biological wastewater treatment, whereas Voulvoulis and Lester (2006) found minimal degradation. In a laboratory study, 50% of TBT and 20% of TPhT applied to soil in biosolids remained in the soil after 2 months (Marcic et al., 2006). The biodegradability of OTs in soil is also reported to decline with increasing substitution (Heroult et al., 2008).

The concentration of OT compounds in sewage sludge (Table 3) has been reported from Switzerland (Mueller, 1987; Fent and Mueller, 1991; Fent, 1996b), Canada (Chau et al., 1992), France (Bancon-Montigny et al., 2000) and the UK (Voulvoulis et al., 2004; Voulvoulis and Lester, 2006). There is no consistent trend in the concentration data, however, values for the main OTs rarely exceed 1 mg kg^{-1} dw in contemporary sludge samples, which may reflect the declining use of these compounds (Table 3).

2.5. Phthalate acid esters (PAEs)

Phthalate acid esters (PAEs) have been in use for over 50 years, mainly in the manufacture of resins and plastics such as PVC (Fromme et al., 2002). PAEs are also used in other non-PVC applications such as paints, rubber products, adhesives and some cosmetics. PAEs soften plastic resins without chemically binding with them and as a consequence leach from plastic products into the surrounding environment. The PAE content of plastic generally ranges from 20 to 40%, but in

some cases is as high as 55% (Fatoki and Vernon, 1990). The most common PAE is di(2-ethylhexyl)phthalate (DEHP) and approximately 95% of DEHP production is directed towards plasticizer use, particularly in PVC products such as tubing and medical device components. The chemical structure and properties of DEHP are given in Table 1. Recently, use of DEHP has declined to an extent, due to concerns that it may disrupt endocrine systems, and the use of other phthalate plasticizers has increased, in particular diisononyl phthalate (DINP) and diisodecyl phthalate (DIDP), which may, in fact be more environmentally persistent (Cadogan, 2002). There is contradictory information published regarding the impact and significance of phthalates for human health and the environment. A recent Chinese study found that significant contamination of the aerial components of vegetable plants occurred from a plastic based mulch with a DEHP concentration of 16.5% (Du et al., 2009). The results of this study appear contrary to currently accepted models of organic pollutants movement into plants (Briggs et al., 1982; Travis and Arms, 1988) and other experimental investigations that show there is minimal uptake of DEHP by crop plants (Schmitzer et al., 1988; Yin et al., 2003). Given that Du et al. (2009) report that the DEHP concentration was smaller in the roots of the plant it is possible that volatilisation or direct contamination might have played a significant role in the accumulation of DEHP in leaves and stems in this case. However, evidence from biosolids-amended systems indicates the minimal uptake of DEHP into plants (Aranda et al., 1989). In contrast to the apparent uptake of DEHP by vegetables from plastic in direct contact with the soil and crop, the sorption of DEHP onto the sludge matrix may thus control its bioavailability preventing movement and transfer into crops. Nevertheless, further research is warranted to clarify the bioavailability of DEHP to plants and whether contamination of aerial plant components can occur from contaminated soil. There is no reported evidence that DEHP has a negative impact upon the soil ecosystem (Kirchmann et al., 1991; Cartwright et al., 2000; Jensen et al., 2001).

PAEs are not environmentally persistent and are readily degraded in soils and sewage sludge under both aerobic and anaerobic conditions (Keyser et al., 1976; Walker et al., 1984; Shanker et al., 1985; Group, 1986; Staples et al., 1997). They are readily metabolised and do not accumulate in mammals even when fed artificially in diets high in PAEs (Giam et al., 1984). Organisms ingesting high doses of PAEs were quickly able to remove phthalate compounds from their body tissues once the chemical was excluded from the feed (Giam et al., 1984).

Only a relatively small number of international studies were found reporting PAE concentrations in sewage sludge. It may be expected that PAEs would increase in sewage sludge and the environment generally in proportion to their use within society, however, there is no evidence indicating that this is the case. The concentration of PAEs in sludge is reported from Canada (Webber and Lesage, 1989; Bright and Healey, 2003), Germany (Fromme et al., 2002), Finland (Marttinen et al., 2003a,b), Spain (Abad et al., 2005), the UK (Gibson et al., 2005; Oliver et al., 2005), Australia (Tan et al., 2007; Clarke et al., 2008b) and the USA (US EPA, 2009). A summary of DEHP concentration data is compiled in Table 3.

Abad et al. (2005) reported the concentration of DEHP in Spanish sewage sludge ($n = 139$) over a number of years and for different WWTPs ($n = 20$). The concentration of DEHP was highly variable ranging from 1.5 to 3513.8 mg kg⁻¹ dw. DEHP concentrations varied from 18.4 → 16.9 → 76.8 → 3513.8 → 157.4 → 11.4 mg kg⁻¹ dw over a two-year period at one WWTP demonstrating the high variability in the DEHP content of sewage sludge. Nevertheless, the DEHP concentration was typically between 10 and 50 mg kg⁻¹ dw (Abad et al., 2005) and was consistent with other survey data for DEHP in sewage sludge (Marttinen et al., 2004; Bago et al., 2005; Gibson et al., 2005; Oliver et al., 2005).

PAEs are well known to degrade during wastewater treatment and degradation under both aerobic and anaerobic conditions increases with water solubility (Shelton et al., 1984; Ziogou et al., 1989; Jianlong

et al., 2000; Fauser et al., 2003; Marttinen et al., 2004; Amir et al., 2005). PAEs characteristically have high log K_{ow} values >4 and therefore partition strongly to the sewage sludge during wastewater treatment (Table 1). They are rapidly degraded by aerobic sludge treatment processes, such as composting (Cheng et al., 2008; Pakou et al., 2009), but reported removals by anaerobic digestion vary from no observed anaerobic biodegradation up to 23–61% removal of DEHP in digested sludge (Fountoulakis et al., 2006). Consequently DEHP is typically present in sewage sludge from the low mg kg⁻¹ to <200 mg kg⁻¹ dw range (Table 3).

2.6. Polybrominated diphenyl ethers (PBDEs)

PBDEs are a class of brominated fire retardants (BFRs) that were used in plastics, textiles, electronic circuitry, and other materials. There are 209 PBDE congeners and they are numbered according to the IUPAC system for PCBs (Ballscmitter and Zell, 1980). PBDEs were sold in three commercial formulations; each named for the prominent homologue in the mixture viz., pentaBDE (BDE 47, 99, 100, 153, 154), octaBDE (BDE 183), and decaBDE (BDE 209) (Sjödin et al., 1998). Despite the commercial formulation names each contains many BDE congeners (BSEF, 2005). PBDEs have low vapour pressures ($4.69 \times 10^{-5} - 6.59 \times 10^{-6}$ Pa) and are highly lipophilic (log K_{ow} values of 5.9–10) (IPCS, 1994; Braekveit et al., 2003). Due to the potential threat to human health and the environment, PBDEs (specifically pentaBDE and octaBDE) were listed as United Nations Environment Programme (UNEP) Persistent Organic Pollutants (POPs) in 2008 (UNEP, 2001; UNEP, 2009).

Assessment of health risks associated with human exposure and accumulation of PBDEs is complicated and has not been adequately characterized. However, the potential risks associated with exposure to the most bioactive congeners (tri- to octa-BDE) include thyroid hormone disruption, neuro-developmental defects and cancer. Several studies have shown that PBDEs share similar general properties to organo-halogenated compounds as *in vivo* exposure of rodents resulted in reduction of serum total and free thyroid hormone (thyroxine T4) levels (Darnerud et al., 2001; McDonald, 2002). Altered thyroid hormone function, particularly during development, is profound and has been hypothesized to lead to disrupted brain development and permanent neurological damage (Legler and Brouwer, 2003).

PBDEs are routinely detected in sewage sludge in the low mg kg⁻¹ dw range (Table 3) and values have been reported from Sweden (Nylund et al., 1992; Sellstrom et al., 1999; Law et al., 2006), USA (de Carlo, 1979; Hale et al., 2001; North, 2004), Germany (Knoth et al., 2007), The Netherlands (de Boer et al., 2003), China (Wang et al., 2007), Australia (Clarke et al., 2008c), Kuwait (Gevao et al., 2008) and Antarctica (Hale et al., 2008).

Three congeners account for the majority of Σ PBDEs in sewage sludge and include: BDE47, 99 (pentaBDE) and BDE209 (decaBDE); the concentrations of these congeners are summarised in Table 3. PBDE congeners representative of the pentaBDE (BDE47, 99, 100, 153, 154) formulations are often present at similar concentrations regardless of the catchment type indicating domestic origin (Hale et al., 2001; Hale et al., 2008). The primary congener of the decaBDE formulation, BDE209, is consistently detected in the highest concentrations in sewage sludge and, in national surveys, its concentrations are also highly variable, suggesting important industrial inputs (Clarke et al., 2008c). Trace PBDE amounts (ng L⁻¹) have also been detected in treated effluent (de Boer et al., 2000; Hamm, 2004; North, 2004; Knoth et al., 2007) and recent studies have demonstrated this as a point source of environmental PBDE contamination (Toms et al., 2006; Toms et al., 2008). The contamination of sludges and effluents with PBDEs could therefore have potential implications for disposal and beneficial reuse strategies. However, action has been taken in Europe to significantly restrict the use of pentaBDE and octaBDE and the placing on the market of articles containing one or both of these

substances taking effect from 15 August 2004 (EPCEU, 2003). PentaBDE is also a WFD PHS (EPCEU, 2001). Therefore, emissions to wastewater and presence in sludge are expected to decrease through source controls thus reducing the significance of these compounds for agricultural recycling of biosolids.

2.7. Polychlorinated alkanes (PCAs)

Technical mixtures of polychlorinated alkanes (PCAs), often referred to as chlorinated paraffins (CPs), are a class of industrial chemicals comprising of chlorinated straight-chain hydrocarbons. They have been produced since the 1930s for use as extreme pressure lubricant additives, plasticizers, flame retardants, and paint additives (IPCS, 1996). World production of PCAs was estimated to be 300,000 t in 1985 (IPCS, 1996) and a similar amount was produced in 2009 (Eurochlor, 2009). They are manufactured in the European Union, North America, South Africa, Australia, India, China, Taiwan and Japan (Eurochlor, 2009). Following their widespread and unrestricted use, PCAs are now present in a range of environmental compartments (Campbell and McConnell, 1980). They have been detected in human milk (Thomas et al., 2006), environmental samples (Campbell and McConnell, 1980; Bayen et al., 2006) and in air over the UK, including remote regions (Peters et al., 2000). The International Agency for Research into Cancer (IARC) has classified PCAs as Class 2B “possibly carcinogenic to humans” (IARC, 1998).

PCAs are chlorinated linear chain alkanes with the general formula $C_xH_{(2x+2)-y}Cl_y$. They are produced from the n-alkane fractions derived from petroleum distillation by chlorination, with the degree of chlorination ranging between 30% and 72% by weight (IPCS, 1996). They are divided into three groups: short-chain PCAs (noted as sPCAs or SCCPs) comprising 10 to 13 carbon atoms, medium-chain PCAs (mPCAs or MCCPs) comprising 14 to 17 carbon atoms and long-chain PCAs (lPCAs or LCCPs) with 18 or more carbon atoms. PCAs are viscous liquid or yellowish dense oils, except for some lPCAs (C20- to C30- with a chlorine content of >70%) which are solid and are practically insoluble in water (IPCS, 1996). There are many possible positions for the chlorine atoms and presence of chiral carbon atoms lead to a large number of potential positional isomers, enantiomers and diastereoisomers. PCAs therefore represent a difficult analytical problem because of the complexity inherent in industrial mixtures. The total number of possible congeners is unknown, but far exceeds 10,000 (Eljarrat and Barceló, 2006).

Reports of PCA concentrations in sewage sludge samples are very limited in the literature (Table 3). In the early 1990s sPCAs were measured in two German sludge samples from an industrial area, which contained 65 and 47 mg kg⁻¹ dw of sPCAs (Rieger and Ballschmiter, 1995). Concentrations of mPCAs in samples of digested sludge (n=9) from the UK were in the range 1.8 to 93 mg kg⁻¹ dw (Nicholls et al., 2001). In another UK survey (n=14), sPCAs and mPCAs concentrations were between 7–200 mg kg⁻¹ dw and 30–9700 mg kg⁻¹ dw, respectively (Stevens et al., 2003). Comparison of data from the limited amount of information available is difficult.

sPCAs are an identified WFD PHS and are therefore subject to controls that should ultimately lead to the cessation or phasing out of discharges, emissions and losses to the environment (EPCEU, 2001). Nevertheless, concentrations of PCAs reported in sludge remain significantly higher than the regulated PCBs, which are typically <1 mg kg⁻¹ dw in sludge.

A preliminary human health exposure assessment, comparing the ratio of the PCA contaminant concentration in biosolids to the tolerable daily intake of 100 µg kg⁻¹ day⁻¹, demonstrated that PCAs could potentially represent a risk to human health from worst case exposure by the direct ingestion pathway for biosolids (IPCS, 1996). The mean concentration in UK sludge was used for the calculations (1800 mg kg⁻¹ dw from Stevens et al., 2003). Thus, the direct ingestion of 100 mg day⁻¹ of sludge (US EPA, 1997) by a child with

a body weight of 15 kg would contribute 12% of the child's tolerable daily intake of PCAs. By contrast, a 15 kg child suffering from the pica medical condition (deliberate ingestion of soil) is assumed to ingest 10 g of soil per day (US EPA, 1997) and would therefore be exposed to 514% of their tolerable daily exposure to PCAs. The concentrations of PCAs in sewage sludge, evidence of accumulation in human and environmental biota, as well as toxicity data indicate that further research is necessary to assess the risk to human health and the environment from the industrial use of this chemical group.

2.8. Polychlorinated naphthalenes (PCNs)

Technical mixtures of PCNs have been used since the early 1900s as dielectric fluids, engine oil additives, electroplating masking compounds, wood preservatives, lubricants, and for dye production (Falandysz, 1998). The historical use of PCNs precedes PCBs, however their applications are similar. They are also structurally similar and have similar physico-chemical properties. There are 75 PCN congeners, substituted with one to eight chlorine atoms per naphthalene molecule (Table 1). They were voluntarily phased out in the 1970s in the USA, but global production of PCN mixtures was estimated to be approximately 150,000 t in the 1990s (Falandysz, 1998).

PCNs are ubiquitous environmental contaminants sharing many of the characteristics of UNEP POPs (Alcock and Jones, 1999): bioaccumulation (Falandysz, 1998), toxicity (Blankenship et al., 2000), long-range atmospheric transport (Harner et al., 1998) and environmental persistence. Several PCN congeners exhibit dioxin-like toxicity and have been assigned TEF values similar to the coplanar PCBs (Blankenship et al., 2000; Villeneuve et al., 2000). There are three known main sources of PCNs in the environment: technical PCN formulations, technical PCB formulations, and thermal (e.g. combustion, roasting, metal reclamation) and other processes (e.g. chloro-alkali industry) in the presence of chlorine (Falandysz, 1998).

Very limited data on PCN concentrations in sludge was found in the literature (Table 3), with reports from Sweden (Nylund et al., 1992), the UK (Stevens et al., 2003) and recently, China (Guo et al., 2008). In the early 1990s, the ΣPCN (9 congeners) concentration in sewage sludge from Sweden ranged between 3.2 and 5.9 µg kg⁻¹ dw (Nylund et al., 1992). Slightly higher concentrations were reported in the UK where the mean ΣPCN value was 83 µg kg⁻¹ dw and range was 50 to 190 µg kg⁻¹ dw (Stevens et al., 2003). Total PCN concentrations in sludge samples from eight Chinese WWTPs, measured for >70 of the 75 PCN congeners, were between 1.48 and 21.21 µg kg⁻¹ dw, and the ‘dioxin’ toxicity equivalence (TEQ) was in the range 0.11–2.45 ng WHO₀₅ kg⁻¹ dw (Guo et al., 2008). This contribution of dioxin-like PCNs is not high when compared to recently reported dioxin-like compound concentrations in English and European sludges (Stevens et al., 2001).

2.9. Polydimethylsiloxanes (PDMSs)

Polydimethylsiloxanes (PDMSs) are man-made organosilicone compounds that range from low molecular weight volatile materials to high molecular weight polymeric substances (Fendinger et al., 1997). They are widely used in industrial applications and consumer products, such as textile treatments, household and personal care products and antifoams for food processing or WWTP (Griessbach and Lehmann, 1999). These applications all result in discharges to WWTP and the potential to enter the environment as a component of effluents and in sewage sludge (Fendinger et al., 1997). PDMSs have been detected in environmental samples such as surface water, sediments and fish tissue (Watanabe et al., 1984, 1988). PDMSs have low ecological toxicity, which occurs at higher concentrations than those observed in the environment (Hobbs et al., 1975), and are not considered to pose an ecologically significant threat (Frye, 1988).

PDMSs have a very low water solubility and are primarily removed by sorption to solids during wastewater treatment (Varaprath et al., 1996; Fendinger et al., 1997). At least 94% of PDMSs are unchanged during wastewater treatment because of high chemical and thermal stabilities (Watts et al., 1995; Fendinger et al., 1997). PDMSs are not toxic to wastewater microbial communities and do not affect treatment performance (Watts et al., 1995). PDMSs not removed on the sludge solids are present in wastewater treatment effluent as a component of the suspended solids (Fendinger et al., 1997) and this accounts for their environmental distribution in sediments and surface waters (Watanabe et al., 1988).

PDMSs degrade in the soil environment as a result of abiotic processes rather than biodegradation, and have a half-life estimated to range from 4 to 28 days (Carpenter et al., 1995; Lehmann and Miller, 1996; Lehmann et al., 1998; Griessbach and Lehmann, 1999). Increased soil moisture retards the degradation of PDMSs in soil (Griessbach and Lehmann, 1999). Clays appear to be the component responsible for catalysing oligomerization and hydrolysis reactions of PDMSs in soil (Buch and Ingebrigtsen, 1979; Xu et al., 1998). A multiyear field dissipation study and laboratory studies, to evaluate the persistence or degradation of PDMSs in biosolids-amended soils, found that soil moisture was the factor controlling the persistence or degradation of PDMSs and that there was no direct effect of biosolids on PDMS persistence or degradation (Traina et al., 2002). As may be expected, the half-life for PDMS determined under field conditions (876 to 1443 days) was longer than that estimated in laboratory studies and this was attributed to the higher moisture contents of the field soils (Traina et al., 2002). PDMSs eventually mineralise in soil to carbon dioxide and silicic acid (Stevens, 1998). A study examining the ecotoxicity of PDMS found no effects on seed germination, seed survival, plant yields or soil microorganisms (Tolle et al., 1995).

Few studies have reported the concentrations of PDMSs present in sewage sludge (Table 3). The most comprehensive study available (Fendinger et al., 1997) indicated PDMS concentrations in US sludges were in the range 290 and 5155 mg kg⁻¹ dw with a mean value of 1120 mg kg⁻¹ dw. This is approaching a magnitude higher than the concentration (144 mg kg⁻¹ dw) reported in a Japanese sludge (Watanabe et al., 1984). Further work would appear to be necessary to determine the concentrations of PDMSs in sewage sludges internationally.

2.10. Perfluorochemicals (PFCs)

Perfluorochemicals (PFCs) are a family of anthropogenic chemicals that have been used since the late 1950s to make products resistant to heat, oil, stains, grease and water. Common applications include non-stick cookware, breathable membranes for clothing, stain-resistant carpets and fabrics, components of fire fighting foam, surfactants and other industrial applications (US EPA, 2008). They have been used in many industry sectors, including the aerospace, automotive, building/construction, chemical processing, electronics, semiconductors, and textile industries (US EPA, 2008).

PFCs are persistent and widely dispersed in the environment (Giesy and Kannan, 2001; Kannan et al., 2001). Accumulation of PFCs has been detected in ocean animals, such as birds and mammals, and in human tissues throughout the world (Olsen et al., 2003; Kannan et al., 2004). The human and environmental toxicological response to such exposure is not known, but could include endocrine disruption (Lau et al., 2004).

The chemical structures of PFCs make them very resistant to degradation in the environment; the carbon–fluorine bonds are extremely strong and are stronger relative to other commonly used halogens viz., bromine and chlorine. Consequently, perfluorocarbon chains do not readily biodegrade and any biodegradation may be limited to attached hydrocarbon moieties. The two most common groups of PFCs that are measured and detected in environmental matrices are:

- Perfluoroalkyl sulphonates (PFASs) – perfluorooctane sulphonate (PFOS), perfluorohexane sulphonate (PFHxS), perfluorooctane sulphonamide (PFOSA)
- Perfluoroalkyl carboxylates (PFACs) – perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA)

PFASs and PFACs are synthetic chemicals that do not occur naturally in the environment (US EPA, 2008). They are employed as a base chemical in the preparation of fluoropolymers and upon degradation can release the environmentally persistent monomers (Boulanger et al., 2005).

Directive 2006/122/EC (EPCEU, 2006) places restrictions on the marketing and use of PFOS and there are also voluntary reductions on PFOA although it is still manufactured. PFOS is also under review for possible identification as a WFD Priority Substance or PHS (EPCEU, 2008). The EU is currently assessing PFOA and, whilst there are no restrictions in place in the EU at present, a ban could be imposed in the future. However, these substances have been extensively used in the built environment and therefore could represent a significant, long-term diffuse input into wastewater and sludge.

The earliest available report of PFCs in sewage sludge is a study of sludges collected from six USA cities (3M Environmental Laboratory, 2001). PFOS and PFOA were the most common PFCs present and were detected in all samples analysed at the low µg kg⁻¹ dw range (Table 3). A sludge sample was tested from a WWTP serving a fluorochemical manufacturer and this correlated with substantial increases in PFOS (2980 µg kg⁻¹ dw) and PFOA (173 µg kg⁻¹ dw) concentrations. This study demonstrated that PFCs, in particular PFOS and PFOA, are likely to be present in sludge in Western countries where PFCs are manufactured and used. Domestic sources are also likely to be a major contributor of PFCs in sewage sludge.

Other studies in the USA (Schultz et al., 2006; Sinclair and Kannan, 2006; Loganathan et al., 2007) and Europe (Bossi et al., 2008) report similar concentration ranges. No differences in PFC levels were apparent in sludges from urban and rural WWTPs, however, major seasonal variations in concentrations were observed (Loganathan et al., 2007). Whilst PFCs have been investigated at WWTPs in other countries (Alzaga and Bayona, 2004; Boulanger et al., 2005), sewage sludge concentrations have mainly been reported in the US. This is largely a consequence of the analytical difficulties associated with quantification of PFCs in sewage sludge matrices.

Mass balance studies of PFCs at WWTP commonly report higher mass loadings of PFOA and PFOS in WWTP effluent compared to raw influent (Schultz et al., 2006; Sinclair and Kannan, 2006; Loganathan et al., 2007). This suggests the degradation of other fluorinated organic compounds (i.e. fluoropolymers) into PFOA and PFOS may take place during wastewater treatment (Loganathan et al., 2007).

Field investigations have demonstrated that PFCs in sludge-amended soil can be mobilised by rainfall (Gottschall et al., 2010).

2.11. Quaternary ammonium compounds (QACs)

Quaternary ammonium compounds (QACs) are cationic surfactants. The molecules contain at least one hydrophobic hydrocarbon alkyl chain linked to a positively charged nitrogen atom. The other alkyl groups are typically short-chain substituents such as methyl or benzyl groups. Cationic surfactants are positively charged in aqueous solutions (Madsen et al., 2001; Ying, 2006). QACs are commonly used in domestic products such as fabric softeners, hair conditioners and other hair preparations. Other applications of cationic surfactants include disinfectants and biocides, emulsifiers, wetting agents, and processing additives. By volume, the most important cationic surfactants in household products are the alkyl ester ammonium salts that are used in fabric softeners (Madsen et al., 2001).

Ditallow dimethyl ammonium chloride (DTDMAC) was the most widely used active ingredient in fabric softeners. However DTDMAC is toxic to aquatic organisms (Roghair et al., 1992; Versteeg et al., 1992) and was therefore voluntarily phased out by industry following an ecological risk assessment (van Leeuwen et al., 1992) in preference of more biodegradable QACs, such as diethyl ester dimethyl ammonium chloride (DEEDMAC) (Giolando et al., 1995). Fernandez et al. (1996) monitored the DTDMAC concentrations in sewage sludge in Switzerland, which declined from 3.67 g kg⁻¹ dw in 1991 to 0.21 g kg⁻¹ dw in 1994 due to its replacement with the alternative QACs.

Domestic use of QACs is the primary source of these compounds entering WWTPs. Due to their positive charge, cationic surfactants sorb strongly to the negatively charged surfaces of sludge, soil and sediments (Madsen et al., 2001; Ying, 2006; Clara et al., 2007), and it is estimated that ~90% of QACs associate with sludge during wastewater treatment (van Leeuwen et al., 1992). Removal of QACs in wastewater treatment can also be attributed to biodegradation (Nishiyama et al., 1995), where degradation rates are typically reported in days or hours (Giolando et al., 1995; Sütterlin et al., 2008). WWTP discharges can result in environmental contamination of marine sediments with QACs (Li and Brownawell, 2009).

There are very few reports of QAC concentrations in sludge in the scientific peer-reviewed literature (Table 3). A recent study from Austria reported that the total QAC concentration was in the range 22 to 103 mg kg⁻¹ dw (Martínez-Carballo et al., 2007), suggesting further substantial reductions in QACs have taken place compared to earlier work addressing the impact of phasing out DTDMAC (Fernandez et al., 1996).

It has been controversially argued that QACs have biocidal properties that may confer antibiotic resistance to bacteria (Gaze et al., 2005; Gaze, 2008). However, it is difficult to reconcile this firstly because QACs are not disinfectants, but are in fact surfactants with low toxicity and ecotoxicity (Giolando et al., 1995). Secondly, modern QAC formulations are designed to rapidly biodegrade during wastewater treatment and anaerobic digestion, and are almost completely removed by these processes (Giolando et al., 1995). In soil QACs rapidly degrade with short half-lives (17–40 d) (Giolando et al., 1995).

2.12. Steroids

Natural endogenous (17β-oestradiol, oestrone, oestriol) and synthetic steroids (17α-ethinyloestradiol, mestranol) are excreted by humans and WWTP effluent is the primary source of synthetic steroids entering the environment (Snyder et al., 2001). Livestock (particularly lactating cows) are also a major source of endogenous oestrogen inputs to soil (Kolodziej et al., 2004). Over 99% of oestrogenic activity in sewage effluents and surface waters may be attributable to the presence of 17β-oestradiol (E2) and 17α-ethinyloestradiol (EE2) at concentrations in the ng L⁻¹ range (Snyder et al., 2001). Oestrogenic activity in WWTP effluents has resulted in adverse effects on environmental biota (Jobling et al., 1998). Natural and synthetic steroids are excreted from the human body as inactive polar conjugates, but are present in sewage influent and effluent as free, active steroids (Belfroid et al., 1999). Once released from the body conjugated oestrogens undergo chemical or enzymatic dissociation in bacterial sludge and re-form as active oestrogens (Belfroid et al., 1999; Reddy et al., 2005).

Several investigations have examined the fate of oestrogens in WWTPs, however, few studies have measured the concentrations of oestrogenic compounds in sludge due to the analytical difficulties involved (Gomes et al., 2004). The presence of 'free' oestrogens in WWTP effluents and receiving waters is commonly reported (Shore et al., 1993; Desbrow et al., 1998; Ternes et al., 1999b), demonstrating that the conversion of oestrogen metabolites into active forms occurs somewhere between the domestic discharge and WWTP outlet. However, the degradation of all steroid-like compounds (natural and synthetic) occurs rapidly and typically within a few days or less

(Ternes et al., 1999a; Korner et al., 2000; Layton et al., 2000; Hashimoto and Murakami, 2009). Natural oestrogens biodegrade in the order: 17β-oestradiol → oestrone → oestriol and at a faster rate compared to synthetic mestranol and 17α-ethinyloestradiol (Ternes et al., 1999a). Research at full-scale WWTPs indicates that operational retention times may only allow partial degradation as concentrations of oestrone measured in treated effluent are frequently increased compared to the influent raw wastewater (Barontri et al., 2000).

Mass balance studies of WWTPs consistently show high removal rates of oestrogens during wastewater treatment, equivalent to: 64–99.9% for 17β-oestradiol, and 78–83% for oestrone and 17α-ethinyloestradiol (Ternes et al., 1999b). While many studies do not distinguish between degradation and sludge partitioning (Schlusener and Bester, 2008), laboratory experiments indicate the high biodegradability of steroids (Ternes et al., 1999a). Such laboratory studies indicate that degradation processes contribute significantly to steroid removal (Ternes et al., 1999a). The removal rate of the natural hormones 17β-oestradiol, oestrone and 17α-ethinyloestradiol during wastewater treatment was >90% and only 5% of the mass of oestrogens entering the WWTP was sorbed to the final digested sewage sludge, indicating high biodegradability in the WWTP (Andersen et al., 2003).

Few studies have reported steroid concentrations in sewage sludge due to analytical difficulties, as well as high degradation rates. The concentrations of oestrone (<2–37 μg kg⁻¹ dw), 17β-oestradiol (5–49 μg kg⁻¹ dw), 17α-ethinyloestradiol (<4–17 μg kg⁻¹ dw) and mestranol (<2 μg kg⁻¹ dw) were measured by Ternes et al. (2002) in German sludges (Table 3). A recent survey of US sludges (n = 84) quantified 25 steroids (US EPA, 2009), but only three steroid compounds (i.e., campesterol, cholestanol, and coprostanol) were found (US EPA, 2009). 17α-ethinyloestradiol was not detected in any sample and five hormones were found in fewer than six samples. Other oestrogenic steroids were not frequently detected and when detected were in the low μg kg⁻¹ dw concentration range (US EPA, 2009).

These low concentrations, combined with fast biodegradation rates in WWTP mass balance and laboratory studies suggest that steroids are unlikely to pose a risk to human health or the environment when land applying biosolids.

2.13. Synthetic musks

Synthetic musks are inexpensive substitutes for natural musks and have been used since the 1930s as fragrances in a variety of domestic and industrial products viz., detergent, cosmetics, shampoo, perfume, food and cigarette additives (Rimkus, 1999). Synthetic musks can be broadly categorised into two groups: nitromusks and polycyclic musks. Nitro musks were the first commonly used synthetic musks, but their use was reduced in the 1950s due to evidence of toxicity to humans and the environment (Tas et al., 1997). They were largely replaced with the polycyclic musks, which account for approximately 85% of worldwide production, while nitro musks account for the remaining 15% (Tas et al., 1997).

The most commonly used synthetic musks are:

- nitromusks – musk moskene (MM), musk tibetene (MT), musk xylene (MX), musk ketone (MK) and musk ambrette (MA).
- polycyclic musks – Galaxolide™ (HHCB), Tonalide™ (AHTN), Celestolide™ (ADBI), Phantolide™ (AHMI), Cashmeran™ (DPMI) and Traseolide™ (ATII).

The majority of synthetic musk applications occur in the domestic environment and are found in personal care products that will be released into the sewer after use and, consequently, the principal source of synthetic musks to the environment is thought to be WWTP effluents and sludges (Rimkus, 1999). WWTP mass balance studies indicate removal rates of between 83 and 91%, whereas

approximately 40% of AHTN and HHCb were eliminated during anaerobic sludge digestion (Balk and Ford, 1999a). The concentrations of synthetic musks in sewage sludge have been reported from Switzerland (Herren and Berset, 2000), Germany (Heberer, 2002), Spain (Llompart et al., 2003), UK (Stevens et al., 2003), China (Zeng et al., 2005) and Hong Kong (Shek et al., 2008).

The nitromusks (MA, MX, MM, and MT) were generally not detected in these studies and when present were found at low $\mu\text{g kg}^{-1}$ dw values. This is consistent with use patterns of nitromusks that have largely been replaced by the polycyclic musks (Rimkus, 1999). MX is currently under review for possible identification as a WFD Priority Substance or PHS (EPCEU, 2008).

The polycyclic musks most frequently detected and at the highest concentrations in sludge are HHCb ($0.1\text{--}81 \text{ mg kg}^{-1}$ dw) and AHTN ($0.03\text{--}16 \text{ mg kg}^{-1}$ dw) (Table 3). Again, this is consistent with use patterns as, together, HHCb and AHTN account for >95% of the market share of polycyclic musks (Tas et al., 1997). Current German proposals are to set biosolids limits for HHCb and AHTN of 10 mg kg^{-1} dw and 15 mg kg^{-1} dw, respectively (BMU, 2007). Other polycyclic musks (ADBI, DPMI) are typically present in lower concentration ranges ($<1 \text{ mg kg}^{-1}$ dw). The mean HHCb concentration in UK sewage sludges (27 mg kg^{-1} dw) was larger than in other European sludges, such as Switzerland (4.85 mg kg^{-1} dw), Germany (8.26 mg kg^{-1} dw) or Spain (0.16 mg kg^{-1} dw). However, concentrations of HHCb measured in sludge in China and Hong Kong are similar to the UK. Soil ecotoxicity studies report a PNEC for HHCb and AHTN of 0.32 mg kg^{-1} dw that incorporates a fifty fold safety factor and is based upon earthworm and springtail no observable effects concentrations of 45 mg kg^{-1} dw for both chemicals (Balk and Ford, 1999b). The concentrations of these compounds found in biosolids are relatively high when compared to other POPs and therefore, further investigation is warranted to elucidate their fate, behaviour, and persistence in biosolids-amended soils (Stevens et al., 2003).

2.14. Triclosan and triclocarban

Triclosan (TCS; 5-chloro-2-[2,4-dichloro-phenoxy]-phenol) and triclocarban (TCC; 3,4,4'-trichlorocarbanilide) are antimicrobial agents widely used in personal care products such as shampoos, soaps, deodorants, cosmetics, skin-care lotions and creams, mouth rinses, and toothpastes. These domestic applications are likely to be

the major source of TCS and TCC to WWTPs (Bester, 2003). The amount of TCS and TCC used in consumer products typically ranges from 0.1 to 0.3% (w/w). At these levels they exhibit a broad-spectrum activity against bacteria, molds and yeasts (McAvoy et al., 2002).

TCS and TCC are both relatively hydrophobic, with $\log K_{ow}$ values at neutral pH of 3.5–4.8 and 4.9, respectively (Halden and Paull, 2005; Snyder et al., 2010). The octanol–water distribution of TCS, however, depends on the pH of the environmental matrix that the compounds are exposed to, since the hydroxyl groups ($-\text{OH}$) in the molecule are capable of deprotonation allowing water solubility.

Mass balance studies at WWTP show the incomplete removal of TCC and TCS during wastewater treatment (Heidler et al., 2006; Sapkota et al., 2007). Therefore, TCS and TCC are released into the environment in WWTP effluents and by land application of biosolids. Consequently, they are regularly detected in surface waters receiving WWTP inputs (Halden and Paull, 2004; Halden and Paull, 2005). However, they can also be found upstream of WWTP discharges indicating environmental persistence (Sapkota et al., 2007). Potential environmental problems from the release of TCC and TCS from WWTP effluent in surface water include bioaccumulation in algae and snails (Coogan et al., 2007; Coogan and La Point, 2008), algal growth inhibiting effects (Yang et al., 2008) and endocrine disruption (Ahn et al., 2008).

A mass balance for TCC at a WWTP showed that approximately 75% of the initial mass of TCC was recovered in sludge, with a concentration of 51 mg kg^{-1} dw (Heidler and Halden, 2007). It is commonly reported that TCS and TCC partition to sludge during wastewater treatment (Ying and Kookana, 2007). Mass balance studies indicate that losses of between 48 and 65% occur possibly due to volatilization or biodegradation (Bester, 2003; Heidler and Halden, 2007), but biodegradation is the more likely removal mechanism (Ying and Kookana, 2007). Laboratory studies have also confirmed the degradation of TCS (Federle et al., 2002; Stasinakis et al., 2007). Biosolids were identified as a source of TCS in agricultural soil contributing to the bioaccumulation of TCS in earthworm tissue with bioaccumulation factors ranging from 0.05 to 27 (Kinney et al., 2008). Furthermore, two recent studies have detected the movement of several pharmaceuticals and personal care products from agricultural areas amended with biosolids into tile drainage water (Lapen et al., 2008) and runoff (Topp et al., 2008). Water run-off experiments found that TCS leached only from surface applied biosolids and not

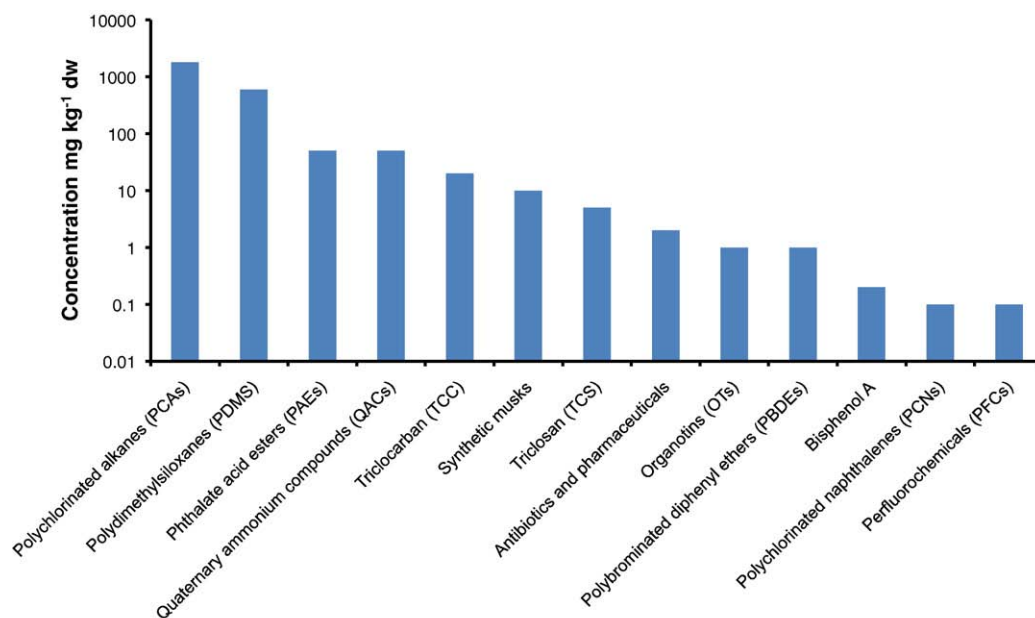


Fig. 1. Typical concentrations of selected 'emerging' organic contaminants in sewage sludge (mg kg^{-1} dw).

Table 4

Assessment matrix to determine research priorities for selected 'emerging' organic contaminants in sewage sludge with respect to their potential significance for agricultural utilisation.

Emerging organic contaminant	Persistent in soil (>6 months) 2 – yes 1 – uncertain 0 – no	Human food chain 2 – possible 1 – uncertain 0 – no	Ecological bioaccumulation 2 – yes 1 – possible 0 – no	Soil ecotoxicity 2 – yes 1 – uncertain 0 – no	Research quality 3 – lack of empirical data 2 – few reported studies 1 – a number of consistent studies 0 – many consistent studies	Score (/11)
Antibiotics and pharmaceuticals ^a	0	2	0	1	2	5
Benzothiazoles	1	1	0	1	3	6
Bisphenol A	0	0	0	0	2	2
Organotins	1	1	2	1	2	7
Phthalate acid esters (PAEs)	0	0	0	0	1	1
Polybrominated diphenyl ethers (PBDEs)	2	2	2	1	0	7
Polychlorinated alkanes (PCAs)	2	2	1	1	3	9
Polychlorinated naphthalenes (PCNs)	2	2	1	1	3	9
Polydimethylsiloxanes (PDMSs)	0	0	0	0	1	1
Perfluorochemicals (PFCs)	2	2	2	1	3	10
Quaternary ammonium compounds (QACs)	0	0	0	0	2	2
Steroids	0	0	0	0	2	2
Synthetic musks	1	0	1	0	1	3
Triclosan	1	0	2	2	2	7
Triclocarban	1	0	2	2	2	7

^a The chemical properties of antibiotics and pharmaceuticals and subsequent behaviour in the environment can vary greatly. The scores are considered generally for antibiotics and pharmaceuticals, certain exceptions such as carbamazepine may exhibit longer soil persistence.

from subsurface application. Therefore, land application practices for biosolids in the UK are likely to prevent surface water contamination from TCS or TCC, although a potential risk to groundwater by leaching has been suggested (Topp et al., 2008).

Reports of TCS and TCC concentrations in activated sludges and biosolids have been reported from the USA (McAvoy et al., 2002; Heidler et al., 2006; Kinney et al., 2006; Sapkota et al., 2007; US EPA, 2009), Germany (Bester, 2003), Spain (Morales et al., 2005), Canada (Chu and Metcalfe, 2007), Australia (Ying and Kookana, 2007) and Greece (Gatidou et al., 2007; Pothitou and Voutsas, 2008; Stasinakis et al., 2008). The ranges of concentrations reported for TCS and TCC are 0.5–16 mg kg⁻¹ dw and 4–50 mg kg⁻¹ dw, respectively (Table 3). Recent biosolids surveys in the USA show that TCC and TCS are routinely detected in biosolids and that the concentrations are higher than previously reported (McAvoy et al., 2002; Heidler et al., 2006; Kinney et al., 2006; Sapkota et al., 2007; US EPA, 2009).

3. Assessment and ranking of emerging contaminants

3.1. Typical concentrations of organic contaminants in sewage sludge

A summary of reported concentrations of the selected 'emerging' contaminants examined here is presented in Fig. 1. The concentrations in Fig. 1 are presented on a logarithmic scale in descending order of reported mean concentrations, ranging from PCAs (g kg⁻¹ dw) to PFCs (µg kg⁻¹ dw). The concentrations of the industrial chemical PCAs are much higher compared to contaminants from domestic sources i.e., QACs, TCS. These concentrations suggest significant industrial discharges of PCAs occur into the wastewater collection system. Chemicals utilized in the domestic environment were in the next 'high-level' concentration range (PAEs, QACs, TCC, synthetic musks, TCS), followed by less commonly used industrial and domestic chemicals (OTs, PBDEs, PCNs, PFCs).

3.2. Matrix assessment

Research and monitoring priorities were identified from an assessment matrix consisting of five criteria applied to the selected 'emerging' contaminants (Table 4). These included:

- environmental persistence in soil environment (>6 months);
- potential for human health impacts resulting from the land application biosolids;

- evidence or likelihood of bioaccumulation in humans or the environment;
- evidence of ecotoxicity, and
- the quality of empirical data and trends on the contaminant in biosolids internationally.

3.3. Assessment results

The 'emerging' contaminants were ranked in decreasing order of priority as follows (maximum possible score of 11):

- perfluorinated chemicals (10)
- polychlorinated alkanes (9), polychlorinated naphthalenes (9)
- polybrominated diphenyl ethers (7), organotins (7), triclosan (7), triclocarban (7)
- benzothiazoles (6)
- antibiotics and pharmaceuticals (5)
- synthetic musks (3)
- bisphenol A (2), quaternary ammonium compounds (2), steroids (2)
- phthalate acid esters (1), polydimethylsiloxanes (1).

PFCs represent a potentially major environmental uncertainty. They have been detected in human blood (Olsen et al., 2003) and environmental samples (Giesy and Kannan, 2001) throughout the world. They have a unique chemistry that facilitates a degree of water solubility, which is not observed with other POPs. Thus, the barriers that normally prevent human and ecological exposures to POPs in biosolids-amended soil may not prevent movement of PFCs. For example, increased water solubility raises the likelihood of exposure through all pathways – water contamination, plant accumulation and grazing animal accumulation (Chaney et al., 1996). Water contamination and plant accumulation are considered to be the priority pathways of concern, as grazing animal accumulation is not unique to PFCs. Whilst measures are in place in Europe to restrict the major exposure risks to PFOS (EPCEU, 2006), and could be extended to PFOA in future, transfer to biosolids could continue to occur over a long period due to the ubiquitous use of these substances within the built environment.

PCAs and PCNs were both detected in recent surveys of UK biosolids (Stevens et al., 2003). PCAs were found at relatively high concentrations (mean concentration 1800 mg kg⁻¹ dw), while PCNs were typically less than 1 mg kg⁻¹ dw. By comparing the concentrations of these

compounds to PCBs and PCDD/Fs it is apparent that PCAs warrant further investigation, as the concentrations of PCAs are three orders of magnitude higher than those of PCBs found in contemporary sewage sludge. Also, there is mounting evidence that PCAs are accumulating in the human population and the impact of these chemicals on human health and the environment has not yet been explored (Nicholls et al., 2001; Thomas et al., 2006). A preliminary human health exposure assessment indicated that the concentrations of PCAs in sludge could be harmful to human health through the direct ingestion exposure pathway. The concentrations of PCNs are small in contrast and are unlikely to pose a threat to human health or the environment at the concentrations measured in sewage sludge internationally.

PBDEs have been the subject of increasing international research (Clarke et al., 2008c). PBDE concentrations in biosolids may be similar throughout the world, however, manufacturing can add significant quantities of PBDEs to the burden derived from domestic sources (Fabrellas et al., 2004). Furthermore, restrictions applied to the penta- and octa-PBDE commercial formulations may increase demand and consumption of the decaBDE formulation. The deca-formulation is the source of BDE 209, the major congener detected in sludges internationally and field studies have shown that this compound is persistent in the environment (Eljarrat et al., 2008).

Antimicrobial agents TCS and TCC have received increasing research attention internationally (Singer et al., 2002; Ying and Kookana, 2007) and are capable of ecological toxicity (Yang et al., 2008) and bioaccumulation in environmental biota (Kinney et al., 2008). However, a recent risk assessment (Reiss et al., 2009) showed that TCS was unlikely to pose a significant problem for sensitive environmental compartments from the agricultural use of biosolids. Nevertheless, field studies have demonstrated that TCS and TCC may exhibit some environmental persistence in biosolids-amended soil (Topp et al., 2008; Cha and Cupples, 2009). The biodegradation of these compounds is reported in laboratory (McAvoy et al., 2002) and WWTP studies (Ying and Kookana, 2007), but further evaluation of the persistence and toxicity of TCS and TCC in biosolids-amended soil is required. It is also important to recognise that the primary route of human exposure to TCS and TCC will occur in the domestic environment and not as a result of land applying biosolids.

Organotin compounds were present in small concentrations in biosolids in the studies reported, but are potentially ecotoxic. There are limited empirical biosolids measurements internationally, so it is difficult to draw definitive conclusions about typical biosolids concentrations. Furthermore, they warrant further investigation to determine their significance in biosolids-amended soil.

Limit values for benzothiazoles in sludge are proposed and under consideration by the German Government. However, there are insufficient data concerning these chemicals to make an informed assessment of their potential significance. Nevertheless, benzothiazoles have not been detected in environmental media and laboratory studies indicate that they are biodegradable (de Wever and Verachert, 1997; Gaja and Knapp, 1998). Therefore, it would appear that there are no specific scientific grounds for regulating this group of OCs in biosolids. Nevertheless, further research is required to determine typical concentrations in sewage sludge, environmental persistence, ecological toxicity and persistence in biosolids-amended soil.

A recent risk assessment completed in Norway evaluated over 1400 pharmaceutical compounds based upon a tiered approach where chemicals were screened against consumption, volume wastewater of influent, human metabolism, biodegradation and behaviour in WWTP (Eriksen et al., 2009). Fourteen pharmaceuticals were identified for further risk assessment investigation as the only compounds that potentially may pose risk to human health or the environment. These were: atorvastatin, carisoprodol, chlorprothixene, ciprofloxacin, dipyrindamole, fexofenadinesotalol, gabapentin, levetiracetam, losartan, mesalazin, metoprolol, ranitidine and tetracycline. The estimated soil concentrations of drug substances (PEC) were low (concentration range

0.01–1 mg kg⁻¹ dw) and well below the estimated PNEC values. Therefore Eriksen et al. (2009) concluded that drug substances in sewage sludge constitute a low risk to the soil compartment.

The polycyclic musks are unlikely to pose a risk to human health from land application of biosolids and the majority of human exposure will occur in the domestic environment. The environmental risk assessment of AHTN and HCHB indicates that these substances are unlikely to pose a significant hazard to the ecosystem at the concentrations likely to be found in biosolids-amended soil (Balk and Ford, 1999a, 1999b). Therefore, proposals for AHTN and HCHB contaminant limits in biosolids appear unfounded. However, further research is warranted to understand the ecological risk of polycyclic musks with respect to fate, mobility and persistence given the relatively high concentrations of these compounds detected in sludge.

Four high-volume production chemicals were included for assessment and all were indicated as having a low research priority. Concern has been raised over QACs due to potential development of antibiotic resistance and endocrine disruption for bisphenol A and phthalates. However, these chemicals are not environmentally persistent, they biodegrade rapidly and humans are routinely exposed to these compounds through more important pathways via dermal and ingestion routes in the domestic environment. PDMS is another high-volume production chemical that is not considered to pose a risk to humans or the environment when land applying biosolids due to low toxicity.

Steroid concentrations in sludge were only found in two international studies. There are analytical difficulties that may prevent the detection and measurement of this group of compounds, however, they are also readily degradable and are therefore not present in significant quantities in stabilised biosolids.

4. Conclusions

There are 50 million chemicals entered in the Chemical Abstracts Registry and of these 143,000 chemicals are registered with the European Chemicals Agency for industrial use (ECHA, 2009; Toussant, 2009). Once discharged from industrial, domestic and urban sources into the urban wastewater collection system they may transfer to the residual sewage sludge during wastewater treatment. Continued vigilance is therefore required to monitor and determine the significance and implications of 'emerging' OCs for the land application of treated wastewater biosolids. The significance of a selection of key contaminants for the agricultural use of biosolids was examined here based upon persistence, human health impacts, bioaccumulation, ecotoxicity and quality of published research for the given chemicals. The concentrations of the 'emerging' OCs in biosolids were also considered.

Two chemical classes warrant particular note. These are the PFCs and PCAs. PFCs are an emerging environmental concern as they have been detected in human blood (Olsen et al., 2003) and environmental samples (Giesy and Kannan, 2001) throughout the world. They have a unique chemistry for a chemical defined as a POP that facilitates a degree of water solubility, and therefore, there is an increased likelihood of exposure through all pathways (water contamination, plant accumulation and grazing animal accumulation) compared to other POPs (Chaney et al., 1996). PCAs were found at relatively high concentrations in sludge (mean concentration 1800 mg kg⁻¹ dw). Comparison of the concentrations of these compounds to PCBs and PCDD/Fs shows that the PCA content in sludge is three orders of magnitude higher than PCB values for instance, and signals the importance of further investigations into the significance of PCAs in biosolids for land application.

Recycling biosolids on land is recognised internationally as the most sustainable option for managing the residual sludge from urban wastewater treatment and most risk assessments demonstrate that this practice does not place human health at risk from the OCs studied.

However, continued vigilance in assessing the significance and implications of 'emerging' OCs in sludge is necessary to support and ensure the long-term sustainability and security of the beneficial agricultural route for biosolids management. The research and monitoring priorities for 'emerging' OCs in sludge we have identified and discussed here would further contribute to the technical evidence base to protect human health and the environment when sewage sludge biosolids are recycled in agriculture as soil improvers and fertilisers.

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