SCREENING OF ORGANIC POLLUTANTS IN SEWAGE SLUDGE AMENDED ARABLE SOILS

NATUR VÅRDS VERKET
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WSP

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Sammanfattning
Spridning av organiska miljögifter i åkermark från användning av slam från reningsverk som gödningsmedel har studerats i en screeningstudie åt Naturvårdsverket. Huvudsakliga syftet med utredningen är att:

- Insamla miljödata på organiska föroreningar i jord och biota från åkermark gödslad med slam från reningsverk.

- Utvärdera uppmätta halter av organiska föroreningar mot tidigare resultat och modelluppskattningar.

Studien utgörs av ett nationellt program med totalt 48 prover. Prover insamlades från två fältförsök i Skåne: Igelösa och Petersborg, och två privata fält, Taxinge och ”Sörmland”. Huvudsaklig matris var jord (n=42), men även prover av slam (n=2), gröda (n=3) och mask (n=1) har ingått i studien.

Undersökningen visar att PFOS, PBDE (47, 99, 100 and 209), galaxolid, tonalid och DEHP anrikas i jord behandlad med slam. Det finns även enstaka indikationer på att halter av DIDP, nonylfenol och TCPP kan öka, men dessa observationer är osäkra eftersom de inte är vanligt förekommande.

Jämförelse mellan modellberäkningar och fältdata visar på bra överensstämmelse när det gäller PFOS och BDE-209 d.v.s. dessa ämnen ackumuleras i jorden över tid vid upprepad slambehandling. De ämnen som generellt inte detekteras, alkylfenoler, dibutylftalat och LAS, stämmer också bra överens med tidigare uppskattningar och mätresultat.

I kärnor från höstvete provtaget vid Petersborg detekterades ingen av de analyserade organiska ämnena. Resultatet stödjer tidigare modellberäkningar och upptagsstudier: ackumulationen av många organiska ämnen från jord till gröda är begränsad.

I mask från Taxingefältet påträffades PFOS och PBDE (47, 99 och 100) i detekterbara halter, vilket visar att dessa ämnen kan bioackumuleras i biota. Beräknade bioackumulationsfaktorer (BSAF) från jord till biota ligger i linje med tidigare resultat.

Riskkarakteriseringen för jordekosystem och människor exponerade via intag av grödor visar att halter i jord efter lång tids gödsling med slam inte utgör en risk för jordekosystemet eller människor. Dessa resultat överensstämmer väl med tidigare resultat.

SUMMARY

An environmental screening study was performed on organic pollutants in arable land where municipal sewage sludge is used as fertiliser. The study is part of the national environmental monitoring program run by the Swedish EPA. The major goals of the study were to:

- Provide data on environmental levels of organic contaminants in soil and biota from arable land amended with sewage sludge.
- Evaluate measured field levels of organic contaminants against previous results and model predictions.

The study consists of a national program with a total of 48 samples. Samples were collected from two experimental arable fields in Skåne: Igelösa and Petersborg, and two private farm fields, Taxinge and “Sörmland”. The three first fields represent long time sewage sludge addition and the last field a first time sludge addition. Major matrix was soil (n=42), but also samples of sewage sludge (n=2), crops (n=3) and earthworms (n=1) were studied.

The following substance groups were studied in all matrices except of earthworms; phthalates, LAS, alkylphenols, antibacterial substances (triclosan, triclocarban), musk compounds, organophosphates, perfluoroalkyls (PFOS, PFOA) and brominated flame retardants (PBDE). Musk compounds, perfluoroalkyls and brominated flame retardants were studied in earthworms.

The study shows that PFOS, PBDE (47, 99, 100 and 209), galaxolide, tonalide and DEHP are enriched in sludge amended soils, as compared to control soils. There are also single indications that concentrations of DIDP, nonylphenol and TCPP may increase, but these observations are uncertain as they are not generally observed.

The results indicate that the concentrations of certain contaminants increase with increasing sludge addition rate and that the impact primarily, but not exclusively, is in the upper most soil layers (0-0,3 m). Simple mass balance demonstrates that single additions of sludge will for many substances result in soil concentrations so low (sub µg/kg dw) that they cannot be detected at the analytical reporting limits used.

When comparing model predictions with the present field data, PFOS and BDE-209 behaves as expected, i.e. they accumulate in soil upon repeated applications of sludge over time. The general non-detects of alkylphenols, dibutylphthalate and LAS are also in agreement with predictions and earlier findings. The non-detects of organophosphates is difficult to evaluate because of relatively high analytical reporting limits compared to maximum expected levels. The general presence of galaxolide in the present sludge amended soils is however in contrast to model predictions. At three sites, galaxolide is enriched in amended soils relative to control fields. Whether galaxolide accumulates or not, following repeated applications, cannot be assessed from this data due to contradictory results. Earlier model predictions and degradation studies do, however, suggest that galaxolide should not accumulate but persist for about one year.

In grains of winter wheat grown at the experimental fields in Petersborg, none of the organic substances were detected. These results are in line with earlier findings. When comparing the non-detects to theoretically expected concentrations in crops, it is apparent that expected concentrations in crops are very low (ng/g ww). For most substances expected levels in crops would be so low that they cannot be detected within the analytical reporting limits. The results thus confirm previous model assessments and laboratory uptake studies: many organic pollutants accumulate poorly from soil to crops.

In earthworms from Taxinge PFOS and PBDEs (47, 99 and 100) were detected, showing that these substances can bioaccumulate. Calculated biota to soil accumulation factors (BSAF) are in line with earlier findings.

To characterize the risk for the soil ecosystem and for humans exposed via intake of crops, observed concentrations of organic substances in soil amended with sewage sludge were evaluated against critical concentrations. The results indicate that levels in soil after long term sludge additions do not pose a risk to the soil ecosystem or humans. These findings are in line with earlier findings.

SAMMANFATTNING

Spridning av organiska miljögifter i åkermark från användning av slam från reningsverk som gödningsmedel har studerats i en screeningstudie åt Naturvårdsverket. Huvudsakliga syftet med utredningen är att:

- Insamla miljödata på organiska föroreningar i jord och biota från åkermark gödslad med slam från reningsverk.
- Utvärdera uppmätta halter av organiska föroreningar mot tidigare resultat och modelluppskattningar.

Studien utgörs av ett nationellt program med totalt 48 prover. Prover insamlades från två fältförsök i Skåne: Igelösa och Petersborg, och två privata fält, Taxinge och ”Sörmland”. De tre första fälten representerar åkermark behandlad med slam under lång tid, medan det sista fältet gödslats med slam endast en gång. Huvudsaklig matris var jord (n=42), men även prover av slam (n=2), gröda (n=3) och mask (n=1) har ingått i studien.

Följande ämnesgrupper studerades i samtliga matriser med undantag av mask; ftalater, LAS, alkylfenoler, antibakteriella substanser (triclosan, triclokarban), doftämnen, organofosfater, perfluorerade ämnen (PFOS, PFOA) och bromerade flamskyddsmedel (BDE). I mask studerades myskämnen, perfluorerade ämnen och bromerade flamskyddsmedel.

Undersökningen visar att PFOS, PBDE (47, 99, 100 and 209), galaxolid, tonalid och DEHP anrikas i jord behandlad med slam jämfört med obehandlad jord. Det finns även enstaka indikationer på att halter av DIDP, nonylfenol och TCPP kan öka, men dessa observationer är osäkra eftersom de inte är vanligt förekommande.

Resultaten tyder på att halter i jord av vissa ämnen ökar med ökad slamdos och att påverkan framförallt, men inte uteslutande, sker i de översta jordlagren (0-0,3 m). För många ämnen visar enkla massbalansberäkningar att enstaka slambehandlingar resulterar i så låga halter i jord (sub µg/kg ts) att de inte kan detekteras inom analysmetodens rapporteringsgräns.

Jämförelse mellan modellberäkningar och fältdata visar på bra överensstämmelse när det gäller PFOS och BDE-209 d.v.s. dessa ämnen ackumuleras i jorden över tid vid upprepad slambehandling. De ämnen som generellt inte detekteras, alkylfenoler, dibutylftalat och LAS, stämmer också bra överens med tidigare uppskattningar och mätresultat. Organofosfater detekteras generellt inte, vilket är svårt att utvärdera eftersom rapporteringsgränsen var relativt hög. Den generella förekomsten av galaxolid i slambehandlad jord överensstämmer inte med tidigare modellberäkningar. Om galaxolid ackumuleras i jord eller inte efter upprepad slambehandling kan inte bedömas utifrån nuvarande studie. Tidigare modellberäkningar och nedbrytningsstudier tyder dock på att galaxolid inte ackumuleras.

I kärnor från höstvete provtaget vid Petersborg detekterades ingen av de analyserade organiska ämnena. Dessa resultat överensstämmer väl med tidigare studier. En teoretisk beräkning av halter av undersökta ämnen i grödor visar att förväntade halter är mycket låga

(ng/g vv). För de flesta ämnena är förväntade halter så låga att de inte kan detekteras med analysmetodens rapporteringsgräns. Resultatet stödjer tidigare modellberäkningar och upptagsstudier: ackumulationen av många organiska ämnen är begränsad från jord till gröda

I mask från Taxingefältet påträffades PFOS och PBDE (47, 99 och 100) i detekterbara halter, vilket visar att dessa ämnen kan bioackumuleras i biota. Beräknade bioackumulationsfaktorer (BSAF) från jord till biota ligger i linje med tidigare resultat.

Riskkarakteriseringen för jordekosystem och människor exponerade via intag av grödor visar att halter i jord efter lång tids gödsling med slam inte utgör en risk för jordekosystemet eller människor. Dessa resultat överensstämmer väl med tidigare resultat.

INNEHÅLL

1. INTRODUCTION

Mineral fertiliser is the main phosphorus source for agricultural land in Sweden as well as in many other countries. However, due to expected global shortage of mineable mineral phosphorus (“Peak phosphorus”) there is a need for increased recycling of phosphorus and other nutrients from the society. Sewage sludge is rich in phosphorus and is frequently used as a fertiliser in arable soils.

Sludge that is produced in municipal waste water treatment plants does however contain numerous unwanted pollutants, many of which have persistent, toxic or bioaccumulative properties. Hundreds of pollutants have been identified in sewage sludge at levels from sub-µg/kg to tens of mg/kg. This includes e.g. modern use chemicals such as phthalates, pharmaceuticals, flame retardants, biocides and pesticides, perfluorinated substances, as well as banned chemicals such as PCBs.

The use of sewage sludge as a fertiliser on arable land does thus pose a potential threat to contaminate soils and to negatively affect soil biota, food production and human health. To what extent contamination and risks realises depends on numerous factors such as pollutant levels in sludge, sludge application (rate and frequency), harvest rates etc. The Swedish regulation for sludge application is, in an international perspective, fairly restricted and studies have shown positive effects of sludge application on biological activity in soils (Dahlin et al., 1997; review in Swedish in Sternbeck and Österås, 2013). However, persistent chemicals may accumulate in soils upon repeated applications and Swedish studies have demonstrated increased concentrations of PBDE and PCBs in both soils and earthworms compared to control fields (Matcheko et al., 2002; Sällström et al., 2005).

Risk assessments have been performed for numerous metals and organic chemicals for Swedish conditions (Sternbeck m.fl., 2011; 2013). The assessments were based on mass balance calculations as well as risk assessment methods for soil biota, certain predators and human health. Accumulation in soil and in crops were calculated for a 100 year scenario where sludge was applied every fifth year. The results showed that:

- PFOS, PFDA, PCDD/Fs, PCBs and SCCPs tend to accumulate in soil upon repeated applications of sludge
- Substances such as PAHs, phthalates, nonylphenol and bisphenol A show low or no tendency to long-term accumulation
- The accumulation in crops does not constitute a serious health risk (for those compounds included in the study).

The model calculations are of course subject to uncertainties in scenario, parameter data etc and validation is, as for all modelling efforts, desirable. In Sweden only few data of organic chemicals in sludge amended soils are available (e.g. Matcheko et al., 2002; Sällström et al., 2005; Hörsing et al., 2014). These studies also include data on pollutants in crops or earthworms. Field studies in other countries as well as laboratory experiments do show that certain organic pollutants may accumulate from sludge amended soil to crops (e.g. Hale et al., 2012; Lechner and Knapp, 2011; Macherius et al., 2012; Trapp and Eggen, 2013). Commonly, sludge application rates or sludge pollutant levels were higher in these studies than what is typical in Sweden.

As an assignment from the Swedish Environmental Protection Agency, WSP Environmental has during 2014-2015 performed a national screening investigation of organic pollutants found in sewage sludge in the Swedish environment.

The goals of this study are to:

- Provide data on environmental levels of organic contaminants in soil and biota from arable land amended with sewage sludge.
- Evaluate measured field levels of organic contaminants against previous results and model predictions.

2. PROPERTIES

A summary of physico-chemical properties and ecotoxicity of the studied compounds is presented In Table 1. Due to the large number of compounds, these aspects are not presented in detail in this report. However, it can be mentioned that the very low PNEC value for tricresylphosphate is derived with the EqP method using toxicity data for fish and using an additional assement factor of 10 due to the high lipophilicity (Sternbeck m.fl., 2013).

Table 1. Summary of physico chemical properties and ecotoxicity for the studied compounds.

Group/Substance	K_D arable soils (l/kg)	$T_{1/2}$ in soil (days)	BCF root crops (kg dw soil/ kg ww plant)	BCF cereals (kg soil dw /kg plant ww)	$PNEC_{soil}$ (mg/kg dw)
Phthalates					
DEHP	156000[3]	300[3]	0,00008[3]		13[3]
DIDP	2900-15900[6]	300[6]			100[6]
DINP	2900-15900[7]	300[7]			30[7]
LAS					
C10-C14 Linear alkylbenzene sulfonic acid	50[3]	8[3]	0,14[3]		35[3]
Alkylphenols					
4-tert-octylphenol	33[3]	10[3]	0,33[3]		0,0067[3]
4-iso-nonylphenol	139[3]	10[3]	0,083[3]		0,3[3]
OP1EO		5[3]			
NP1EO		5[3]			
Antibacterial substances					
Triclosan	178-264[8]	120[4]			0,196[5]
Triclocarban	763-1187[8]	120[4]			0,0373[5]
Musk substances					
Galaxolide	1600[1]	150[1]	0,096[1]	0[1]	0,31[1]
Tonalide	1900[1]	150[1]	0,074[1]	0[1]	0,31[1]
Galaxolide lactone					

Group/Substance	K_D arable soils (l/kg)	$T_{1/2}$ in soil (days)	BCF root crops (kg dw soil/ kg ww plant)	BCF cereals (kg soil dw /kg plant ww)	$PNEC_{soil}$ (mg/kg dw)
Organophosphates					
Tricresylphosphate	120[1]	30[1]			0,00039[1]
Tris(2-chloroisoproyl)phosphate	5[1]	180[1]	2,8[1]	0,095[1]	1,7[1]
Tris(2-butoxyethyl)phosphate					
2-Ethylhexyl diphenyl phosphate	250[2]	300[1]	0,04[1]		0,044[1]
Perfluoroalkyls					
PFOS	57[1]		0,0009/0,04[1, 2]	0,0086[1]	0,01[1]
PFOA					0,016[7]
Brominated flame retardants					
BDE-209	52000[3]	3200[3]	0,00025[3]		>98[3]
BDE-47	5720[3]	385[3]	0,0023[3]		
BDE-99	13000[3]	707[3]	0,001[3]		0,38[3]
BDE-100		707[3]			

[1] Sternbeck et al 2013. [2] Potato/carrot.

[3] Sternbeck et al 2011. Kd calculated from Koc with an assumed organic matter content of 2,6 %.

[4] Remberger et al 2014.

[5] © European Chemicals Agency, 2007-2015.

[6] EC, 2003a. Kd calculated from Koc with an assumed organic matter content of 2,6 %.

[7] EC, 2003b. Kd calculated from Koc with an assumed organic matter content of 2,6 %.

[8] Wu et al, 2009.

3. PREVIOUS ENVIRONMENTAL STUDIES

This chapter gives a brief description of previous studies and gives examples of observed concentrations in sludge amended soil, earthworms and crops. Impact on surface water quality would in theory also be possible, depending on local hydrological conditions. In the literature search, however, we found no reports on this subject.

3.1. Soil

Whether or not certain pollutants accumulate in soils upon repeated applications of sludge is a key issue. This aspect has been investigated in several field studies where sludge has been applied to arable soils. Result from some of these studies, where accumulation was confirmed in soil, is summarized in Table 2. Substances shown to accumulate in soil on a long-term basis are persistent organic substances such as PBDE, PCB and PFOS. Shortly after sludge amendment less persistent substances such as the musk substances galaxolide and tonalide have also been found in soil.

Table 2. Examples of concentrations in soil that were repeatedly amended with sewage sludge. Only data from studies where accumulation was confirmed is included, and no corrections for background concentrations were performed. All concentrations in dry weight.

Group/Substance	Accumulation confirmed	Concentrations	Country	Reference
PBDE (47, 99, 100)	Yes	PBDE47: 0,026-0,30 μg/kg PBDE99: 0,03-0,35 μg/kg	Sweden	Matcheko et al, 2002
PBDE-209	Yes	0,028-1,0 μg/kg	Sweden	Sällström et al 2005
PCB	Yes	PCB153: 0,15-0,82 μg/kg	Sweden	Matcheko et al, 2002
Galaxolide	Yes	4-15 μg/kg; 25-75-percentile	Sweden	SWECO, 2010
Tonalide	Yes	4,7-22 μg/kg; 25-75-percentile	Sweden	SWECO, 2010
Perfluorinated sulfonic acids	Yes	PFOS: 80-219 μg/kg MeFOSAA: 63-143 μg/kg	USA	Sepulvado et al., 2010
Perfluorinated carboxylic acids	Yes	PFOA: 8-68 μg/kg	USA	Sepulvado et al., 2010

Several studies have also demonstrated that many non-halogenated substances (e.g. LAS, nonylphenol and DEHP) that occur at high concentrations in sewage sludge, do not accumulate in soil upon repeated sludge amendments (Table 3). Nevertheless those substances are commonly present in soil during the first months or the first year following amendment, but their concentrations progressively decline during this period.

Table 3. Substances that did not show evidence of long term accumulation in sludge treated soils**.**

Reference	LAS	Nonylphenol	DEHP	Triclosan
Landesanstalt für Umweltschutz, Baden-Württemberg, 2003	x	x	x	
Petersen et al., 2003	x	x	x	
Xia et al., 2010		x		x

In Sweden a long-term field study with sewage sludge amendment has been performed in Skåne county (eg. Andersson, 2012). Fields at this site are also included in the present study. In this field, accumulation of PBDE and PCB has been shown (Matcheko et al 2002, Sällström et al 2005). A recent Swedish study in the same field could not detect PAH, octylphenol, nonylphenol, triclosan, PFOS, PFOA or bisphenol A in soils that were amended with sludge for the past 30 years (Hörsing et al., 2014). Analytical reporting limits for the alkylphenols were similar to those in the present study, whereas they were higher for PFOS, PFOA and triclosan.

Musk substances in Swedish arable soil that were fertilised with sludge were studied by SWECO (2010). Sampling after ploughing, within weeks after application, showed the presence of galaxolide, the degradation product galaxolide lactone, and tonalide. However, none of the substances could be detected (i.e. <1-2 µg/kg dw) in samples taken one year after the sludge was applied. This suggests degradation. Laboratory studies have also shown rapid degradation of galaxolide in sludge amended soil (DiFrancesco et al., 2004).

3.2. Plants

There are few field studies performed were organic pollutants have been examined in crops after sludge amendment. Most studies on plant uptake have also been performed with higher sludge additions than what are representative for Swedish legislation. The uptake of organic pollutants in plants grown in soil treated with sewage sludge was recently reviewed within an assignment for the Swedish Environmental Protection Agency (Sternbeck and Österås, 2013). In Table 4 results from this review are summarized. The few studies performed, with sludge additions representative for Swedish legislation, show no uptake of organic substances in crops.

Table 4. Summary of field studies with sewage sludge where plant uptake of organic substances has been studied.

Country	Sweden	Sweden	Denmark	USA
Crops	Sugar beets	Wheat, Rape	Carrot Barley	Maize
Sludge addition	0-12 ton dw/ha	0 and 4 ton dw/ha	0-90 ton dw/ha	0-8,5 ton dw/ha
Cultivation after amendment	3 years	1 and 4 years	Directly	Directly
Yield	2011	1990-1993	2000	2004-2007
Substances analysed	PCB7, PAH16, Bisphenol-A, triclosan, octyl- and nonylphenol, PFOS, PFOA	BTEX, dioxins, PCB7	DEHP, LAS, nonylphenol	PBDE
Results crops	Increase in octyl-and nonylphenol at the highest sludge addition in combination with mineral fertilizer	No differences., PCB < 0,2 mg/kg dw	Generally no correlation between concentrations in crop and sludge addition	No PBDE detected in crops
Ref.	Hörsing et al, 2014	Malmöhus läns Hushållningssällskap, 1994	Laturnus et al, 2007	Hale et al, 2012

Within the Skåne field experiment organic pollutants (BTEX, PCB and dioxins) have been examined in crops (spring and autumn wheat and autumn rape) yielded in 1990 and 1993 (one respectively four year after sludge treatment). No differences in levels of the examined substances between sewage sludge treated (4 t dw/ha/year) and control fields could be observed.

Organic pollutants (alkylphenols, PAH, PCB, PFOS, PFOA and triclosan) were examined in soil and sugar beets from the field experiments in Skåne in 2011 (Hörsing et al, 2014). The last treatment with sewage sludge on the sampled fields was performed in 2009. At the highest sewage sludge addition (12 tons/ha) in combination with mineral fertiliser, 4-nonylphenol and 4-octylphenol were detected in sugar beets at 3,9 and 35 µg/kg dw, respectively. It was concluded that organic substances with surface-active properties, e.g. certain alkylphenols, can be assimilated in crops like sugar beets. It was also concluded that the source of these substances most probably is sewage sludge and that the uptake is stimulated by addition of mineral fertilizers together with sewage sludge.

Sternbeck and Österås (2013) also reviewed other studies and compilations for plant uptake of organic substances. Of some of the studied substances in this report the following was found:

- Brominated flame retardants and musk substances – the accumulation in plants tend to be low.
- Perfluoroalkyls – can accumulate in plants.
- Triclosan – can accumulate in plants, also as conjugates.

3.3. Earthworms

Examples of concentrations in earthworms from field collected sludge treated soils are given in Table 5. The general theory is that concentrations in the worms are in equilibrium with those in soil. A biota soil accumulation factor (BSAF) can thus be calculated from empirical data. BSAF values are relevant when comparing different studies or different compounds because the influence of varying soil concentrations is accounted for. BSAF values, when present, are also given in Table 5.

Table 5. Examples of pollutant concentrations in earthworms and biota to soil accumulation factors (BSAF). Please note that different units are used for different substance groups. OM = soil organic matter or ignition loss.

Substance	Concentration	Unit conc.	BSAF	Unit BSAF	Country	Reference
BDE47	7,9-37	ng/g lw	5	OM/lw	Sweden, Skåne	Sellström et al, 2005
BDE 99	14-38	ng/g lw	4,2	OM/lw	Sweden, Skåne	Sellström et al, 2005
BDE 100	2,7-9,6	ng/g lw	4,6	OM/lw	Sweden, Skåne	Sellström et al, 2005
BDE 209	3,7-7,1	ng/g lw	0,3	OM/lw	Sweden, Skåne	Sellström et al, 2005
Galaxolide	3340, 131	µg/kg dw	0,05-3,1	dw/dw	USA	Kinney et al, 2008
Tonalide	279, 75	µg/kg dw	0,1-1	dw/dw	USA	Kinney et al, 2008
PFOS	683	ng/g ww	18	dw/dw	USA	Rich et al, 2015
PFOA	4,76	ng/g ww	2,1	dw/dw	USA	Rich et al, 2015

4. SAMPLING STRATEGY AND STUDY AREAS

The study consists of a national program financed by the Swedish EPA. The program aims at giving an overview of the occurrence of organic pollutants in arable land treated with sewage sludge. Soils from arable land subjected to different intensity of sludge treatment were sampled to study differences in accumulation of organic substances over time. The dispersion of and elimination of contaminants from soil was also addressed by sampling of soil at different soil depth and time after sludge application. Furthermore, the possible up-take of organic contaminants in biota and crop was within the scope of the investigation. To meet this aim, crop samples and earthworms were collected.

Samples were collected from two experimental arable fields in Skåne: Igelösa and Petersborg, and two private farm fields in Sörmland: Taxinge and a farm which, due to requests of confidentiality, will be termed with the fictional name "Sörmland" throughout the report. The total number of samples per matrix is summarised in Table 6. In total 48 samples were analysed. A complete sample list is given in appendix 2. Further details about the fields and samples collected at the different sites are outlined below.

Table 6. Total number of samples.

Matrix	Number of samples
Sewage sludge	2
Topsoil (0-0,2/0,3 m)	23
Subsoil (0,2/0,3-0,4/0,6 m)	19
Plants	3
Earthworms	1

4.1. Skåne field experiment

The Skåne field experiment includes two fields: Igelösa and Petersborg, where long term field studies of the effects of sludge amendment on arable land have been conducted since year 1981 (e.g. Andersson, 2012). Satellite pictures from the two fields are presented in Figure 1. The total area of each field is 36 x 120 m (4320 m^2). The two fields are equally divided into 36 sections. Every section is 6 x 20 m (120 m^2). In each section a defined amount of sludge: 0 (control), 4 or 12 tons dw/ha, has been applied repeatedly every fourth year since the experiments started. The latest sludge application was in August year 2013 (SLU, 2013). A schematic picture of the design of the two fields is presented in Table 7.

The soil characteristics in the two fields are:

- Igelösa: 3-6 % organic matter, 25-40 % clay, pH 7,0
- Petersborg: 2-3 % organic matter, 15-25 % clay, pH 6,8

The sludge delivered to Igelösa was from Källby STP in the city of Lund and the sludge applied in Petersborg was from the Sjölunda STP in Malmö.

The two fields are cultivated according to a field specific crop rotation. In year 2014 the crops were winter wheat in Petersborg and winter oilseed rape in Igelösa.

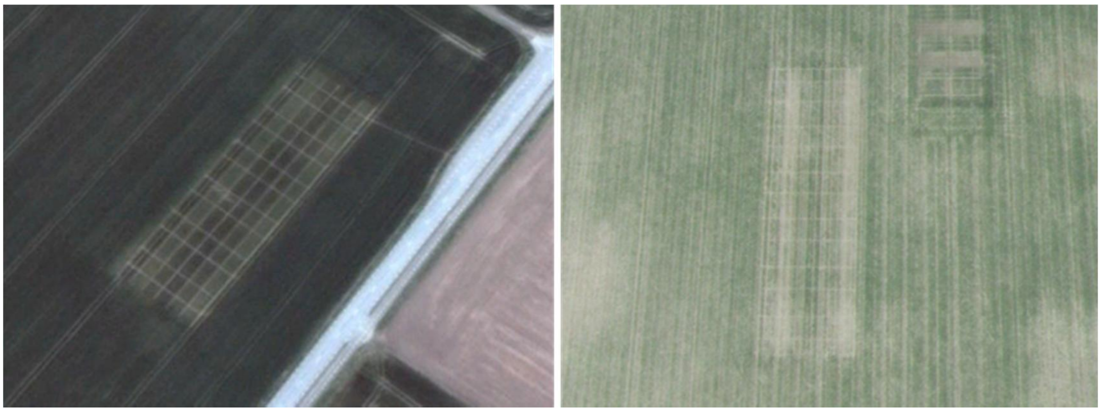


Figure 1. Satellite pictures of the two experimental fields in Skåne. Left: Igelösa (Source: Google earth, Digital globe). Right: Petersborg (Source: Google earth, Lantmäteriet/Metria).

In the present study soil was sampled from five sections in each of the Skåne fields as shown in Table 7. Soil from untreated sections (control) were sampled from 0-0,3 m depth. Soil from sections receiving 4 and 12 tons dw sludge/ha were sampled from 0-0,3 and 0,3-0,6 m depth.

Table 7. Schematic design of the study fields in Skåne. Each numbered square (1-36) represents a field section of 6x20 m, which is fertilised with the combination of sludge and mineral NPK (N=nitrogen, P=phosphorus, K=potassium) fertiliser given by the first column and header row. The squares highlighted in grey marks the sections sampled in the present screening study.

NPK fertiliser[1]	Sludge addition rate (tons/ha) every fourth year					
	0	4	12	4	12	0
None	1	7	13	19	25	31
PK, N½	2	8	14	20	26	32
NPK	3	9	15	21	27	33
PK, N½	4	10	16	22	28	34
None	5	11	17	23	29	35
NPK	6	12	18	24	30	36

[1] PK, N ½ = PK to crop demand, N half of crop demand; NPK = NPK to crop demand.

Sampling of soil was performed on two occasions after sludge application, first 2014-04-08 in both fields, and later 2014-07-27 and 2014-07-29 in Petersborg and Igelösa respectively. The sampling was hence approximately eight and twelve months after sludge was applied to the field. The second sampling was following a whole year including crucial events such as main crop growth, longer period of leaching or runoff with precipitation and the higher temperature span during summer, all of which provide conditions favoring the elimination of contaminants from soil.

Originally it was intended to sample the sewage sludge that was applied to the fields. However, this was not possible since sludge had already been applied when this study was initiated, and no sludge had been sampled and saved from the applied batches.

In Petersborg crop samples consisting of winter wheat grain were taken from each sludge application level 0, 4 and 12 tons dw/ha. Unfortunately, due to poor plant establishment and crop failure in Igelösa, the intended sampling of winter oilseed rape could not be performed.

The detailed sampling designs for Igelösa and Petersborg are shown in Table 8 and Table 9. At each location soil was sampled in one control section, two sections of each 4 and 12 tons/ha every fourth year, and at two occasions after sludge addition (8 and 12 months). At the last occasion of soil sampling at Petersborg, winter wheat grain was also sampled.

Table 8. Sampling design at Petersborg. S=soil, P=plant. Last sludge application was in August 2013.

Sludge rate (tons/ha/4years)	Section (#)	Sampling depth (m)	Sludge application	Sampling time and matrix		
			2013-08	2014-04-08	2014-07-27	
0	3	0-0,3		S	S	P
4	9, 12	0-0,3		S	S	P
		0,3-0,6		S	S	
12	15, 18	0-0,3		S	S	P
		0,3-0,6		S	S	

Table 9. Sampling design at Igelösa. S=soil. Last sludge application was in August 2013.

Sludge rate (tons/ha/4years)	Section (#)	Sampling depth (m)	Sludge application	Sampling time and matrix	
			2013-08	2014-04-08	2014-07-29
0	3	0-0,3		S	S
4	9, 12	0-0,3		S	S
		0,3-0,6		S	S
12	15, 18	0-0,3		S	S
		0,3-0,6		S	S

The Skåne field experiment gives a possibility to examine the occurrence of sludge contaminants in soil and crops in a long term sludge amended area with two different application rates of sludge as well as a control field.

4.2. Taxinge

The Taxinge field has a long history of sludge amendment. According to personal information from Per Fimmerstad at the Taxinge farm, the field has received sludge every sixth year since the late 1970s. During the last decade the field has received sludge twice, the latest application was in year 2012. The total amount of sludge applied to the field during the last ten years was 220 kg P/ha, which approximately equals 4 tons sludge per ha and application. No details about the long term application rate of sludge expressed as tons dw/ha were obtained. A satellite picture of the sampled area and the view over the sampling spot is presented in Figure 2.

Figure 2. A satellite picture (Source: Google earth, DigitalGlobe) (left) and the view over the sampling area (right) at the Taxinge field. The sampling area is marked as a rectangle (left).

The soil at Taxinge is clayey. No other details about soil characteristics were obtained for this field.

The sludge delivered to the field the last two occasions was from Käppala STP in the municipality of Lidingö.

In the Taxinge field, soil was sampled in 2015-04-09, approximately three years after the latest sludge application (Table 10). One sample was prepared from each of the soil depth 0-0,2 and 0,2-0,4 m.

A sample of more than 100 earthworms was obtained from the 0-0,2 m soil depth from the whole sampling area (Figure 2).

The Taxinge field provides a possibility to examine the occurrence of sludge contaminants in soil and biota in a long term, and by farming practice, sludge amended area.

Table 10. Sampling design at the Taxinge field. S=soil, E=Earthworms. Last sludge addition was in 2012, approximately 3 years before sampling.

Sampling depth (m)	Sludge application	Sampling time and matrix
	2012	**2015-04-09**
0-0,2		S, E
0,2-0,4		S

4.3.Sörmland

The Sörmland field was subjected to its first sludge application ever during the course of the present study. Sludge was delivered and piled in a stack along the border of the field 2013-08-16 and 2013-08-23. The stack was covered with plastic film (Figure 3). Thereafter the sludge was stored in the stack during one year until it was applied to the field in September 2014. The sludge application rate was 2,6 tons dw/ha.

Sludge samples from the time of delivery to the field stack and after one year storage in the field stack was examined. This was made possible since samples of sludge that was delivered to the field, before this study was initiated, had been sampled and stored frozen from the time it was generated at the STP. Sludge from the stack stored in field was sampled 2014-09-03 a few days before it was applied to and mixed into the field soil.

Figure 3 The sludge stack at the Sörmland field after one year of in-field storage.

The soil characteristics of the Sörmland field are: 4 % organic matter, ca 25 % clay, pH 6,7.

Soil was sampled before (2014-09-03) and after sludge application (2014-09-15). The soil was sampled at depth 0-0,2 and 0,2-0,4 m.

Table 11. Sampling design at the Sörmland field. S=soil, Z=sludge. Sludge was applied approximately one week before the second sampling of soil.

Sampling depth (m)	Date of sampling and sludge addition, and matrix		
	2014-09-03	Sludge application 2014-09	2014-09-15
0-0,6 (in sludge stack)	Z		
0-0,2	S		S
0,2-0,4	S		S

The Sörmland field provides an opportunity to study the content of sludge contaminants in soil before and after the first sludge amendment to a field. Also the level of contaminants in soil in relation to the concentration of contaminants in sludge applied to the field may be studied. Furthermore the level of contaminants in sludge before and after one year of storage in field is examined, giving a picture of the degree of possible elimination of compounds from the sludge during such handling.

5. METHODS

5.1. Sampling

Soil, sludge and crop samples were collected in glass jars with plastic lids sealed with a Teflon lining, provided by the analysing laboratory ALS Scandinavia. For samples intended for analysis of fluorinated compounds the plastic lids were without Teflon lining. Earthworms were collected in a plastic diffusion resistive Rilsan bag for soil samples.

5.1.1. Skåne field experiment

The sampling of soil and crops in Skåne was performed by staff at Hushållningssällskapet in Malmöhus county. Soil was sampled using a handheld soil drill made of steel. WSP designed the sampling strategy and provided thorough instructions regarding the sampling procedures.

Each soil sample from 0-0,3 m was prepared as a composite sample of 5 subsamples and each soil sample from 0,3-0,6 m was prepared from 3 subsamples.

Winter wheat grains was sampled as composite samples from the sections 3 (control), 9+12 (4 tons/ha) and 15+18 (12 tons/ha) shown in Table 7.

5.1.2. Private farmers

The sampling of soil at the private farmers in Taxinge and Sörmland was performed by staff from WSP. Sampling of soil was performed using a handheld larger steel shovel for digging holes and a smaller steel spade for collecting soil samples.

Each soil sample from the two fields was prepared from 12 subsamples of soil thoroughly mixed in a diffusion resistive Rilsan sample bag before collected in glass jars delivered from the laboratory as described in 5.1.

Sludge applied to the Sörmland field was sampled daily by staff at the STP during the production of the batch delivered to the field stack. Subsamples were stored in refrigerator until the sludge batch was delivered; thereafter composite samples were prepared and stored frozen at the analyzing laboratory. A composite sample of sludge from the batches was prepared for the present study by staff at the STP. Sludge stored one year in the field stack was sampled at site by staff from WSP. Sludge samples were taken from the stack using a steel core sampler and transferred directly to a glass jar. A composite sample was prepared from twelve subsamples from the stack.

Earthworms from Taxinge were picked while wearing one-use nitrile gloves. In total >100 earthworms amounting to a total weight of ~110 g were collected in a Rilsan bag. There was no determination of species of the earthworms. The minimum size of the worms was approximately 3 cm.

The worms were carefully washed from soil, organic debris and castings with cold tap water approximately one hour after sampling. Thereafter the earthworms were transferred to a new Rilsan bag and kept without food substrate in a refrigerator for eight days. Twice daily, the earthworms were gently rinsed from freshly excreted worm castings with cold tap water. At each occasion, the apparent visual status of the earthworms was checked to ensure the worms were kept alive. No earthworm was identified as dead. When the production of worm castings had stopped, as judged by the visible content of castings in the Rilsan bag, the whole sample of earthworms were frozen to -18 °C and sent to the laboratory for chemical analysis on a fresh weight basis.

During the rinsing period the total sample weight decreased from ~110 g to 100 g, which is attributed to the excretion of gastrointestinal tract content from the worms.

5.2. Chemical analysis

Chemical analyses were performed by ALS Scandinavia in cooperation with GBA Germany. Reporting limits for the different media are presented in Appendix 1.

Samples are homogenized and a representative amount (see below) for each analysis is taken out. For PFC, LAS and BFR samples are dried at room temperature prior to analysis.

The methods for the different substances are given below.

- Phthalates: The sample amount was 2 g dry matter. Internal standard is added to the sample and extracted with ethylacetate. The sample is centrifuged and analyzed via GC-MS.
- Alkylphenols: The sample amount was 10 g dry material. Internal standard is added to the sample and the sample is extracted with NaCl/Acetone/Hexane. After extraction the sample is cleaned up on a silica gel column. The measurement is performed on GC-MS.
- Triclosan and Triclocarban: The sample amount was 5 g dry material. Internal standard is added to the sample and extracted with methanol. Acetic anhydride is added, extracted with hexane and analyzed via GC-MS.
- LAS: The sample amount was 5 g dry material. Internal standard is added to the sample and extracted with methanol. After centrifugation the sample is analyzed via HPLC.
- Musk compounds: The sample amount was 10 g original sample. Internal standard is added to the sample and extracted with water/acetone/hexane. The sample is cleaned up over silica gel and analyzed via GC-MS.
- Organophosphates: The sample amount was 2 g original sample. Internal standard is added to the sample and extracted with acteone/water/cyclohexane and analyzed via GC-MS.

- Perfluoroalkyls: The sample amount was 2 g dry material. Internal standard is added to the sample and extracted with methanol. After a SPE enrichment step the sample is analyzed via LC-MS.
- Brominated flame retardants: The sample amount was 2,5 g dry matter. Internal standard is added to the sample and extracted with toluene via soxhlet extraction and treated with cyclohexane/$NaSO_4$. The sample is cleaned up with H_2SO_4 and analyzed via GC.

5.2.1. Quality assurance

All sample jars were provided by the laboratory. At each analytical event, control samples are also used. Generally, these were internationally certified reference materials and laboratory blanks. No laboratory blank problems were observed.

6. RESULTS

This section presents a general overview of the levels of the studied compounds for each matrix. Sample details are given in Appendix 2 and all data are presented in Appendix 3. A discussion on spatial trends, environmental partitioning and possible risks to the health and environment is given in chapter 7.

In total the national program covered 48 samples. Table 12 summarizes which groups of compounds that were analysed and found at detectable concentrations in the different matrices included in this study.

Table 12. Overview of the compound groups detected (x) or non-detected (-) in the different matrices in the present study. NA=not analysed.

Substance group	Sludge (n=2)	Soil (n=42)	Crop (n=3)	Earthworm (n=1)
Phthalates	x	x	-	NA
Linear alkylbenzene sulfonates	x	-	-	NA
Alkylphenols	x	x	-	NA
Antibacterial compounds	x	-	-	NA
Musk compounds	x	x	-	-
Organophosphates	x	x	-	NA
Perfluoroalkyls	x	x	-	x
Polybrominated diphenyl ethers	x	x	-	x

6.1. Sewage sludge

The result from the sludge analysis is presented in Table 13. In total 29 different compounds were detected and 21 compounds were reported as below the reporting limit. The concentrations of detected compounds were comparable to those reported previously in Swedish sludge (Sternbeck et al, 2013).

Sludge was sampled both at the time for delivery to field stack and after one year storage in the field stack just before it was applied to the field. The remaining percentage of each compound in relation to its initial concentration at the STP is displayed in Table 13. Phthalates and musk compounds showed the highest degree of elimination (> 80%) between the two sampling occasions. Approximately half of the compounds were eliminated to less than 60 % of its initial concentration after one year storage.

LAS showed negligible concentration change during the storage, although these substances are generelly considered readily biodegradable. The lack of degradation is probably due to anaerobic conditions within the stack, since LAS is much more persistent under those conditions.

Some compounds and perhaps most notably PBDEs, showed higher concentrations after storing of sludge in field than its initial concentrations. It is not possible to assess whether or not this is due to sampling and/or analytical variability, enrichment due to degradation of organic material in the sludge or the formation of the compounds in the sludge stack.

Table 13. Concentration of compounds (mg/kg dw) in Sludge samples from Sörmland. All data with concentrations above the reporting limit in any of the two sludge samples are included. The remaining percentage (%) of the initial concentration after one year storage in the field is also presented.

Substances	Concentrations		Percentage (%) of initial concentration after one year in field stack
	Initial 2013-08-(12 – 23)	After one year 2014-09-03	
dw_105°C	27,7 %	26,6 %	
di-isobutylphthalate	0,43	<0,060	<14%
di-(2-etylhexyl)phthalate	87	9,2	11%
di-isodecylphthalate	<10	5,3	
di-isononylphthalate	76	9,1	12%
LAS C10	25	13	52%
LAS C11	88	72	82%
LAS C12	110	100	91%
LAS C13	78	100	128%
LAS C14	<10	1,5	
LAS C10-C14, sum	300	290	97%
4-tert-octylphenol	0,44	0,25	57%
4-tert-OF-monoethoxylate	0,058	0,031	53%
4-nonylphenols	8,3	5,8	70%
4-NF-monoethoxylate	1,2	0,58	48%
4-NF-diethoxylate	0,15	<0.11	<73%
triclosan	1,3	0,76	58%
galaxolide	14	1,7	12%
tonalide	3,5	0,16	5%
galaxolide lactone	2,0	0,53	27%
TCPP	0,52	0,75	144%
TDCP	0,061	<0.050	<82%
TEHP	0,37	0,23	62%
EHDPhP	0,36	0,2	56%
PFOS	0,022	0,0095	43%
PFOA	0,0014	0,0015	107%
BDE 209	0,28	0,82	293%
BDE 47	0,020	0,022	110%
BDE 99	0,0072	0,021	292%
BDE 100	0,0023	0,0049	213%

6.2. Soil

6.2.1. Igelösa and Petersborg, Skåne

Twelve different compounds were detected in at least one sample from the two examined fields and two sampling occasions. The results for these compounds are presented in Table 14 and Table 15. However, the majority of the analysed compounds were below the reporting limit in all analysed soil samples from Igelösa and Petersborg.

In control fields no compound was detected in either Igelösa or Petersborg at the first sampling occasion in April 2014. At the second sampling, performed in August 2014, detectable levels of di-isodecylftalat (DIDP), galaxolide and tonalide were found in Igelösa, and tonalide in Petersborg.

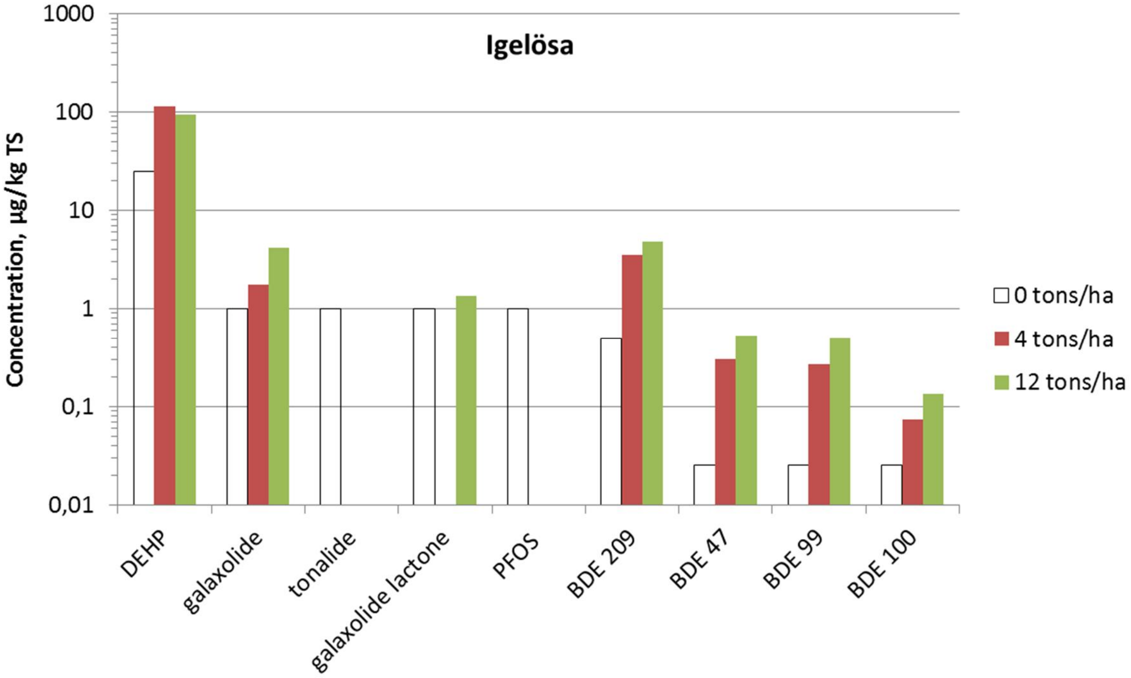
In soil receiving 4 or 12 tons sludge/ha, brominated flame retardants (BDE209, 47, 99 and 100), musk compounds (galaxolide, tonalide and galaxolide lactone) and DEHP were frequently found in detectable concentrations. PFOS was found primarily in soil from fields receiving 12 tons/ha. The levels of the above mentioned compounds found in topsoil at the first sampling occasions in Igelösa and Petersborg are given in Figure 4. In addition to the above mentioned compounds, each of the compounds DIDP, 4-nonylphenol and tris(chloroisopropyl)phosphate (TCPP) were found in detectable amounts once in separate soil samples from sludge amended fields.

In general, the levels of DEHP, galaxolide, galaxolide lactone, PFOS and BDE were higher in sludge amended fields than in control fields, where most of the compounds were below the reporting limit. The levels of these substances were thus estimated to be at least 2-3 times higher in sludge amended fields than in control fields. For tonalide data from the second sampling event had to be excluded due to contamination problems. With these data excluded, levels found at Petersborg were slightly higher than the control.

The concentrations of contaminants are generally higher in soils amended with 12 tons sludge/ha than in soils with 4 tons/ha. In those cases where a comparison of soil concentrations was possible, the BDE concentrations were a factor of 2-3 higher in the 12 tons/ha sections than in the ones receiving 4 tons/ha.

All detectable compounds except for DEHP showed a tendency of occurring in higher concentrations in the upper (0-0,3 m depth) than in deeper soil layers (0,3-0,6 m depth). For DEHP there was no clear distribution with soil depth.

In general, the concentrations of substances in soil were higher 12 months after sludge application than 8 months after application. This may be due to analytical uncertainty, since soil samples collected after 8 and 12 months were analysed at different occassions. True variability due to heterogenity could possibly also contribute.



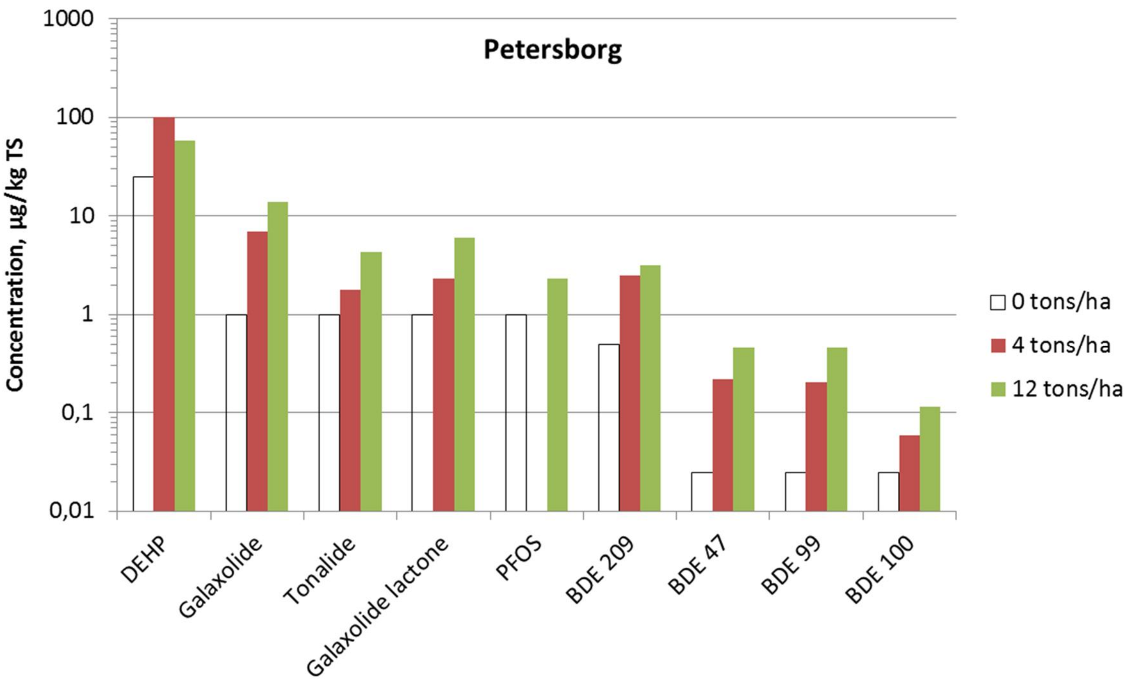


Figure 4. Levels of different organic compounds in topsoil (0-0,3 m) of fields treated with different doses of sewage sludge at Igelösa and Petersborg 8 months after treatment. In control field (0 tons/ha) where none of the compounds were detected half the detection limit is shown as comparison.

Table 14. Concentrations (µg/kg dry weight) of compounds in soil from Igelösa, Skåne. Grey shading highlights concentrations above the reporting limit. The table includes all compounds found in detectable amounts in any of the analysed soil samples from the two Skåne research fields. The field area ID denotes a number specific to different field areas in the long term Skåne experiments, which are continuously amended with a given sludge level (tons/ha). The sludge application rate refers to the amount of sludge (tons/ha) applied to the sampled field area every fourth year.

Igelösa	**Sludge application rate**	**0 tons/ha**	**4 tons/ha**				**12 tons/ha**			
Month after	**Sample depth (m)**	**0,0-0,3**	**0,0-0,3**		**0,3-0,6**		**0,0-0,3**		**0,3-0,6**	
sludge application	**Field area ID**	**I_3**	**I_9**	**I_12**	**I_9**	**I_12**	**I_15**	**I_18**	**I_15**	**I_18**
8	DEHP	<50	89	140	<50	230	77	110	100	130
12	DEHP	<50	<50	<50	<50	140	89	74	<50	<50
8	DIDP	<2500	<2500	<2500	<2500	<2500	<2500	<2500	<2500	<2500
12	DIDP	3100	<2500	<2500	<2500	<2500	5200	<2500	<2500	<2500
8	4-nonylphenol	<10	<10	<10	<10	<10	<10	<10	<10	<10
12	4-nonylphenol	<10	<10	<10	<10	<10	<10	<10	<10	<10
8	galaxolide	<2,0	<2,0	2,5	<2,0	<2,0	2,8	5,6	<2,0	<2,0
12	galaxolide	2,6	4,6	5,1	<2,0	<2,0	12	11	<2,0	<2,0
8	tonalide	<2,0	<2,0	<2,0	<2,0	<2,0	<2,0	<2,0	<2,0	<2,0
12	tonalide	28	6,5	5,4	<2,0	<2,0	13	18	4,4	<2,0
8	galaxolide lactone	<2,0	<2,0	<2,0	<2,0	<2,0	<2,0	2,7	<2,0	<2,0
12	galaxolide lactone	<2,0	2,7	4,5	<2,0	<2,0	15	5,9	4,6	<2,0
8	TCPP	<20	<20	20	<20	<20	<20	<20	<20	<20
12	TCPP	<50	<50	<50	<50	<50	<50	<50	<50	<50
8	PFOS	<2,0	<2,0	<2,0	<2,0	<2,0	<2,0	<2,0	<2,0	<2,0
12	PFOS	<2,0	2,5	2,0	<2,0	<2,0	3,3	3,7	2,3	2,0
8	BDE 209	<1,0	2,9	4,2	3	1,6	5,8	3,8	1,1	2,2
12	BDE 209	<1,0	6	4,5	2,1	1	14	12	16	4,6
8	BDE 47	<0,050	0,3	0,31	0,10	0,10	0,54	0,51	0,19	0,18
12	BDE 47	<0,050	0,21	0,39	0,11	0,085	0,39	0,33	0,2	0,17
8	BDE 99	<0,050	0,27	0,27	0,083	0,084	0,52	0,49	0,19	0,17
12	BDE 99	<0,050	0,2	0,37	0,12	0,088	0,42	0,35	0,23	0,21
8	BDE 100	<0,050	0,076	0,072	<0,050	<0,050	0,14	0,13	0,051	<0,050
12	BDE 100	<0,050	<0,050	0,068	<0,050	<0,050	0,094	0,069	<0,050	<0,050

Table 15. Concentrations (µg/kg dw) of compounds in soil from Petersborg, Skåne. Grey shading highlights detected compounds. The table includes all compounds found in measurable amounts in any of the analysed soil samples from the two Skåne research fields. The field area ID denotes a number specific to different field areas in the long term Skåne experiments, which are continuously amended with a given sludge level (tons dw/ha). The sludge application rate refers to the amount of sludge (tons dw/ha) applied to the sampled field area every fourth year.

Petersborg	**Sludge application rate**	**0 tons/ha**	**4 tons/ha**				**12 tons/ha**			
Month after	**Sampling depth (m)**	**0,0-0,3**	**0,0-0,3**		**0,3-0,6**		**0,0-0,3**		**0,3-0,6**	
Sludge application	**Field area ID**	**P_3**	**P_9**	**P_12**	**P_9**	**P_12**	**P_15**	**P_18**	**P_15**	**P_18**
8	DEHP	<50	110	92	<50	<50	92	<50	100	65
12	DEHP	<50	71	<50	<50	<50	<50	<50	<50	<50
8	DIDP	<2500	<2500	<2500	<2500	<2500	<2500	<2500	<2500	<2500
12	DIDP	<2500	<2500	<2500	<2500	<2500	<2500	<2500	<2500	<2500
8	4-nonylphenol	<10	<10	<10	<10	<10	16	<10	<10	<10
12	4-nonylphenol	<10	<10	<10	<10	<10	<10	<10	<10	<10
8	Galaxolide	<2,0	13	<2,0	<2,0	<2,0	10	18	<2,0	<2,0
12	Galaxolide	<2,0	<2,0	<2,0	<2,0	<2,0	26	9,9	24	70
8	Tonalide	<2,0	2,6	<2,0	<2,0	2,8	2,9	5,7	2,9	<2,0
12	Tonalide	16	7,6	8,2	2,9	10	29	7,1	25	41
8	Galaxolide lactone	<2,0	3,7	<2,0	<2,0	<2,0	5,8	6,2	<2,0	<2,0
12	Galaxolide lactone	<2,0	<2,0	<2,0	<2,0	<2,0	2,3	<2,0	3,5	13
8	TCPP	<20	<20	<20	<20	<20	<20	<20	<20	<20
12	TCPP	<50	<50	<50	<50	<50	<50	<50	<50	<50
8	PFOS	<2,0	<2,0	<2,0	<2,0	<2,0	2,0	2,7	<2,0	<2,0
12	PFOS	<2,0	<2,0	<2,0	<2,0	<2,0	5,1	3,7	2,4	2,3
8	BDE 209	<1,0	3,6	1,4	<1,0	<1,0	3,5	2,8	1,2	1,2
12	BDE 209	<1,0	2,1	1,7	1,5	2	5,9	5,1	3,3	2,8
8	BDE 47	<0,050	0,21	0,23	0,089	<0,050	0,51	0,4	0,13	0,14
12	BDE 47	<0,050	0,1	0,12	0,061	0,054	0,24	0,26	0,14	0,14
8	BDE 99	<0,050	0,19	0,22	0,078	<0,050	0,5	0,41	0,13	0,13
12	BDE 99	<0,050	0,098	0,11	0,071	0,053	0,26	0,28	0,16	0,17
8	BDE 100	<0,050	0,055	0,064	<0,050	<0,050	0,13	0,1	<0,050	<0,050
12	BDE 100	<0,050	<0,050	<0,050	<0,050	<0,050	<0,050	0,061	<0,050	<0,050

6.2.2. Taxinge and Sörmland

The concentrations of compounds detected in soil at any of the Taxinge and Sörmland fields are presented in Table 16.

At Taxinge, which has been fertilised with sewage sludge since 1978, musk compounds, PFOS and polybrominated diphenyl ethers were found in concentrations above the reporting limit in the soil approximately three years after sludge addition (Table 16).

At Sörmland, which is a field with a first time application of sludge, only musk compounds and BDE47 were found in soil, shortly after sludge addition (Table 16). Galaxolide and tonalide were found above the reporting limit in soil both before and after sludge addition. After sludge addition the levels of galaxolide were about 7 times higher in the topsoil and also the degradation product galaxolide lactone was detected.

Table 16. Concentrations (µg/kg dw) of compounds found above the reporting limit in any of the analysed soil samples from the Taxinge and Sörmland fields. Grey shading highlights concentrations of compounds above the reporting limit. In Sörmland the sludge was applied at 2014-09-11. At Taxinge the sludge was applied in autumn 2012.

Field location	**Taxinge**		**Sörmland**			
	Approx 3 years after sludge appl.		**Before sludge**		**After sludge**	
Sampling date	**2015-04-09**		**2014-09-03**		**2014-09-15**	
Sampling depth (m)	**0-0,2**	**0,2-0,4**	**0-0,2**	**0,2-0,4**	**0-0,2**	**0,2-0,4**
Galaxolide	1,7	2,6	2,8	2,5	22	<2.0
Tonalide	2,8	5,9	<2.0	3,8	4,2	<2.0
Galaxolide lactone	1,4	<1.0	<2.0	<2.0	7,4	<2.0
PFOS	2,6	<1.0	<1.0	<1.0	<1.0	<1.0
BDE 209	12	<5.0	<5.0	<5.0	<5.0	<5.0
BDE 47	0,1	0,052	<0,05	<0,05	0,11	<0,05
BDE 99	<0.050	0,068	<0,05	<0,05	<0,05	<0,05

6.3. Plant

None of the analysed substances were found at measurable levels in grain from winter wheat sampled at the Skåne field experiment in Petersborg (Appendix 2).

6.4. Earthworms

In earthworms from Taxinge PFOS and BDE 47, 99 and 100 were above the reporting limit (Table 17). PFOA, BDE 209 and musk compounds were below the reporting limits.

Table 17. Concentrations (µg/kg ww) of compounds analysed in earthworms. Grey shading highlights compounds above the reporting limit. Fat content is displayed as %.

Substance	Concentration
PFOS	33
PFOA	<10
BDE 209	<5.0
BDE 47	0,42
BDE 99	0,38
BDE 100	0,11
Galaxolide	<1.0
Tonalide	<1.0
Galaxolide lactone	<1.0
Fat	1,1 %

7. DISCUSSION

7.1. Levels compared to predicted levels

Whether or not certain pollutants accumulate in soils upon repeated applications of sludge is a key issue. Highly persistent substances may persist in soils for long time after sludge amendment. Concentrations of persistent substances thus increase upon each sludge application. For less persistent substances, concentrations may progressively decline following a sludge application. Thus, the concentrations in soil of a certain pollutant also depend on when the samples was taken in relation to the last sludge application. These two types of behavior are illustrated in Figure 5. The results of the present study will now be discussed in relation to this theoretical background.

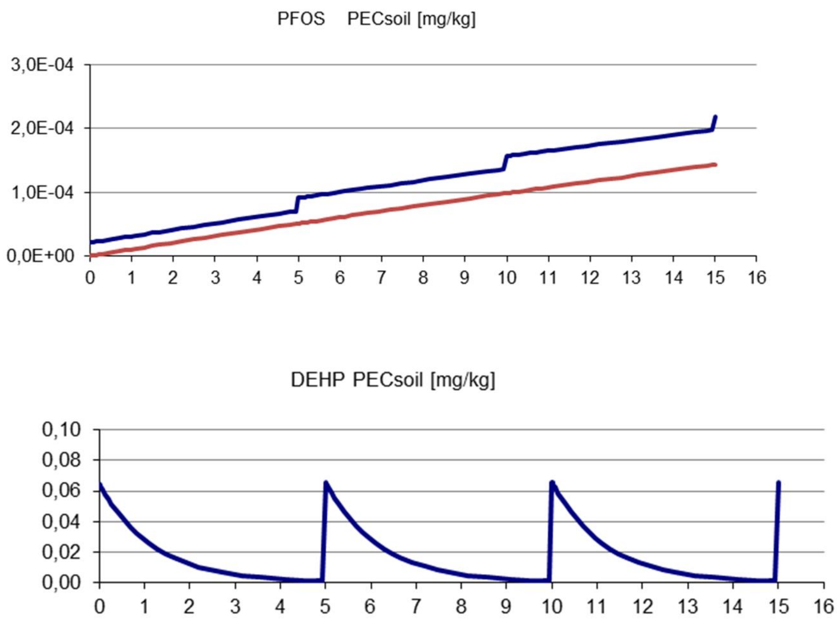


Figure 5. Model calculated concentrations of PFOS and DEHP in soil that is amended with sewage sludge every fifth year. The x-axis shows years and the y-axis concentrations in mg/kg dw. For the upper graph, the red line denotes concentrations due to atmospheric deposition only, whereas the blue line also included sludge amendment.

The present study demonstrates that the following substances were enriched in sludge amended soils, as compared to control soils: PFOS, PBDEs (47, 99, 100 and 209), galaxolide, tonalide and DEHP. There are also single indications that concentrations of DIDP, nonylphenol and TCPP may increase, but these observations are uncertain as they are not generally observed.

The impact of sludge amendments on the occurrence of organic compounds in soil is related to the degradability or persistence of each compound. Model calculations have earlier been performed and can be generalised as follows (Sternbeck et al., 2011; 2013; Figure 5):

- PFOS and BDE-209 should accumulate in soil, and their concentrations are likely to increase upon repeated applications of sludge.
- Lower brominated BDEs such as #47 and #99 do not show this tendency but are likely to persist in soil due to continuous supply by atmospheric deposition.
- DEHP should be detectable during the first two years following sludge application.
- Galaxolide and tonalide should be detected during the first year following sludge application.
- Certain organophosphates, like TCPP, have a slight tendency to accumulate in soil.
- Nonylphenol, LAS and dibutylphthalate should be detected during the first weeks or months following sludge application.

When comparing model predictions with the present field data, PFOS and BDE-209 behaves as expected. The general non-detects of alkylphenols, dibutylphthalate and LAS is also in agreement with predictions. The non-detects of organophosphates is difficult to evaluate because of relatively high analytical reporting limits.

The general presence of galaxolide in the sludge amended soils is however in contrast to model predictions. Predicted degradation is supported by several laboratory studies showing rapid degradation of galaxolide with half lifes much less than a year (DiFrancesco et al., 2004). Tonalide was however more persistent in that study. An in-depth review of galaxolide degradation was performed within the European risk assessment of galaxolide (EU RAR, 2008). The conclusion was that galaxolide should be degraded in soils within about a year. A Swedish screening study of musk substances found detectable levels in soils of galaxolide, galaxolide lactone and tonalide within a few weeks following sludge amendment (Sweco, 2010). At the time of sampling, the soil had also undergone mixing by ploughing. However, none of these substances were detected a year later.

The role of repeated applications can also be assessed by comparing the different fields in this study. In the Sörmland field, sludge was added for the first time, whereas the other fields had been amended for decades. Soil sampling at Sörmland was performed within approximately one week after sludge application. The effect of degradation should thus be minimal even for labile substances. The expected soil concentrations were calculated by simply accounting for the dilution of sludge in the topsoil. Calculated and measured concentrations are compared in Table 18. For many substances it is apparent that single additions of sludge are expected to result in soil concentrations so low that they cannot be detected within the analytical reporting limits. The musk substances galaxolide, tonalide and galaxolide lactone were detected at concentrations slightly higher than predicted.

Table 18. Measured vs calculated concentrations in soil (µg/kg dw) at the Sörmland site.

Substance	Calculated levels	Measured levels
di-(2-etylhexyl)phthalate	46	<50
di-isodecylphthalate	27	<2500
di-isononylphthalate	46	<2500
LAS C10	65	<5000
LAS C11	360	<5000
LAS C12	500	<5000
LAS C13	500	<5000
LAS C14	7,5	<5000
4-tert-octylphenol	1,3	<1
4-tert-OF-monoethoxilate	0,16	<10
4-nonylphenols	29	<15
4-NF-monoethoxilate	2,9	<100
Triclosan	3,8	<10
Galaxolide	8,5	22
Tonalide	0,8	4,2
galaxolide lactone	2,7	7,4
TCPP	3,8	<20
TEHP	1,2	<50
EHDPhP	1	<20
PFOS	0,048	<1
PFOA	0,0075	<1
BDE 209	4,1	<5
BDE 47	0,11	<0,2
BDE 99	0,105	<0,2
BDE 100	0,025	<0,2

The concentrations measured in long term sludge amended soil can be compared to calculated concentrations for 25 years of sludge application. Calculated concentrations were taken from Sternbeck et al. (2011; 2013), or by using the same model. The field data is from three fields where sludge was applied at approximately 4 tons/ha/yr every fourth year for about 30 years. The calculated levels also include atmospheric deposition as a source for PBDE and PFOS. Swedish average concentrations in sludge were used in the calculations. Because we do not know the historical concentrations in sludge that have been applied at these sites, a perfect match cannot be expected.

The measured data show fairly good agreement with model calculations (Table 19). Tonalide and galaxolide have anticipated degradation half-lives less than one year. Their concentrations in soil will thus decrease progressively over time after the sludge application, and this may explain the slightly larger deviations as compared to the more persistent PBDEs. For PFOS, the low concentration in sludge infers that anticipated concentrations in soil are very low even after many repeated amendments. The analytical reporting limits (2 µg/kg dw) where thus not low enough.

Table 19. Comparing measured vs calculated concentrations in sludge amended (4 tons/ha) arable top soils. The measured data represent five samples from fields were sludge has been applied repeatedly every fifth year for ca 30 years. Duplicate samples are represented as average values. “Calculated 25 years” means that sludge has been applied every fifth year for 25 years in the model calculations. Calculated concentrations from Sternbeck et al. (2011, 2013).

Substance	**Measured**		**Calculated 25 years**
	median	Min-max	
BDE47, µg/kg dw	0,22	0,1-0,3	0,077
BDE99, µg/kg dw	0,21	<0,05-0,27	0,11
BDE209, µg/kg dw	3,5	1,9-12	1,6
Galaxolide, µg/kg dw	1,8	1,7-7	9,2
Tonalide, µg/kg dw	<2	<2-2,8	0,75
PFOS, µg/kg dw	<2	<2-2,6	0,33

7.2. Levels compared to earlier findings

There are only few preceding Swedish studies of organic pollutants in arable soils that were subject to sludge applications (see chapter 3). Two of those studies include the fields in Igelösa and Petersborg, that are also investigated in the present study. Concentrations of PBDEs in soils from this area were investigated in 2000 (Sellström et al., 2005). These data are compared to the present data in Figure 6 and Figure 7. BDE47 and 99 are moderately higher in 2015 as compared 2000. Although some difference could be attributed to different analytical laboratories in the two studies, the difference indicates that BDE47 and 99 accumulate in soils as a result of the additional sludge applications that have occurred since 2000.

A North American field study, where sludge had been applied for 33 years and at much higher application rates than in Skåne, did also show accumulation over time for BDE47 and 99 (Xia et al., 2010). The result is, however, in contrast to model calculations which suggest that these congeners should degrade during the time between to sludge application events (Schafer et al, 2008; Sternbeck et al., 2011).

The concentrations of BDE209 in soils are much higher at present than in 2000 (Figure 6 and Figure 7). Qualitatively this is in agreement with model calculations showing that BDE209 will persist in soils and thus accumulate over time upon repeated sludge amendments (Sternbeck et al., 2011). A Spanish field study also demonstrated that BDE209 accumulates more strongly in soil than BDE47 or BDE99 (Eljarrat et al., 2008).

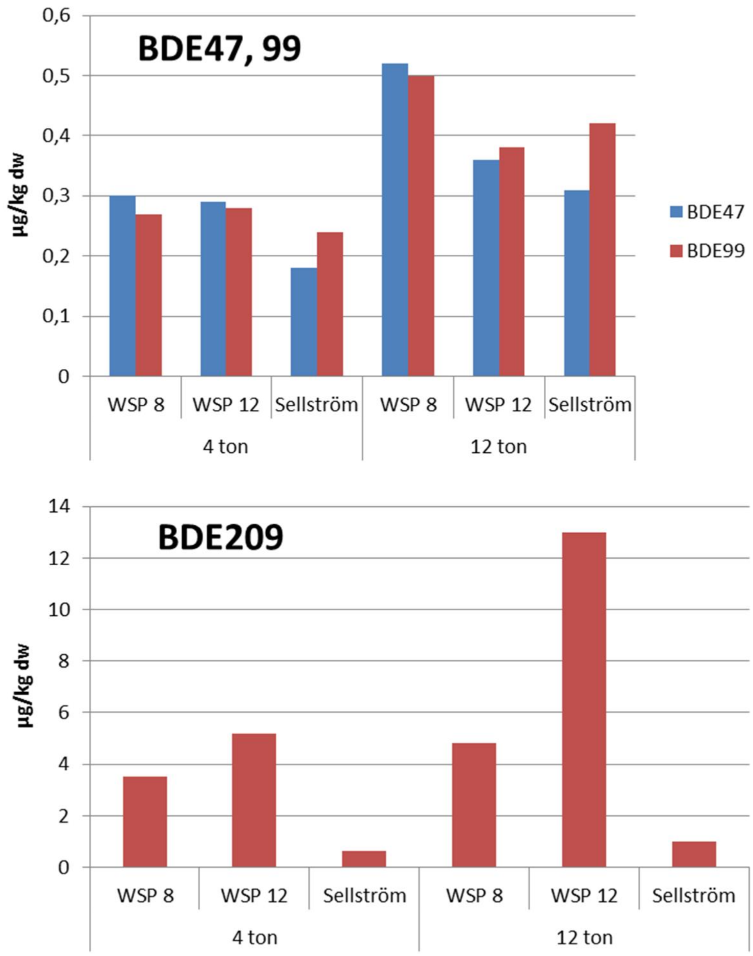
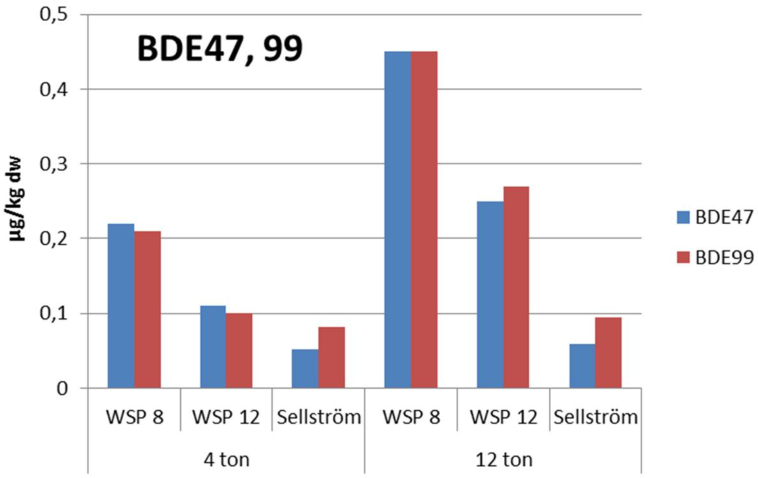


Figure 6. Concentrations of PBDE in soils from Igelösa. WSP 8 means sampling 8 months after sludge application, and WSP 12 means after 12 months. Data represent fields with different application rates: 4 ton/ha and 12 ton/ha. The data from Sellström (2005) represent samples from the year 2000.



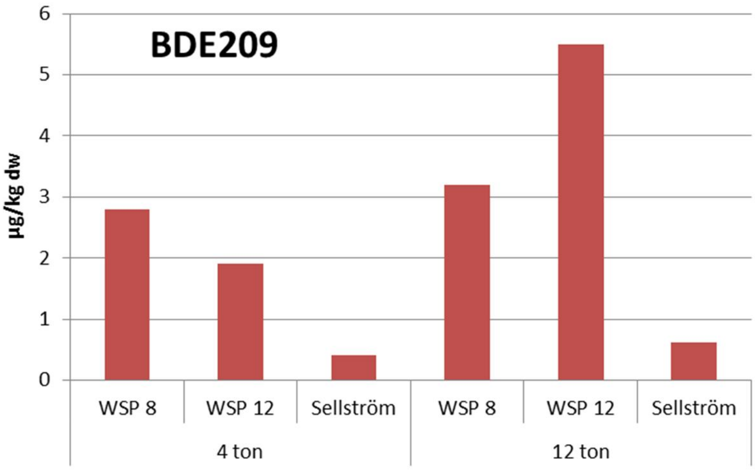


Figure 7. Concentrations of PBDE in soils from Petersborg. WSP 8 means sampling 8 months after sludge application; WSP 12 means after 12 months. The data from Sellström (2005) represent samples from the year 2000.

The results of two other Swedish studies are summarised and compared to the present data in Table 20. Alkylphenols and PFOS were analysed in the sludge amended soils from Igelösa and Petersborg by Hörsing et al. (2014). None of these substances were detected in the samples. In the present study, nonylphenol was detected in one out of several samples. PFOS was detected in several samples but not in the controls, suggesting an influence of sludge amendment. The detected levels are lower than the analytical reporting limit used by Hörsing et al (2014), which explains why they did not detect PFOS. Nevertheless, the detected levels are higher than could be expected based on mass balance for 25 years of sludge amendment (see Table 19). Hypothetically, this could also be due to the fact that PFOS concentrations in sludge were higher in the past (Woldegiorgis et al, 2014). Large scale atmospheric deposition also contributes to a background concentration in Swedish soils (Filipovic et al., 2015). The musk substances occur irregularly in sludge amended soils. Concentrations are in the same range as the previous study.

Table 20. Comparing concentrations in sludge amended soil from the present study with earlier Swedish studies (alkylphenols and PFOS from Hörsing et al., 2014; Musk substances från Sweco, 2010). Data from the present study contains both topsoil and the underlying layer. Duplicate samples are presented as average values. All concentrations in µg/kg dw.

Substances	Earlier Swedish data	Present study			
		Igelösa	Petersborg	Taxinge	Sörmland
OP	<1	<1	<1	<1	<1
NP	<10	<10	<10-16	<10	<10
PFOS	<10	<2-3,7	<2-4,4	<1-2,6	<1
Galaxolide	4-15[A]	<2-4,2	<2-48	1,7-2,6	<2-22
Tonalide	4,7-22[A]	<2	2,3-4,3	2,8-5,9	<2-4,2

A. 25-75- percentiles.

7.3. Bioaccumulation and human exposure

There is a concern that crops grown in sludge amended arable soils will accumulate pollutants from the sludge and thereby contribute to human exposure. None of the analysed organic compounds in the present study were detected in grains from winter wheat grown at the experimental fields in Petersborg, Skåne. This is in line with earlier findings (Hörsing et al 2014, Laturnus et al 2007, Hale et al 2012), which shown no or low uptake of organic pollutions in crops with sludge additions representative to Swedish legislation. However, in the present study only one type of plant species were examined. There are large differences in plant accumulation of organic pollutants, accumulation is generally higher in root crops.

The non-detects in crops can be compared to theoretically expected concentrations. Expected concentrations in root crops and cereals have been calculated based on the highest measured levels in topsoil, where sewage sludge has been applied according to Swedish legislations. Calculations were performed for those substances with available bioconcentration factors (BCF; from Sternbeck et al., 2011, 2013). In general calculated levels in plants are very low (ng/g ww) and it is apparent that expected concentrations for most substances are so low that they cannot be detected within the analytical reporting limits (Table 21). The highest accumulation that should be possible to measure in root crops with the used detection limit is TCPP, which is the substance with highest potential of bioaccumulation in crops of the studied compounds. However, in the present study TCPP was only detected in one soil sample and at the same level as the detection limit.

In summary, that fact that none of the analysed substances could be detected in crops confirms laboratory uptake studies and model calculations: many organic pollutants are not efficiently accumulated from sludge amended soil to crops.

Table 21. Expected levels in plants as calculated from the highest measured levels in top-soil, where sewage sludge has been applied according to Swedish legislations. BCF values from Table 1. The current detection limits for plants are also shown.

Substance	**Max conc in topsoil** µg/kg dw	**Calc. conc. in root crops** µg/kg ww plant	**Calc. Conc. in cereals** µg/kg ww plant	**Detection limit** µg/kg ww
DEHP	140	0,012		50
Galaxolide	13	1,2		2
Tonalide	5,7	0,4		2
TCPP	20	56	1,9	20
PFOS	2,5	0,10	0,022	1
BDE-209	12	0,0030		10
BDE-47	0,39	0,0009		0,1
BDE-99	0,37	0,0005		0,1

Earthworms of many species are ubiquitous organisms in arable soils. They feed by digesting organic detritus and are directly exposed to soil by their burrowing activities. These animals have thus been used in monitoring pollutants in soils, including sludge amended soil (e.g. Matcheko et al., 2002; Kinney et al., 2008, 2010). In contrast to conventional analysis of soil, the analysis of earthworms gives a measure of the bioavailable concentration. Furthermore, earthworms are important feed to a number of species and the pollutants in earthworms may thus be of ecological relevance.

In the present study PFOS and BDE 47, 99 and 100 were detected in earthworms from Taxinge, showing that these substances can bioaccumulate in biota. All of these substances, with exception of BDE 100, were also detected in soil from Taxinge. Calculated biota to soil accumulation factors (BSAF) from measured levels or used detection limits in soil and earthworms are shown in Table 22 together with BSAF values from previous studies. In general calculated BSAF values are in line with earlier findings. The substances with highest BSAF values are also the ones that were detected in earthworms in the present study. Neither galaxolide nor tonalide were detected in earthworms.

Table 22. Calculated biota to soil accumulation factor (BSAF) for earthworms compared to reference values of BSAF (Table 5). The units differ for different substances.

Substance	**Calculated BSAF**	**Reference BSAF**	**Unit BSAF**
Galaxolide	<2	0,05-3,1	dw/dw
Tonalide	<2	0,1-1	dw/dw
PFOS	63	18	dw/dw
BDE47	21	5	OM/lw[1]
BDE 99	<38	4,2	OM/lw[1]
BDE 100	<6	4,6	OM/lw[1]
BDE 209	<2	0,3	OM/lw[1]

[1] Organic matter content (OM) for Taxinge field was assumed to be 5 % in lack of site specific values.

7.4. Environmental significance

To characterize the risk to health or the soil ecosystem, concentrations of those organic pollutants that were found in elevated levels in sludge amended soil have been compared to critical soil concentrations for soil ecosystem and humans. The highest measured levels in soil, where sewage sludge has been applied according to Swedish legislations, were used in this assessment. The risk level is given as a risk quota in Figure 8, which is calculated as the maximum found concentration in topsoil divided with the critical concentration for soil ecosystem or humans exposed via intake of crops ($RQ=C_{soil}/PNEC$ or $C_{crit\ humans}$). Critical concentrations for soil ecosystem are outlined in Table 1 and critical concentrations for humans have earlier been calculated and presented in Sternbeck et al, 2011 and 2013.

A RQ>1 indicates that levels in soil can pose negative effects to soil ecosystem or humans exposed via crops. None of the calculated risk quota exceeds 1 (Figure 8), indicating that the observed levels in soil after long term sludge additions do not pose a risk to the soil ecosystem or humans.

All risk quotas for human exposure via crops are less than 1. The safety margin to the critical concentration for humans is at least 400 times. For the soil ecosystem the safety margin is in general less. The safety margin for soil ecosystem is at least 20 for all compounds except PFOS. For PFOS the safety margin is 4. These results are in line with earlier risk assessment of organic substances applied to arable land via sewage sludge addition (Sternbeck et al, 2011 and 2013).

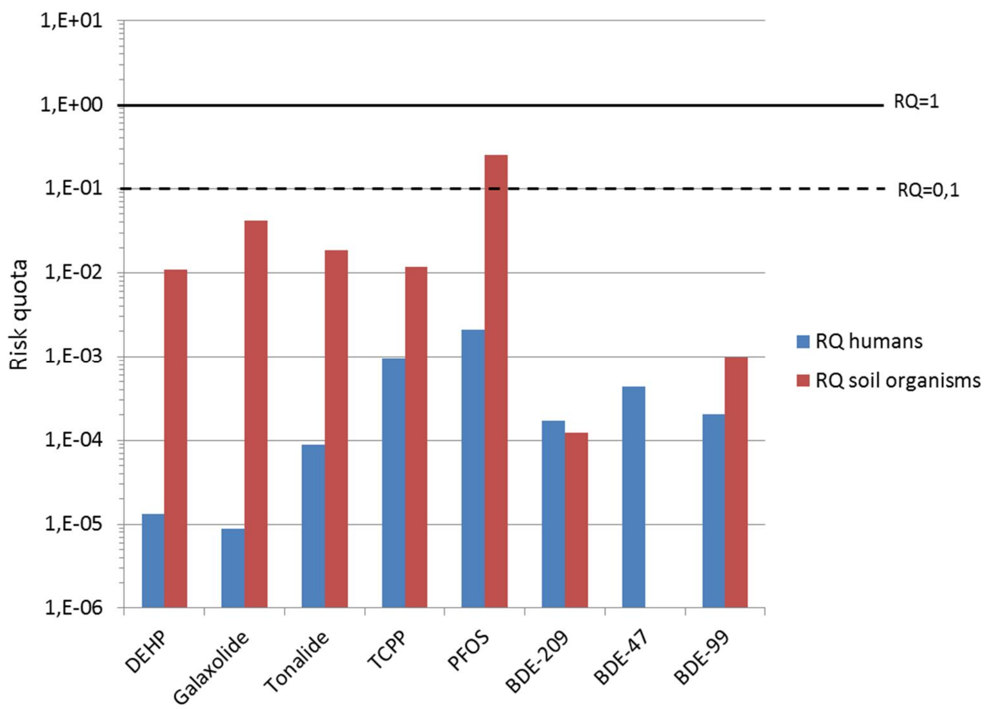


Figure 8. Calculated risk quota (RQ) for humans and soil ecosystem for organic substances found in elevated levels in soil amended with sewage sludge. Maximum levels in topsoil have been used. For BDE-47 no RQ for soil organisms could be calculated due to lack of critical concentration (PNEC).

8. CONCLUSIONS

- PFOS, PBDEs (47, 99, 100 and 209), galaxolide, tonalide and DEHP are enriched in sludge amended soils. The impact is primarily, but not exclusively, in the upper most soil layers (0-0,3 m).
- Concentrations of DIDP, nonylphenol and TCPP may also increase in sludge amended soils, but these observations are uncertain as they are not generally observed.
- For many of the examined substances it is apparent that single additions of sludge will result in soil concentrations so low (sub µg/kg dw) that they cannot be detected within the analytical reporting limits.
- The found accumulation of PFOS and BDE-209 in soil upon repeated applications of sludge over time is in line with previous model predictions.
- The general non-detects of alkylphenols, dibutylphthalate and LAS in sludge amended soils is also in line with earlier predictions and findings.
- Galaxolide was commonly found in enriched levels in sludge amended soils relative to control fields. Whether galaxolide accumulates or not, following repeated applications, cannot be assessed from this study. Earlier model predictions and degradation studies do, however, suggest that galaxolide should not accumulate.
- None of the analysed organic substances were detected in grains of winter wheat grown at the experimental fields in Petersborg. The results confirm previous model assessments and uptake studies: many organic pollutants accumulate poorly from soil to crops.
- PFOS and PBDE (47, 99 and 100) were detected in earthworms from Taxinge, showing that these substances can bioaccumulate in biota. Calculated biota to soil accumulation factors (BSAF) are in line with earlier findings.
- The risk characterization shows that levels in soil do not pose a risk to the soil ecosystem or humans, not even after long term sludge additions. These findings are in agreement with earlier findings.

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WSP Environmental, 2015-11-24

Ann Helén Österås Mats Allmyr John Sternbeck

APPENDIX 1. REPORTING LIMITS FOR ANALYSED COMPOUNDS

This appendix shows the estimated reporting limits specified by ALS for the herein analysed compounds. Note that the estimated reporting limit may be over or under the analytical reporting limit.

Table A1. Estimated reporting limits for the analysed compounds in the present study.

Compound	Sludge (mg/kg dw)	Soil (µg/kg dw)	Crop (µg/kg)	Earthworm (µg/kg)
di-(2)etylhexyl phthalate (DEHP)	0,05	50	50	
di-isodecyl phthalate (DIDP)	2,5	2500	2500	
di-isononyl phthalate (DINP)	2,5	2500	2500	
Dimethylphthalate	2,5	2500	2500	
Diethylphthalate	2,5	2500	2500	
Di-n-Propylphthalate	2,5	2500	2500	
Di-iso-Butylphthalate	2,5	2500	2500	
Di-n-Butylphthalate	2,5	2500	2500	
Butylbenzylphthalate	2,5	2500	2500	
Di-Cyclohexylphthalate	2,5	2500	2500	
Di-n-Octylphthalate	2,5	2500	2500	
C10-C14 Monoalkylbenzene sulfonic acid, sodium salt (LAS)	5	5000	5000	
4-tert-octylphenol	0,002	1	1	
4-iso-nonylphenol	0,020	10	10	
OP1EO, OP2EO, OP3EO	0,020	10	10	
NP1EO, NP2EO, NP3EO	0,200	100	100	
Triclosan	0,100	10	10	
Triclocarban	1,000	100	100	
galaxolide	0,010	2	2	5
tonalid	0,010	2	2	5
galaxolide lactone	0,010	2	2	10
TCrP (tri-cresyl phosphate)	0,100	20	20	
TCEP (tris (2-chloroethyl) phosphate)	0,050	50		
TDCP (tris (dichlorpropyl) phosfate)	0,050	50		
TBP (tris-n-butyl phosfate)	0,050	50		
TBEP (tris(2-butoxyethyl) phosphate)	0,050	100		
TEHP (tris(2-ethylhexyl)phosphate	0,050	50		
TIBP (tri-iso-butylfphosphate)	0,050	50		
ToCrP (trs-o-cresyl phosphate)	0,050	50		
TPhP (trifphenyl phosphate)	0,050	50		
DBPhP (dibutyl phenyl phosphate)	0,10	100		
DPhBP (diphenylbutylphosphate)	0,050	50		
TCPP (tris(2-chloroisopropyl)phosphate)	0,100	20	20	
TBEP (tris(2-butoxyethyl) phosphate)	0,200	50	50	
EHDPhP (2-Ethylhexyl diphenyl phosphate)	0,100	20	20	
PFOS	0,002	2	1	1
PFOA	0,002	2	1	1
BDE209	0,025	10	10	10
BDE47	0,00025	0,05	0,1	0,1
BDE99	0,00025	0,05	0,1	0,1
BDE100	0,00025	0,05	0,1	0,1

APPENDIX 2. SAMPLE DETAILS

This appendix shows sample details for all samples. The analytical results for the corresponding samples are shown in appendix 3.

Table A2. Sample details

Sample no	County	Municipality	Media	Site name	SWEREF 99, N	SWEREF 99, E	Sampling date	Sample details
WSP6232-1	Confidential	Confidential	Sludge	"Sörmland"	Confidential	Confidential	2014-08-(12-23)	Sludge from STP (frozen)
WSP6232-2	Confidential	Confidential	Sludge	"Sörmland"	Confidential	Confidential	2014-09-03	Sludge stack depth: 0-0,6 m
WSP6232-3	Skåne	Malmö	Crop	Petersborg	6156900	373630	2014-07-27	Sludge: 0 t dw/ha/4 yr, Field ID: 3, Winter wheat, crop
WSP6232-4	Skåne	Malmö	Crop	Petersborg	6156900	373630	2014-07-27	Sludge: 4 t dw/ha/4 yr, Field ID: 9+12, Winter wheat, crop
WSP6232-5	Skåne	Malmö	Crop	Petersborg	6156900	373630	2014-07-27	Sludge: 12 t dw/ha/4 yr, Field ID: 15+18, Winter wheat, crop
WSP6232-6	Stockholm	Nykvarn	Earthworm	Taxinge	6568580	631490	2015-04-09	Soil depth: 0-0,2 m
WSP6232-7	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-04-08	Sludge: 0 t dw/ha/4 yr, Field ID: 3, Soil depth: 0-0,3 m
WSP6232-8	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-04-08	Sludge: 4 t dw/ha/4 yr, Field ID: 9, Soil depth: 0-0,3 m
WSP6232-9	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-04-08	Sludge: 4 t dw/ha/4 yr, Field ID: 9, Soil depth: 0,3-0,6 m
WSP6232-10	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-04-08	Sludge: 4 t dw/ha/4 yr, Field ID: 12, Soil depth: 0-0,3 m
WSP6232-11	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-04-08	Sludge: 4 t dw/ha/4 yr, Field ID: 12, Soil depth: 0,3-0,6 m
WSP6232-12	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-04-08	Sludge: 12 t dw/ha/4 yr, Field ID: 15, Soil depth: 0-0,3 m
WSP6232-13	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-04-08	Sludge: 12 t dw/ha/4 yr, Field ID: 15, Soil depth: 0,3-0,6 m
WSP6232-14	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-04-08	Sludge: 12 t dw/ha/4 yr, Field ID: 18, Soil depth: 0-0,3 m
WSP6232-15	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-04-08	Sludge: 12 t dw/ha/4 yr, Field ID: 18, Soil depth: 0,3-0,6 m
WSP6232-16	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-07-27	Sludge: 0 t dw/ha/4 yr, Field ID: 3, Soil depth: 0-0,3 m
WSP6232-17	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-07-27	Sludge: 4 t dw/ha/4 yr, Field ID: 9, Soil depth: 0-0,3 m
WSP6232-18	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-07-27	Sludge: 4 t dw/ha/4 yr, Field ID: 9, Soil depth: 0,3-0,6 m
WSP6232-19	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-07-27	Sludge: 4 t dw/ha/4 yr, Field ID: 12, Soil depth: 0-0,3 m
WSP6232-20	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-07-27	Sludge: 4 t dw/ha/4 yr, Field ID: 12, Soil depth: 0,3-0,6 m
WSP6232-21	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-07-27	Sludge: 12 t dw/ha/4 yr, Field ID: 15, Soil depth: 0-0,3 m
WSP6232-22	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-07-27	Sludge: 12 t dw/ha/4 yr, Field ID: 15, Soil depth: 0,3-0,6 m
WSP6232-23	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-07-27	Sludge: 12 t dw/ha/4 yr, Field ID: 18, Soil depth: 0-0,3 m
WSP6232-24	Skåne	Malmö	Soil	Petersborg	6156900	373630	2014-07-27	Sludge: 12 t dw/ha/4 yr, Field ID: 18, Soil depth: 0,3-0,6 m
WSP6232-25	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-04-08	Sludge: 0 t dw/ha/4 yr, Field ID: 3, Soil depth: 0-0,3 m
WSP6232-26	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-04-08	Sludge: 4 t dw/ha/4 yr, Field ID: 9, Soil depth: 0-0,3 m

Sample no	County	Municipality	Media	Site name	SWEREF 99, N	SWEREF 99, E	Sampling date	Sample details
WSP6232-27	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-04-08	Sludge: 4 t dw/ha/4 yr, Field ID: 9, Soil depth: 0,3-0,6 m
WSP6232-28	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-04-08	Sludge: 4 t dw/ha/4 yr, Field ID: 12, Soil depth: 0-0,3 m
WSP6232-29	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-04-08	Sludge: 4 t dw/ha/4 yr, Field ID: 12, Soil depth: 0,3-0,6 m
WSP6232-30	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-04-08	Sludge: 12 t dw/ha/4 yr, Field ID: 15, Soil depth: 0-0,3 m
WSP6232-31	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-04-08	Sludge: 12 t dw/ha/4 yr, Field ID: 15, Soil depth: 0,3-0,6 m
WSP6232-32	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-04-08	Sludge: 12 t dw/ha/4 yr, Field ID: 18, Soil depth: 0-0,3 m
WSP6232-33	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-04-08	Sludge: 12 t dw/ha/4 yr, Field ID: 18, Soil depth: 0,3-0,6 m
WSP6232-34	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-07-29	Sludge: 0 t dw/ha/4 yr, Field ID: 3, Soil depth: 0-0,3 m
WSP6232-35	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-07-29	Sludge: 4 t dw/ha/4 yr, Field ID: 9, Soil depth: 0-0,3 m
WSP6232-36	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-07-29	Sludge: 4 t dw/ha/4 yr, Field ID: 9, Soil depth: 0,3-0,6 m
WSP6232-37	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-07-29	Sludge: 4 t dw/ha/4 yr, Field ID: 12, Soil depth: 0-0,3 m
WSP6232-38	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-07-29	Sludge: 4 t dw/ha/4 yr, Field ID: 12, Soil depth: 0,3-0,6 m
WSP6232-39	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-07-29	Sludge: 12 t dw/ha/4 yr, Field ID: 15, Soil depth: 0-0,3 m
WSP6232-40	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-07-29	Sludge: 12 t dw/ha/4 yr, Field ID: 15, Soil depth: 0,3-0,6 m
WSP6232-41	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-07-29	Sludge: 12 t dw/ha/4 yr, Field ID: 18, Soil depth: 0-0,3 m
WSP6232-42	Skåne	Lund	Soil	Igelösa	6176700	392920	2014-07-29	Sludge: 12 t dw/ha/4 yr, Field ID: 18, Soil depth: 0,3-0,6 m
WSP6232-43	Confidential	Confidential	Soil	"Sörmland"	Confidential	Confidential	2014-09-03	Soil depth: 0-0,2 m
WSP6232-44	Confidential	Confidential	Soil	"Sörmland"	Confidential	Confidential	2014-09-03	Soil depth: 0,2-0,4 m
WSP6232-45	Confidential	Confidential	Soil	"Sörmland"	Confidential	Confidential	2014-09-15	Soil depth: 0-0,2 m
WSP6232-46	Confidential	Confidential	Soil	"Sörmland"	Confidential	Confidential	2014-09-15	Soil depth: 0,2-0,4 m
WSP6232-47	Stockholm	Nykvarn	Soil	Taxinge	6568580	631490	2015-04-09	Soil depth: 0-0,2 m
WSP6232-48	Stockholm	Nykvarn	Soil	Taxinge	6568580	631490	2015-04-09	Soil depth: 0,2-0,4 m

APPENDIX 3. ANALYTICAL DATA

Table A3. Concentrations of analysed compounds in sludge (mg/kg dw), biota (µg/kg ww), soil (µg/kg dw).

Sample no	Media	Depth (m)	Dry matter (%) Earthworm: fat (%)	Dimethyl phthalate	Diethyl phthalate	Di-n-Propyl phthalate	Di-iso-Butyl phthalate	Di-n-Butyl phthalate	Di-pentyl phthalate	Di-n-oktyl phthalate
WSP6232-1	Sludge		27,7	<0,20	<0,20	<0,20	0,43	<0,20	<0,20	<1,0
WSP6232-2	Sludge	0,0-0,6	26,6	<0,050	<0,050	<0,050	<0,060	<0,050	<0,050	<0,10
WSP6232-3	Crop		87,1	<50	<50	<50	<50	<50	<50	<50
WSP6232-4	Crop		87,1	<50	<50	<50	<50	<50	<50	<50
WSP6232-5	Crop		87,1	<50	<50	<50	<50	<50	<50	<50
WSP6232-6	Earthworm	0,0-0,2	1,1	NA	NA	NA	NA	NA	NA	NA
WSP6232-7	Soil	0,0-0,3	86,5	<50	<50	<50	<50	<50	<50	<50
WSP6232-8	Soil	0,0-0,3	85,6	<50	<50	<50	<50	<50	<50	<50
WSP6232-9	Soil	0,3-0,6	85,9	<50	<50	<50	<50	<50	<50	<50
WSP6232-10	Soil	0,0-0,3	86	<50	<50	<50	<50	<50	<50	<50
WSP6232-11	Soil	0,3-0,6	86,9	<50	<50	<50	<50	<50	<50	<50
WSP6232-12	Soil	0,0-0,3	85	<50	<50	<50	<50	<50	<50	<50
WSP6232-13	Soil	0,3-0,6	86,8	<50	<50	<50	<50	<50	<50	<50
WSP6232-14	Soil	0,0-0,3	85,4	<50	<50	<50	<50	<50	<50	<50
WSP6232-15	Soil	0,3-0,6	86,4	<50	<50	<50	<50	<50	<50	<50
WSP6232-16	Soil	0,0-0,3	86,5	<50	<50	<50	<50	<50	<50	<50
WSP6232-17	Soil	0,0-0,3	86,5	<50	<50	<50	<50	<50	<50	<50
WSP6232-18	Soil	0,3-0,6	88,9	<50	<50	<50	<50	<50	<50	<50
WSP6232-19	Soil	0,0-0,3	86,2	<50	<50	<50	<50	<50	<50	<50
WSP6232-20	Soil	0,3-0,6	89	<50	<50	<50	<50	<50	<50	<50
WSP6232-21	Soil	0,0-0,3	85,5	<50	<50	<50	<50	<50	<50	<50
WSP6232-22	Soil	0,3-0,6	88,7	<50	<50	<50	<50	<50	<50	<50
WSP6232-23	Soil	0,0-0,3	85,7	<50	<50	<50	<50	<50	<50	<50
WSP6232-24	Soil	0,3-0,6	87,5	<50	<50	<50	<50	<50	<50	<50
WSP6232-25	Soil	0,0-0,3	81,2	<50	<50	<50	<50	<50	<50	<50
WSP6232-26	Soil	0,0-0,3	81	<50	<50	<50	<50	<50	<50	<50
WSP6232-27	Soil	0,3-0,6	84,3	<50	<50	<50	<50	<50	<50	<50
WSP6232-28	Soil	0,0-0,3	79	<50	<50	<50	<50	<50	<50	<50
WSP6232-29	Soil	0,3-0,6	82,8	<50	<50	<50	<50	<50	<50	<50
WSP6232-30	Soil	0,0-0,3	79,5	<50	<50	<50	<50	<50	<50	<50
WSP6232-31	Soil	0,3-0,6	82,8	<50	<50	<50	<50	<50	<50	<50
WSP6232-32	Soil	0,0-0,3	79,2	<50	<50	<50	<50	<50	<50	<50
WSP6232-33	Soil	0,3-0,6	84,3	<50	<50	<50	<50	<50	<50	<50
WSP6232-34	Soil	0,0-0,3	81,1	<50	<50	<50	<50	<50	<50	<50
WSP6232-35	Soil	0,0-0,3	80,1	<50	<50	<50	<50	<50	<50	<50
WSP6232-36	Soil	0,3-0,6	83	<50	<50	<50	<50	<50	<50	<50
WSP6232-37	Soil	0,0-0,3	78,5	<50	<50	<50	<50	<50	<50	<50
WSP6232-38	Soil	0,3-0,6	83,1	<50	<50	<50	<50	<50	<50	<50
WSP6232-39	Soil	0,0-0,3	79	<50	<50	<50	<50	<50	<50	<50
WSP6232-40	Soil	0,3-0,6	83,4	<50	<50	<50	<50	<50	<50	<50
WSP6232-41	Soil	0,0-0,3	78,3	<50	<50	<50	<50	<50	<50	<50
WSP6232-42	Soil	0,3-0,6	83,5	<50	<50	<50	<50	<50	<50	<50
WSP6232-43	Soil	0,0-0,2	84,3	<50	<50	<50	<50	<50	<50	<50
WSP6232-44	Soil	0,2-0,4	87,2	<50	<50	<50	<50	<50	<50	<50
WSP6232-45	Soil	0,0-0,2	83,6	<50	<50	<50	<50	<50	<50	<50
WSP6232-46	Soil	0,2-0,4	87,5	<50	<50	<50	<50	<50	<50	<50
WSP6232-47	Soil	0,0-0,2	78,5	<50	<50	<50	<50	<50	<50	<50
WSP6232-48	Soil	0,2-0,4	79,4	<50	<50	<50	<50	<50	<50	<50

Table A3. *(Cont.)* Concentrations of analysed compounds in sludge (mg/kg dw), biota (µg/kg ww), soil (µg/kg dw).

Sample no	Media	Depth (m)	DEHP Di-(2)etylhexyl phthalate	Butylbenzyl phthalate	Di-cyclohexyl phthalate	Di-isodecyl phthalate	Di-isononyl phthalate	C10- Monoalkylbenzene sulfonic acid, sodium salt	C11- Monoalkylbenzene sulfonic acid, sodium salt	C12- Monoalkylbenzene sulfonic acid, sodium salt
WSP6232-1	Sludge		87	<0,20	<0,20	<10	76	25	88	110
WSP6232-2	Sludge	0,0-0,6	9,2	<0,050	<0,050	5,3	9,1	13	72	100
WSP6232-3	Crop		<50	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-4	Crop		<50	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-5	Crop		<50	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-6	Earthworm	0,0-0,2	NA	NA	NA	NA	NA	NA	NA	NA
WSP6232-7	Soil	0,0-0,3	<50	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-8	Soil	0,0-0,3	110	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-9	Soil	0,3-0,6	<50	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-10	Soil	0,0-0,3	92	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-11	Soil	0,3-0,6	<50	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-12	Soil	0,0-0,3	92	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-13	Soil	0,3-0,6	100	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-14	Soil	0,0-0,3	<50	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-15	Soil	0,3-0,6	65	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-16	Soil	0,0-0,3	<50	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-17	Soil	0,0-0,3	71	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-18	Soil	0,3-0,6	<50	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-19	Soil	0,0-0,3	<50	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-20	Soil	0,3-0,6	<50	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-21	Soil	0,0-0,3	<50	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-22	Soil	0,3-0,6	<50	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-23	Soil	0,0-0,3	<50	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-24	Soil	0,3-0,6	<50	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-25	Soil	0,0-0,3	<50	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-26	Soil	0,0-0,3	89	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-27	Soil	0,3-0,6	<50	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-28	Soil	0,0-0,3	140	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-29	Soil	0,3-0,6	230	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-30	Soil	0,0-0,3	77	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-31	Soil	0,3-0,6	100	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-32	Soil	0,0-0,3	110	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-33	Soil	0,3-0,6	130	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-34	Soil	0,0-0,3	<50	<50	<50	3100	<2500	<500	<500	<500
WSP6232-35	Soil	0,0-0,3	<50	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-36	Soil	0,3-0,6	<50	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-37	Soil	0,0-0,3	<50	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-38	Soil	0,3-0,6	140	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-39	Soil	0,0-0,3	89	<50	<50	5200	<2500	<500	<500	<500
WSP6232-40	Soil	0,3-0,6	<50	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-41	Soil	0,0-0,3	74	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-42	Soil	0,3-0,6	<50	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-43	Soil	0,0-0,2	<50	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-44	Soil	0,2-0,4	<50	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-45	Soil	0,0-0,2	<50	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-46	Soil	0,2-0,4	<50	<50	<50	<2500	<2500	<5000	<5000	<5000
WSP6232-47	Soil	0,0-0,2	<50	<50	<50	<2500	<2500	<500	<500	<500
WSP6232-48	Soil	0,2-0,4	<50	<50	<50	<2500	<2500	<500	<500	<500

Table A3. *(Cont.)* Concentrations of analysed compounds in sludge (mg/kg dw), biota (µg/kg ww), soil (µg/kg dw).

Sample no	Media	Depth (m)	C13- Monoalkylbenzene sulfonic acid, sodium salt	C14- Monoalkylbenzene sulfonic acid, sodium salt	LAS C10-C14, Monoalkyl-benzene sulfonic acid, sodium salt	4-tert-octylphenol	OP1EO 4-tert- octylphenol monoethoxylate	OP2EO 4-tert-octylphenol diethoxylate	OP3EO (4-tert-octylphenol-triethoxylate)	4-nonylphenol (technical mix)
WSP6232-1	Sludge		78	<10	300	0,44	0,058	<0,010	<0,010	8,3
WSP6232-2	Sludge	0,0-0,6	100	1,5	290	0,25	0,031	<0,010	<0,010	5,8
WSP6232-3	Crop		<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-4	Crop		<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-5	Crop		<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-6	Earthworm	0,0-0,2	NA	NA	NA	NA	NA	NA	NA	NA
WSP6232-7	Soil	0,0-0,3	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-8	Soil	0,0-0,3	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-9	Soil	0,3-0,6	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-10	Soil	0,0-0,3	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-11	Soil	0,3-0,6	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-12	Soil	0,0-0,3	<5000	<5000	NA	<1,0	<10	<10	<10	16
WSP6232-13	Soil	0,3-0,6	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-14	Soil	0,0-0,3	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-15	Soil	0,3-0,6	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-16	Soil	0,0-0,3	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-17	Soil	0,0-0,3	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-18	Soil	0,3-0,6	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-19	Soil	0,0-0,3	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-20	Soil	0,3-0,6	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-21	Soil	0,0-0,3	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-22	Soil	0,3-0,6	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-23	Soil	0,0-0,3	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-24	Soil	0,3-0,6	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-25	Soil	0,0-0,3	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-26	Soil	0,0-0,3	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-27	Soil	0,3-0,6	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-28	Soil	0,0-0,3	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-29	Soil	0,3-0,6	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-30	Soil	0,0-0,3	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-31	Soil	0,3-0,6	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-32	Soil	0,0-0,3	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-33	Soil	0,3-0,6	<5000	<5000	NA	<1,0	<10	<10	<10	<10
WSP6232-34	Soil	0,0-0,3	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-35	Soil	0,0-0,3	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-36	Soil	0,3-0,6	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-37	Soil	0,0-0,3	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-38	Soil	0,3-0,6	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-39	Soil	0,0-0,3	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-40	Soil	0,3-0,6	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-41	Soil	0,0-0,3	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-42	Soil	0,3-0,6	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-43	Soil	0,0-0,2	<5000	<5000	NA	<2,0	<10	<10	<10	<20
WSP6232-44	Soil	0,2-0,4	<5000	<5000	NA	<2,0	<10	<10	<10	<20
WSP6232-45	Soil	0,0-0,2	<5000	<5000	NA	<1,0	<10	<15	<15	<15
WSP6232-46	Soil	0,2-0,4	<5000	<5000	NA	<1,0	<10	<10	<20	<10
WSP6232-47	Soil	0,0-0,2	<500	<500	NA	<1,0	<10	<10	<10	<10
WSP6232-48	Soil	0,2-0,4	<500	<500	NA	<1,0	<10	<10	<10	<10

Table A3. *(Cont.)* Concentrations of analysed compounds in sludge (mg/kg dw), biota (µg/kg ww), soil (µg/kg dw).

Sample no	Media	Depth (m)	NP1EO (4-nonylophenol-monoethoxylate)	NP2EO (4-nonylophenol-diethoxylate)	NP3EO (4- nonylophenol-triethoxylate)	triclosan	triclocarban	galaxolide	tonalide	galaxolide lactone
WSP6232-1	Sludge		1,2	0,15	<0,800	1,3	<1,0	14	3,5	2,0
WSP6232-2	Sludge	0,0-0,6	0,58	<0,11	<0,30	0,76	<1,0	1,7	0,16	0,53
WSP6232-3	Crop		<100	<100	<100	<10	<100	<10	<10	<10
WSP6232-4	Crop		<100	<100	<100	<10	<100	<10	<10	<10
WSP6232-5	Crop		<100	<100	<100	<10	<100	<10	<10	<10
WSP6232-6	Earthworm	0,0-0,2	NA	NA	NA	NA	NA	<1,0	<1,0	<1,0
WSP6232-7	Soil	0,0-0,3	<100	<100	<100	<10	<100	<2,0	<2,0	<2,0
WSP6232-8	Soil	0,0-0,3	<100	<100	<100	<10	<100	13	2,6	3,7
WSP6232-9	Soil	0,3-0,6	<100	<100	<100	<10	<100	<2,0	<2,0	<2,0
WSP6232-10	Soil	0,0-0,3	<100	<100	<100	<10	<100	<2,0	<2,0	<2,0
WSP6232-11	Soil	0,3-0,6	<100	<100	<100	<10	<100	<2,0	2,8	<2,0
WSP6232-12	Soil	0,0-0,3	<100	<100	<100	<10	<100	10	2,9	5,8
WSP6232-13	Soil	0,3-0,6	<100	<100	<100	<10	<100	<2,0	2,9	<2,0
WSP6232-14	Soil	0,0-0,3	<100	<100	<100	<10	<100	18	5,7	6,2
WSP6232-15	Soil	0,3-0,6	<100	<100	<100	<10	<100	<2,0	<2,0	<2,0
WSP6232-16	Soil	0,0-0,3	<100	<100	<100	<10	<100	<2,0	16	<2,0
WSP6232-17	Soil	0,0-0,3	<100	<100	<100	<10	<100	<2,0	7,6	<2,0
WSP6232-18	Soil	0,3-0,6	<100	<100	<100	<10	<100	<2,0	2,9	<2,0
WSP6232-19	Soil	0,0-0,3	<100	<100	<100	<10	<100	<2,0	8,2	<2,0
WSP6232-20	Soil	0,3-0,6	<100	<100	<100	<10	<100	<2,0	10	<2,0
WSP6232-21	Soil	0,0-0,3	<100	<100	<100	<10	<100	26	29	2,3
WSP6232-22	Soil	0,3-0,6	<100	<100	<100	<10	<100	24	25	3,5
WSP6232-23	Soil	0,0-0,3	<100	<100	<100	<10	<100	9,9	7,1	<2,0
WSP6232-24	Soil	0,3-0,6	<100	<100	<100	<10	<100	70	41	13
WSP6232-25	Soil	0,0-0,3	<100	<100	<100	<10	<100	<2,0	<2,0	<2,0
WSP6232-26	Soil	0,0-0,3	<100	<100	<100	<10	<100	<2,0	<2,0	<2,0
WSP6232-27	Soil	0,3-0,6	<100	<100	<100	<10	<100	<2,0	<2,0	<2,0
WSP6232-28	Soil	0,0-0,3	<100	<100	<100	<10	<100	2,5	<2,0	<2,0
WSP6232-29	Soil	0,3-0,6	<100	<100	<100	<10	<100	<2,0	<2,0	<2,0
WSP6232-30	Soil	0,0-0,3	<100	<100	<100	<10	<100	2,8	<2,0	<2,0
WSP6232-31	Soil	0,3-0,6	<100	<100	<100	<10	<100	<2,0	<2,0	<2,0
WSP6232-32	Soil	0,0-0,3	<100	<100	<100	<10	<100	5,6	<2,0	2,7
WSP6232-33	Soil	0,3-0,6	<100	<100	<100	<10	<100	<2,0	<2,0	<2,0
WSP6232-34	Soil	0,0-0,3	<100	<100	<100	<10	<100	2,6	28	<2,0
WSP6232-35	Soil	0,0-0,3	<100	<100	<100	<10	<100	4,6	6,5	2,7
WSP6232-36	Soil	0,3-0,6	<100	<100	<100	<10	<100	<2,0	<2,0	<2,0
WSP6232-37	Soil	0,0-0,3	<100	<100	<100	<10	<100	5,1	5,4	4,5
WSP6232-38	Soil	0,3-0,6	<100	<100	<100	<10	<100	<2,0	<2,0	<2,0
WSP6232-39	Soil	0,0-0,3	<100	<100	<100	<10	<100	12	13	15
WSP6232-40	Soil	0,3-0,6	<100	<100	<100	<10	<100	<2,0	4,4	4,6
WSP6232-41	Soil	0,0-0,3	<100	<100	<100	<10	<100	11	18	5,9
WSP6232-42	Soil	0,3-0,6	<100	<100	<100	<10	<100	<2,0	<2,0	<2,0
WSP6232-43	Soil	0,0-0,2	<100	<100	<100	<10	<100	2,8	<2,0	<2,0
WSP6232-44	Soil	0,2-0,4	<100	<100	<100	<10	<100	2,5	3,8	<2,0
WSP6232-45	Soil	0,0-0,2	<100	<100	<100	<10	<100	22	4,2	7,4
WSP6232-46	Soil	0,2-0,4	<100	<100	<100	<10	<100	<2,0	<2,0	<2,0
WSP6232-47	Soil	0,0-0,2	<100	<100	<100	<100	<100	1,7	2,8	1,4
WSP6232-48	Soil	0,2-0,4	<100	<100	<100	<100	<100	2,6	5,9	<1,0

Table A3. *(Cont.)* Concentrations of analysed compounds in sludge (mg/kg dw), biota (µg/kg ww), soil (µg/kg dw).

Sample no	Media	Depth (m)	TCPP (tris(2-chloro-isopropyl)phosphate)	TCEP (tris (2-chloroethyl) phosphate)	TDCP (tris (dichlorpropyl) phos-fate)	TBP (tris-n-butyl phosfate)	TBEP (tris(2-butoxyethyl) phosphate)	TEHP (tris(2-ethylhexyl)phosphate)	TIBP (tri-iso-butylfphosphate)
WSP6232-1	Sludge		0,52	<0,050	0,061	<0,050	<0,35	0,37	<0,050
WSP6232-2	Sludge	0,0-0,6	0,75	<0,050	<0,050	<0,050	<0,050	0,23	<0,20
WSP6232-3	Crop		<50	NA	NA	NA	<100	NA	NA
WSP6232-4	Crop		<50	NA	NA	NA	<150	NA	NA
WSP6232-5	Crop		<50	NA	NA	NA	<100	NA	NA
WSP6232-6	Earthworm	0,0-0,2	NA	NA	NA	NA	NA	NA	NA
WSP6232-7	Soil	0,0-0,3	<20	<50	<50	<50	<100	<50	<50
WSP6232-8	Soil	0,0-0,3	<20	<50	<50	<50	<100	<50	<50
WSP6232-9	Soil	0,3-0,6	<20	<50	<50	<50	<100	<50	<50
WSP6232-10	Soil	0,0-0,3	<20	<50	<50	<50	<100	<50	<50
WSP6232-11	Soil	0,3-0,6	<20	<50	<50	<50	<100	<50	<50
WSP6232-12	Soil	0,0-0,3	<20	<50	<50	<50	<100	<50	<50
WSP6232-13	Soil	0,3-0,6	<20	<50	<50	<50	<100	<50	<50
WSP6232-14	Soil	0,0-0,3	<20	<50	<50	<50	<100	<50	<50
WSP6232-15	Soil	0,3-0,6	<20	<50	<50	<50	<100	<50	<50
WSP6232-16	Soil	0,0-0,3	<50	NA	NA	NA	<100	NA	NA
WSP6232-17	Soil	0,0-0,3	<50	NA	NA	NA	<100	NA	NA
WSP6232-18	Soil	0,3-0,6	<50	NA	NA	NA	<100	NA	NA
WSP6232-19	Soil	0,0-0,3	<50	NA	NA	NA	<100	NA	NA
WSP6232-20	Soil	0,3-0,6	<50	NA	NA	NA	<100	NA	NA
WSP6232-21	Soil	0,0-0,3	<50	NA	NA	NA	<100	NA	NA
WSP6232-22	Soil	0,3-0,6	<50	NA	NA	NA	<100	NA	NA
WSP6232-23	Soil	0,0-0,3	<50	NA	NA	NA	<100	NA	NA
WSP6232-24	Soil	0,3-0,6	<50	NA	NA	NA	<100	NA	NA
WSP6232-25	Soil	0,0-0,3	<20	<50	<50	<50	<100	<50	<50
WSP6232-26	Soil	0,0-0,3	<20	<50	<50	<50	<100	<50	<50
WSP6232-27	Soil	0,3-0,6	<20	<50	<50	<50	<100	<50	<50
WSP6232-28	Soil	0,0-0,3	20	<50	<50	<50	<100	<50	<50
WSP6232-29	Soil	0,3-0,6	<20	<50	<50	<50	<100	<50	<50
WSP6232-30	Soil	0,0-0,3	<20	<50	<50	<50	<100	<50	<50
WSP6232-31	Soil	0,3-0,6	<20	<50	<50	<50	<100	<50	<50
WSP6232-32	Soil	0,0-0,3	<20	<50	<50	<50	<100	<50	<50
WSP6232-33	Soil	0,3-0,6	<20	<50	<50	<50	<100	<50	<50
WSP6232-34	Soil	0,0-0,3	<50	NA	NA	NA	<100	NA	NA
WSP6232-35	Soil	0,0-0,3	<50	NA	NA	NA	<100	NA	NA
WSP6232-36	Soil	0,3-0,6	<50	NA	NA	NA	<100	NA	NA
WSP6232-37	Soil	0,0-0,3	<50	NA	NA	NA	<100	NA	NA
WSP6232-38	Soil	0,3-0,6	<50	NA	NA	NA	<100	NA	NA
WSP6232-39	Soil	0,0-0,3	<50	NA	NA	NA	<100	NA	NA
WSP6232-40	Soil	0,3-0,6	<50	NA	NA	NA	<100	NA	NA
WSP6232-41	Soil	0,0-0,3	<50	NA	NA	NA	<100	NA	NA
WSP6232-42	Soil	0,3-0,6	<50	NA	NA	NA	<100	NA	NA
WSP6232-43	Soil	0,0-0,2	<20	<50	<50	<50	<60	<50	<200
WSP6232-44	Soil	0,2-0,4	<20	<50	<50	<50	<50	<50	<50
WSP6232-45	Soil	0,0-0,2	<20	<50	<50	<50	<50	<50	<200
WSP6232-46	Soil	0,2-0,4	<20	<50	<50	<50	<50	<50	<200
WSP6232-47	Soil	0,0-0,2	<20	NA	NA	NA	<80	NA	NA
WSP6232-48	Soil	0,2-0,4	<20	NA	NA	NA	<50	NA	NA

Table A3. *(Cont.)* Concentrations of analysed compounds in sludge (mg/kg dw), biota (µg/kg ww), soil (µg/kg dw).

Sample no	Media	Depth (m)	TCrP (tri-cresyl phosphate)	ToCrP (trs-o-cresyl phosphate)	TPhP (trifphenyl phosphate)	DBPhP (dibutyl phenyl phosphate)	DPhBP (diphenylbutylphosphate)	EHDPhP (2-Ethylhexyl diphenyl phosphate)	PFOS	PFOA
WSP6232-1	Sludge		<0,50	<0,050	<0,050	<0,10	<0,050	0,36	0,022	0,0014
WSP6232-2	Sludge	0,0-0,6	<0,10	<0,02	<0,050	<0,10	<0,050	0,2	0,0095	0,0015
WSP6232-3	Crop		<80	NA	NA	NA	NA	<50	<2,0	<2,0
WSP6232-4	Crop		<80	NA	NA	NA	NA	<50	<2,0	<2,0
WSP6232-5	Crop		<80	NA	NA	NA	NA	<50	<2,0	<2,0
WSP6232-6	Earthworm	0,0-0,2	NA	NA	NA	NA	NA	NA	33	<10
WSP6232-7	Soil	0,0-0,3	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-8	Soil	0,0-0,3	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-9	Soil	0,3-0,6	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-10	Soil	0,0-0,3	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-11	Soil	0,3-0,6	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-12	Soil	0,0-0,3	<100	<50	<50	<100	<50	<25	2	<2,0
WSP6232-13	Soil	0,3-0,6	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-14	Soil	0,0-0,3	<100	<50	<50	<100	<50	<25	2,7	<2,0
WSP6232-15	Soil	0,3-0,6	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-16	Soil	0,0-0,3	<500	NA	NA	NA	NA	<50	<2,0	<2,0
WSP6232-17	Soil	0,0-0,3	<500	NA	NA	NA	NA	<50	<2,0	<2,0
WSP6232-18	Soil	0,3-0,6	<500	NA	NA	NA	NA	<50	<2,0	<2,0
WSP6232-19	Soil	0,0-0,3	<500	NA	NA	NA	NA	<50	<2,0	<2,0
WSP6232-20	Soil	0,3-0,6	<500	NA	NA	NA	NA	<50	<2,0	<2,0
WSP6232-21	Soil	0,0-0,3	<500	NA	NA	NA	NA	<50	5,1	<2,0
WSP6232-22	Soil	0,3-0,6	<500	NA	NA	NA	NA	<50	2,4	<2,0
WSP6232-23	Soil	0,0-0,3	<500	NA	NA	NA	NA	<50	3,7	<2,0
WSP6232-24	Soil	0,3-0,6	<500	NA	NA	NA	NA	<50	2,3	<2,0
WSP6232-25	Soil	0,0-0,3	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-26	Soil	0,0-0,3	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-27	Soil	0,3-0,6	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-28	Soil	0,0-0,3	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-29	Soil	0,3-0,6	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-30	Soil	0,0-0,3	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-31	Soil	0,3-0,6	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-32	Soil	0,0-0,3	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-33	Soil	0,3-0,6	<100	<50	<50	<100	<50	<25	<2,0	<2,0
WSP6232-34	Soil	0,0-0,3	<500	NA	NA	NA	NA	<50	<2,0	<2,0
WSP6232-35	Soil	0,0-0,3	<500	NA	NA	NA	NA	<50	2,5	<2,0
WSP6232-36	Soil	0,3-0,6	<500	NA	NA	NA	NA	<50	<2,0	<2,0
WSP6232-37	Soil	0,0-0,3	<500	NA	NA	NA	NA	<50	2	<2,0
WSP6232-38	Soil	0,3-0,6	<500	NA	NA	NA	NA	<50	<2,0	<2,0
WSP6232-39	Soil	0,0-0,3	<500	NA	NA	NA	NA	<50	3,3	<2,0
WSP6232-40	Soil	0,3-0,6	<500	NA	NA	NA	NA	<50	2,3	<2,0
WSP6232-41	Soil	0,0-0,3	<500	NA	NA	NA	NA	<50	3,7	<2,0
WSP6232-42	Soil	0,3-0,6	<500	NA	NA	NA	NA	<50	2	<2,0
WSP6232-43	Soil	0,0-0,2	<100	<0,02	<50	<100	<50	<20	<1,0	<1,0
WSP6232-44	Soil	0,2-0,4	<20	<0,050	<50	<100	<50	<20	<1,0	<1,0
WSP6232-45	Soil	0,0-0,2	<100	<0,02	<50	<100	<50	<20	<1,0	<1,0
WSP6232-46	Soil	0,2-0,4	<100	<0,02	<50	<100	<50	<20	<1,0	<1,0
WSP6232-47	Soil	0,0-0,2	<20	NA	NA	NA	NA	<20	2,6	<1,0
WSP6232-48	Soil	0,2-0,4	<20	NA	NA	NA	NA	<20	<1,0	<1,0

Table A3. *(Cont.)* Concentrations of analysed compounds in sludge (mg/kg dw), biota (µg/kg ww), soil (µg/kg dw).

Sample no	Media	Depth (m)	BDE 209	BDE 47	BDE 99	BDE 100
WSP6232-1	Sludge		0,28	0,02	0,0072	0,0023
WSP6232-2	Sludge	0,0-0,6	0,82	0,022	0,021	0,0049
WSP6232-3	Crop		<5,0	<0,20	<0,20	<0,20
WSP6232-4	Crop		<5,0	<0,20	<0,20	<0,20
WSP6232-5	Crop		<5,0	<0,20	<0,20	<0,20
WSP6232-6	Earthworm	0,0-0,2	<5,0	0,42	0,38	0,11
WSP6232-7	Soil	0,0-0,3	<1,0	<0,050	<0,050	<0,050
WSP6232-8	Soil	0,0-0,3	3,6	0,21	0,19	0,055
WSP6232-9	Soil	0,3-0,6	<1,0	0,089	0,078	<0,050
WSP6232-10	Soil	0,0-0,3	1,4	0,23	0,22	0,064
WSP6232-11	Soil	0,3-0,6	<1,0	<0,050	<0,050	<0,050
WSP6232-12	Soil	0,0-0,3	3,5	0,51	0,5	0,13
WSP6232-13	Soil	0,3-0,6	1,2	0,13	0,13	<0,050
WSP6232-14	Soil	0,0-0,3	2,8	0,4	0,41	0,1
WSP6232-15	Soil	0,3-0,6	1,2	0,14	0,13	<0,050
WSP6232-16	Soil	0,0-0,3	<1,0	<0,050	<0,050	<0,050
WSP6232-17	Soil	0,0-0,3	2,1	0,1	0,098	<0,050
WSP6232-18	Soil	0,3-0,6	1,5	0,061	0,071	<0,050
WSP6232-19	Soil	0,0-0,3	1,7	0,12	0,11	<0,050
WSP6232-20	Soil	0,3-0,6	2	0,054	0,053	<0,050
WSP6232-21	Soil	0,0-0,3	5,9	0,24	0,26	<0,050
WSP6232-22	Soil	0,3-0,6	3,3	0,14	0,16	<0,050
WSP6232-23	Soil	0,0-0,3	5,1	0,26	0,28	<0,050
WSP6232-24	Soil	0,3-0,6	2,8	0,14	0,17	<0,050
WSP6232-25	Soil	0,0-0,3	<1,0	<0,050	<0,050	<0,050
WSP6232-26	Soil	0,0-0,3	2,9	0,3	0,27	0,076
WSP6232-27	Soil	0,3-0,6	3	0,1	0,083	<0,050
WSP6232-28	Soil	0,0-0,3	4,2	0,31	0,27	0,072
WSP6232-29	Soil	0,3-0,6	1,6	0,1	0,084	<0,050
WSP6232-30	Soil	0,0-0,3	5,8	0,54	0,52	0,14
WSP6232-31	Soil	0,3-0,6	1,1	0,19	0,19	0,051
WSP6232-32	Soil	0,0-0,3	3,8	0,51	0,49	0,13
WSP6232-33	Soil	0,3-0,6	2,2	0,18	0,17	<0,050
WSP6232-34	Soil	0,0-0,3	<1,0	<0,050	<0,050	<0,050
WSP6232-35	Soil	0,0-0,3	6	0,21	0,2	<0,050
WSP6232-36	Soil	0,3-0,6	2,1	0,11	0,12	<0,050
WSP6232-37	Soil	0,0-0,3	4,5	0,39	0,37	0,68
WSP6232-38	Soil	0,3-0,6	1	0,085	0,088	<0,050
WSP6232-39	Soil	0,0-0,3	14	0,39	0,42	0,094
WSP6232-40	Soil	0,3-0,6	16	0,2	0,23	<0,050
WSP6232-41	Soil	0,0-0,3	12	0,33	0,35	0,069
WSP6232-42	Soil	0,3-0,6	4,6	0,17	0,21	<0,050
WSP6232-43	Soil	0,0-0,2	<5,0	<0,050	<0,050	<0,050
WSP6232-44	Soil	0,2-0,4	<5,0	<0,050	<0,050	<0,050
WSP6232-45	Soil	0,0-0,2	<5,0	<0,11	<0,050	<0,050
WSP6232-46	Soil	0,2-0,4	<5,0	<0,050	<0,050	<0,050
WSP6232-47	Soil	0,0-0,2	12	0,1	<0,050	<0,050
WSP6232-48	Soil	0,2-0,4	<5,0	0,052	0,068	<0,050