



SSPI-CT-2003-502411

HORIZONTAL - ORG

**HORIZONTAL STANDARDS ON ORGANIC MICRO-POLLUTANTS FOR IMPLEMENTATION OF
EU DIRECTIVES ON SLUDGE, SOIL AND TREATED BIO-WASTE**

Instrument: STREP

Thematic Priority: PRIORITY 8.1 STREP Topic 1.5
Environmental assessment

**Deliverable D2.6 Research report on the
application of activity models for organic
parameters in sludges, treated biowastes and
soil**

Due date of deliverable:

Actual submission date:

Start date of project: 1-10-2003

Duration: 3 years

Organisation name of lead contractor for this
deliverable: UREADS

Revision [draft, 1]

Project co-funded by the European Commission within the Sixth Framework Programme (2002-2006)		
Dissemination Level		
PU	Public	PU
PP	Restricted to other programme participants (including the Commission Services)	
RE	Restricted to a group specified by the consortium (including the Commission Services)	
CO	Confidential, only for members of the consortium (including the Commission Services)	

Disclaimer

The European Commission does not accept any liability for the information contained in this document resulting from work carried out under contract with the FP 6 program of the European Commission,. The conclusions herein reflect at this point the opinion of the authors. After consultation, it can be considered to reflect the consensus opinion of the CEN bodies involved in project HORIZONTAL. The opinions are not necessarily consistent with the views of the European Commission. HORIZONTAL-ORG

WP2-d2.5a: Research Report

Draft Version July 2007

HORIZONTAL-ORG WP2

Research report on the application of activity models for organic parameters in sludges, treated biowastes and soil

Authors: D C Lambkin¹, T C White², and S Nortcliff¹

¹Department of Soil Science, The University of Reading

Whiteknights, Reading, UK

²Marquis and Lord, Stratford upon Avon, UK



Summary

A task of the Sampling Work Package was to investigate how many samples of treated sewage sludge, treated biowaste or soil would be required to evaluate a particular parameter with a specified confidence level and interval. Sampling designs can be investigated by analysing samples using microbiological/chemical methods and this has been done for metals, *E. coli*, PAH and PCB. However, it is not practical to analyse samples for all relevant organic micropollutants for reasons of cost and/or lack of well-defined analytical methods.

It was hypothesized that an indirect analytical approach, based on some generic rules relating to low-cost test parameters, might provide a tool to guide the choice of soil sampling schemes and consequently to improve confidence in the sampling results.

The number of samples required to estimate a population mean characteristic with specified confidence (e.g. mean Loss on Ignition in a field) increases as the within-population variability of the characteristic increases. It was supposed that, if concentrations of organic micropollutants were related to low-cost test parameters such as the pH and/or organic matter content of the matrix, then:

- (i) knowledge of pH and/or organic matter variability might be used to estimate the minimum number of samples needed to estimate mean organic micropollutant concentrations for a population with specified confidence;
- (ii) knowledge of pH and/or organic matter distribution within a population might be used to target sampling for organic micropollutants.

This report describes the use of Structure-Activity modelling to identify those compounds whose measured variability in concentration in a given matrix might correlate with low-cost, easily measured parameters, such as pH and organic carbon content.

Limited datasets for sewage sludge, compost and soil were used to test the possibility of using pH and LOI to inform the sampling strategy for PAH and PCB concentrations in these matrices.

Main Conclusions

pH-dependent variability in solubility and K_{OC} was modelled for six groups of compounds. For three groups the model did not identify pH-dependent variability in either measurement. For NPE, NP and LAS pH-dependent variability was predicted at pH extremes.

Laboratory data (PAH, PCB, LOI, pH) were examined to assess if variability in organic carbon and modelled K_{OC} or pH could explain variation in compound concentration. There were insufficient analytical data for rigorous testing of the results, but examination of the available data provided no evidence to support co-variation in sludge, co-composted sludge, or soil.

Further examination of the data indicated that, for the two Wastewater Treatment Works investigated, there was strong correlation between concentrations of different substances within a group of compounds, i.e. either PAH or PCB. This may be due to lack of variability in feed stock and/or the high degree of mixing during processing, either of which could have produced a high degree of homogeneity in the treated material that was sampled.

Only one soil data set was examined, but, as for the sludge, there was strong correlation between concentrations of different substances within a group of compounds. It was hypothesized that the factors

controlling PCB spatial variability in the field are (i) variability in the applied material and (ii) variability in the application rate. Support for this hypothesis was demonstrated using example data.

Although almost 600 samples from 13 sites were collected and analysed for PAH and PCB few of these contained PAH and PCB above the limit of quantification. Consequently the number of sites for which adequate data was available for examination was limited. This means that the results have to be treated with caution and may not be transferable to other sites or, more importantly, to other compound groups.

It has been recommended in earlier work that, since the number of samples required for a given precision varies between sites, treatment sites should be characterised during commissioning. Based on this work, it is likely that the number of samples required for organic micropollutant analysis can be determined by measurement of a small number such compounds.

Recommendations

1. Further sampling and analysis should be carried out to test and confirm the results from this study, which was limited by the lack of available data sets.
2. This work has demonstrated a strong correlation between concentrations of PAH groups of compounds and between PCB groups of compounds. Future sampling should include matrices with measurable concentrations of both PAH and PCB to enable investigation of between group correlation.
3. Once a treatment site has been characterised, it is likely that the variability of concentrations of many substances can be inferred from the characteristics of perhaps one or two compounds, unless there are major changes to the feedstock.
4. The chemical analysis should be extended to other groups of compounds to test for correlation within and between groups of compounds.

Contents

SUMMARY	I
CONTENTS	III
LIST OF TABLES	IV
LIST OF FIGURES	V
1 INTRODUCTION	1
1.1 General.....	1
1.2 Aim	1
1.3 Scope	1
1.4 Limitations and assumptions	2
2 EXPERIMENTAL	3
2.1 Introduction	3
2.2 Method	4
2.2.1 General.....	4
2.2.2 Identification of compounds for modelling	5
2.2.3 Modelling solubility and K_{oc}	5
2.2.4 Sample collection and analysis.....	6
2.3 Results	6
2.3.1 Identification of compounds for modelling	6
2.3.2 Modelling	6
2.4 Translation to Field Studies	17
2.4.1 Investigation of PAH and PCB variability	17
2.4.2 Investigation of pH-dependent compounds	22
2.5 Conclusions	23
3 RECOMMENDATIONS	24
REFERENCES	25
ANNEX A ACD/SOLUBILITY DB	29
A.1 Introduction	29
A.2 Key Capabilities	29
A.3 Input/Output.....	29
A.4 Comparison with experiment.....	29
ANNEX B TYPICAL ENVIRONMENTAL CONCENTRATIONS	31
ANNEX C MODELLED COMPOUNDS: K_{oc} AND SOLUBILITY AT PH 7	37
ANNEX D FIELD DATA	40
D.1 Introduction	40

List of Tables

Table 2.1	NPE compounds: Key for figures 2.4 and 2.5	12
Table 2.2	Summary of K_{OC} and Solubility sensitivity to pH.....	13
Table 2.3	Soil (Spain Field 2): PCB concentration – Correlation Matrix	18
Table 2.4	Co-composted sewage sludge (STWB): PCB concentration – Correlation Matrix.....	19
Table 2.5	Thermally dried sewage sludge (STWE): PAH concentration – Correlation Matrix	20
Table 2.6	Example 1: Compound concentrations in sludge	21
Table 2.7	Example: Application rates and resulting compound concentrations	21
Table A.1	ACD/Solubility DB - Accuracy of calculations	30
Table B.1	Typical Environmental Concentrations: Linear Alkyl Sulfonates	31
Table B.2	Typical Environmental Concentrations: Furans and dioxins.....	32
Table B.3	Typical Environmental Concentrations: Nonylphenols and nonylphenol ethoxylates	33
Table B.4	Typical Environmental Concentrations: Phthalates	34
Table B.5	Typical Environmental Concentrations: Brominated Fire Retardants	35
Table B.6	Typical Environmental Concentrations: Pharmaceuticals	36
Table C.1	Modelled K_{OC} and Solubility at pH 7	37
Table D.1	Spain Field 2: Analytical results.....	41
Table D.2	STWB: Analytical results	45
Table D.3	STWE: Analytical results	46

List of Figures

Figure 2.1	Solubility and K_{OC} at pH 7: Ranges for compound groups studied.....	8
Figure 2.2	PCB compounds: Solubility and K_{OC} at pH 7.0.....	9
Figure 2.3	PAH compounds: Solubility and K_{OC} at pH 7.0.....	10
Figure 2.4	NPE compounds: Range of solubility and K_{OC} at pH 7.0	11
Figure 2.5	NPE compounds: Solubility and K_{OC} variability with pH.....	14
Figure 2.6	NP isomers: Solubility and K_{OC} variability with pH	15
Figure 2.7	LAS compounds: solubility and K_{OC} variability with pH	16
Figure 2.8	Soil (Spain Field 2): Concentration of six PCB compounds relative to PCB 180.....	18
Figure 2.9	Co-composted sewage sludge (STWB): Concentration of six PCB compounds relative to PCB 101.....	19
Figure 2.10	Thermally dried sewage sludge (STWE): Concentration of 11 PAH compounds relative to pyrene	20
Figure 2.11	Example: Relationship between sampled compound concentrations	22

1 INTRODUCTION

1.1 General

A task of the Sampling Work Package was to investigate how many samples of treated sewage sludge, treated biowaste or soil would be required to evaluate a particular parameter with a specified confidence level and interval.

Sampling designs can be investigated by analysing samples using microbiological/chemical methods and this has been done for metals, *E. coli*, PAH and PCB [Lambkin et al., 2004, 2006, 2007]. However, it is not practical to analyse samples for all relevant organic micropollutants for reasons of cost and/or lack of well-defined analytical methods.

It was hypothesized that an indirect analytical approach, based on some generic rules relating to low-cost test parameters, might provide a tool to guide the choice of soil sampling schemes and consequently to improve confidence in the sampling results.

The number of samples required to estimate a population mean characteristic with specified confidence (e.g. mean pH in a field) increases as the within-population variability of the characteristic increases. It was supposed that, if concentrations of organic micropollutants were related to low-cost test parameters such as the pH and/or organic matter content of the matrix, then:

- (i) knowledge of pH and/or organic matter variability might be used to estimate the minimum number of samples needed to estimate mean organic micropollutant concentrations for a population with specified confidence;
- (ii) knowledge of pH and/or organic matter distribution within a population might be used to target sampling for organic micropollutants.

1.2 Aim

Structure-Activity modelling is a well-established method used by industry, such as pharmaceutical and pesticide research and development. The aim of this work was to use a well-established chemometric model to identify those compounds whose measured variability in concentration in a given matrix might correlate with low-cost, easily measured parameters, such as pH and organic carbon content.

1.3 Scope

The scope of the investigation was:

1. To identify a list of organic compounds, that are likely to be of regulatory interest, for which analytical methods were being investigated and developed within project Horizontal.
2. To use structure-activity modelling to determine which of the compounds, or groups of compounds, would be likely to vary spatially due to the influence of pH and organic matter content.
3. To determine if confidence levels might be improved by amending the sampling scheme for those compounds where spatial variability was likely to be influenced by pH and organic matter content.
4. To make recommendations on how such information might assist the development of a sampling plan.

1.4 Limitations and assumptions

Sludge and treated biowaste sample collection was limited to the UK; therefore the range of materials and sampling sites chosen reflects those that are readily available in the UK. The results may not be transferable to other sites in the UK or to sites in other countries.

Similarly, adequate data for organic micropollutants in soil was available for only one field, located in Spain.

The range of materials examined reflects the availability of suitable samples, i.e. sludge and soil samples with analytical results above the limit of quantification.

Sampling had to be practicable. Since sampling was carried out at commercial sites some limitations were imposed on where and when samples could be collected. Wherever possible, samples were collected at sampling locations where regulatory samples would normally be collected – as advised by site staff.

All of the samples were analysed by a single laboratory operating recognised quality assurance and control procedures. Therefore it was considered justified to assume that the precision of sample preparation, extraction and analysis was consistent for all testing and that any differences in data sets were attributable to the sampling methodologies.

2 EXPERIMENTAL

2.1 Introduction

A task of the Sampling work package was to investigate how many samples of sludge, treated biowaste or soil should be collected and analysed to estimate the concentration of organic micropollutants with quantified precision.

Sampling designs can be investigated by analysing samples using microbiological, physical and chemical methods and this has been done for physicochemical parameters, metals, *E. coli*, PAHs and PCBs (Lambkin et al., 2004 and 2006, 2007). However, it is not practical to analyse samples for all relevant organic micropollutants for reasons of cost and/or lack of well-defined analytical methods. A non-analytical approach may provide an alternative method of predicting the distribution of these compounds and consequently the number of samples to be taken.

The number of samples required ultimately depends on the variability of organic pollutants in the material to be sampled. Many factors can determine variability and these are not the same for all materials. The main differences will be between materials that are to be landspread, i.e. sludge and treated biowaste, and the land to which they are applied.

Factors affecting variability in applied materials include:

- Variability in feedstock over different timescales, e.g. hours, weeks, seasons;
- The degree of mixing during processing;
- Variability in chemical factors affecting losses during processing, i.e. adsorption and solubility.

Factors affecting variability in soils to which the materials are applied include:

- Variability in the materials applied;
- Variability in application rates;
- Variability in mixing, e.g. during ploughing;
- Variability in factors affecting environmental fate, i.e. adsorption, solubility and degradation.

It might be assumed that most factors affecting variability are common to all organic micropollutants for example, degree of mixing and application rates. If that is the case then it follows that the main factors affecting variability are those affecting losses during processing and environmental fate, i.e. adsorption, solubility and degradation. These are in turn controlled, in the main, by organic carbon content and pH.

Measured K_{OC} (organic carbon adsorption coefficient) and solubility have not been reported for all organic micropollutants. Reported values of specific physicochemical properties can vary by several orders of magnitude due to differences in analytical methodology and the constraints of analytical equipment (Cousins and Mackay, 2000). pH-dependent K_{OC} and solubility measurements are rarely reported in the literature, probably because many measurements would be close to the limit of analytical detection. Chemical modelling provides a method of producing pH-dependent data that is free of the limitations of laboratory analysis.

Quantitative structure-property relationships (QSPR), also called structure-activity relationships (QSAR) when biological processes are included, use chemometric methods to describe how a given activity or physicochemical property varies as a function of molecular descriptors that describe the chemical structure of the molecule (de Lima Ribeiro et al., 2003). In this way it is possible to replace costly biological tests or experiments to measure physicochemical properties with calculated descriptors, which can be used to predict responses for other compounds. For example, Ferreira (2001) used QSAR methodology to predict environmentally relevant physicochemical properties of PAHs and toxicity to *Daphnia magna*, a freshwater organism.

Since QSAR can be used for the prediction of biological activity, it lends itself readily to the prediction of environmental toxicity and as an alternative to animal testing for toxicity. Over the last 20 years environmental QSAR has increased steadily in importance and has reached the stage where some regulatory agencies, such as USEPA, routinely use some QSAR-predicted toxicities for regulatory purposes (Dearden, 2002), for example in development of new compounds such as pharmaceuticals and pesticides. In a review of the regulatory use of QSARs for ecologic effects and environmental fate, Cronin et al. (2003) discussed their use by three EU Member States (Denmark, Germany and the Netherlands), the USA, Canada, Australia and Japan.

Chemical modelling has been used to predict for organic compounds aqueous solubility (Dunnivant et al., 1992) and sorption coefficients (Meylan et al., 1992). Predictions of physicochemical properties using QSARs have been used to aid the screening and prioritisation of a range of chemicals that may enter wastewater treatment works (Webber et al., 1996). Physicochemical properties can be used to predict the likelihood of loss during wastewater treatment. For example, García et al. (2002) used QSAR to investigate the effect of chain length and water hardness on sorption of linear alkylbenzene sulfonates (LAS) on sewage sludge. A wide range of potential organic pollutants (POPs) have been studied, including PCB (Dunnivant et al., 1992; Öberg, 2001), PAH (Ferreira, 2001; de Lima Ribeiro and Ferreira, 2003); phthalates (Cousins and MacKay, 2000) and linear alkylbenzene sulfonates (García et al., 2002).

The fate of organic micropollutants is determined to a great extent by the physicochemical properties of the chemical, such as solubility and sorption. Matrix properties, such as organic carbon content (OC) and pH, which affect solubility and sorption and hence retention, vary spatially; therefore it is likely that the distribution of organic micropollutants also varies spatially.

If compound adsorption and solubility are not affected by pH, then variability in the sampled materials should be independent of pH. However, if adsorption and solubility are pH-dependent the compounds might exhibit greater spatial variability than that predicted from the variability of soil organic carbon.

To address the specific aspect of sampling sludge, soil and treated biowaste for organic micropollutants, samples were collected and analysed for pH, Loss on Ignition (LOI, as a surrogate measurement of organic carbon), PAHs and PCBs. These parameters are relatively easy and cheap to measure (pH and Loss on Ignition) and well-defined analytical methods are available (PAHs and PCBs).

A commercially available model was used to predict the variation of solubility and sorption with pH of several groups of organic micropollutants. The results are discussed in relation to variability of PAHs and PCBs. The aim was to investigate how the knowledge of variability in pH, LOI, PAHs and PCBs could be extended to a wider range of compounds.

2.2 Method

2.2.1 General

The work was broken down into four stages:

- (i) Identify compounds for modelling;
- (ii) Model the effect of pH on compound solubility and K_{OC} ;
- (iii) Collect samples of soil, sludge and treated biowaste and analyse for pH, LOI, PAHs and PCBs;
- (iv) Evaluate the results of the modelling in light of the field data.

2.2.2 Identification of compounds for modelling

A range of candidate compounds was investigated:

- linear alkylbenzene sulfonates (LAS);
- brominated fire retardants (BFRs);
- furans and dioxins (PCDF and PCDD);
- polychlorinated diphenyl ethers (PCDEs);
- pharmaceuticals;
- nonylphenols;
- phthalates.

These compounds are not routinely analysed in agricultural soils, treated biowaste or sewage sludge by UK laboratories and it was recognised that many commercial laboratories would probably regard these analysis as non-standard.

They were considered for suitability for chemical modelling based on the following criteria:

- (i) The compounds are the subject of reports and draft EN standards produced by project Horizontal [<http://www.ecn.nl/horizontal/phase2/Organ/>];
- (ii) Evidence of their existence in the matrices of interest should be available in the literature;
- (iii) Sufficient physicochemical data should be available for modelling.

Chemical modelling requires reliable data on the physicochemical properties of the compounds to be modelled. A literature search was conducted for each group of compounds to collate information about (a) physicochemical properties and (b) the levels to be expected in each of the matrices of interest. Based on the amount of information available, a shortlist of candidate compounds was selected.

2.2.3 Modelling solubility and K_{oc}

The model suite ACD/Solubility DB v8.07 (Advanced Chemistry Development Inc., Toronto, Canada) was used to predict solubility and K_{oc} over the full pH range. This program calculates aqueous solubility values at any pH at 25°C and zero ionic strength. The accuracy of calculations for simple structures is usually better than 0.2-0.5 logS units for simple structures and better than 0.5-1.0 logS units for complex structures.

Additional information on the ACD/Solubility DB model is given in Annex A.

The original plan had been to use PCKOCWIN, a programme in the EPI Suite™ [<http://www.epa.gov/opptintr/exposure/pubs/episuite.htm>], to estimate the soil adsorption coefficient, K_{oc} . PCKOCWIN estimates K_{oc} from the first-order molecular connectivity index and 27 polar, structural correction factors (Meylan et al., 1992). However, on further investigation, it was discovered that PCKOCWIN calculates K_{oc} at pH 7 and does not produce values of K_{oc} at a range of pH, which was required to fulfil the aims of modelling.

ACD/Solubility DB was used to estimate both solubility and the soil adsorption coefficient, K_{oc} over the full pH range.

2.2.4 Sample collection and analysis

Sample collection and analysis is described elsewhere (Lambkin et al., 2007). Analytical data were available for a range of materials: Soils collected from Poland and Spain, and sludge and treated biowaste from the UK. The samples had been analysed at a commercial laboratory for loss on ignition, pH, moisture content, and a range of PAHs (EPA 16) and PCBs (IES 7). Full results are listed in Lambkin et al. (2007).

2.3 Results

2.3.1 Identification of compounds for modelling

After an initial screening of the literature it was decided to exclude two compound groups from further study:

- brominated fire retardants (BFRs); and
- pharmaceuticals;

Firstly, each of these groups includes a wide range of substances and secondly, few published studies were found that provided evidence of their widespread existence in the matrices of interest. Also, it was considered that there is currently no regulatory interest in these compounds.

It was therefore decided that the following criteria would be used to populate the lists of compounds to be modelled. The substances chosen should be:

- compounds of regulatory interest as named substance; and/or
- proposed as calibration standards for analytical methods being developed in Project Horizontal; and
- implicated in the literature as environmental contaminants in association with substances identified in one or both of the above two qualifying categories.

In addition, but of secondary importance, availability of some measured physicochemical parameters was considered desirable as input to the model software to allow improved self-correction.

In reality, for some substances there was a paucity of information, such as melting points, which the software needed to generate an improved prediction. This was because, for some substances, the parameters have not been measured; for example, melting points have not been measured for many of the nonylphenol isomers.

Also, in some cases, two or more measurements for a given compound were reported in the literature, although in most cases the variation was slight. Assessing the variability in measurements was outside the scope of this work and it was considered that bias might be introduced by applying a correction for the variability. Instead it was decided to run the model in its default condition and allow the software to operate without manual entry of melting points so that there was consistent operation across all the groups of compounds studied.

The substances selected are listed in Appendix 2.

2.3.2 Modelling

Solubility and K_{OC} at pH7

For each group of compounds the value of solubility and K_{OC} at pH 7 was modelled. The results for individual compounds are in Table C.1 (Annex C) and summarised in Figure 2.1.

Solubility and K_{OC} at pH 7 for PCB and PAH compounds are shown in Figures 2.2 and 2.3. For these two groups of compounds solubility decreased and K_{OC} increased with molecular weight. The same result was found for phthalates, although only two phthalate compounds were modelled, and for LAS (three compounds) and NPE compounds (Figure 2.4). Similar observations have been reported widely in the literature, e.g. García et al. (2002) for LAS and Ferreira (2001) for PAH.

NP is a mixture typically of at least 13 isomers with branched C_9 - C_{19} -chains; the two main isomers have two methyl groups at the C_1 position in the chain. As pointed out by Fremmersvik and Hansen (2004) there may be confusion about the identity of nonylphenols because the CAS numbers are not very precise. There are two CAS numbers for nonylphenols: 104-40-5, described as '4-nonylphenol', and 25154-52-3, described as 'nonylphenol' or '4-nonylphenol, mixture of isomers'.

Fifteen nonylphenol isomers were identified that it was reported could be separated using a published method (Gundersen, 2001; Ieda et al., 2005). Since they are isomers with the same molecular formula, there could be no relationship between modelled values and molecular weight. However, K_{OC} decreased and solubility increased as the degree of branching in the nonyl side chain increased.

Two NPEs were identified as being of regulatory interest and included in analytical method development and ruggedness testing within project Horizontal. The compounds listed were: Nonylphenol-monoethoxylates (NP1EO) and Nonylphenol-diethoxylates (NP2EO), both of which were listed as a mixture of isomers.

Modelled K_{OC} and solubility at pH 7 varied within and between groups of compounds. Mean K_{OC} decreased in the order Phthalate > PCB > PAH > NP > NPE > LAS. Mean solubility increased in the order PCB < Phthalate < PAH < NPE < LAS < NP.

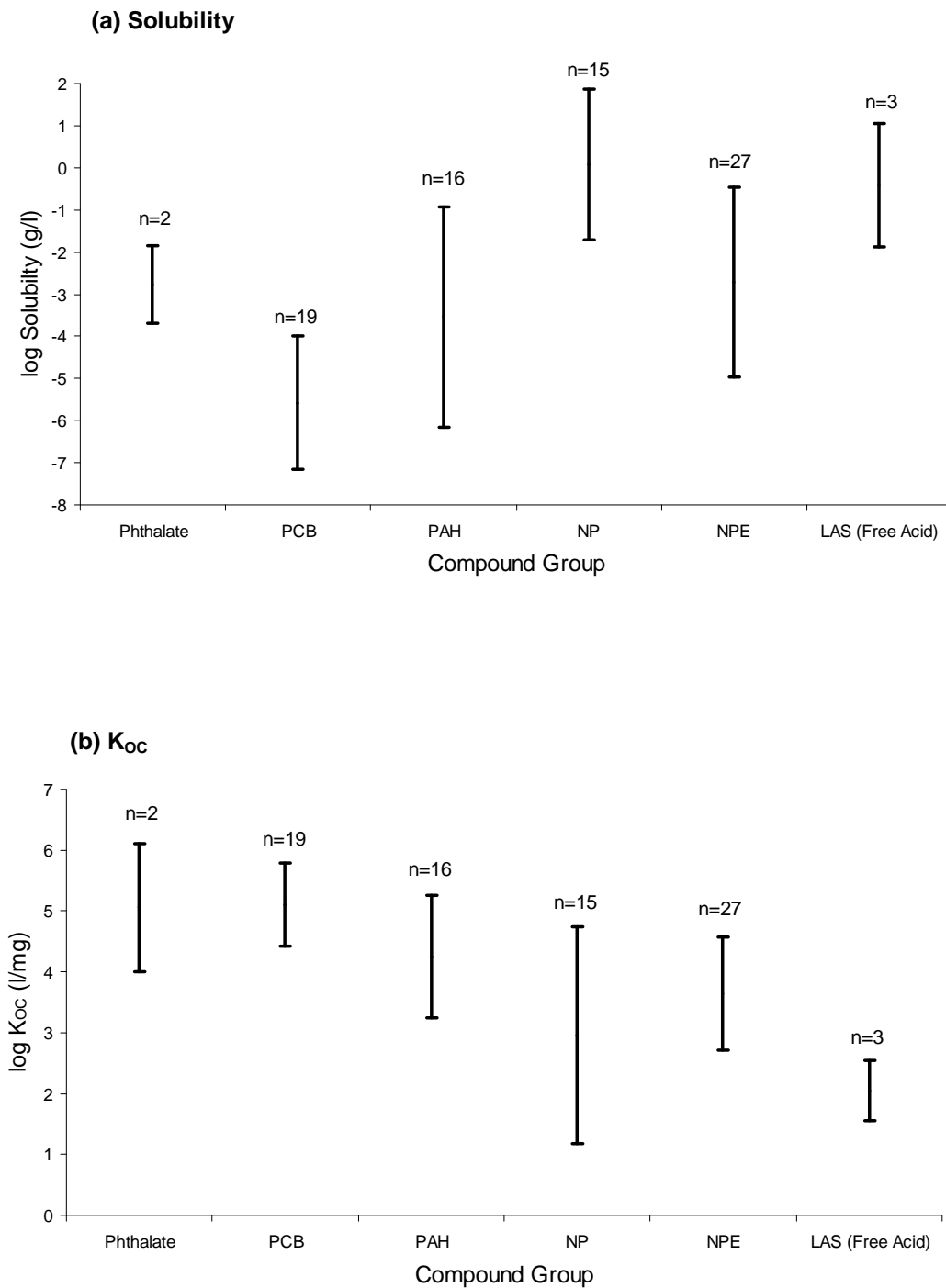
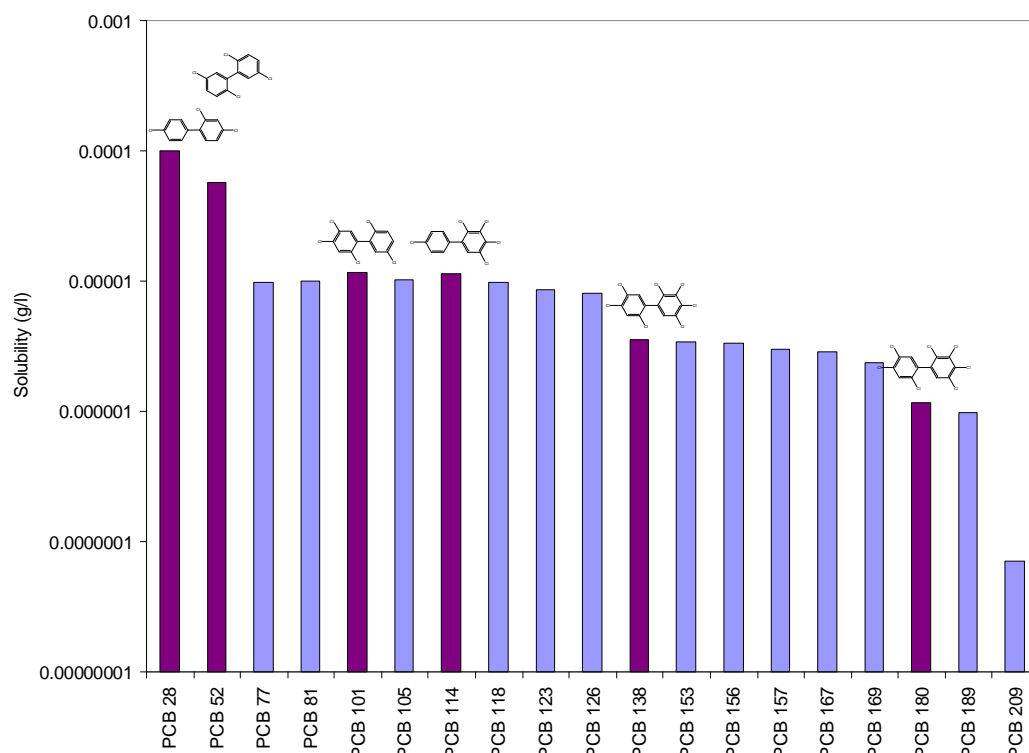


Figure 2.1 Solubility and K_{OC} at pH 7: Ranges for compound groups studied

(a) Solubility



(b) K_{oc}

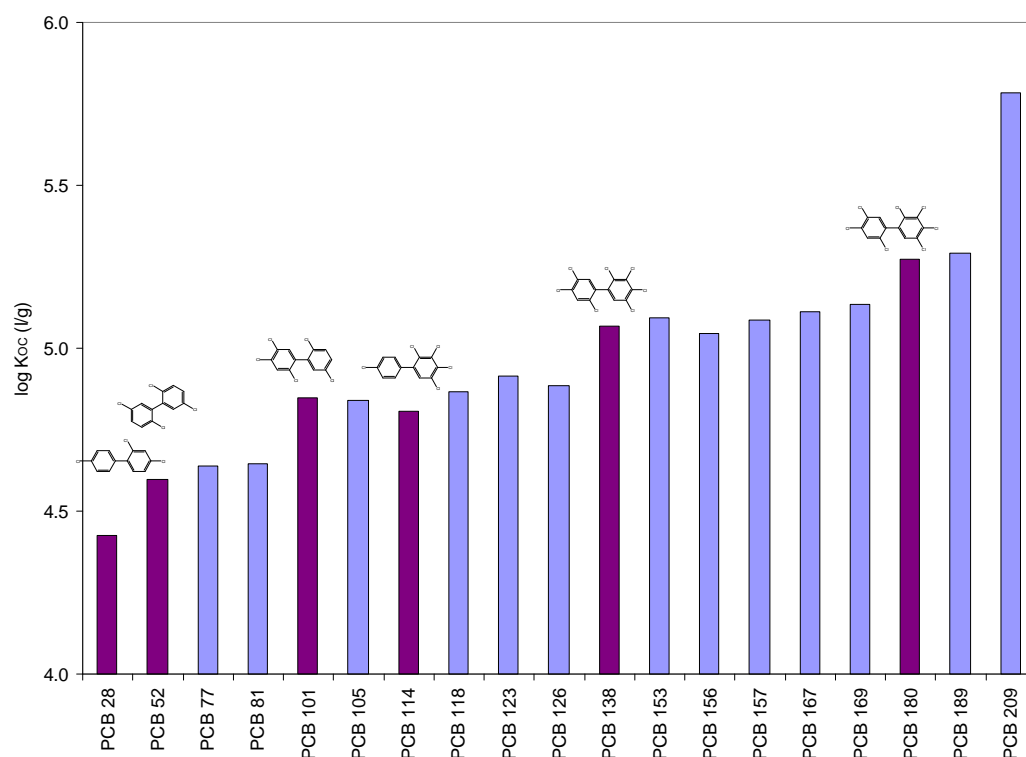
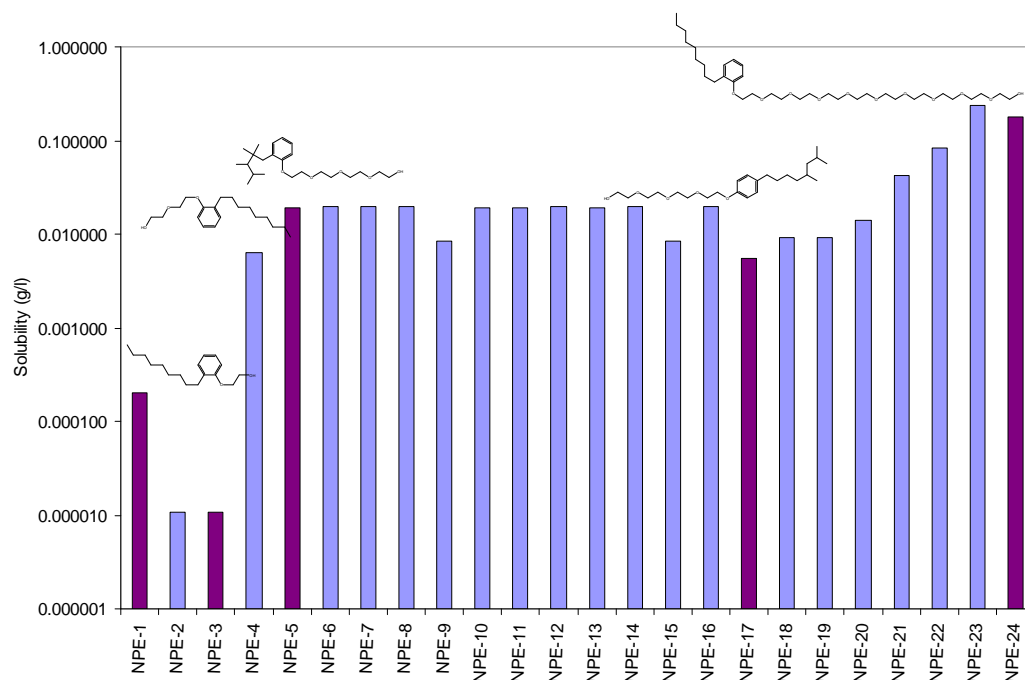


Figure 2.2 PCB compounds: Solubility and K_{oc} at pH 7.0

(a) Solubility

(b) K_{oc}

(a) Solubility



(b) K_{OC}

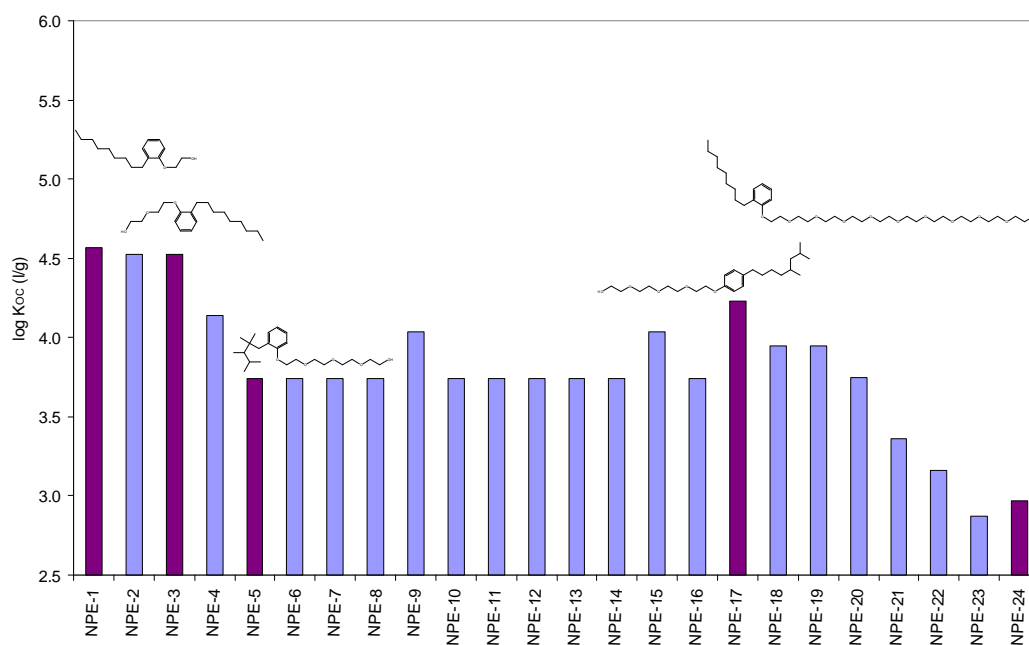


Figure 2.4 NPE compounds: Range of solubility and K_{OC} at pH 7.0

Table 2.1 NPE compounds: Key for figures 2.4 and 2.5

Key	NPE Compound
NPE-1	2-(nonylphenoxy)ethanol
NPE-2	2-[2-(4-nonylphenoxy)ethoxy]ethanol
NPE-3	2-[2-(nonylphenoxy)ethoxy]ethanol
NPE-4	2-[2-[2-[2-(4-nonylphenoxy) ethoxy]ethoxy]ethoxy]ethanol
NPE-5	2-(2-(2-(2-(2-(2,2,3,4-tetramethylpentyl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol
NPE-6	2-(2-(2-(2-(2-(2,3,5-trimethylhexan-2-yl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol
NPE-7	2-(2-(2-(2-(2-(3,3,5-trimethylhexan-2-yl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol
NPE-8	2-(2-(2-(2-(2-(4,4,5-trimethylhexan-2-yl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol
NPE-9	2-(2-(2-(2-(2-(7-methyloctyl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol
NPE-10	2-(2-(2-(2-(4-(2,2,3,4-tetramethylpentyl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol
NPE-11	2-(2-(2-(2-(4-(2,2,4,4-tetramethylpentyl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol
NPE-12	2-(2-(2-(2-(4-(2,4,5-trimethylhexan-2-yl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol
NPE-13	2-(2-(2-(2-(4-(3,3,4,4-tetramethylpentyl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol
NPE-14	2-(2-(2-(2-(2-(3,4,5-trimethylhexan-2-yl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol
NPE-15	2-(2-(2-(2-(4-(7-methyloctyl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol
NPE-16	2-(2-(2-(2-(4-(3,4,5-trimethylhexan-2-yl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol
NPE-17	2-(2-(2-(2-(4-(5,7-dimethyloctyl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol
NPE-18	3,6,9,12-Tetraoxatetradecan-1-ol, 14-(4-nonylphenoxy)-
NPE-19	3,6,9,12-Tetraoxatetradecan-1-ol, 14-(nonylphenoxy)-
NPE-20	3,6,9,12,15-Pentaoxaheptadecan-1-ol, 17-(nonylphenoxy)-
NPE-21	3,6,9,12,15,18,21-Heptaoxatricosan-1-ol, 23-(nonylphenoxy)-
NPE-22	3,6,9,12,15,18,21,24-Octoaxahexacosan-1-ol, 26-(nonylphenoxy)-
NPE-23	3,6,9,12,15,18,21,24,27-Nonaoxanonacosan-1-ol, 29-(isononylphenoxy)-
NPE-24	3,6,9,12,15,18,21,24,27-Nonaoxanonacosan-1-ol, 29-(nonylphenoxy)-

pH-dependent solubility and K_{OC}

For each group of compounds solubility and K_{OC} were modelled over the full pH range to identify those groups of compounds for which solubility and K_{OC} were likely to be pH dependent. Solubility and K_{OC} did not vary with pH for PAH, PCB or phthalates (data not shown). For the other compound groups (NPE, NP, LAS) both solubility and K_{OC} varied with pH. The detailed model output is presented graphically in Figures 2.5 to 2.7 and summarised in Table 2.2.

Table 2.2 Summary of K_{OC} and Solubility sensitivity to pH

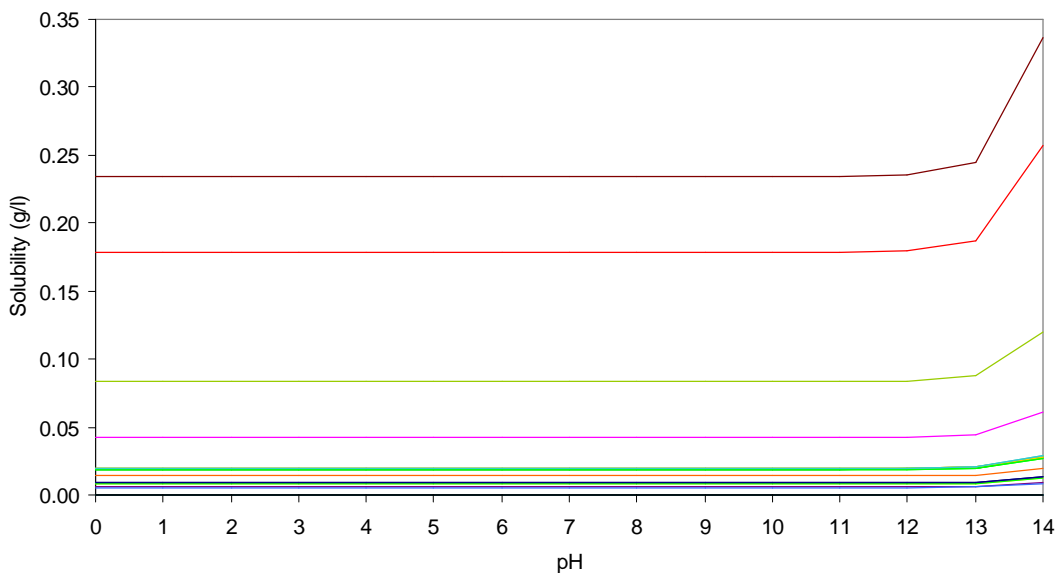
Substance Grouping	K_{OC} pH sensitive?	Solubility pH sensitive?	Comment
PCB	No	No	
PAH	No	No	
Phthalates	No	No	
Nonylphenol ethoxylates	No	No	There is a small decrease in modelled K_{OC} above pH 12 of approximately 0.15 log K_{OC} . There is a corresponding small increase in solubility of up to 0.1 g/l.
Nonylphenol	Yes	Yes	Modelled K_{OC} decreases gradually above pH 8 and decreases steeply above pH 10, with a total change of about 3 log K_{OC} . Solubility increases gradually above pH 8 and increases steeply above pH 10, with a total change of three orders of magnitude.
Linear Alkyl Benzene Sulfonates (LAS)	Yes	Yes	K_{OC} decreases with pH up to pH 2 – 4, where the response levels out. Solubility increases with pH up to pH 2 – 5, where the response levels out.

That solubility and K_{OC} for PAH, PCB and phthalates do not vary with pH is expected since these are non-polar compounds.

For NPEs and nonylphenols, the variable response to pH is probably linked to the degree of dissociation of the hydroxyl (-OH) group in more alkaline conditions; this is likely to increase solubility. The critical conditions are pH 12 for NPEs and pH 8 for nonylphenols. K_{OC} also appears to be influenced by compound structure irrespective of pH.

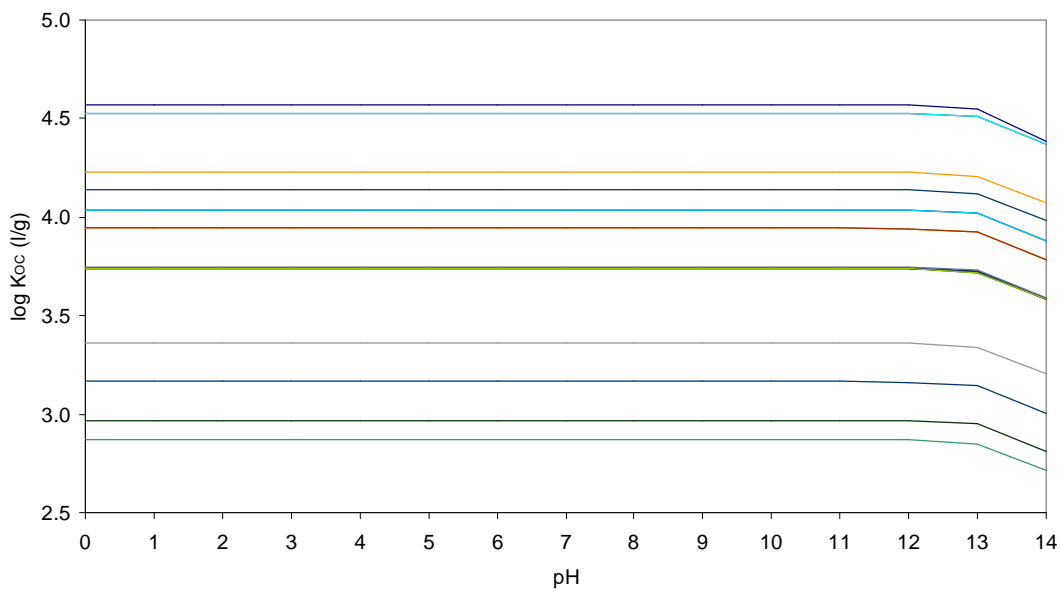
LAS are anionic detergents with a hydrophobic and a hydrophilic end to the molecule. Since one end of the molecule is non-polar, it will adsorb to the organic component of the matrix. However, due to the polar sulphonate group, LAS will bind to organic matter less strongly than non-polar contaminants, such as PAH and PCB.

(a) Solubility



- NPE-1 NPE-2 NPE-3 NPE-4 NPE-5 NPE-6 NPE-7 NPE-8
- NPE-9 NPE-10 NPE-11 NPE-12 NPE-13 NPE-14 NPE-15 NPE-16
- NPE-17 NPE-18 NPE-19 NPE-20 NPE-21 NPE-22 NPE-23 NPE-24

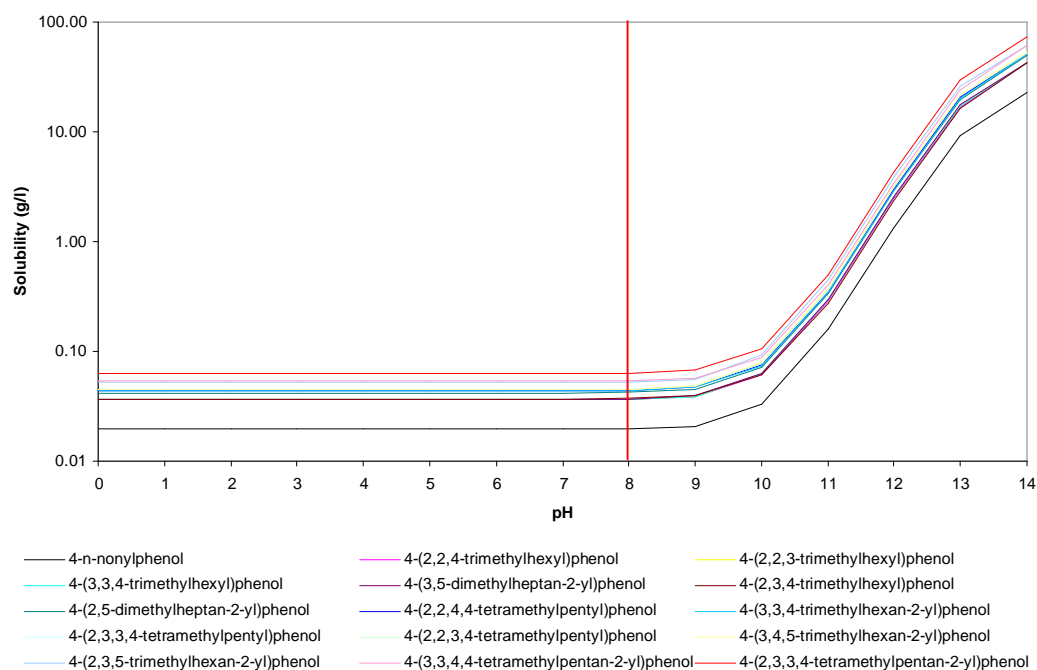
(b) K_{OC}



- NPE-1 NPE-2 NPE-3 NPE-4 NPE-5 NPE-6 NPE-7 NPE-8
- NPE-9 NPE-10 NPE-11 NPE-12 NPE-13 NPE-14 NPE-15 NPE-16
- NPE-17 NPE-18 NPE-19 NPE-20 NPE-21 NPE-22 NPE-23 NPE-24

Figure 2.5 NPE compounds: Solubility and K_{OC} variability with pH

(a) Solubility



(b) K_{OC}

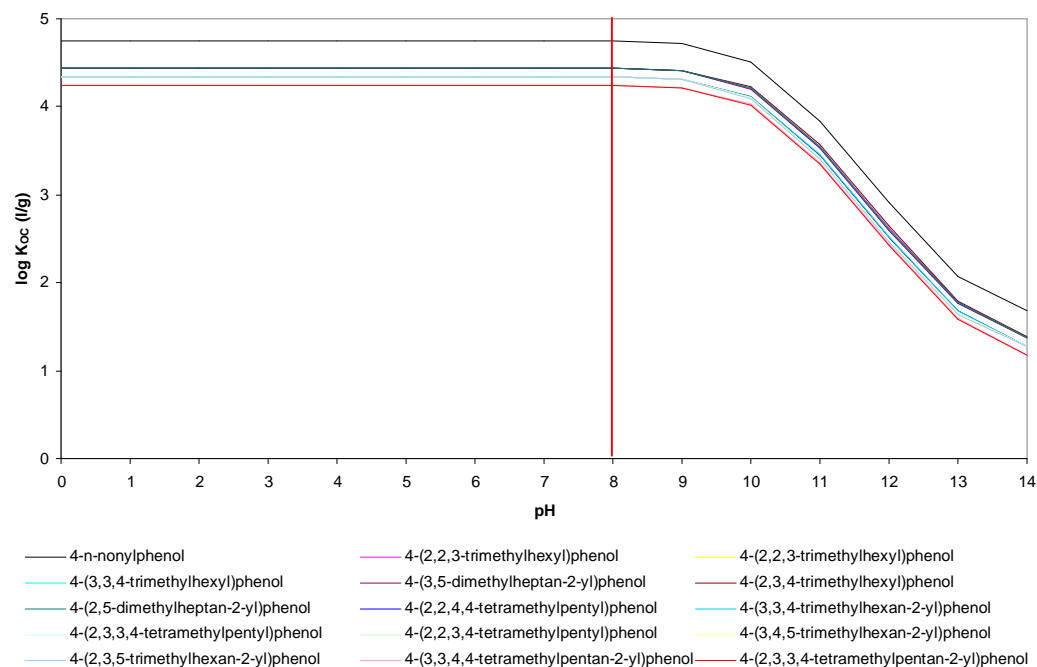


Figure 2.6 NP isomers: Solubility and K_{OC} variability with pH

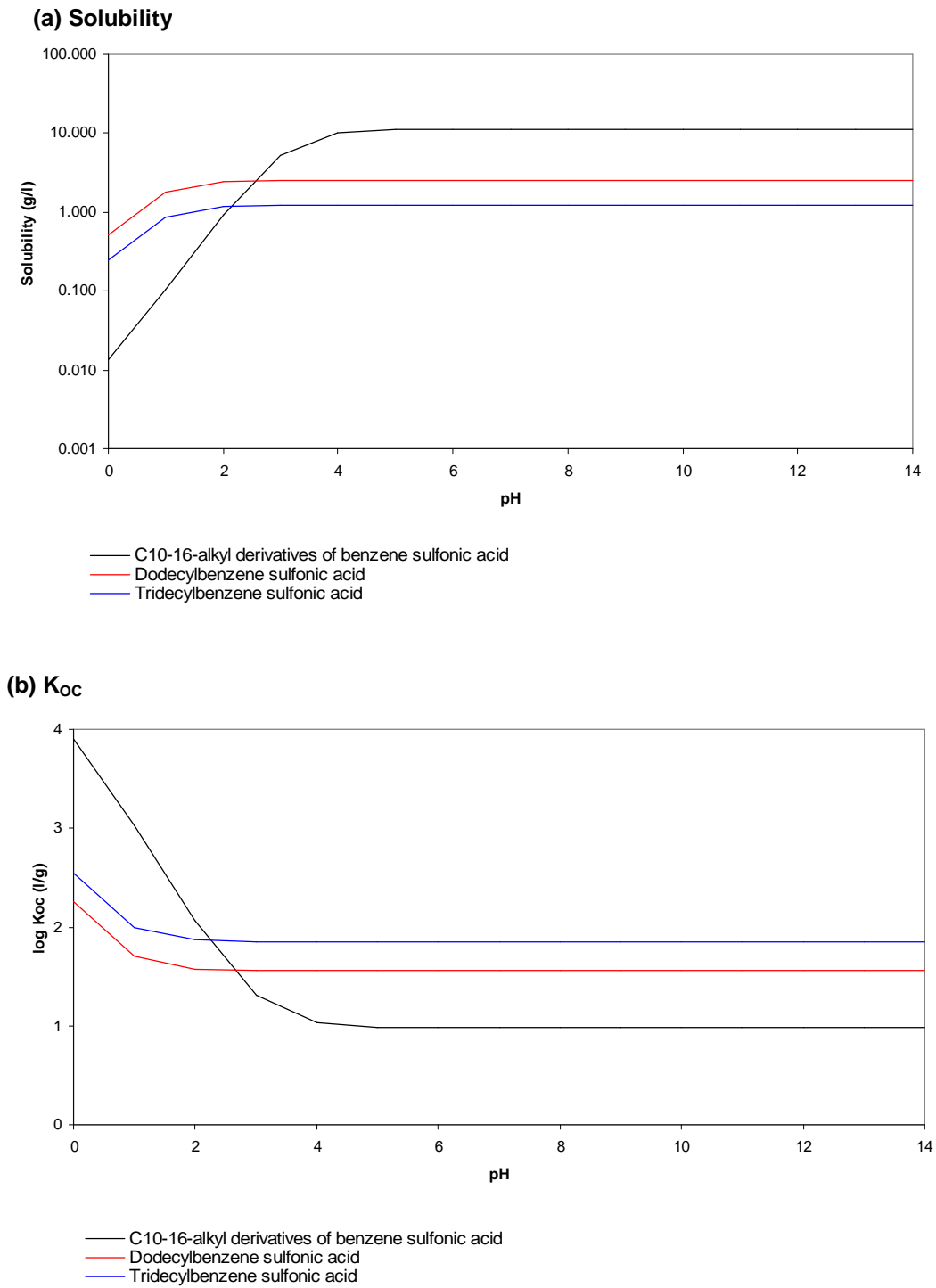


Figure 2.7 LAS compounds: solubility and K_{OC} variability with pH

2.4 Translation to Field Studies

It is hypothesized that, in a given material, variability in organic micropollutant concentration depends on compound solubility and K_{OC} . If solubility and K_{OC} are pH-dependent then variability in concentrations will depend on the pH and organic carbon content of the matrix (sludge, treated biowaste or soil).

The aim was to test this hypothesis using the predicted pH effects from the chemical modelling and the results of field sample analyses.

2.4.1 Investigation of PAH and PCB variability

Many samples had been collected and analysed from several Wastewater Treatment Works, composting plants and agricultural fields (Lambkin et al., 2007). It was anticipated that sampling would have produced a large data set for statistically testing the relationship between PAH/PCB concentration and Loss on Ignition. However, few of the data sets had PAH/PCB concentrations above the limit of quantification. Consequently, the statistical analysis was limited to those data that could be regarded as robust (Tables D.1-3, Annex D):

- (i) Soil with regular sewage sludge applications (Spain Field 2: PCB);
- (ii) Co-composted sewage sludge (STWB: PCB);
- (iii) Thermally dried sewage sludge (STWE: PAH).

Since PAH and PCB solubility and K_{OC} do not vary with pH, a statistical relationship between concentration and matrix pH was not expected. In all cases no significant relationship with pH was found.

Correlation coefficients were calculated for each data set, comparing between compound concentrations and concentration to Loss on Ignition (LOI). LOI was used as a measure of organic carbon since it is easier and cheaper to determine than chemical methods. It is recognized that LOI is only an approximate measure of organic carbon in soils and that its value depends on a number of factors such as the sequioxide, calcium carbonate and clay content of the soil. However, it has been demonstrated that, over an area of land where the soil type is similar and the land is managed for the same purpose, there is a strong relationship between the two measures (Frogbrook and Oliver, 2001).

The results are shown in Tables 2.3 – 2.5 and graphically in Figures 2.8 – 2.10 (LOI not shown). It should be noted that not all the data sets were complete because some reported concentrations were below the detection limit. Consequently, the data in Figures 2.8 – 2.10 are plotted against the compound for which all concentrations were above the detection limit. Some compounds were detected in very few samples in a given data set (e.g. 1 or 2 of 16). In this case the data were omitted from the correlation analysis.

Correlation between compound concentrations and LOI was low and, in almost all cases, was not significant. It had been expected that, if PAH/PCB variability was controlled by adsorption, then there would be a strong correlation with LOI.

For thermally dried sewage sludge (STWE) lack of correlation with LOI might be explained by the lack of variability in LOI, which varied by less than one percentage point (75.7-76.4%). The same explanation cannot be used for the other two data sets, which have a much wider range of LOI (soil (Spain Field 2): 5.82-11.8% and co-composted sludge (STWB): 49.6-62.9%).

Table 2.3 Soil (Spain Field 2): PCB concentration – Correlation Matrix

PCB	PCB						LOI%
	52	101	118	138	153	180	
28	***0.989	***0.894	-0.049	***0.890	***0.899	***0.922	0.217
52		***0.928	-0.248	***0.934	***0.934	***0.949	0.186
101			***0.820	***0.990	***0.996	***0.991	0.193
118				***0.704	***0.785	**0.701	*0.257
138					***0.997	***0.995	0.213
153						***0.995	0.223
180							0.215

Significance: * p < 0.05; ** p < 0.01; *** P < 0.001; all other values are non-significant

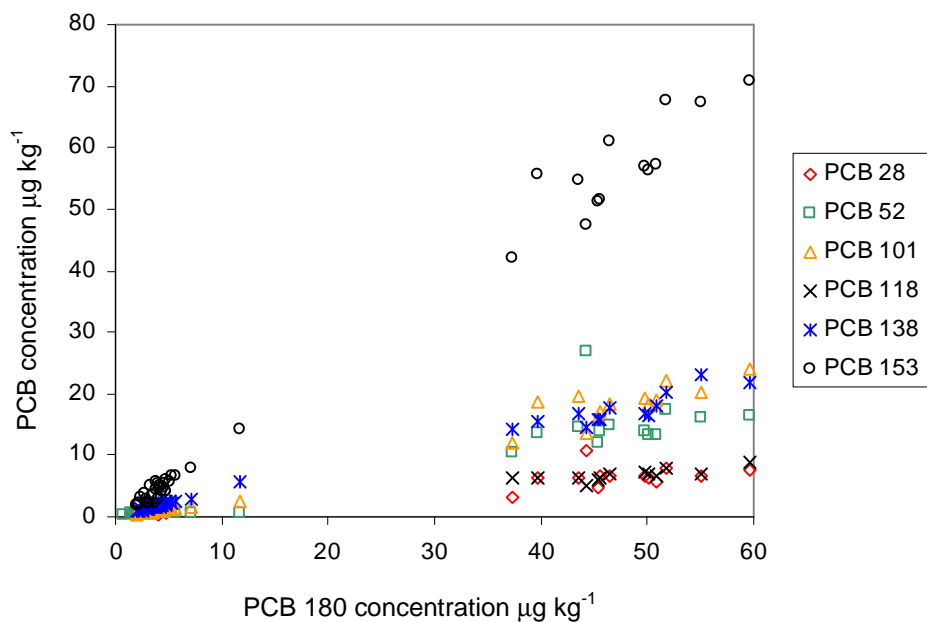


Figure 2.8 Soil (Spain Field 2): Concentration of six PCB compounds relative to PCB 180

Table 2.4 Co-composted sewage sludge (STWB): PCB concentration – Correlation Matrix

PCB	PCB						LOI%
	52	101	118	138	153	180	
28	*0.549	***0.749	0.057	**0.715	0.253	-0.061	0.081
52		***0.768	***0.951	***0.881	**0.721	0.133	-0.462
101			*0.734	***0.907	**0.779	0.245	-0.213
118				***0.950	**0.776	-0.039	-0.439
138					**0.729	0.079	-0.430
153						0.197	-0.101
180							-0.219

Significance: * p < 0.05; ** p < 0.01; *** P < 0.001; all other values are non-significant

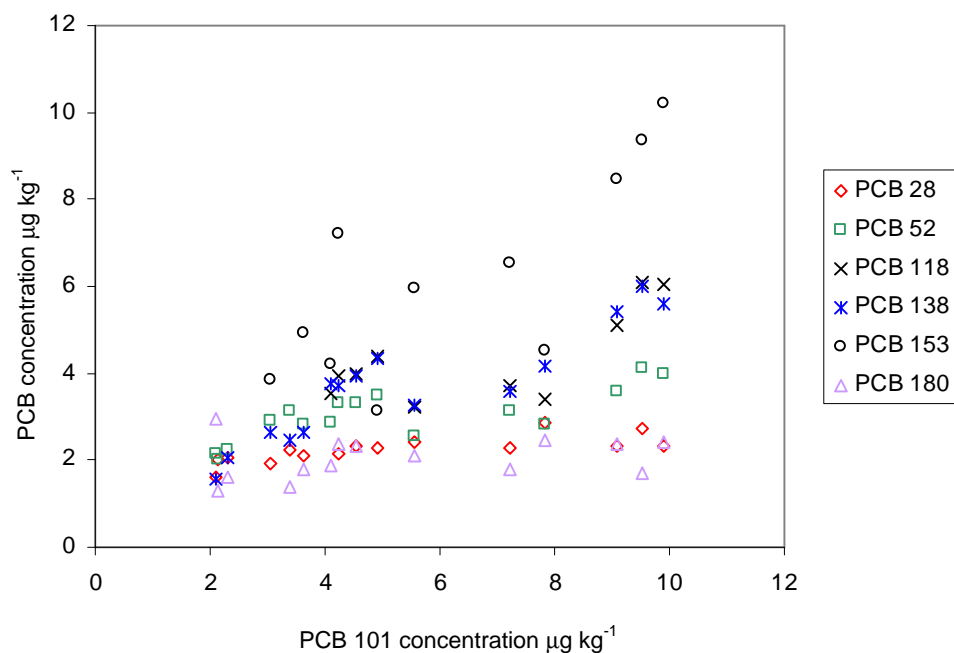


Figure 2.9 Co-composted sewage sludge (STWB): Concentration of six PCB compounds relative to PCB 101

Table 2.5 Thermally dried sewage sludge (STWE): PAH concentration – Correlation Matrix

	Benzo (a) Pyrene	Benzo (b&k) Fluoranthene	Benzo (g,h,i) Perylene	Fluoranthene	Indeno (1,2,3-cd) pyrene	Phenanthrene	PAH Total (EPA 16)	LOI%
Pyrene	***0.919	***0.820	***0.937	***0.952	***0.898	***0.936	***0.947	0.177
Benzo (a) Pyrene		***0.926	***0.967	***0.922	***0.964	***0.829	***0.958	0.088
Benzo (b&k) Fluoranthene			***0.985	***0.862	***0.960	***0.774	***0.956	0.345
Benzo (g,h,i) Perylene				***0.964	***0.985	***0.864	***0.994	0.188
Fluoranthene					***0.941	***0.895	***0.964	0.203
Indeno (1,2,3-cd) pyrene						***0.805	***0.970	0.038
Phenanthrene							***0.878	0.200
PAH Total (EPA 16)								0.158

Significance: * p < 0.05; ** p < 0.01; *** P < 0.001; all other values are non-significant

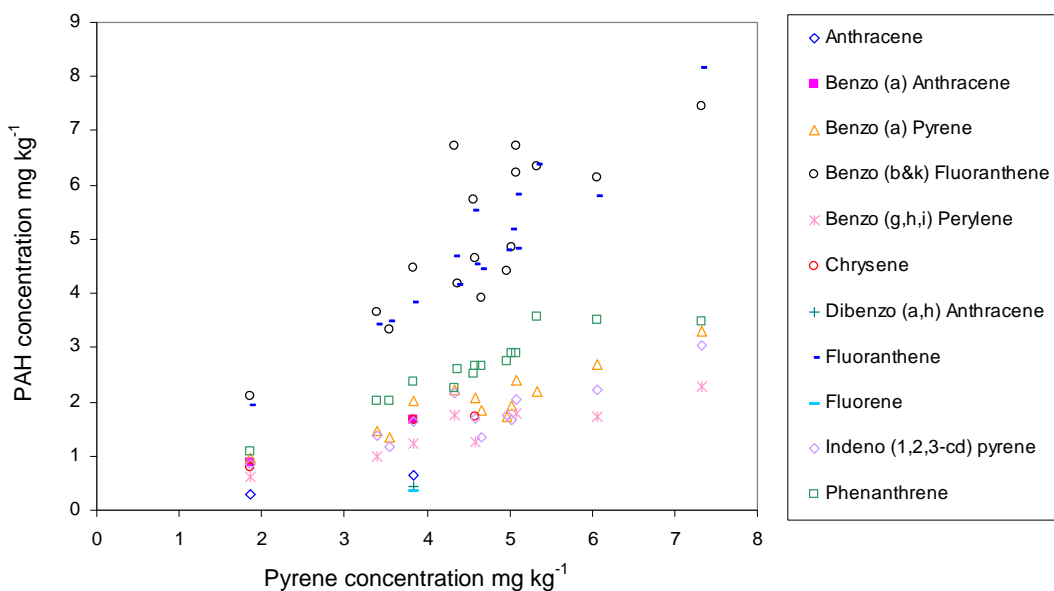


Figure 2.10 Thermally dried sewage sludge (STWE): Concentration of 11 PAH compounds relative to pyrene

Correlation between compound concentrations was high and statistically significant for almost all comparisons. This result indicates that, although concentrations vary between samples, the ratio between compound concentrations is maintained.

For co-composted (STWB) and thermally dried (STWE) sewage sludge, possible reasons include:

- (i) Relative concentrations in the feedstock are constant, i.e. output reflects input;
- (ii) Differential losses during processing are constant, i.e. compounds with low solubility and high K_{OC} will be retained;
- (iii) The degree of mixing during processing.

In soil (Spain Field 2), the results can be explained by:

- (i) Variability in levels of concentration are due to variability in sludge application rate or dilution factor (i.e. water content);
- and,
- (ii) There is little difference in concentration ratios from one application to the next.

This theory is illustrated by the following example.

Example

Sludge is applied to a field on four occasions, but application rate varies across the field. Although the concentration of Compound 1 is different for each of the four applications, the concentration ratio of each of the other 4 compounds to Compound 1 is always constant.

Table 2.6 Example 1: Compound concentrations in sludge

Compound	Concentration			
	Year 1	Year 2	Year 3	Year 4
1	0.1	1.2	0.25	1.7
2	0.05	0.6	0.125	0.85
3	0.12	1.44	0.3	2.04
4	0.14	1.68	0.35	2.38
5	0.2	2.4	0.5	3.4

Compound concentrations are calculated by multiplying compound concentration by application received (generated using random number tables), see Table 2.7.

Table 2.7 Example: Application rates and resulting compound concentrations

Sample point	Application received				Compound Concentration				
	Year 1	Year 2	Year 3	Year 4	C1	C2	C3	C4	C5
1	6	2	2	6	13.7	6.9	16.4	19.2	27.4
2	10	7	5	2	14.1	7.0	16.9	19.7	28.1
3	2	9	7	7	24.7	12.3	29.6	34.5	49.3
4	5	3	1	4	11.2	5.6	13.4	15.6	22.3
5	10	4	7	6	17.8	8.9	21.3	24.9	35.5
6	7	10	8	8	28.3	14.2	34.0	39.6	56.6
7	8	9	2	3	17.2	8.6	20.6	24.1	34.4
8	3	5	3	3	12.2	6.1	14.6	17.0	24.3
9	4	7	9	1	12.8	6.4	15.3	17.9	25.5
10	2	6	7	10	26.2	13.1	31.4	36.6	52.3

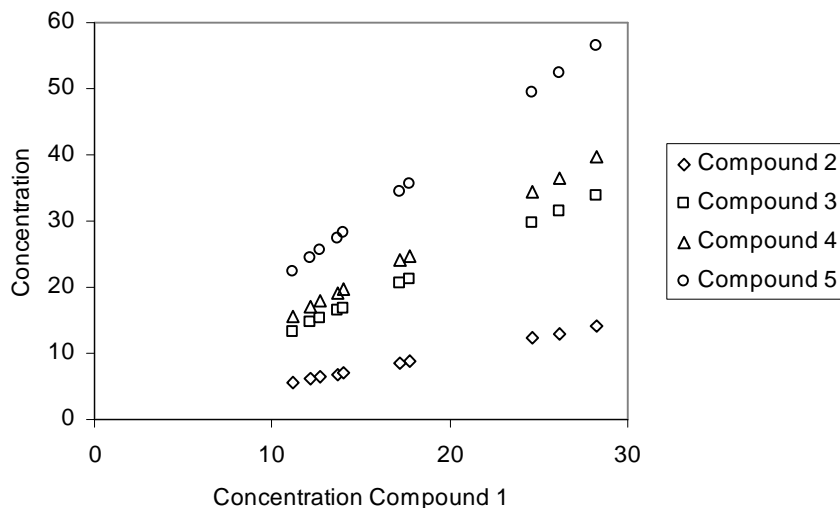


Figure 2.11 Example: Relationship between sampled compound concentrations

The hypothetical results plotted in Figure 2.11 produce similar patterns to those seen in Figure 2.8. The results reproduce:

- (i) the linear relationship between compound concentrations;
- (ii) the paucity of data at some concentrations.

Deviations from perfect linearity in the analytical data (Figure 2.8) are likely to be due to variation in concentration caused by small differences in sample support and laboratory sub-sampling and analysis.

Modelled solubility and K_{OC} of phthalates did not vary with pH therefore it is likely that phthalate concentrations vary in a similar way to PAH and PCB. Since phthalates are used as plasticizers, they are common in the environment and it is difficult to control cross-contamination between samples for analysis. Consequently, it might be difficult to test.

2.4.2 Investigation of pH-dependent compounds

Three of the modelled compound groups have pH-dependent solubility and K_{OC} : NPE, NP and LAS.

NPE compounds

Modelled solubility and K_{OC} for NPE compounds does not vary over the normal pH range for soils. There is a small increase in solubility and decrease in K_{OC} above pH12. This could have some consequences for NPE removal from lime-treated sludge, which is raised above pH12 during processing, due to losses from the aqueous fraction. However,

- (i) the change in solubility between pH12 and pH13 is quite small and the greatest change is above pH13 for some of the NPE compounds;
- (ii) the change in K_{OC} between pH12 and pH13 is quite small and most of the change is above pH13;

Changes at such high pH are only likely to be of concern in lime-treated sludges. However, the pH conditions are usually well-controlled during lime treatment for operational and economic reasons. Therefore it would be unlikely that any sampling variability could be differentiated from variability due to laboratory preparation procedures such as sub-sampling. Also, lime is usually added after sludge dewatering and the material is well-mixed during the liming process, so losses from the aqueous fraction will be small.

Once applied to soil, the pH will reduce over time towards the soil pH due to the buffering capacity. This change in pH would decrease solubility and increase K_{OC} , so immobilising NPE.

NP compounds

Modelled solubility and K_{OC} for NP compounds does not vary over the most of the normal pH range for soils. There is a small change between pH8 and pH9, but the greatest change is above pH9. Therefore pH is unlikely to be a major factor in controlling NP variability in most agricultural soils. One exception may be sodic soils, where the pH could exceed 11.

As with NPE, the change in solubility and K_{OC} could have consequences for NP concentrations in lime-treated sludge. The predicted changes above pH8 are greater than for NPE, so the effect on NP fate in lime-treated sludges might be more significant, especially in sludges with higher water content.

LAS compounds

Modelled solubility and K_{OC} for LAS compounds does not vary over the normal pH range for soils. There is a decrease in solubility below pH 2-4 and a corresponding increase in K_{OC} , but such low soil pH is not found in agricultural soils, although very acidic soil can be found elsewhere, such as metaliferous mine sites. The pH of sludges and treated biowastes is typically above pH5.

2.5 Conclusions

1. pH-dependent variability in solubility and K_{OC} was modelled for six groups of compounds. For three groups the model did not identify pH-dependent variability in either measurement. For NPE, NP and LAS pH-dependent variability was predicted at pH extremes.
2. Laboratory data (PAH, PCB, LOI, pH) were examined to assess if variability in organic carbon and modelled K_{OC} or pH could explain variation in compound concentration. There were insufficient analytical data for rigorous testing of the results, but examination of the available data provided no evidence to support co-variation in sludge, co-composted sludge, or soil.
3. Further examination of the data indicated that, for the two Wastewater Treatment Works investigated, there was strong correlation between concentrations of different substances within a group of compounds, i.e. either PAH or PCB. This may be due to lack of variability in feed stock and/or the high degree of mixing during processing, either of which could have produced a high degree of homogeneity in the treated material that was sampled.
4. Only one soil data set was examined, but, as for the sludge, there was strong correlation between concentrations of different substances within a group of compounds. It was hypothesized that the factors controlling PCB spatial variability in the field are (i) variability in the applied material and (ii) variability in the application rate. Support for this hypothesis was demonstrated using example data.
5. Although almost 600 samples from 13 sites were collected and analysed for PAH and PCB few of these contained PAH and PCB above the limit of detection. Consequently the number of sites for which adequate data was available for examination was limited. This means that the results have to be treated with caution and may not be transferable to other sites or, more importantly, to other compound groups.

6. It has been recommended in earlier work that, since the number of samples required for a given precision varies between sites, treatment sites should be characterised during commissioning. Based on this work, it is likely that the number of samples required for organic micropollutant analysis can be determined by measurement of a small number such compounds.

3 Recommendations

1. Further sampling and analysis should be carried out to test and confirm the results from this study, which was limited by the lack of available data sets.
2. This work has demonstrated a strong correlation between concentrations of PAH groups of compounds and between PCB groups of compounds. Future sampling should include matrices with measurable concentrations of both PAH and PCB to enable investigation of between group correlation.
3. Once a treatment site has been characterised, it is likely that the variability of concentrations of many substances can be inferred from the characteristics of perhaps one or two compounds, unless there are major changes to the feedstock.
4. The chemical analysis should be extended to other groups of compounds to test for correlation within and between groups of compounds.

REFERENCES

- [1] Abad E, Martínez K, Planas C, Palacios O, Caixach J and Rivera J (2005) Priority organic pollutant assessment of sludges for agricultural purposes. *Chemosphere*, **61**: 1358-1369.
- [2] Bagó B, Martín Y, Mejía G, Broto-Puig F, Díaz-Ferrero J, Agut M and Comellas L (2005) Di-(2-ethylhexyl)phthalate in sewage sludge and post-treated sludge: Quantitative determination by HRGC-MS and mass spectral characterization. *Chemosphere*, **59**: 1191-1195.
- [3] Carlsen L, Metzton M-B, Kjelsmark J (2002) Linear alkylbenzene sulfonates (LAS) in the terrestrial environment. *The Science of the Total Environment*, **290**: 225-230.
- [4] Cavalli L and Valtorta L (1999) Surfactants in sludge-amended soil. *Tenside Surfactants Detergents*, **36**: 22-28.
- [5] Céspedes R, Lacorte S, Ginebreda A and Barceló D (2006) Chemical monitoring and occurrence of alkylohenols, alkylophenol ethoxylates, alcohol ethoxylates, phthalates and benzothiazoles in sewage treatment plants and receiving waters along the Ter River basin (Catalonia, NE Spain). *Analytical and Bioanalytical Chemistry*, **385**: 992-1000.
- [6] Cousins I and MacKay D (2000) Correlating the physical-chemical properties of phthalate esters using the 'three solubility' approach. *Chemosphere*, **41**: 1389-1399.
- [7] Cronin, MTD, Walker, JD, Jaworska, JS, Comber, MHI, Wattes, CD and Worth, AP (2003) Use of QSARs in international decision-making frameworks to predict ecologic effects and environmental fate of chemical substances. *Environmental Health Perspectives*, **111**: 1376-1390.
- [8] Daughton CG and Ternes TA (1999) Pharmaceuticals and personal care products in the environment: Agents of subtle change? *Environmental Health Perspectives*, **107**: 907-938.
- [9] Dearden JC (2002) Prediction of environmental toxicity and fate using quantitative structure-activity relationships (QSARs). *Journal of the Brazilian Chemical Society*, **13**: 754-762.
- [10] De Lima Ribeiro FA and Ferreira MMC (2003) QSPR models of boiling point, octanol-water partition coefficient and retention time index of polycyclic aromatic hydrocarbons. *Journal of Molecular Structure (Theochem)*, **663**: 109-126.
- [11] Dunnivant FM, Elzerman AW, Jurs PC and Hasan MN (1992) Quantitative structure-property relationships for aqueous solubilities and Henry's Law constants of polychlorinated biphenyls. *Environmental Science and Technology*, **26**: 1567-1573.
- [12] Eljarrat E, Caixach J and Rivera J (2003) A comparison of TEQ contributions from PCDDs, PCDFs and dioxin-like PCBs in sewage sludges from Catalonia, Spain. *Chemosphere*, **51**: 595-601.
- [13] Fabrellas B, Larrazabal D, Martinez MA, Eljarrat E and Barcelló D (2004) Presence of polybrominated diphenyl ethers in Spanish sewage sludges: Important contribution of DECA-BDE. *Organohalogen Compounds*, **66**: 375-3760.
- [14] Ferreira MMC (2001) Polycyclic aromatic hydrocarbon: a QSPR study. *Chemosphere*, **44**: 125-146.
- [15] Fountoulakis M, Drillia P, Pakou C, Kampioti A, Stamatelatou K and Lyberatos G (2005) Analysis of nonylphenol and nonylphenol ethoxylates in sewage sludge by high performance liquid chromatography following microwave-assisted extraction. *Journal of Chromatography*, **1089**: 45-51.

- [16] Fremmersvik, G and Hansen, N (2004) *Desk Study: LAS and Nonylphenols*. Project HORIZONTAL Desk Study 13.
- [17] Frogbrook ZL and Oliver MA (2001) Comparing the spatial predictions of soil organic matter determined by two laboratory methods. *Soil Use and Management*, **17**: 235-244.
- [18] Fromme H, K uchler T, Otto T, Pilz K, M uller J and Wenzel A (2002) Occurrence of phthalates and bisphenol A and F in the environment. *Water Research*, **36**: 1429-1438.
- [19] Garc a MT, Compos E, Dalmau M, Ribosa I and S anchez-Leal J (2002) Structure-activity relationships for association of linear alkylbenzene sulfonates with activated sludge. *Chemosphere*, **49**: 279-286.
- [20] Gatidou G, Thomaidis NS, Stasinakis AS, Lekkas TD (2007) Simultaneous determination of the endocrine disrupting compounds nonylphenol, nonylphenol ethoxylates, triclosan and bisphenol A in wastewater and sewage sludge by gas chromatography–mass spectrometry. *Journal of Chromatography*, **1138**: 32-41.
- [21] Gibson R, Wang M-J, Padgett E and Beck AJ (2005) Analysis of 4-nonylphenols, phthalates, and polychlorinated biphenyls in soils and biosolids. *Chemosphere*, **61**: 1336-1344.
- [22] Green NJL, Hassanin A, Johnston AE and Jones KC (2004) Observations on historical, contemporary, and natural PCDD/Fs. *Environmental Science and Technology*, **38**: 715-723.
- [23] Gundersen JL (2001) Separation of isomers of nonylphenol and select nonylphenol polyethoxylates by high-performance liquid chromatography on a graphitic carbon column. *Journal of Chromatography A*, **914**: 161–166.
- [24] Harrad S and Hunter S (2006) Concentrations of polybrominated diphenyl ethers in air and soil on a rural-urban transect across a major UK conurbation. *Environmental Science & Technology*, **40**: 4548-4553.
- [25] Hassanin, A, Breivik, K, Meijer, SN, Steinnes, E, Thomas, GO, Jones, KC (2004) PBDEs in European background soils: levels and factors controlling their distribution. *Environmental Science and Technology*, **38**: 738-745.
- [26] Hassanin A, Lee RGM, Steinnes E and Jones KC (2005) PCDD/Fs in Norwegian and UK soils: Implications for sources and environmental cycling. *Environmental Science and Technology*, **39**: 4784-4792.
- [27] Herberer T (2002) Occurrence, fate, and removal of pharmaceutical residues in the aquatic environment: a review of recent research data. *Toxicology Letters*, **13**: 5-17.
- [28] IC Consultants Ltd. (2001) *Pollutants in urban wastewater and sewage sludge*. Final report. Prepared by ICON, IC Consultants Ltd, London, UK for Directorate-General Environment, 273pp. Office for Official Publications of the European Communities, Luxembourg.
- [29] Ieda T, Horii Y, Petrick G, Yamashita N, Ochiai N, and Kannan K (2005) Analysis of Nonylphenol Isomers in a Technical Mixture and in Water by Comprehensive Two-Dimensional Gas Chromatography-Mass Spectrometry. *Environmental Science and Technology*, **39**: 7202-7207.
- [30] Jacobsen AM, Mortensen GK, and Bruun Hansen HC (2004) Degradation and mobility of linear alkylbenzene sulfonate and nonylphenol in sludge-amended soil. *Journal of Environmental Quality*, **33**: 232-240.

- [31] Jensen J and Jepsen S-E (2005) The production, use and quality of sewage sludge in Denmark. *Waste Management*, **25**: 239-247.
- [32] Johnson A and Jürgens M (2003) Endocrine active industrial chemicals: release and occurrence in the environment. *Pure and Applied Chemistry*, **75**: 1895-1904.
- [33] Kloepper-Sams P, Torfs F, Feijtel T, and Gooch J (1996) Effect assessments for surfactants in sludge-amended soils: a literature review and perspectives for terrestrial risk assessment. *The Science of the Total Environment*, **185**: 171-185.
- [34] Lambkin DC, Evans TD, Nortcliff S and White TC (2004) *Towards producing harmonised methods, with quantified precision, for sampling sludges, treated biowaste, and soils in the landscape*. Project HORIZONTAL. Final Report WP2.
- [35] Lambkin DC, Nortcliff S and White TC (2006) *Report on fieldwork to test statistical performance of sampling strategies for sludge and treated biowaste*. Project HORIZONTAL Draft report HYG WP1-2.
- [36] Lambkin DC, Nortcliff S and White TC (2007) *Research report on the relationship of sampling protocols to the analytical results obtained for soils and treated biomaterials (biowastes and sludges)*. Project HORIZONTAL Draft report ORG WP2-6.
- [37] Landesanstalt für Umweltschutz. (2003) *Concise Report: Contaminants in arable soils in Baden-Württemberg fertilised with sewage sludge*. 15pp. The Ministry for Environment and Transport, Baden-Württemberg, Karlsruhe, Germany.
- [38] Law RJ, Allchin CR, de Boer J, Covaci A, Herzke D, Lepom P, Morris S, Tronczynski J, de Wit CA (2006) Levels and trends of brominated retardants in the European environment. *Chemosphere*, **64**: 187-208.
- [39] Marttinen SK, Hänninen K and Rintala JA (2004) Removal of DEHP in composting and aeration of sewage sludge. *Chemosphere*, **54**: 265-272.
- [40] Meylan W, Howard PH and Boethling RS (1992) Molecular topology/fragment contribution method for predicting soil sorption coefficients. *Environmental Science and Technology*, **26**: 1560-1567.
- [41] Öberg T (2001) Prediction of physical properties for PCB congeners from molecular descriptors. *Internet Journal of Chemistry*, Volume **4**, Article No 11.
- [42] Öberg K, Warman K, and Öberg T (2002) Distribution and levels of brominated flame retardants in sewage sludge. *Chemosphere*, **48**: 805-809.
- [43] Oliver R, May E and Williams J (2005) The occurrence and removal of phthalates in a trickle filter STW. *Water Research*, **39**: 4436-4444.
- [44] Oliver R, May E and Williams J (2007) Microcosm investigations of phthalate behaviour in sewage treatment biofilms. *Science of the Total Environment*, **372**: 605-614.
- [45] Petersen SO, Henriksen K, Mortensen GK, Krogh PH, Brandt KK, Sorensen J, Madsen T, Petersen J, and Gron C (2003) Recycling of sewage sludge and household compost to arable land: fate and effects of organic contaminants, and impact on soil fertility. *Soil & Tillage Research*, **72**: 139-152.
- [46] Schmid P, Gujer E, Zennegg M, Bucheli TD, Desaulles A (2005) Correlation of PCDD/F and PCB concentrations in soil samples from the Swiss soil monitoring network (NABO) to specific parameters of the observation sites. *Chemosphere*, **58**: 227-234.

- [47] Sellström U, de Wit CA, Lundgren N, Tysklind M (2005) Effect of sewage-sludge application on concentrations of higher-brominated diphenyl ethers in soils and earthworms. *Environmental Science and Technology*, **39**: 9064-9070.
- [48] Stumpf M, Ternes TA, Wilken R-D, Rodrigues SV, and Baumann W (1999) Polar drug residues in sewage sludge and natural waters in the state of Rio de Janeiro, Brazil. *The Science of the Total Environment*, **225**: 135-141.
- [49] Vikelsøe J, Thomsen M and Carlsen L (2002) Phthalates and nonylphenols in profiles of differently dressed soils. *The Science of the Total Environment*, **296**: 105-116.
- [50] Vogelsang C, Grung M, Jantsch TG, Tollefsen KE and Liltved H (2006) Occurrence and removal of selected organic micropollutants at mechanical, chemical and advanced wastewater treatment plants in Norway. *Water Research*, **40**: 3559-3570.
- [51] Webber MD, Rogers HR, Watts CD, Boxall ABA, Davis RD and Scoffin R (1996) Monitoring and prioritisation of organic contaminants in sewage sludges using specific chemical analysis and predictive, non-analytical methods. *The Science of the Total Environment*, **185**: 27-44.
- [52] de Wit CA (2001) An overview of brominated flame retardants in the environment. *Chemosphere*, **46**: 583-624.
- [53] Zuccato E, Calamari D, Natangelo M, and Fanelli R (2000) Presence of therapeutic drugs in the environment. *The Lancet*, **35**: 1789-1790.

ANNEX A

ACD/SOLUBILITY DB

A.1 Introduction

The following paragraphs briefly describe the functionality of the structure-activity model used in this study. The information has been extracted from the manufacturer's website [accessed 13-04-07]:

http://www.acdlabs.com/products/phys_chem_lab/aqsol/

A.2 Key Capabilities

ACD/Solubility DB software determines the aqueous solubility at 25°C of organic compounds for pH values from 0.0 to 14.0. The accuracy of calculations for simple structures is usually better than 0.2-0.5 logS units; for complex structures it is better than 0.5-1.0 logS units.

A.3 Input/Output

Users can choose to either draw their structures using an array of tools including pre-made chemical templates, or import the structures from any popular third-party formats such as ChemDraw, molfile, or ISIS Draw.

Chemical structures and calculated properties can be transferred to the ACD/ChemSketch to be saved as Adobe PDF reports, or copied and pasted into Microsoft Office applications. MDL SDF files are also accepted. Up to 999 structures can be imported directly into the History window to generate selected properties for several molecules at a time. For larger files, ACD/PhysChem Batch is recommended.

A.4 Comparison with experiment

Solubility calculated using ACD/Solubility DB were compared with experimental data. Correlation of the experimental logS vs. calculated logS values for several classes of chemical compounds is shown in Table A.1. Compound solubility is being predicted within 1.0 log unit for greater than 87% of compounds, and within 0.3 log units for 45% of compounds in the data set.

Table A.1 ACD/Solubility DB - Accuracy of calculations

	Number of compounds	Std Dev	R, correlation
All compounds	993	0.58	0.957
Monofunctional Aromatic, Aliphatic, and Unsaturated compounds	223	0.34	0.971
Hydrocarbons	68	0.31	0.971
- Aliphatic and unsaturated hydrocarbons	31	0.19	0.982
- Aromatic hydrocarbons	37	0.39	0.960
- Nonfused aromatics	19	0.21	0.975
- Fused aromatics	18	0.52	0.921
Halogenated hydrocarbons	150	0.42	0.985
- Chlorinated hydrocarbons	99	0.44	0.984
- Chlorinated aliphatic and unsaturated hydrocarbons	36	0.32	0.970
- Chlorinated aromatic hydrocarbons	58	0.45	0.971
- Polychlorinated biphenyls	44	0.43	0.954
Nitrocompounds	16	0.49	0.918
Steroids	47	0.51	0.769
Ketones	27	0.38	0.976
Esters	47	0.40	0.969
Alcohols and phenols	87	0.34	0.969
Mono- and diacids	27	0.40	0.950
N- and C-containing compounds	90	0.66	0.941
- Amines and anilines (including hydrazines and anilines with polyaromatic system)	43	0.54	0.960
- Heteroaromatic N-containing compounds (including pyridines, pteridines, pyrroles, diazines, di- and triazoles)	33	0.47	0.974
Sugars and carbohydrates	8	0.56	0.832
All amide-containing compounds (heteroatoms and more complex functional groups containing NHCO fragment also allowed)	136	0.65	0.914
- Ureas	9	0.56	0.967
- Mono- and di- amides and carbamates	29	0.58	0.919
Phosphoroorganics (mainly pesticides)	20	0.60	0.925

ANNEX B

TYPICAL ENVIRONMENTAL CONCENTRATIONS

Table B.1 Typical Environmental Concentrations: Linear Alkyl Sulfonates

Sample Type	Location	ΣLAS	Units	Reference
Compost (household)	Denmark	<10	mg/kg dw	Petersen et al., 2003
Sewage sludge	Denmark	2870	mg/kg dw	Petersen et al., 2003
Sewage sludge	Denmark	110	mg/kg dw	Petersen et al., 2003
Sewage sludge	Italy	11500-14000	mg/kg	Cavalli et al., 1993 cit. in Cavalli & Valtorta, 1999
Sewage sludge	Spain	12100-17800	mg/kg	Prats et al., 1993 cit. in Cavalli & Valtorta, 1999
Sewage sludge	Italy	6000+/-1200	mg/kg	Di Corcia et al., 1994 cit. in Cavalli & Valtorta, 1999
Sewage sludge	Netherlands	3400-5930	mg/kg	Feijtel et al., 1995 cit. in Cavalli & Valtorta, 1999
Sewage sludge	Netherlands	205	mg/kg	Feijtel et al., 1995 cit. in Cavalli & Valtorta, 1999
Sewage sludge	Denmark	11-<500	mg/kg	VKI, 1997 cit. in Cavalli & Valtorta, 1999
Sewage sludge	Denmark	1000-16100	mg/kg	VKI, 1997 cit. in Cavalli & Valtorta, 1999
Sewage sludge	Austria	2199-17955	mg/kg ds	Scharf et al., 1997 cit. in IC Consultants Ltd., 2001
Sewage sludge	Germany	50-16000	mg/kg ds	Drescher-Kaden et al., 1992 cit. in IC Consultants Ltd., 2001
Sewage sludge	Denmark	11-16100	mg/kg ds	Torslov et al., 1997 cit. in IC Consultants Ltd., 2001
Sewage sludge	UK	60-18800	mg/kg ds	Holt et al., 1992 cit. in IC Consultants Ltd., 2001
Sewage sludge	Denmark	50-1507	mg/kg dw	Jensen & Jepsen, 2005 (data for 2002)
Sewage sludge	Denmark	16.95-3335	mg/kg	Carlsen et al., 2002
Soil	Denmark	<10	mg/kg dw	Petersen et al., 2003
Sludge-amended soil	Spain	0.7	mg/kg	Prats et al., 1993 cit. in Kloepper-Sams et al., 1996
Soil (0-10cm)	Denmark	0.40-11.23	mg/kg	Carlsen et al., 2002
Sludge-amended soil	UK	0-8	mg/kg	Holt & Bernstein, 1992 cit. in Cavalli & Valtorta, 1999

Table B.2 Typical Environmental Concentrations: Furans and dioxins

Sample type	Location	Σ PCDD	Σ PCDF	Σ PCDD/F	Units	Reference
Fresh sewage sludge	Spain			7-160	I-TEQ (pg/g dw)	Eljarrat et al., 1999 cit. in IC Consultants Ltd., 2001
Archived sewage sludge	Spain			29-8300	I-TEQ (pg/g dw)	Eljarrat et al., 1999 cit. in IC Consultants Ltd., 2001
WWTW effluent	Italy	0.024-16.9			ug/L	Italian RPA cit. in IC Consultants Ltd., 2001
Sewage sludge	Germany			15-45	mg/kg dw	Leschber, 1997 cit. in IC Consultants Ltd., 2001
Sewage sludge	Spain			7-160	mg/kg dw	Eljarrat et al., 1999 cit. in IC Consultants Ltd., 2001
Sewage sludge	Spain			29-8300	mg/kg dw	Eljarrat et al., 1999 cit. in IC Consultants Ltd., 2001
Sewage sludge	Sweden			23-25	mg/kg dw	Rappe et al., 1989 cit. in IC Consultants Ltd., 2001
Sewage sludge	UK			7.6-192	mg/kg dw	UKWIR, 1995 cit. in IC Consultants Ltd., 2001
Sludge – composted	Spain			7.73-78.34	I-TEQ (ng/kg)	Abad et al., 2005
Sludge – thermally dried	Spain			3.14-88.21	I-TEQ (ng/kg)	Abad et al., 2005
Sludge - raw	Spain			6.04-263.84	I-TEQ (ng/kg)	Abad et al., 2005
Sewage sludge	Spain	527-1603	47.1-1173	645-2776	pg/g dw	Eljarrat et al., 2003
Soil from sewage sludge fields	Germany			2.7-5.2	ng TE/kg	Landesanstalt für Umweltschutz, 2003
Soil - no sludge	Germany			1.8	ng TE/kg	Landesanstalt für Umweltschutz, 2003
Soil - archived	Europe			<15-1700	pg/g dw	Green et al., 2004
Soil	UK			44-3300	pg/g dw	Hassanin et al., 2005
Soil	Norway			8.5-2275	pg/g dw	Hassanin et al., 2005
Soil	Switzerland			72-703	ng/kg	Schmid et al., 2005

Table B.3 Typical Environmental Concentrations: Nonylphenols and nonylphenol ethoxylates (NP and NPE)

Sample type	Location	NP	NP1EO	NP2EO	Σ NPE	Units	Reference
Sewage sludge	Denmark	60				mg/kg dw	Petersen et al., 2003
Sewage sludge	Denmark	12.5				mg/kg dw	Petersen et al., 2003
Sewage sludge	Austria					<69, mean 12 mg/kg dw	Scharf et al., 1997 cit. in IC Consultants Ltd, 2001
Sewage sludge	Germany					60-120 mg/kg dw	Leschber, 1997 cit. in IC Consultants Ltd, 2001
Sewage sludge	Germany					3.8-96.3 mg/kg dw	Hessische Landesanstalt fur Umwelt (1991-96) cit. in IC Consultants Ltd, 2001
Sewage sludge	Germany		5-80	<3-80		mg/kg dw	Drescher-Kaden et al., 1992 cit. in IC Consultants Ltd, 2001
Sewage sludge	Denmark					0.3-67 mg/kg dw	Torslov et al., 1997 cit. in IC Consultants Ltd, 2001
Sewage sludge	Sweden					26-1100 mg/kg dw	Wahlberg et al., 1990 cit. in IC Consultants Ltd, 2001
Sewage sludge	Sweden					13-27 mg/kg dw	Staitika Meddelanden, 1998 cit. in IC Consultants Ltd, 2001
Sewage sludge	UK					256-824 mg/kg dw	Sweetman 1994 cit. in IC Consultants Ltd, 2001
Sewage sludge - stored 1 year	Denmark	1.8		6.7		mg/kg	Vikelsøe et al., 2002
Sewage sludge	Greece	0.11	1.01	2.89		µg/kg dw	Gatidou et al., 2007
Composted sewage sludge	Spain					17.9-363.4 mg/kg	Abad et al., 2005
Sewage sludge - thermally dried	Spain					14.3-3150 mg/kg	Abad et al., 2005
Sewage sludge	Greece	3.6-93				12.8-233.5 mg/kg	Fountoulakis et al., 2005
Sewage sludge	Denmark					1-25 mg/kg dw	Jensen and Jepsen, 2005 (data for 2002)
Sewage sludge	Denmark					60 mg/kg dw	Jacobsen et al., 2004
Compost (household)	Denmark	2.7				mg/kg dw	Petersen et al., 2003
Soil - Uncultured	Denmark	0.47		2.9		µg/kg dw	Vikelsøe et al., 2002
Soil - Manured 40 years	Denmark	0.47		2.6		µg/kg dw	Vikelsøe et al., 2002
Soil - Manured 5 years	Denmark	0.98		2.9		µg/kg dw	Vikelsøe et al., 2002
Soil - Art. Fertilised	Denmark	0.24		3.6		µg/kg dw	Vikelsøe et al., 2002
Soil - Low sludge	Denmark	0.04		3.1		µg/kg dw	Vikelsøe et al., 2002
Soil - Normal sludge	Denmark	0.01		16		µg/kg dw	Vikelsøe et al., 2002
Soil - High sludge	Denmark	1450		1170		µg/kg dw	Vikelsøe et al., 2002
Soil - High sludge 2 years later	Denmark	2430		2240		µg/kg dw	Vikelsøe et al., 2002
Soil	Denmark					<0.05 mg/kg dw	Petersen et al., 2003

Table B.4 Typical Environmental Concentrations: Phthalates

Sample type	Location	DEHP	DEP	DBP	Reference
Sewage effluent	UK	1-182 µg/L			Oliver et al., 2007
Sewage - Raw	UK	28-122 µg/L	1-74 µg/L		Oliver et al., 2007
Soil ±manure/sludge	Denmark	12-1900 µg/kg		0.3-453 µg/kg	Vikelsøe et al., 2002
WWTW influent	Norway	<0.5-21 µg/L	1.3-4.9 µg/L		Vogelsang et al., 2006
WWTW effluent	Norway	<0.5-34 µg/L	<0.5-9.9 µg/L		Vogelsang et al., 2006
Sewage - raw	UK	23.6±12.2 µg/L	250±16.7 µg/L	2.54±3.52 µg/L	Oliver et al., 2005
Sewage Sludge - Treated	Germany	27.9-154 mg/kg		0.2-1.7 mg/kg	Fromme et al., 2002
WWTW effluent	Spain		<0.01-13.3 µg/L	<0.03-2.78 µg/L	Céspedes et al., 2006
Soil ± sludge	UK	22-550 µg/kg	0.2-0.9 µg/kg	7.9-11.8 µg/kg	Gibson et al., 2005
Sewage sludge	UK	62482 µg/kg	18 µg/kg	393 µg/kg	Gibson et al., 2005
Composted sewage sludge	Spain	2.2-120.5 µg/kg			Gibson et al., 2005
Sewage sludge – thermally dried	Spain	1.5-3514 mg/kg			Gibson et al., 2005
Sewage sludge - raw	Spain	2.0-258 mg/kg			Gibson et al., 2005
Sewage sludge - treated	Spain	38-267 mg/kg			Bagó et al., 2005
Sewage sludge	Finland	71-88 µg/g			Martinen et al., 2004
Sewage sludge effluent	UK/Germany	<2.4-182 µg/L		<1-14 µg/L	Johnson & Jürgens, 2003
Sewage sludge	Denmark	27-55 mg/kg			Petersen et al., 2003
Household compost	Denmark	18 mg/kg			Petersen et al., 2003

Table B.5 Typical Environmental Concentrations: Brominated Fire Retardants (polybrominated diphenyl ethers, PBDE) ng/g dw

Sample type	Location	#47	#85	#99	#100	#138	#153	#154	#183	#209	ΣPBDE Reference
Sewage sludge	Sweden	15		19	3.5						38 Dodder et al., 2000 cit. in de Wit, 2001
Sewage sludge	Sweden	22		18	5.4						45.4 Nylund et al., 1992 cit. in de Wit, 2001
Sewage sludge	Sweden	53		53	13						119 Sellström & Jansson, 1995; Sellström, 1999 cit. in de Wit, 2001
Sewage sludge	Sweden	39-91		48-120	11-28						98-239 Sellström et al., 1999 cit. in de Wit, 2001
Sewage sludge	Germany										0.4-15 Hagenmaier et al., 1992 cit. in de Wit, 2001
Sewage sludge	Sweden	7.0-100		8.1-150	1.5-22		0.8-18	0.6-10		5.6-1000	18-260 Law et al., 2006
Sewage Sludge	UK	<0.3-48	<0.3-2.7	<0.3-320	<0.3-11	<0.3-<1	<0.3-5.5	<0.3-5		<0.6-390	Öberg, 2002
Sewage sludge	Spain	1.8- 83.6	0.6-1.6	23.4- 64.2	0.2-14.0		2.7-16.4	1.7-10.4		786- 18032	844- 18147 Fabrellas et al., 2004
Soil	UK	0.035- 0.91		0.088- 1.71	0.017- 0.392		0.014- 0.097	0.0066- 0.357			0.073- 3.89 Harrad & Hunter, 2006
Soil - grassland	UK	0.007- 0.52	0.009- 0.18	0.078- 3.2	0.008- 0.47	0.026- 0.068	0.019- 0.60	0.008- 0.24	0.010- 0.90		0.065- 6.0 Hassanin et al., 2004
Soil – woodland	UK	0.050- 1.4	0.007- 1.7	0.190- 3.2	0.011- 0.36	0.013- 0.27	0.038- 1.2	0.014- 0.42	0.010- 7.0		0.110-12 Hassanin et al., 2004
Soil - woodland	Norway	0.012- 0.86	0.011- 0.090	0.063- 1.4	0.018- 0.23	0.020- 0.14	0.011- 0.27	0.013- 0.31	0.009- 0.13		0.130- 3.0 Hassanin et al., 2004
Soil	Sweden	0.015- 450	<0.001- 17	0.011- 590	<0.001- 120		<0.001- 52	<0.001- 46	<0.002- <3	0.071- 2200	0.12- 3900 Sellström et al., 2005

Table B.6 Typical Environmental Concentrations: Pharmaceuticals

Sample type	Location	Ciprofloxacin	Erythromycin	Phenazone	Salicylic Acid	Propranolol	Carbamazepine	Ketoprofen	Bezafibrate	Diclofenac	Reference
WWTW Effluent	Germany								4.6		µg/L Ternes, 1998 cit. in Daughton & Ternes, 1999
WWTW influent	Brazil							0.5	1.2	0.8	µg/L Stumpf et al., 1999
Effluent-biological filter	Brazil							0.3	0.8	0.7	µg/L Stumpf et al., 1999
Effluent-activated sludge	Brazil							0.2	0.6	0.2	µg/L Stumpf et al., 1999
POTW max. effluent	unspecified			0.41	0.14	0.29	6.3	0.38			2.1 µg/L Daughton & Ternes, 1999
WWTW influent	Switzerland									500-1800	ng/L Daughton & Ternes, 1999
WWTW influent	Germany				4.6						3.02 µg/L Herberer, 2002
WWTW Effluent	Germany				0.04						2.51 µg/L Herberer, 2002
Wastewater effluent	Switzerland	249-405									ng/L Herberer, 2002
River sediments	Spain		10-630							nd-130	ng/kg Zuccato et al., 2000

ANNEX C

MODELLED COMPOUNDS: K_{OC} and Solubility at pH 7Table C.1 Modelled K_{OC} and Solubility at pH 7

Group	Compound investigated		CAS Number	Molecular weight g/mole	Modelled solubility at pH 7 (g/l)	Modelled K_{OC} at pH 7 (l/g)
PCB	PCB 28	2,4,4'-trichlorobiphenyl	7012-35-5	255.961	1.00E-4	2.67E+4
	PCB 52	2,2',5,5'-tetrachlorobiphenyl	35693-99-3	289.922	5.67E-5	3.95E+4
	PCB 77	3,3',4,4'-tetrachlorobiphenyl	32598-13-3	289.922	9.74E-6	4.36E+4
	PCB 81	3,4,4',5-tetrachlorobiphenyl	70362-50-4	289.922	1.01E-5	4.43E+4
	PCB 101	2,2',4,5,5'-pentachlorobiphenyl	37680-73-2	326.433	1.17E-5	7.01E+4
	PCB 105	2,3,3',4,4'-pentachlorobiphenyl	32598-14-4	326.433	1.03E-5	6.89E+4
	PCB 114	2,3,4,4',5-pentachlorobiphenyl	74472-37-0	326.433	1.14E-5	6.39E+4
	PCB 118	2,3',4,4',5-pentachlorobiphenyl	31508-00-6	326.433	9.83E-6	7.36E+4
	PCB 123	2,3',4,4',5-pentachlorobiphenyl	65510-44-3	326.433	8.65E-6	8.19E+4
	PCB 126	3,3',4,4',5-pentachlorobiphenyl	57465-28-8	326.433	8.12E-6	7.69E+4
	PCB 138	2,2',3,4,4',5'-hexachlorobiphenyl	35065-28-2	360.878	3.58E-6	1.16E+5
	PCB 153	2,2',4,4',5,5'-hexachlorobiphenyl	35065-27-1	360.878	3.42E-6	1.24E+5
	PCB 156	2,3,3',4,4',5-hexachlorobiphenyl	38380-08-4	360.878	3.35E-6	1.11E+5
	PCB 157	2,3,3',4,4',5'-hexachlorobiphenyl	69782-90-7	360.878	2.98E-6	1.22E+5
	PCB 167	2,3',4,4',5,5'-hexachlorobiphenyl	52663-72-6	360.878	2.85E-6	1.30E+5
	PCB 169	3,3',4,4',5,5'-hexachlorobiphenyl	32774-16-6	360.878	2.36E-6	1.36E+5
	PCB 180	2,2',3,4,4',5,5'-heptachlorobiphenyl	35065-29-3	395.323	1.18E-6	1.87E+5
	PCB 189	2,3,3',4,4',5,5'-heptachlorobiphenyl	39635-31-9	395.323	9.81E-7	1.96E+5
	PCB 209	2,2',3,3',4,4',5,5',6,6'-heptachlorobiphenyl	2051-24-3	498.658	7.04E-8	6.08E+5
PAH	Acenaphthene		83-32-9	154.208	3.40E-3	4.52E+3
	Acenaphthylene		208-96-8	152.192	1.60E-3	4.98E+3
	Benzo(a)anthracene		56-55-3	228.288	1.46E-5	3.91E+5
	Chrysene		218-01-9	228.288	9.63E-6	3.91E+5
	Dibenzo(a,h)anthracene		53-70-3	278.347	6.84E-7	1.83E+5
	Fluoranthene		206-44-0	202.251	1.00E-4	1.55E+4
	Fluorene		86-73-7	166.219	2.10E-3	4.39E+3
	Phenanthrene		85-01-8	178.229	5.00E-4	8.37E+3

Group	Compound investigated	CAS Number	Molecular weight g/mole	Modelled solubility at pH 7 (g/l)	Modelled K _{OC} at pH 7 (l/g)
PAH	Pyrene	129-00-0	202.251	4.36E-5	1.55E+4
	Benzo(a)pyrene	50-32-8	252.309	1.86E-6	7.24E+4
	Benzo(g,h,i)perylene	191-24-2	276.331	1.63E-6	1.34E+5
	Indeno(1,2,3-cd)pyrene	193-39-5	276.331	1.89E-6	1.34E+5
	Anthracene	120-12-7	178.229	5.00E-4	8.37E+3
	Naphthalene	91-20-3	128.171	1.21E-1	1.79E+3
	Benzo(b)fluoranthene	205-99-2	252.309	5.16E-6	7.24E+4
	Benzo(k)fluoranthene	207-08-9	252.309	3.25E-6	7.24E+4
NP	4-n-nonylphenol		220.35	1.95E-2	5.56E+4
	4-(2,2,4-trimethylhexyl)phenol	104-40-5	220.35	3.61E-2	2.79E+4
	4-(2,2,3-trimethylhexyl)phenol		220.35	3.61E-2	2.79E+4
	4-(3,3,4-trimethylhexyl)phenol		220.35	3.61E-2	2.79E+4
	4-(3,5-dimethylheptan-2-yl)phenol		220.35	3.62E-2	2.78E+\$
	4-(2,3,4-trimethylhexyl)phenol		220.35	3.68E-2	2.78E+4
	4-(2,5-dimethylheptan-2-yl)phenol		220.35	4.18E-2	2.79E+4
	4-(2,2,4,4-tetramethylpentyl)phenol		220.35	4.38E-2	2.22E+4
	4-(3,3,4-trimethylhexan-2-yl)phenol		220.35	4.39E-2	2.22E+4
	4-(2,3,3,4-tetramethylpentyl)phenol		220.35	4.47E-2	2.22E+4
	4-(2,2,3,4-tetramethylpentyl)phenol		220.35	4.47E-2	2.22E+4
	4-(3,4,5-trimethylhexan-2-yl)phenol		220.35	4.48E-2	2.21E+4
	4-(2,3,5-trimethylhexan-2-yl)phenol		220.35	5.17E-2	2.22E+4
	4-(3,3,4,4-tetramethylpentan-2-yl)phenol		220.35	5.32E-2	1.77E+4
	4-(2,3,3,4-tetramethylpentan-2-yl)phenol		220.35	6.27E-2	1.77E+4
NPE	Ethanol, 2-(nonylphenoxy)-	27986-36-3	264.403	2.00E-4	3.71E+4
	Ethanol, 2-[2-(4-nonylphenoxy)ethoxy]-	20427-84-3	308.456	1.07E-5	3.38E+4
	Ethanol, 2-[2-(nonylphenoxy)ethoxy]-	27176-93-8	308.456	1.07E-5	3.38E+4
	Ethanol, 2-[2-[2-(4-nonylphenoxy)ethoxy]ethoxy]ethoxy]-	7311-27-5	396.561	6.30E-3	1.38E+4
	2-(2-(2-(2-(2-(2,3,4-tetramethylpentyl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol		396.561	1.91E-2	5.49E+3
	2-(2-(2-(2-(2-(2,3,5-trimethylhexan-2-yl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol		396.561	1.95E-2	5.49E+3
	2-(2-(2-(2-(2-(3,3,5-trimethylhexan-2-yl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol		396.561	1.96E-2	5.49E+3
	2-(2-(2-(2-(2-(3,4,5-trimethylhexan-2-yl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol		396.561	1.98E-2	5.48E+3

Group	Compound investigated	CAS Number	Molecular weight g/mole	Modelled solubility at pH 7 (g/l)	Modelled K_{oc} at pH 7 (l/g)
NPE	2-(2-(2-(2-(4,4,5-trimethylhexan-2-yl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol		396.561	1.96E-2	5.49E+3
	2-(2-(2-(2-(7-methyloctyl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol	37205-87-1	396.561	8.40E-2	1.09E+4
	2-(2-(2-(2-(2,2,3,4-tetramethylpentyl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol		396.561	1.91E-2	5.49E+3
	2-(2-(2-(2-(2,2,4,4-tetramethylpentyl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol		396.561	1.90E-2	5.50E+3
	2-(2-(2-(2-(2,4,5-trimethylhexan-2-yl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol		396.561	1.95E-2	5.49E+3
	2-(2-(2-(2-(3,3,4,4-tetramethylpentyl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol		396.561	1.90E-2	5.50E+3
	2-(2-(2-(2-(3,4,5-trimethylhexan-2-yl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol		396.561	1.98E-2	5.48E+4
	2-(2-(2-(2-(3,4,5-trimethylhexan-2-yl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol		396.561	1.98E-2	5.48E+3
	2-(2-(2-(2-(7-methyloctyl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol		396.561	8.40E-3	1.09E+4
	2-(2-(2-(2-(5,7-dimethyloctyl)phenoxy)ethoxy)ethoxy)ethoxy)ethanol		410.587	5.50E-3	1.69E+4
	3,6,9,12-Tetraoxatetradecan-1-ol,14-(4-nonylphenoxy)-	20636-48-0	440.613	9.10E-3	8.79E+3
	3,6,9,12-Tetraoxatetradecan-1-ol,14-(nonylphenoxy)-	26264-02-8	440.613	9.10E-3	8.79E+3
	3,6,9,12,15-Pentaoxaheptadecan-1-ol,17-(nonylphenoxy)-	27111-01-1	484.666	1.40E-2	5.61E+3
	3,6,9,12,15,18,21-Heptaoxatricosan-1-ol,23-(nonylphenoxy)-	27177-05-5	572.771	4.25E-2	2.29E+3
	3,6,9,12,15,18,21,24-Octaoxahexacosan-1-ol,26-(nonylphenoxy)-	26571-11-9	616.823	8.37E-2	1.46E+3
3,6,9,12,15,18,21,24,27-Nonaoxanonacosan-1-ol,29-(isononylphenoxy)-	65455-72-3	660.876	2.35E-1	7.41E+2	
3,6,9,12,15,18,21,24,27-Nonaoxanonacosan-1-ol,29-(nonylphenoxy)-	27177-08-8	660.876	1.79E-1	9.33E+2	
LAS	C10-16-alkyl derivatives of benzene sulfonic acid	68584-22-5	298.441	1.12E+1	9.64E+0
	Dodecylbenzene sulfonic acid	27176-87-0	326.494	2.54E+0	3.65E+1
	Tridecylbenzene sulfonic acid	25496-01-9	340.521	1.23E+0	7.10E+1
Phthalate	DEHP (Diethylhexylphthalate)	117-81-7	390.556	2.00E-4	1.30E+6
	DBP (Dibutylphthalate)	84-74-2	278.344	1.46E-2	1.00E+4

ANNEX D

FIELD DATA

D.1 Introduction

The data in the following tables are extracted from the Horizontal-ORG sampling report (Lambkin et al., 2007).

For many samples, the concentrations of PAH and PCB compounds were below the limit of quantification (LOQ). Those data above the LOQ are highlighted in blue.

Table D.1 Spain Field 2: Analytical results (as reported)

	Sample Number															
	A1	A2	A3	A4	A5	A6	A7	A8	B1	B2	B3	B4	B5	B6	B7	B8
pH	8.33	8.22	8.19	8.17	8.29	8.38	8.06	8.29	8.26	8.31	8.17	8.17	8.10	7.96	8.45	7.82
DS%	91.7	94.6	88.1	93.1	87.9	93.2	89.9	90.7	92.3	93.9	91.9	90.1	92.7	90.2	92.0	92.8
LOI%	9.15	7.20	6.83	12.3	7.11	6.58	7.34	6.08	8.15	5.94	7.93	7.63	5.82	9.35	8.07	7.23
PAH (mg kg⁻¹)																
Acenaphthene	<0.30	<0.25	<0.30	<0.30	<0.28	<0.30	<0.28	<0.30	<0.29	<0.29	<0.27	<0.32	<0.28	<0.28	<0.25	<0.30
Acenaphthylene	<0.30	<0.25	<0.30	<0.30	<0.28	<0.30	<0.28	<0.30	<0.29	<0.29	<0.27	<0.32	<0.28	<0.28	<0.25	<0.30
Anthracene	<0.30	<0.25	<0.30	<0.30	<0.28	<0.30	<0.28	<0.30	<0.29	<0.29	<0.27	<0.32	<0.28	<0.28	<0.25	<0.30
Benzo (a) Anthracene	<0.30	<0.25	<0.30	<0.30	<0.28	<0.30	<0.28	<0.30	<0.29	<0.29	<0.27	<0.32	<0.28	<0.28	<0.25	<0.30
Benzo (a) Pyrene	<0.30	<0.25	<0.30	<0.30	<0.28	<0.30	<0.28	<0.30	<0.29	<0.29	<0.27	<0.32	<0.28	<0.28	<0.25	<0.30
Benzo (b&k) Fluoranthene	<0.30	<0.25	<0.30	<0.30	<0.28	<0.30	<0.28	<0.30	<0.29	<0.29	<0.27	<0.32	<0.28	<0.28	<0.25	<0.30
Benzo (g,h,i) Perylene	<0.30	<0.25	<0.30	<0.30	<0.28	<0.30	<0.28	<0.30	<0.29	<0.29	<0.27	<0.32	<0.28	<0.28	<0.25	<0.30
Chrysene	<0.30	<0.25	<0.30	<0.30	<0.28	<0.30	<0.28	<0.30	<0.29	<0.29	<0.27	<0.32	<0.28	<0.28	<0.25	<0.30
Dibenzo (a,h) Anthracene	<0.30	<0.25	<0.30	<0.30	<0.28	<0.30	<0.28	<0.30	<0.29	<0.29	<0.27	<0.32	<0.28	<0.28	<0.25	<0.30
Fluoranthene	<0.30	<0.25	<0.30	<0.30	<0.28	<0.30	<0.28	<0.30	<0.29	<0.29	<0.27	<0.32	<0.28	<0.28	<0.25	<0.30
Fluorene	<0.30	<0.25	<0.30	<0.30	<0.28	<0.30	<0.28	<0.30	<0.29	<0.29	<0.27	<0.32	<0.28	<0.28	<0.25	<0.30
Indeno (1,2,3- cd) Pyrene	<0.30	<0.25	<0.30	<0.30	<0.28	<0.30	<0.28	<0.30	<0.29	<0.29	<0.27	<0.32	<0.28	<0.28	<0.25	<0.30
Naphthalene	<0.30	<0.25	<0.30	<0.30	<0.28	<0.30	<0.28	<0.30	<0.29	<0.29	<0.27	<0.32	<0.28	<0.28	<0.25	<0.30
Phenanthrene	<0.30	<0.25	<0.30	<0.30	<0.28	<0.30	<0.28	<0.30	<0.29	<0.29	<0.27	<0.32	<0.28	<0.28	<0.25	<0.30
Pyrene	<0.30	<0.25	<0.30	<0.30	<0.28	<0.30	<0.28	<0.30	<0.29	<0.29	<0.27	<0.32	<0.28	<0.28	<0.25	<0.30
PAH Total (EPA 16)	<4.5	<3.75	<4.5	<4.5	<4.2	<4.5	<4.2	<4.5	<4.35	<4.35	<4.05	<4.8	<4.2	<4.2	<3.75	<4.5
PCB (mg kg⁻¹)																
PCB 28	<1.2	<1	<1.2	<1.2	<1.12	<1.2	<1.12	<1.2	<1.16	<1.16	<1.08	<1.28	<1.12	10.9	<1	<1.2
PCB 52	<1.2	<1	<1.2	<1.2	<1.12	<1.2	<1.12	<1.2	<1.16	<1.16	<1.08	<1.28	<1.12	26.9	<1	<1.2
PCB 101	1.43	1.46	<1.2	<1.2	<1.12	<1.2	<1.12	<1.2	<1.16	<1.16	<1.08	<1.28	<1.12	13.7	1.18	1.37
PCB 118	<1.2	<1	<1.2	<1.2	<1.12	<1.2	<1.12	<1.2	<1.16	<1.16	<1.08	<1.28	<1.12	5.01	<1	<1.2
PCB 138	2.11	2.23	1.71	1.61	<1.12	<1.2	<1.12	1.29	1.22	1.57	2.02	2.37	<1.12	14.6	2.49	2.59
PCB 153	5.15	4.03	3.74	<1.2	1.87	<1.2	<1.12	2.30	3.02	2.13	2.93	5.30	<1.12	47.5	6.75	6.06
PCB 180	3.35	4.67	2.75	3.04	2.23	2.06	2.18	3.07	2.42	3.24	4.00	3.97	1.53	44.3	5.75	4.81

Table D.1 (cont.) Spain Field 2: Analytical results (as reported)

	Sample Number															
	C1	C2	C3	C4	C5	C6	C7	C8	D1	D2	D3	D4	D5	D6	D7	D8
pH	7.88	8.04	8.13	7.98	7.58	7.73	8.15	8.19	8.11	8.12	8.23	8.22	8.27	7.77	7.73	8.31
DS%	94.1	91.7	95.5	92.3	91.5	90.5	93.4	89.7	93.2	94.9	94.0	95.4	95.1	93.5	92.4	95.1
LOI%	10.0	7.05	6.21	5.95	7.48	7.58	8.29	7.03	11.8	8.42	8.16	7.83	8.24	9.00	7.43	9.87
PAH (mg kg⁻¹)																
Acenaphthene	<0.28	<0.28	<0.25	<0.30	<0.32	<0.29	<0.30	<0.32	<0.25	<0.25	<0.30	<0.26	<0.27	<0.26	<0.25	0.32
Acenaphthylene	<0.28	<0.28	<0.25	<0.30	<0.32	<0.29	<0.30	<0.32	<0.25	<0.25	<0.30	<0.26	<0.27	<0.26	<0.25	<0.28
Anthracene	<0.28	<0.28	<0.25	<0.30	<0.32	<0.29	<0.30	<0.32	<0.25	<0.25	<0.30	<0.26	<0.27	<0.26	<0.25	<0.28
Benzo (a) Anthracene	<0.28	<0.28	<0.25	<0.30	<0.32	<0.29	<0.30	<0.32	<0.25	<0.25	<0.30	<0.26	<0.27	<0.26	<0.25	0.61
Benzo (a) Pyrene	<0.28	<0.28	<0.25	<0.30	<0.32	<0.29	<0.30	<0.32	<0.25	<0.25	<0.30	<0.26	<0.27	<0.26	<0.25	<0.28
Benzo (b&k) Fluoranthene	<0.28	<0.28	<0.25	<0.30	<0.32	<0.29	<0.30	<0.32	<0.25	<0.25	<0.30	<0.26	<0.27	<0.26	<0.25	<0.28
Benzo (g,h,i) Perylene	<0.28	<0.28	<0.25	<0.30	<0.32	<0.29	<0.30	<0.32	<0.25	<0.25	<0.30	<0.26	<0.27	<0.26	<0.25	<0.28
Chrysene	<0.28	<0.28	<0.25	<0.30	<0.32	<0.29	<0.30	<0.32	<0.25	<0.25	<0.30	<0.26	<0.27	<0.26	<0.25	<0.28
Dibenzo (a,h) Anthracene	<0.28	<0.28	<0.25	<0.30	<0.32	<0.29	<0.30	<0.32	<0.25	<0.25	<0.30	<0.26	<0.27	<0.26	<0.25	<0.28
Fluoranthene	<0.28	<0.28	<0.25	<0.30	<0.32	<0.29	<0.30	<0.32	<0.25	<0.25	<0.30	<0.26	<0.27	<0.26	<0.25	0.63
Fluorene	<0.28	<0.28	<0.25	<0.30	<0.32	<0.29	<0.30	<0.32	<0.25	<0.25	<0.30	<0.26	<0.27	<0.26	<0.25	<0.28
Indeno (1,2,3- cd) Pyrene	<0.28	<0.28	<0.25	<0.30	<0.32	<0.29	<0.30	<0.32	<0.25	<0.25	<0.30	<0.26	<0.27	<0.26	<0.25	<0.28
Naphthalene	<0.28	<0.28	<0.25	<0.30	<0.32	<0.29	<0.30	<0.32	<0.25	<0.25	<0.30	<0.26	<0.27	<0.26	<0.25	<0.28
Phenanthrene	<0.28	<0.28	<0.25	<0.30	<0.32	<0.29	<0.30	<0.32	<0.25	<0.25	<0.30	<0.26	<0.27	<0.26	<0.25	0.71
Pyrene	<0.28	<0.28	<0.25	<0.30	<0.32	<0.29	<0.30	<0.32	<0.25	<0.25	<0.30	<0.26	<0.27	<0.26	<0.25	0.58
PAH Total (EPA 16)	<4.2	<4.2	<3.75	<4.5	<4.8	<4.35	<4.5	<4.8	<3.75	<3.75	<4.5	<3.9	<4.05	<3.9	<3.75	4.25
PCB (mg kg⁻¹)																
PCB 28	<1.12	<1.12	<1	<1.2	<1.28	<1.16	<1.2	<1.28	<1	<1	<1.2	<1.04	<1.08	<1.04	<1	<1.12
PCB 52	<1.12	<1.12	<1	<1.2	<1.28	<1.16	<1.2	<1.28	<1	<1	<1.2	<1.04	<1.08	<1.04	<1	<1.12
PCB 101	1.51	<1.12	<1	<1.2	1.59	1.22	<1.2	<1.28	1.3	<1	<1.2	<1.04	1.14	<1.04	<1	2.54
PCB 118	<1.12	<1.12	<1	<1.2	<1.28	<1.16	<1.2	<1.28	<1	<1	<1.2	<1.04	<1.08	<1.04	<1	<1.12
PCB 138	1.76	1.41	<1	<1.2	2.94	1.80	<1.2	2.09	1.93	1.68	<1.2	<1.04	1.72	<1.04	<1	5.55
PCB 153	3.50	<1.12	<1	<1.2	8.00	4.26	<1.2	4.71	5.10	2.93	<1.2	<1.04	4.66	<1.04	<1	14.2
PCB 180	3.39	2.38	<1	1.90	7.17	4.71	2.99	4.60	4.23	2.88	2.15	2.53	4.12	2.39	1.94	11.8

Table D.1 (cont.) Spain Field 2: Analytical results (as reported)

	Sample Number															
	E1	E2	E3	E4	E5	E6	E7	E8	F1	F2	F3	F4	F5	F6	F7	F8
pH	8.16	7.57	7.88	8.06	8.20	7.99	8.07	8.31	7.68	8.52	7.93	8.49	8.07	8.00	8.08	8.06
DS%	94.9	94.8	93.8	94.5	93.3	94.6	93.3	96.1	95.5	93.1	94.2	95.2	93.1	96.3	90.9	94.1
LOI%	9.87	10.3	8.76	7.36	7.51	7.83	6.77	7.44	9.51	9.04	8.88	8.44	7.05	7.22	6.96	8.55
PAH (mg kg⁻¹)																
Acenaphthene	<0.30	<0.25	<0.26	<0.27	<0.28	<0.30	<0.28	<0.28	<0.25	<0.27	<0.29	<0.29	<0.25	<0.30	<0.28	<0.26
Acenaphthylene	<0.30	<0.25	<0.26	<0.27	<0.28	<0.30	<0.28	<0.28	<0.25	<0.27	<0.29	<0.29	<0.25	<0.30	<0.28	<0.26
Anthracene	<0.30	<0.25	<0.26	<0.27	<0.28	<0.30	<0.28	<0.28	<0.25	<0.27	<0.29	<0.29	<0.25	<0.30	<0.28	<0.26
Benzo (a) Anthracene	<0.30	<0.25	<0.26	<0.27	<0.28	<0.30	0.87	<0.28	<0.25	<0.27	<0.29	<0.29	<0.25	<0.30	<0.28	<0.26
Benzo (a) Pyrene	<0.30	<0.25	<0.26	<0.27	<0.28	<0.30	<0.28	<0.28	<0.25	<0.27	<0.29	<0.29	<0.25	<0.30	<0.28	<0.26
Benzo (b&k) Fluoranthene	<0.30	<0.25	<0.26	<0.27	<0.28	<0.30	<0.28	<0.28	<0.25	<0.27	<0.29	<0.29	<0.25	<0.30	<0.28	<0.26
Benzo (g,h,i) Perylene	<0.30	<0.25	<0.26	<0.27	<0.28	<0.30	<0.28	<0.28	<0.25	<0.27	<0.29	<0.29	<0.25	<0.30	<0.28	<0.26
Chrysene	<0.30	<0.25	<0.26	<0.27	<0.28	<0.30	<0.28	<0.28	<0.25	<0.27	<0.29	<0.29	<0.25	<0.30	<0.28	<0.26
Dibenzo (a,h) Anthracene	<0.30	<0.25	<0.26	<0.27	<0.28	<0.30	<0.28	<0.28	<0.25	<0.27	<0.29	<0.29	<0.25	<0.30	<0.28	<0.26
Fluoranthene	<0.30	<0.25	<0.26	<0.27	<0.28	<0.30	<0.28	<0.28	<0.25	<0.27	<0.29	<0.29	<0.25	<0.30	<0.28	<0.26
Fluorene	<0.30	<0.25	<0.26	<0.27	<0.28	<0.30	<0.28	<0.28	<0.25	<0.27	<0.29	<0.29	<0.25	<0.30	<0.28	<0.26
Indeno (1,2,3- cd) Pyrene	<0.30	<0.25	<0.26	<0.27	<0.28	<0.30	<0.28	<0.28	<0.25	<0.27	<0.29	<0.29	<0.25	<0.30	<0.28	<0.26
Naphthalene	<0.30	<0.25	<0.26	<0.27	<0.28	<0.30	<0.28	<0.28	<0.25	<0.27	<0.29	<0.29	<0.25	<0.30	<0.28	<0.26
Phenanthrene	<0.30	<0.25	<0.26	<0.27	<0.28	<0.30	<0.28	<0.28	<0.25	<0.27	<0.29	<0.29	<0.25	<0.30	<0.28	<0.26
Pyrene	<0.30	<0.25	<0.26	<0.27	<0.28	<0.30	<0.28	<0.28	<0.25	<0.27	<0.29	<0.29	<0.25	<0.30	<0.28	<0.26
PAH Total (EPA 16)	<4.5	<3.75	<3.9	<4.05	<4.2	<4.5	<4.2	<4.2	<3.75	<4.05	<4.35	<4.35	<3.75	<4.5	<4.2	<3.9
PCB (mg kg⁻¹)																
PCB 28	<1.2	<1.0	<1.04	<1.08	<1.12	<1.2	<1.12	<1.12	<1.0	<1.08	<1.16	<1.16	<1.0	<1.2	<1.12	6.40
PCB 52	<1.2	<1.0	<1.04	<1.08	<1.12	<1.2	<1.12	<1.12	<1.0	<1.08	<1.16	<1.16	<1.0	<1.2	<1.12	13.6
PCB 101	<1.2	<1.0	<1.04	<1.08	<1.12	1.36	<1.12	1.22	<1.0	<1.08	<1.16	<1.16	<1.0	<1.2	<1.12	18.6
PCB 118	<1.2	<1.0	<1.04	<1.08	<1.12	<1.2	<1.12	<1.12	<1.0	<1.08	<1.16	<1.16	<1.0	<1.2	<1.12	6.40
PCB 138	<1.2	<1.0	1.52	1.55	<1.12	2.33	1.57	2.38	2.12	1.19	<1.16	<1.16	1.61	1.39	1.97	15.4
PCB 153	<1.2	1.91	<1.04	2.27	<1.12	6.54	<1.12	5.77	5.25	2.28	<1.16	2.27	2.28	<1.2	3.98	55.6
PCB 180	1.87	1.96	3.13	3.68	1.98	5.29	3.52	5.16	4.61	2.85	1.68	2.12	3.03	3.20	4.33	39.7

Table D.1 (cont.) Spain Field 2: Analytical results (as reported)

	Sample Number															
	G1	G2	G3	G4	G5	G6	G7	G8	H1	H2	H3	H4	H5	H6	H7	H8
pH	7.71	7.88	8.12	8.34	8.30	8.11	8.02	8.42	8.15	8.37	8.31	8.24	7.66	8.47	8.13	8.48
DS%	95.0	94.5	93.0	91.7	93.0	95.1	92.8	95.4	95.6	90.4	92.7	89.2	94.2	93.1	91.9	94.5
LOI%	11.6	10.5	9.93	7.05	7.45	8.35	7.39	7.33	9.27	9.56	7.83	8.41	7.94	9.42	8.34	6.55
PAH (mg kg⁻¹)																
Acenaphthene	<0.32	<0.28	<0.32	<0.28	<0.30	<0.27	<0.28	<0.29	<0.25	<0.26	<0.29	<0.32	<0.31	<0.30	<0.28	<0.26
Acenaphthylene	<0.32	<0.28	<0.32	<0.28	<0.30	<0.27	<0.28	<0.29	<0.25	<0.26	<0.29	<0.32	<0.31	<0.30	<0.28	<0.26
Anthracene	<0.32	<0.28	<0.32	<0.28	<0.30	<0.27	<0.28	<0.29	<0.25	<0.26	<0.29	<0.32	<0.31	<0.30	<0.28	<0.26
Benzo (a) Anthracene	<0.32	<0.28	<0.32	<0.28	<0.30	<0.27	<0.28	<0.29	<0.25	<0.26	<0.29	<0.32	<0.31	<0.30	<0.28	<0.26
Benzo (a) Pyrene	<0.32	<0.28	<0.32	<0.28	<0.30	<0.27	<0.28	<0.29	<0.25	<0.26	<0.29	<0.32	<0.31	<0.30	<0.28	<0.26
Benzo (b&k) Fluoranthene	<0.32	<0.28	<0.32	<0.28	<0.30	<0.27	<0.28	<0.29	<0.25	<0.26	<0.29	<0.32	<0.31	<0.30	<0.28	<0.26
Benzo (g,h,i) Perylene	<0.32	<0.28	<0.32	<0.28	<0.30	<0.27	<0.28	<0.29	<0.25	<0.26	<0.29	<0.32	<0.31	<0.30	<0.28	<0.26
Chrysene	<0.32	<0.28	<0.32	<0.28	<0.30	<0.27	<0.28	<0.29	<0.25	<0.26	<0.29	<0.32	<0.31	<0.30	<0.28	<0.26
Dibenzo (a,h) Anthracene	<0.32	<0.28	<0.32	<0.28	<0.30	<0.27	<0.28	<0.29	<0.25	<0.26	<0.29	<0.32	<0.31	<0.30	<0.28	<0.26
Fluoranthene	<0.32	<0.28	<0.32	<0.28	<0.30	<0.27	<0.28	<0.29	<0.25	<0.26	<0.29	<0.32	<0.31	<0.30	<0.28	<0.26
Fluorene	<0.32	<0.28	<0.32	<0.28	<0.30	<0.27	<0.28	<0.29	<0.25	<0.26	<0.29	<0.32	<0.31	<0.30	<0.28	<0.26
Indeno (1,2,3- cd) Pyrene	<0.32	<0.28	<0.32	<0.28	<0.30	<0.27	<0.28	<0.29	<0.25	<0.26	<0.29	<0.32	<0.31	<0.30	<0.28	<0.26
Naphthalene	<0.32	<0.28	<0.32	<0.28	<0.30	<0.27	<0.28	<0.29	<0.25	<0.26	<0.29	<0.32	<0.31	<0.30	<0.28	<0.26
Phenanthrene	<0.32	<0.28	<0.32	<0.28	<0.30	<0.27	<0.28	<0.29	<0.25	<0.26	<0.29	<0.32	<0.31	<0.30	<0.28	<0.26
Pyrene	<0.32	<0.28	<0.32	<0.28	<0.30	<0.27	<0.28	<0.29	<0.25	<0.26	<0.29	<0.32	<0.31	<0.30	<0.28	<0.26
PAH Total (EPA 16)	<4.8	<4.2	<4.8	<4.2	<4.5	<4.05	<4.2	<4.35	<3.75	<3.9	<4.35	<4.8	<4.65	<4.5	<4.2	<3.9
PCB (mg kg⁻¹)																
PCB 28	7.74	6.50	<1.28	<1.12	<1.2	5.56	6.55	6.61	4.84	6.19	3.13	<1.28	7.87	<1.2	6.61	6.50
PCB 52	16.4	13.4	<1.28	<1.12	<1.2	13.2	16.2	13.8	11.9	14.4	10.3	<1.28	17.4	<1.2	14.8	13.9
PCB 101	24.1	18.4	<1.28	<1.12	<1.2	19.0	20.3	19.2	16.2	19.5	12.1	<1.28	22.2	<1.2	18.3	17.0
PCB 118	8.83	6.94	<1.28	<1.12	<1.2	6.70	6.83	7.13	6.05	6.34	6.44	<1.28	8.00	<1.2	6.83	6.45
PCB 138	21.9	16.5	<1.28	<1.12	<1.2	18.1	23.1	16.8	15.7	16.9	14.4	1.71	20.1	1.99	17.9	16.1
PCB 153	71.0	56.0	<1.28	<1.12	<1.2	57.2	67.2	56.9	51.5	54.6	41.9	4.57	67.6	5.57	61.0	51.5
PCB 180	59.6	50.2	2.69	1.49	1.51	50.8	55.1	49.7	45.3	43.6	37.2	3.77	51.7	3.77	46.5	45.5

Table D.2 STWB: Analytical results (as reported)

	Sample Number															
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
pH	5.28	6.37	6.42	6.31	5.56	6.50	6.25	5.30	6.27	5.22	6.12	5.78	5.97	5.97	6.04	5.58
DS%	40.6	46.0	48.2	40.3	40.8	41.1	42.8	41.4	42.8	40.1	41.3	41.1	40.9	44.0	41.9	40.4
LOI%	57.2	56.4	62.9	62.9	52.8	54.3	57.3	49.6	57.3	57.0	57.8	58.4	56.9	58.6	57.0	60.5
PAH (mg kg⁻¹)																
Acenaphthene	0.68	0.58	<0.30	1.50	<0.28	<0.30	<0.30	<0.37	<0.31	<0.28	<0.34	0.44	<0.30	<0.30	<0.34	<0.28
Acenaphthylene	<0.26	<0.27	<0.30	<0.37	<0.28	<0.30	<0.30	<0.37	<0.31	<0.28	<0.34	<0.32	<0.30	<0.30	<0.34	<0.28
Anthracene	<0.26	<0.27	<0.30	<0.37	<0.28	<0.30	<0.30	<0.37	<0.31	<0.28	<0.34	<0.32	<0.30	<0.30	<0.34	<0.28
Benzo (a) Anthracene	<0.26	<0.27	0.34	<0.37	<0.28	0.58	<0.30	0.79	<0.31	0.50	<0.34	<0.32	0.44	0.35	<0.34	<0.28
Benzo (a) Pyrene	<0.26	<0.27	<0.30	<0.37	<0.28	<0.30	<0.30	<0.37	<0.31	0.29	<0.34	<0.32	<0.30	<0.30	<0.34	<0.28
Benzo (b&k) Fluoranthene	0.74	0.65	0.90	0.93	0.72	0.79	0.40	0.98	0.43	1.03	0.78	0.80	0.94	0.75	0.61	0.60
Benzo (g,h,i) Perylene	<0.26	<0.27	<0.30	<0.37	<0.28	<0.30	<0.30	<0.37	<0.31	<0.28	<0.34	<0.32	<0.30	<0.30	<0.34	<0.28
Chrysene	<0.26	<0.27	<0.30	<0.37	<0.28	<0.30	<0.30	<0.37	<0.31	0.31	<0.34	<0.32	<0.30	<0.30	<0.34	<0.28
Dibenzo (a,h) Anthracene	<0.26	<0.27	<0.30	<0.37	<0.28	<0.30	<0.30	<0.37	<0.31	<0.28	<0.34	<0.32	<0.30	<0.30	<0.34	<0.28
Fluoranthene	0.74	0.94	1.23	1.35	0.67	0.80	0.71	0.69	0.66	0.92	1.23	0.92	1.28	1.03	0.73	0.85
Fluorene	0.42	0.72	<0.30	0.73	<0.28	<0.30	<0.30	<0.37	<0.31	<0.28	<0.34	<0.32	<0.30	<0.30	<0.34	<0.28
Indeno (1,2,3- cd) Pyrene	<0.26	<0.27	<0.30	<0.37	<0.28	<0.30	<0.30	<0.37	<0.31	<0.28	<0.34	<0.32	<0.30	<0.30	<0.34	<0.28
Naphthalene	0.53	0.89	<0.30	1.43	<0.28	<0.30	<0.30	<0.37	<0.31	<0.28	<0.34	0.40	<0.30	<0.30	<0.34	<0.28
Phenanthrene	0.50	0.63	<0.30	1.01	0.30	0.70	0.41	<0.37	<0.31	0.28	0.34	0.86	0.56	<0.30	<0.34	0.30
Pyrene	0.60	0.81	1.09	1.17	0.59	0.71	0.62	0.61	0.56	0.80	0.97	0.77	1.10	0.89	0.65	0.68
PAH Total (EPA 16)	5.24	6.30	5.21	9.59	<4.20	5.07	<4.50	<5.50	<4.65	5.25	5.19	5.63	5.82	4.67	<5.10	<4.20
PCB (mg kg⁻¹)																
PCB 28	<1.04	2.43	2.05	2.85	2.35	2.31	1.62	2.29	2.00	2.74	2.25	2.35	2.16	2.09	1.93	2.30
PCB 52	2.85	2.53	2.26	2.80	3.59	4.00	2.13	3.50	2.00	4.10	3.13	3.32	3.32	2.80	2.93	3.14
PCB 101	4.11	5.56	2.30	7.84	9.07	9.90	2.10	4.91	2.14	9.52	3.38	4.53	4.24	3.62	3.05	7.22
PCB 118	3.55	3.24	<1.20	3.41	5.12	6.06	<1.20	4.37	<1.24	6.10	<1.36	3.97	3.93	<1.20	<1.36	3.73
PCB 138	3.75	3.28	2.05	4.18	5.40	5.60	1.55	4.34	<1.24	5.99	2.47	3.93	3.71	2.66	2.64	3.57
PCB 153	4.22	5.94	<1.20	4.51	8.45	10.2	<1.20	3.12	<1.24	9.35	<1.36	<1.28	7.20	4.93	3.85	6.55
PCB 180	1.87	2.12	1.62	2.46	2.36	2.42	2.95	<1.48	1.29	1.72	1.41	2.35	2.36	1.80	<1.36	1.79

Table D.3 STWE: Analytical results (as reported)

	Sample Number															
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
pH	5.82	5.94	5.98	5.95	5.97	6.81	6.82	6.13	5.98	5.97	6.03	5.93	6.01	5.94	6.81	6.80
DS%	90.8	89.6	90.8	90.9	90.5	91.6	90.6	91.5	89.3	90.6	90.7	92.1	89.9	91.7	91.4	91.3
LOI%	76.1	75.8	76.3	76.4	76.2	75.8	76.0	75.8	75.8	76.1	76.3	75.7	75.8	76.3	75.7	75.7
PAH (mg kg⁻¹)																
Acenaphthene	<0.78	<1.60	<3.50	<1.70	<1.70	<0.28	<1.20	<1.70	<1.70	<1.70	<1.40	<0.93	<1.70	<1.20	<0.26	<0.64
Acenaphthylene	<0.78	<1.60	<3.50	<1.70	<1.70	<0.28	<1.20	<1.70	<1.70	<1.70	<1.40	<0.93	<1.70	<1.20	<0.26	<0.64
Anthracene	<0.78	<1.60	<3.50	<1.70	<1.70	0.63	<1.20	<1.70	<1.70	<1.70	<1.40	<0.93	<1.70	<1.20	0.29	<0.64
Benzo (a) Anthracene	<0.78	<1.60	<3.50	<1.70	<1.70	1.66	<1.20	<1.70	<1.70	<1.70	<1.40	<0.93	<1.70	<1.20	0.89	<0.64
Benzo (a) Pyrene	1.47	1.73	<3.50	<1.70	2.23	2.03	2.68	3.30	2.40	2.20	1.92	1.34	<1.70	1.84	0.96	2.08
Benzo (b&k) Fluoranthene	3.64	4.40	6.72	5.74	6.71	4.46	6.13	7.45	6.21	6.33	4.84	3.33	4.17	3.92	2.11	4.64
Benzo (g,h,i) Perylene	0.99	<1.60	<3.50	<1.70	1.74	1.23	1.73	2.28	1.79	<1.70	<1.40	<0.93	<1.70	<1.20	0.63	1.27
Chrysene	<0.78	<1.60	<3.50	<1.70	<1.70	1.68	<1.20	<1.70	<1.70	<1.70	<1.40	<0.93	<1.70	<1.20	0.80	1.73
Dibenzo (a,h) Anthracene	<0.78	<1.60	<3.50	<1.70	<1.70	0.45	<1.20	<1.70	<1.70	<1.70	<1.40	<0.93	<1.70	<1.20	<0.26	<0.64
Fluoranthene	3.43	4.78	4.83	5.52	4.68	3.83	5.79	8.16	5.82	6.36	5.18	3.49	4.16	4.45	1.92	4.54
Fluorene	<0.78	<1.60	<3.50	<1.70	<1.70	0.37	<1.20	<1.70	<1.70	<1.70	<1.40	<0.93	<1.70	<1.20	<0.26	<0.64
Indeno (1,2,3- cd) Pyrene	1.38	1.75	<3.50	<1.70	2.15	1.63	2.21	3.04	2.06	<1.70	1.67	1.18	<1.70	1.35	0.86	1.70
Naphthalene	<0.78	<1.60	<3.50	<1.70	<1.70	<0.28	<1.20	<1.70	<1.70	<1.70	<1.40	<0.93	<1.70	<1.20	<0.26	<0.64
Phenanthrene	2.02	2.76	<3.50	2.52	2.25	2.36	3.50	3.48	2.88	3.56	2.90	2.02	2.59	2.66	1.07	2.67
Pyrene	3.39	4.97	5.08	4.56	4.33	3.83	6.06	7.32	5.09	5.34	5.02	3.55	4.37	4.67	1.86	4.58
PAH Total (EPA 16)	19.4	27.6	<53.0	27.7	30.9	24.6	32.9	41.8	33.1	32.3	27.8	19.1	<26.0	24.3	12.0	25.5
PCB (mg kg⁻¹)																
PCB 28	<3.10	<6.40	<14.0	<6.60	<6.60	1.45	<4.60	<6.60	<6.80	<6.60	<5.40	<3.70	<6.80	<4.80	<1.00	<2.60
PCB 52	<3.10	<6.40	<14.0	<6.60	<6.60	<1.10	<4.60	<6.60	<6.80	<6.60	<5.40	<3.70	<6.80	<4.80	<1.00	<2.60
PCB 101	<3.10	<6.40	<14.0	<6.60	<6.60	1.42	<4.60	<6.60	<6.80	<6.60	<5.40	<3.70	<6.80	<4.80	<1.00	<2.60
PCB 118	<3.10	<6.40	<14.0	<6.60	<6.60	1.35	<4.60	<6.60	<6.80	<6.60	<5.40	<3.70	<6.80	<4.80	<1.00	<2.60
PCB 138	<3.10	<6.40	<14.0	<6.60	<6.60	1.39	<4.60	<6.60	<6.80	<6.60	<5.40	<3.70	<6.80	<4.80	<1.00	<2.60
PCB 153	<3.10	<6.40	<14.0	<6.60	<6.60	<1.10	<4.60	<6.60	<6.80	<6.60	<5.40	<3.70	<6.80	<4.80	<1.00	<2.60
PCB 180	<3.10	<6.40	<14.0	<6.60	<6.60	1.49	<4.60	<6.60	<6.80	<6.60	<5.40	<3.70	<6.80	<4.80	<1.00	<2.60

