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**HORIZONTAL STANDARDS ON ORGANIC MICRO-POLLUTANTS FOR IMPLEMENTATION OF  
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WP2-7: Research Report

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## **HORIZONTAL-ORG WP2**

### **Report: Comparison of 0-25cm soil sampling with 0-35cm sampling**

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## Summary

The task of the Sampling Work Package had been to investigate the differences in concentrations of organic micropollutants in sludge-amended soil in samples taken at 0-25 cm and 0-35 cm depth. However, earlier work showed that concentrations in most samples taken at 0-25 cm were close to, or below, the limit of quantification. It was decided that the original plan was not feasible and that the subject would be investigated with the use of simple models and results of experiments reported in the literature.

The first part of this report investigates why such low concentrations were found in soil samples using actual data collected for Project Horizontal and simple modelling. It is concluded that the likely cause was the dilution effect of mixing sludges (containing elevated levels of organic micropollutants) with soil (with very low levels).

A (non-exhaustive) search of the literature shows that metals and organic micropollutants tend to be retained in the mixing layer associated primarily with the sludge organic matter. There is evidence of losses by volatilization, degradation or leaching down the soil profile in solution or attached to soil colloids or dissolved organic carbon. However, the highest concentration is always found in the mixing layer.

Some exemplar situations are used to examine the effect of inappropriate sampling that doesn't take into account the mixing depth. Finally the results are discussed in the context of the wording of the Sludge Directive.

The following recommendations are made.

- In agricultural soils the sampling depth should be no greater than the mixing depth. In grassland soils the sampling depth should be no deeper than 10 cm.
- Samples for metals and organic micropollutants should be taken to the same depth since the highest concentrations of both are found in the mixing zone.
- The current Sludge Directive states that samples should be taken to a maximum 25 cm depth. This could be extended to 35 cm to reflect current practices in some Member States, where ploughing or sludge injection is deeper than 25 cm. This would not affect practices in those member States where ploughing is less deep.

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## 1 Introduction

### 1.1 General

The purpose of this work was to compare soil sampling at 0-25 cm, as specified in the Sludge Directive (86/278/EC), with soil sampling at 0-35 cm and to evaluate the implications for reported average concentrations of organic micropollutants and metals.

The Sludge Directive (86/278/EC) on the use of sewage sludge in agriculture (CEC, 1986) prescribes limit values for heavy metals in soils and sewage sludges and is the basis of current national legislation within the EU. This directive has been under review and the latest Working Document on Sludge (3rd draft, unpublished; CEC, 2000) proposed limit values for several groups of organic compounds in sludge: AOX (adsorbable organic halogens), LAS (linear alkylbenzene sulphonate), DEHP (diethylhexylphthalate), NPE (nonylphenol ethoxylates), PAH (polyaromatic hydrocarbons), PCB (polychlorinated biphenyls), and PCDD/PCDF (dioxins/ furans). Some countries already regulate organic pollutants in sludge, e.g. Denmark (Danish Ministry of Environment and Energy, 1996), France (JORF, 1997 & 1998), Germany (AbfKlärV, 1992), and Sweden (SNV, 1995 & 1996). Although the Working Document on Sludge did not propose that soil is analysed for these compounds, such analysis is specified in some national legislation, e.g. *Verordnung über Belastungen de Bodens* (Switzerland: VBBo, 1998) and *Bundesbodenschutz und Altlastenverordnung* (Germany: BBodSchV, 1999).

Current EC regulations prescribe sampling of soil prior to the first application of sludge (to establish base level concentrations) and thereafter at intervals. The Sludge Directive 86/278/EEC states: "The samples must be taken to a depth of 25 cm unless the depth of the surface soil is less than that value; however, the sampling depth in the latter case must not be less than 10 cm." The same wording was proposed in the Working Document on Sludge.

As stated in the Sludge Directive (CEC, 1986), the sampling depth depends on the depth of the surface soil. At the time the Directive was formulated maximum surface soil depth was typically 25 cm, i.e. the plough layer. However, farming practices have changed and it is not uncommon now to find ploughing to 35 cm in some member states. There are probably many reasons why plough depth has increased. These may include a reduction in nitrate leaching (Nortcliff & Amlinger, 2001), breaking of impermeable layers (plough pan), improved drainage, and better incorporation of organic matter. A change in the ploughing depth may alter the vertical distribution of pollutants (metals and organic compounds) applied to the soil in sludge or treated biowastes. Consequently, it might be appropriate to increase the maximum surface soil depth for sampling from 25 cm to 35 cm.

### 1.2 Evidence for change in plough depth

Nieder and Richter (2000) reported that plough depth had increased in intensively cultivated areas of West Germany between the 1960s, when typical plough depth was  $\leq 25$  cm, and the 1980s, when plough depth was typically 35 cm although, in other countries, e.g. UK, Denmark, France, Belgium, and Sweden, typical plough depths were 15-25 cm and, in East Germany, they rarely exceeded 25 cm.

Sleutel et al. (2006) resampled 116 Belgian soils, covering seven textural classes, to examine trends in soil organic carbon over a 50-year period. One of the parameters they recorded was plough layer depth, observed by inspecting auger samples for colour changes. This information had also been recorded in earlier surveys of the same soils. In 2003, plough depth ranged from 29.8 to 33.6 cm (mean 30.8 cm). This was not significantly different from plough depth measured in 1990 (mean 31.9 cm), but an increase of almost 10 cm over measurements made in 1952.

### 1.3 Aim

The original aim had been to sample soils at two depths (0-25 cm and 0-35 cm), analyse the samples for organic micropollutants and examine the results for correlation between paired samples. However, it was decided, based on previous work (Lambkin et al., 2007), that this was not feasible because, in that work, concentrations of organic micropollutants were near or below the limit of quantification in most soil samples that were analysed. Instead, the aim was revised such that it would be based on information found in published literature.

For this reason the aims were revised:

- (a) Perform some simple modelling to estimate concentrations of PAHs and PCBs in soil;
- (b) Summarize the results of published reports on vertical distributions of compounds in soil following sludge application.

To answer the questions:

- (a) Why did we find low concentrations in our soil samples?
- (b) Are metals and organic micropollutants distributed in the same way?
- (c) What would be the effect of extending the maximum sampling depth to 35 cm?
- (d) How should the sampling depth vary?

### 1.4 Scope

The scope of the investigation was:

1. To investigate why low concentrations of organic micropollutants were found in the soil samples collected by Project Horizontal.
2. To use the literature and simple modelling to investigate the likely differences between sampling to 25 cm and sampling to 35 cm.
3. To determine if metals and organic micropollutants should be sampled differently.

## 2 Concentrations of PAHs and PCBs in soil

### 2.1 The purpose of soil sampling

Agricultural soil is sampled prior to sludge application for two reasons. Firstly, to assess the current nutrient status so that the amount of sludge applied is sufficient to fulfil the nutrient requirement, i.e. the beneficial use. Secondly, soil samples are analysed to assess the risk of harm to the environment, crops and food chains, i.e. protection of public health and the environment; examples include leaching to groundwater (particularly nitrogen), uptake by plants, and ingestion by animals or humans (directly by soil ingestion or indirectly by consumption of plant-derived foods).

### 2.2 Sludge and biowaste application and incorporation

Application and incorporation methodologies depend on the type of biowaste (e.g. liquid, solid, semi-solid, granular), when the material is applied, and the equipment available to undertake the task (see Table 2.1).

**Table 2.1 Methods used for application and incorporation**

Operation	Typical Methods
<b>Primary cultivation</b>	Sub-soiler; Mouldboard plough; Heavy disc; Rigid tine cultivator; Powered cultivator; Chisel plough
<b>Secondary cultivation</b>	Cultivator; Harrow; Roller; Combination cultivator/ drills; Disk harrow; Direct drill
<b>Types of material</b>	Liquid sludge or digestate; Sludge cake; Thermally dried granular/ extruded pellets; Co-composted sewage sludge (+straw/ bark/ green waste); Composted biowaste
<b>Application</b>	Broadcast; Band spread; Shallow injection; Deep injection; Incorporation by ploughing/ cultivation

The extent of mixing depends in the type of soil and the tillage practice. Some cultivation methods do not invert the soil, e.g. mouldboard ploughing inverts the soil, but chisel ploughing doesn't. Consequently, mixing depths vary. Some examples of mixing depths are listed in Table 2.2.

**Table 2.2 Typical mixing depths for different incorporation methods**

Method	Depth / cm
Mouldboard ploughing	15-25
Shallow plough	10
Deep plough	15-30
Reduced cultivation	10-15
Minimum tillage	5-7
Shallow injection	5-8
Deep injection	12-30

When sludge or biowaste is added to agricultural soil the concentration of potential polluting elements/compounds will vary spatially and temporally at several scales. The factors affecting concentrations in soils include:

- (i) Initial concentration in the soil – spatially variable;
- (ii) Concentration in the sludge or treated biowaste that is applied – temporally and spatially variable;
- (iii) Application method – uniformity of application;
- (iv) Incorporation method – depth and uniformity of incorporation;
- (v) Dilution factor – a function of the application rate and mixing depth;
- (vi) Losses due to leaching in solution and colloid-facilitated transport through macropores;
- (vii) Physical, chemical and biological characteristics of the constituents – e.g. pH-dependent solubility and  $K_{OC}$ ;
- (viii) Losses due to degradation and volatilization;
- (ix) Soil organic matter – adsorption;
- (x) Bioturbation – mixing by soil fauna;
- (xi) Tillage operations and soil erosion.

The concentration of various constituents in the land-applied materials will vary between sources and between seasons. Variability within a material will also depend on the processing method, e.g. the degree of mixing during processing.

### **2.3 Why did we find low concentrations in our soil samples?**

Most potential pollutants (metals and organic micropollutants) are present in higher concentrations in biosolids than in agricultural soils. Therefore it is to be expected that repeated biosolids applications would increase the concentrations of these compounds in soil. Extensive research has demonstrated that most of them are quite strongly held by soil and they tend to remain where they are placed, although movement down the soil profile in leaching water and through bioturbation and losses from degradation and volatilization are possible. However, on balance, it would be expected that increases in soil concentrations would be expected within the depth of incorporation.

Consequently the factors of interest for estimating the concentration in soil at a given point is the dilution factor, which is a function of the application rate and mixing depth, i.e. the application and incorporation methods.

As stated in the introduction, the original aim had been to sample soils at two depths (0-25 cm and 0-35 cm), analyse the samples for organic micropollutants and examine the results for correlation between paired samples. However, it was decided, based on previous work (Lambkin et al., 2007), that this was not feasible because, in that work, concentrations of organic micropollutants were near or below the limit of quantification in most soils that were analysed.

Some exemplar calculations of concentrations of PAHs and PCBs are presented in Annex A. These are based on typical application rates, mixing to 25 cm depth and the highest concentrations found when analysing sludge materials collected for Project Horizontal. It is assumed that there are no losses by

degradation, volatilization or leaching and no mixing to depths below 25 cm either by agricultural activity or bioturbation.

From these calculations, it can be seen that the predicted concentrations in soil are between one and two orders of magnitude lower than reported Limits of Quantification (LOQ). The concentrations are based on a single application of sludge and demonstrate, even when losses by degradation, volatilization and leaching are not taken into account, that it might take in excess of 10, and up to 300, applications of sludge to reach concentrations that could be reported above the LOQ. This goes some way to explaining why such low concentrations of PAHs and PCBs were found in the soil samples collected for the Sampling Work Package (WP2).

### 3 Evidence from the literature

#### 3.1 Why sampling depth is important

Following application to land the contaminants in sludge are subject to various chemical or biological transformation processes, as well as physical transfer processes (i.e. leaching, run-off, erosion, and volatilization) that will influence their concentration in the soil. Schowanek et al. (2004) provides a number of calculations that could be used to assess concentrations in soil at any given time and under steady state conditions. For example, the steady state after multiple additions is given as:

$$PEC_{\text{soil-steady state}} = [F_{\text{air}} + F_{\text{sludge}}] / [k * H * R],$$

where

- PEC = predicted environmental concentration
- $F_{\text{air}}$  = chemical flux from air ( $\text{kg m}^{-2} \text{d}^{-1}$ )
- $F_{\text{sludge}}$  = chemical flux from sludge application ( $\text{kg m}^{-2} \text{d}^{-1}$ )
- $k$  = total removal rate (1/d) = sum of first order biodegradation, leaching and evaporation rate
- $H$  = soil depth (m)
- $R$  = soil density ( $\text{kg m}^{-3}$ )

So soil depth is important for estimating concentrations; if samples are taken too deep then PEC will be underestimated. On the other hand, if samples are not taken deep enough PEC could be underestimated, e.g. in the case of deep injection of sludge to, say 30 cm. As a consequence, if samples are not taken at an appropriate depth, judgements could be made on unrealistic concentrations.

In the absence of experimental data, a (non-exhaustive) search was made of the literature for experimental evidence of depth distribution of concentrations in soil following sludge application. Not all experiments were conducted under realistic conditions, i.e. chemicals were added to the soil dissolved in either water or methanol or in a spiked sludge matrix (e.g. McLaren et al., 2005) or not under field conditions, i.e. in columns or lysimeters (e.g. Jacobsen et al., 2004). Other experiments were directed at evaluation of losses by leaching rather than movement within the soil profile (e.g. Gove et al., 2001; Keller et al., 2002).

Frequently the incorporation or the ploughing and mixing depths are not stated in the literature, often because they are not known. For example, Stehouwer (2003) investigated trace element distribution with depth in 20 fields to which biosolids had been applied and 20 paired fields with no biosolids applied and compared concentrations at three depths: 0-10, 10-20, and 20-40 cm. They found that concentrations of Cu, Cr, Pb and Mo were greater in biosolids fields and this effect was observed at all depths, but the actual biosolids mixing depths for the fields were not known. Consequently they could not exclude deeper mixing as a cause for their observations, which could be the result of biosolids incorporation to below 20 cm or occasional deep tillage operations, even though the results could indicate movement of trace elements deeper into the profile due to the leaching action of percolating water.

#### 3.2 Evidence for movement of metals

There are many reports of investigations of the vertical distribution of metals in soils following biosolids application. In the main, the findings were that the location of the metals depended on the application and mixing method and the tillage system used following biosolids application. The following summaries provide examples of different tillage systems.

Davis et al. (1988) investigated 18 experimental plots at each of two sites that were planted with ryegrass and had received annual surface applications of liquid digested sludge over 3-4 years. Although increases in metal contents were found to a depth of 30 cm, the increases were only significant to a depth of 10 cm. It was concluded that the findings supported the UK Ministry of Agriculture recommendation of a sampling depth of 7.5 cm for long-term grassland with surface applications of sludge. They concluded that sample depths greater than 7.5 cm would include the sludge metal, but would not realistically represent true metal accumulation in the soil profile because the sludge effect is diluted by soil close to background levels further down the profile.

Yingming and Corey (1993) studied vertical movement of Cd, Cu and Zn in soil that had received a heavy sludge application incorporated into the 0-20 cm plough layer 11 years previously. Samples were collected at four depths (0-20 cm, 20-30 cm, 30-40 cm, and 40-50 cm). The results showed that, on average, 13% of the total metals applied had migrated below the incorporated layer and there was no significant enrichment below 30 cm. The highest metal concentrations were found in the plough layer. Although it was acknowledged that some enrichment of metals below 20 cm could be due to variations in plough depth, which was noted to be 21 or 22 cm in places, every effort was made to identify such occurrences and exclude plough soil from the 20-30 cm sampling depth. They concluded that that particulate transport through cracks and root channels along with ant and earthworm activity might move more metals than leaching, which probably played an insignificant part in metals movement.

Berti and Jacobs (1998) investigated vertical distribution of six metals in soil samples collected at 5 depths. The shallowest sample (0-15 cm) came from the plough layer, which was 23 cm deep. The second sample (15-30 cm) contained either equal amounts to or less than the first sample. No enrichment was found below 30 cm depth. It was concluded that metals were distributed in the profile by post-application ploughing and tillage, to depths of 20-25 cm.

Hoss et al. (2001) examined the vertical distribution of metals in a long-term experiment using three different tillage systems: conventional tillage (CT), characterised by topsoil turning; reduced non-inversion tillage (RT) by chisel ploughing with a wing share cultivator; no-tillage (NT). The soils were sampled at 0-3, 3-10 and 10-25 cm. Differences in the total metal content depended on the tillage system; the deeper the mixing depth, the deeper the increase in metal content.

Sukkariyah et al. (2005) investigated vertical distribution of metals in soils to which a single application biosolids had been incorporated to 14-16 cm depth and tilled annually to 15 cm with a rotavator. Analysis of soil cores, taken 17 years after biosolids application, showed that the biosolids-applied metals were still concentrated in the top 20 cm with minor enrichment down to 25 cm.

Similar results have been demonstrated for liquid sludge applications and in short-rotation energy forestry (willow). For example, Baveye et al. (1999) investigated metal distribution in soil profiles (0-75 cm) seven years after cessation of sludge application in the form of annual applications of liquid sludge over a period of 14 years. They found some enrichment of metals in deeper layers and estimated that there had been losses by leaching, but the highest concentrations were found in the two topmost layers analysed (0-15 cm and 15-30 cm). Hasselgren (1999) found that, in general, metals accumulated in the layer in which the sludge was applied and metal transportation below this point was not pronounced.

### **3.3 Evidence for movement of organic micropollutants**

In common with metals, the concentration of organic chemicals in soil resulting from land application of sludge will probably be lower in soils due to dilution, which is a function of the initial concentration in the sludge and soil, the rate of application and management practices, and losses through degradation, volatilization and leaching processes (Harrison et al., 2006). Adsorption to organic matter is an important factor in the immobilization of metals, but even more so for organic compounds. Several studies have demonstrated that organic micropollutants are strongly adsorbed to sewage sludge and remain strongly adsorbed to the sludge when it is mixed into soil. For example, Hesselsøe et al. (2001) found that mobility of NP from sludge aggregates to the surrounding soil is negligible.

Degradation and volatilization are important processes for some organic compounds. Harrison et al. (2006) estimated that a single application of sludge tilled into the soil would be diluted 100-fold and concentrations in the soil would increase with repeated applications only when losses were less than inputs from applied sludge. Concentrations would be higher in surface soils if the sludge was not tilled into the soil, for example in no-till operations and pasture application.

Vikelsøe et al. (2002a) demonstrated the effect of degradation and volatilization losses in their investigation of the geographical and vertical distribution of nonylphenols and phthalates in soil following sludge application. They concluded that sludge amendment below a certain limit did not lead to elevated levels of nonylphenols and phthalates in the soils, whereas heavy sludge amendment led to the accumulation of these contaminants, indicating that at low and medium sludge loads the biodegradation, elution or other removal mechanisms had been able to keep pace with the influx of phthalates and nonylphenols, preventing a build-up of high concentrations.

Vikelsøe et al. (2002b) studied the effect of sludge loading on the vertical distribution of a wide range of organic compounds (PAH, NPE, phthalates, PCB, PCN and PBDE). They found that following low sludge applications (0.7t DM ha<sup>-1</sup> yr<sup>-1</sup>) the compounds were found predominantly in the top 0-10 cm. Following high sludge applications the compounds had reached a depth of 40-50 cm. However, the authors cautioned that, at the high-sludge site, sludge had been applied over a period of 25 years (average 17t DM ha<sup>-1</sup> yr<sup>-1</sup>), a much greater rate than is normal in Danish agriculture and the location has effectively been used as a sludge-dumping site rather than for normal agricultural use.

The influence of long-term (9-17 years) tillage systems on the behaviour of PCBs in soils with different tillage practices was evaluated by Düring et al. (2002) (see also Düring & Gäth, 2002). Tillage intensity was considered to decrease in the following sequence: conventional plough tillage (CT: mouldboard plough to 25 cm followed by rotary harrow to 10 cm), reduced tillage (RT: chisel plough to 25 cm), and no-till (NT: direct seeding). Soil samples were collected at three depths (0-3, 3-10 and 10-25 cm). It was found that in ploughed soils the mixing effect resulted in homogeneous distribution of PCBs within a soil depth of 25 cm whereas in no-till soils most of the PCBs remained in the top 3 cm. The authors cautioned against transferring a soil quality criterion which refers to deeper soil sampling depths (e.g. 0-25 cm) to non-tilled soils since this would underestimate the PCB concentration.

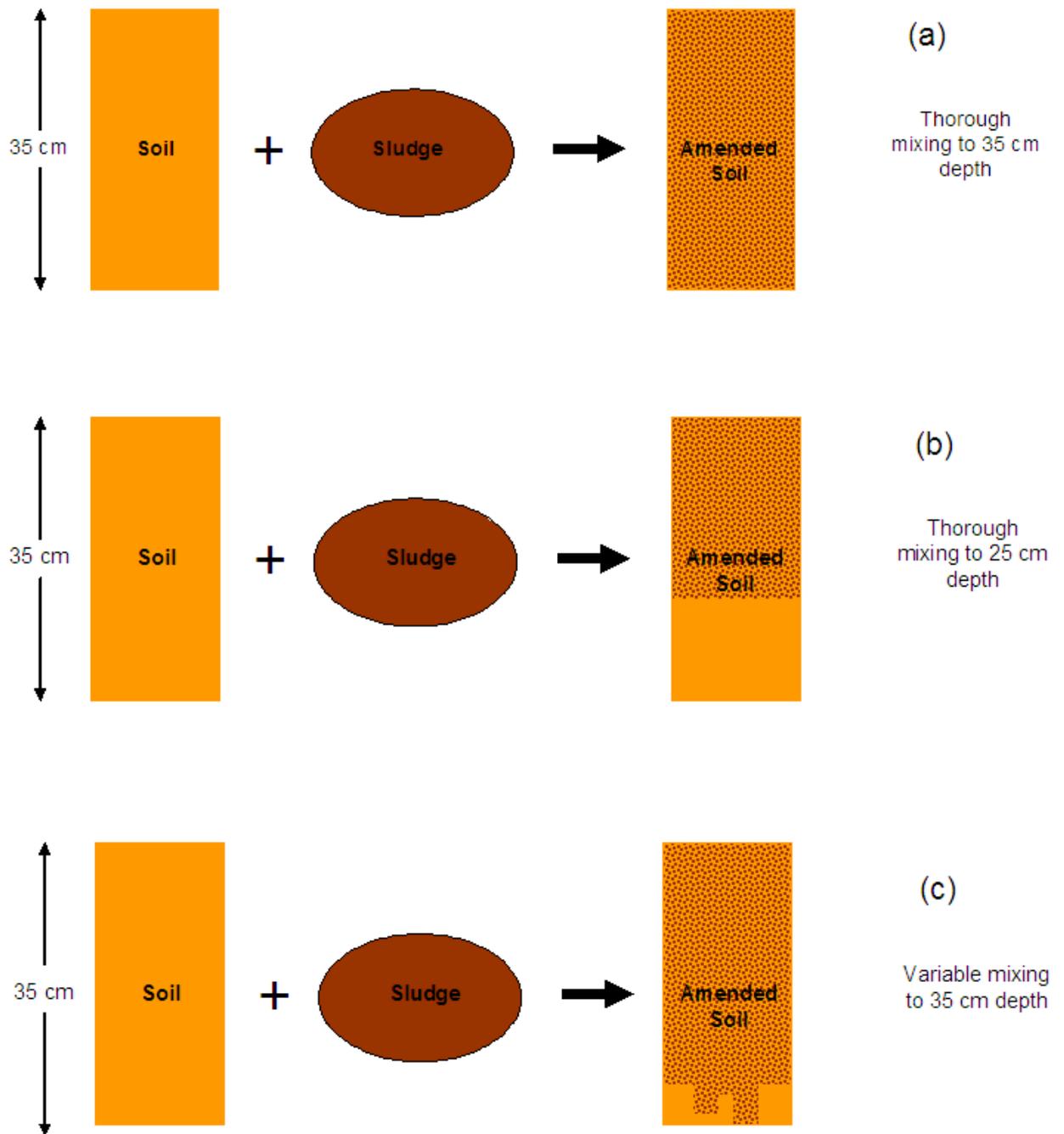
It has also been demonstrated (Jacobsen et al., 2004) that linear alkylbenzene sulfonate (LAS) and nonylphenol (NP) contained in sewage sludge applied to soil are retained in the mixing layer (15 cm) with no measurable losses by leaching, but significant losses by degradation. This experiment was carried out in lysimeters (outdoors) and the soil and sludge were well mixed prior to packing in the lysimeter. The authors recognized that conditions might be different under field conditions where degradation, an aerobic process, may be slower due to low oxygen content in larger sludge aggregates and leaching may happen as colloid-facilitated transport through macropores. They also recognized that it is possible that surfactants are more mobile in soil during autumn and winter, with low temperatures, no or little plant growth, and downward water transport.

The degree of mixing as well as the depth of mixing plays an important part in the degradation and mobility of some organic compounds. For example, Hesselsøe et al. (2001) demonstrated that degradation of NP, which is enhanced under aerobic conditions, increases as the size of sludge aggregates decreases. Sludge aggregates are broken up by mechanical mixing (e.g. ploughing) and bioturbation.

### **3.4 The effect of extending the sampling depth to 35 cm**

The effect of extending the sampling depth to 35 cm will depend on the depth to which the sludge is incorporated into the soil. Figure 1 illustrates three examples.

In Fig 1 (a) the sludge is thoroughly mixed to a depth of 35 cm, as might happen in Western Germany (see 1.2). In this case sampling to 25 or 35 cm would be expected to produce the same result.



**Figure 1** Effect of different mixing

In Fig 1(b) the sludge is thoroughly mixed to a depth of 25 cm, as might happen in some other Member States. In this case sampling to 35 cm would produce lower concentrations than sampling to 25 cm because the mixing layer is diluted by lower background concentrations below 25 cm.

The effect of variable mixing depth is illustrated in Fig 1(c). This example demonstrates the effect of spatial variation in plough depth, which has been reported, for example, by Yingming and Corey (1993). In this case sampling to 35 cm will produce a lower result than sampling to, say, 25 or 30 cm due to the dilution effect in the lower few cms.

These simple examples demonstrate that to produce precautionary results (i.e. the worst case) the practical sampling depth should be appropriate to the theoretical mixing depth, i.e. 25 or 35 cm, and should take into account spatial variation in mixing depth, i.e. sample to 20 or 30 cm.

### 3.5 How should the sampling depth vary?

In the examples cited from the literature, the highest concentrations of metals and organic micropollutants were found in the mixing zone. Therefore, if precautionary assessment of the metals content in soil is to be made, it is important to sample within that zone. Samples taken that include soil below the mixing zone will include soil with lower metals content and underestimate the true concentration.

The effect of taking samples to inappropriate depths and not taking into account spatial variability in mixing was illustrated in 3.4.

This is reiterated by Sastre et al. (2001) who concluded that:

- (i) A unique, common sampling strategy cannot be followed for different land uses, which may have a greater influence than soil type.
- (ii) The type of pollutant is another factor to consider in sampling design, although in most cases sampling strategy should be designed for mixed contaminated scenarios, or for pollutants with different interactions with the soil, as is the case of mobile/ non-mobile trace elements.
- (iii) In arable lands depth is not a crucial factor, as long as the sample belongs to the ploughed layer (usually down to 20-25 cm).
- (iv) Finally, when dealing with semi-natural, undisturbed systems (grasslands), the sampling depth must account for vertical variability. Samples of 0-10 cm depth are recommended since they prevent underestimation due to dilution effects when sampling deeper layers.

## 4 Conclusions

The measured concentrations of organic micropollutants found in agricultural soils investigated by the Horizontal Sampling Work Package (WP2) were close to or below the limit of quantification. Using typical application rates and mixing to 25 cm depth it was shown that the likely cause of such low concentrations was dilution. The likelihood is that samples taken to 35 cm depth from the same sites would have been even more diluted and no organic micropollutants would have been detectable.

The evidence from the literature shows that the highest concentrations of metals and organic micropollutants are to be found in the mixing zone, with some movement down the profile caused by leaching (in solution or adsorbed to colloids) or bioturbation. High concentrations were only found below the mixing zone when exceptionally high amounts of sludge (17 t DM ha<sup>-1</sup> yr<sup>-1</sup>) had been applied over a long time (25 years) to land that was regarded as a sludge-dumping site rather than for normal agricultural use.

Using some exemplar situations it was shown that to produce precautionary results (i.e. the worst case) the practical sampling depth should be appropriate to the theoretical mixing depth, i.e. 25 or 35 cm, and should take into account spatial variation in mixing depth, i.e. sample to 20 or 30 cm.

Since the sampling depth should reflect the mixing depth, it should differ depending on the land use and the management method. In arable soils the sample should be taken from within the plough layer; in undisturbed systems (grasslands) they should be taken within the top 10 cm.

The Sludge Directive 86/278/EEC states: "The samples must be taken to a depth of 25 cm unless the depth of the surface soil is less than that value; however, the sampling depth in the latter case must not be less than 10 cm."

Based on the results of this work there is no real benefit to increasing the maximum sampling depth to 35 cm, except that this would better reflect modern farming practices in some Member States.

Increasing the maximum sampling depth to 35 cm would not affect areas where soils are ploughed to shallower depths because the current wording is "...25 cm unless the depth of the surface soil is less than that value...".

## **5 Recommendations**

- In agricultural soils the sampling depth should be no greater than the mixing depth. In grassland soils the sampling depth should be no deeper than 10 cm.
- Samples for metals and organic micropollutants should be taken to the same depth since the highest concentrations of both are found in the mixing zone.
- The current Sludge Directive states that samples should be taken to a maximum 25 cm depth. This could be extended to 35 cm to reflect current practices in some Member States, where ploughing or sludge injection is deeper than 25 cm. This would not affect practices in those member States where ploughing is less deep.

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## Annex A

### Predicted concentrations of PAH and PCB in soil

#### A.1 Dilution factors in soil

In the following sections typical values for application rates and soil mixing depth are used to estimate the concentrations in soil of exemplar PAH and PCB compounds after one application of sewage sludge, assuming thorough mixing in the top 25 cm.

##### A.1.1 Example 1: Liquid sludge

**Sludge:** Application rate:  $150 \text{ m}^3 \text{ ha}^{-1}$  (Typical rate; information from UK Water Utility company).

Dry solids: 2% dry solids, equivalent to an application rate of  $3 \text{ m}^3 \text{ ha}^{-1}$  DM.

**Soil:**  $1 \text{ ha} = 10,000 \text{ m}^2$ ; Assume a mixing depth of 25cm

Soil volume =  $2500 \text{ m}^3 \text{ ha}^{-1}$

**Dilution factor:**

Dilution is 3 parts sludge to 2500 parts soil (1:833).

##### A.1.2 Example 2: Dewatered sludge

**Sludge:** Application rate:  $8\text{-}12 \text{ Mg ha}^{-1}$  (Rate used on Spain Field 1; Lambkin et al., 2007).

**Soil:**  $1 \text{ ha} = 10,000 \text{ m}^2$ ; Assume a mixing depth of 25cm;

Soil volume =  $2500 \text{ m}^3 \text{ ha}^{-1}$ ; Assume soil density =  $1300 \text{ kg m}^{-3}$  ( $1.3 \text{ g/cm}^3$ )

Soil mass =  $3250 \text{ Mg ha}^{-1}$

**Dilution factor:**

Dilution is 8-12 parts sludge to 3250 parts soil (1:271 – 1:406).

##### A.1.3 Predicted concentrations of PAH and PCB in soil

Based on the dilution factors calculated in Examples 1 and 2, the concentration in soil was calculated for the highest PAH and PCB concentrations found in sewage sludge samples analysed by project Horizontal (see Table A.1).

Even for these compounds, which might be taken to represent a worst-case scenario, the concentrations are between one and two orders of magnitude lower than the limits of quantification quoted by the commercial laboratory that analysed the Sampling materials ( $1.1\text{-}1.3 \text{ } \mu\text{g kg}^{-1}$  for PCB compounds and  $0.25\text{-}0.35 \text{ mg kg}^{-1}$  for PAH compounds) (Lambkin et al., 2007).

**Table A.1** Effect of dilution factors on sludge applied to soil

	Sludge $\mu\text{g kg}^{-1}$ DM	Soil $\mu\text{g kg}^{-1}$ DM based on Liquid Sludge :Soil (3:2500)	Soil $\mu\text{g kg}^{-1}$ DM based on Dewatered Sludge :Soil (8-12:3250)	Analytical LOQ <sup>(2)</sup>
Fluoranthene <sup>(1)</sup>	2431	2.9	6.0-9.0	250-350 $\mu\text{g kg}^{-1}$
Benzo(b&k)fluoranthene <sup>(2)</sup>	8160	9.8	20.1-30.1	250-350 $\mu\text{g kg}^{-1}$
PCB 52 <sup>(1)</sup>	57	0.068	0.140-0.210	1.1-1.3 $\mu\text{g kg}^{-1}$
PCB 153 <sup>(2)</sup>	10.2	0.012	0.026-0.038	1.1-1.3 $\mu\text{g kg}^{-1}$

(1) Validation materials (Gawlik, 2007)

(2) Sampling materials (Lambkin et al., 2007)