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# Data sheet

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Abstract: The topic is risk assessment of sludge supply to agricultural soil in relation to xenobiotics. A large variety of xenobiotics arrive to the wastewater treatment plant in the wastewater. Many of these components are hydrophobic and thus will accumulate in the sludge solids and are removed from the plant effluent. The focus in this work is the top soil as this layer is important for the fate of a xenobiotic substance due to the high biological activity. A simple model for the top soil is used where the substance is assumed homogeneously distributed as suggested in the European Union System for the Evaluation of Substances (EUSES). It is shown how the fraction of substance mass, which is leached, from the top soil is a simple function of the ratio between the degradation half lifetime and the adsorption coefficient. This model can be used in probabilistic risk assessment of agricultural soils and different examples are shown using a Bayesian type of modelling approach.

Keywords: Mathematical modelling, sludge, soil, probabilistic risk assessment, xenobiotics

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# Contents

# Summary

*Nutrient reading*

Sludge amendment of agricultural soil is a obvious way of nutrient recycling and furthermore a cheap way of waste management. However, aspects need to be considered regarding the content of xenobiotics in the sludge. A large variety of cenobiotics arrives to the wastewater treatment plant in the wastewater. Many of these components are hydrophobic and thus will accumulate in the sludge solids and be removed from the plant effluent. This may yield a sufficient cleaning of the effluent and a good water quality, but the resulting accumulation in the sludge many cause problems in the sludge disposal. The agricultural re-use of the sludge nutrients needs to be associated with a risk assessment concerning the accumulated xenobiotic substances.

*Risk assesment*

*Top soil model*

Exposure assessment for the soil compartment (root zone) is important in relation to terrestrial organisms and human consumption of agricultural products originating from sludge amended soils. The top soil in form of the ploughing zone represents the upper 20 cm layer and is the most biologically active zone, because crop detritus and sludge is introduced directly within the soil. The biological activity wills typical decrease rapidly with depth under the top soil layer and the total root zone depth for agricultural crops is typical about one meter. The focus in this work is the top soil as this layer is important for the fate of a xenobiotic substance due to the high biological activity. Furthermore, hydrophobic substances will tend to accumulate in the layer in which it is supplied by ploughing and the top layer will thus be a good indicator for soil accumulation and the potential for leaching (ground water contamination). The local Predicted Environmental Concentration (PEC) level for the soil compartment as calculated in the European Union System for the Evaluation of Substances (EUSES) is likewise related to the upper 20 cm layer of top soil *(EC, 1996)*.

*EUSES*

A simple model for the top soil is used where the substance is assumed homogeneously distributed as suggested in the European Union System for the Evaluation of Substances (EUSES). It is shown how the fraction of substance mass, which is leached, from the top soil is a simple function of the ratio between the degradation half lifetime and the adsorption coefficient. When the main part of the substance mass is degraded in the top soil the following relationship is valid:

*Leached fraction*

$$X_L \approx \frac{q}{\ln(2) \cdot \rho \cdot x_1} \cdot \frac{T_{½}}{K_d}$$

where $X_L$ is the mass ratio between the degraded and the leached mass ($X_L$=0 means no leaching), $q$ is the mean flux of water leaching from the top soil (cm/d), $\rho$ is the dry bulk density (kg/dm$^3$), $x_1$ is the top soil depth (cm), $T_{½}$ is the degradation half lifetime for the substance in the top soil (d), and $K_d$ is the adsorption coefficient (l/kg). Using numeric values representative for Danish condition this equation predict the leached mass ratio to be about 0.3% of the numeric value for the $T_{½} / K_d$ ratio.

*Completely mixed top soil*

As illustrated, the model becomes rather simple and easy to use when the top soil is considered to be homogenous (completely mixed conditions), however, a stratification may be formed in the layer due to penetration of water. Such stratification may violate the validity of the assumed homogenous concentration distribution. This problem is analysed and the conclusion is that the completely mixed condition is a valid assumption when the retention factor ($R$) in the top soil is in the magnitude for 50 or higher, when the ploughings takes place once a year. The retention factor is approximately related to the adsorption coefficient ($K_d$) as $R=\rho\cdot K_d$, where the numeric value for $\rho$ is about 1.5 kg/dm$^3$. Thus, the completely mixed condition in the top soil is valid for both slightly and highly hydrophobic substances.

*Model application*

The simple top soil model is applied in an analysis of a field scale measurements of DEHP contamination in sludge amended agricultural soil *(Vikelsøe et al., 2000)*. The intensity of the actual sludge amendment (load) is calculated based on the input parameters: (1) The number of person equivalent (pe) connected to the wastewater treatment plant; (2) The sludge production per pe; (3) The concentration of DEHP in the sludge *(Vikelsøe et al., 2000)*. This calculation is subject to some uncertainty and the value of this is estimated using considerations about the uncertainty in each input parameter yielding a log-normal distribution for possible sludge load values. The measured concentration values are also associated with some variability and a normal distribution is estimated based on the recorded measurements.

The top soil model can relate the sludge load to a resulting soil concentration value which is compared to the measured concentrations by using a first order disappearance coefficient ($K_{dis}$). The calculated concentration values can be compared to the measurements and thus it is possible to adjust the $K_{dis}$ value in order to obtain the best agreement between the model calculations and the measurements. However, this calibration is not straightforward because both the results from the top soil model and the measurements are distribution functions and not fixed values. The top soil model result is a distribution function because the sludge load value enters the model as a distribution function. Thus, an inverse modelling method is developed based on maximum likelihood to calculate the probability distribution of the $K_{dis}$ values, which yields the best coincidence between the model and the measurements. The result is a close to normal distribution for the $K_{dis}$ values and the interval for about 95% of the probability is identified to be a half lifetime from 7 to 38 years.

*Probalistic risk assessment*

This model can be used in probabilistic risk assessment of agricultural soils and different examples are shown. The results are strongly related to the experimental values which have be used to calibrate the $K_{dis}$ distribution function. Thus, the shown use of the model is mainly a numerical example of probabilistic risk assessment. Based on the soil quality criterion, the maximum allowable soil concentration level is identified and is a key parameter in the risk assessment. The recovery time period needed for a polluted soil to recover after contamination, in which the soil concentration decreases below the soil quality criterion is identified as a distribution function. It is shown how missing information about the soil concentration level at the beginning of the recovery time period can lead to much longer estimated time periods if the prediction needs to be safe (low probability for under estimation of the recovery time period). The maximum period of sludge amendment is calculated for three different load intensities. The results is compared to the critical load where the soil quality criteria is fulfilled at infinite time (the disappearance sufficient to remove the substance from the top soil at the same rate as the load). It is shown that this critical load value in this case is much lower than the tolerable load for a long time period (up to 48 years).

# 1 Introduction

*Agrucultural use of sludge*

*Risk assessment*

*Selection of simple top soil model*

A large amount of sludge is produced from the wastewater treatment facilities and sludge disposal costs occupy a major part of the total costs for the wastewater treatment. One attractive solution of disposal is agricultural soil amendment using the sludge, due to low cost and reuse of the nutrients in the sludge. Nevertheless, the reuse of sludge involves a risk for contamination of the soil system by undesirable contaminants, which may accumulate in the soil as a result of repeating supplies. Thus risk assessment appears necessary. This risk assessment needs methods to predict soil concentration levels and mathematical models seem attractive in this context. A large number of soil contaminant models have been suggested during the last 20 years and many of them are highly complicated deterministic models e.g. the Macro model *(Jarvis, 1994)* and the PESTLA model *(Boesten and van der Linden, 1991)* both being used in simulation of pesticide leaching. However, the risk assessment will typically operate without the detailed information needed for the complicated models. The natural variability and uncertainty due to lack of knowledge of the model parameters can easily overshadow the improvements of the modelling result obtained by replacing a simple model by a more complicated one. The model in the presented analysis is similar to the simple local soil compartment model suggested in EUSES *(Vermeire et al., 1997 and EUSES, 1997)* as a one box model for the upper 20 cm of top soil. However, the calculations as suggested by *EUSES (1997)* are static number calculations without explicit to take the great uncertainty related to the model parameters into account. The model uncertainty is a critical part of the risk analysis, hence, a stochastic modelling approach, reflecting the uncertainty will be an significant improvement of the risk assessment methodology. The precautionary principle in environmental management, where harmlessness has to be proven at a specific level of significance, taking the uncertainties into account, can only be possible by using a stochastic approach.

*Stochastic approach*

In the area of pesticide leaching, efforts have been made to develop stochastic model methodologies for assessment of the degree of leaching *(Loague et al., 1989; Jury and Gruber, 1989; Van Der Zee and Boesten, 1991; Petac et al., 1991; Loague, 1991; Loague et al., 1996; Di and Aylmore, 1997)*. The pesticides in all these investigation are assumed to be spread on the soil surface, while the sludge in this investigation is assumed to be dispersed in the upper 20 cm of the soil matrix by ploughing. The pesticide models assumes different kind of probability density distribution function (often as normal distribution functions) for the input parameters, and calculates the resulting distribution function of the model result. In contrast to this the present investigation calculates the distribution function for the model parameters based on a inverse modelling technique, where results from a field investigation *(Vikelsøe et al., 2000)* are used.

*Field measurement*

Sludge was continuously loaded to the agricultural soil during a period of 25 years. Soil samples in different depths are taken and analysed for DEHP 6 and 8 years after the end of the 25 years period of sludge load, respectively. The results are interpreted with respect to the upper 20 cm layer using the one box soil model. The supply of DEHP during 25 years is obviously rather difficult to quantify. Thus, a probability distribution function has been identified including the estimated uncertainty. The load and the measured concentration levels are used to determine the field disappearance coefficient (including degradation and wash out) for the upper 20 cm layer. The uncertainties related to as well the estimation of load as the mean soil concentration are included in the analysis by using a stochastic modelling approach. Finally, the developed model will be used in different aspects of risk assessment.

# 2 Model formulation and discussion of complexity level

The model system is illustrated in Figure 2.1.

Top soil thickness $x_1$: 20 cm

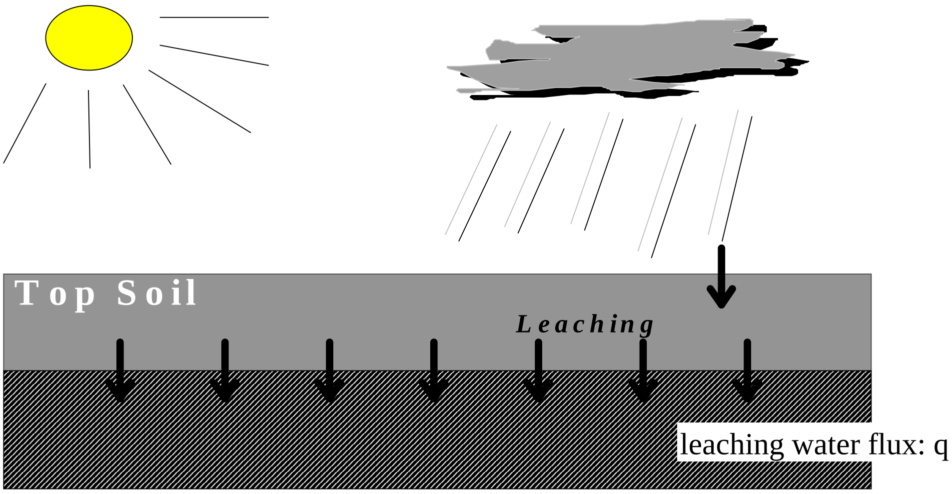


***Figure 2.1*** *Model system.*

*Homogen distribution in top soil*

The sewage sludge is assumed equally distributed on the field and subsequently the field is ploughed. This results in a homogeneous plough layer (20 cm top soil) with the applied substances being assumed homogeneously distributed in the layer immediately after the application. Occasionally rain will lead to downwards water flow through the soil and may bring part of the substances present with it. The remaining part of the substances is either degraded or adsorbed to the soil particles.

## 2.1 Basic assumptions

*Assumptions*

The overall assumptions for the models involved in the analysis are:

1. No macro pore flow in the top soil (preferential flow)
2. Sludge and soil homogeneously mixed in the top soil, i.e. no influence from sludge lumps in the soil.
3. Constant soil humidity (field capacity, 30% volume)
4. Homogeneous soil, i.e. degradation and adsorption independent of depth
5. First order degradation assuming constant rate coefficient
6. Linear and reversible adsorption

*Macro pore and sludge lumps*

The single assumptions can not be considered separately. E.g., if a significant part of the water is by passed through the macro pores (assumption No. 1 invalid) the transmission of substance from the sludge, included in the soil matrix, into the macro pore water may still be limited in case of well-mixed conditions (Assumption No. 2). This can lead to decreased leaching due to macro pores. On the other hand, in cases where both macro pores and sludge lumps (hot spots) prevail the situation may be totally different. In these cases erosion is possible directly from sludge lumps into the macro pores.

*Soil humidity*

The soil humidity is assumed constant at a field capacity, which correspond to approximately pF=2 (*Marshall et al., 1996*, p. 250. Using the pF=2 value, soil data from *Jacobsen, (1989)* gives a field capacity in the interval of 25-30%. If nothing is mentioned a value of 30% is used. In the summer time the humidity will typically only be at field capacity shortly after rain fall and be below field capacity most of the time. The flux of leaching water ($q$) will be limited in the summer season and most water will leave the root zone during the winter season.

*Homogene top soil*

The soil is assumed homogenous independent of depth, which seems to be a valid assumption in the top soil layer where plowing takes place yearly. In the deeper soil layer, i.e., below approximately 20 cm no mixing takes place and stratification may exist. The organic matter content will typically be higher in the top soil compared to the layers below which may influence the adsorption and degradation characteristics significantly. However, the models operate only in the upper layers but not in deeper layers. Thus, the soil layers involved in the present study are considered as homogenous having a dry bulk density $\rho$ of 1.5 kg/dm$^3$. In general the value of the bulk density is subject to rather low uncertainty compared to the other model parameters.

*First order degradation*

The degradation is assumed to be a first order process and the coefficient is assumed constant. Nevertheless, the degradation is influenced by soil temperature and humidity and other less predictable mechanisms such as micro organism adaptation, organic matter type and content etc. Since only limited data are available for most substances, it appears problematic to use more sophisticated degradation models.

*Linear and equilibrium adsorption*

The adsorption is assumed linear and reversible, i.e. system equilibrium, which is a simplification in relation to a soil column system, where local transport mechanisms in the porous structure will tend to introduce a kinetic term of adsorption. Furthermore, the adsorption may not be at equilibrium locally, especially for higher concentrations where micro scale concentration clusters may be formed in the pore water. However, the availability of more data seems critical both in relation to the soil and the substance in order to include such complicated processes in the system description.

## 2.2 Model equations

**One single instantaneous sludge amendment**

*Instant sludge supply*

The governing differential equation is derived using a mass balance for the top soil:

$$\frac{dC_{tot}}{dt} = -\left(\frac{q}{x_1} + k_1\right) \cdot C_{diss} \qquad 2.1$$

where $C_{tot}$ is the total volumetric concentration of substance ($\mu$g/l), $q$ is the mean flux of water leaving the top soil (leaching), $k_1$ is the first order degradation coefficient (1/d), $x_1$ is the top soil layer thickens (m) and $C_{diss}$ is the dissolved concentration of substance ($\mu$g/l). The total concentration is related to the dissolved concentration as

$$C_{tot} = (\theta + \rho \cdot K_d) \cdot C_{diss} \equiv R \cdot C_{diss} \qquad 2.2$$

where $\theta$ is the volume fraction of water (0.3), $\rho$ is the dry bulk density of the soil (1.5 kg/dm$^3$), $K_d$ is the reversible linear adsorption coefficient (l/kg) and $R$ is defined as the retention factor. It shall be noticed that the unit for $C_{tot}$ is mass per volume soil, so the $R$ factor become equal to the porosity ($\theta$) in case of no adsorption ($K_d$=0). Combining the Eqs. 2.1 and 2.2 the following relationship comes out:

*Governing differential equation*

$$\frac{dC_{tot}}{dt} = -\left(\frac{q}{x_1 \cdot R} + k'\right) \cdot C_{tot} \qquad 2.3a$$

$$\frac{dC_{diss}}{dt} = -\left(\frac{q}{x_1 \cdot R} + k'\right) \cdot C_{diss} \qquad 2.3b$$

where k' is the overall degradation rate ($k' = \frac{k_1}{R}$) and related to the total degradation half lifetime ($T_{½}$ in days) as $\frac{\ln 2}{T_{½}}$.

The solution of Eqs. 2.3a and 2.3b assuming constant coefficients ($q$, $R$ and $k'$) and in case of a single instantaneously sludge supply is an exponential function as

*Solution*

$$C = C_o \cdot e^{-\left(\frac{q}{R \cdot x_1} + k'\right)t} = C_o \cdot e^{-K_{dis}t} \qquad 2.4$$

where $C_o$ is either the initial total or the initial dissolved concentration (µg/l) and where the definition a total disappearance coefficient for the top soil is defined as: $K_{dis} \equiv \frac{q}{x_1 \cdot R} + k'$. The total mass leaching for the top soil due to a single sludge amendment is the integral of Eq. 2.4 for $C_{diss}$ multiplied by $q$ as

$$mass_{leach} = \int_0^\infty q \cdot C_{diss} \cdot dt = \frac{q \cdot C_{diss}}{\frac{q}{x_1 R} + k'} \qquad 2.5$$

**Continuously sludge load**

*Constant rate sludge supply*

If the sludge is repeatedly ploughed into the top soil in relatively short time intervals compared to the disappearance time for the substance in the top soil it will be possible to consider the sludge supply as a continuously load to the soil. During the period of sludge amendment a mass balance for a unit field area can be formulated, assuming a constant sludge supply and a first order degradation:

*Governing differential equation*

$$\frac{dC_{tot}}{dt} = \frac{L}{x_1} - \frac{q + k_1}{x_1} \cdot C_{diss} \qquad 2.6$$

where *L* is the load of substance per unit time due to sludge amendment (mg/(m$^2$·d). The solution of this equation including Eq. 2.2 assuming constant *L*, *q*, *k’* and *R* for the initial concentration (before start of loading) to be zero is

*Solution*

$$C_{tot} = \frac{L}{(\frac{q}{x_1 \cdot R} + k')} \cdot \left(1 - e^{-(\frac{q}{x_1 \cdot R} + k') \cdot t}\right) = \frac{L}{K_{dis}} \cdot \left(1 - e^{-K_{diss} \cdot t}\right) \qquad 2.7$$

A long time period of sludge supply compared to the disappearance time results in a steady state concentration ($C_{tot\infty}$) using Eq. 2.7 for $t \rightarrow \infty$ as:

$$C_{tot\infty} = \frac{L}{\frac{q}{x_1 \cdot R} + k'} = \frac{L}{K_{dis}},\ for\ t \rightarrow \infty \qquad 2.8$$

The downward mass flux of substance (µg/(m$^2$·s)) in case of a long-term sludge supply can be calculated using Eq. 2.8 as

$$Flux_{leach} = q \cdot C_{diss\infty} = q \cdot \frac{C_{tot\infty}}{R} = \frac{L \cdot q}{\left(q + R \cdot x_1 \cdot k'\right)} \approx \frac{L \cdot q}{q + x_1 \cdot K_d \cdot \rho \cdot k'} \qquad 2.9$$

where the assumption: $\theta << K_d \cdot \rho$ is used (see Eq. 2.2).

### Termination of a continuous sludge supply period

*End of constant rate sludge supply periode*

During the period after termination of the sludge supply it is assumed that the concentration decreases following a simple 1. order disappearance:

$$C_{tot} = \frac{L}{x_1 \cdot (\frac{q}{R} + k')} \cdot \left(1 - e^{-(\frac{q}{R} + k') \cdot t_{load}}\right) \cdot e^{-(\frac{q}{R} + k') \cdot t} = \frac{L}{x_1 \cdot K_{dis}} \cdot \left(1 - e^{-K_{dis} \cdot t_{load}}\right) \cdot e^{-K_{dis} \cdot t} \qquad 2.10$$

where the initial concentration is equal the final concentration during the sludge supply period (Using Eq. 2.7) and where $t_{load}$ is the time period for the load to take place and *t* is the time period after termination of loading.

### Fraction of leaching

*Leaching*

The ratio between the total amount of substance supplied and the amount of substance leached from the plough layer, $X_L$, can be calculated for a single sludge amendment as

$$X_L = \frac{mass_{leach}}{x_1 \cdot R \cdot C_{diss,o}} = \frac{q}{q + k' \cdot R \cdot x_1} \approx \frac{q}{q + k' \cdot K_d \cdot \rho \cdot x_1} \qquad 2.11$$

where the assumption: $\theta << K_d \cdot \rho$ and Eq. 2.5 is used. An equivalent relationship can be derived in case of continuously sludge supply as the ratio between the flux of substance entering the soil by sludge supply (The load $L$) and the flux of substance released from the top soil by leaching ($Flux_{leach}$). This can be seen using Eq. 2.9 as:

$$\frac{Flux_{leach}}{L} \approx \frac{L \cdot q}{L \cdot (q + x_1 \cdot K_d \cdot \rho \cdot k')} = \frac{q}{q + x_1 \cdot K_d \cdot \rho \cdot k'} \qquad 2.12$$

The Eqs. 2.11 and 2.12 are seen to be equal.

If the degradation is insignificant ($q << x_1 \cdot K_d \cdot \rho \cdot k'$) the ratio $X_L$ approach one i.e. all the substance supplied is leached. The equation can be rewritten using the relationship for $T_{½}$:

*Fractions leached*

$$\frac{T_{½}}{K_d} = \frac{\ln(2) \cdot \quad \cdot x_1}{q} \cdot \frac{X_L}{1 - X_L} \qquad 2.13$$

Eq. 2.13 predicts a unique combination of $K_d$ and $T_{½}$ to be related to specific fractions of leached material. The units for $K_d$ and $T_{½}$ are l/kg and days, respectively. The equation is visualized in Fig. 2.1 applying $q$=0.06 cm/d (20 cm/year), $\rho$ =1.5 kg/dm$^3$ and $x_1$ =20 cm.

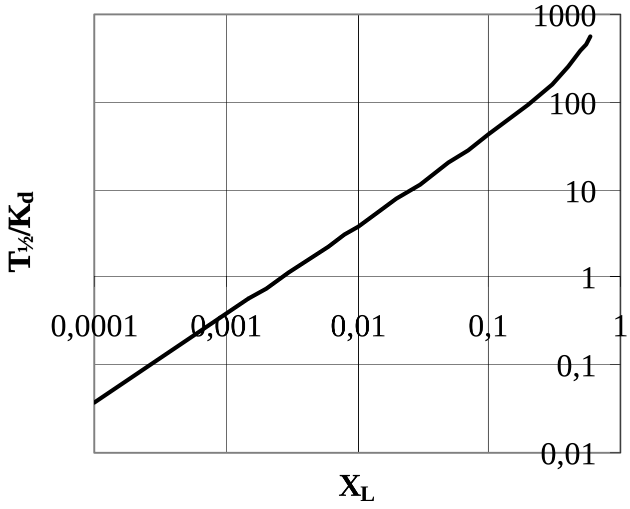


***Figure 2.1*** *The relationship between totally leached fraction ($X_L$) and the ratio $T_{½} / K_d$.*

## 2.3 The validity of the assumed complete mixed conditions in the top soil

The fraction of a given substance leached is approximately zero for $T_{½}/K_d$ ratios smaller than 1. Thus, if e.g. the $K_d$ value is 3000 nearly all the substance will be degraded in the top soil if the half life time for degradation is below 3000 days. Consequently the removal of substance from the top layer will be governed by a simple 1. order degradation removal following a simple exponential concentration fraction decline in dependent on the actual $q$ value.

The water flow dynamics are selected for analysis in this investigation in relation to two mechanisms: (1) Dispersion by water flow in the porous soil and (2) The temporally change in water flow.

*Dispersion*

The dispersion in the soil is primary a result of different water flow velocities locally in the pores of the soil. This will smoothen out spatial concentration gradients and thereby equalize spatial concentration value differences.

*Change during time*

The temporally changes in the water flow value results in removal of substance in different periods of time. The degradation process will decrease the concentration level during time, so, different time scales for the water flow to happen will result in a wash out of different amounts of substance from the top soil.

*Assumed infinite high dispersion*

*The time scale of mixing*

*The concept for the analysis*

The top layer is described as one completely mixed box, thus, the concentration from top to bottom is equal at any time. This is equivalent to assume infinite high dispersion in the top layer. In reality, however, there will exist some degree of advective water transport yielding a concentration profile in the top soil layer. The ploughing is actual a mixing of the top soil, so, the ploughing sequence yields the time scale for analysing the assumption of completely mixed conditions in the top soil. If a stratification in concentration level can be established in the time period between two ploughing events due to the vertical water flow then the assumption of completely mixed conditions is invalid. The time period between the ploughings will typical be about one year, so this is the time scale for analysing the completely mixed assumption. The assumption of completely mixed conditions in the top layer will be investigated in the following analysis. In the analysis the full mixed condition will be tested against a situation of solely advective transport (piston water flow). In reality there will be a specific degree of dispersion in the top soil where concentration gradients will tend to smooth out due to differences in local water flow velocities in the soil pores. This dispersion will act as a kind of mixing in the top soil where a infinite degree of dispersion will yield completely mixed conditions. So the actual state is between the two extremes of completely mixing and piston flow and the difference between those yields the maximum discrepancy due to the assumption of completely mixing in the top soil.

## Advective flow model

*Neglecting dispersion*

The transport equation, where both degradation and dispersion are neglected, can recognize the general pattern of the concentration profile. Subsequently the degradation is included in the final advective flow model description.

The substance transport equation neglecting dispersion and degradation is

$$\frac{\partial C_{tot}}{\partial t} = -\frac{\partial (q_x \cdot C_{dis})}{\partial x} \qquad 2.14a$$

*Assuming constant $q_x$*

where $q_x$ is the downward water flux (dm$^3$/(dm$^2$·d)) at position $x$ from the soil surface. The $q_x$ can vary as a function of depth. The highest value ($q_{max}$) will be at the soil surface and numerically close to the precipitation. In the following the $q_x$ value is assumed constant and equal to $q_{max}$ in order to analyze the case of most dominating advective transport. In this way Eq. 2.14a can be simplified as

$$\frac{\partial C_{tot}}{\partial t} = -q_{\max} \cdot \frac{\partial C_{dis}}{\partial x} \tag{2.14b}$$

Eqs. 2.14b and 2.2 is combined to the equation for advective transport:

$$\frac{\partial C_{tot}}{\partial t} = \frac{q_{\max}}{\mathrm{R}} \cdot \frac{\partial C_{tot}}{\partial x} \tag{2.15}$$

*Characteristics*

Now we will define a curve in the two dimensional time-space plan ($x$,$t$) along which the total concentration ($C_{tot}$) is constant. Such a curve is typically denoted a characteristic and the index $_{char}$ is in used for at value along this curve in the following. The characteristics are defined as

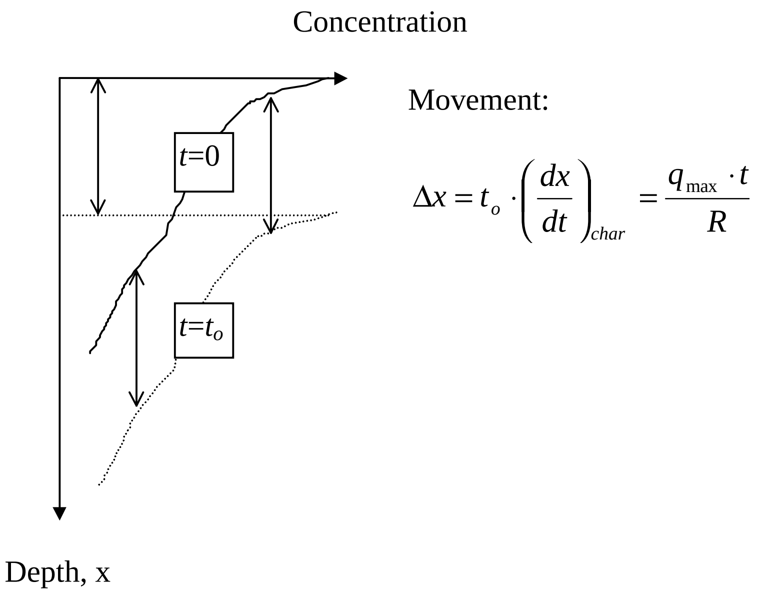
$$\left(\frac{dC_{tot}}{dt}\right)_{char} = \frac{\partial C_{tot}}{\partial x}\left(\frac{dx}{dt}\right)_{char} + \frac{\partial C_{tot}}{\partial t} \equiv 0 \tag{2.16}$$

*Speed of substance migration*

where the chain rule for partial derivatives are used. The total time derivative ($\frac{dC_{tot}}{dt}$) in Eq. 2.16 is equal zero along the characteristic curve, because $C_{tot}$ is defined to be constant along the curve. The dx/dt relationship is simply the downward movement of the characteristic in the soil. Combining Eqs. 2.15 and 2.16 gives

$$\left(\frac{dx}{dt}\right)_{char} = \frac{q_{\max}}{R} \tag{2.17}$$

From Eq. 2.17 it is seen that any characteristic will move downward of the same velocity of $\frac{q_{\max}}{R}$ if the parameters $q_{max}$, and $R$ are assumed constant with respect to $x$. A graphically interpretation of this special case having constant parameters is shown in Fig. 2.2. The shape of the concentration profile will, therefore, be preserved during time.



$$\Delta x = t_o \cdot \left(\frac{dx}{dt}\right)_{char} = \frac{q_{\max} \cdot t}{R}$$

***Figure 2.2.*** *Graphically interpretation of Eq. 2.17 for constant parameters. All concentration values on the concentration profile curve moves downward of the same velocity.*

*Including degradation*

The degradation mechanism will cause a decrease in the concentration level which can be superposed to the downward moving concentration profile. Thus, in case of an initial uniform concentration profile in the top soil the concentration will change during time as illustrated in Fig. 2.3. The uniformly shaped concentration profile will be preserved but the concentration level will decrease due to degradation.

*Time for complete wash out*

The substance will not be uniformly distributed in the top soil layer, instead there exist an upper area where all the substance has disappeared and a lower area, where there the substance remains. However, after a specific time period ($t_1$) the upper boundary of the substance profile ($x$ in Fig. 2.3) will reach the lower boundary of the top soil ($x_1$) and all substance will be completely removed from the top layer. The value for $t_1$ can simply be calculated using Eq. 2.17 as

$$t_1 > \frac{x_1 \cdot R}{q_{\max}} \qquad 2.18$$

The value for $t_1$ can easily by many years for hydrophobic substances, where the retention coefficient $R$ is large. For $q_{max}$ = 70 cm/year, $x_1$ = 20 cm, and a value for $R$ of 1000 will predict the top soil to be completely washed out after more than 286 years. Often the agricultural use of the soil will disturb the top layer structure, by ploughing, so the predicted long time periods are unrealistic. This very slow movement indicates that degradation easily can be the most important mechanism responsible for removal of substance from the top soil. In the following the time value is assumed to be smaller than or equal to $t_1$.

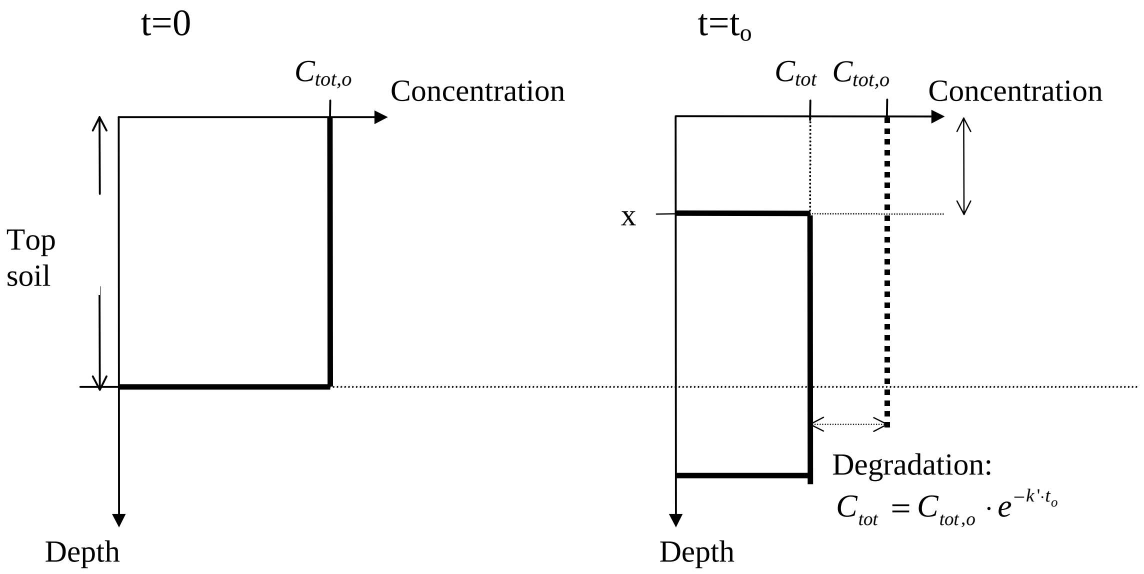


***Figure 2.3.*** *The change in concentration level as function of time and depth in case of linear instant adsorption and no dispersion.*

In the following, $x$ denotes the upper end of the contaminated soil zone, cf. Fig. 2.3. In the time period when $x<x_1$ the mean concentration in the top soil including degradation is

$$C_{tot,m}^{conv} = \frac{x_1 - x}{x_1} \cdot C_{tot,o} \cdot e^{-k' \cdot t} \qquad 2.19$$

where $C_{tot,m}^{conv}$ is the spatial mean concentration in the top soil calculated using the convection model and $x$ is the movement of the substance as illustrated in Fig. 2.3. This equation can be rewritten using Eq. 2.17 as

*Advective flow model*

$$C_{tot,m}^{conv} = \left(1 - \frac{q_{\max} \cdot t}{R \cdot x_1}\right) C_{tot,o} \cdot e^{-k' \cdot t} \qquad 2.20$$

The relative difference (Δ) between the box model (completely mixed) in form of Eq. 2.4 and Eq. 2.20, respectively is

*Difference between advective and dispersive flow*

$$\Delta = \frac{C_{tot}^{mixed} - C_{tot,m}^{conv}}{C_{tot}^{mixed}} = \frac{e^{-\left(\frac{q}{R \cdot x_1} + k'\right) \cdot t} - \left(1 - \frac{q_{\max} \cdot t}{R \cdot x_1}\right) \cdot e^{-k' \cdot t}}{e^{-\left(\frac{q}{R \cdot x_1} + k'\right) \cdot t}} \qquad 2.21$$

$$= 1 - \left(1 - \frac{q_{\max} \cdot t}{R \cdot x_1}\right) \cdot e^{\frac{q \cdot t}{R \cdot x_1}}$$

The following Taylor series will be used for analysis of Eq. 2.21:

$$e^u = 1 + u + \frac{u^2}{2!} + \frac{u^3}{3!} \ldots \qquad 2.22$$

where $u$ is an arbitrary variable. This series will be used for the exponential expression: $e^{\frac{q}{R\cdot x_1}\cdot t}$ in Eq. 2.21 neglecting second order and higher terms. The second order and higher terms in Eq. 2.22 are negligible when the u values are ‘much’ lower than unity. The value of $\frac{q_{\max}\cdot t}{R\cdot x_1}$ is always smaller than unity because otherwise the time period $t$ is so long that all the substances are washed out of the top soil according to Eq. 2.17. The $q_{max}$ value is always larger than the $q$ value (typical range 2-3 times larger) so the term $\frac{q\cdot t}{R\cdot x_1}$ will also be smaller than unity in a higher degree than $\frac{q_{\max}\cdot t}{R\cdot x_1}$. Using the part $e^u=1+u$ from Eq. 2.22 in Eq. 2.21 yields

$$\Delta \approx \frac{q_{\max}\cdot t}{R\cdot x_1}-\frac{q\cdot t}{R\cdot x_1}+\frac{q_{\max}\cdot t}{R\cdot x_1}\cdot\frac{q\cdot t}{R\cdot x_1} \qquad 2.23$$

All the ratios in this equation are lower than unity as discussed above, so the following simple relationship reveals:

$$\Delta < \frac{q_{\max}\cdot t}{R\cdot x_1}, \quad for\ R >> \frac{q_{\max}\cdot t}{x_1} \qquad 2.24$$

where the demand on $R$ to be much larger than the ratio $\frac{q\cdot t}{R\cdot x_1}$ is needed for the Taylor assumption to be valid ($u$<<1).

The numbers: $q_{max}$=700 mm/year, $t$= 1 year (the period between ploughing) and $x_1$=20 cm yields:

*Simple rule for the relative difference*

$$\Delta < \frac{3,5}{R}, \quad for\ R >> 3.5 \qquad 2.25$$

Thus, if a slightly hydrophobic substance is associated whit a $R$ value of 1000, then the maximal discrepancy due to the assumption of completely mixed conditions in the top soil is 0.35% compared to calculation where the true (but unknown) dispersion is used.

# 3 Experimental results

Field scale measurement of the top soil contamination has been undertaken by *Vikelsøe et al., (2000)*. These data will be further interpreted in an analysis using the simple model for the top soil.

## 3.1 The chemical properties of DEHP

The general structure of the phthalates is shown in Figure 3.1 and the specific structure for DEHP is shown in Figure 3.2. DEHP is mainly hydrophobic exhibiting high sorption affinity to soils.

The basic environmental properties of DEHP are shown in Table 3.1.

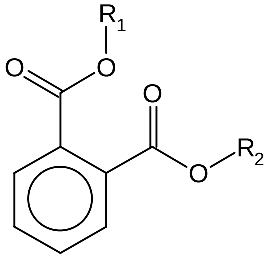


***Figure 3.1*** *Structure of phthalates. R1 and R2 may be alkyl or aryl groups*

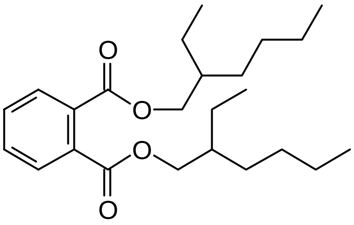


***Figure 3.2*** *Di-2-ethylhexyl phthalate (CAS 117-81-7)*

***Table 3.1*** DEHP properties

| ***Property*** | | ***References*** |
|---|---|---|
| CAS number | 117-81-7 | |
| Formula | $C_{24}H_{38}O_4$ | |
| MW (g/mol) | 390.6 | |
| Mp ($^{o}$C) | -45, -46, -55, -50 | Furtmann (1996) |
| Bp ($^{o}$C) | 370, 385 | Furtmann (1996) |
| Aqueous solubility (mol/L) | $4.7\cdot10^{-7}$ | Thomsen and Carlsen (1997) |
| Aqueous solubility (mg/L) | 0.18 | |
| Henry’s law constant | 0.03, 18.6, 64.1, 4.3 (293 K), 7.8 (298 K), | Furtmann (1996) |
| log $K_{ow}$ ( | 9.64, 7.94, 5.11, 7.5, 7.45, 7.4, 5.11, 3-9.6 | Thomsen and Carlsen (1998) |
| log $K_{oc}$ | 5.48, 5.94, 5.41, 6, 4.94, 5.8 | Thomsen and Carlsen (1998) |
| $K_d$ (cm$^3$/g) | 452, 1000-5000, 1390, 4830, 5860 | Thomsen and Carlsen (1998) |
| $T_{½}$ (days) | approximately 400 | Roslev et al. (1998) |
| Density (g/cm$^3$) at 25 $^{o}$C | 0.981, 0.98-0.985, 0.986 | Furtmann (1996) |
| Surface tension (N/m) | 1.5, 32.2 mN/m | Furtmann (1996) |
| Flash point (K) | 488 | Furtmann (1996) |
| Dynamic viscosity (mPa·s) | 58 (25 $^{o}$C) | Furtmann (1996) |
| Molar volume (mL/mol) | 525 | Furtmann (1996) |

## 3.2 Field scale measurements

*Sampling strategy*

Soil samples are taken 6 and 8 years after the termination of the sludge loading period respectively. Two different sample sites were selected for sampling in both years, where the vertical concentration profiles (0-50 cm) were investigated *(Vikelsøe et al., 2000)*. In the present analysis the data for the top layer (0-20 cm) are used to estimate the contamination level in the top soil. It was not possible to observe any significant difference between the two years of sampling and between the depth of 10 and 20 cm *(Vikelsøe et al., 2000)*. Thus, results from both years are pooled in order to estimate the soil contamination after 7 years, corresponding to a value between the two years of sampling. The concentration measurement results are shown in Table 3.2.

***Table 3.2*** The measured soil concentration levels *(Vikelsøe et al., 2000).*

| ***Year (month)*** | ***Sample site number*** | ***Concentration (mg/kg dw)*** | ***Concentration (mg/kg dw)*** |
|---|---|---|---|
| **1996 (August)** | 1 | 480 | 1700 |
| | | 1120 | 1700 |
| | 2 | 770 | 1800 |
| | | 1600 | 1600 |
| **1998 (October)** | 3 | 1526 | 1868 |
| | | 1816 | 2290 |
| | 4 | 1000 | 1324 |
| | | 1440 | 1273 |

*Variability in concentration levels*

The mean concentration is 1457 mg/kg dw and the standard deviation is 453 mg/kg dw. The purpose of the analysis is to compare the mean soil concentration to modeling results for the top soil. However, the mean is associated to some uncertainty, which has to be taken into account. The concentration values are assumed normal distributed. Thus, the mean value is also normal distributed and the density function for the mean value will be used in the analysis.

## 3.3 Estimation of DEHP loading

*The site of wastewater treatment facility*

The field has received all sludge produced at Roskilde municipal water treatment plant during 25 years ending in 1990. Today, the actual amount of wastewater generated from the area in Roskilde is 80.000 Pe (Person equivalents). However, the amount of wastewater has obviously changed during the 25 years period. The wastewater treatment technology has also changed from a mixture of both mechanical, biofilm and activated sludge facilities to only activated sludge.  Furthermore, expansion has happened of both the numbers of connected inhabitants and industries.

The sludge load to the soil is calculated using estimates for wastewater load and sludge production per load unit. The wastewater load is calculated using estimated figures for the number of person equivalents (*pe*). The sludge production is calculated using an estimate for the amount of sludge generated per day from one pe. Obviously significant uncertainties are related to all the estimates, so, the input numbers will be treated as intervals and the final load calculation will thus be a probability density function. The calculation is summarized in the following equation.

*Sludge load*

$$L = \frac{N_{pe} \cdot SP_{pe} \cdot C_{pe}}{A} \qquad 3.1$$

where $L$ is the daily load of substance per unit area (mg/(m$^2$·d)), $N_{pe}$ is the number of person equivalents (*pe*), $SP_{pe}$ is the sludge production per day per pe (kg dw/(pe·d)), $C_{pe}$ is the substance (DEHP) concentration in the sludge (mg/kg dw) and $A$ is the field area receiving the sludge (m$^2$). The unit for the load ($L$) becomes mg substance per m$^2$ per day. Thus, the load can be interpreted as a flux of substance to the field.

*Value intervals for load estimate*

There is no reason to assume that the waste water load during the period of sludge supply has been larger than the actual load of today (80.000 pe). So, the maximum waste water load in the period of sludge supply is estimated as 80.000 *pe*. The minimum waste water load is estimated to be half of the maximum (40.000 *pe*). The sludge production from one *pe* is estimated as the interval between 30 and 60 g dw/d *(Christensen and Sørensen, 1994)*, where the value depends on the load of matter per *pe*, the total yield coefficient for the plant and the degree of cleaning. The DEHP content from experimental results of two fresh sludge samples from two similar wastewater treatment plants are 25 and 29 mg/kg dw respectively *(Vikelsøe et al., 2000)*. However, due to the limited data set and the fact that the load of sludge has been taken place in a longer time period the concentration estimate will be the interval of 10-50 mg/kg dw. The field area is 100 ha.

*Distribution function for load estimate*

Parameter values are randomly selected from the identified intervals using a simple Monte Carlo technique (using 10000 realizations). The resulting load calculations using Eq. 3.1 are shown in Fig. 3.3.

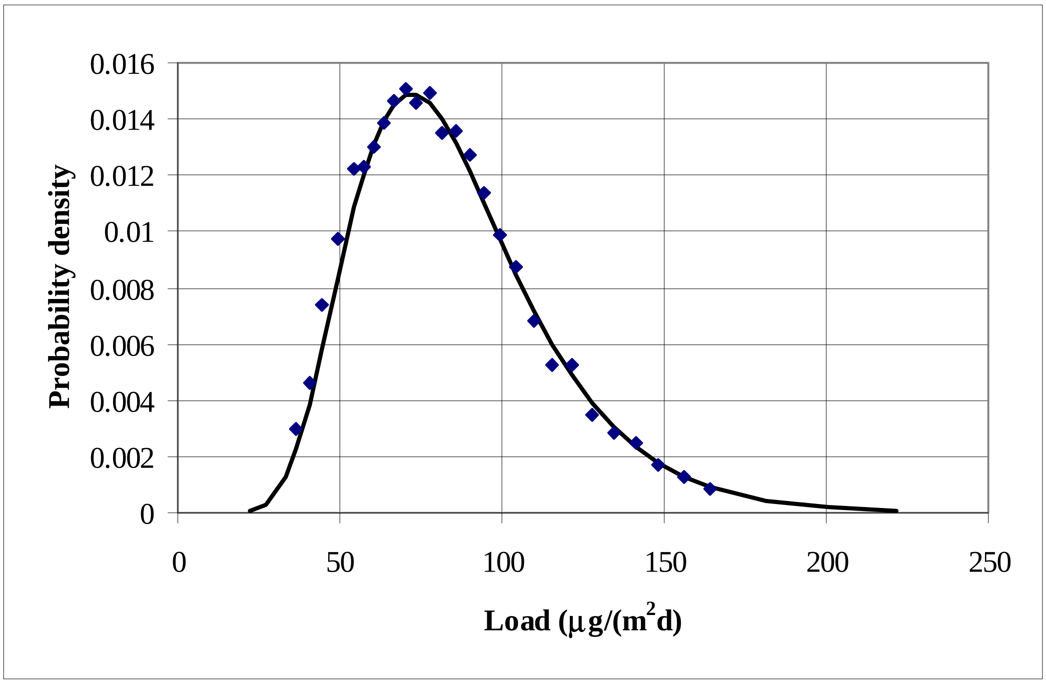


***Figure 3.3*** *Load density function as a lognormal distribution (curve) compared to Monte Carlo estimates (data points).*

A lognormal distribution seems to make a suitable fit to the Monte Carlo results (cf. Fig. 3.3) In the load estimate (Eq. 3.1), the parameters are multiplied and such a product of a series of stochastic numbers will tend be lognormal distributed as seen in Fig. 3.3.

# 4 Determination of the field half life time

*Bayesian approach*

The agricultural area has been subject to sludge amendment during 25 years until year 1990 where the load was ended. The measurements were taken during the 90'ties. Therefore, the used mathematical description of the top soil assumes a termination of a continuously sludge load period (Eq. 2.10) and will be used in a Bayesian type statistical approach in order to identify the resulting variability introduced in the top soil model from the data input. This variability is partly a result of a 'true' variability in a heterogeneous environment and partly a result in experimental uncertainty. In this analysis the total variability will be considered as the condition for the model predictions.

The probability density function for the load ($f_L$) is described by a log-normal distribution (Fig. 3.3) and Eq. 2.10 relates a load value to a soil concentration level. Thus, for known values of $t_{load}$, $t$ and $K_{dis}$ it is possible to calculate a probability density function for soil concentration levels, $f_{c,cal}$. The probability for the load value $L$ is $f_L$ and the probability density function for the concentration value $C$ is $f_{c,cal}$. The concentration and load are interrelated as

$$f_L \cdot dL = f_{c,cal} \cdot dC \Leftrightarrow f_{c,cal} = f_L \cdot \frac{dL}{dC} \qquad 4.1$$

Combining Eqs. 4.1 and 2.10 yields

$$f_{c,cal} = f_L \cdot \frac{x_1 \cdot K_{dis}}{(1 - e^{-K_{dis} \cdot t_{load}})} \cdot e^{K_{dis} \cdot t} \qquad 4.2$$

*Density function for concentration estimates*

where the dissipation coefficient $K_{dis}$ is used as aggregated parameter for $\frac{q}{R} + k'$ and $L$ in $f_L$ is calculated as a function of $C$. Eq. 4.2 can be considered as a probability density function for model estimates of concentration values. Two probability density functions for $C$ can be made: (1) based on the measurements and (2) Based on Eq. 4.2. From these two functions, the probability for a concentration value $C$ to be both measured and calculated is determined in the next paragraph.

*Joint probability*

The joint probability for at concentration value $C$ to be included in both the measurements ($f_c$) and the calculation ($f_{c,cal}$) is the product: $f_c \cdot f_{c,cal}$ (Fig. 3.4).

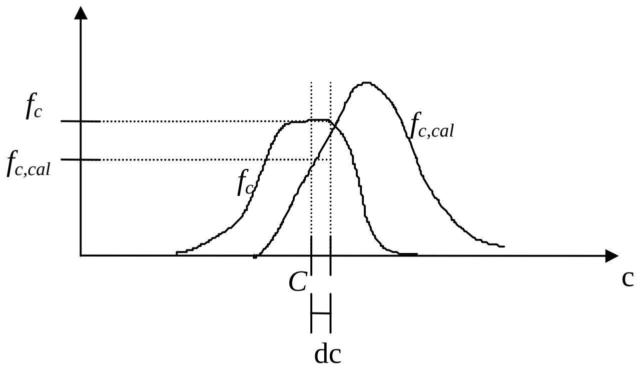


***Figure 4.1** The principle of calculating the probability for co to be included in both $f_c$ and $f_{c,cal}$.*

The probability (*m*) for any concentration value to be included in both the measurements and the calculation is the sum of the joint probabilities for all concentration values. Thus, *m* is calculated by the integral equation as

$$m = \int_0^{\infty} f_c \cdot f_{c,cal} \, dC \qquad 4.3$$

*Likelihood function for $K_{dis}$*

A function of this type is denoted the marginal distribution by *Berger (1985)*. The *m* values appear as a function of a series of parameters out of which only $K_{dis}$ is unknown. The *m* function can therefore be taken as a measure for the relevance of a specific $K_{dis}$ value, where higher *P* values means higher relevance for the associated $K_{dis}$ values. In this way the *m* function is a likelihood function *(Berger, 1985)*. The *m* function calculated by numerical integration of Eq. 4.3. is shown in Fig. 4.2. as a function of $K_{dis}$.

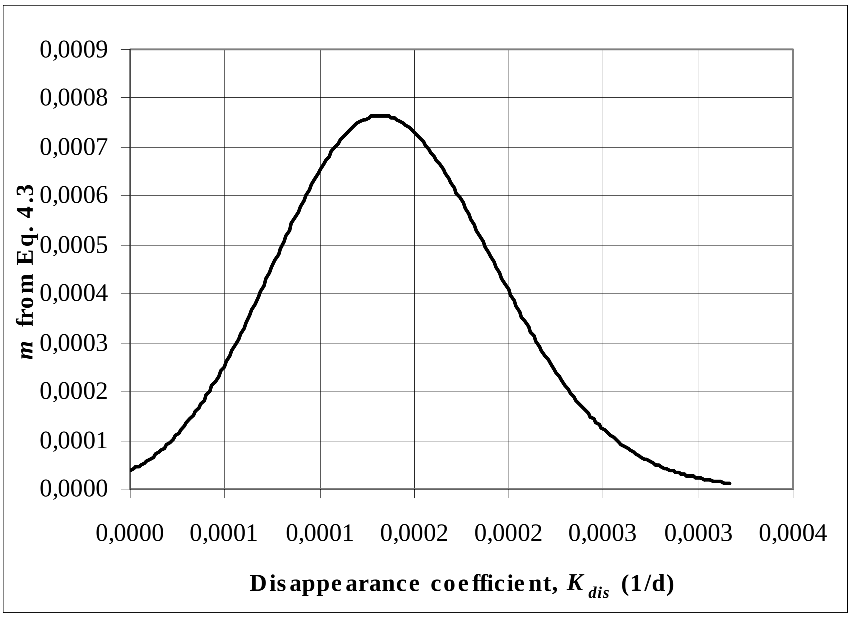


***Figure 4.2** The m values (Eq. 4.3) as a function of $K_{dis}$.*

*Interpreted of the m function*

The *m* function can be interpreted graphically (Fig. 4.3). The model over estimates the concentration values for small values of $K_{dis}$ (small disappearance from top soil). Contrary, the model under estimates the measurements for larger values of $K_{dis}$ (high disappearance from top soil). $K_{dis}$ value between these extremes apparently gives a more realistic coincidence between the model predictions and the measurements.

It is emphasized that the *m* function is not an probability density function as the area under the curve (Fig. 3) is not unity. However, the shape of the curve is close to be similar to a normal distribution. The curve may, however, be normalized in order to obtain a area under the curve of unity. Thereby, the curve will form a probability density function for $K_{dis}$. The interpretation of the *m* function has to be done with caution, since the *m* function is discontinuous around zero. In our case the $K_{dis}$ value needs to be positive or zero but not negative so the intersection on the y-axis at $K_{dis}$ equal zero is a discontinuity. This discontinuity is in our case, however, small (cf. Fig. 4.2) and will not be taken into account.

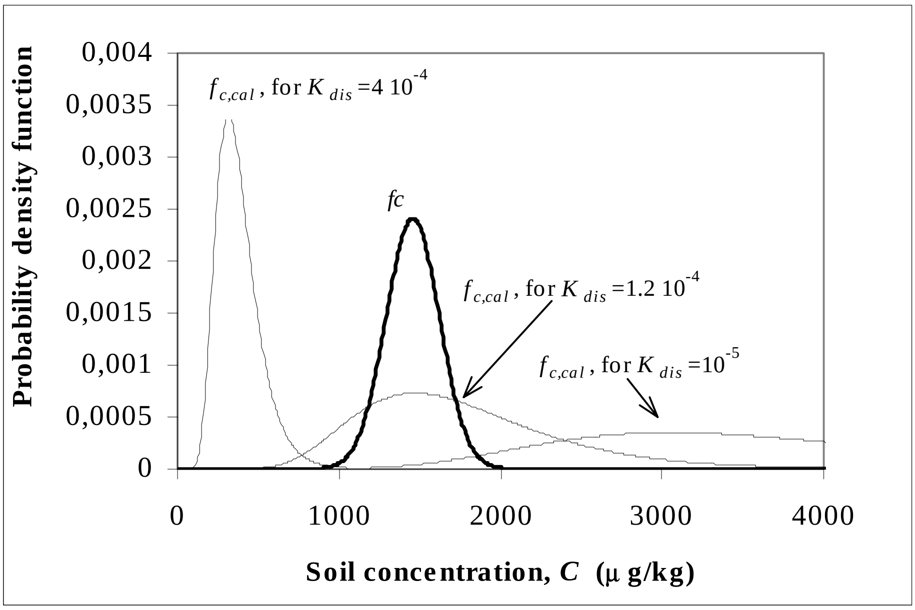


***Figure 4.3*** *The principle of calculating the probability density function for $K_{dis}$ from the two density functions for C (the measured and calculated using Eq. 4.3) at different fixed values of $K_{dis}$.*

*Probability function for $K_{dis}$*

When the *m* function is normalized to have an under laying area of unity it can be used to generate an accumulated probability curve for a specific $K_{dis}$ value or smaller (cf. Fig. 4.4). An realistic interval for $K_{dis}$ could be from 0.00005 to 0.00027 1/d. In terms of the field half life this corresponds to an interval from 7 to 38 years, which indicates that DEHP is rather persistent.

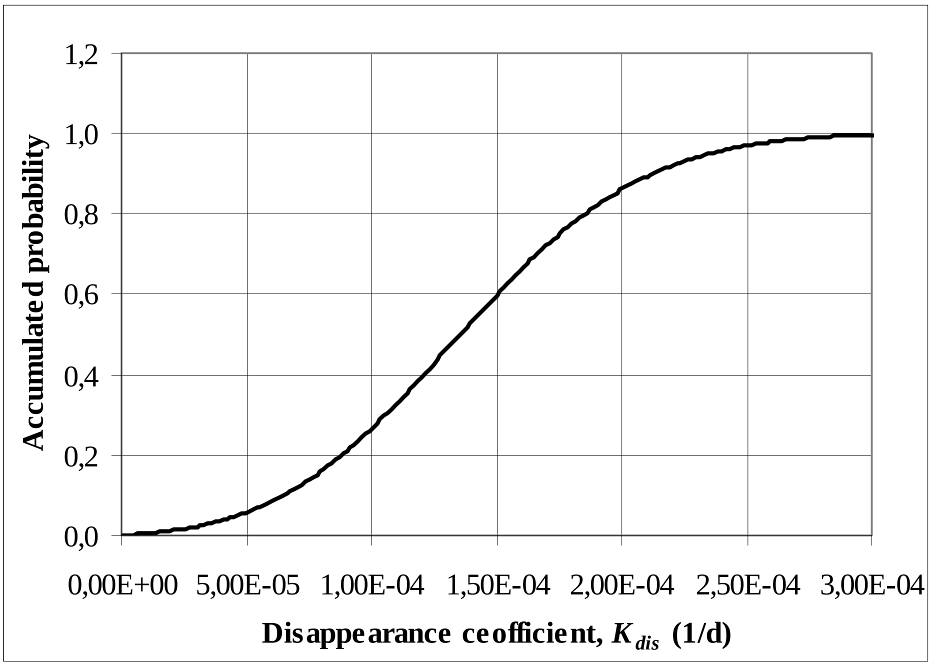


***Figure 4.4.*** *The accumulated probability curve for* $K_{dis}$ *to be smaller than the specific value on the x-axis.*

# 5 Application of the soil model

*Top soil model as decision support*

The model for the top soil is useful as decision support for the management of sludge amendment of agricultural soils. The estimated distribution function for $K_{dis}$ will in the following be used in three examples in order to illustrate different use of the model. The basic concept is a stochastic modeling approach, where the uncertainty of the key input parameters result in a probability distribution function for the model result.

## 5.1 Determination of recovery time values

*Known initial soil concentration*

*Unknown initial soil concentration*

When soil has been contaminated to an extent where the concentration level appears to be above the recommended soil quality criteria it will be useful to estimate the time period necessary for the soil to recover. Two examples of such estimates will be given in the following. In the first example the initial soil concentration is assumed known in form of the experimental results from *Vikelsøe et al. (2000)*. In the second example the initial concentration is assumed unknown and the load is estimated as above for the Roskilde area. The two examples results in different uncertainty, as the first example avoids the uncertain estimate of the load during 25 years and uses more confident field concentration measurements. However, both scenarios do implicit have uncertainty arising from the load estimates, since this uncertainty was involved in the estimate of $K_{dis}$.

**Known initial concentration levels**

In this case the initial soil concentration is assumed known in form of the results in Table 1. The problem is then to estimate the recovery time period ($t_{rec}$) necessary for the soil concentration value to drop below a specific soil quality criteria ($c_{sqc}$). The soil quality criterion is based on the concentration relative to the mass of dry soil (µg/kg dw) and not the volumetric concentration (µg/l). However, in this analysis the volumetric concentration can simply be replaced by the concentration based on soil mass using the density of the dry soil. The disappearance is assumed to be a first order so $t_{rec}$ and $c_{sqc}$ are assumed related as

$$c_{sqc} = c_o \cdot e^{-K_{dis} \cdot t_{rec}} \qquad 5.1$$

where $c_o$ is the initial soil concentration in relation to the mass of dry soil (µg/kg dw) and calculated using Eq. 2.4. Eq. 5.1 can be rewritten to

*Time for recovery*

$$t_{rec} = \frac{1}{K_{dis}} \cdot \ln\left(\frac{c_o}{c_{sqc}}\right) \qquad 5.2$$

Both the parameters $K_{dis}$ and $c_o$ in Eq. 5.2 are described as distribution functions, so the equation has to be treated in a stochastically way. A convenient way to handle Eq. 5.2 is a Monte Carlo simulation as done in

Fig. 5.1, where 10000 realizations of Eq. 5.2 were performed for the three soil quality criteria ($c_{sqc}$): 500, 1000 and 1500 µg/kg dw.

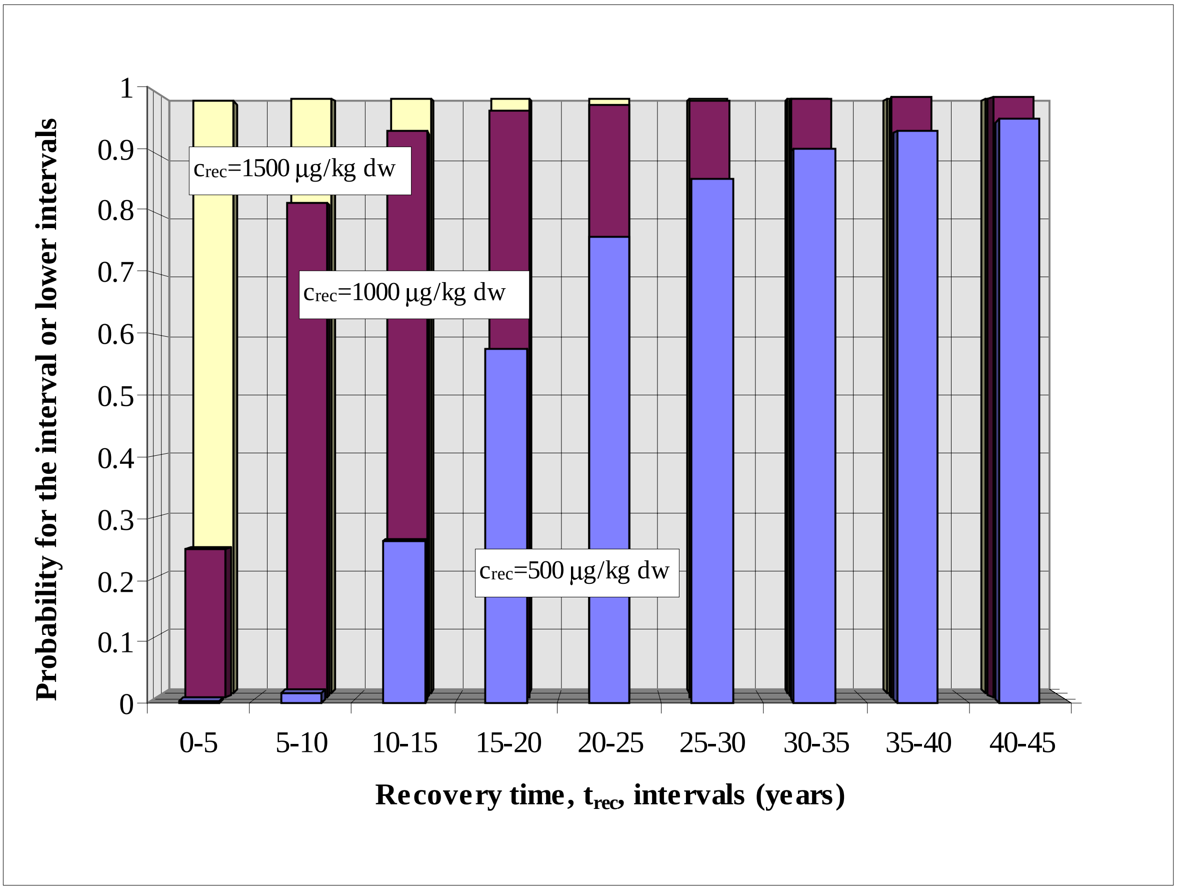


***Figure 5.1*** *The accumulated probability for the trec value to be in the interval. Calculated based on 10000 Monte Carlo realizations of Eq. 5.2, where the initial soil concentration is assumed known with out uncertainty.*

*Recovery time estimates*

The $c_{sqc}$ value of 1500 µg/kg dw is met already in the first time period interval (0-5 years), which is not surprising, because the mean value for $C_o$ is below 1500 µg/kg dw. For the $C_{sqc}$ value of 1000 µg/kg dw a recovery time period of 10-15 years is needed in order to meet a 95% confidence level and for a $c_{sqc}$ value of 500 mg/kg dw the 95% confidence level is reached after 35-40 years.

## Unknown initial concentration levels

When the initial soil concentration value is unknown it obviously needs to be calculated using estimated values for the load and time period of loading. Using Eq. 2.10 and the relationship $C = \sigma \cdot c$ the recovery time equation becomes

*Time of recovery*

$$t_{rec} = \frac{1}{K_{dis}} \cdot \ln\left( \frac{L}{c_{sqc} \cdot \sigma \cdot x_1 \cdot K_{dis}} \cdot \left(1 - e^{-K_{dis} \cdot t_{load}}\right) \right) \qquad 5.3$$

The parameters $L$ and $K_{diss}$ are described by probability functions, so the use of Eq. 5.3 needs to be in terms of probabilities. A Monte Carlo technique is again applied for the calculations.

In the following analysis the value for $c_{sqc}$ are sat equal to respectively 500, 1000 and 1500 µg/kg dw and the loading time ($t_{load}$) is 25 years. 10000 Monte Carlo realizations are made using the two probability distribution functions identified earlier for respectively $L$ and $K_{dis}$. The results are summarized in Fig. 5.2.

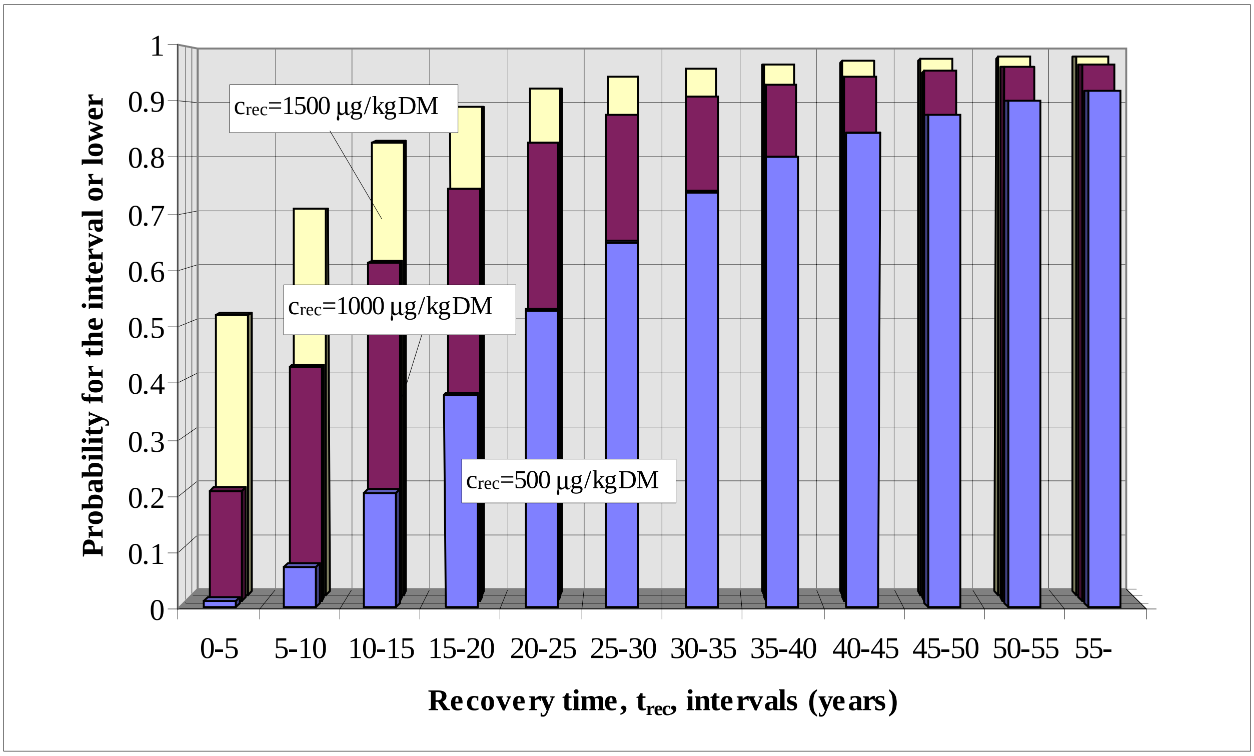


***Figure 5.2*** *The accumulated probability for the* $t_{rec}$ *value to be in the interval. Calculated based on 10000 Monte Carlo realizations of Eq. 5.3, where the initial soil concentration is estimated from the load.*

*Longer estimated recovery time period for unknown initial concentration level*

When the results in Fig. 5.2 are compared to those in Fig. 5.1 it becomes clear that the probability build up for increasing time values in Fig. 5.2 are less pronounced compared to Fig. 5.1. This difference is easily explained as a result of the higher degree of uncertainty in the calculation of Fig. 5.2, where the initial soil concentration is unknown in contrast to Fig. 5.1.

If the uncertainty in the calculations leading to Fig. 5.2 has to be reduced, more historic information about the load needs to be collected. This will further reduce the uncertainty in the estimate of $K_{dis}$ as well and thereby also reduce the uncertainty in the results summarized in Fig. 5.1.

## 5.2 Prediction of maximum amendment period

The maximum sludge amendment period may be calculated in order to secure the soil concentration to be below the soil quality criteria ($c_{sqc}$). During the sludge amendment period the concentration development in the top soil is described by Eq. 2.7.

*Maximal loading for infinite time periode*

As a first step in the analysis the infinite loading time concentration value ($C_{tot\infty}$) is compared to the soil quality criteria concentration value ($c_{sqc}$). Any loading time is acceptable if (using Eq. 2.8):

$$c_{sqc} < \frac{L}{\rho \cdot x_1 \cdot K_{diss}} \qquad 5.4$$

where the relationship: $c_{sqc} \cdot \rho = C_{tot\infty}$ is used. Therefore, the critical load, $L$, below which the soil quality criteria never will be exceeded is calculated using a rewritten form of Eq. 5.4 as

$$L = c_{sqc} \cdot \rho \cdot x_1 \cdot K_{dis} \qquad 5.5$$

This equation shows how the x-axis at the accumulated probability distribution function for $K_{dis}$ (Fig. 4.4) can be multiplied by the factor of to form a similar curve for the critical load. The critical load is identified using a specific significance level for which $L$ is exceeded. Thus, the value for $K_{diss}$ is about $5 \cdot 10^{-5}$ 1/d on a significant level of 0.05 (Fig. 4.4). This $K_{dis}$ value results in a $L$ value of 15µg/(m$^2$·d) for $c_{sqc}$ = 1000 µg/kg dw, which is a low value compared to the estimated values for $L$ in Fig.3.3.

*Maximal load in a finite time periode*

More realistic calculations is possible where loading time period and concentration build up are related using Eq. 2.7. In a rewritten version of Eq. 2.7, the time period for a load to take place ($t_{max}$) to reach the soil quality criteria concentration ($C_{sqc}$) is calculated as

$$t_{max} = \frac{1}{K_{dis}} \cdot ln\left( \frac{1}{1 - \frac{c_{sqc} \cdot \rho \cdot x_1 \cdot K_{dis}}{L}} \right) \qquad 5.6$$

The disappearance coefficient $K_{dis}$ is a stochastic variable while $c_{sqc}$ and $L$ are fixed values in this analysis.

Eq. 5.6 is used for $c_{sqc}$=1000 µg/kg dw and load values of respectively 25, 50 and 75 µg/(m$^2$·d). 10000 Monte Carlo realizations are made, the results being shown in Fig. 5.3. A cautious analysis will allow the high load (75 µg/(m$^2$·d)) to proceed for 8 years whereas the lower load of 50 µg/(m$^2$·d) may proceed for 16 years. The lowest load of 25 µg/(m$^2$·d) may proceed for all 48 years involved in the problem investigation indicating that the maximum load value of 15 µg/(m$^2$·d) as predicted using Eq. 5.6 is rather conservative.

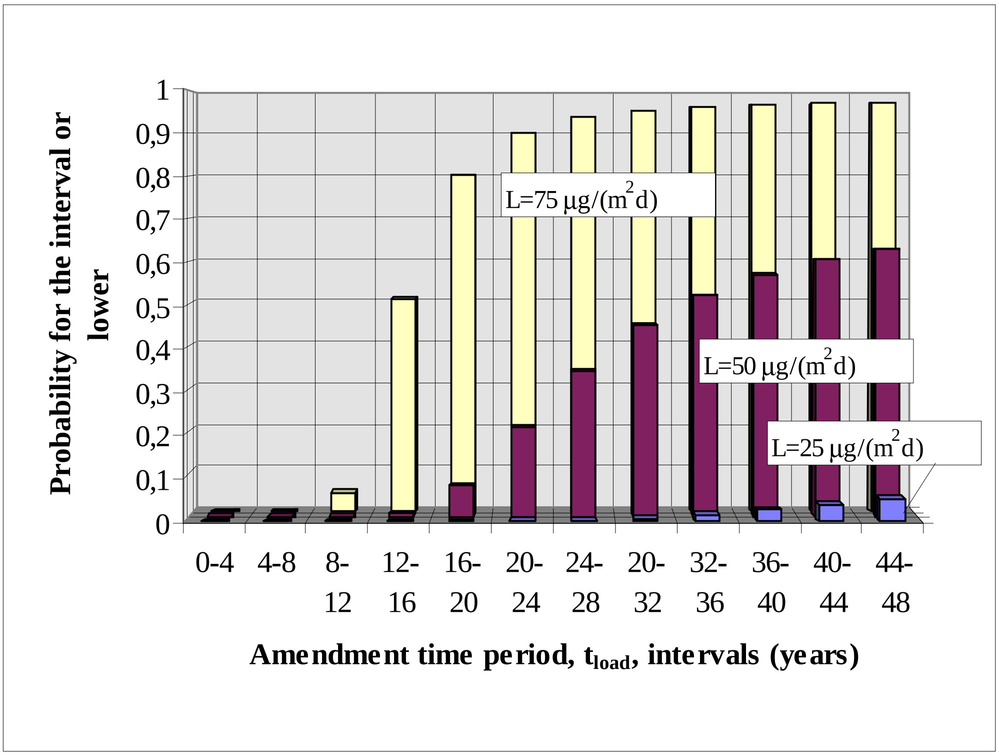


***Figure 5.3*** *The predicted accumulated probability for the loading time period needed to reach a soil quality criteria of 1000 µg/kg dw.*

# 6 Symbols

| | | |
|---|---|---|
| $A$ | field area receiving the sludge | ($m^2$) |
| $C_{diss}$ | dissolved concentration of substance | ($\mu$g/l) |
| $C_{tot}$ | total volumetric concentration of substance | ($\mu$g/l) |
| $C_o$ | either the initial total or the initial dissolved concentration | (µg/l) |
| $C_{pe}$ | substance (DEHP) concentration in the sludge | (mg/kg dw) |
| $C_{tot\infty}$ | steady state concentration | (µg/l) |
| $C^{conv}$ or $C^{mixed}$ | Concentration calculate using either the advective flow model or the completely mixed top soil model respectively | (µg/$dm^3$) |
| $c_{sqc}$ | soil quality criteria | (µg/kg dw) |
| $c_{"indeks"}$ | Concentration relative to dry soil mass | (µg/kg dw) |
| $f_{c,cal}$ | probability density function for the concentration value $C$ | (-) |
| $f_L$ | probability density function for the load | (-) |
| $Flux_{leach}$ | the flux of substance released by leaching | (µg/($dm^2$·d)) |
| $K_d$ | reversible linear adsorption coefficient | (l/kg) |
| $K_{dis}$ | total disappearance coefficient for the top soil | (1/d) |
| k’ | overall degradation rate ($k' = \frac{k_1}{R}$) | (1/d) |
| $k_1$ | first order degradation coefficient | (1/d) |
| $L$ | load of substance per unit time due to sludge amendment | (mg/($m^2$·d) |
| $m$ | marginal distribution of $f_{c,cal}$ and $f_L$ | (-) |
| $N_{pe}$ | number of person equivalents (*pe*) | (-) |
| $q$ | mean flux of water leaving the top soil (leaching) | (dm/d) |
| $q_{max}$ | maximal water flux in the soil (at the soil surface) | (dm/d) |
| $q_x$ | downward water flux at position $x$ from the soil surface | (dm/d) |
| $R$ | retention factor | (-) |
| $SP_{pe}$ | sludge production per day per *pe* | (kg dw/(pe·d)) |
| $T_{½}$ | total degradation half lifetime | (d) |
| $t_{load}$ | time period for the load to take place | (d) |
| $t_{max}$ | the time period for a load to take place | (d) |
| $t_{rec}$ | recovery time period | (d) |
| $u$ | arbitrary variable in Taylor series | (-) |
| $X_L$ | ratio between the amount supplied and the amount leached | (-) |
| $x_1$ | top soil layer thickens | (dm) |

| | | |
|---|---|---|
| $\Delta$ | relative difference between box model and advective flow model | (-) |
| $\rho$ | dry bulk density of the soil | (1.5 kg/$dm^3$) |
| $\theta$ | volume fraction of water | (-) |

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