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Uncertainty and stochastic sensitivity analysis of the GeoPEARL pesticide leaching model

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ABSTRACT

GeoPEARL is a spatially distributed model describing the fate of pesticides in the soil–plant system. It calculates the drainage of pesticides to local surface waters and the leaching into groundwater. GeoPEARL plays an important role in the evaluation of Dutch pesticide policy plans. This study analysed how uncertainties in soil and pesticide properties propagate through GeoPEARL for three representative pesticides. The GeoPEARL output considered is the 90th percentile of the spatial distribution of the temporal median of the leaching concentration (P90). The uncertain pesticide properties are the coefficient of sorption on organic matter and the half-life of transformation in soil. Both were assumed uncorrelated in space and were represented by lognormal probability distributions. Uncertain soil properties considered were horizon thickness, texture, organic matter content, hydraulic conductivity and the water retention characteristic. Probability distributions were derived from meta-data stored in the Dutch soil information system. A regular grid sample of 258 points covering the agricultural area in the Netherlands was randomly selected. At the grid nodes, realizations from the probability distributions of uncertain inputs were generated and used as input to a Monte Carlo uncertainty propagation analysis. The results show large uncertainties in P90, with interquartile ranges larger than the median for all three pesticides. Taking input uncertainty into account also leads to a systematic shift of the P90 towards greater values. Stochastic sensitivity analysis showed that the pesticide half-life is the main source of uncertainty and that the coefficient of sorption to organic matter and uncertainty in soil organic matter contribute to a lesser extent. Uncertainty contributions from other soil properties were negligible. These results suggest that improved assessment of soil properties will only marginally improve the accuracy of the predicted pesticide leaching. Instead, more accurate assessment of the pesticide properties, in particular the pesticide half-life, is required. This is, however, difficult, because the pesticide half-life depends on highly variable soil microbial properties in a way that is as yet poorly understood.

1. Introduction

Evaluation of the leaching potential of pesticides into the groundwater is an important aspect of pesticide registration procedures in the Netherlands (Van Der Linden et al., 2004), among others because groundwater is a major source of drinking water. Leaching of pesticides to the groundwater is assessed with the GeoPEARL model, which is a spatially distributed model that combines a one-dimensional, process-based leaching model with spatially distributed input data stored in a Geographical Information System (Tiktak et al., 2002). A pesticide can only be registered for use in the Netherlands if the GeoPEARL analysis shows that the concentration of the pesticide is below 0.1 μg/L for at least 90% of the intended use area. This regulatory endpoint can be directly inferred from the spatial cumulative frequency distribution of the predicted leaching concentration in the intended use area.

Uncertainty is not explicitly addressed in the pesticide risk assessment. However, understanding the consequences of uncertainty is important to improve risk assessment as a decision-support tool (Brown and Heuvelink, 2005). The different sources of uncertainty in pesticide fate modelling were reviewed by Dubus et al. (2003). Uncertainty is contained in environmental input data (e.g. soil properties and climatic data) (e.g. Diaz-Diaz and Loague, 2000), in pesticide properties (particularly the pesticide half-life and the coefficient for sorption on organic matter) (Boesten, 1991; Dubus et al., 2003), it can result from the use of imperfect pedotransfer functions (Tiktak et al. 1999; Dubus et al., 2003) or be caused by spatial interpolation errors (Brus and Jansen, 2004). Model structure and the numerical solution of the model provide additional sources of uncertainty (Addiscott et al., 1995; Refsgaard et al., 2006).

This paper presents a Monte Carlo uncertainty and stochastic sensitivity analysis of GeoPEARL. Only the propagation of uncertainty

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in model inputs is considered. GeoPEARL has a large number of inputs, many of which are spatially distributed. An uncertainty analysis with all model inputs treated uncertain would be too complex and would require too much computation time. The analysis is therefore restricted to those inputs that have substantial uncertainty and for which the model was shown to be sensitive (Boesten, 1991). The model output considered is the 90th percentile of the spatial distribution of the PEC50 in the intended use area (P90). Here, PEC50 is defined as the temporal median of the annual average leaching concentration at 1 m depth. To get a stable estimate of PEC50, the median annual average leaching concentration is obtained for a weather time series of 20 years (FOCUS, 2000). In this study, the intended use area over which the PEC50 is aggregated is taken as the entire agricultural area of the Netherlands.

The objectives of this work are twofold. First, it aims to quantify the uncertainty about the P90, as caused by uncertainty about pesticide properties and soil characteristics. This is useful to decide whether the GeoPEARL output is sufficiently reliable for decision making. Second, it assesses the contribution of (groups of) individual uncertain inputs to the total uncertainty in P90. This provides valuable insight into which inputs must be more accurately estimated to improve the quality of the GeoPEARL output.

2. Identification and stochastic simulation of uncertain GeoPEARL inputs

2.1. Characterizing uncertain soil properties with probability distributions

Eight soil properties were considered uncertain: soil horizon thickness, clay, silt, sand, organic matter content, median particle size of the sand fraction (M50), the hydraulic conductivity and the water retention characteristic. The first six of this list are obtained from direct measurements ('basic soil properties'). The remaining two properties are derived from these basic soil properties using pedotransfer functions (Wösten et al., 1994). Organic matter content is used in GeoPEARL to calculate the Freundlich coefficient for sorption on organic matter. Clay content, sand content and organic matter content are needed for the calculation of heat transfer in soil. Most of the soil properties vary in space as well as in depth, which has to be considered when defining probability distributions. There are also cross-correlations. For instance, clay content, silt content, M50 and organic matter content partly determine the water retention characteristic and hydraulic conductivity. In addition, the water retention characteristic and hydraulic conductivity are functions rather than variables. This must also be incorporated when probability distributions are defined. The proper definition and identification of the uncertain soil inputs are therefore a complex task and substantial simplifications are made to ensure that the pdf parameters can be estimated from the limited information available. One important assumption made here that simplifies the vertical uncertainty quantification is that soil horizons are assumed perfectly homogeneous. In other words, the values of a soil property at an arbitrary location are assumed identical at all depths within the same horizon.

For a soil property at a single location and horizon, the unknown (because of uncertainty) value is represented by a random variable. Important parameters of the probability distribution function (pdf) of the random variable are its mean, which represents the expected or average value, and its standard deviation, which characterizes the variation or spread around the mean. Because the soil properties are spatially distributed and vary per horizon, the marginal (univariate) pdf must be specified for each location and horizon. Neither the mean nor the standard deviation needs to be constant in space and depth but will typically vary laterally and vertically. In this study, the locations for which calculations are done lie on a coarse square grid with a grid mesh of 9.5 km (see Section 2.3). The distances between locations are sufficiently large that spatial correlation in the uncertainty about soil properties may safely be ignored. In addition, it was also decided to ignore correlation between uncertainty in soil properties of different horizons at the same location. This greatly facilitates the uncertainty analysis, although a critical analysis of the validity of this decision and its consequences for the results of the uncertainty analysis would be sensible. To address the within-horizon heterogeneity and between-horizon correlations would require a complex 3D geostatistical analysis which was beyond the scope of this work.

The spatial variation of basic soil properties per soil horizon was determined in De Vries (1999) for all soil types of the Dutch 1:50,000 soil map given in Fig. 1. In fact, a much finer classification was used in which 330 soil classes that differ in physical–chemical characteristics were distinguished. For each of the six basic soil properties, spatial variation was summarized for each combination of 330 soil types and a maximum of 7 horizons by computing and storing the minimum, maximum and 10, 50 and 90th percentiles. Uncertainty about the soil properties is caused by the spatial variation of these properties within soil mapping units. Lack of knowledge about the distribution of the highs and lows within mapping units means that the mean or representative value is used as best guess for all locations within the mapping unit. This implies that the error or deviation from that value is dictated by the degree of spatial variation within the unit. Thus, the probability distribution associated with the soil property at an arbitrary location equals the spatial distribution of the soil property within the unit. In order to derive the marginal pdf from the minimum, maximum and three percentiles, all uncertain basic soil properties including soil horizon thickness were described with a (truncated) lognormal distribution. The lognormal distribution was used because the variables tend to be positively skewed and are not properly represented by a symmetric probability distribution. The parameters of the truncated lognormal distribution were chosen such that the median of the soil property equalled the median of the lognormal distribution and that the probability of a value greater than the 10th percentile and smaller than the 90th percentile equalled 0.80. This was achieved by choosing the mean μ_Y and standard deviation σ_Y of the log-transformed soil property such that:

$$\mu_Y = \log(q_{50}) \tag{1}$$

$$F\left(\frac{\log(q_{90}) - \log(q_{50})}{\sigma_Y}\right) - F\left(\frac{\log(q_{10}) - \log(q_{50})}{\sigma_Y}\right) = 0.80 \tag{2}$$

where q_{50} is the median, q_{10} the 10th and q_{90} the 90th percentile of the soil property and where F is the cumulative standard-normal distribution. Note that this ensures that the probability of values falling in the $q_{10} - q_{90}$ range of 'frequently observed values' equals 0.80, but does not guarantee that the probability of values smaller than q_{10} (or greater than q_{90}) equals 0.10. The resulting lognormal distribution was truncated at the specified minimum and maximum, meaning that values smaller than the minimum and greater than the maximum were assigned a zero probability density. In practice, the truncation is achieved by discarding simulated values that are smaller than the minimum or greater than the maximum (see Section 2.3).

Since soil horizon thickness is uncertain, the sum of the soil horizon thicknesses is uncertain as well and need not equal the standard fixed profile depth of 1.20 m required by GeoPEARL. This problem was solved by assuming that the thickness of the bottom horizon equals 1.20 m minus the cumulative thickness of all horizons above. In the exceptional case where this would yield a negative thickness for the bottom horizon, it was assigned a zero value and the horizon just above it was corrected to match the 1.20 m depth criterion.

Soil properties can also be cross-correlated. For instance, clay, silt and sand are negatively correlated because their sum must always equal 100%. Part of the correlations between soil properties are explained by the fact that soil properties and their associated probability

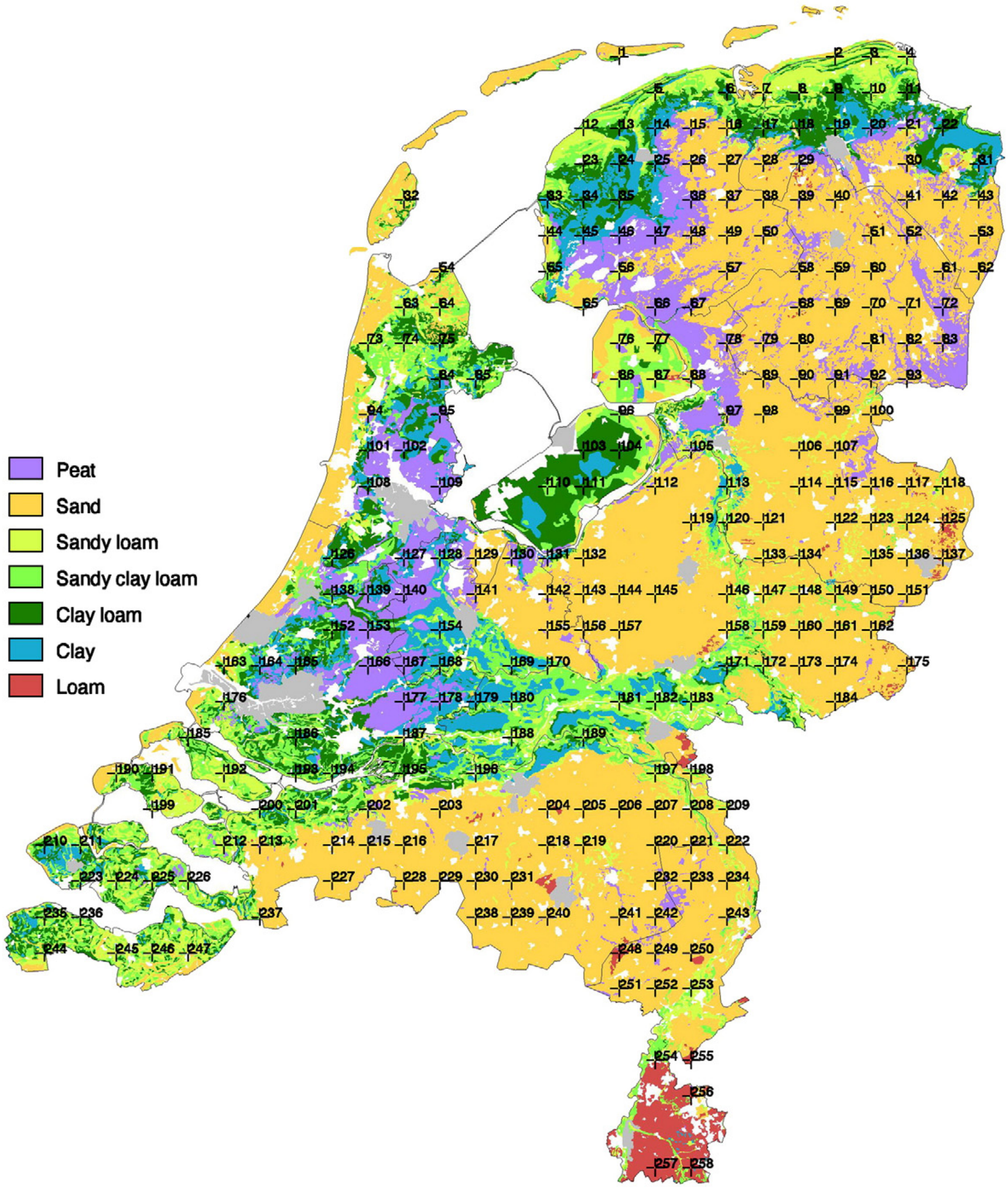

Fig. 1. Randomly placed square grid sample of 258 locations, overlaid on the Dutch 1:50,000 soil map.

distributions depend on soil type, but the deviations from the map unit representative value may also be cross-correlated. However, these within-unit cross-correlations were judged small and were therefore ignored, except for soil texture. For soil texture, sand content was simply defined as 100–clay–silt, thus creating negative correlations between sand and clay and between sand and silt.

Soil hydraulic properties and their uncertainty were derived from the so-called Staring series. The Staring series is based on a database containing 832 measured water retention and hydraulic conductivity curves of Dutch soils (Wösten et al. 1994). Each of the soil samples is allocated to one of the 36 different so-called 'building blocks', derived from a classification of soils with similar soil physical and hydraulic properties. In addition, upper horizons have different building blocks than lower horizons. The soil samples and associated curves per building block were treated as a random sample from all curves that populate the building block. Thus, the differences between curves of a building block represent the uncertainty about the 'true' curve at a location within the mapping unit corresponding with the building block. However, the variation between curves was not the only source of uncertainty in soil hydraulic parameters, because the building blocks themselves were uncertain as well. The building block at any location and horizon is determined by entering a scheme that uniquely derives the building block from parent material, texture, organic matter and M50 (Van Den Berg et al., 2008). Some of these inputs are uncertain and consequently so is the building block. This was all taken into account. The probability distribution of soil hydraulic properties is therefore partly derived from the probability distribution of the building blocks, which in turn depends on those of the basic soil properties, and partly from the random sample of curves corresponding with each building block. It is not easy to derive the exact pdfs for each of the soil hydraulic properties because of the complicated way in which these are defined, but in fact there is no need to know because all that is needed is a way to sample from the pdfs. This is much easier, because all that needs to be done is to enter the scheme with the simulated basic soil properties, derive the building block, and next randomly select a curve from the set of curves associated with it (see Section 2.3).

2.2. Probability distributions of pesticide properties

Two pesticide properties were considered uncertain: the half-life of the pesticide (DT50, days) and its coefficient of sorption to organic matter (Kom, L/kg). Both properties are pesticide-dependent and may also vary with location. In principle they may even vary with depth, but this was not considered here. The half-life of a pesticide in soil differs between soil types, but it is not known in what manner because of lack of experimental data. One of few studies quantified the effect of soil on the rate of degradation of various pesticides in 18 soils (Allen and Walker, 1987; Walker and Thompson, 1977). The degradation had a skew distribution with an average coefficient of variation of about 25%. Similar results were obtained for the sorption coefficient on organic matter (Walker and Thompson, 1977; Allen and Walker, 1987).

In this study, three hypothetical example pesticides were taken, i.e. pesticides A, B and D (FOCUS, 2000). These example pesticides cover a wide and relevant range of values for DT50 and Kom. Both were assumed lognormally distributed, with parameters given in Table 1. The coefficient of variation was taken as 25% for all cases. Note that it was assumed that DT50 and Kom are independent of soil type and soil properties. Moreover, it was also assumed that DT50 and Kom are spatially uncorrelated and not cross-correlated. These assumptions are perhaps not realistic, but lack of data enforced these simplifications.

2.3. Stochastic simulation of uncertain GeoPEARL inputs

Running the Monte Carlo uncertainty analysis at a high spatial resolution was neither feasible nor necessary. The target output (P90) is a spatial aggregate and for this reason a moderate spatial sample approximates the entire Dutch agricultural area sufficiently well. Therefore, a sample of 258 locations was selected by placing a square grid with a grid distance of 9500 m randomly over the Netherlands (Fig. 1). Gaps represent areas without agriculture. The Monte Carlo analysis was done for all 258 locations, thus effectively providing an uncertainty analysis of the PEC50 at each location (for results on the PEC50, see Van Den Berg et al., 2008). The results were then merged to analyse how uncertainties in soil and pesticide properties propagate to the P90.

The Monte Carlo analysis requires random sampling (i.e., stochastic simulation) from the pdfs of the uncertain inputs at all 258 locations (Heuvelink et al., 2007). The basic soil properties were simulated by taking the antilog of normally distributed simulations, thus yielding simulations from the lognormal distribution. Truncation was achieved simply by discarding simulated values that were greater than the maximum or smaller than the minimum and repeating simulations until the required number of simulations is reached. To sample from the pdfs of the soil hydraulic properties, first the building block was determined by entering the classification scheme mentioned in Section 2.2 with the simulated basic soil properties. Next, hydraulic conductivity and water retention characteristic curves were randomly selected from the set of curves associated with the block. In total, $N=1000$ simulations were made for each location. As an example, Fig. 2 shows histograms of the 1000 simulated values of three soil properties for two location–horizon combinations. The truncation effect is clearly visible for the clay content of the second horizon of location 208, whereas the positive skewness is evident from the histograms of organic matter.

In summary, the following steps compose the simulation of soil and pesticide properties at each of the 258 locations:

1. determine the soil mapping unit (one of the 330 classes) at the location;
2. determine the number of soil horizons in the representative soil profile description for the unit;
3. take the first soil horizon (from the top);
4. draw a value from the (truncated lognormal) probability distribution associated with the soil horizon thickness;
5. repeat step 4 for M50, silt, clay and organic matter content;
6. calculate sand by subtracting the simulated silt and clay from 100%;
7. redo sampling for realizations that have unacceptable values (i.e., values that are smaller than the minimum or greater than the maximum);
8. determine the soil building block from the simulated basic soil properties, the soil mapping unit at the location and the horizon code (i.e., upper or lower);
9. draw one sample from the group of soil samples of this building block, and determine the water retention and hydraulic conductivity curves for this sample;
10. repeat steps 4 to 9 for all remaining soil horizons;
11. sample from the pdfs of the pesticide properties;
12. repeat steps 3 to 11 as many times as required by the Monte Carlo uncertainty analysis (in this case $N=1000$ times).

Table 1
Mean values and standard deviation (SD) for the properties of the three selected pesticides.

Pesticide	Kom (L/kg)		DT50 (days)	
	Mean	SD	Mean	SD
A	60	15	60	15
B	10	2.5	20	5
D	35	8.75	20	5

3. Monte Carlo uncertainty and stochastic sensitivity analysis

The Monte Carlo uncertainty analysis runs the GeoPEARL model N times, each time using one of the N simulated set of inputs. Sample properties of the N resulting P90 values are then computed, such as the mean, standard deviation and percentiles. The standard deviation is a measure of the propagated input uncertainty. The sample properties yield only estimates of the pdf parameters of P90, because the number of Monte Carlo runs is limited (here $N=1000$ was taken, mainly for computational reasons). The sampling error can be computed and used to compute interval estimates of the properties. In case of simple random sampling as used here, the variance of the sample mean m_{P90} and sample variance S^2_{P90} are given by (Heuvelink 1998):

$$var(m_{P90}) = \frac{\sigma^2_{P90}}{N} \tag{3}$$

$$var(S^2_{P90}) = \frac{1}{N}\left(\tau^4_{P90} - \sigma^4_{P90}\cdot\frac{N-3}{N-1}\right) \tag{4}$$

where σ_{P90} is the standard deviation and τ^4_{P90} is the fourth central moment of P90. In practice, these are estimated from the sample, but this has a negligible influence when N is large.

The contribution of individual sources of uncertainty to the uncertainty of P90 was analysed using a regression-free stochastic sensitivity analysis (Jansen, 1999). The uncertainty contribution of a subset of the uncertain inputs can be expressed using the top marginal variance (TMV), also known as the percentage of the output variance accounted for by the subset (Saltelli et al., 2000). In a regression-free approach it is necessary to define independent input groups, where a

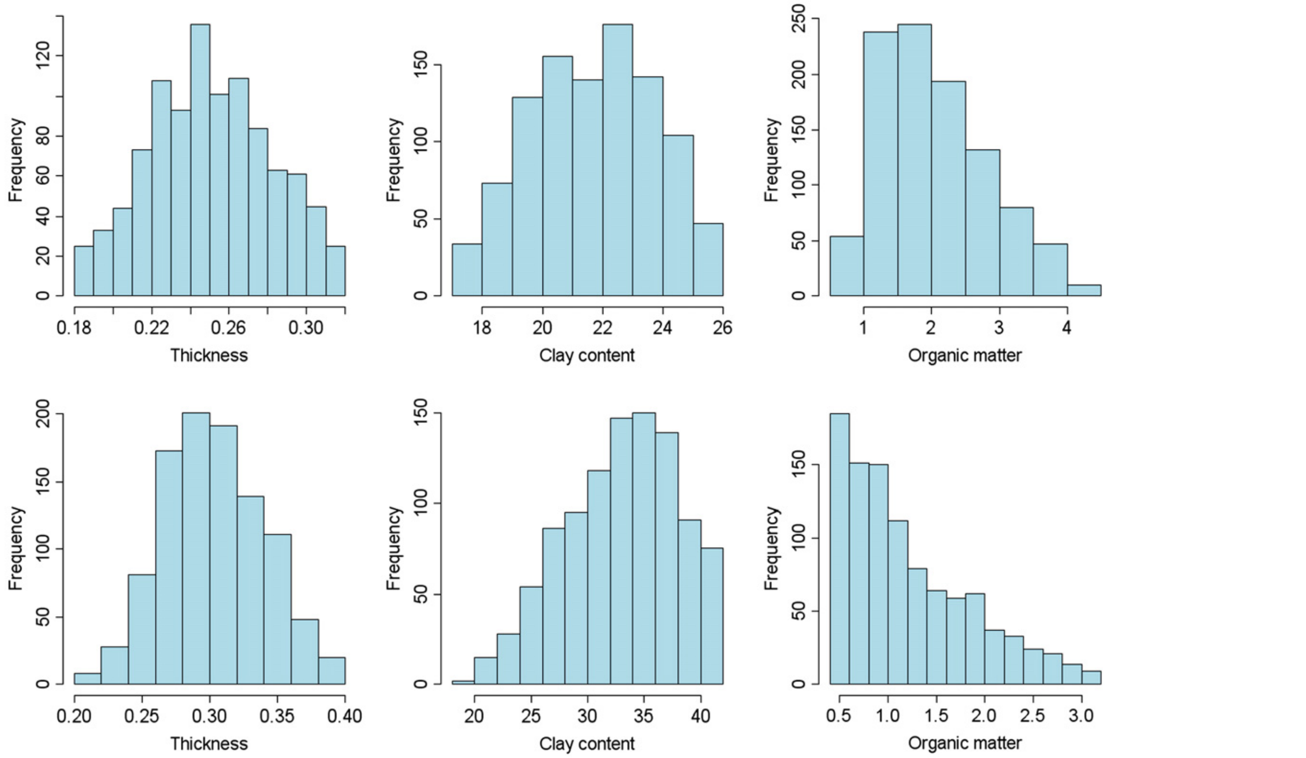

Fig. 2. Histograms of 1000 simulated realizations of thickness, clay content and organic matter content of the first horizon of location 12 (top) and the second horizon of location 208 (bottom).

group can also be one variable. In this study, four groups of independent input variables were defined:

1. organic matter content;
2. texture and soil hydraulic properties (clay, silt and sand content, median particle size of sand fraction, soil horizon thickness, water retention characteristic and hydraulic conductivity);
3. half-life of the pesticide (DT50);
4. sorption coefficient of the pesticide (Kom).

In order to assess the TMV for each group, the Monte Carlo uncertainty analysis had to be modified. The total of 1000 Monte Carlo runs was divided into five groups of 200 runs each. The first group of 200 input values are random draws from all uncertain inputs. The second group of 200 input values have the same organic matter content as the first group but are different for all other variables. The third group of 200 input values have the same texture and soil hydraulic properties as the first group but are different for all other variables. The fourth group has the same DT50 values as the first group and the fifth group has the same Kom value as the first group. This set-up was achieved by taking the 1000 random draws for all inputs and overwriting the second group of 200 values for organic matter with the first 200 organic matter values, and so on. Uncertainty contributions of each of the four groups as quantified by the TMV can then be estimated by comparison of variations between and within each of the five groups. Using bootstrap techniques (Efron and Tibshirani, 1993), the sampling error was identified and used to compute interval estimates of the TMVs.

4. Results

For each Monte Carlo run a few locations might be missing due to crashed GeoPEARL runs (e.g. this might occur when the hydrological module fails to converge in time). The number of missing locations varied between runs, and had a maximum of 16. Since the number of missing locations is small, the effect on the results was judged small.

4.1. Uncertainty analysis

Table 2 presents the mean and variance of P90 for the three pesticides and their associated sampling error standard deviations, computed with Eqs. (3) and (4). The sampling errors are small compared to the means, which indicates that $N = 1000$ runs was sufficiently large for accurate estimation of the mean and variance of P90. Fig. 3 presents the uncertainty of the P90 as box plots. The differences between the three pesticides are large. Pesticide D has the smallest mean and standard deviation, but the P90 still has a 100% probability of being above the regulatory limit of 0.1 μg/L. The uncertainty about P90 is large in all cases, particularly for pesticide A.

Fig. 4 shows the spatial cumulative frequency distribution of the PEC50, estimated from the 258 locations, both for the Monte Carlo simulations and the deterministic run (i.e., a run that assumes that none of the GeoPEARL inputs is uncertain). The P90 can be derived from this figure by taking a horizontal cross-section at the 90th percentile. For all three pesticides, the spatial cumulative frequency distribution of the PEC50 obtained with Monte Carlo simulation shows more spatial variation than the deterministic simulation, as expressed by the gentler slope of the frequency distributions. Uncertainty introduces variability and this increases the spread of the spatial distribution. It has important consequences for the P90, which is systematically shifted towards larger values. The 80% prediction zones seem not very wide, although the cross-sections at the 90th percentile yield the wide distributions presented in Fig. 3.

Table 2
Estimated mean and variance of P90 and associated sampling error standard deviations.

Pesticide	Mean of P90 (μg/L)		Variance of P90 $(\mu g/L)^2$	
	Mean	SD	Mean	SD
A	7.168	0.033	1.063	0.045
B	4.202	0.016	0.258	0.012
D	0.3922	0.0034	0.0119	0.0007

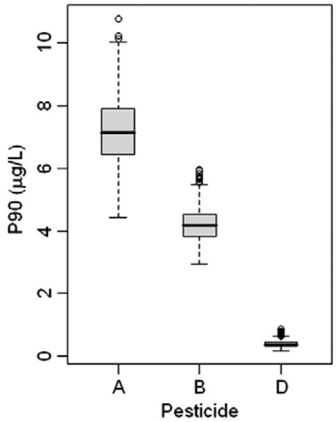

Fig. 3. Box plot of the 1000 simulated P90 values of the leaching concentrations (μg/L), for pesticides A, B and D.

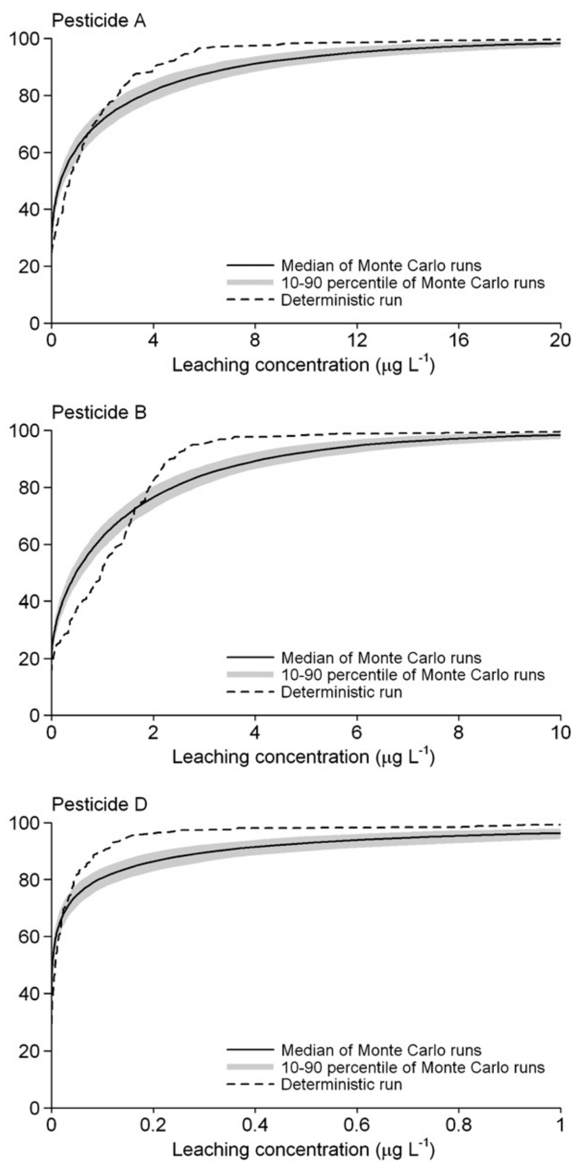

Fig. 4. Spatial cumulative frequency distribution (%) of the PEC50 for pesticides A, B and D estimated from GeoPEARL outputs at 258 locations.

4.2. Stochastic sensitivity analysis

The TMV for (groups of) soil and pesticide properties is presented in Table 3. In all three cases DT50 is the main source of uncertainty, followed by Kom and organic matter. Uncertainty in texture and soil physical properties hardly contributes to P90 uncertainty. The sum of the TMVs is much smaller than 100% in all three cases, indicating that due to interactions uncertainty contributions are not additive and TMV values must be interpreted with care (Jansen, 1999). Negative values of the bootstrap estimates of the lower confidence bound were replaced by zero. In all cases the confidence intervals are wide. More accurate estimation of the TMV requires a substantial increase of the number of Monte Carlo runs.

5. Discussion and conclusions

The uncertainty in P90 as caused by uncertainty in soil and pesticide properties is large. The width of the interquartile range is 15% of the median P90 for pesticides A and B and approximately 40% for pesticide D. Comparison with results at point locations (i.e., the PEC50) shows that spatial aggregation reduces uncertainty (Heuvelink and Pebesma, 1999; Van Den Berg et al., 2008), but the uncertainty remains large. It is therefore important to explore methods to reduce uncertainty. Since the degradation half-life is by far the most important source of uncertainty, strategies to reduce uncertainty should therefore first be directed towards obtaining better estimates of the degradation half-life and its dependence on soil type. This would require field measurements of this parameter for multiple soil types, as well as a method to quantify the effect of relevant soil properties on the degradation half-life. It is generally accepted that variability of the degradation half-life is caused to a large extent by properties that affect the microbial population (Walker and Brown, 1983). Description of pesticide degradation and sorption could benefit from experimental research into the interaction of soil properties, microbial activity and pesticides. The contributions of the sorption coefficient (Kom) and organic matter to the total uncertainty in P90 are smaller than that of the pesticide half-life, but improved estimates of these inputs will also reduce P90 uncertainty. The contribution of uncertainty in texture and soil hydraulic properties is generally small. These findings confirm results of earlier (stochastic) sensitivity analyses by Boesten (1991) and Dubus et al. (2003).

Many assumptions and simplifications were made to be able to undertake the uncertainty and stochastic sensitivity analysis. It was assumed that there was no spatial autocorrelation in the uncertainty of the soil and pesticide properties, which was justified by the large spacing of the grid. However, if an assessment was needed on the effect of uncertainty in the properties on a finer scale, smaller grid spacing would be required, so that spatial autocorrelation can no longer be ignored. Vertical spatial correlation (i.e., between soil layers) may also need to be included. This is important, because the uncertainty contribution of soil physical and hydraulic properties was underestimated in this study due to an exaggerated vertical 'averaging

Table 3
Top marginal variances (%) of P90 for four groups of uncertain inputs.

Pesticide	TMV	Organic matter	Texture and soil hydraulic properties	DT50	Kom
A	Estimate	7.4	2.8	31.9	5.3
	Lower	0.0	0.0	22.0	0.0
	Upper	17.4	13.9	42.4	17.6
B	Estimate	7.6	0.0	50.3	7.1
	Lower	0.0	0.0	39.4	0.0
	Upper	17.8	11.8	60.3	16.3
D	Estimate	4.6	3.7	37.3	9.7
	Lower	0.0	0.0	26.2	0.0
	Upper	15.0	13.3	48.3	19.7

Lower and upper are bounds of the 90% bootstrap confidence interval.

out' effect. As yet, little to no attention has been paid to this complex issue and data were lacking to investigate it. Cross-correlation between uncertain inputs was also ignored, except for soil texture. This was partly because some of the uncertainties are truly independent or because their correlation is negligibly small, but partly also because little information was available to estimate the correlations. This study also assumed that the Dutch 1:50,000 soil map was free of errors. However, it is well known that the purity of soil maps is often poor, with impurities that are typically 30% or greater (Visschers et al., 2007).

All quantitative uncertain variables included in this study were assumed to be lognormally distributed. Truncation was applied where minima and maxima were known. The choice for the lognormal distribution was inspired by the fact that many of the variables were known to be positively skewed. The lognormal distribution is flexible because it can accommodate highly skewed distributions as well as distributions that are near-symmetric. Parameter estimation and stochastic simulation are also straightforward. However, alternative distributions such as the beta-distribution for DT50 and the normal distribution for Kom have also been used in the past (e.g. Dubus et al., 2003). With little experimental data available, it is difficult to decide which parametric form is best. Analysis of the sensitivity of the results to the choice of distribution and all other assumptions made would therefore be sensible. Beulke et al. (2006) argue that user-subjectivity in Monte Carlo analyses can have a marked effect on the results of Monte Carlo uncertainty analyses.

This study also showed a systematic shift of the P90 towards greater values when uncertainty of pesticide and soil properties is taken into account. For instance, the P90 of pesticide D increased from 0.1 μg/L to approximately 0.4 μg/L when uncertainty in soil and pesticide properties is taken into account. This can be explained as follows. Each Monte Carlo run samples from the probability distribution of the uncertain properties at each of the 258 locations. Different values are drawn within a run, thus increasing the spread of the spatial distribution of PEC50. Since P90 is defined as the 90th percentile of this distribution, an increase of the spread will result in a shift away from the mean. For P90, this results in greater values. Similar results were obtained by Jury and Gruber (1989) and Van Der Zee and Boesten (1991), who analysed the effect of heterogeneities on leaching at the field scale and concluded that stochastic simulations are likely to generate extreme events that are not captured within an 'average' deterministic simulation. Leterme et al. (2007) applied the GeoPEARL model to the Dyle catchment in Belgium and also found a shift of the leaching concentration towards greater values. Although the effect is known, the implications for policy making seem not have been identified. Systematic underestimation of the P90 causes bias in the regulatory evaluation. Policy makers should be aware of this effect and take it into account when protection limits are defined.

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