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Spatial uncertainty analysis of pesticide leaching using a metamodel of PEARL

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Abstract

Spatially distributed modelling has recently been introduced in pesticide registration procedures and policy evaluations. The assessments are quite demanding with respect to computational efforts, which probably is the reason that little attention has been paid so far to uncertainty analyses of leaching concentrations at the regional scale. Recently, a metamodel of pesticide leaching was developed and calibrated with results of the spatially distributed model EuroPEARL (Tiktak, et al. 2004; Tiktak, et al. (acc.)). The metamodel is based on an analytical expression of the transport of solutes through porous media and calculates the fraction of the applied amounts which leach to a reference depth in the soil profile. Important inputs to the metamodel are the half-life and sorption constant of a pesticide, soil organic carbon content and soil moisture at field capacity, as well as long-term averages of temperature and precipitation surplus. The metamodel explained more than 90% of the variance in the simulation results and thus might be considered a good approximation of the original model. As opposed to the original model, the metamodel is very inexpensive with regard to computational efforts, which makes it suitable for calculations at high resolutions (i.e. small grid sizes) or uncertainty analyses using Monte Carlo techniques. Using the metamodel, this paper investigates the uncertainty in pesticide leaching assessments due to uncertainty in the half-life and sorption constant of pesticides and uncertainty and spatial variability in soil parameters and climatic conditions. The magnitude of uncertainty in the annual leached pesticide is computed for the temperate region of the EU and the relative contributions of individual uncertain inputs are compared. Possible implications for policy evaluations are discussed.

Keywords: groundwater, spatial correlation, error propagation, Monte Carlo simulation

1 Introduction

Leaching to groundwater is one of the key aspects in the authorization procedures of plant protection products, both at the European and the member state level. As part of the process of harmonizing evaluation procedures, standard scenarios have been defined at the European level for use in the leaching evaluation procedure (FOCUS, 2000). At the European level, the scenarios are being used to decide whether an active substance can be included in Annex I of Directive 91/414; the list of active substances which can be considered for authorization at the memberstate level (EU, 1991). The procedure evaluates whether safe use in one or more climatic zones of the European Union is possible. Use is considered safe when the calculated annual average leaching concentration, according to the FOCUS procedure[1], is below the value of 0.1 μg/L for realistic worst case conditions within the climatic zone. The soils of the FOCUS scenarios were selected to approximately represent the 80th percentile in vulnerability within a climatic zone, considering soil texture and soil organic matter content. The combination of the selected soil within the climatic zone and the calculation procedure is assumed to lead to a leaching concentration representative of the leaching concentration under realistic worst case conditions for that climatic zone. Realistic worst case conditions are now taken to represent the 90th percentile in vulnerability of all possible conditions within the climatic zone. Crops, on which the plant protection product are used, were not considered when selecting the scenario; calculations have to be performed for each crop listed on the registration application form. Properties of the plant protection product are not included in the vulnerability definition of the scenarios as these are subject to the evaluation.

In the second tier of the evaluation procedure in the Netherlands, leaching is calculated using GeoPEARL (van der Linden et al., subm.). GeoPEARL is a spatially distributed model in which the PEARL model is coupled to soil and climate databases. Calculations are performed for unique combinations of input parameters and 20 yearly applications of the plant protection product. The target quantity of the calculations is the 90th percentile of the area weighted median leaching concentrations of all unique input combinations, which are called plots. The median concentration is taken because it is less influenced by possible extreme input combinations. This 90th percentile of the leaching is considered the realistic worst case situation for the area of use of the plant protection product. For the registration, it is evaluated whether this 90th percentile is exceeding the value of 0.1 μg dm^{-3}. If not, the product can be registered with respect to leaching.

GeoPEARL is quite demanding in computational effort and therefore less suitable for use in uncertainty analyses. Recently however, a metamodel of GeoPEARL was derived (Tiktak et al, 2006). The metamodel is based on an analytical expression of the convection-dispersion-equation and explained more than 90% of the observed variance in simulation results of the original model. This and the efficiency of the metamodel with respect to computational effort raised the question whether this metamodel could be used in uncertainty analyses at regional and national level. More specifically the paper addresses:

- The influence of uncertainty and variability in the pesticide properties half-life and sorption constant, the soil properties organic matter and bulk density and the climate

[1] The FOCUS procedure involves the calculation of the leaching for a period of 20 annual, biennial or triannual application periods. The 80th percentile of the results is the target concentration which is used in the evaluation procedure.

properties temperature and precipitation on the uncertainty in the target quantity on the evaluation procedure.

- The influence of the half-life and the sorption constant on the uncertainty in the calculation results.
- The influence of moving from a data poor situation (a schematization with a low resolution) to a data rich situation (a schematization with a high resolution.

2 Materials and methods

2.1.1 The metamodel

Tiktak et al. (2006) derived a process-based metamodel of the EuroPEARL model:

$$\ln(C) = \alpha_0 - \alpha_1 \cdot X_1 - \alpha_2 \cdot X_2 \tag{1}$$

Where

$$X_1 = \frac{\mu^* \cdot \theta}{q} \tag{2}$$

$$X_2 = \frac{\mu^* \cdot \rho \cdot K}{q} \tag{3}$$

And

$$\mu^* = f_T \cdot \mu = \exp\left(\frac{E_a}{R}\left(\frac{1}{T_r} - \frac{1}{T}\right)\right) \cdot \frac{\ln(2)}{DT_{50}} \tag{4}$$

$$K = f_{om} \cdot K_{om} \tag{5}$$

Where:

C – the annual average concentration of the substance in the leachate, (μg dm^{-3})
α – a regression parameter, (-)
μ – the reference first-order transformation rate coefficient, (d^{-1})
θ – the average volumetric soil moisture content, (m^3 m^{-3})
q – the annual average water flux at the bottom of the soil profile, (m d^{-1})
ρ – the soil dry bulk density, (kg m^{-3})
f_T – function for the influence of temperature on the transformation process, (-)
E_a – the activation energy of the transformation process, (J mol^{-1})
R – the gas constant, (J mol^{-1} K^{-1})
T – the temperature with suffix r denoting reference conditions, (K)
DT_{50} – the half-life of the substance under reference conditions, (d)
f_{om} – the fraction soil organic matter, (kg kg^{-1})
K_{om} – the soil organic matter sorption coefficient of the substance, (dm^3 kg^{-1}).

This metamodel was used in this study; the parameterization of this model is described below.

All parameters, except for the regression parameters, were taken from the databases underlying the GeoPEARL model for the Netherlands and EuroPEARL model for 15 member states of the EU (Tiktak et al., 2004). The regression parameters α_0, α_1 and α_2 were derived by fitting equation 1 to the results obtained with the EuroPEARL model for 56 hypothetical substances, using S-PLUS version 6.2 (Insightful Corp.) and a robust regression algorithm (Yohai and Zamar, 1997). Default settings for the robust regression algorithm were used. Tiktak et al. (2006) divided the data into four sets representing different climatic zones, 1) warm and wet, 2) warm and dry, 3) temperate and wet, and 4) temperate and dry), were distinguished, which resulted in 4 sets of α-parameters.

For the purpose of this study the temperate zones (zones 3 and 4) were clustered and a set of α-parameters for the temperate region (temperature below 12.5 ºC) was derived (Table 1). The reason for the clustering is that for the analysis of scale effects the Netherlands was chosen as the study area. The long term average precipitation in the Netherlands varies around 800 mm, which is the division value used by Tiktak. The aim of the study of Tiktak et al. (2006) was to predict the 80th percentile of the leaching concentrations at 1 m depth. Instead of using the 80th percentile of the leaching concentrations for each plot, in this study we used the 50th percentile concentrations. This choice is consistent with the choice in Dutch pesticide registration, where also the median leaching concentration for each plot is chosen. The R^2 of the regression, 0.89, is lower than the values obtained by Tiktak, but still considered acceptable for this study.

Table 1 Metamodel regression parameters* for the temperate region of Europe, spring application.

Parameter	nominal value	standard deviation
$\alpha0$	4.33	0.022
$\alpha1$	0.0221	0.0017
$\alpha2$	0.2947	0.0011

R^2 of the regression = 0.89 (robust version as defined by Yohai and Zamar (1997).

2.1.2 Uncertainty analyses

The metamodel has three different types of input: 1) the pesticide properties DT50, Kom and activation energy, 2) the soil properties organic matter content and dry bulk density, and 3) the climatic properties temperature and precipitation surplus. Table 2 gives an overview of the parameters, their ranges and the uncertainty distribution. For the purpose of this study, we assume that the properties are uncorrelated with respect to each other and with respect to space. The latter is assumed because of the resolution of the schematisation; being greater than pesticide properties v The metamodel Sensitivity and uncertainty analysis at the point scale has revealed that the pesticide properties DT_{50} and K_{om} generally contribute most to the uncertainty in the results, but their relative contributions are dependent on their nominal values (Tiktak et al., 1994; Dubus, 2002). For this reason, in this study several nominal values were chosen to cover a wide range of these parameters (see Table 2).

Table 2 Overview of the most important properties of the pesticides considered in this study.

Parameter[a] (unit)	nominal value / range	distribution	variation coefficient
DT_{50} (d)	20 - 100	lognormal	0.25
K_{om} (dm^3 kg^{-1})	20 - 100	lognormal	0.25
E_a (J mol^{-1})	54000	normal	0.1
f_{om} (-)	map dependent	normal	0.25
θ (m^3 m^{-3})	map dependent	normal	0.21
ρ (dm^3 kg^{-1})	map dependent	normal	0.1
T (K)	map dependent	normal	0.02
q (m d^{-1})	map dependent	normal	0.04

[a] refer to the equations for a description of the parameters

The metamodel uses the long term average temperature and long term average precipitation surplus. Average values of these parameters for each of the plots were taken from the EuroPEARL results. The standard deviations of these parameters were calculated from the FOCUS Hamburg scenario weather file and water balance results of a standard FOCUS calculation for the Hamburg scenario. Variability in soil organic matter contents and soil bulk density was estimated using the soil database underlying the GeoPEARL model. For different soil types (for example: sand, silt loam, loam) the fraction soil organic matter and the dry bulk density were gathered and their means and standard deviations calculated. The average of the standard deviations was used in this study as an overall indication of the variability. DT_{50} and K_{om} are usually taken to have a lognormal distribution (Dubus et al., 2002). The values of DT_{50} should be positive and the values of K_{om} should be positive or zero. A lognormal distribution ensures this. The coefficient of variation was estimated from a few data series contained in the database of the Board for the Registration of Pesticides of the Netherlands (Dorgelo, 2006).

All analyses were performed using Matlab ® 7.0.1. Each analysis consisted of at least 1000 calculations. 1000 calculations were found to give consistent results.

2.1.3 Coarse and fine schematization on the Netherlands

The EuroPEARL output is on a grid basis of 10 by 10 km, so just over 900 grids for the Netherlands. In the schematization 24 plots are considered, each covering 2 to 208 grids. The schematization of the Netherlands in GeoPEARL is based on 6405 plots. The average size of a plot is approximately 440 ha; the minimum size is 25 ha.

3 Results and discussion

Figure 1 gives an example of the cumulative distribution of pesticide leaching concentrations. Calculations were performed for the EuroPEARL schematization of the Netherlands and all parameters were taken uncertain, except the activation energy used in the Arrhenius equation (equation 4). The target quantity for the registration in the Netherlands, that is 90^{th} percentile of the concentrations under the area of use, is approximately 7.2 µg/L when all parameters are fixed to their nominal value. Taking uncertainty into account, the calculated 90^{th} percentile goes up to a value of 11.4 µg/L, approximately three times higher. The uncertainty is indicated in the same figure; the dotted lines indicate the results plus or minus one standard deviation. It is very obvious here that there is a large uncertainty around the predicted average values. At the value of 0.1 µg/L, the critical value in the approval process, the uncertainty range is from 0.03 to 0.3 µg/L. The relative uncertainty declines with increasing concentration. For the calculation in the Netherlands

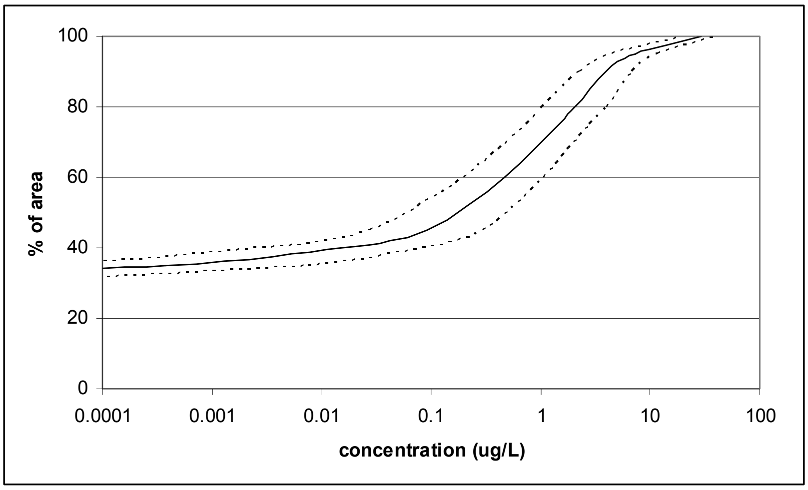

Figure 1 Cumulative distribution of leaching concentrations (µg/L) as calculated with the metamodel for the EuroPEARL schematization. Solid line: average value; dotted lines average ± standard deviation.

Table 3 compares the 90th percentile of the leaching concentrations of the deterministic case with their uncertain equivalents, for various combinations of DT_{50} and K_{om}. Taking uncertainty into account raises the 90th percentile in all cases. The relative difference becomes larger when the concentrations decrease. At the critical level of 0.1 µg/L, the difference is about a factor of two.

Table 3 Regression parameters of the metamodel for the temperate region of Europe, spring application.

Case	DT_{50}	K_{om}	P90 deterministic	P90 uncertain
1	20	60	0.0033	0.010
2	60	20	24.6	27.5
3	60	60	2.71	3.87
4	60	100	0.294	0.540
5	100	60	10.2	12.5

The relative influence of uncertainty is different for each of the parameters and also depends on the nominal value of the parameter. Table 4 gives the mean of the 90th percentile concentrations (µg/L) for when varying the parameters individually. The parameters Ea, θ, ρ, T and q have rather little influence on the calculated 90th percentile concentration. DT50 and Kom have a comparable effect; the influence of uncertainty in DT50 tends to have a larger effect than K_{om} at higher predicted concentrations whereas the opposite is true at lower concentration. The uncertainty in f_{om} shows to have the largest effect in all three cases.

Table 4 Relative influence of individual parameters.

Parameter	Case 1	Case 2	Case 3
DT_{50}	0.0037	24.9	2.77
K_{om}	0.0039	24.7	2.75
E_a	0.0032	24.6	2.69
f_{om}	0.0057	25.7	3.19
θ	0.0034	24.6	2.69
ρ	0.0033	24.6	2.69
T	0.0033	24.6	2.66
q	0.0033	24.5	2.64

Going to a situation in which input is available at a higher resolution, one expects that the uncertainty in the predicted 90th percentile of the leaching concentration will become less. Figure 2 gives the cumulative distribution of the average leaching concentrations (solid line) and the spreading around the average (dotted lines) for the same substance as shown in Figure 1. Indeed, the spreading around the average has decreased. However, the curve has flattened considerably; the range in predicted leaching concentration has grown. Very small concentrations are predicted at the left hand side of the curve (concentrations far out of the range of the graph) and higher concentrations are predicted at the right hand side of the graph. The 90th percentile is higher than in the situation with far less input data. The reasons for this shift in concentration has to be studied in more depth. Possible explanations for this are: correlation between input parameters at this higher resolution and overestimation of the uncertainty in the input parameters. These two possibilities will be studied in more depth.

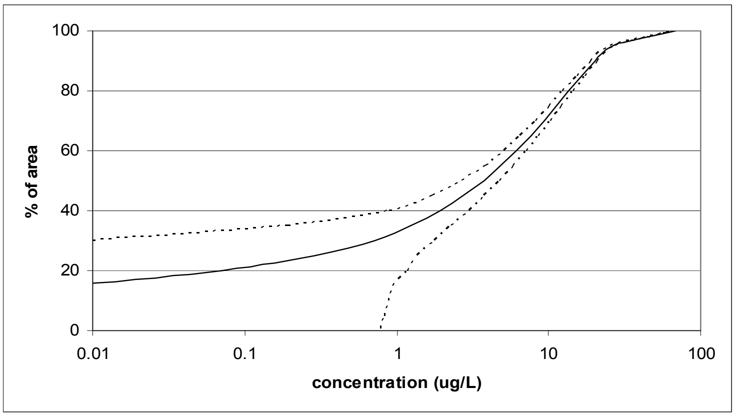

Figure 2 Cumulative distribution of leaching concentrations (µg/L) as calculated with the metamodel for the GeoPEARL schematization. Solid line: average value; dotted lines average ± standard deviation.

In the current evaluation procedures, average values for the sorption and transformation parameters are used and the result of the leaching calculations are compared to the drinking water standard. If uncertainties are taken into account in the decisions on authorization, plant protection products will fail to comply with the drinking water standard more often when the same comparison is made. If the procedure is not changed, more data, especially on the transformation and the sorption properties will be necessary, in order to lower the uncertainty.

4 Conclusions and future research

Taking uncertainty in pesticide parameters and soil parameters into account in most cases leads to higher estimates of the 90th percentile of the leaching concentrations. In this study DT50, Kom and fom showed to have the largest effects; taking into account uncertainty in all three parameters had much larger effect than each of the parameters separately. The relative contribution of the uncertainty to the leaching results was much less dependent on the nominal values of the parameters.

This study showed that a process based metamodel, which still includes the basic processes and parameters of the leaching process, can be used to investigate uncertainty. Many calculations can be performed in a relatively short time, which is a prerequisite in studying uncertainty.

Going from a data poor environment to a data rich environment resulted in less uncertainty around the calculated 90th percentile of the leaching concentrations. The 90th percentile was higher than in the data poor environment. The reasons are not yet understood. As this study is part on ongoing research, this will be studied in more detail. Next steps will include spatial correlation of soil properties and further refinement of the resolution.

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