"Assessing pesticide leaching at the regional scale : a case study for atrazine in the Dyle catchment/"

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ABSTRACT

The overall objective of this thesis is to better understand and assess pesticide leaching at the regional scale, using both the analysis of monitoring data and spatially distributed modelling. Atrazine contamination of the Brusselian aquifer (central Belgium) is poorly understood. Considerable uncertainty surrounds whether the pollution is agricultural or non-agricultural in origin. The spatial and temporal covariance of atrazine concentrations was studied by fitting semivariogram models to monitoring data. Correlation ranges were found to be 600 metres and 600-700 days. A non-parametric one-way ANOVA found a strong relationship between mean concentrations and land use, whilst other environmental variables were found to be less important. Higher levels of pollution were detected in areas dominated by urban land use suggesting that atrazine residues in groundwater resulted from non-agricultural applications. Modelling pesticide leaching at the regional scale (Dyle catchment) was used to assess groundwater vulnerability. Different approaches to process soil information were tested with both a linear (modified Attenuation Factor) and a non-linear (GeoPEARL) leaching model. The CI (calculate first, interpolate later) and IC (interpolate first, calculate later) approaches were identical for the linear model, but differences in the amount of leaching were found for the non-linear model. The CI approach would be expected to give better results than IC, but the CA (calculate alone) approach is probably the best method if no spatial output is required. Finally, a methodology was ...

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Chapter 7

Including Spatial Variability in Monte Carlo Simulations of Pesticide Leaching

The previous chapter showed that the choice of interpolating or calculating first may have significant consequences in the case of non-linear GeoPEARL simulations. However, this represents only a small part of the uncertainty associated with spatially distributed simulations of GeoPEARL. In this chapter, a stochastic methodology is proposed to adapt a classical parameter uncertainty analysis to the spatial assessment of pesticide leaching potential.

7.1 Outline

A methodology is developed to quantify the uncertainty in a pesticide leaching assessment arising from the spatial variability of non-georeferenced parameters. A Monte Carlo analysis of atrazine leaching is performed in the Dyle river catchment (Belgium) with pesticide half-life (DT50) and topsoil organic matter (OM) content as uncertain input parameters. Atrazine DT50 is taken as a non-georeferenced parameter, so that DT50 values sampled from the input distribution are randomly allocated to spatial plots for every simulation. OM content is a georeferenced parameter, so that each spatial plot has a fixed uncertainty distribution. Spatially variable DT50 values are found to have a significant influence on the amount of simulated leaching. In the stochastic simulation, concentrations exist above the regulatory level of 0.1 μg/L, but virtually no leaching occurs in the deterministic simulation.

It is axiomatic that substance parameters (DT50, sorption coefficient...) are spatially variable, but pesticide registration procedures currently ignore this fact. Including this spatial variability would have significant consequences for future registration policies, especially if risk assessments are implemented in a spatially distributed way[1].

7.2 Introduction

Leaching modelling of plant protection products (PPPs) within the soil-crop continuum is often used in the assessment of the risk of groundwater contamination by surface applied PPPs. For example, the EU registration directive EU/91/414 (European Commission, 1991) stipulates that modelled groundwater concentrations of PPP may not exceed 0.1 μg/L for a single product. Modelling procedures for groundwater risk assessment are standardised by the FOCUS working groups following a tiered approach (FOCUS, 2000). At the European level, leaching models are used in combination with a limited number of standard scenarios to make conservative predictions of PPP concentrations in groundwater, across the european agricultural areas. Four pesticide leaching models are currently used in these predictions: PEARL (Tiktak et al., 2000), MACRO (Larsbo and Jarvis, 2003), PELMO (Klein, 1995) and PRZM (Carsel et al., 1998). At the level of individual member states a variety of assessment methods are applied (e.g. van der Linden et al., 2004). In the Netherlands, for example, assessments are based on the GeoPEARL model (Tiktak et al., 2002, 2003), which couples PEARL with spatial data derived from standard Geographical Information Systems. Tiktak et al. (2004) demonstrated that a similar approach could be adopted at the pan-european level. This would take account of soil and climate variability and thus allow the identification of potential high or low risk areas. The harmonisation of pan-european risk assessment methods is currently in progress (FOCUS, 2007).

Unfortunately, uncertainty is often insufficiently addressed in risk assessment. Dubus et al. (2003b) described the different sources of uncertainty in

[1]This chapter is based on an article by Leterme B., Vanclooster M., van der Linden A.M.A., Tiktak A. and Rounsevell M.D.A.; submitted to *Environmental Science & Technology*.

pesticide fate modelling. Uncertainty is present in the primary data (physical, chemical and environmental properties), arising (i) from the spatial and temporal variability of environmental variables, (ii) from field-based sampling procedures and (iii) from laboratory analysis. There is also uncertainty in the estimation of model input parameters. This includes the direct estimation of e.g. pesticide half-life (DT50) from laboratory data, even though nearly identical results can be obtained with identical or similar boundary conditions and settings (FOCUS, 2006), and the use of indirect methods such as pedotransfer functions and/or the interpolation of spatially referenced variables (Dubus et al., 2003b; Brown and Heuvelink, 2005). The structure and the solution of the model provide additional sources of uncertainty (Addiscott et al., 1995; Brown and Heuvelink, 2005). Dubus et al. (2003b) note that typical probabilistic approaches (e.g. Monte Carlo analysis) often ignore the latter two sources of uncertainty.

Understanding the consequences of uncertainty is needed to improve risk assessment as a decision-support tool (Brown and Heuvelink, 2005). This paper addresses uncertainty in spatially distributed PPP leaching modelling. A distinction is made between georeferenced and non-georeferenced parameters. For a spatially distributed simulation, georeferenced parameters are defined as parameters that were already distributed in deterministic simulations (e.g. soil properties), while non-georeferenced parameters are those that were considered to be spatially constant (e.g. constant pesticide properties). This approach is analogous to the distinction between uncertainty and variability in second-order Monte Carlo simulations (EUFRAM, 2005; Wu and Tsang, 2004). This study differs from second-order Monte Carlo simulations in using spatially distributed parameters. At a given location georeferenced parameters are uncertain, while non-georeferenced parameters display spatial variability.

Studies examining the effect of spatial variability on PPP leaching by stochastic simulation have suggested that spatial variability can significantly affect the leached fractions of PPPs (Jury and Gruber, 1989; van der Zee and Boesten, 1991). In general, stochastic simulations are likely to generate extreme events that are not captured within an 'average', deterministic simulation. Building on this, a number of modelling studies have used Monte Carlo techniques to include the spatial variability of soil and/or pesticide properties in risk assessment methods. Earlier work focused mainly on the

variability of soil properties within soil units, while pesticide properties were kept constant (Carsel et al., 1988; Petach et al., 1991; Foussereau et al., 1993; Zhang et al., 1993). Soutter and Pannatier (1996) performed a Monte Carlo analysis on soil profiles (i.e. on point support) and then interpolated output percentiles to produce a vulnerability map.

Monte Carlo simulation was also applied to non-georeferenced parameters. Zacharias et al. (1999) developed a stochastic framework at the field scale in which multiple realizations of a hypothetical field were undertaken with the same statistical distribution of soil properties. The Monte Carlo simulation, however, considered all soil and pesticide parameters as non-georeferenced (i.e. the outputs can only be examined at the field-scale). Lindahl et al. (2005) combined georeferenced field management parameters (such as pesticide application dose) with a range of non-georeferenced parameters to study the contamination of surface water. They found that simulated values of pesticide loads reached concentrations similar to measurements as a result of summer outflows captured in the stochastic approach.

The objective of this study is to determine whether leaching at the regional scale is significantly affected by the inclusion of the spatial variability of non-georeferenced parameters. This objective has been tackled by refining, applying and evaluating a Monte Carlo approach that samples both georeferenced and non-georeferenced parameters in a spatially distributed assessment of PPP leaching. A spatially distributed stochastic simulation of atrazine (2-chloro-4-ethylamino-6-isopropylamino-1,3,5-triazine) leaching was performed for a well documented case study, with soil organic matter content (OM; georeferenced parameter) and pesticide half-live (DT50; non-georeferenced) as the uncertain input parameters. The issues of reproducibility and truncation levels of the input probability density functions were also addressed. Finally, although data availability limits direct validation, the results are discussed with respect to information about groundwater contamination in the study area.

7.3 Material and methods

The work presented in this chapter was undertaken in the part of the Dyle catchment located in the Walloon region. Chapter 3 provided a description of the study area.

7.3.1 GeoPEARL: model description and parameterisation

GeoPEARL couples the PEARL model (Tiktak et al., 2000) with spatial data derived from standard Geographical Information Systems. PEARL is a one-dimensional, dynamic, multi-layered model of the fate of pesticides and relevant transformation products in the soil-plant system. The model is linked to the Soil Water Atmosphere Plant model (SWAP; Van Dam, 2000) for the hydrology. Soil water flow is described with the Richards' equation and reference evapotranspiration is calculated with the FAO modified Penman-Monteith approach (Allen et al., 1998). Pesticide transport is simulated with the convection-dispersion equation. Sorption onto the soil solid phase is described with a Freundlich isotherm. The degradation of pesticides is described with a first-order rate equation and a number of reduction factors, which account for the influence of temperature, soil moisture and soil depth (Boesten and van der Linden, 1991).

Annual (spring) applications of atrazine were simulated for silage maize cropping. Atrazine coefficient of sorption on organic matter (K_{OM}) was assumed constant (74 L.kg^{-1}). The simulation period was fixed as 1980-2002, including six initialisation years. Annual average concentrations of atrazine at 1 m depth were extracted from the results by dividing the annual cumulative leached amount of pesticide with the annual cumulative water drainage. Climatic variability was ignored, as test simulations in the catchment showed it to be unimportant (results not shown). Over larger areas, climatic conditions become more important (e.g. Bleecker et al., 1995).

Figure 7.1 shows a flow chart of the parameterisation of OM content and texture for the stochastic simulation. Soil properties were interpolated from soil profiles and a digital soil map (scale 1:20,000). The Aardewerk database (Van Orshoven and Vandenbroucke, 1993) for Belgium consists of more than 10,000 soil profile descriptions (texture, organic matter, pH, etc.)

for the different horizons. 393 profiles with arable land use were available within the arable part of the study area. A buffer zone of 4 km around the study area was included because neighbouring soil profiles could provide valuable information. Pedotransfer functions (PTFs) were used to estimate soil input parameters not available in Aardewerk. Soil dry bulk density was calculated with the PTF of Bollen et al. (1995) and parameters of the water retention equations (van Genuchten, 1980) were derived with the PTFs of Wösten et al. (2001).

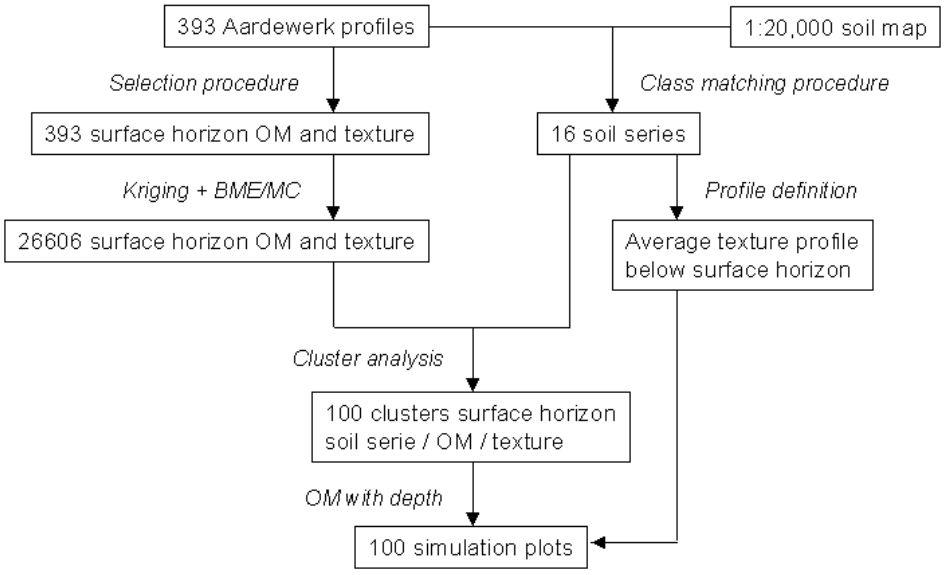


Figure 7.1: Parameterisation of OM content and texture for the simulation profiles.

Organic matter content of the surface horizon was interpolated using ordinary kriging. Topsoil OM has changed since the 1950s (when the data were collected) and so values were adjusted using a correction factor available for each soil association and land use (see van Wesemael et al., 2004). The resulting map is shown in Figure 7.2. Finally, an empirical relationship was used to determine OM content with depth (Sleutel et al., 2003):

$$\mathrm{OM}(z) = \mathrm{OM_b} + (\mathrm{OM_0} - \mathrm{OM_b}) \exp(-\mathrm{k}z) \tag{7.1}$$

where z is the depth (cm); OM_0 and OM_b are the organic matter contents in the top horizon (25 cm) and at the bottom of the soil profile (150 cm), respectively; k is a study area specific constant. The average profile of OM content and the mathematical fit are given in Figure 7.3.

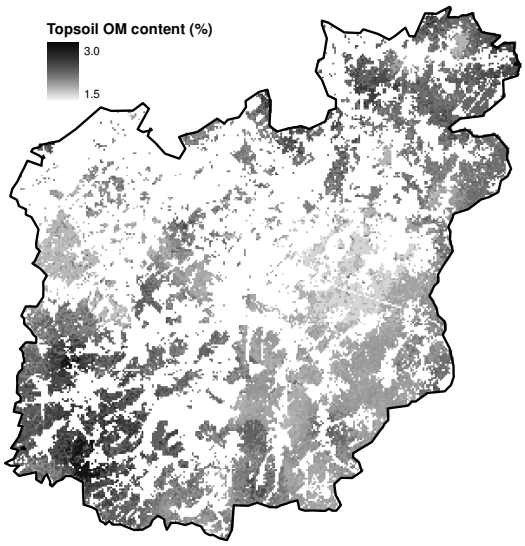


Figure 7.2: Organic matter content (%) in the surface horizon. White areas are non-arable land.

Interpolation of texture fractions in the surface horizons was based on the Bayesian maximum entropy (BME) approach, which allows the inclusion of hard (accurate) and soft (vague) data in a spatial estimation context (Christakos, 1990, 2000). Hard data were the texture fractions of Aardewerk profiles, and soft data consisted of the texture class given by the 1:20,000 soil map at every estimation location. A variant of the regular BME algorithm using a Monte Carlo procedure, called BME/MC (Bogaert and D'Or, 2002), was used because it takes into account the fundamental constraints on the textural fractions (they sum to one and belong to the $[0, 1]$ interval).

The 1:20,000 soil map was then used to define average soil profiles with depth. The Aardewerk profiles were associated with map units using a 'class-matching' procedure (see Van Orshoven, 1993). The following rules

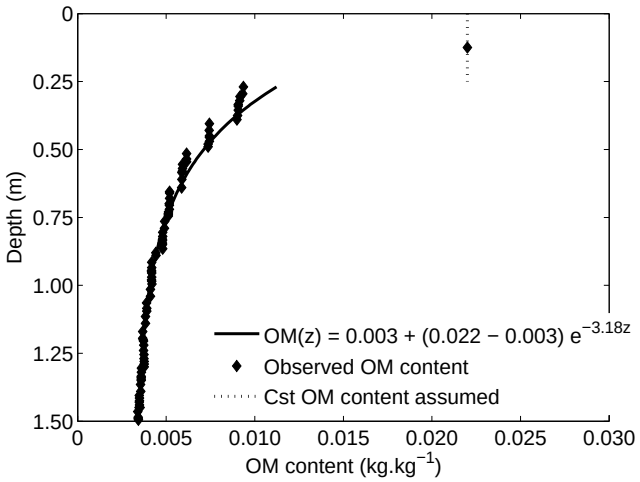


Figure 7.3: Average OM content profile observed in the study area and fit to the data from 0.25 m to the bottom of the soil profile.

were used to determine the groups of soil series needing an average profile with depth:

- Texture class (A, L, Z... from the Belgian texture triangle; Tavernier and Maréchal, 1958)
- Profile development or not (e.g. Aba >< Abp)
- Presence/absence of a sandy substrate (e.g. sAba2 >< xAba2)
- Some groups were split into further subgroups if these had a high number (>30) of Aardewerk profiles available to parameterise the subgroups (e.g. Aba → Aba and Aba(b))

This led to the definition of 16 groups of soil series. The drainage class was not taken into account because most of the Aardewerk profiles were classified as 'well-drained' and the number of profiles in other classes was not sufficient to create a new group. For each group, the available Aardewerk profiles were used to calculate average values of the texture fractions and depth to

a sandy substrate for the deeper horizons (from 25 to 150 cm). Variable horizon thicknesses were accounted for by discretising the soil profiles.

Due to the relatively small variability in OM and texture over the study area, it was possible to reduce the number of plots from 26606 to 100 using k-means cluster analysis. Test simulations proved that the results would not be significantly affected by this clustering (results not shown). This substantially reduced the run-time of the Monte Carlo analysis. The variables used in the cluster analysis were (i) the soil series group (ensuring the correct retention of these groups in the cluster analysis), (ii) OM content, (iii) silt fraction, and (iv) clay fraction, in the top horizon (r = 0.0344 between silt and clay fractions).

7.3.2 Uncertainty analysis

The uncertainty analysis was based on a Monte Carlo (MC) approach, modified to account for the spatial variability of non-georeferenced parameters. The whole procedure can be divided into five steps: (i) selection of the parameters to be included in the MC process, (ii) attribution of probability density functions (PDFs) to the stochastic parameters, (iii) sampling, (iv) automatic running of GeoPEARL, and (v) statistical evaluation of the outputs.

Selection of the parameters for the Monte Carlo procedure

To evaluate the methodology, only two parameters were included in the uncertainty analysis (one georeferenced and one non-georeferecend). The choice of the parameters was based on a previous sensitivity analysis performed on PESTLA (a predecessor of PEARL) (Dubus et al., 2003a), which found the Freundlich exponent, K_{OM} and DT50 to be the most sensitive pesticide parameters, and OM content and bulk density the most sensitive soil parameters. Thus, the analysis presented here focused, a priori, on the uncertainty of atrazine DT50 and OM content, i.e. a non-georeferenced and a georeferenced parameter respectively.

Definition of the PDFs

OM content is a georeferenced parameter, because the interpolation using ordinary kriging led to a spatially distributed characterisation of OM content. Following the cluster analysis, 100 values of OM content were assigned to the corresponding clusters. For each cluster, the uncertainty distributions around these values were obtained using block kriging. In other words, the ordinary kriging (together with other variables) allowed the clusters delineation, while block kriging performed on each cluster was used to estimate the value (and its variance) over this cluster.

DT50 is a non-georeferenced parameter, because the initial parameterisation of GeoPEARL assumed a constant value. However, it is widely recognized that pesticide DT50 values can show a large variability at the field or catchment scale (Walker and Brown, 1983; Smith et al., 1987; Charnay et al., 2005). This variability is mainly related to the properties that condition microbial activity, such as the physico-chemical environment (determined by pedoclimatic conditions) or agricultural practices (Charnay et al., 2005). Coquet et al. (2005) argued that whenever available, site-specific data should be preferred over databases to limit bias in pesticide leaching risk assessments at the catchment scale. Therefore, data from Pussemier et al. (1997) were used to assess the distribution of DT50 values. These data consist of 33 laboratory measurements of the atrazine degradation rate in loam or sandy loam soils; most located in the Dyle catchment. DT50 of less than 10 days was found in more than 60% of the soil samples. Figure 7.4 shows these data and the lognormal fit to them, together with the data of the Dutch registration dossier ($n = 27$; Dorgelo, 2006). The site-specific DT50 measurements of Pussemier et al. (1997) are much lower than the Dutch dossier data. The rapid dissipation observed by Pussemier et al. (1997) was linked to the adaptation of microbial communities resulting from repeated pretreatments with atrazine (intensive maize cropping). Such observations are excluded from the Dutch registration dossier if the effect is known to occur. Moreover, the all data collected for the Dutch registration dossier do not necessarily come from the same soil types than in the study area. The coefficients of variation for the two datasets are 117% (Pussemier et al.) and 63% (Dutch dossier). Some samples taken from fields with less frequent atrazine application produced higher DT50 values, which account for the larger coefficient of variation.

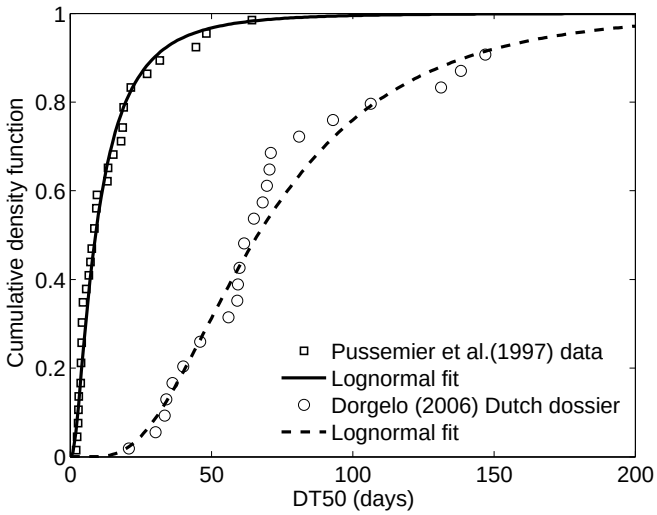


Figure 7.4: Cumulative density functions and lognormal fits of DT50 values for atrazine using site-specific data (squares; Pussemier et al., 1997) or the Dutch national database (circles; Dorgelo, 2006).

Parameter sampling

OM content and DT50 input parameters were generated using a Latin Hypercube Sampling (LHS) procedure. LHS uses a probabilistic sampling scheme that provides better coverage of the input distribution and has been shown to be more efficient than the random or stratified sampling in MC analysis (Helton and Davis, 2003).

A single MC simulation equates to one GeoPEARL simulation over the study area. This corresponds to 100 PEARL runs (one for each unique cluster) over the whole study area. The total number of PEARL runs is therefore equal to $N \times 100$, where N is the number of simulations in the MC procedure.

For each unique cluster, the input distribution of the georeferenced parameter (OM content) was sampled N times. Furthermore, for each of the N simulations, 100 values from the input distribution of the non-georeferenced parameter (DT50) were drawn with LHS and randomly allocated to the 100 unique clusters. Thus a major difference between the georeferenced and non-georeferenced parameters lies in the way their PDFs were sampled in the MC analysis.

Truncation levels were arbitrarily set to the 1^{st} and 99^{th} percentiles of the normal distributions used for OM content, and to the 5^{th} and 95^{th} percentiles of the lognormal distribution used for atrazine DT50. The justification for truncation stems from the modeller opinion that extremely small or large values sampled from the (log-)normal distributions would be unrealistic.

Automatic running of GeoPEARL

A Matlab™ (version 7.0) routine was created for the generation of LHS input parameters and these replaced the default values in the GeoPEARL input files. The N MC simulations (i.e. $N \times 100$ model runs) were processed on a grid of 256 Central Processing Units (CPUs) using batch files.

Stability of the results was tested by iteratively increasing the number of samples, because the accuracy of the MC technique is inversely proportional to the square root of the number of runs N (Brown and Heuvelink, 2005). Accuracy means here the closeness with which the MC samples represent

the joint distribution of uncertain inputs, and from which depends the level of risk associated with a decision. N was successively fixed at 10, 50, 100, 150 and 200.

Analysis of the model outputs

An indicator of atrazine leaching potential was defined as the 80^{th} percentile of the annual average concentrations at 1 m depth (for a total of 17 simulation years), i.e. for a given location in the catchment the 80^{th} percentile of leaching due to variations in weather conditions (FOCUS, 2000). This indicator could be mapped for any of the N MC simulations. An indicator of leaching risk at the catchment scale was chosen as the 80^{th} percentile in space (Vanclooster et al., 2003). Minimum, median and maximum values of the latter indicator were reported against the increase in the number N of simulations and were compared to the results of a deterministic GeoPEARL assessment with average OM and DT50 values.

Another indicator that is potentially relevant for decision makers is the area of the catchment where the regulatory limit of 0.1 μg/L is exceeded. Again, minimum, median and maximum values of this indicator were computed for increasing N to estimate the minimum required number of simulations.

7.3.3 Reproducibility and the effect of truncation

Reproducibility should be examined when undertaking uncertainty analysis. The study of reproducibility (or replicability) of MC simulations looks at the influence that the generation of a random sample with a particular seed number may have on the overall outcome of MC modelling (Dubus and Janssen, 2003). The robustness of the MC analysis was assessed for $N = 50$ by running the simulations with six different seed numbers in the LHS method. The assessment of stability was restricted to median values of model outputs, as the minimum and maximum values would be expected to vary in an unpredictable way.

A major issue concerning the influence of modeller subjectivity is the truncation level of the input PDFs (Beulke et al., 2006). The MC analysis

for $N = 50$ was repeated with truncation set to the 1^{st} and 99^{th} percentiles of the lognormal distribution for atrazine DT50. Truncation was kept to the level of the 1^{st} and 99^{th} percentiles of the normal distributions for OM content.

7.4 Results and discussion

7.4.1 Deterministic simulation

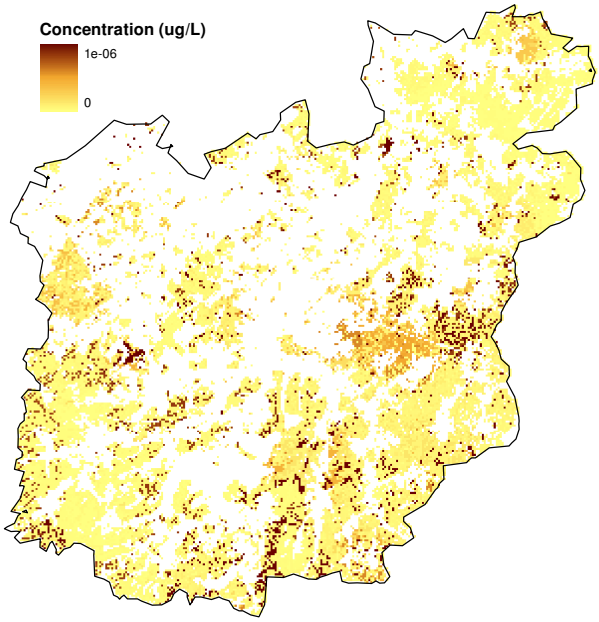
The 80^{th} percentile of the annual average concentrations at 1 m depth was lower than 1×10^{-6} μg/L for all the study area using average values for OM and DT50, i.e. several orders of magnitude below standard detection limits (Figure 7.5(a)). This result is explained by the low mean DT50 value of 8.97 days, which was derived from the distribution fitted to the data of Pussemier et al. (1997, Figure 7.4).

7.4.2 Stochastic simulation

Table 7.1 summarises the results of the Monte Carlo simulations. The two leaching indicators are significantly different than in the deterministic simulation. The median 80^{th} percentile in space is always around 0.0030 μg/L, about two orders of magnitude lower than the regulatory limit of 0.1 μg/L. The median value of this indicator appears to be stable with increasing N. Furthermore, the range between the minimum and maximum values tends to increase with a higher number of simulations (N). This could be expected because as N increases, so does the probability that the (random) spatial allocation of DT50 values to the 100 plots will produce extreme cases (e.g. high DT50 values located in areas with low OM content, leading to higher vulnerability).

Figure 7.5(b) shows the spatial pattern of leaching in the MC analysis, by mapping the maximum simulated values of the 80^{th} percentile of the annual average concentrations of atrazine (μg/L) at 1 m depth. The location of high concentrations is similar between the stochastic and deterministic simulations (Figure 7.5(a)), although the stochastic simulations give much larger absolute values. This suggests that the spatial pattern of leaching

(a)



(b)

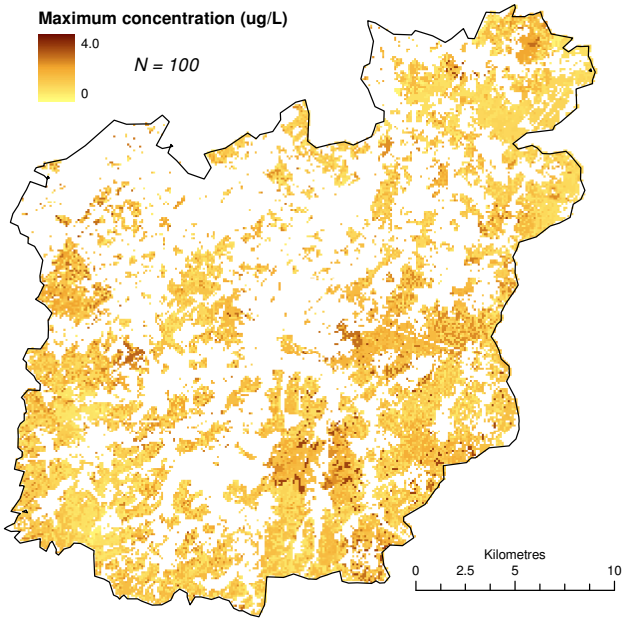


Figure 7.5: (a) 80^{th} percentile of the annual average concentrations at 1 m depth for the deterministic GeoPEARL assessment. (b) Maximum simulated values of the 80^{th} percentile of the annual average concentrations of atrazine (μg/L) at 1 m (Monte Carlo analysis; $N = 100$ simulations). White areas are non-arable land.

Table 7.1: Results of the Monte Carlo analysis.

N		10	50	100	150	200
80^{th} percentile in space (μg/L)	Min.	.0014	.0015	.0010	.0011	.0010
	Median	.0028	.0031	.0033	.0033	.0034
	Max.	.0046	.0092	.0073	.0097	.0102
Part of the study area > 0.1 μg/L (%)	Min.	6.8	6.7	5.1	6.2	5.7
	Median	9.2	8.9	8.6	8.7	8.6
	Max.	10.7	10.6	12.1	11.9	12.3

Each column corresponds to a full set a N new simulations (i.e. no serial correlation).

risk does not depend on the adoption of a probabilistic approach. The risk appears to be negatively correlated with organic matter content in the surface horizon (Figure 7.2).

Figure 7.6 displays the lower, median and upper cumulative density functions of atrazine leaching with $N = 100$. For clarity only the $[0.8-1]$ interval is shown. The 80^{th} percentile in space remains below the 0.1 μg/L limit (cf. Table 7.1). Moreover, Figure 7.6 shows the percentage of the study area above the latter threshold. The exact values are presented in Table 7.1 and vary between 5.1 and 12.1% (median = 8.6%) for $N = 100$. The median value of the area above 0.1 μg/L is also found to be relatively stable for all N. The results suggest that the median indicator values can be estimated with confidence using $N = 50$ ($= 25 \times$ number of parameters).

The simulation of atrazine leaching was strongly affected by the inclusion of spatial variability in DT50. The uncertainty in OM content (a georeferenced parameter) had a negligible influence on the results, due to a lower coefficient of variation (3.9% on average). This does not seem to support the choice of OM content as the most important georeferenced parameter. This conclusion is probably specific to the study area, where the limited variability in OM content leads to low block kriging variances.

The deterministic GeoPEARL assessment produced virtually no leaching, due mainly to the very low average DT50 value extracted from the field data of Pussemier et al. (1997). However, significant leaching was simulated in the MC analysis using a lognormal distribution of DT50. Thus, the in-

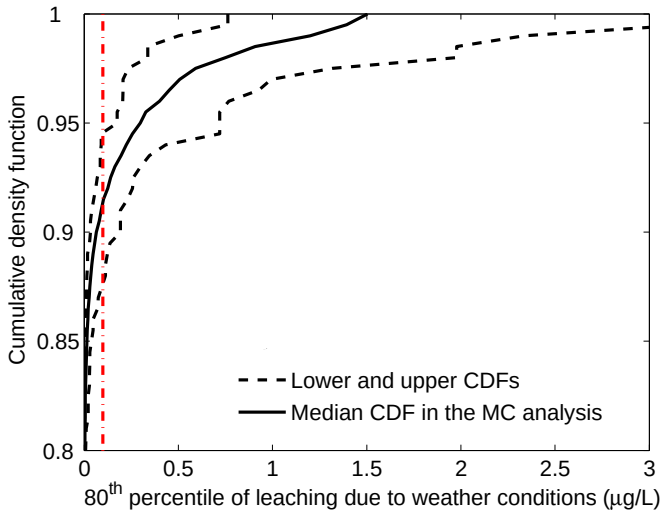


Figure 7.6: Cumulative density functions of the results of the Monte Carlo simulations with $N = 100$. The vertical dash-dotted line indicates the regulatory limit of 0.1 μg/L. Note that the y-axis starts at the 80$^{\text{th}}$ percentile.

clusion of spatial variability of any non-georeferenced parameters is likely to modify the conclusions drawn from simulations performed with spatially distributed models. In the context of pesticide registration, this finding needs to be taken into account if future risk assessments are to be implemented in a spatially distributed way.

7.4.3 Reproducibility and truncation

Table 7.2 presents the results of the simulations repeated with different seed numbers and with a different truncation of the DT50 PDF for $N = 50$. The different seed numbers did not appear to modify the examined outputs in a significant way, especially considering the median values. The 80$^{\text{th}}$ percentile in space was stable around 0.0031 and the percentage of the study area above 0.1 μg/L remained between 8.5 and 9%. These results suggest that the MC analysis was robust with respect to the choice of the seed number.

Table 7.2: Reproducibility of the Monte Carlo analysis and the effect of truncation ($N = 50$).

Seed number		1	2	3	4	5	6	1
Percentiles of PDF truncation for DT50		5-95	5-95	5-95	5-95	5-95	5-95	1-99
80^{th} percentile in space (μg/L)	Min.	.0015	.0014	.0013	.0016	.0015	.0015	.0030
	Median	.0031	.0035	.0029	.0031	.0034	.0032	.0071
	Max.	.0092	.0087	.0070	.0062	.0077	.0066	.0182
Part of the catchment with > 0.1 μg/L (%)	Min.	6.7	5.7	7.0	6.1	6.1	6.0	9.2
	Median	8.9	8.5	8.6	8.4	8.8	8.8	12.0
	Max.	10.6	11.1	11.2	10.1	11.0	10.8	15.2

Table 7.2 also gives the results of the MC analysis using a lower level of truncation in the sampling of the DT50 PDF. When the 1^{st} and 99^{th} percentiles of the lognormal distribution were chosen, the sampled values of atrazine DT50 reached a maximum at about 78 days, while the maximum was at about 41 days for the initial configuration (truncation at the 5^{th} and 95^{th} percentiles). This led to a significant increase in the amount of simulated pesticide leaching. The 80^{th} percentile in space doubled and the area above 0.1 μg/L increased to 12%.

This finding illustrates the importance of the subjective choices made by the modeller in typical MC analyses, which introduce variability in the probabilistic modelling. Beulke et al. (2006) argued that user subjectivity biases truncation, type and parameterisation of the distributions, correlation between parameters, methods for sampling and size of random samples. In the present study, truncation of the input PDFs clearly had an impact on the results, but it is rather difficult to define truncation thresholds in an objective way. However, performing the Monte Carlo analysis with a different level of truncation for atrazine DT50 provided information on the impact of this choice on the simulation outputs.

7.4.4 Field samples vs. databases.

Coquet et al. (2005) recommended field-sample measurements rather than databases to parameterise the distribution of DT50 and this was found to be important here. Using DT50 data from the Dutch registration dossier led to concentrations above 0.1 μg/L for more than 80% of the study area, in all MC simulations with $N = 50$ (results not shown). However, the contamination of groundwater by atrazine is believed to be mainly from non-agricultural sources (CERVA, 2004; Leterme et al., 2006a). Thus, the DT50 of Pussemier et al. (1997) produced more realistic simulations i.e. rarely exceeding 0.1 μg/L for the agricultural sources. This supports the appropriateness of field-sample measurements when deriving pesticide parameters. It should be noted, however, that model error was not assessed in this study (among other sources of uncertainty), e.g. GeoPEARL does not account for preferential flow. A good match of modelled and field observations would, therefore, not be expected.