

Chapter 9

Conclusions and Perspectives

9.1 Main findings

This thesis has focused on the assessment of groundwater vulnerability to contamination by pesticides, in the Walloon part of the Dyle river catchment. A review of the literature suggested that methods using process-based models have the best potential to meet current requirements in the assessment of pesticide leaching risk. It also emphasized a number of important issues associated with spatially distributed modelling of pesticide leaching, mainly related to scale and uncertainty.

Our first main objective was to determine what information could be obtained about the processes of groundwater contamination by atrazine in the study area, through a complete analysis of conventional monitoring data. The analysis was restricted to atrazine because of a lack of data for other pesticides. Despite the good availability of data describing atrazine concentrations within the study area, relatively little was known about atrazine pollution at the scale of the Brusselian aquifer.

In chapter 4, we attained our first specific objective by showing that the source of atrazine contamination in the Brusselian aquifer was non-agricultural. The application of a declustering algorithm and a non-parametric analysis of variance allowed the establishment of a link between atrazine concentrations in groundwater and land use at the soil surface. Higher levels of pollution were detected in areas dominated by urban land use suggesting that atrazine residues in groundwater resulted from non-agricultural applications (urban weed control programmes, individual use in gardens, etc.). These conclusions are consistent with a modelling study

performed by Loague and Soutter (2006), who showed that leaching from non-point sources are unlikely to be the cause of contamination hot spots.

Although certain well-documented cases had already shown the importance of non-agricultural pesticide contamination, this thesis has extended the analysis of atrazine contamination to the entire Brusselian aquifer. Differences in application rates between farmers and ‘urban’ atrazine users could explain the predominance of non-agricultural sources (Lapworth and Gooddy, 2006)¹. Differences in the physical environment may also play a role. For instance, urban settings are mostly located in the valleys bottom, where the relatively protective loess layer is generally eroded (cf. soil map in chapter 3—Figure 3.4), although available data did not permit such a relationship to be detected in the one-way analysis of variance performed in chapter 4. Other local characteristics, for example organic matter content on the exact sites of non-agricultural applications, probably play a part but these data are difficult to collect at the regional scale.

Another specific objective related to the analysis of monitoring data was to describe the spatial and temporal dynamics of atrazine concentrations in the aquifer, using geostatistics. Correlation ranges of 600 m and 600-700 days were fitted to the monitoring data. This provided valuable insight into the possible residence time of atrazine in the aquifer, knowing that virtually no degradation occurs in saturated conditions. It was thus suggested that the observed pollution is likely to persist at a significant level for at least several years after the ban on atrazine inputs (in effect since September 2005).

Our second main objective was to assess pesticide leaching potential from agricultural use in the Dyle catchment. This objective was fulfilled in chapters 6 to 8 by the production of several vulnerability maps based on the assessment of atrazine leaching potential.

¹Farmers are generally constrained by economic considerations that limit excessive pesticide usage, whereas non-agricultural applications often follow incorrect application rates or timing. Cost considerations are less important for occasional, non-agricultural uses compared to the large quantities applied by farmers. Also, urban pesticide users are generally less informed than farmers and may therefore be inclined to think that ‘a little more than the dose’ will ensure perfect results. Furthermore, when private companies are engaged to apply pesticides on public or private property, they have no rigorous incentive to limit pesticide dosage as these costs are passed onto the client. These hypotheses remain untested, but could be addressed through detailed user interviews.

However, a more specific objective of this thesis was to determine the advantages and consequences of different modelling approaches, in terms of spatialisation of point information—CA (calculate alone), CI and IC (calculate or interpolate first). Chapter 5 presented the three methods in a simple, synthetic case study. Chapter 6 applied these concepts to a real case study, comparing the results for two pesticide leaching models (a linearised version of AF, and GeoPEARL). The CI and IC approaches were found to be identical for a linear model, but significant differences in the amount of leaching were evident for the non-linear model. The CI approach would be expected to give better results than the IC approach, especially if the uncertainty about the interpolated input values is not processed in the model run. However, in practice, the IC approach would be easier to implement in an uncertainty analysis because fitting semivariogram models and interpolation can hardly be automated in, for example, a Monte Carlo analysis. In the context of decision making, the CA approach is probably the best method if no spatial output is required, considering that CI results will not contain more information than CA when computing global estimates. The CA approach has the advantage that no regression towards the mean is made, but the output is not available in a spatially distributed way (unless appropriate ancillary information—typically a soil map—is available).

A further specific objective was to apply a probabilistic methodology for the assessment of leaching potential in the study area. The IC approach was chosen, because the uncertainty analysis mainly addressed the ‘C’ step (model run). Indeed, the CI approach would have been hard to implement, requiring the interpolation step to be repeated manually a large number of times. The methodology developed in these chapters focused on the distinction between georeferenced and non-georeferenced parameters. Soil properties were considered to be georeferenced, because they were spatially distributed following interpolation. Pesticide properties (and dispersion length) were considered to be non-georeferenced, because no information is available about their spatial distribution. The main issue was to determine whether leaching at the regional scale is significantly affected by the inclusion of the spatial variability of non-georeferenced parameters. In chapters 7 and 8, a probabilistic assessment of atrazine leaching potential was undertaken in the Dyle river catchment using Monte Carlo simulations. It was shown that introducing spatial variability of non-georeferenced pa-

rameters greatly affected the simulation of pesticide leaching potential. In particular, this is the first time (to our knowledge) that pesticide half-life (DT50) and sorption parameters (K_{OM} and Freundlich exponent) were simulated with a given spatial distribution without complete aggregation of the model outputs.

The spatial pattern of leaching was not strongly affected, but the simulated concentrations increased significantly when looking at the 80th percentile, as in current registration procedures. That upper percentiles increase as more variability is introduced into the uncertainty analysis was to be expected, but this study has been able to quantify the extent of this increase. The distribution of atrazine DT50 in the stochastic simulations was estimated using site-specific data and this had a profound influence on the modelling results. The importance of DT50 parameterisation was underlined by the sensitivity analyses performed on the simulation results. This parameter was found to be the most sensitive in the MC analysis, although interaction between parameters was also believed to play an important role.

A final objective was to assess whether existing groundwater monitoring data could be used for the validation of leaching modelling at the regional scale. Here a major issue raised from chapter 4 conclusions about the origin of groundwater contamination by atrazine in the study area. Indeed, atrazine monitoring data cannot reasonably be used to validate the assessment of leaching from agricultural sources if it is accepted that most of the pollution results from non-agricultural applications. This issue is further developed in section 9.3 below.

9.2 Implications for science and policy

The management of water resources in the European Union today is influenced increasingly by the guidelines of the Water Framework Directive (WFD; EU, 2000), although the 91/414/EEC Directive (European Commission, 1991) is still important for the management of groundwater contamination by pesticides. The member states have been translating the WFD regulatory aspects into their own legislations, and common environmental objectives (water quality, quantity...) are defined. For instance, one of the most cited quote of the WFD states that (ground)water resources have

to reach a ‘good chemical status’ by 2015. To achieve good groundwater chemical status, concentrations of pollutants should not exceed any of the quality standards (e.g. $0.1 \mu\text{g/L}$ for pesticides).

However, this thesis has shown that the time lags necessary for the dilution of pollutants in groundwater may hamper the achievement of good chemical status within the period defined by the WFD. While pollution events often occur as short-lived peaks in surface waters, correlation ranges found for atrazine concentrations in a sandy aquifer indicate that groundwater contamination (where it is observed today) is likely to persist over many years. This should be taken into consideration when evaluating the efficiency of policies to reduce groundwater contamination.

The conclusions drawn from chapter 4 also indicate the need for greater controls on non-agricultural applications of pesticides. Although non-agricultural use of atrazine represents approximately only 25% of total applications (at the time this substance was authorised; CERVA, 2004), this study has shown it to be an important cause of groundwater contamination. Similar conclusions have been suggested for other compounds, such as simazine (CERVA, 2004). Clearly there is need of better procedures for tracing pesticides from the producer to the final user. In this respect, society would certainly benefit from open access to pesticide sales and application data, which are currently confidential. This lack of transparency probably favours illegal applications of atrazine (suggested by occasional pollution peaks in surface water; Debongnie, pers. comm.) since this pesticide was banned for non-agricultural uses.

In the case of pesticides used for both agricultural and non-agricultural applications, the development of scenarios to assess the risk of leaching from non-agricultural applications could be considered. These scenarios would be constructed after a thorough investigation of application rates, surface types, timing, etc. Analysis of these scenarios could be imposed for the most problematic substances, for example before the renewal of registration licences. Deciding which compounds should be tested could be made on a national basis, using monitoring data to assess the frequency that regulatory thresholds are surpassed or based on the occurrence of contamination events for which evidence of contamination from non-agricultural source exists.

The registration of new compounds is currently regulated by the 91/414/

EEC Directive and the FOCUS group have elaborated a number of procedures to implement this Directive (FOCUS, 1995, 2000, 2006). Scientists generally agree that decision making processes should evolve towards the adoption of probabilistic approaches for pesticide leaching modelling (EU-FRAM, 2005). This thesis has presented a stochastic methodology for the spatially distributed assessment of pesticide leaching risk. Both spatial variability (of non-georeferenced parameters) and uncertainty (of georeferenced parameters) have been taken into account, in a way that is analogous to the concepts developed in second-order Monte Carlo analyses. The main advantage of the approach applied here is that it generates a detailed (depending on the scale of the study) spatial outcome and allows a relative assessment of the spatial distribution of risk.

This thesis has also stressed the importance of the choices made for the parameterisation of DT50 and K_{OM} distributions. Current registration procedures use a constant value for these parameters, although their properties have been shown to display considerable spatial variability. Even if no information is available to precisely locate high or low DT50 areas, it has been shown that using a constant value of DT50 may lead to erroneous conclusions about the risk of pesticide leaching. In this context, environmental policies may be forced to adopt stochastic procedures to implement more realistic spatial vulnerability assessments.

When available, site-specific data on pesticide degradation and sorption may further affect the results of risk assessments. For example, microbial adaptation has been demonstrated to lead to rapid degradation of atrazine in the study area (Pussemier et al., 1997). Significant differences in pesticide leaching assessments may then arise, depending on the source used for the parameterisation of DT50 variability (site-specific data vs. generic databases).

These two points highlight the need to find a balance between harmonised procedures and realistic assumptions. Registration procedures may be put into question if risk assessments are contradicted by real observations. In this respect, this thesis has shown that it may be relevant to adopt stochastic approaches that take account of spatial variability of the most sensitive parameters (e.g. DT50 and K_{OM}). There is a trend in undertaking more realistic leaching assessments, particularly in the correct representation of spatial variability (Beven, 2001a), but a compromise has yet to be

reached as ‘to what extent’ this is feasible. For instance, trying to include crop rotations in pesticide leaching assessment did not seem appropriate in our case because it would probably have just added noise to the simulated spatial pattern due to insufficient data.

Finally, the fact that non-georeferenced parameters were found to be the most sensitive parameters explaining the results of the stochastic simulations does not diminish the value of spatially distributed modelling. Indeed, the study area was characterised by a low variability of soil texture properties, and the conclusions about the relative importance of non-georeferenced (pesticide) and georeferenced (soil) parameters may therefore be very different in larger or more heterogeneous areas.

9.3 Limitations of the study and perspectives for future research

In spite of the achievements described previously, this study has several limitations.

First, the information gained from the analysis of historical monitoring data was restricted to atrazine, due to data availability. Conclusions about the spatio-temporal dynamics of pesticide concentrations in groundwater would have benefited from the study of other compounds. It is not known to what extent the conclusions about residence time and dilution are specific to atrazine.

As additional data become available in the future, especially with enhanced monitoring networks required by the WFD, it may be possible to apply the geostatistical tools presented in this thesis to other pesticides. For the main types of groundwater bodies (classified after their hydrogeological characteristics), monitoring data should be collected at an increasingly finer spatio-temporal resolution, until general spatial and temporal correlation ranges are identified. The frequency and density of routine sampling could then be defined from these results. This may provide important information in better defining reasonable goals for the implementation of the WFD, and ‘good quality’ objectives in particular.

More data are also needed about the vertical distribution of pesticide

concentrations in the saturated zone because the extent of vertical dilution is still largely unknown at the scale of the aquifer. Finally, where technically possible, sample mixing from the individual wells of a monitoring station should be avoided because this removes valuable information that could be gained from this configuration.

However, it remains uncertain whether a sufficient number of measurements above the detection limit will be collected, as atrazine has been known to be the most problematic substance in the study area. Here lies a disturbing paradox for scientists: groundwater contamination is sometimes ‘useful’ to understand the processes involved, and eventually help to prevent further pollution. . .

Although valuable information for the delineation of well protection zones was derived from the correlation ranges, these results are highly dependent on the local hydrogeological conditions. The Brusselian aquifer is a sandy, unconfined, and quite homogeneous groundwater body. If conducted over a wider (e.g. national) area, very different hydrogeological conditions (karst systems, confining beds. . .) would likely blur the spatial and temporal patterns of pesticide concentrations. Thus, the methodology used in this thesis is probably applicable only to aquifers expected to display ‘smooth’ patterns of groundwater flow variation and connectivity.

A second limitation of this work appears when confronting the two parts of the thesis. On the one hand, the analysis of historical monitoring data showed that groundwater contamination by atrazine is from non-agricultural source. On the other hand, the complete modelling study and vulnerability assessment were based on the simulation of atrazine leaching from agricultural sources only.

So, how can we explain this apparent contradiction? Without exception, all leaching models have been developed for the simulation of pesticide fate deriving from applications to agricultural land (arable land use and pasture). The main reason for this is that agricultural use accounts for greater pesticide loads than non-agricultural use (lower dosage, but over a much larger area). Parallel to the development of pesticide leaching models, registration procedures for new substances have increasingly relied on the use of those models. Indeed, more attention is devoted to potential public exposure to pesticides through the consumption of agricultural goods. Also, the

laboratory and field experiments necessary for the development of leaching models are more easily controlled in agricultural conditions than elsewhere and have a better potential for generalisation and replicability (application rate and timing is more easily monitored, etc.). The large variability in non-agricultural applications makes them difficult to simplify and represent in process-based models in an efficient way.

Therefore, alternative approaches are needed to take non-agricultural use of pesticides into account in vulnerability assessments. A possible way of doing this is the adaptation of existing models to specific non-agricultural applications, provided that sufficient data are available for model parameterisation. For example, process-based models have been applied to study leaching in railway embankments (e.g. Jarvis et al., 2006). However, these developments remain marginal because different (unknown or unpredictable) processes are believed to occur in such applications. Trying to calibrate ‘classical’ models to a monitored experiment in railway embankments, Jarvis et al. (2006) identified long-term retention/sorption process not included in any of the tested models, and also suggested the formation of ‘protected’ residues.

To include urban areas in vulnerability assessments, Ducommun and Zwahlen (2006) proposed to adapt models developed for the simulation of pesticide transport in karstic environments. In any case, integrated vulnerability assessments should make use of advanced mapping tools to merge both agricultural and non-agricultural sources (e.g. fuzzy logic, or GIS-based models as in Thomas and Tellam, 2006). However, validation of such approaches at a fine resolution will certainly remain very difficult, as a multitude of non-locatable point sources may exist in the capture zone of a single well.

The discrepancy between models applied exclusively on agricultural land and actual groundwater contamination from different sources has already been pinpointed as a major obstacle to the validation of pesticide leaching models at the regional scale (Holman et al., 2004). This highlights the next limitation identified in this thesis, namely the absence of model validation.

Although it has been claimed that the validation of environmental models is an impossible task (see section 2.2.3 in chapter 2), the GeoPEARL simulations presented in this thesis were not confronted with monitoring

data. Although we have discussed the relative agreement between simulations and observations by comparison of the orders of magnitude of atrazine concentrations, no detailed comparison was made of the spatial patterns of leaching. As Beven (2001a) noted, there are few assessments of distributed models that have included spatially distributed observations in either calibration or evaluation. The main problem is the mismatch between model predictions and available observations. The present work focused on atrazine concentrations at the bottom of the soil profile, while monitoring data are derived from wells, galleries or springs. Similarly, measured data are point samples in time rather than annual average concentrations. There is also a considerable variability in sampling frequency between different monitoring stations.

Finding a framework for the evaluation (or validation) of leaching studies at the regional scale is indubitably a major challenge in hydrological science. The main difficulty is that the output of model simulations is much less accessible for direct measurement in the field than for example pesticide loads to surface waters. Remote sensing techniques do not provide a solution, as they are limited to the first few centimetres of the soil surface. Monitoring data obtained from wells have already undergone mixing and dilution in groundwater. Therefore, pesticide leaching models should be coupled to distributed groundwater models for a robust comparison of model predictions and monitoring data. This is not always feasible at the regional scale, given the amount of data required to apply such models, and in any case such a simulation would not validate the pesticide leaching model alone.

We believe that a first step is to implement model evaluation in the form of relative comparisons (i.e. the examination of spatial patterns). Vulnerability maps may for example be compared to point monitoring data using a score matrix to quantify the correspondence between simulations and observations (Sulmon et al., 2006). If the zones with a high potential for leaching correspond to the sites of actual groundwater contamination this is relevant for the model evaluation. However, as explained previously, this seems inadequate in the case of atrazine because of the discrepancy between non-agricultural contamination of the aquifer and model simulations on agricultural land use.

This thesis has focused on atrazine concentrations in leaching, but a classical approach is to include other variables in the model validation. For

example, groundwater recharge rate is often used to increase the number of evaluation measures.

Another possibility is to extend the model evaluation to other substances, focusing by preference on pesticides used only for agricultural applications. Other substances may behave differently (pH dependence of sorption, etc.), but the main conclusions of this thesis concerning the modelling study should be similar. If the simulated relative ranking of pesticides after their leaching potentials corresponds to observed contamination levels in the aquifer, this comparison would enhance the confidence in the vulnerability assessment. However, to apply this strategy, more monitoring data than are currently available would be needed, to allow significant statistical comparisons.

Following model simulations, the areas of highest and lowest leaching potential could easily be identified. Pesticide concentrations at the bottom of the soil profile could then be measured in these areas, provided that similar boundary conditions apply (e.g. crop rotation pattern, dosage...). For atrazine, this is no longer relevant since the ban on atrazine inputs, but this approach may be useful for other compounds. If significant differences in leaching potential were confirmed by field measurements, this approach would provide a positive evaluation of model simulations.

We believe that making an absolute comparison of pesticide concentrations in leaching at the regional scale is irrelevant because it is almost impossible to obtain sufficient data for an exact, realistic parameterisation. In our case study, for instance, detailed information was not available for the patterns of crop rotation. Adding to this the problem of the confidentiality of pesticide sales and applications data, and it soon becomes clear that simulations replicating the reality of atrazine applications are not conceivable.

A further shortcoming concerns the uncertainty analysis. The Monte Carlo simulations dealt mainly with the uncertainty in model input parameters, while other sources of uncertainty such as model error were ignored. Beven (2005) insisted on the difficulty of separating the different sources of errors in non-linear dynamic cases and there is currently no general theory available for doing this.

A frequent method is to perform validation tests in which the failures of model predictions are declared as model structural or conceptual errors. However, model error can thus only be assessed with respect to the output

variables for which field data are available (Højberg and Refsgaard, 2005). Another approach comprises the comparison between different models, but this says nothing about the validity of model simulations compared to reality. Models with structural errors could agree with the validation data if these errors were (at least partly) compensated for by other sources of uncertainty. This leads us back to the issue of equifinality (see section 2.2.4.1 in chapter 2), in which many different model structures and parameter sets may be accepted as ‘behavioural’ (Beven, 2001a).

For example, GeoPEARL (in the version used for this thesis) certainly introduced model conceptual error by ignoring preferential flow processes. Nevertheless, simulation results could be judged as ‘behavioural’, because very low amounts of atrazine leaching were modelled, agreeing with the information derived from monitoring data. This evaluation is however based on a very loose validation, so additional monitoring data or simulations with different boundary conditions could undermine this statement.

Finally, the uncertainty associated with interpolation (as examined in chapters 4 and 5) was not included in the MC analysis of chapters 6 and 7—except in the parameterisation of the uncertainty of OM content, which was derived from block kriging variance. Indeed, doing so would engender high computation costs. In this respect, the substitution of GeoPEARL simulations by a metamodel would be an interesting avenue of further research, provided that a metamodel could represent the system with sufficient accuracy. This would allow the inclusion of more parameters in the stochastic simulations as well as the incorporation of different modelling approaches (e.g. CI and IC) or model structures in the model space of Beven (2001a).

9.4 Final remarks

Monitoring data and sales projections indicate that pesticides will be present in our environment for the foreseeable future. Past applications have already contaminated many groundwater resources, sometimes at a level assuring long-term pollution. We consider therefore that investigating pesticide fate in the environment and preventing further leaching of pesticide residues is a societal duty.

In this context, the Water Framework Directive and its offshot the

Groundwater Directive provide major opportunities for the European research community, notably as more data become available with the enhancement of monitoring networks and strategies. Moreover, the objectives defined by the WFD should be perceived as stimulating challenges to improve groundwater quality.

Finally, the development of more efficient procedures for the registration of new compounds is an essential aspect of groundwater management and protection. In this respect, the effect of the complexity of processes determining the environmental fate of pesticides must not be underestimated, as illustrated in this thesis by the consequences of spatial variability in pesticide properties. Assuming it is not laced with uncertainty, this complexity should be integrated in registration procedures.

