"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 1

Introduction

1.1 General introduction

The expansion and intensification of agriculture has been a predominant feature of the twentieth century. The Green Revolution is estimated to have doubled food production since the 1960s (Chrispeels, 2000). Intensification of agriculture has meant the use of high-yielding crop varieties, fertilization, irrigation, and pesticides. This tremendous increase was, however, not sufficient as an estimated eight hundred million people on earth are poor and malnourished[1] (Chrispeels, 2000).

At the same time, our society is confronted with a number of harmful consequences of agriculture intensification on the environment. Major areas of concern include biodiversity losses, decreasing soil productivity, soil erosion and salinization, and water contamination by nutrients and pesticides (Matson and Parton, 1997). Thus, the pressure on the environmental resources may threaten our ability to produce sufficient food and may also worsen the quality levels of air, soil and water[2].

This thesis addresses more specifically the issue of groundwater contamination by plant protection products—commonly known as pesticides.

[1]Today, the defenders of genetically modified crops call for a 'Doubly Green Revolution' (Conway, 1999; Huang et al., 2002) to meet future food demand, while their opponants claim that better redistribution of the surplus and communities empowerment could suffice.

[2]For example, global pesticide production ($3.75\ 10^6$ tons in 2000) has increased for the past 40 years, and projections for 2050 range from 1.9- to 4.8-fold increase (Tilman, 2001). Should these trends be verified, by 2050, humans and other living organisms in various ecosystems would be exposed to markedly high levels of pesticides.

Pesticides are used not only to sustain agricultural production, but also in a variety of non-agricultural applications (e.g. railway track treatment, urban weed control programmes, individual use in gardens, etc.).

However, a fraction of pesticide that is applied (and some of its metabolites) can leach through the soil with infiltrating water and eventually reach the groundwater table. Some of the pesticide residues found in groundwater are hormone mimics or immuno-suppressants that may have significant implications for public health (Matson and Parton, 1997). More generally, all pesticide compounds pose an environmental health hazard as they are, to greater or lesser degree, chemically tailored to be toxic and persistent (Foster and Chilton, 2003). This issue becomes particularly important given the part of groundwater in the distribution of drinking water (e.g. over 80% of the total volume supplied in the Walloon Region; DGRNE, 2005).

It is therefore essential to test and improve our ability to scientifically assess the potential groundwater contamination by pesticides. In Europe, the Water Framework Directive (EU, 2000) and its offshot the Groundwater Directive (EC, 2003) provide the member states with the major objectives and guidelines of water management. The general objective is to ensure that adequate supplies of water of good quality are maintained for the entire population, while preserving the hydrological, biological and chemical functions of ecosystems. Appropriate action is needed because pesticide concentrations in groundwater repeatedly exceed the regulatory limit of 0.1 μg/L in many aquifers across Europe (Tappe et al., 2002; IFEN, 2004; DGRNE, 2005). For pesticides, specific management objectives and registration procedures are still defined within the 91/414/EEC Directive (European Commission, 1991).

In this context, models simulating the transport and fate of pesticides are important tools for groundwater management and protection. Usually a number of climatic, land use, soil and substance parameters are needed as input to allow the assessment of pesticide transport to the different parts of the soil-plant-atmosphere system. These leaching models—and the soil-column or lysimeter experiments often associated with them—are involved in regulatory procedures such as the registration of new compounds. Modelling results offer glimpses into the future (e.g. by the use of 'what if' scenarios) and allow extrapolation to unmonitored areas, whereas monitoring of groundwater contamination reflects activities of the past (Corwin et al., 1999a). These two sources of information are complementary and constitute the core of the research presented here.

1.2 Problem statement

The presence of pesticide residues in groundwater is generally revealed by the analysis of samples taken in wells or at the spring of a groundwater body. These monitoring data may be part of routine measurements (usually performed by drinking water companies) or may be undertaken as a specific field campaign. In many cases, however, the origin of groundwater contamination may remain unclear (Loague and Soutter, 2006), especially for compounds used in both agricultural and non-agricultural applications. Even for pesticides used exclusively in agriculture, leaching may have occurred from a point source such as accidental spill or intended down-well dumping (e.g. after a pesticide label is cancelled), rather than from field applications by farmers (Loague and Soutter, 2006).

In central Belgium, atrazine contamination of the Brusselian aquifer is already described for certain local, well-documented pollution events (Hallaux, 1995; Debongnie et al., 1996), but it is not clearly understood for the entire groundwater body. In particular, considerable argument has concerned the agricultural vs. non-agricultural origin of the pollution; and although atrazine use has been banned for several years, the concentrations measured in certain wells remain above the regulatory limits (DGRNE, 2005).

The scientific literature provides a few examples of studies trying to differentiate between the two contamination sources, generally by looking for significant statistical relationships between atrazine monitoring data and other environmental variables (Maas et al., 1995; Kolpin, 1997; Burkart et al., 1999b). However, these investigations were not always successful and the conclusions can hardly be transferred from one case to another. The first part of this thesis is therefore devoted to the study of atrazine contamination in the Brusselian aquifer.

More generally, little is currently known about the spatial and temporal dynamics of contaminants in large, regional aquifers. The evolution of pesticide concentrations is monitored, but not much information is available about e.g. the time lag necessary for the complete dilution of the pollution.

The existing literature is often restricted either to detailed field-scale studies of a plume movement involving a density of measurements generally not available (e.g. Albrechtsen et al., 2001), or to the description of data sets (e.g. Lapworth et al., 2006; Morvan et al., 2006).

The second part of this thesis is concerned with the modelling of pesticide leaching to groundwater at the regional scale. A recurrent issue in the modelling of soil processes is the discrepancy of the spatial support for which a process model was conceived and the support needed for practical soil and water management (Refsgaard and Butts, 1999; Vanclooster et al., 2004a). Soil process models are usually developed at the scale of soil profiles or pedons, while the geographical area over which models are used has gradually increased (Addiscott and Tuck, 1996). Current needs e.g. for environmental management, have extended the range of model applications across several spatial scales (catchment, regional, global). However, non-linearity in environmental modelling prevents the direct application of models over different spatial scales (Beven, 2001a). There is currently no framework (i.e. a scaling theory) to allow the definition of grid scale equations and parameter values on the basis of small scale (point) measurements (Blöschl, 2001; Beven, 2001a).

In practice, three different approaches are possible before the aggregation of model outputs (Addiscott and Tuck, 1996; Heuvelink and Pebesma, 1999). (i) Model runs are performed on all available point inputs followed by interpolation of the model outputs. (ii) Model input parameters are first interpolated, and subsequently the model is run for all points. (iii) A process model is run on point support within a block without interpolating input or point simulated output. These different approaches have been tested in different applications (e.g. Stein et al., 1991; Sinowski et al., 1997; Van Bodegom et al., 2002b), but the conclusions varied and no consensus was reached to prefer one approach over the others.

Furthermore, the issue of uncertainty is a key theme in pesticide leaching modelling. Understanding the consequences of uncertainty is needed to improve the contribution of process-based models as decision-support tools (Brown and Heuvelink, 2005). A number of methods are available to study the propagation of uncertainty in environmental modelling (Dubus et al., 2002b), but pesticide leaching assessment at the regional scale requires further consideration of the issue of spatial variability. Indeed, studies using stochastic simulations have suggested that spatial variability of environmental properties can significantly affect the leached fractions of pesticides (Jury and Gruber, 1989; van der Zee and Boesten, 1991). 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). However, pesticide properties have also been shown to be spatially variable at different scales (Walker and Brown, 1983; Smith et al., 1987; Novak et al., 1997; Coquet, 2002), but it is still largely unknown how pesticide leaching assessments might be affected. A few studies performed stochastic simulations with variable pesticide properties, but the results were aggregated over the whole study area and thus spatial information was lost (e.g. Zacharias et al., 1999).

Therefore, methods for the assessment of pesticide leaching at the regional scale need to take into account both the modelling uncertainty and the spatial variability of soil and pesticide properties. In this way, the total amount of pesticide leaching to groundwater may be estimated, while groundwater vulnerability may still be assessed on a relative basis between different locations.

1.3 Research objectives

The overall aim of this thesis is to improve our understanding and assessment of pesticide leaching at the regional scale, using both the analysis of monitoring data and spatially distributed modelling.

This work has mainly been carried out with the data of a specific region, i.e. the Walloon part of the Dyle river catchment (580 km^2; central Belgium). This study area is extended in one chapter to include the complete underlying aquifer (Brusselian sands). Another specificity of the thesis is the restriction to atrazine as the pesticide under study. Indeed, the analysis has to cope with the limits imposed by relative data scarcity, even though atrazine is often the best monitored pesticide.

This thesis has two main objectives and several specific objectives.

The first main objective is to determine what information can be extracted from conventional monitoring data sets. The hypothesis is made that a complete analysis of historical monitoring data can serve to obtain information about the processes of groundwater contamination by atrazine in the study area. In particular, a specific objective is to assess the relative importance of non-agricultural vs. agricultural sources of contamination. Currently, catchment- or regional-scale simulations of groundwater contamination focus on agricultural use and the diffuse pollution of pesticides. However, in many urbanized areas, such as the study area, non-agricultural uses of pesticides can have a significant impact on groundwater quality.

Another specific objective is to describe the spatial and temporal dynamics of atrazine concentrations in the Brusselian aquifer, using geostatistics. In the study area, the frequency of observations is rarely higher than 1 to 3 measurements per year for a given monitoring station, because companies usually just comply with the requirements of the regional authorities. A higher sampling frequency allows more comprehensive studies of the temporal trends, including methods such as autoregressive modelling (Jones and Smart, 2005).

The second main objective of the thesis is to assess pesticide leaching potential from agricultural use in the Dyle catchment. This objective is addressed by the implementation of a groundwater vulnerability assessment at the regional scale. Assessing groundwater vulnerability requires by definition to evaluate the likelihood of contaminants to reach a certain target of the groundwater body. A variety of methods are available to do so. The National Research Council (1993) makes the distinction between (i) overlay and index methods, (ii) methods employing process-based simulation models, and (iii) monitoring based statistical inference methods. In this thesis, the second approach is adopted, with the process-based GeoPEARL model (Tiktak et al., 2003).

Given the scale issues involved by the application of GeoPEARL at the regional scale, a specific objective of this thesis is to determine the advantages and consequences of different modelling approaches, in terms of spatialisation of point information (Heuvelink and Pebesma, 1999). These modelling approaches can be tested with simple models in the first place, before their application to a real case in the Dyle catchment area.

A further specific objective in the second part of this thesis is to apply a probabilistic methodology for the assessment of leaching potential in the study area. In particular, we want to address the issue of spatial variability in the assessment of groundwater vulnerability at the catchment scale. Here the issue is to determine whether leaching at the regional scale is significantly affected by the inclusion of the spatial variability of non-georeferenced parameters (i.e. those that were considered to be spatially constant in deterministic simulations).

A final specific objective is to assess whether existing groundwater monitoring data can be used for the validation of leaching modelling at the scale of the study area. As Burkart et al. (1999a) suggested, regional estimates of pesticides concentrations in groundwater should not be expected to compare precisely with the measured values from surveys. Yet, there is a need to evaluate our ability to simulate the right orders of magnitude and the spatial pattern of pesticide leaching to groundwater.

1.4 Outline of the thesis

This thesis (except chapters 2, 3 and 5) is based on articles published or submitted to international peer reviewed journals. This may in some cases lead to repetitions in the text, although in this way the different chapters of the thesis may be read independently.

Chapter 2 gives an overview of the scientific literature on groundwater vulnerability assessment and pesticide leaching modelling. In the review of concepts associated with the modelling of the environmental fate of pesticides, emphasis will be put on the problems of scale, uncertainty, and spatial variability.

Chapter 3 presents the main features (land use, hydrogeology, soils) of the Dyle catchment, which is the study area for most of the following chapters.

Chapter 4 presents an analysis of the spatial and temporal dynamics of atrazine concentrations in the Brusselian aquifer. Using monitoring data collected by water distribution companies and a number of environmental data, we then try to discriminate between agricultural and non-agricultural sources of groundwater contamination in the study area.

Chapters 5 to 8 deal with the spatially-distributed modelling of pesticide leaching. In chapter 5, a synthetic basic case is constructed to analyse the effect of interpolation on spatially-distributed models. In particular, we investigate with simple models the potential effect of interpolating or calculating first on spatial simulations (i.e. similar to the approach followed in the assessment of groundwater vulnerability).

Chapter 6 explores the same concepts as the previous chapter, but in a real-case study of groundwater vulnerability assessment in the Dyle catchment. Using linear and non-linear models, the influence of interpolating or calculating first is evaluated with a number of statistical indicators.

A spatially-distributed uncertainty analysis for the assessment of pesticide leaching is presented in chapter 7. The probabilistic methodology, which consists of Monte Carlo simulations of the GeoPEARL model, is adapted to allow the inclusion of spatial variability of parameters previously assumed constant. In this chapter, the approach is tested with pesticide half-live and organic matter content as the input parameters considered in the uncertainty analysis.

In chapter 8, we apply the same methodology with a higher number of input parameters in the uncertainty analysis. We then examine and discuss the influence of each of them on the total variance of the response variable in the Monte Carlo simulations.

Chapter 9 gives a synthetic view of the main findings of the different chapters. The principal contributions of the thesis are presented, as well as the implications for science and policy, before a section devoted to the limitations and perspectives for future research.