EFFECTIVE APPROACHES FOR PREDICTING ENVIRONMENTAL CONCENTRATIONS OF PESTICIDES: THE APECOP PROJECT

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ABSTRACT

Environmental fate modelling is now actively used for the registration of plant protection products (PPPs). Yet, to be efficient in a harmonised registration process, the quality of PEC modelling needs to be assured. Quality assurance of mathematical modelling implies the validation, documentation and maintenance of the modelling codes and scenarios. The FOrum for the Co-ordination of pesticide fate models and their USe (FOCUS) defined the procedures for realising tier 1 PEC groundwater calculations for active substances at the pan-European level. The FOCUS working groups also identified a range of uncertainties related to the validity of the leaching models and scenarios. To mitigate some of these problems, the EU project APECOP (Effective approaches for predicting environmental concentrations of pesticides) was designed. The major objective of the project is to improve modelling concepts for PEC groundwater and air, and to increase the validity status of the modelling codes. In addition a methodology was developed and implemented to evaluate the representativity of the current tier 1 groundwater scenarios such as recommended by the FOCUS groundwater scenarios working group. In this paper, we summarise the methodologies that were used in this project. More detailed information is given in the subsequent papers.

KEY WORDS: Pesticide fate modelling, validation, scenario evaluation, FOCUS.

INTRODUCTION

For reducing the groundwater contamination risk by residues of toxic plant protection products, appropriate agricultural management at the European scale is needed. The Council Directive 91/414/EEC related to the registration of active substances of plant protection products is one of the instruments which has been developed to meet sustainability criteria in the agricultural sector. The directive envisages, amongst others, to implement uniform principles for assessing the risk associated with the use of plant protection products, and this to support a harmonised registration at the EU level. Predicting the environmental concentrations of pesticides in the water resources by means of mathematical models is an essential part of such a risk assessment.

In recognition of the fact that there was no agreed methodology for PEC calculation, the European Commission (DG-SANCO) set up the *FO*rum for the *C*o-ordination of pesticide fate models and their *US*e (FOCUS). FOCUS has published a general guidance document and reports on the use of mathematical models for predicting PECs in groundwater, surface water and soil (e.g. FOCUS, 1995). Limited number of standardised worst-case scenarios, to be used in the penultimate calculations for PEC groundwater and guidance to model selection, parameter selection and scenario selection became available in 2000 (FOCUS, 2000). Standardised scenarios are needed because they increase the uniformity of the regulatory evaluation process by minimising the influence of the person that performs the PEC groundwater calculation and because they make PEC calculations and their interpretation much easier for administrators, regulators and industry (Boesten et al., 2000).

The PEC groundwater calculation model and the scenarios should apply to the whole EU, and this implies the consideration of the variability of soil, crop, climate, hydrology and hydrogeology, agricultural practice, land use and pesticide use at the community level. An appropriate PEC groundwater model should therefore be sufficiently validated for the different conditions in the EU, and compiled databases on soil, climate, hydrology and hydrogeology at EU level, allowing to generate realistic worst case input for the PEC models, should be available and compatible with the identified PEC groundwater models. Yet, a series of limitations to this was identified by the FOCUS groundwater working group and addressed in some cautionary notes related to the uncertainty of the proposed procedures (FOCUS, 2000). With respect to groundwater, the major uncertainties are related to the validation status of present PEC models and scenarios.

Indeed, validating a PEC groundwater model implies the quantification of the error that is made when predicting e.g. leaching with different leaching modelling codes. Quantifying this modelling error for all possible scenarios in which the model may potentially be used is simply not possible since experimental data for all possible scenarios are not available. Hence, when evaluating the model performance for a new condition (e.g. the fate of a new substance at a new site), there is always a possibility that the model error will become unacceptably large, and that the modelling code should be considered as invalidated for this site. Therefore, within the framework of FOCUS, one considers rather the overall validation status which will increase if a model has successfully passed several validation tests. If this occurs, the probability of success in a new scenario with similar properties will increase. The PEC groundwater models proposed by FOCUS have been through several validation exercises before (e.g. Styczen, 1995; Thorsen et al. 1998; Vanclooster *et a*l., 2000). However, the exercises documented so far represent a very limited number of cases as compared to the number of potential scenarios for which the models will be used in the registration context, and the results of such exercises are rather variable. Therefore scope exists to increase the number of validation studies of the FOCUS PEC groundwater models.

In contrast to the studies related to the validation of the PEC groundwater modelling codes, little attention in the relevant literature has been devoted to the validation of modelling scenarios. Within the framework of the Tier 1 FOCUS PEC groundwater procedure, vulnerability of ground water to contamination resulting from the use of an active substance is represented by means of nine realistic worst-case scenarios. Collectively, these nine scenarios represent agriculture across Europe, for the purposes of a Tier 1 EU-level assessment of leaching potential. The realistic worst case was identified by the concept that scenarios should correspond to 90th percentile vulnerability situations (FOCUS, 2000). This is, in reality, a function of all system properties (weather, soil, groundwater, crop, substance application and chemical properties). A correct theoretical identification of this realistic worst case would therefore imply the development of a few hundred scenarios at the EU-level, which should all be run for the specified substance. A 90th percentile vulnerable scenario could then be identified from the resulting frequency distribution. At the time of the start of the FOCUS activity, Pan-European climate and soil databases, which are required to develop this large number of scenarios, were not available. Therefore, a statistical approach to infer optimal scenarios could not be adopted, thereby adding uncertainty to the Tier 1 assessment. Again, scope exists to reduce this source of uncertainty, hereby improving the quality of PEC groundwater modelling in the Tier 1 level risk assessment.

Further, in contrast to the PEC groundwater, surface water and soil which was addressed by the previous FOCUS working groups, little attention was paid so far to the calculation of PEC to air in a harmonised way. This is a little bit surprising given the considerable human toxicological and ecotoxicological impacts of the volatile components of PPPs in air.

In order to mitigate the above mentioned uncertainty problems and to make a major step forward in PEC air calculations, the project entitled 'Effective approaches for predicting environmental concentrations of pesticides, APECOP' was executed within the framework of the EU-FP5 Quality of Life Program. The major objectives of the project are i) to evaluate the validation status of actual models and scenarios for predicting, within the framework of Council Directive 91/414/EEC, environmental concentrations of PPPs in groundwater; and ii) to reduce the uncertainties in the predictions, by improving the description of preferential transport of plant protection products in soils and volatilisation of these substances to air. For realising these project objectives, experimental field studies were either compiled or carried out to improve the process understanding of pesticide fate and behaviour in the soil-plant-atmosphere system and to allow to evaluate the performance of local scale pesticide fate and behaviour models. Modelling studies at the local scale envisaged to evaluate the performance of local scale models and modelling tools. Modelling studies at the regional scale allowed to evaluate the appropriateness of the scenarios. In this paper, we shortly summarise the methods which were implemented. More information can be found at the project web page (http://www.geru.ucl.ac.be/recherche/projets/apecop-pub/), the project final report, and in the papers further in this section.

IMPROVEMENT OF THE CURRENT PEC MODELS

FOCUS (2000) recommends the use of the PEARL, PELMO, MACRO and PRZM modelling codes in the Tier 1 evaluation of active substances. Guidelines and shells for using the models were designed by the FOCUS groundwater scenario working group and are presently available (http://arno.ei.jrc.it/focus/index.html). However, given the similarity between the PRZM and PELMO model, it was decided to consider only PEARL, MACRO and PELMO in the project.

Preferential flow in soil is well known to affect PPP movement to groundwater but is poorly represented in the actual PEC groundwater modelling codes. It is a kinetic process by which pesticides are leached from the root zones of crops by mechanisms that cannot be explained by classical soil matrix flow theory. Preferential flow can be driven by the presence of structural pores in a macroporous soil, by extreme variability of porosity in the soil matrix, and by the hydrophobicity creating unstable wetting fronts and fingers. Within the project, new process descriptions for preferential flow were analysed and, if appropriate, included in the present PEC models. The flow modules of the MACRO model, which already considered preferential flow, were upgraded. In particular, modules were included which considered an improved description of the boundary conditions, surface tillage and surface sealing effects. The PEARL module for transport in a cracking clay soil is based on the module for water flow in cracking clay soil in the FLOCR model. The model includes both permanent and dynamic macropores which are either continuous up to the groundwater depth or stop at shallower depths. PELMO uses a different approach. Since this model does not solve the Richards equation, a simple preferential flow module, compatible with the field capacity concept was included. Details on these modelling approaches are given by Jarvis et al. (2003, this issue).

Pesticide emission by volatilisation is another major process which is poorly represented in the present PEC procedures. Volatilisation is a major source of pesticides residues in air, fog and rain, and thus may lead to long range transport of pesticide residues remote from their application. Volatilisation is therefore likely to have a major impact on environmental balance of pesticides. In the project, it was envisaged to use data from a detailed experimental program on pesticide volatilisation, to improve the present process knowledge on pesticide volatilisation as a basis for including pesticide volatilisation in present PEC groundwater models. To study the process studies affecting pesticide volatilisation from soil first, a photovolatility chamber was used. Under defined conditions (radiation, soil moisture, air humidity, soil temperature and wind velocity) experiments were performed to analyse the different impacts of environmental parameters on the volatilisation from soil. Further, wind tunnel experiments were carried out to study volatilisation under simulated field conditions, using a UV-transparent wind tunnel, which was set up above a lysimeter with a soil surface area of 0.5 m^2 containing an undisturbed soil core with or without vegetation. Finally field experiments were performed in which volatilisation rates of pesticides were determined by two micro-meteorological methods. The inclusion of a description for the increase in the sorption coefficient at low soil water contents resulted in a better description of the volatilisation under dry soil surface conditions. Details on the volatilisation experiments are given by van den Berg et al. (2003, this issue) for soils and by Wolters et al. (2003, this issue) for plants.

Process knowledge generated in the volatilisation process studies were used to upgrade the PEC air modules of the considered models. A simple empirical approach was included in MACRO to deal with pesticide volatilisation from the soil surface, whereby a fraction of the applied dose is lost to the atmosphere, based on the proportion calculated to be present in the air phase of the soil. The volatilisation module included in PELMO and PEARL was improved, e.g. the moisture dependence of the soil adsorption coefficients at low water content was taken into consideration. The volatilisation from plant surfaces and competing processes (uptake, photo-transformation, wash-off) have also been considered in PELMO and in a plant volatilisation model that will be implemented in PEARL. Details on the modelling approaches for volatilisation from plants to air are given by Wolters et al. (2003, this issue) and those for volatilisation from soils to air are given by van den Berg et al. (2003, this issue).

VALIDATION OF THE LOCAL SCALE PEC MODELS

The performance of the local scale PEC models was evaluated using experimental data from seven field sites. The major characteristics of these sites are summarised in Table 1. The major criterion for considering the site was the availability of data appropriate for model validation. This implies the availability of high quality data on water, solute, and heat transport in the soil, and data on pesticide fate and transport in soil and groundwater (if applicable). The availability of support from the data set provider was considered to be essential. The data sets for the model validation are now fully documented.

A multistage validation approach is considered, similar to the one used in a previous COST model evaluation exercise (Vanclooster et al., 2000). The procedure is described in detail by Trevisan et al., (2001). The different components of the emission models are validated separately in a sequential process. The originality of the procedure resides in the adoption of a blind validation strategy for the evaluation of the models on some of the field sites. For this blind validation exercise, no field data were made available to the model users. Hence, it was evaluated how good PEC models describe field behaviour if only laboratory data or generic data are made available. Although it is well known that calibration may improve considerably the model performance, field data are generally not available and the blind validation level is therefore the most appropriate validation level to be considered for a real life registration exercise.

The preliminary results show that the validation status of the three FOCUS model is quite variable depending on the scenario simulated. No large differences are found between models, even if Richards equation based models are better able to simulate water behaviour. Prior knowledge of real data improves the performance of models. In general, the predictive ability of the models is still poor, and more research is needed to identify the reasons for this: is it the models themselves, or a lack of data to enable appropriate parameterisation? The improvements in model performance that can be demonstrated following 'physically justifiable calibration', suggest that it is mostly the latter. Thus, comprehensive datasets, such as those reported here for Andelst, Bologna, Lanna and Vredepeel are required for proper validation of FOCUS PEC models. Details of the model validation results are given in Trevisan et al. (2003, this issue).

Table 1: Major characteristics of the APECOP experimental field sites

Name	Country	Climate	Soil type	Available reference
Lanna	Sweden	Cold humid	Silty clay over clay	Larsson and Jarvis, 1999
Brimstone	United Kingdom	Moderate sea climate	Cracking heavy clay	Bromilow et al., 1998; Harris and Catt, 1999; Armstrong et al., 2000.
Andelst	the Netherlands	Moderate sea climate	Silty clay loam	In press.
Vredepeel	the Netherlands	Moderate sea climate	Humic sand	Boesten and van der Pas, 1999, 2000
Lebrija	Spain	Mediterranean	Silty (drained marshland)	Andreu et al., 1996, Rieu et al, 1998;
Coria	Spain	Mediterranean	Silty clay loam	In press.
Bologna	Italy	Mediterranean	Loam	Araldi, 1997, Scarabello, 1999

PEC GROUNDWATER SCENARIO VALIDATION / EVALUATION

In contrast with the large number of publications available on model validation, only few studies dealt with the validation/evaluation of scenarios. However, the efficiency of a PEC groundwater assessment in a Tier 1 exercise will depend on both, the validity of the PEC groundwater model and the correctness of the modelling scenarios. As already reported in the introduction, only a limited number of groundwater scenarios were, for pragmatic reasons, identified in the FOCUS PEC groundwater procedure. Given the limited availability of pan-European environmental data at the start of the FOCUS activity, a statistical approach to infer optimal scenarios could not be adopted. The present PEC calculations may therefore be biased in representing worst case scenarios, given the under-sampling of the population of European soil types, climatic conditions and agricultural practices. Therefore, it is of paramount importance to make progress in the validation of the groundwater scenarios and to quantify/reduce reduce this bias.

A scenario in the context of the FOCUS procedure is defined as a representative combination of crop, soil, climate and agronomic parameters to be used in modelling. In this context representative means that the selected scenarios should represent physical sites known to exist, i.e. the combination of crop, soil, climate and agronomic conditions should be realistic (FOCUS, 1995). For PEC groundwater calculations, FOCUS intends to construct scenarios that represents an overall vulnerability approximating the 90th percentile leaching of all possible situations (this percentile is often referred to as a realistic worst case) (FOCUS, 2000). They further assumed that the vulnerability was equally attributed to soil and climate. To achieve this, they first defined nine so-called FOCUS areas. Within each of these areas, they selected an approximate 80% vulnerable soil, which implies that the concentration of a PPP should be less than the EU drinking water limit in at least 80% of the corresponding area. Hence, a FOCUS groundwater scenario is a combination of parameter values selected in such a way that the leaching concentration calculated with a FOCUS PEC groundwater model equals the 90th percentile of leaching inside the corresponding FOCUS area. It was the objective to validate this scenario definition statement in the APECOP project.

Details on the scenario validation protocol can be retrieved in Piñeros Garcet et al., 2001. A brief summary is given here. The key question to answer is: 'Are FOCUS scenario combinations of parameter values selected in such a way that when used in combination with a FOCUS PEC groundwater model, the calculated leaching values correspond to the real 90th percentile of leaching?'. To answer this question, it is essential to define the 'real' 90th percentile of leaching. In an ideal situation, this 90th percentile could be obtained from a detailed monitoring of the presence of active substance in groundwater. However, our observational skills are limited in time and space, and therefore the 'real' 90th percentile is unknown. If the 90th percentile of leaching cannot be obtained from direct measurements, an alternative is offered by approximating these leaching values using a 'detailed' assessment technique. This technique comes down to approximating the 'real' 90th percentile of the leaching concentration by means of a spatially distributed leaching model in combination with Pan-European soil, climate and agricultural databases. Obviously, this type of validation has a lower power than a validation in which the 90th percentile of leaching is estimated from direct measurements, but it is the only pragmatic way to proceed with scenario validation at this time. Given the computational burden associated with the use of spatially distributed modelling technique, the validation could only be applied to a limited number of pesticides and to two major agricultural crops (maize and winter wheat). This implies that the vulnerability is considered to be driven mainly by soil and climate, and not by crop properties.

Simulations were performed by means of the PEARL model (i.e. the EuroPEARL model) for 1062 unique combinations of soil type, climate and country, sampled throughout Europe. Soil properties, including soil horizon designations, were obtained from the Soil Profile Analytical Database of Europe. Daily weather data were obtained from the MARS database. Other data like irrigation data, crop data and pesticide properties have been compiled from various sources, such as inventories, field-studies and the literature. The 1062 unique combinations together represent at maximum 75% of the total agricultural area of the European Union. Austria, Sweden and Finland could not be included in the simulations, because there was insufficient soil profile information for these countries. However, to consider also the soil-climate and crop combinations for which no profile data where available, a meta PEC groundwater model was derived by interpolating EuroPEARL modelling results in the input parameter space using radial basis functions neural networks. The combination of the process based deterministic EuroPEARL model with the meta-model allows: (1) to use (for leaching simulations) the European 1:1 000 000 soil map, which covers 97% of the European agricultural area instead of the 75% covered by the profile database; (2) to take into account spatial variability of leaching inside the mapping units; and (3) to test the validity of the scenarios in a statistical way.

Results of the EuroPEARL model are presented in maps with a resolution of 10x10 km^2, which is the highest justifiable resolution based on the vectorial EU soil map 1:1 000 000. The results indicate that the leaching concentration generally increases with precipitation and irrigation and decreases with increasing organic matter content. Because of the strong sensitivity of the leaching concentration to soil properties, there is a strong variability of the calculated leaching concentration at relatively short distances. Results further indicate that due to large irrigation amounts combined with large temporal variation of rainfall in the Southern European countries, the trend in the calculated leaching risks from North to South is less extreme than expected. This implies that areas of high leaching risk ('hotspots') as assessed by means of the EuroPEARL model occur in all countries of the European Union, including the Southern European countries. Confronting results of the EuroPEARL model with FOCUS calculations shows that, for the limited cases which were analysed, the FOCUS groundwater scenarios for Southern European countries may be biased and should be considered for revision. Comparing the meta-modelling results with the FOCUS scenarios shows that the inclusion of the additional soil variability substantially increases the leaching percentiles. Details on the EuroPEARL modelling results can be retrieved in Tiktak et al. (2003) and Tiktak et al. (2003; this issue). Details of the combination of EuroPEARL with the meta-model can be found back in Piñeros Garcet et al. (2003, this issue).

CONCLUSION

In this paper, we summarise the methodologies that were used and some preliminary results that were obtained within the framework of the EU-FP5 project "Effective approaches for predicting environmental concentrations of pesticides: APECOP". The project was designed to improve PEC modelling procedures to groundwater and air as a basis for the harmonised registration of PPPs. To this end, process descriptions for preferential flow and volatilisation were improved in the current local scale PEC groundwater models. The local scale PEC models were further validated using a series of field studies, and the representativity of current recommended PEC modelling scenarios (i.e. the FOCUS groundwater scenarios) were critically analysed. For this latter analysis, the PECs of a fully spatially distributed modelling approach were compared to the PECs as obtained using the actual recommended FOCUS procedures.

The new approaches for describing preferential flow in soil and volatilisation from soil and crop canopies result in new tools for including two extremely relevant emission routes in exposure assessment procedures. The new approaches should be further considered for inclusion in the actual recommended FOCUS exposure assessment procedures. The critical results of the local scale PEC model validation confirm the uncertainty associated with present PEC modelling procedures. It emphasizes the need i) to further improve the process description in exposure models; ii) to establish high quality data for parametrizing and evaluating fate and transport models; and iii) to explore alternative robust strategies for PEC calculations. The results of the scenario validation/evaluation analysis suggest that some recommended FOCUS scenarios for PEC groundwater, in particular those for Southern European countries, should be considered for revision. It also elucidates the problem of introducing small scale variability in a robust PEC assessment procedure.

ACKNOWLEDGEMENT

This research is carried out in the framework of the EU-FP5 R&D project: "Effective approaches for predicting environmental concentrations of pesticides: APECOP", QLRT-CT1998-01238.

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