



ELSEVIER

Agricultural Water Management 44 (2000) 1–19

Agricultural  
water management

www.elsevier.com/locate/agwat

## A European test of pesticide-leaching models: methodology and major recommendations

M. Vanclooster<sup>a,\*</sup>, J.J.T.I. Boesten<sup>b</sup>, M. Trevisan<sup>c</sup>, C.D. Brown<sup>d</sup>,  
E. Capri<sup>c</sup>, O.M. Eklo<sup>e</sup>, B. Gottesbüren<sup>f</sup>, V. Gouy<sup>g</sup>,  
A.M.A. van der Linden<sup>h</sup>

<sup>a</sup>*Department of Environmental Sciences and Land Use Planning, Université Catholique de Louvain,  
Place Croix du Sud 2 BP2, B-1348 Louvain-la-Neuve, Belgium*

<sup>b</sup>*ALTERRA P.O. Box 125, 6700 AC Wageningen, Netherlands*

<sup>c</sup>*Istituto Chimica Agraria ed Ambientale, Università Cattolica del Sacro Cuore,  
Via Emilia Parmense 84, I-29100 Piacenza, Italy*

<sup>d</sup>*Soil Survey and Land Research Centre (SSLRC) Cranfield University, Silsoe, Bedfordshire, MK45 4DT, UK*

<sup>e</sup>*The Norwegian Crop Research Institute, Plant Protection Centre, Fellesbygget, N-1432 As, Norway*

<sup>f</sup>*BASF AG, Agrarzentrum Limburgerhof, Postfach 120, D-67114 Limburgerhof, Germany*

<sup>g</sup>*CEMAGREF, Agricultural and Environmental Engineering Research, 3bis Quai Chauveau,  
F-69336 Lyon, France*

<sup>h</sup>*RIVM, P.O. Box 1, NL-3720 BA Bilthoven, Netherlands*

---

### Abstract

Testing of pesticide-leaching models is important in view of their increasing use in pesticide registration procedures in the European Union. This paper presents the methodology and major conclusions of a test of pesticide-leaching models. Twelve models simulating the vertical one-dimensional movement of water, solute, heat, and, in particular, pesticides, through the soil profile were used by 36 different modellers. The adopted modelling codes differ in terms of modelling concepts and modelling hypothesis. Modellers were affiliated to industry and to the scientific community as well. Four quality datasets were identified to perform the analysis. The dataset included field and lysimeter data, collected in the Netherlands, Germany, Italy and the UK. As well, non-structured as structured soils were available in the dataset. To elucidate the ability to model correctly water transport, solute transport, heat transport and pesticide transport in soils, a stepwise evaluation approach was followed. Splitting up the experimental dataset enabled us to quantify the calibration capability and the prediction capability of the models. The simulations were performed by different model users enabling us also to characterise output variability in terms of user dependent interpretation of the model input and parameters. Recommendations are formulated for

---

\* Corresponding author. Tel.: +32-10-473710; fax: +33-10-473833.

E-mail address: vanclooster@geru.ucl.ac.be (M. Vanclooster).

improving the quality of modelling datasets, and the process description of water, solute, and heat transport in a pesticide-leaching model, plus the process description of pesticide fate. Application of the principles of good modelling practice (GMP) is briefly described. © 2000 Elsevier Science B.V. All rights reserved.

*Keywords:* Pesticide-leaching modelling; Model validation; Risk assessment

---

## 1. Introduction

Losses of pesticides from agricultural soils may influence the long-term quality of both surface and groundwater. Residues of chemicals have been retrieved in surface and groundwater bodies all over the world (IUPAC, 1987; Leistra and Boesten, 1989; Faasen, 1995) and the pesticide loads frequently exceed the drinking water limit set by the EU at 0.1 µg/l.

Uniform principles have recently been accepted by the European Commission to implement a coherent registration of pesticide products at the European level, envisaging a high level of protection of surface and groundwater under the conditions of use. Only those products will be accepted for registration for which the risk of exceeding the drinking water limit is insignificant. For the assessment of the leaching risk, the EU-directive 91/414 (EU, 1997) suggests the performance of simulation studies. Such simulation studies should be carried for realistic worst case scenarios, using 'validated' simulation models. The simulation model should be reliably validated with measurements carried out under realistic circumstances. Given the wide range of conditions within the EU, there is also a wide range of validity needed for an appropriate use in the EU pesticide registration procedure. Moreover, the EU drinking water limit of 0.1 µg/l corresponds with an amount leached that is in the order of 1 g/ha. This is 0.1% of a typical dose of 1 kg/ha. This implies that the validation status of pesticide-leaching models is most relevant at leaching levels of 0.1% of the dose. Modelling leaching at such low levels requires a rigorous understanding of all relevant pesticide transport and fate processes in soil (Brown et al., 1995). However, site specific information on such processes is often lacking and the validation status of pesticide-leaching models for the intended use in pesticide registration at the EU level is therefore low.

Compared to the number of products what need to be registered, and compared to the number of available simulation codes, limited attention has been devoted to model testing. The lowest level of model testing is testing a single model by a single user against experimental data. A more elaborated test, however, may include the comparison of different potential simulation codes. Such model comparison exercises have been the subject of some projects in the early 1990s. Pennel et al. (1993), for instance, used field measurements of aldicarb and bromide to compare the performance of five pesticide fate models. They made recommendations for the selection of the models, based on objective performance criteria, data requirements and intended model use. Bergström and Jarvis (1994) compared the performance of seven models using lysimeter data. They observed that the experimental leaching rates varied tremendously among the lysimeters, which they attributed to spatial variability of the transport and transformation processes and

preferential flow phenomena. These phenomena were poorly represented with the available codes. Diekkrüger et al. (1995) compared, amongst other agro-chemicals, three pesticide-leaching models on a field site where methabenzthiazuron, metamitron, and simazine were measured. They observed small differences between different models when analysing the short-term simulation results, but suspected deviations to become more important in the long-term. Given the limited dataset, the long-term behaviour could not appropriately be evaluated.

Modelling technology is, however, changing rapidly. Whenever new process insights, and modelling techniques are acquired, scope exists for additional model testing studies. Further, in contrast to the pioneering modelling activity of the scientific community to unravel the system processes, current activity evolves within an engineering context. Nowadays, modelling is performed by people from academic, regulatory, and industrial backgrounds who have often diverging objectives and different skills in modelling. The use of complicated pesticide-leaching models by people having different backgrounds is not without danger. Diekkrüger et al. (1995), for instance, stated that the experience of the modeller applying the model may be as important as the difference among the modelling approaches. Different subjective interpretations of modelling results, input and parameters may induce diverging assessments of pesticide-leaching. In order to cope with such a ‘user-specific variability’, good modelling practices (GMPs) are required. Ad hoc working groups in the EU (FOCUS, Boesten et al., 1995) and US (EMWG, Exposure modelling working group, 1995) are currently defining the protocols and guidelines for soundly using these models. A ‘high level’ model test will, therefore, not only include the evaluation of the applicability of the model concept, but also the way how model input is generated and used by the model user.

Within this special issue, an attempt is made to present a ‘high level’ pesticide-leaching model test. The adoption of different modelling codes and different datasets allows us to compare the quality of different models for different edapho-climatic conditions. Adopting a stepwise test procedure, allows qualification of different essential components of pesticide-leaching models and the components describing water flow, solute flow, heat flow and pesticide fate in soils. Splitting up of the experimental data, allows characterisation of the impact of calibration on modelling results. The use of the same model by different users allows us finally to characterise the impact of subjective decisions in the overall modelling process, and on model output. To our knowledge, no studies have been presented that include all these aspects of model testing for the pesticide-leaching modelling process. The objective of this paper is to present our methodology, the implementation structure, and the overall recommendations formulated by the contributors at the end of the exercise.

## **1. 2. Materials and methods**

### *2.1. The models*

A detailed description of available models to simulate pesticide-leaching is not the subject of the present paper. For such a description, the interested reader is referred to

review papers such as those presented by Jury and Ghodrati (1989), Jarvis et al. (1995), Thorsen et al. (1996) and Vanclooster et al. (2000b). The interested reader is also referred to the compiled databases of leaching models such as reported by FIFRA (EMWG, Exposure modelling working group, 1995), Camase (1996), FOCUS (Boesten et al., 1995) or COST (Vanclooster et al., 2000b).

The models that have been used in our exercise are given in Table 1. For a more detailed description of the models, the reader is referred to the accompanying papers and the reference lists included therein. All models of Table 1 calculate pesticide-leaching as a one-dimensional flow process within the unsaturated soil root zone of the crop. The selected models ignore the horizontal flow component that may occur in the unsaturated zone. Neither, do they appeal to any stochastic theory to consider a full three-dimensional description of the flow phenomenon. The models operate at least on a daily time step and integrate the daily leaching rate to obtain the total leaching amount. We further observe that both mechanistic leaching models (PESTLA, PESTRAS, MACRO, LEACHP, WAVE, SIMULAT, CRACKP) and functional, yet more empirical, models (PRZM2, VARLEACH, PLM, PELMO, GLEAMS) are considered. The former type of models is traditionally used in an academic context, whereas the latter type of models is more popular for registration and management applications. In addition, models are retained that explicitly account for preferential flow through macropores (MACRO, CRACKP, PLM).

## *2.2. The model users and model co-ordinators*

The models were used by different model users to simulate pesticide-leaching. The list of the model users is included in the references of Table 1. It should be noted that model users are from the academic and industrial world, allowing to pay particular attention to model testing when modelling is guided by sometimes diverging objectives. For each model, a model co-ordinator was appointed. The model co-ordinator was responsible for providing the appropriate version of the model and for streamlining the work to be carried out with the code in the present analysis. The model co-ordinator was either the model developer or a dedicated model user having specific technical know-how of the model. The model co-ordinator was also responsible for the summary report of the performance of the model.

## *2.3. The datasets*

Four ‘high quality’ datasets were considered in the modelling test. In view of the stepwise test procedure, only datasets were considered that contained data on soil hydrology, solute transport behaviour, soil temperature, and, of course, soil pesticide fate. Further, given the European dimension of the project, datasets from different edapho-climatic regions of Europe were considered. In addition, given the availability of models explicitly considering macroporous flow, data from a typical macroporous soil were included. Finally, to evaluate the modelling performance of the balance terms, and to test the applicability of the modelling methodology to describe field behaviour, both field and lysimeter datasets were included.

Table 1  
Models and major characteristics considered within the modelling test

Subject	CRACKP	GLEAMS	LEACHP	MACRO	PELMO	PESTLA	PESTRAS	PLM	PRZM2	SIMULAT	VARLEACH	WAVE
Reference	Armstrong et al., 2000b	Rekolainen et al., 2000	Dust et al., 2000	Jarvis et al., 2000	Klein et al., 2000	Boesten and Gottesbüren, 2000	Freijer et al., 1996	Nicholls et al., 2000	Trevisan et al., 2000	Aden and Diekkrüger, 2000	Trevisan et al., 2000	Vancloster et al., 2000a
Water flow governing equation	Capacity type	Capacity type	Richards equation	Richards equation	Capacity type	Richards equation	Richards equation	Capacity type	Capacity type	Richards equation	Capacity type	Richards equation
Solute flow governing equation	Convective	Convective	Convection dispersion equation (Yes)	Convection dispersion equation (Yes)	Convection	Convection dispersion equation (No)	Convection dispersion equation (No)	Convection	Convection	Convection dispersion equation (Yes)	Convection	Convection dispersion equation (Yes)
Preferential flow Sorption	Yes Linear	No Linear	Freundlich	Linear	No Freundlich	No Freundlich	No Freundlich	(Yes) Linear	No Linear	Yes Freundlich	No Linear	Yes Linear
Degradation	First-order	First-order	First-order	First-order	First-order	First-order	First-order	First-order	First-order	First-order	First-order	First-order
Plant uptake	No	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	No	Yes	Yes
Volatilisation	No	No	Yes	No	No	No	Yes	No	No	No	No	No

Table 2  
Datasets considered in the modelling exercise

Dataset	Type	Reference of dataset description	Reference to major modelling results
Vredepeel	Field	Boesten and van der Pas, 2000	Tiktak, 2000; Vanclooster and Boesten, 2000
Weierbach	Field/lysimeter	Schierholz et al., 2000	Gottesbüren et al., 2000
Tor Mancina	Lysimeter	Francaviglia and Capri, 2000	Francaviglia et al., 2000
Brimstone	Field	Harris et al., 2000	Armstrong et al., 2000a

The references to the four datasets are given in Table 2. For each dataset, a dataset coordinator was appointed, who communicated experimental details from the data provider to the model users. They also distributed the data in a rigid format, and described the dataset in an accompanying paper. For each dataset, the performance of the different models was compared (see references in Table 1).

#### 2.4. The modelling protocol

Any model can be tested using a scheme as depicted in Fig. 1. Such a protocol consists of two subsequent phases. In a first phase, the model's fitting capacity is analysed. In the second phase, the ability of the model to extrapolate or to predict, is analysed. Calibration in this context is considered as a parameter estimation technique. During calibration, an object function is optimised such that the difference between model calculated and observed pesticide concentration is minimised. Such an optimisation can be done by changing the model input parameters by trial and error or by more elaborate statistical optimisation techniques. During the second phase, no re-adjustment of the model parameters is done. At this stage, independent data are used to test the model performance.

It should be noted that, in a regulatory context, calibration as suggested here will in general not be possible, because it requires several field experiments for the same pesticide/soil combination that are usually not available. In pesticide registration procedures, a first assessment is made on the basis of pesticide properties measured under well-defined laboratory conditions. So for decision-makers, it is crucial to know the potential of a model to predict pesticide behaviour without calibration. Moreover, the Vredepeel dataset (see Table 2) consists of only one field experiment. The calibration procedure proposed in Fig. 1 could, therefore, not exactly be applied to this study. Another complication is that pesticide models consist of sub-models for simulating water flow, solute flow, heat flow, and pesticide behaviour. A method of testing should, therefore, be pursued allowing us to evaluate all sub-models in subsequent steps. Given this dilemma, the following pragmatic solution was applied in our model testing exercise. First, test the whole model for uncalibrated parameter values and report the results. Then, calibrate the sub-models in the sequence water flow, solute flow, heat flow, pesticide behaviour; and finally test the whole model with calibrated parameter values if data are available.

In model testing, particular attention needs to be paid to the target quantity to be considered. The sub-models for simulating water flow, solute flow, heat flow, and

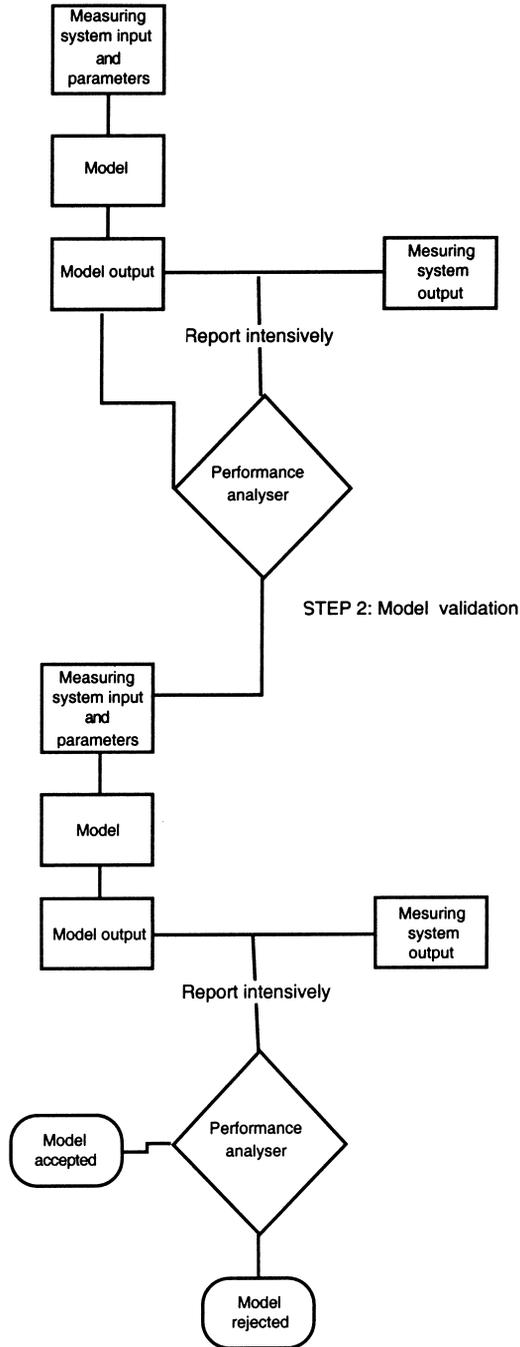


Fig. 1. Flow chart for evaluating pesticide-leaching models.

pesticide behaviour could be tested using system-state variables such as the soil moisture content, the soil solute concentration, the soil temperature, and the soil pesticide concentration. Alternatively, balance terms such as the cumulative soil water drainage or the cumulative amount of pesticide-leached can be used. The balance terms may be more appropriately analysed in a risk analysis context. Yet the balance terms are hard to measure on a field scale, which makes the test of pesticide-leaching models at this scale cumbersome. Correct cumulative amounts leached can be inferred from the lysimeter datasets. However, flow behaviour in lysimeters does not reflect the complete variability of pesticide flow that might occur in the field, and bias may result in the risk assessment. To cope with aforementioned dilemma, a pragmatic solution was proposed. For the field datasets, the simulated state variables were compared with the field measured state variables in space and time. Adopting such an approach, one tacitly assumes that if the model matches measured state variables in space and time, the model will have a large probability of simulating the balance terms appropriately. For the lysimeter datasets, the different models calculated balance terms that were compared to the balance terms measured in the lysimeters. In this local scale approach, field heterogeneity — a phenomenon that undoubtedly dominates pesticide fate and transport — is not explicitly accounted for during the test.

The appropriateness of the model calculation can be either assessed by modelling statistics or simulation graphics. When both system output and model output are considered to be deterministic, the arithmetic difference between the measured and calculated output is the most straightforward measure of the model deviation. The large amount of data, however, enhances the use of more comprehensive model deviation indicators. A selection of statistical criteria, used in the current modelling exercise, is given in Table 3.

Table 3  
Selection of statistical model indicators

Overall fit		
Maximum error	$ME = \max P_i - O_i  \quad \forall i$	Loague et al., 1989
Root mean square error	$RMSE = (100/\bar{O}) \sqrt{\frac{\sum (P_i - O_i)^2}{n}}$	Loague et al., 1989
Modelling efficiency	$EF = \frac{(\sum (O_i - \bar{O})^2 - \sum (P_i - O_i)^2)}{\sum (O_i - \bar{O})^2}$	Loague et al., 1989
Nash–Sutcliffe coefficient	$CNS = 1 - \frac{\sum (O_i - P_i)^2}{\sum (O_i - \bar{O})^2}$	Nash and Sutcliffe, 1970
Average difference	$AVDIF = \frac{\sum (O_i - P_i)}{n}$	
Total soil residue		
Coefficient of residual mass	$CRM = \frac{(\sum O_i - \sum P_i)}{\sum O_i}$	Loague et al., 1988
Distribution of model error		
Coefficient of determination	$CD = \frac{\sum (O_i - \bar{O})^2}{\sum (P_i - \bar{O})^2}$	Loague et al., 1988

## 2.5. *The implementation*

Three workshops were organised to implement the modelling test. In the first workshop (Limburgerhof, Germany, autumn 1995) the participants were introduced to the modelling protocol. Datasets were presented and distributed after the initial workshop. During the second workshop (Piacenza, Italy, summer 1996), an intermediate evaluation of the study was done and participants presented their results related to soil hydrology, solute and heat transport. In a final workshop (Louvain-la-Neuve, Belgium, spring 1997), the pesticide modelling results were presented and overall conclusions and recommendations were drawn on the basis of discussions in four subgroups. In between the workshops, considerable exchange of information between model users, model co-ordinators, data co-ordinators, model evaluators and the organising committee took place. A web page and news server were established at the University of Prague to promote the exchange of information.

## 3. Recommendations

### 3.1. *The significance of a high quality dataset*

The four selected datasets were initially estimated to be appropriate for a modelling test. However, during the realisation of the test, the quality of the available datasets was questioned more than once. A dataset was considered to be qualified for a modelling test if the experimental protocols met the recommended guidelines, and if all relevant model input requirements could be inferred from the dataset. Both quality criteria are important.

#### 3.1.1. *The preparation of the dataset*

Before starting the experiment it is important to know the history of the agricultural field. Major historical perturbations need to be mapped when deciding the suitability of the site for setting-up an experimental programme. The installation of artificial drainage, for instance, will disturb the structure of the soil profile and may create permanent macropores down to the tiles in the backfill. A major shift in land use may influence nutrient status and hydrology. Recent amendments of organic fertiliser will change the soil nutrient status and its physical properties. Set aside or new crops may have an impact on soil chemistry, and hence pesticide mobility. Pesticide residues from previous years can still be present when new dressings start.

In addition, during the preparatory phase of an experimental programme, a lot could be learned from a priori model simulations, parameterised with generic modelling data. Generic soil data can be inferred from soil map information using pedo-transfer functions; climatic data can be generated with a weather generator; physico-chemical pesticide data can be estimated from desktop reference databases. These hypothetical simulation runs may address the expected variations of the state variables of soil moisture and pesticide concentration in space and time, and so allow optimisation of the sampling programme.

### *3.1.2. The experimental setup*

Particular attention needs to be devoted to the correct monitoring of the boundary conditions. If we do not know exactly what goes in the system, it will be difficult to predict what goes out. Daily climatic data were felt to be sufficient for feeding the hydrological component of the matrix based flow models. High resolution data of incoming water flux is, however, critical for macropore flow models, but these data were not available within the test dataset. In addition, there is still a lot of uncertainty involved with the estimates of evapotranspiration rates determining to a large extent the drainage term. Uncertainty of evaporation rates was addressed for the Vredepeel and the Tor Mancina dataset. It was argued that the collection of meteorological data should start some months before the application of the compounds and be extended some months after the last soil or groundwater sample had been taken. The early collection of meteorological data is needed to start the simulation sometime before pesticide application takes place. Such a ‘warming up’ period allows the state variables of the model to attain equilibrium before pesticide application takes place. Finally, given the spatial variability of meteorological data, weather data should be collected as close as possible to the experimental site. To make an appropriate solute and pesticide balance, it is obligatory to quantify correctly the total application rates of pesticide and tracer. Filter paper or petri-dishes filled with soil, distributed randomly on the plot, may give some valuable information on the variability of application within the field plot. A special boundary problem is related to the monitoring of surface run-off fluxes during periods of heavy rainfall. In lysimeter experiments, overflow may often be inappropriately monitored. Similar errors may take place in small field plots, especially when the surface infiltration rates are limited by ice and frost. In addition to run-off, losses by wind-erosion (soil dust) may contribute to disappearance of a compound and result in poor mass balance.

When conceiving the experimental programme, particular attention needs to be devoted to the spatio-temporal variability of soil processes. To avoid gaps in soil description, we recommend a complete soil profile description, with samples and soil parameters for all relevant soil horizons. Most models can handle the depth dependency of soil properties and consider the soil horizon as the carrier of the soil information. Soil samples should, therefore, at best be representative for a given soil horizon. To comply with the spatial variability of the properties within the soil horizons, a significant number of replications need to be available. It was felt by the participants that soil moisture and temperature should be measured at the field site itself. To comply with the dynamic nature of the system, enough measurements during a year must be made. The participants suggested that at least six observations of soil moisture and pesticide status are needed in a year. However, for correctly evaluating the soil temperature, it was felt that more detailed data are needed.

It was concluded that information on soil hydrology, soil heat and tracer behaviour is essential to validate a leaching model. However, ionic tracers may also be liable to other complicating factors. They may show uptake by plants and anion exclusion. There also seems to be some sorption of the tracers in some layers, especially layers with organic matter. For the agronomic variables, it was felt that a detailed monitoring of the leaf area index, together with the rooting depth, could be very useful.

### 3.1.3. *The presentation and interpretation of the dataset*

For the purpose of modelling testing the raw data as well as the interpretation of the data should be represented. It was suggested that we add expert judgements from the dataset provider concerning the quality of the data. All individual measurements should be tagged with a label indicating the quality or the reliability of the individual measurement. Initially, no guidance was given to the modellers on methods of interpretation of the data, although such guidance would clearly be beneficial and would help to eliminate much of the user-introduced subjectivity. Mass balance of the experiments should further be checked to see if the recovery allows sound conclusions to be drawn from the results.

It is an interesting question as to whether laboratory or field transformation data are most appropriate as input values for simulations at the field scale. Laboratory data are collected under controlled conditions that allow influences such as those of soil temperature and moisture content to be quantified. In addition, data are collected for a large number of compounds using the same methodology and this allows ready comparison. However, transformation rates derived from field studies should be a better representation of the behaviour in the field where conditions are not constant and fluctuations in, for example, soil moisture content may have a profound effect upon various relevant processes. Pesticide transformation in the field is generally lumped into an overall dissipation rate, which includes losses from a large number of processes including volatilisation, plant uptake, runoff and leaching. Where transformation is the dominant process, use of field dissipation rates as an input for models may be justified. This may best represent behaviour in the field. But in the case where losses via other processes are significant, rates of transformation will be over-estimated. The extent to which laboratory data can be interpreted and used to simulate field behaviour have been highlighted in the exercise. For sorption, the choice of isotherm type is important. But a major question is how closely laboratory data measured at large water/soil ratios can be expected to mimic the field situation that is much drier. For transformation processes, a number of general rules might have improved the match between laboratory and field behaviour. Firstly, where a decrease in the rate of transformation with time is observed, one can exclude transformation data for times greater than 100 days after the start of the experiment. This will limit the effect of adverse changes in soil microbial viability with time resulting from the closed and constant nature of the experimental setup. One can exclude transformation studies conducted at low temperatures where overall transformation is relatively small and the measurement errors are, thus, relatively large. Where only a limited number of transformation rates at different temperatures and moisture contents are measured, parameters describing the variation in rate with soil conditions might be better obtained from literature values.

## 3.2. *Water, solute and heat transport processes in pesticide-leaching models*

### 3.2.1. *Soil hydrology*

There is a general agreement that pesticide-leaching models preferably should not be used if the hydrological component is not reliable. The rationale behind this is that

pesticide-leaching fluxes will be assessed wrongly if soil water drainage fluxes are wrongly predicted.

Yet, there is no agreement about the level of detail needed in modelling the soil hydrology for assessing pesticide-leaching risk in a regulatory context. The relative ranking of pesticide mobility with leaching models containing a simplified flow module could be similar to that obtained with more complex mechanistic leaching models. All the models based on the Vredepeel dataset for instance were able to target significant mobility differences between bentazone and ethoprophos, regardless of the hydrological description used. This may confirm, as already reported by Van der Zee and Boesten (1991), that the sensitivity of the simulated pesticide-leaching fluxes to the water flow parameters is an order of magnitude lower than the sensitivity to the sorption and transformation parameters.

Mechanistic models describe flow behaviour based on the governing flow equation. The use of these models within a regulatory context is often jeopardised by non-user-friendly modelling codes, and by the difficulty in obtaining the appropriate model parameters. However, much progress has recently been made in developing generic databases and user-friendly software codes, allowing mechanistic models to be used much more easily in an engineering context. Simplified models, based on the field-capacity concept, are already available to circumvent the aforementioned problems. Detailed comparison of simulation results obtained with the Vredepeel dataset, however, has shown that the use of simplified ‘capacity type models’ may be biased under certain circumstances (Vanclooster and Boesten, 2000).

Particular attention was devoted to the issue of macroporous flow. Macropores are important for assessing pesticide-leaching, as it was clearly illustrated for the Brimstone and Tor Mancina dataset. Yet, it is suspected that preferential flow could also occur at other experimental sites. The significance of macroporous flow is in agreement with the conclusion of an earlier modelling test presented by Bergström and Jarvis (1994). The correct conception of macroporous flow in a pesticide-leaching model remains a matter of debate. Outputs from the macropore flow models are sensitive to parameters related to the macropore region, but this information is generally lacking in the datasets. This may lead to high levels of predictive uncertainty compared to the use of models in non-structured soils. Many measurements have been made on macroporosity in soils. But there is an urgent need to collate these data into existing generic databases to allow the development of robust pedo-transfer functions for macroporosity. A plea is also made for including more quantitative structural descriptions to consider macroporosity in a regulation and land management context.

### 3.2.2. *Solute transport processes*

Some description of the solute transport concept is necessary for pesticide transport modelling. Pesticide components contribute to sub-surface water contamination risk if they are dissolved in the soil solution. However, the process of solute transport at the field scale is currently far from being fully understood, and, therefore, no agreement exists on the level of detail needed to model solute transport in a pesticide-leaching model. All models consider convective motion as being the major factor driving solute transport in soils. The more mechanistic codes, however, consider an additional contribution to solute

transport from hydrodynamic dispersion and/or chemical diffusion. The most complicated models consider non-equilibrium phenomena that influence solute transport. Unfortunately, the tracer data available from field experimental datasets did not allow us to elucidate what modelling concept is driving solute transport in soils. Also, it did not allow us to estimate appropriately solute transport parameters, such as the effective hydrodynamic dispersion coefficients or effective pore water velocity. Therefore, much more detailed experimental information of solute flow, especially the variability of it in the field, must be obtained.

### 3.2.3. *Heat transport processes*

Most pesticide-leaching models consider temperature as being a controlling factor for modelling pesticide dissipation. Poor temperature simulations were only reported for the LEACHP model with underestimation by 6–9°C. There was a consensus that the uncertainty in estimation of the impact of temperature on transformation is far more uncertain than the estimation of temperature itself. It should be noted, however, that freezing conditions were not considered within the test datasets and that the potential of the models to describe appropriately heat flow under these conditions could not be evaluated. It was concluded that appropriate heat flow modelling in soil is essential within a pesticide-leaching modelling study. But it was not felt as being a major bottleneck within the state of our current know-how.

### 3.2.4. *Scale issues and uncertainty aspects*

To estimate correctly the pesticide load leaving the soil profile at the field scale, and especially to target the small quantities required within EU regulation, pesticide fluxes should be assessed in combination with pesticide concentrations. Given the heterogeneity of the flow at the field scale, appropriate evaluation of field scale pesticide flux remains difficult. Resident pesticide concentration data, such as determined at the field scale in Vredepeel, are only available for limited times and soil depths. So there is considerable uncertainty in the estimation of pesticide fluxes, especially when non-equilibrium flow, such as in macropores, takes place. The measured concentrations at the leading edge of the pesticide-leaching front are usually very variable. Given the variability occurring within the field, current predictions of the fluxes should be considered as uncertain.

Notwithstanding this uncertainty in absolute prediction, current models are successful in screening the potential mobility of pesticides. In addition, if a deterministic model is technically sound and if it gives sufficient support to the user during the parameterisation process, then it will often predict the observed experimental data within the range of variability. This was clearly demonstrated with the simulations at the Vredepeel, the Tor Mancina, and the Weiherbach field sites. In the case of the Brimstone dataset, the drain flow measurements integrated the spatial variability over the whole plot. The macropore flow model MACRO predicted five times too large concentrations in the drainflow, although the timing of the breakthrough was remarkably well predicted.

Further scientific developments in pesticide-leaching modelling must seek to reduce the uncertainty in estimated fluxes. One way of dealing with uncertainty in management applications is to combine model predictions with available monitoring and experimental data, using Bayesian statistics to assign confidence levels to both. Another way is to

account directly for variability in input parameters in deterministic models. This would involve either a full stochastic treatment, or more simply and more practically, running of a ‘worst-case’, ‘average-case’ and ‘best-case’ simulations to try to cover the range of possible outcomes.

The lysimeter data, such as available in the Tor Mancina dataset, did allow us to assess correctly the leaching flux. Yet this flux does not represent the complete variability occurring within the field. Lysimeters are only local samples sampled from a heterogeneous agricultural field. In addition, the data available allowed us only to estimate flux properties in the upper part of the soil profile. Models could not be tested to estimate fluxes in the underlying sub-surface vadose zone. Further research must, therefore, envisage the upscaling of current know-how to the larger scales, say from the field scale up to the scale of the complete geo-pedological formation, or region.

### 3.3. Pesticide sorption and transformation

The models reported in Table 1 contain broadly similar descriptions of pesticide transformation and sorption on soil. Transformation can always be described with first-order kinetics with most models which allow the rate of transformation to be modified according to soil temperature and moisture content, following the Arrhenius law and the formulation proposed by Walker (1974). The latest version of PRZM2 enables a dual-phase transformation to be modelled by combining two first-order processes with different rate constants. In SIMULAT, seven approaches are available to describe the transformation of biodegradable material.

Observed patterns of transformation frequently do not simply follow the widely used first-order kinetics. Simulations based upon first-order kinetics often result in a small residue of pesticides that persists long after actual residues have been fully degraded. This is particularly significant in sub-soil layers where transformation is generally slower. Further, the Arrhenius law, which allows us to estimate parameters from transformation experiments in laboratory with varying temperatures, may be limited in describing field dissipation as well. Alternative approaches such as the O’Neill function (Richter et al., 1996) could remediate such a problem, in which an increase of microbial activity with increasing temperature and decreasing activity is considered, if an optimal temperature is reached or passed. Similar problems are present when using the Walker function as a humidity-response function to describe the rate dependency on soil water content. It is recommended to carry out well-designed incubation experiments, allowing one to estimate model parameters for a wide range of transformation models, and to examine in more detail the transfer of laboratory results to field studies. The datasets used in this exercise, however, did not allow us to estimate reliable parameters for more advanced transformation kinetics. With such limited information, the use of more complicated transformation models does not seem to be feasible yet.

There is a roughly an equal split between models that describe pesticide sorption with a linear isotherm and those that use a Freundlich isotherm (Table 1). In both cases, sorption is assumed to be fully reversible, and no hysteresis by desorption characteristic is considered. Two models (VARLEACH and PLM) allow for an increase in the sorption coefficient with time. Generally, sorption is assumed to be instantaneous.

But a number of the models (e.g., LEACHP, WAVE, SIMULAT) include parameters which simulate two-site sorption with a proportion of instantaneous sorption followed by a kinetic sorption process according to a first-order rate constant. SIMULAT offers the possibility to simulate linear, Freundlich and kinetic sorption at three different sorption sites.

Failure to describe hysteresis in adsorption/desorption isotherms and to provide rigorous descriptions of the increase in sorption with time, which is frequently observed, are important limitations for modelling pesticide sorption. However, the sorption descriptions built into the various models are based upon the data that are derived from standard laboratory experiments. As such, most of the parameters are readily measurable and the impact of each on simulations of pesticide-leaching is understood. Although more rigorous descriptions of pesticide sorption may become more important with time, it was felt by that the inclusion of more elaborate descriptions is presently unjustified considering engineering applications.

An area in which modelling is still weak is that of predicting rates of transformation in sub-soils. Data are generally unavailable and modellers frequently resort to an assumed correlation between soil organic carbon content, microbial population, and the rate of transformation. An effect of increased availability of sorbed compounds in less organic sub-soil layers is sometimes also considered. Where leaching of chemicals out of the topsoil occurs, lack of knowledge about behaviour in the sub-soil is an important limitation. Generating data to allow prediction of half-lives in sub-soils from topsoil values is a research priority. It is worth noting that given the wide range of chemical properties relevant to pesticides, it is unlikely that a general applicable rule could be developed.

Two environmental processes that may be important in some cases are plant uptake and volatilisation. Plant uptake is considered by many models although VARLEACH, SIMULAT and PLM are exceptions. Uptake is often simulated using a factor to describe the relative uptake of the compound into the plant roots compared to the removal of water. A value of 1 would equate to simple mass flow with the uptake of water. Although included in many models, plant uptake is often not simulated because of a lack of information for specific compounds. The process will have limited relevance for autumn-applied compounds where any leaching will occur over the winter months when plant growth is slow. However, uptake of the bromide tracer by plants may account for a significant proportion of amounts applied to soil. For this tracer solute and for those pesticides that are more readily taken up, plant uptake should be given greater attention.

Volatilisation of pesticides is simulated by approximately half of the models using either Henry's constant, or vapour pressure. The effective diffusion coefficient in air is also often required. As most of the loss under field conditions has been shown to occur within the first week after application, models that do not simulate volatilisation can be 'corrected' by using an application rate adjusted for the expected loss. Relative to the other processes included in the various models, volatilisation has received relatively little research attention. Selection of input parameters can make it difficult to simulate this process from first principles. Further work on volatilisation is required to increase confidence in model predictions.

### 3.4. *Good modelling practice in pesticide-leaching modelling*

Good modelling practice (GMP) has been recently defined as the development, maintenance, distribution, and use of computer simulation models whereby the integrity of the model, its various improvements, and utilisation is assured. The overall objective of GMP's is the complete transparency of all steps of the modelling process (Resseler et al., 1997). The modelling exercise has to be made transparent, understandable, and repeatable by other experienced user. GMP's should be used by the model developer, the model maintainer, and the model user.

The main objective of a GMP is to describe what has been done, to justify why it has been done, and to document the modelling process such that the model user and any independent persons may repeat the modelling exercise and achieve the same result (Travis, 1995). It is envisaged that one can attach a quality label to the modelling effort. It is recommended that the model user receive the following responsibilities for obtaining a GMP.

#### 3.4.1. *The model user is responsible for understanding the model and its appropriate usage*

This demands that the user should thoroughly understand the assumptions and limitations of the model. The assumptions should be examined with each new study to assure that the scenario being examined does not violate any of the model assumptions. The scenario should also be examined to confirm that it is within the limitations of the model. The model user should choose geographical areas, weather information, chemical properties, and environmental conditions that adequately represent the scenario being simulated.

#### 3.4.2. *The model user is responsible for estimating the model parameters and the input for a selected scenario*

When input information is unavailable, appropriate indirect estimates of model parameters and the model input procedure should be selected. The uncertainty of the estimate on the modelling results must be characterised. This may involve performing simulations over a range of values for the uncertain input parameter. Model outputs must be examined to confirm that the results are reasonable. Unjustified extrapolation of model results must be avoided.

#### 3.4.3. *The model user must further keep in touch with accepted versions of the model and model documentation*

The user is responsible for assuring source-code integrity. Any changes to the source code should be documented in the modelling report. Modified versions of the source code must be identified clearly to maintain model integrity in the public domain.

#### 3.4.4. *The user is finally responsible for developing modelling reports that contain sufficient information for an independent person to reproduce the results*

The report should clearly state the objectives of the modelling study and identify both the model and the version used. It should give a detailed description of the adopted

dataset and provide a detailed report on the parameter and input estimation procedure. The calibration procedure should be described and give a detailed evaluation and interpretation of the modelling results. If the model was modified, a copy of the modified source code should be included with the report.

Although a range of laboratory and field data was available for the present exercise, it was concluded that the subjectivity in the process of parameter estimation and input selection is critical in pesticide-leaching modelling (Boesten, 2000). It is, therefore, recommended that expert judgement on model parameters should be given by the provider of the dataset to improve the GMP. It was felt that model calibration only allows us to do a 'cold validation'. In such a case, the extrapolative capacity of the model still has to be demonstrated with independent modelling results. Calibration should, therefore, be avoided as much as possible. Changing of the initial laboratory data to match observed field data should not be carried out unless a sound basis for such a change is given.

#### **4. Conclusion**

Within this paper, the methodology of a detailed pesticide-leaching modelling test was presented. Twelve models were tested by 36 model users, using experimental data collected at four different sites. An attempt was made to qualify both the models and model users. To our knowledge, this test is the most extensive that has been carried with pesticide-leaching models in Europe so far. In addition to the presentation of the adopted methodology, major conclusions of the modelling test are summarised. Recommendations are formulated for improving the quality of modelling datasets, the process description of water, solute, and heat transport in a pesticide-leaching model, plus the process description of pesticide fate. The strong impact of the modeller on the modelling results, requires also the development of robust techniques which can easily be applied in an engineering context. A plea is therefore made to the application of the principles of GMP when modelling pesticide behaviour in a regulatory context.

#### **Acknowledgements**

This study was carried out within the framework of the mathematical modelling working group of COST Action 66 'Pesticide fate in the soil environment' organised by DGXII of the EU. Special thanks goes to all modellers and dataset providers who contributed to the exercise. Their work, which was discussed in the different workshops, offered the materials that allow us to synthesise the present paper. B. Clothier (Palmerstone North Research Centre, New Zealand) and Mary Beth Kirkham (Kansas State University, USA) are acknowledged for their valuable comments on the manuscript.

#### **References**

- Aden, K., Diekkrüger, B., 2000. Modeling pesticide dynamics of four different sites using the model system SIMULAT. *Agric. Water Manage.* 44, 337–355.

- Armstrong, A., Aden, K., Amraoui, N., Dieckrüger, B., Jarvis, N., Mouvet, C., Nicholls, P., Wittwer, C., 2000a. Comparison of pesticide leaching models: results using the Brimstone Farm data set. *Agric. Water Manage.* 44, 153–181.
- Armstrong, A.C., Matthews, A.M., Portwood, A.M., Jarvis, N.J., Leeds-Harrison, P.B., 2000b. CRACK-NP: A pesticide leaching model for cracking clay soils. *Agric. Water Manage.* 44, 183–199.
- Bergström, L.F., Jarvis, N.J., 1994. Evaluation and comparison of pesticide leaching models for registration purposes. *J. Environ. Sci. Health* 29, 1061–1072.
- Boesten, J.J.T.I., Gottesbüren, B., 2000. Testing PESTLA by two modellers for bentazone and ethoprophos in a sandy soil. *Agric. Water Manage.*, this issue.
- Boesten, J.J.T.I., Businelli, M., Delmas, A., Edwards, V., Helweg, A., Jones, R., Klein, M., Kloskowski, R., Layton, R., Marcher, S., Schäfer, H., Smeets, L., Styczen, M., Russell, M., Travis, K., Walker, A., Yon, D., 1995. Leaching models and EU registration. The final report of the work of the Regulatory Modelling Work Group of FOCUS, Forum for the Co-ordination of pesticide fate models and their use. Guidance document 4952/VI/95, Commission of the European Communities, DGVI, Brussels, 123 pp.
- Boesten, J.J.T.I., van der Pas, L.J.T., 2000. Movement of water bromide ion and the pesticides ethoprophos and bentazone in a sandy soil: description of the Vredepeel dataset. *Agric. Water Manage.* 44, 21–42.
- Boesten, J.J.T.I., 2000. Modeller subjectivity in estimating pesticide parameters for leaching models using the same laboratory data set. *Agric. Water Manage.* 44, 389–409.
- Brown, C.D., Carter, A.D., Hollis, J.M., 1995. Soils and pesticide mobility. In: Roberts, T.R., Kearny, P.C. (Eds.), *Environmental Behaviour of Agrochemicals*. Wiley, pp. 132–184.
- Camase, 1996. Register of agro-ecosystems models. DLO Research Institute for Agrobiology and Soil Fertility, Wageningen, the Netherlands. <http://www.bib.wau.nl/camase>
- Council of the European Union, 1997. Proposal for a council directive establishing annex VI to directive 91/414/EEC concerning the placing of the plant protection products on the market. *Official Journal of the European Communities* C240, 1–23.
- Dieckrüger, B., Söndgerath, D., Kersebaum, K.C., McVoy, C.W., 1995. Validity of agro ecosystems models: a comparison of results of different models applied to the same data set. *Ecol. Modelling* 81, 3–29.
- Dust, M., Baran, N., Errera, G., Hutson, J.L., Mouvet, C., Schäfer, H., Vereecken, H., Walker, A., 2000. Simulation of water and solute transport in field soils with the LEACHP model. *Agric. Water Manage.*, this issue.
- EMWG, Exposure modelling working group, 1995. Primary, secondary and screening models for pesticide registration. Report. The FIFRA modeling Work Group. Washington, USA.
- Faasen, R., 1995. Agricultural pesticide use... a threat to European environment? *Eur. Water Pollut. Control* 5, 33–40.
- Francaviglia, R., Capri, E., Klein, M., Hosang, J., Aden, K., Trevisan, M., Errera, G., 2000. Comparing and evaluating pesticide leaching models: results for the Tor Mancina data set (Italy). *Agric. Water Manage.* 44, 135–151.
- Francaviglia, R., Capri, E., 2000. Lysimeter experiments in Tor Mancina (Italy). *Agric. Water Manage.* 44, 63–74.
- Freijer, J.L., Tiktak, A., Hassanizadeh, S.M., van der Linden, A.M.A., 1996. PESTRAS v3.1: A one-dimensional model for the assessment of the fate of pesticides in soil. Report 715501007. National Institute of Public Health and the Environment, Bilthoven, Netherlands, 130 pp.
- Gottesbüren, B., Aden, K., Bärlund, I., Brown, C., Dust, M., Görlitz, G., Jarvis, N., Rekolainen, S., Schäfer, H., 2000. Comparison of pesticide leaching models: results using the Weiherbach data set. *Agric. Water Manage.* 44, 153–181.
- Harris, G.L., Catt, J.A., Bromilow, R.H., Armstrong, A.C., 2000. Evaluating pesticide leaching models: the Brimstone farm dataset. *Agric. Water Manage.* 44, 75–83.
- IUPAC, 1987. IUPAC report on pesticides: potential contamination of groundwater by pesticides. *Pure Appl. Chem.* 10, 1419–1446.
- Jarvis, N., Brown, C., Granitzka, E., 2000. Sources of error in model predictions of pesticide leaching: a case study using the MACRO model. *Agric. Water Manage.* 44, 247–262.
- Jarvis, N.J., Bergström, L.F., Brown, C.D., 1995. Pesticide leaching models and their use for management purposes. In: Roberts, T.R., Kearny, P.C. (Eds.), *Environmental Behaviour of Agrochemicals*. Wiley, pp. 196–220.

- Jury, W.A., Ghodrati, R., 1989. Overview of organic chemical environmental fate and transport modeling approaches. In: *Reactions and movement of organic chemicals in soils*, Vol. 22. Soil Science Society of America, Special publication, pp. 271–304.
- Klein, M., Hosang, J., Schäfer, H., Ergräber, B., Ressler, H., 2000. Comparing and evaluating pesticide leaching models. Results of simulations with PELMO. *Agric. Water Manage.* 44, 135–151.
- Loague, K., Yost, R.S., Green, R.E., Liang, T.C., 1989. Uncertainty in a pesticide leaching assessment for Hawaii. *J. of Contam. Hyd.* 4, 139–161.
- Leistra, M., Boesten, J.J.T.I., 1989. Pesticide contamination of groundwater in Western Europe. *Agric. Ecosyst. Environ.* 26, 269–389.
- Nash, J.E., Sutcliffe, J.V., 1970. River flow forecasting through conceptual models. 1. Discussion of principles. *Journal of Hydrology* 10, 282–290.
- Nicholls, P.H., Harris, G.L., Brockie, D., 2000. Simulation of pesticide leaching at Vredepeel and Brimstone farm using the macropore model PLM. *Agric. Water Manage.* 44, 307–315.
- Pennel, K.D., Hornsby, A.G., Jessup, R.E., Rao, P.S.C., 1993. Evaluation of five simulation models for predicting aldicarb and bromide under field conditions. *Water Resource Res.* 26(11), 2679–2693.
- Rekolainen, S., Gouy, V., Francaviglia, R., Eklo, O.M., Bärlund, I., 2000. Simulation of soil water bromide and pesticide behaviour in soil with the GLEAMS model. *Agric. Water Manage.*, this issue.
- Ressler, H., Schäfer, H., Görlitz, G., Hermann, M., Hosang, J., Kloskowski, R., Marx, R., Sarafin, R., Stein, B., Winkler, R., 1997. Recommendations for conducting simulation calculations for the registration procedure. *Nachrichtenblatt des Deutschen Pflanzenschutzdienstes* 49(12), 305–309.
- Richter, O., Dieckrüger, B., Nörtersheuser, P., 1996. *Environmental Fate Modelling of Pesticides: From the Laboratory to the Field Scale*. VCH, Weinheim, 281 pp.
- Schierholz, I., Schäfer, H., Kollé, K., 2000. The Weiherbach data set: an experimental data set for pesticide model testing on the field scale. *Agric. Water Manage.* 44, 43–61.
- Thorsen, M., Feyen, J., Styczen, M., 1996. Agrochemical modelling. In: Abbott, M.B., Refsgaard, J.C. (Eds.), *Distributed Hydrological Modelling*. Kluwer Academic Publishers, Dordrecht, pp. 121–141.
- Tiktak, A., 2000. Application of nine pesticide leaching models to the Vredepeel dataset. II Pesticide fate. *Agric. Water Manage.* 44, 119–134.
- Trevisan, M., Errera, G., Goerlitz, G., Remy, B., Sweeney, P., 2000. PRZM-2 evaluation using Vredepeel dataset. *Agric. Water Manage.* 44, 317–335.
- Trevisan, M., Errera, G., Viscetti, A., Walker, A., 2000. Varleach evaluation. *Agric. Water Manage.*, this issue.
- Travis, K.Z., 1995. Recommendations for the correct use of models and reporting of modelling results. In: Boesten, J.J.T.I., Businelli, M., Delmas, A., Edwards, V., Helweg, A., Jones, R., Klein, M., Kloskowski, R., Layton, R., Marcher, S., Schäfer, H., Smeets, L., Styczen, M., Russell, M., Travis, K., Walker, A., Yon, D. (Eds.), *Leaching Models and EU Registration. The final report of the work of the Regulatory Modelling Work Group of FOCUS, Forum for the Coordination of pesticide fate models and their use*. Guidance document 4952/VI/95, Commission of the European Communities, DGVI, Brussels, 123 pp.
- Vanclooster, M., Boesten, J.J.T.I., 2000. Application of nine pesticide leaching models to the Vredepeel dataset. I Water and solute fate. *Agric. Water Manage.* 44, 105–117.
- Vanclooster, M., Ducheyne, S., Dust, M., Vereecken, H., 2000a. Simulating pesticide dynamics with the WAVE-model. *Agric. Water Manage.* 44, 371–388.
- Vanclooster, M., Boesten, J.J.T.I., Trevisan, M., 2000b. Mathematical modelling for assessing pesticide leaching in agricultural soils at the field scale. In: Jamet, P., Cornejo, J. (Eds.), *Methods in Pesticide Science*, in press.
- Van der Zee, S.E.A.T.M., Boesten, J.J.T.I., 1991. Effects of soil heterogeneity on pesticide leaching to groundwater. *Water Resource Res.* 27, 3051–3063.
- Walker, A., 1974. A simulation model for prediction of herbicide persistence. *J. Environ. Qual.* 3, 396–401.