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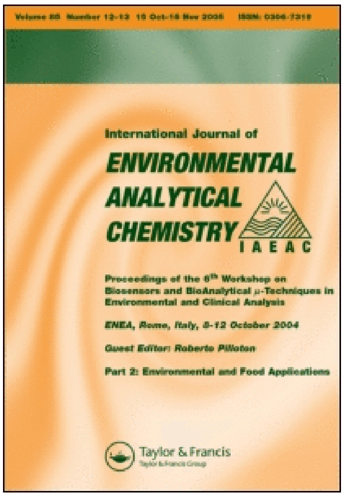

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Leaching of Pesticides from Clayey Soils. Comparison of Simulated and Determined Concentrations

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LEACHING OF PESTICIDES FROM CLAYEY SOILS. COMPARISON OF SIMULATED AND DETERMINED CONCENTRATIONS

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Leaching of the herbicides atrazine (6-chloro-N^2-ethyl-N^4-=isopropyl-1,3,5-triazine-2,4-diamine) and hexazinone (3-cyclohexyl-6-dimethylamino-1-methyl-1,3,5-triazine=2,4(1H,2H)-dione) was measured in samples taken from drainage water from two plantations with 7–10-year-old *Abies nordmanniana* (Steven) Spach grown on clayey soils.

Leaching of atrazine was almost the same in both plantations showing highest concentrations at the beginning of the 2-year sampling period. Leaching of hexazinone from the plantations showed fluctuating concentrations.

A relatively simple deterministic model, CMLS[1], was chosen to see if existing simulation models could simulate the observed concentrations.

An extension was added to the CMLS model in order to simulate dispersion of pesticides in soil. The simulation results of CMLS with dispersion are in better agreement with the observed concentrations.

KEY WORDS: Leaching, simulation, dispersion, pesticide, CMLS.

INTRODUCTION

Leaching of the herbicides atrazine (6-chloro-N^2-ethyl-=N^4-isopropyl-1,3,5-triazine-2,4-diamine), and hexazinone (3-cyclohexyl-6-dimethylamino-1-methyl-1,3,5-triazine-=2,4(1H,2H)-dione) from two clayey soils both situated on a glacier deposit in Denmark was studied[2]. Over a period of two years concentrations of herbicide in the leachate from the root zone were determined. The herbicides were chosen because they are relatively persistent[3,4] and only moderately adsorbed[5,6] by the soil, which means that there is a risk of leaching. In many countries atrazine is one of the most commonly detected pesticides in ground water[7,8,9]. Numerous models simulating the fate and transport through the unsaturated zone have been developed to predict the impact of soil tillage practices on ground water.

Melancon *et al.*[10], Sauer *et al.*[11] and Smith *et al.*[12] have all three evaluated the pesticide transport model PRZM on soil columns or soil cores from field plots applicated with atrazine.

Simulations of the CMLS model have been compared with field measurements of atrazine and hexazinone in leachate at the Department of Chemistry, University of Copenhagen and at the Department of Weed Control and Pesticide Ecology at the Danish Institute of Plant and Soil Science.

EXPERIMENTAL METHODS

Two plantations of *Abies nordmanniana* (Steven) Spach both situated on a flat, temporarily saturated clayey soil were selected for the leaching study[2]. The plantations situated at Bremersvold and Køge have both been sprayed with atrazine approximately 3.6 kg active ingredient per hectare, respectively in April 1984 and April 1982.

In the spring from 1985 onwards both plantations have been sprayed with hexazinone, approximately 2 kg active ingredient per hectare. In the period from April 1988 to March 1990 when the soils were saturated water samples were taken below soil surface from 100–125 cm via three tubes. They were filtered, extracted and concentrated immediately after sampling. The contents of atrazine and hexazinone and some of the principal degradation products of the herbicides were measured by gas chromatography/mass spectrometry (GC/MS)[2].

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MODEL DESCRIPTION

The relatively simple model, CMLS (Chemical Movement in Layered Soils) developed by Nofziger and Hornsby[1], was selected for comparison of simulated and determined concentrations. One of the assumptions in the model is transport in a coarse grained soil in the unsaturated zone. The soil textural class in the plantations is a sandy loam/sandy clay loam, and the pesticide transport will be a combination of unsaturated and saturated transport, because the soil is temporarily saturated during the winter. These deviations from the model assumptions imply that the pesticide front will be delayed in relation to the simulated time. Since the number of input data is small it was possible to determine many of the input data instead of using values from the literature. In connection with comparison of the determined and simulated concentrations it was necessary to extend the model with simulation of dispersion and diffusion, which was done using Fick's 2nd law. The dispersion and diffusion coefficients were calculated as described by Leistra[13,14].

Dispersion and diffusion

The pesticide front will disperse in the vertical direction because pesticides diffuse upwards or downwards in the soil water and more importantly, because the flow of water which transports the pesticides in the soil is not uniform, but is dispersed around the mean flow rate as a result of the differences in size and directions of the pores in the soil.

Diffusion and dispersion can be described by Fick's 2nd law,

$$\frac{\delta C}{\delta t} = - D_{disp} \frac{\delta^2 C}{\delta x^2} - D_{dif} \frac{\delta^2 C}{\delta x^2} = - D \frac{\delta^2 C}{\delta x^2}$$

where

C:	concentration of pesticide in soil water, $\mu g \cdot l^{-1}$
t:	time, d
D_{disp}:	coefficient of convective dispersion, $cm^2 d^{-1}$
x:	depth below surface, cm
D_{dif}:	coefficient of diffusion in soil water, $cm^2 d^{-1}$
D:	sum of D_{disp} and D_{dif}.

Only the sum of the diffusion coefficient (D_{dif}) and dispersion coefficient (D_{disp}) is needed in the simulation. We assume that diffusion in the soil air is unimportant for the pesticides considered here.

The magnitude of D_{disp} and D_{dif} can be estimated from the following equations given by Leistra[13,14]:

$$D_{disp} = L_d \cdot |J_l| \qquad and\ D_{dif} = \tau \varepsilon D_{diff}$$

where

L_d:	dispersion length, cm
J_l:	infiltration rate, $cm \cdot d^{-1}$
τ:	tortuosity factor
ε:	soil water content.

Using a dispersion length L_d of 5 cm and an infiltration rate J_l of 0.05 $cm \cdot d^{-1}$ as an average over both time and depth, D_{disp} will be 0.25 $cm^2 d^{-1}$. For the tortuosity parameter τ Leistra used values between 0,03 and 0,5, while the water content of the soil ε is between 0.1 and 0.4. As the diffusion coefficient for neutral organic compounds in water D_{diff} is around 0.5 $cm^2 d^{-1}$, the resulting estimate of D_{dif} is 0.0015 to 0.1 $cm^2 d^{-1}$.

These assumptions give a sum D of between 0.25 and 0.35 $cm^2 d^{-1}$ where dispersion is much more important than diffusion. Since the simulation uses the total concentration of pesticide in each depth (adsorbed plus dissolved) the effective D is obtained by multiplication with the ratio between the amount of pesticide in the water phase and the total amount of pesticide in each depth. This ratio varies between 0.04 (for atrazin in the upper horizon under wilting conditions) and 0.94 (for hexazinon in the deepest horizon at a water content equal to the field capacity). The effective D to be used in the simulation is thus within the range 0.01 to 0.35 $cm^2 d^{-1}$ and a value of 0.2 $cm^2 d^{-1}$ was arbitrarily chosen for most simulations as representative for conditions with a dispersion in the high end of the normal range.

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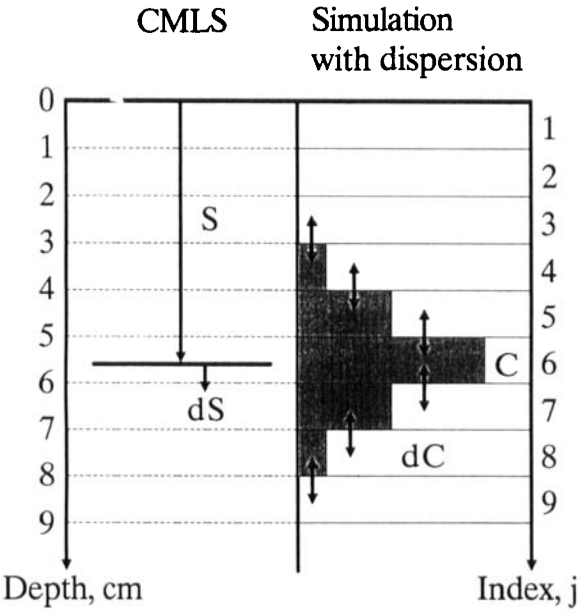
In the simulations we have kept D constant in order to keep the model as simple as possible. The dispersion coefficient is a function of the infiltration rate, and the diffusion coefficient is a function of soil water content, but we do not have experimental determined parameters (L_d and τ) for the actual soil. Introducing too many parameters in a model results in a better fit to field observations, but the interpretation becomes less meaningful. The dispersion is also introduced here in order to simulate dispersion due to upward water movements and macropore flow, which is not taken into account by CMLS. If simulation of dispersion is related closely to water movement as calculated by CMLS, any errors in the CMLS calculations are amplified. Work on a more complex and complete model is in progress.

Simulation with dispersion

The soil is divided into horizontal volume elements with a thickness of 1 cm, numbered with index j, and for each day the CMLS position S and total concentration C_j for all j is calculated in the following way, see scheme 1:



Step 0 Initialize all values

$S = 0$; C_0 = Initial concentration; $C_j = 0$ for $j > 0$

For each day the following steps are calculated:

Step 1 Calculate the depth according to CMLS

$dS = q/((1 + bK_d/Q_{fc})Q_{fc})$

$S_{new} = S + dS$

where q is the infiltration (cm), b is bulk density ($\mathrm{kg\,l^{-1}}$, K_d is the partition coefficient ($\mathrm{l\,kg^{-1}}$), and Q_{fc} is the field capacity ($\mathrm{l\,l^{-1}}$.).

Step 2 When the CMLS depth S passes one of the borders between the volume elements all concentrations are moved to the next volume element:

$$C_j = C_{j-1}$$

Step 3 Calculate dispersion according to the discrete expression of Fick's 2nd law (see Feldberg 1969[15] for an excellent description of digital simulation):

$$dC_j = D((C_{j+1}-C_j)-(C_j-C_{j-1}))$$

$$C_{newj} = C_j + dC_j$$

Step 4 Calculate the degradation of the pesticide for each j (k is equal to $ln2/T_{½}$ at the depth j)

$$C_j = C_j exp(-k)$$

INPUT DATA

The input data can be divided in soil parameters, pesticide parameters, hydrological parameters and dispersion parameters.

Soil parameters

The soil properties may vary through the profile. CMLS can divide the profile in up to 30 layers, within each horizon the properties are assumed to be uniform.

The following data are needed for each horizon: depth of bottom of horizon, percent organic carbon, bulk density and volumetric water content (%) at: the wilting point (–1.5MPa), field capacity (–0.01MPa) and saturation (all pores of the soil are filled).

Pesticide parameters

The adsorption of a pesticide in soil is described by the partition coefficient K_{oc}. When a literature value of K_{oc} is given, the model will normalize the value on organic carbon for each horizon. Values for measured sorption coefficients can be given for each layer. Values for degradation half-lives ($T_{½}$) can also be given for each horizon.

The method used to obtain the adsorption of atrazine has been described by OECD[16]. Half-lives for atrazine in the surface soil has with a few exceptions been obtained as described by Helweg[17]. Unlabelled atrazine has been used and the half-lives are based on extractable residues[18].

The following pesticide related data are needed: application depth of pesticide, application date, the date to end simulation and maximum rooting depth of plant.

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Table 1 Soil Input Parameters.

Horizon *cm*	*oc*[a] *w. %*	*b*[b] *kgl^{-1}*	*Q_w*[c] *vol. %*	*Qfc*[d] *vol. %*	*Porosity* *vol. %*
	Location: Bremersvold				
0–20	1.9	1.45	16.5	36.8	43.3
20–40	1.6	1.53	16.8	34.0	40.7
40–60	0.4	1.63	25.3	37.1	38.2
60–80	0.1	1.72	21.2	36.6	35.0
80–120	<0.05[e]	1.88	10.9	29.0	29.0
	Location: Køge				
0–20	1.3	1.52	11.1	32.7	41.4
20–40	0.3	1.66	12.8	28.5	37.0
40–80	0.1	1.76	18.4	29.5	33.4
80–120	<0.05[e]	1.68	14.3	32.4	36.7

[a]Organic Carbon. [b]Bulk density. [c]Water content at the wilting point. [d]Water content at field capacity. [e]Below the detection limit (0.05 %)

Hydrological parameters

Daily values for precipitation and evaporation are needed to calculate infiltration. Hydrological input data for the locations in the period from the first spraying (1982) to the end of sampling (1990) are used in the simulations.

Table 2 Pesticide Input Parameters.

	Atrazine[a]		*Hexazinone* *[b]		*Hexazinone* #[c]	
Horizon *cm*	*Kd* *lkg^{-1}*	*T½*[d] *days*	*Kd* *lkg^{-1}*	*T½*[d] *days*	*Kd* *lkg^{-1}*	*T½*[d] *days*
	Location: Bremersvold					
0–20	1.68	83	0.475	454	1.026	180
20–40	1.08	83	0.400	454	0.864	180
40–60	0.39	830	0.100	4540	0.216	1800
60–80	0.37	830	0.025	4540	0.054	1800
80–120	0.26	830	0.013	4540	0.027	1800
	Location: Køge					
0–20	1.71	133	0.325	454	0.702	180
20–40	0.48	1330	0.075	4540	0.162	1800
40–80	0.42	1330	0.025	4540	0.054	1800
80–120	0.35	1330	0.013	4540	0.027	1800

[a]K_d Values are determined experimentally on actual soil samples. $T_{½}$ values are determined in the upper horizon. Values for the half-lives were obtained at 25°C with a moisture at 90% of the field capacity. The values have been converted into half-lives at 10°C, which is the average temperature 10 cm below soil surface[19].

[b]Literature values[20,21]. K_d calculated from K_{oc} and *oc*[e].

[c]Literature values[22]. K_d calculated from K_{oc} and *oc*[f].

[d]As the biomass is very important for the degradation of chemical substances we have chosen to let the values for half-lives follow the content of organic matter, like that a low content of organic matter cause that the half-lives obtained in soil surface are multiplied by 10 when they are used to simulate half-lives in subsurface soils (personale communication, Helweg).

[e]The half-live has been determined at 10°C.

[f]The half-live was determined at 20–25°C and has therefore been converted into a half-live value at 10°C.

Dispersion parameters

The calculation of dispersion and diffusion uses one single parameter D (cm^2d^{-1}), which represents the average of the sum of the dispersion coefficient and the coefficient of diffusion for all horizons, see section 3.1.

The actual input parameters are summarized in Table 1 and 2. The net precipitation is calculated from measured precipitation and estimated evapotranspiration[19] and are shown in Figures 4.a and 4.b for the two locations.

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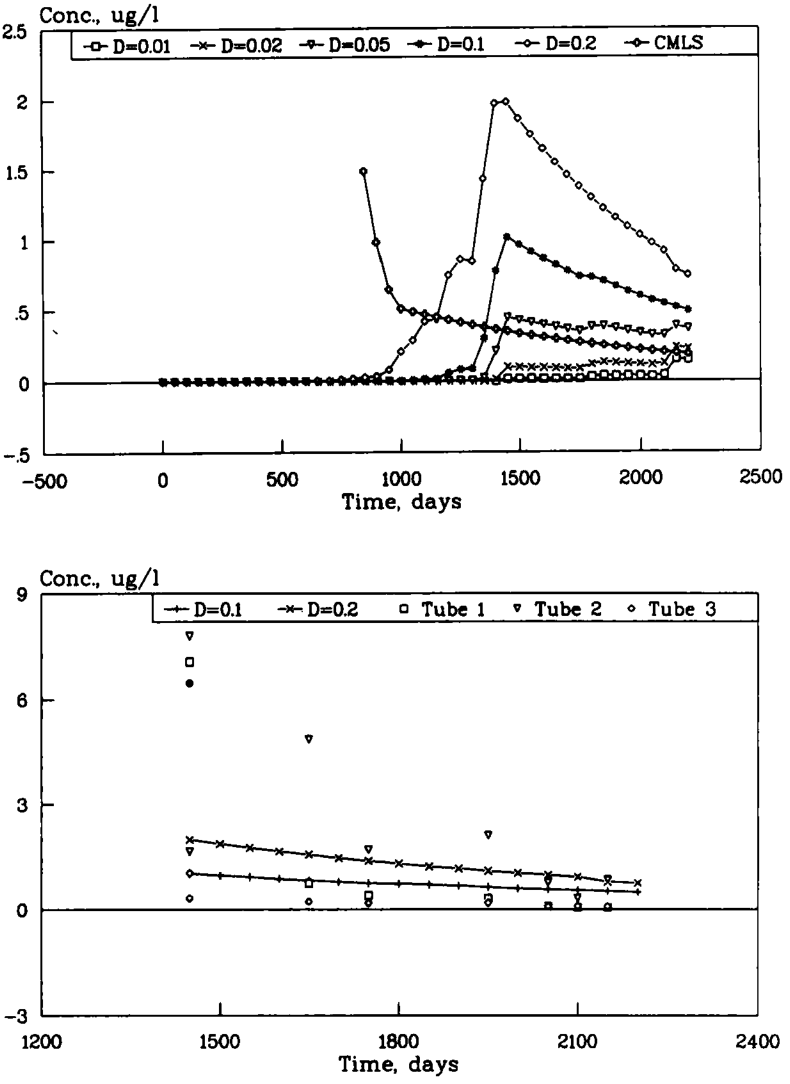

Figure 1 Concentrations of atrazine in soil water from Bremersvold at 100–125 cm calculated with varying dispersion. The content of atrazine in the water samples was measured in the period from 1459 days to 2146 days. A) Dispersion from 0.0 (CMLS) to 0.2 cm^2day^{-1}. B) Comparison of calculated and determined concentrations.

RESULTS AND DISCUSSION

Atrazine

Figure 1.a shows the simulated concentrations of atrazine in water samples, taken 100–125 cm below soil surface at Bremersvold, calculated with a dispersion from 0.01 cm^2d^{-1} to 0.2cm^2d^{-1}. The total concentration calculated with CMLS is also given in the figure. Figure 1.b shows the simulated and determined concentrations (0.06 to 7.79 $(\mu gl^{-1})^2$ for the sampling period. The results in this case show that a large dispersion of the pesticide will cause an increased concentration in the water samples.

This appears clearly from Figure 2.a, where the simulated concentration profiles are shown for atrazine in the soil from Bremersvold. Even though the position of the pesticide is the same for all 5 curves (since it has been calculated with CMLS) the highest concentration was observed deeper down, when dispersion is great, owing to the fact that degradation under the plough layer is slower than in it. The greater the dispersion the greater the amount of the pesticide found in the subsurface soil, where degradation is slow. Our simulations with dispersion indicate that the decrease in concentration, seen from the first determination in April 1988 to the last determination in March 1990, is more likely coursed by to be due degradation than measuring the tail of the pesticide front. Figure 2.b shows how the concentration profiles calculated with D=0.2 cm^2d^{-1} will change in the measuring period as a function of time. This shows that there is no change in the position of the pesticide until very late in the period and that the decreasing concentration primarily arises from degradation of atrazine rather than leaching.

Calculating the amount of atrazine leached from the soil profile at Køge using various dispersion coefficients—Figure 3.a—gives the expected progress. When the pesticide front according to CMLS passes the sampling depth (100–125 cm below soil surface) the highest concentration will be achieved with the lowest dispersion. When the pesticide front is either above or below the sampling depth, then the concentration in the sampling depth is highest with the highest dispersion. Figure 3.b shows that simulations with dispersion gave the same variation as observed and that the differences between the determined values (0.09 to 3.95 $\mu gl^{-1})^2$ and simulated values are significantly lower with dispersion than without it.

Figure 4 shows a survey of the excess precipitation and the simulated values for leaching depth, total concentration and the determined atrazine content. This shows that leaching of atrazine in subsurface soil closely follows the net precipitation which means that the pesticide front is stationary for long periods.

Simulation with dispersion supports the finding that the depths which CMLS calculates for the atrazine leaching, can very well be correct as the relative variation in concentrations with time are reproduced, while simulated concentrations still deviate from the determined concentrations. This indicates that it is the variation of the half-lives with depth (and the temperature) which are liable to the greatest uncertainty.

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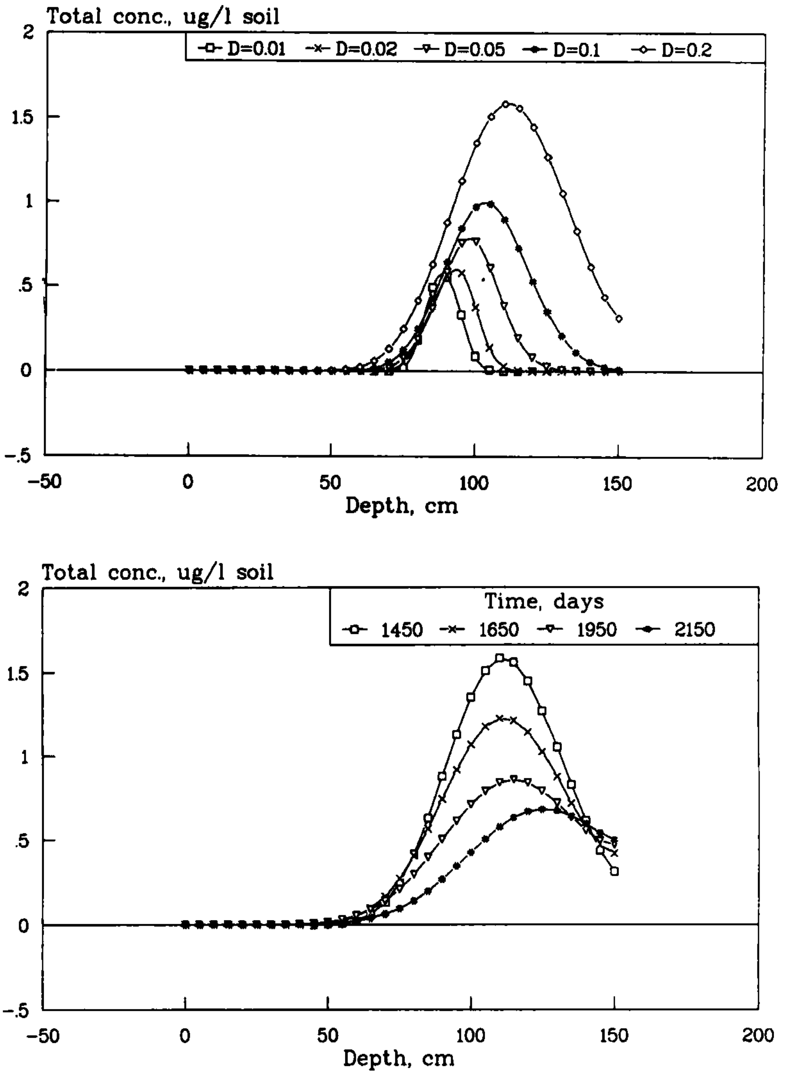

Figure 2 Calculated concentration profiles of atrazine in soil at Bremersvold as function of depth. A) Profiles corresponding to the time of the first sampling. B) Profiles during the sampling period.

Hexazinone

Simulation with dispersion (*D*=0.2 cm^2d^{-1}) of hexazinone leached from locality Bremersvold with two sets of parameters (marked # and *) are shown in Figures 5.a and 5.b. The simulations with # parameters predict a comparatively uniform concentration of

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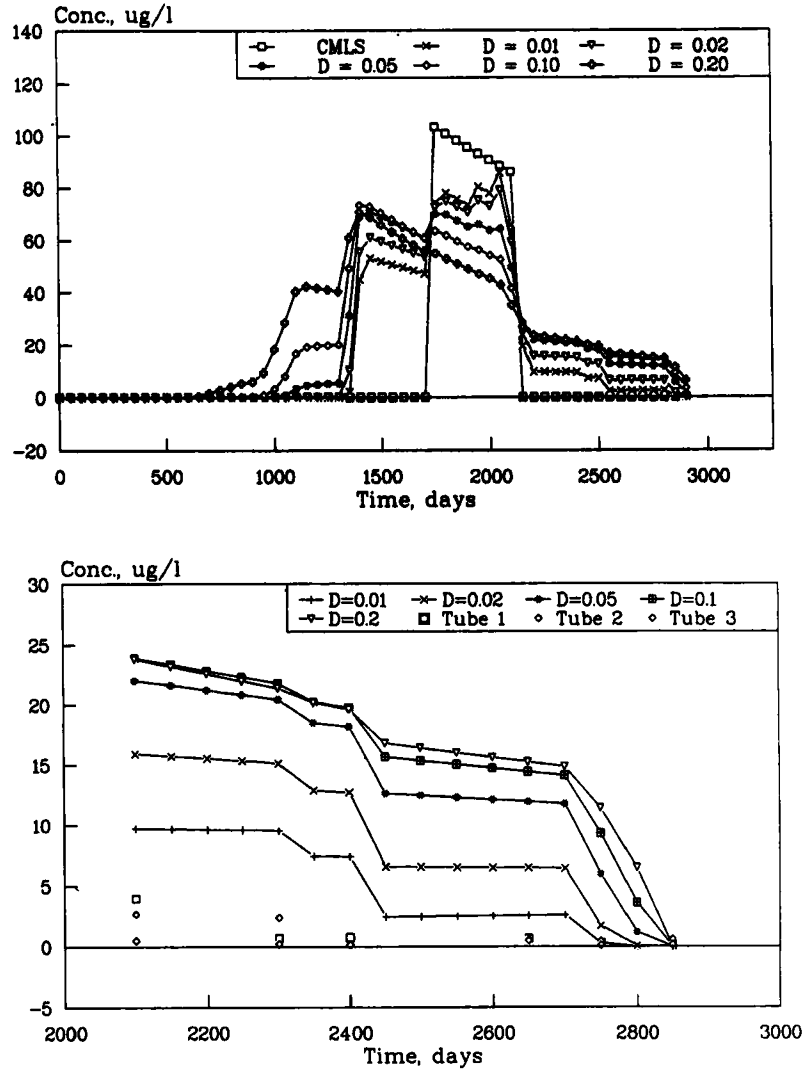

Figure 3 Concentrations of atrazine in soil water from Køge at 100–125 cm calculated with varying dispersion. The content of atrazine in the water samples was measured in the period from 2171 days to 2881 days. A) Dispersion from 0.0 (CMLS) to 0.2 cm^{2}day^{-1}. B) Comparison of calculated and determined concentrations.

hexazinone from 110 to 160 μgl^{-1} in the soil water samples taken below soil surface (100–125 cm) through the measuring period. Use of the * parameters results in higher and more fluctuating concentrations. The concentrations determined at Bremersvold (0.07 to 2.09 μgl^{-1})^2 are all much lower than the simulated ones and without an increasing or a decreasing tendency.

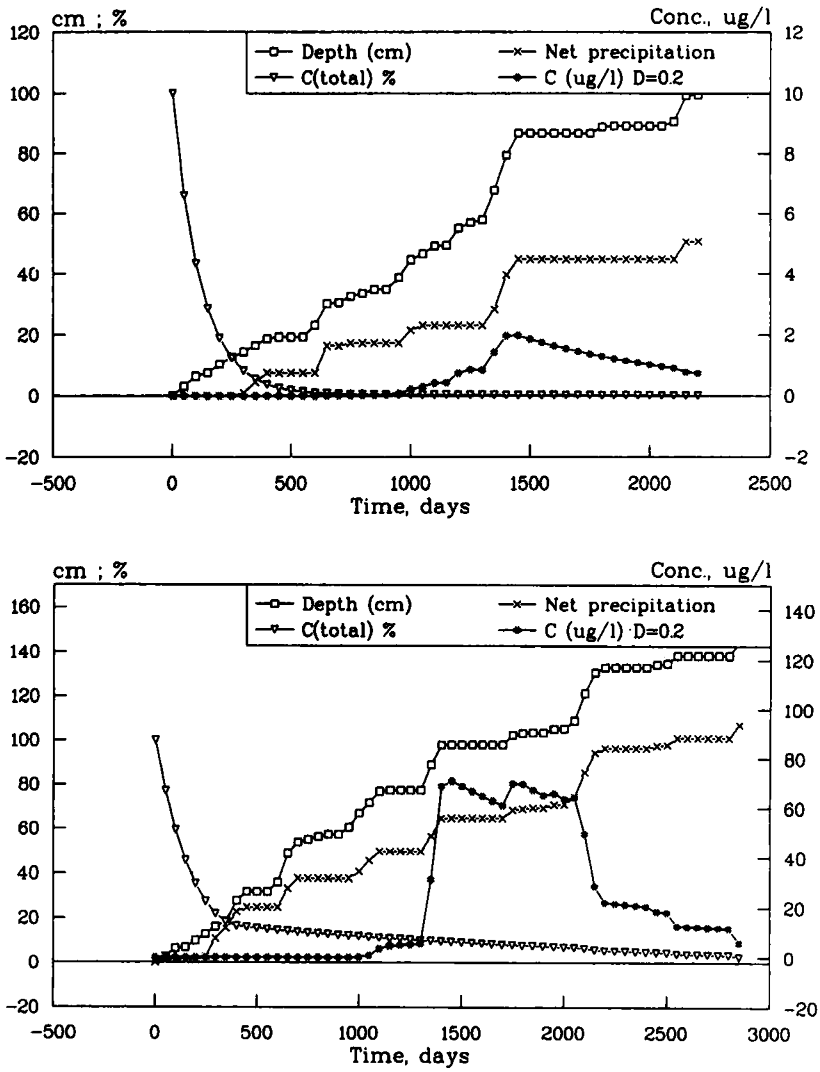

Figure 4 Relation between net precipitation and calculated values of leaching depth (CMLS), total concentration (CMLS) and concentrations in soil water samples (with dispersion D=0.2 $cm^2 day^{-1}$).

The concentrations in the soil profiles at the end of the sampling period are shown in Figures 6.a and 6.b.

It appears from these figures that it is impossible to evaluate if the leaching rate is calculated correctly, because neither a higher nor a lower leaching rate implies lower concentrations between 100 and 125 cm. The results will mainly differ from which year the

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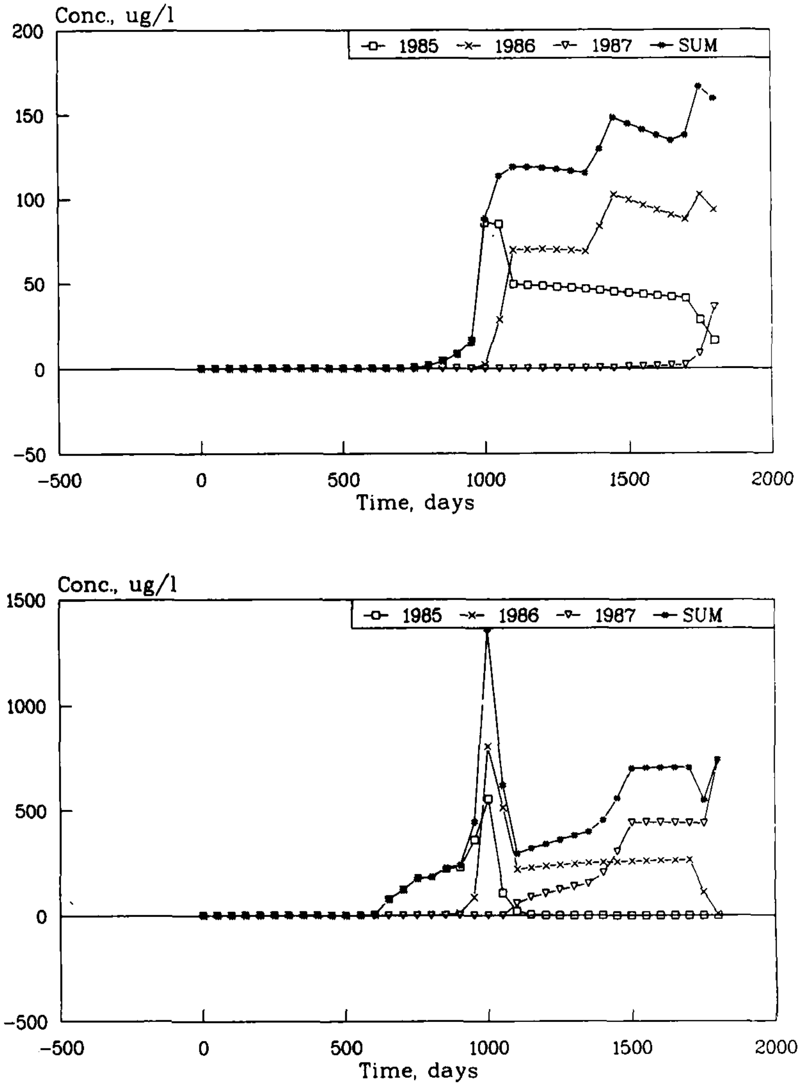

Figure 5 Calculated concentrations of hexazinone in soil water from Bremersvold at 100–125 cm calculated with $D=0.2\ cm^2 day^{-1}$ and A) #-parameters and B) *-parameters.

pesticide contribution derives. The same applies to calculations with a higher or lower dispersion. This will affect the extent of the merging of the pesticide contributions from the years concerned. The simulations with the # parameters are more in accordance with the real situation than calculations with the * parameters.

The simulations with dispersion for hexazinone leaching in the soil from Køge, (Figures 7.a and 7.b plus 8.a and 8.b), produce results, which are quite analogous to the results from

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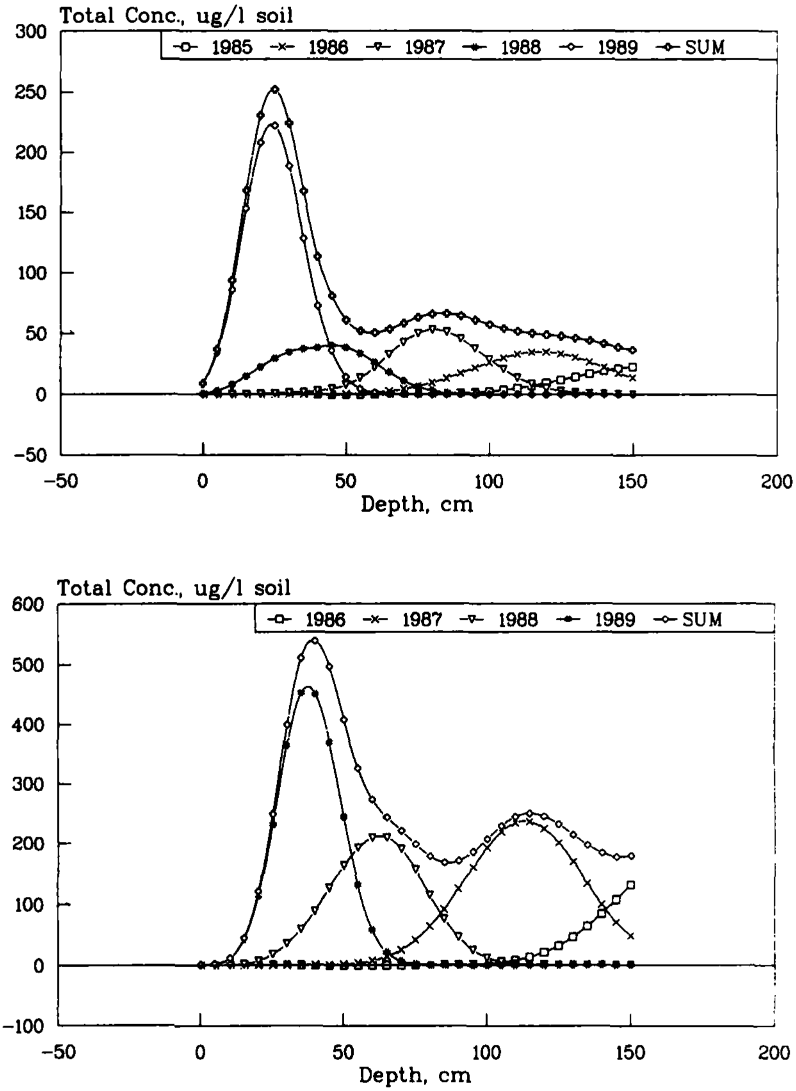

Figure 6 Calculated profiles of total concentrations of hexazinone in soil at Bremersvold for D=0.2 cm^{2}day^{-1} with A) #-parameters and B) *-parameters.

the Bremersvold soil. The simulated concentrations are more in accordance with the determined values (3.47 to 42.66 $(\mu g l^{-1})^2$ using # parameters than * parameters, but the simulated concentration level is overestimated.

How can these great differences for hexazinone be explained? The determined concentrations cannot be low, due to a quicker leaching in the soil than simulated, as it is inconceivable that pesticides used in the most recent years could pass the collection depth

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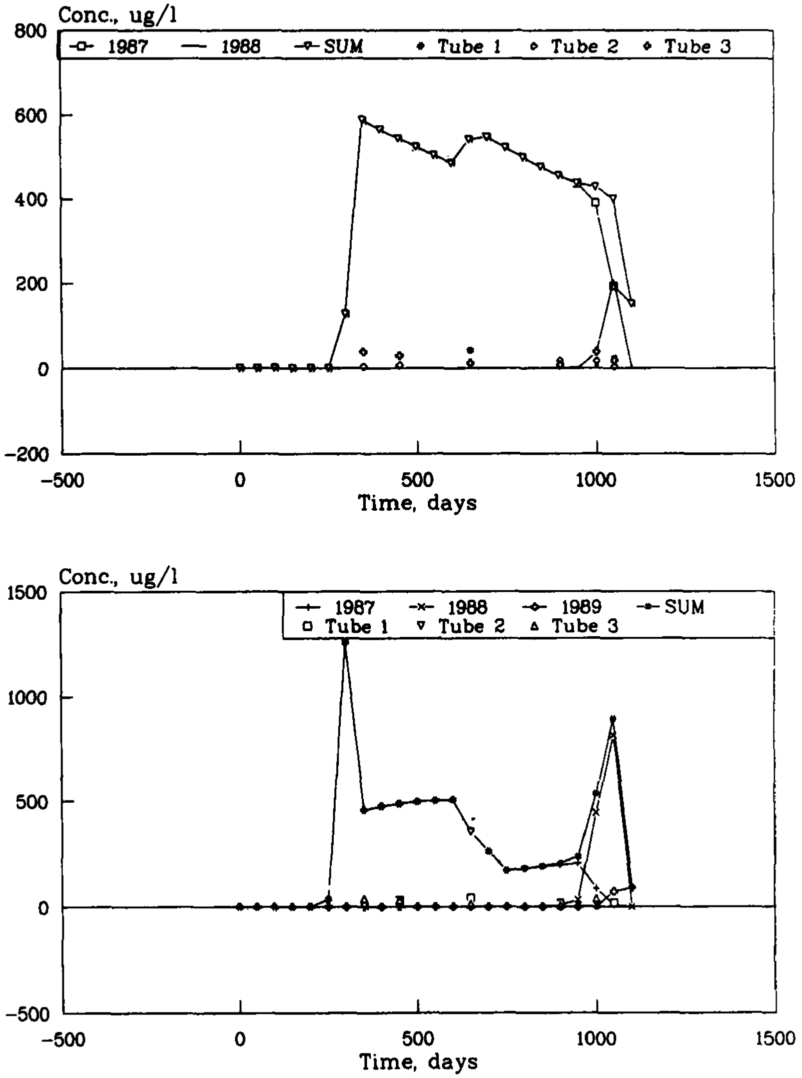

Figure 7 Calculated concentrations of hexazinone in soil water from Køge at 100–125 cm calculated with D=0.2 cm^{2}day^{-1} and A) #-parameters and B) *-parameters.

without being determined. If this after all was the case, the concentrations in the soil water below 125 cm should be extremely high.

However, if the transport is slower than simulated, the pesticides are retained for a longer time in the surface soil, where the degradation is fastest.

It is likely that this could be the case, as we have used K_d-values, which were calculated from K_{oc}-values and the soil content of organic carbon in the simulations. For atrazine where

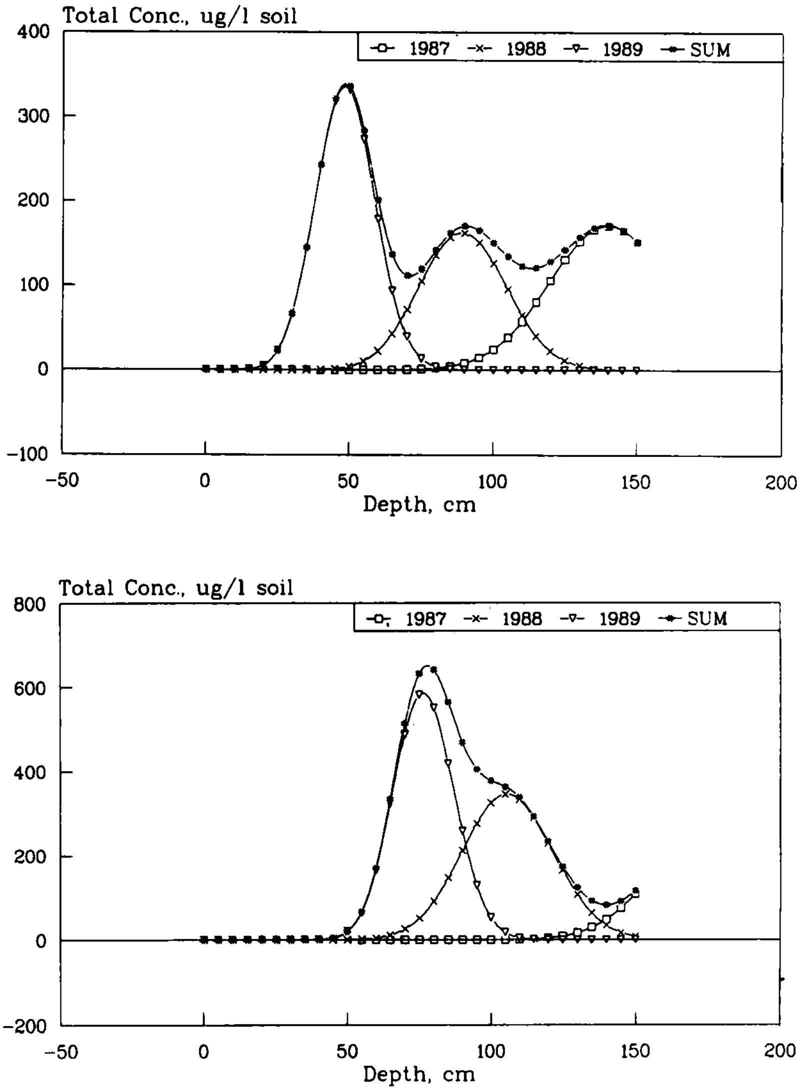

Figure 8 Calculated profiles of total concentrations of hexazinone in soil at Køge for D=0.2 $cm^2 day^{-1}$ with A) #-parameters and B) *-parameters.

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K_d has been determined in the actual soils it appears that this conversion gave K_d-values that were too low in the subsurface soil, where the content of organic carbon is very low.

Concerning the half-lives, which are subject to great variation, they often depend on whether the microorganisms have been exposed to the investigated pesticide or not. It is therefore possible that the actual half-lives in the soils from Køge and Bremersvold are lower than the values from the literature, which have been used for the simulations.

CONCLUSION

Simulation of the transport and degradation of atrazine through the soil profile using the model CMLS extended with dispersion and using experimentally determined values for the pesticide parameters K_d and $T_{½}$ give results which are in fairly good accordance with the determined values.

The conclusion that can be drawn from the simulations of hexazinone is that the determined concentrations can best be explained if the leaching of hexazinone is slower than simulated and that the actual half-lives are lower than the literature values.

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