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LEACHING OF DICHLORPROP AND NITRATE IN STRUCTURED SOIL

Lars Bergström*

Department of Soil Sciences Swedish University of Agricultural Sciences, PO Box 7072, 75007 Uppsala, Sweden

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ABSTRACT

Leaching rates of the herbicide dichlorprop [(±)-2-(2,4-dichlorophenoxy)propanoic acid] and nitrate were measured together in field lysimeters containing undisturbed clay and peat soils. The purpose of the study was to investigate the leaching pattern of the two solutes in structured soils under different precipitation regimes. Spring barley (Hordeum distichum *L.) was sown on each monolith and fertilized with 100 kg N ha^{-1}. Dichlorprop was applied at a rate of 1·6 kg active ingredient (a.i.) ha^{-1}. Each soil type received supplemental irrigation at two levels ('average' and 'worst-case'), giving total water inputs (irrigation and precipitation) of 664 and 749 mm year^{-1}, respectively. The larger water input approximately doubled the nitrate loads, from, on average, 11·6 to 21·8 kg N ha^{-1} year^{-1} in the clay soil and from 37·6 to 65·4 kg N ha^{-1} year^{-1} in the peat soil. In contrast, dichlorprop leaching was reduced by more than one order of magnitude when the water input was increased, from average amounts of 3·22 to 0·26 g a.i. ha^{-1} during an 3-month period in the clay and from 28·9 to 2·67 g a.i. ha^{-1} in the peat. This leaching pattern of dichlorprop was explained in terms of preferential flow. The dried-out topsoil of 'average' watered monoliths may have allowed water flow in cracks, thus moving some of the herbicide rapidly through the topsoil to the subsoil. Once the compound reached the subsoil, degradation rates would be reduced and the herbicide residues would be stored for later leaching. Nitrate was presumably more evenly distributed in the soil matrix; therefore, water rapidly moving through macropores would not carry significant amounts of nitrate. In contrast, leaching would occur more evenly through the soil matrix, causing larger nitrate loads in the 'worst-case' watered monoliths. These results show that wet years may constitute a worst case scenario in terms of nitrate leaching, but not pesticide leaching, if macropore flow exerts a significant influence on leaching.*

INTRODUCTION

Non-point source pollution resulting from agricultural activities and causing widespread pollution of surface waters and groundwaters has received considerable attention during recent years. Much effort has also been made to develop management strategies for reducing loads of agrochemicals to acceptable levels in these environments. However, some factors cannot be easily changed by human activities. Along with climate and geohydrological conditions, soil type is one such factor that exerts a substantial influence on the leaching of chemicals through soils (Bergström & Johansson, 1991). In this respect, sandy soils are usually considered to be very susceptible to leaching (Kissel *et al.*, 1982), mainly owing to their low water-holding capacities. However, substantial chemical movement may also occur in structured soils if continuous macropores allow rapid nonequilibrium flow (Thomas & Phillips, 1979).

Fast responses of percolating water to leaching of surface applications of various chemicals have been noted in several studies (e.g. Richard & Steenhuis, 1988; Everts *et al.*, 1989; Bergström *et al.*, 1990). Kladivko *et al.* (1991) showed that pesticide transport into tile-drains in a silt loam soil was typically event-driven, with peak concentrations occurring at the onset of each new flow period. In contrast, most of the nitrate transport, which was measured in the same study, occurred during extended drainflow periods when concentrations were consistently around 20–30 mg NO_3—N litre^{-1}. It is reasonable to assume that the observed difference in flow behaviour between the surface-applied pesticides and nitrate was due to the fact that exposure to preferential flow processes is greater for pesticides than for nitrate, the latter being more uniformly distributed in the soil matrix. It is also reasonable to assume that intensive rainfall would encourage preferential flow behaviour, so that pesticides are more likely to move through soil when amounts of precipitation are large.

The present study was aimed at investigating the leaching of dichlorprop [(±)-2-(2,4-dichlorophenoxy)-propanoic acid] and nitrate in structured soils under controlled environmental conditions. For this purpose, field lysimeters containing undisturbed soil profiles were used and treated in accordance with normal agricultural practices. In addition to differences in leaching behaviour between the two solutes, the influence of precipitation was followed by using two watering regimes.

* Present address: 116 Sandra Road, Wilmington, Delaware 19803, USA.

Table 1. Soil characteristics of the Lanna and Bälinge soils

Soil	Soil texture[a]	Organic matter (%)	CEC ($cmol_c kg^{-1}$)	Bulk density ($g\ cm^{-3}$)	$\theta_{1\cdot0}$[b] (%)	pH
Lanna						
Topsoil	Clay	3·8	28·4	1·21	46	7·2
Subsoil	Clay	0·0	33·6	1·42	43	7·4
Bälinge						
Topsoil	Peat	88·5	140·3	0·42	70	4·8
Subsoil	Peat	85·4	162·8	0·20	62	5·5

[a] According to the USDA soil classification system.
[b] $\theta_{1\cdot0}$ = Volumetric water content at a tension of 1 m water (estimated field capacity).

MATERIALS AND METHODS

Experimental design

Ten lysimeters containing undisturbed soil profiles were used. Five were filled with a clay soil (Lanna) and five with a peat soil (Bälinge). Some physical and chemical properties of the soils are listed in Table 1. The monoliths were enclosed in 1·18-m deep polyvinyl chloride (PVC) pipes (0·295 m inner diameter) by using a coring technique described by Persson and Bergström (1991). To collect monoliths, the lysimeter casing is inserted in a steel cylinder with four mounted cutting teeth at the bottom. The steel cylinder then rotates around the casing and carves out a soil column that is gently pushed into the casing. After collection, the lysimeters were prepared for free drainage by replacing ca 0·08 m of soil at the bottom of the monoliths with a nylon mesh, a gravel layer and a porous plastic sheet. The lysimeters were then placed in pipes (Fig. 1) permanently installed (below ground) at a lysimeter station in Uppsala, Sweden. One week before installation of the lysimeters in the station (19 April 1989), each monolith was watered until leachate was observed at the lysimeter bottoms. This was done to ensure that the soils were at field capacity shortly before the start of the experimental period. The preparation and installation of the lysimeters are described in more detail by Bergström and Johansson (1991).

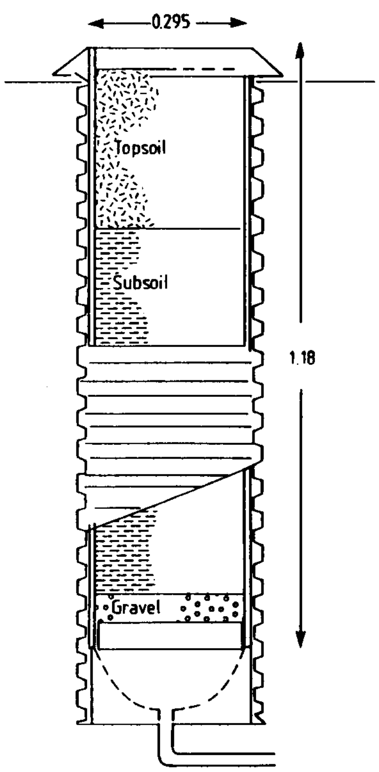

Fig. 1. Lysimeter placed in a below-ground pipe (from Bergström & Johansson, 1991).

Each of the lysimeters was cultivated and sown with spring barley (*Hordeum distichum* L.) on 16 May 1989. Four days prior to sowing (12 May) N-fertilizer was applied at a rate of 100 kg N ha^{-1} as $Ca(NO)_3$. Simultaneously, P and K were applied at a rate of 30 kg ha^{-1} each. All fertilizers were applied in solid form.

On 9 June 1989 (at GS21-25 according to the decimal code growth stages of cereals, Tottman, 1987), dichlorprop was applied at a rate of 1·6 kg active ingredient (a.i.) ha^{-1} using an atomizer and a spray volume of 400 litre ha^{-1} (2·74 ml $lysimeter^{-1}$). This rate represents the recommended normal dose of the compound under Swedish conditions.

The barley was harvested on 23 August 1989, whereupon the soils were hand-cultivated with a spade down to ca 0·2 m depth to simulate conventional ploughing.

Two different watering regimes were used: one corresponding to the long-term average for the Uppsala region (554 mm $year^{-1}$) and the other to the highest precipitation rate recorded in the region during the last 75 years (715 mm $year^{-1}$). Records of cumulative precipitation were compared every 2 weeks with the target values and any calculated deficit was then compensated for by adding regular tap water, which was done with spray bottles. Hereafter, the watering treatments are referred to as 'average' and 'worst-case' precipitation. Lysimeters were watered for 2–6 h each day watering occurred (giving <4 mm h^{-1}) to simulate natural rainfall and prevent ponding on the soil surface. After fertilization, watering was performed on 14 and 18 occasions resulting in total amounts of 190 and 275 mm for the 'average' and 'worst-case' regimes, respectively. After herbicide treatment, 138 and 223 mm were applied on 10 and 14 occasions in the respective regimes.

Leachate sampling and water analyses

Water leaching through the soil columns was led in plastic (polyethylene) pipes to glass sampling bottles (both materials with nonsorptive properties with regard to dichlorprop) placed in a below-ground measuring station. The bottles were weighed each week to determine the leachate volumes. Simultaneously, subsamples were taken from the accumulated leachate for chemical analyses. Sampling started after fertilization and continued until the end of April 1990. However, dichlorprop was analysed only on samples collected between herbicide treatment and 19 February 1990.

In the dichlorprop analyses, water samples were first acidified with concentrated phosphoric acid (ca 5 ml), after which dichlorprop was extracted with dichloromethane (50 + 25 ml). Extractive alkylation with pentafluorobenzyl bromide was carried out according to Åkerblom *et al.* (1983). After extraction, the dichloromethane phase (1·5 ml) was evaporated to dryness. The remainder was dissolved in hexane (1·5 ml). Quantitative determination was by capillary column gas chromatography (Carlo Erba 2130, OV-1 column) with electron capture detection (Carlo Erba HT-25) and a detection limit of 0·5 μg litre^{-1}. The average recovery was ca 70%. The analytical procedure is described in more detail by Åkerblom *et al.* (1990).

Nitrate concentrations were determined by flow injection analysis (Fiastar 5010 Analyzer, Tecator AB) according to the colorimetric Cd-reduction method (APHA, 1985).

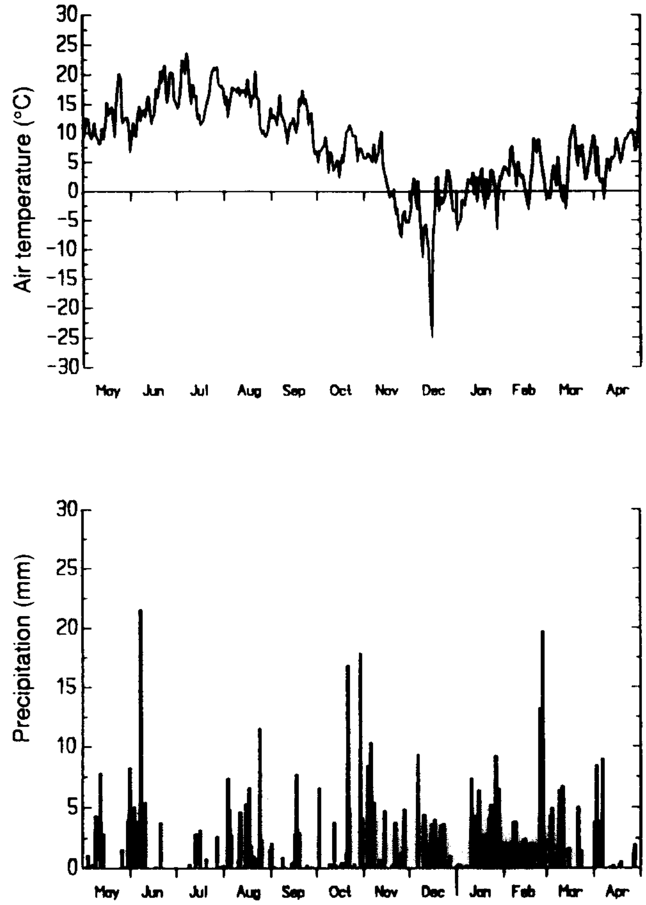

Fig. 2. Daily mean air temperatures (upper) and daily precipitation at a nearby weather station (ca 500 m from the lysimeter station) in Uppsala.

RESULTS

Weather and drainage conditions

The period as a whole was relatively warm and dry (Fig. 2), with the total precipitation of 474 mm between 1 May 1989 and 30 April 1990 being considerably below the long-term average (554 mm) for the region. However, the distribution of precipitation over the year was quite uneven. The amount of rainfall during the growing period (1 May–30 September) was ca 55% of the 30-year normal rainfall, whereas precipitation during the rest of the study period (1 October–30 April) was ca 17% above average. Monthly precipitation from May onwards amounted to 30, 41, 10, 54, 17, 61, 42, 37, 58, 60, 34, and 30 mm.

Supplemental irrigation, together with natural precipitation, resulted in total water inputs to the lysimeters (1 May 1989–30 April 1990) being 664 and 749 mm for the 'average' and 'worst-case' watering regime, respectively. These amounts are clearly above the respective target values. From the spraying of dichlorprop (9 June 1989) until 19 February 1990 (when dichlorprop analyses were terminated), the lysimeters received total water inputs of 505 ('average') and 590 mm ('worst-case'), which are also above the respective target values for that period.

From mid-November to the end of December, precipitation often occurred as snow. However, there was no continuous snowpack on the ground because temperatures commonly rose above zero (Fig. 2).

A drainage pattern with two main flow periods, in autumn and in spring, which is typical for Swedish conditions (Gustafson, 1983), never occurred in this study owing to the extremely dry summer of 1989. Amounts of leachate were very small during and shortly after the cropping season (Fig. 3) regardless of the watering regime. It was only the 'worst-case' watered clay monoliths that produced leachate amounts exceeding 50 mm in autumn (Fig. 3). Once the soils had become completely thawed in the beginning of February, drainflows reached or exceeded 100 mm in both soils and watering treatments within a month (Fig. 3), indicating a fast drainage response once the soils were wetted up. The accumulated average leachate amounts during the experimental period (until 30 April 1990) were 199·6 and 234·3 mm for the 'average' and 'worst-case' watered clay, and 127·8 and 151·0 mm for the corresponding peat monoliths. These amounts are equivalent to ca 20 and 30% of the total water input (both watering treatments) to the peat and clay monoliths, respectively. These figures correspond relatively well with the rainfall recovered in leachate in earlier similar studies using the same type of lysimeters (Bergström, 1987*a*, 1992). Total average amounts of leachate up until the end of February were 158·0 ('average') and 196·2 mm ('worst-case') for the clay soil, and 118·2 ('average') and 128·4 mm ('worst-case') for the peat soil.

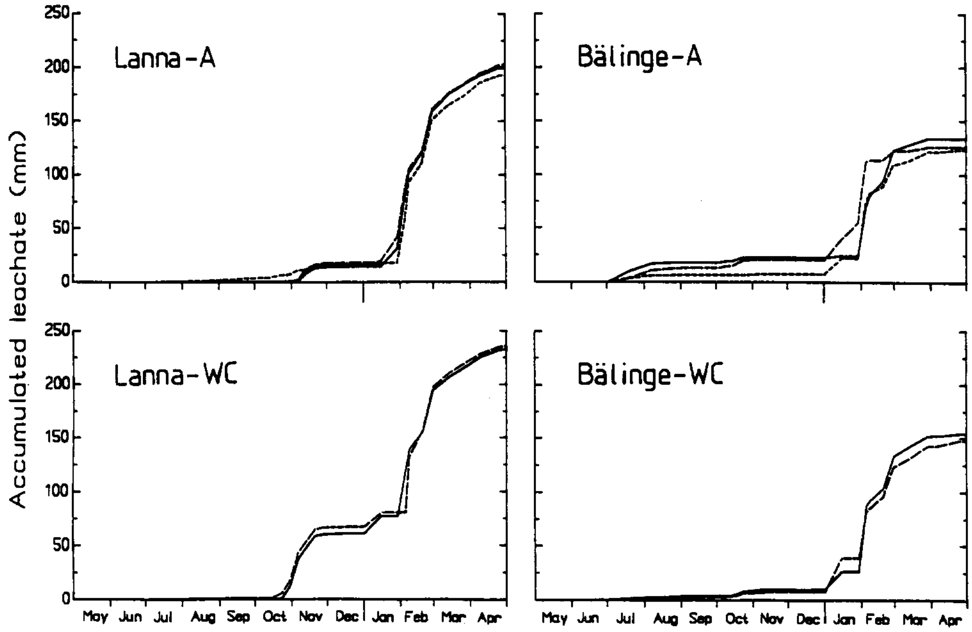

Fig. 3. Accumulated leachate amounts in the two soils (Lanna-clay and Bälinge-peat) during the experimental period (1 May 1989–30 April 1990). All the replicates are shown in each diagram. A = 'average' precipitation and WC = 'worst-case' precipitation.

Nitrate in leachate

Nitrate concentrations were much larger in leachate from the peat soil, reaching concentrations of ca 160 mg N litre^{-1} in both watering treatments (Fig. 4). Nitrate concentrations in clay soil leachate never exceeded 15 mg N litre^{-1}. The difference was most likely due to intensive decomposition of the large quantities of organic matter in the peat soil. In both soils, concentrations were relatively small at the beginning of both main drainflow periods, in spring and in autumn (Fig. 4). Subsequently, as drainage proceeded, nitrate concentrations increased, and in the spring they levelled off at around 100 mg N litre^{-1} in the peat soil and 10 mg N litre^{-1} in the clay soil. This pattern was especially obvious in the peat monoliths. Later in the spring, towards the end of the drainflow period, concentrations tended to decrease, although not quite to the initial levels. There was very little difference between the watering treatments regarding the distribution and levels of nitrate in leachate.

The total average nitrate loads over the period (1 May 1989–30 April 1990) were 11·6 and 21·8 kg N ha^{-1} in the 'average' and 'worst-case' watered clay monoliths, and 37·6 and 65·4 kg N ha^{-1} in the corresponding peat monoliths. The largest monthly loads occurred during February in both soils (between 42 and 61% of the total loads), coinciding with the largest drainflow events (Fig. 3).

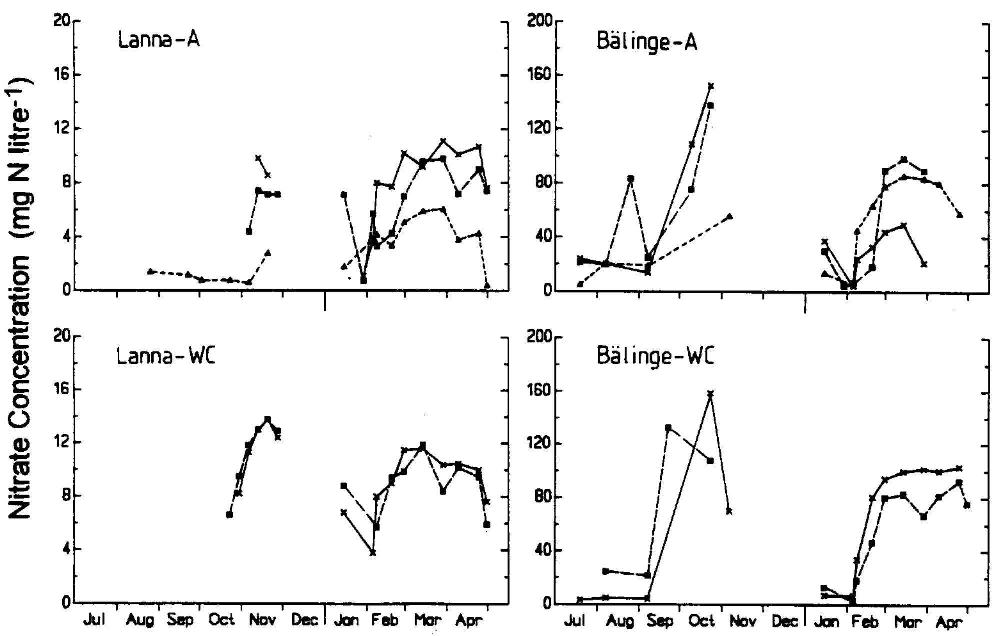

Fig. 4. Nitrate concentrations in leachate from the two soils (Lanna-clay and Bälinge-peat). All the replicates are shown in each diagram. A = 'average' precipitation and WC = 'worst-case' precipitation. Note the different scales on the *Y*-axes.

Table 2. Dichlorprop concentrations in leachate (μg litre^{-1}), presented as mean values (± SD, n = 2–3) for the replicates

	Lanna-Clay		Bälinge-Peat	
Date	A	WC	A	WC
19 Jul.	—[a]	—	269·3 ± 208·7	77·7
7 Aug.	—	—	139·4 ± 87·1	79·6 ± 72·0
25 Aug.	1·4	—	19·3 ± 12·0	—
2 Sept.	—	—	11·3	—
22 Sept.	2·8	—	—	16·7
23 Oct.	3·3	8·0	8·7 ± 0·6	11·0 ± 11·6
30 Oct.	—	<0·5 ± 0·6	—	—
6 Nov.	3·8 ± 0·9	0·0 ± 0·0	27·7	2·9
13 Nov.	2·4 ± 0·6	0·0 ± 0·0	—	—
20 Nov.	3·5 ± 1·9	0·0 ± 0·0	—	—
27 Nov.	2·0	0·0	—	—
15 Jan.	4·8 ± 4·4	0·0 ± 0·0	2·2 ± 2·4	1·4 ± 0·7
29 Jan.	0·6 ± 0·9	0·0	0·0	—
5 Feb.	5·1 ± 3·6	0·0	0·7 ± 0·7	0·0 ± 0·0
8 Feb.	0·0 ± 0·0	0·0 ± 0·0	0·0	0·0 ± 0·0
19 Feb.	1·1 ± 1·8	0·0 ± 0·0	<0·5 ± 0·6	1·4

[a]Dashes indicate that no leachate was available.
A = 'average' precipitation.
WC = 'worst-case' precipitation.
Concentrations below the detection limit (<0·5 μg litre^{-1}) are set to zero when calculating the mean concentrations. Mean concentrations shown as <0·5 μg litre^{-1} indicate that not all replicates had concentrations below the detection limit.

Dichlorprop in leachate

As was the case for nitrate, dichlorprop concentrations were, by far, largest in leachate from the peat soil, reaching a maximum level of, on average, ca 270 μg litre^{-1} (Table 2). This peak concentration occurred in 'average' watered monoliths within 40 days of spraying. Simultaneously, leachate from 'worst-case' watered peat monoliths reached average dichlorprop concentrations in excess of 77 μg litre^{-1} (Table 2). Subsequently, there was a more or less continuous decrease in concentration to levels just above the detection limit (0·5 μg litre^{-1}) in both watering treatments towards the end of the measurement period (Table 2). In the clay soil, a peak dichlorprop concentration of 8 μg litre^{-1} in the first leachate sample collected from one of the 'worst-case' watered monoliths was followed by non-detectable concentrations throughout the rest of the period (Table 2). In contrast, detectable dichlorprop peaks reaching, on average, ca 5 μg litre^{-1} occurred in the 'average' watered clay soil during the whole period (Table 2).

Accumulated average loads of dichlorprop from the time of spraying until 19 February 1990 were 3·22 and 0·26 g a.i. ha^{-1} in the 'average' and 'worst-case' watered clay, and 28·91 and 2·67 g a.i. ha^{-1} in the peat soil with the corresponding watering regimes. These figures represent 0·20, 0·02, 1·81, and 0·17% of the applied active substance. In clear contrast to nitrate loads, maximum monthly dichlorprop loads did not occur during February when drainflows were large (except for the 'average' watered clay soil), but in July (peat soil) and October (clay soil) when peak concentrations occurred, and drainflows were very small (Fig. 3).

DISCUSSION

One flow mechanism that has received widespread attention during recent years is that of preferential movement of solutes through various soil structures (e.g. White, 1985; Priebe & Blackmer, 1989). This type of flow enables solute to bypass the soil matrix, resulting in considerably faster movement than would be expected with classical convective–dispersive flow behaviour (Thomas & Phillips, 1979). In this study, there was a clear difference in leaching behaviour between the two solutes. Preferential flow appeared to be of little significance in nitrate transport, which could be described, more or less, as a simple function of amounts of leachate. In contrast, dichlorprop leaching seemed to be clearly influenced by preferential flow processes. For example, on average, ca 88% of the total dichlorprop load in the 'average' watered peat soil was found in the first 10 mm of leachate in the summer. Kladivko *et al.* (1991) explained this difference in terms of nonequilibrium adsorption/desorption of the pesticide in the preferential flow paths. Immediately after a heavy rainstorm, pesticide residues dissolved in the soil solution are rapidly transported through macropores in the soil profile to the bottom of the lysimeter. Desorption is too slow to maintain an equilibrium solution concentration in new infiltrating water, resulting in decreasing pesticide concentrations in macropore water as the rainstorm continues. A similar explanation was given by Everts *et al.* (1989). It is also reasonable to believe that preferential flow processes normally dominate during a short initial part of a flow event (Hallberg, 1986). Surface-applied chemicals could then move rapidly through the soil to deeper layers. Towards the end of a flow period, when most of the preferential flow paths are closed, much of the water percolating through the soil will interact with the soil matrix, where adsorbed pesticides are less susceptible to leaching. Concentrations of such solutes in leachate will therefore decrease as drainage continues. A nonadsorbed solute, such as nitrate, will continue to leach through soil even when water flow through the soil matrix dominates. Indeed, nitrate concentrations in leachate from both soils in this study increased as the drainflow periods proceeded.

A general problem with lysimeters is that water and water-carried chemicals may flow along the lysimeter wall, thereby affecting solute movement in the same way as macropore flow. This behaviour is extremely important in leaching studies of surface-applied chemicals since the compound is often concentrated within the top few centimeters of soil and may therefore be transported rapidly along the wall as fictitious preferential flow. The extent of this potential problem for the type of lysimeter used in this study has been assessed in an earlier experiment (Bergström *et al.*, 1994) in which monoliths of Lanna clay and a sandy soil were treated with two non-reactive tracers, ^{36}Cl and tritiated water.

Each tracer was applied to an annular zone and a similarly sized disc zone of the monolith surfaces. In both soils, the tracers appeared in leachate simultaneously: in the sand, they both appeared after ca 100 mm of leachate had accumulated, whereas in the clay only a few millimeters of leachate had been collected by the time that they appeared. The latter observation is clearly indicative of macropore flow in the structured clay. Even though there was a difference between tracers in the total amounts of radioactivity recovered owing to the unavoidable evaporation of the tritiated water, these results show that sidewall flow is not a problem for this type of lysimeter.

Based on the above discussion, it is obvious that intensive rainfall favours preferential flow behaviour. Even though the precipitation mostly occurred as low-intensity rainfall events (Fig. 2), the use of supplemental irrigation, at rates of 7–26 mm day^{-1}, allowed simulation of relatively heavy rainstorms. This was accentuated in the 'worst-case' watered monoliths during the summer/autumn period when they received supplemental irrigation in larger amounts and more frequently compared with the 'average' watered monoliths. For example, within 20 days after spraying dichlorprop, the two irrigation treatments had received 22 and 7 mm of supplemental water. Still, for both soils leaching loads of dichlorprop were clearly the largest for the 'average' watered monoliths, which also produced larger concentrations of the compound most of the time. In both soils, cracks, openings and other discontinuities in the soil surface of 'average' watered monoliths were visibly more abundant and wider during the summer months, compared with 'worst-case' watered monoliths. Such cracks presumably encouraged water flow in macropores, moving some of the dichlorprop rapidly through the topsoil to the subsoil. Once the compound reached the subsoil, degradation rates should have been reduced substantially and the transported dichlorprop residues stored until later drainflow events. In 'worst-case' watered monoliths, with fewer cracks, a greater proportion of dichlorprop should have remained in the topsoil where degradation rates were likely to be much higher. For example, in a study by Stenström (personal communication), dichlorprop half-life was estimated to be 5·4 days in Lanna clay topsoil, whereas no detectable degradation was found in subsoil samples during the course of a 13-day experiment. In contrast to dichlorprop, nitrate leaching from both soils was considerably larger in 'worst-case' watered monoliths than in 'average' ones. Nitrate was presumably more evenly distributed in the soil matrix, and water rapidly flowing through macropores should therefore not have carried significant amounts of nitrate. Indeed, nitrate leaching in these two structured soils seemed to occur in the true sense of the word, with increasing loads following increased water inputs. If a ^{15}N-labelled tracer had been used, allowing the recovery of a surface-applied nitrate fertilizer to be determined, it is very likely that the added fertilizer would also have been found to be significantly exposed to preferential flow processes. For example, Priebe and Blackmer (1985) showed that substantial amounts of ^{15}N-labelled urea were lost from the surface metre of soil within just a few weeks of the spring application. However, when using unlabelled nitrogen, as was the case here, these effects are completely masked by the great proportion of nitrate in leachate derived from the decomposition of soil organic matter (see Bergström, 1987*b*).

In a parallel study where dichlorprop and nitrate leaching were measured in sand monoliths, it was found that dichloprop also leached more when 'worst-case' watering was performed, i.e. 0·48 and 0·91 g a.i. ha^{-1} in the 'average' and 'worst-case' watering regime, respectively (Bergström & Jarvis, 1993). This was to be expected, since macropore flow is certainly not a main flow process in sandy soils. Therefore, the leaching of pesticides as well as other solutes should occur more uniformly throughout the profile of such non-structured soils.

CONCLUSIONS

The results of this study clearly show that preferential flow processes may have a substantial influence on pesticide leaching in structured soils. Since dry weather conditions are likely to favour the creation of cracks and other macro-fissures in soil, it is not necessarily true that wet years constitute worst-case conditions in terms of pesticide leaching. Nitrate leaching, on the other hand, seems to increase when rainfall amounts increase. When preferential flow processes exert a significant influence on pesticide movement in soils and exposure of the compound to the soil matrix is thereby reduced, there is reason to believe that the influence of physico-chemical properties of pesticides on leaching loads is also reduced considerably.

ACKNOWLEDGMENTS

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