PESTICIDE LEACHING IN INTACT BLOCKS OF CLAYEY TILL

LA FILTRATION DES PESTICIDES DANS LES BLOCS INTACTS D'ARGILE MORAINIQUE

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SYNOPSIS. Solute transport through intact blocks of clayey till was investigated by leaching of $CaCl_2$ and the herbicides simazine and mecoprop. The block specimens (d = 0.5 m, h = 0.5 m) were installed in triaxial cells simulating in situ conditions of soil pressure and temperature during leaching. The experiments indicated by-pass of the clayey matrix by rapid transport of the solutes in fractures of the till. Furthermore, the intact blocks revealed 3-4 orders of magnitude higher hydraulic conductivities than traditional small sample permeability tests. Together, the blocks revealed a strongly increasing hydraulic conductivity (10^{-7}-10^{-5}m/s) upward in the till profile investigated.

INTODUCTION

Clayey till is often assumed to act as an effective barrier against long-term percolation of pollutants to the groundwater. This assumption is based on laboratory tests which show permeabilities in a range of 10^{-8}-10^{-10}m/s (Fredericia 1990, Foged et.al 1992). These tests are usually performed with relatively small samples and so very few of them represent the natural inhomogenity of clayey till. However the methods developed were extended to the present specimen size, in order to justify the hydraulic properties being found under controlled test conditions. Investigations by Jørgensen (1990), Fredericia (1990, 1991) and Jørgensen & Fredericia (1992) have documented that waterflow in fractures and macropores is a principal mechanism of groundwater infiltration in shallow Danish clayey tills. In this paper the characteristic of waterflow and solute transport in clayey till is quantified by means of laboratory tests applying large intact blocks of clayey till.

DESCRIPTION OF THE INTACT BLOCK SAMPLING SITE

The site of the intact block sampling is an orchard situated at Havdrup 40 km southwest of Copenhagen where the landscape is developed as a slightly undulating Weichselian till plain. Field investigations at the site by Jørgensen & Fredericia (1992) show that the hydraulic activity of groundwater flow is by far most intensive in the upper ten metres of the till. In this zone dual porosity is developed, with an upward increasing density of fracturing, as a result of dessication and glacial jointing. From ground water monitoring in the orchard in 1989 (Jørgensen & Fredericia, 1992) and in 1992 (Jørgensen et. al., in. prep.) rapid leaching of pesticides in fractures is indicated by the migration of simazine and atrazine through 8 metres of clayey till in a period of less than ten years.

METHODS

Sampling of Intact Blocks

The circular blocks were 0.5 m in diameter and 0.5 m high, being large enough to represent fractures and macropores in the till. Sampling depths was 1-1.5, 2-2.5 and 4-4.5 m in a profile excavated in the orchard. During sampling the blocks were embedded in a fluid rubber casing which fixed the blocks after hardening in a combined mould and transport steel cylinder. Before hardening the fluid rubber enters a few millimetres of the till matrix. Thereby the outer surface of the blocks was sealed and problems with flow along this boundary during the experiments were eliminated. After fixation, the blocks were detached from the till formation. The steel cylinder was removed after transport of the blocks. During installation of the blocks in the laboratory they were operated using under pressure of - 30 to - 60 kPa.

Laboratory Set-Up and Experimental Conditions

The intact till blocks were installed in large triaxial cells and connected to a percolation system, Fig. 1. In the triaxial cells the in situ pressure and temperature of the till formation were simulated in order to establish realistic physical conditions during the experiments with the blocks. A percolation experiment was performed with $CaCl_2$. An influent concentration of $CaCl_2$ equivalent to 3000 mg Cl^-/l was applied in the blocks from 1 and 2 m's depth. In the block from 4 m's depth the influent concentration of Cl^- was 1700 mg/l. Afterwards pesticides were percolated. The influent concentration of mecoprop was 54.7 in the block from 1 m's depth and 57.6 mg/l in the blocks from 2 and 4 m's depth. Influent concentration of simazine was 13.6 mg/l in the block from 1 m's depth and 4.8 mg/l in the blocks from 2 and 4 m's depth. In the fluorescent dye experiment Na-fluorescein concentration was 2000 mg/l.

Chemical Analyses and Presentation

In the $CaCl_2$ tracer experiment influent and effluent concentration was monitored with a electric conductivity probe. Effluent conductivities (e_e) were normalized (e^*) to influent conductivities (e_i) with the expression $e^* = (e_e - e_{background})/(e_i - e_{background})$. Determination of simazine and mecoprop in the influent and effluent percolate was carried out with high performance liquid chromatography and UV-detection at 229 nm. Effluent concentrations (C_e) was normalized (C^*) to influent concentration (C_i) as $C^* = C_e/C_i$.

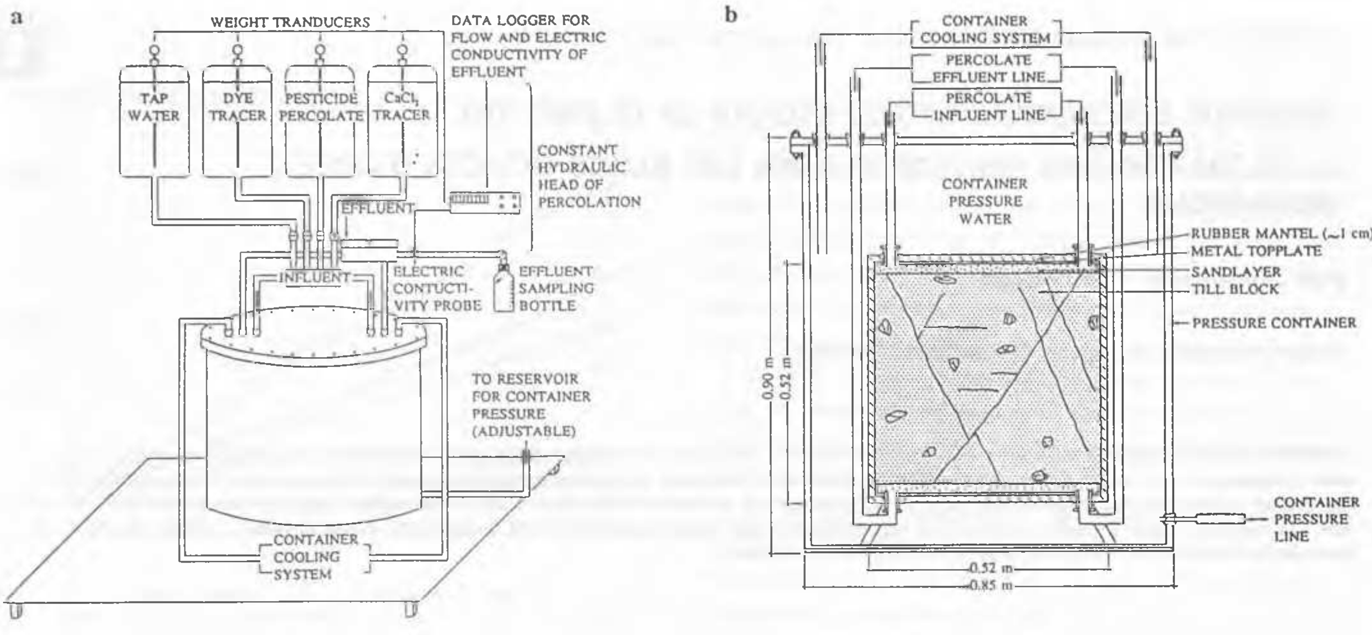

Fig.1. (a) Oblique view of the triaxial cell. (b) Vertical cross-section of triaxial cell and intact block installed for leaching experiments under in situ conditions of soil pressure and temperature.

RESULTS AND DISCUSSION

Bulk Saturated Hydraulic Conductivity, Hydraulic Fracture Aperture and Effective Porosity of Intact Blocks

After installation and water saturation of the blocks in the triaxial cells liniarity af flow (Q) versus hydraulic gradient was tested. One basic demand was the ability of the experimetal set-up to reproduce the Darcy equation for the equivalent flow of dual porosity clayay till

$$Q = k(dh/dx)$$

The result of this test is displayed in Fig.2 which shows the flow rate at two levels of triaxial pressure versus different hydraulic gradients. From the straight line relationship between flow rate and gradient it is demonstrated that the hydraulic performance of the intact block follows the Darcy equation.

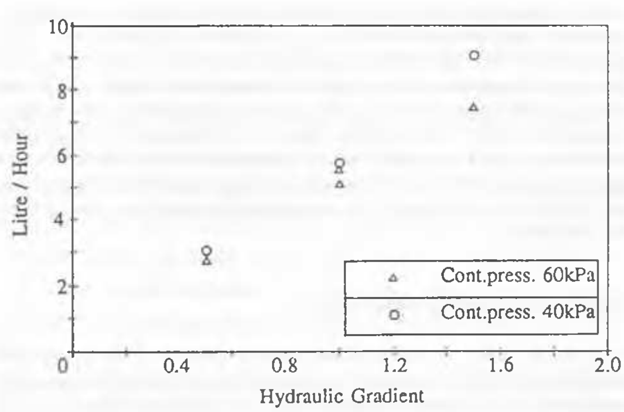

Fig. 2. Flow rate versus hydraulic gradient of the block from 2-2.5 m's depth tested at two levels of triaxial pressure.

Measurements of the saturated hydraulic conductivity (k) of the intact blocks revealed values of 1-5 10^{-7} m/s in the deepest block (4-4.5 m's depth). Above this depth the hydraulic conductivity increased to 2-6 10^{-6} m/s in the block from 2.5 m's depth and to 1-5 10^{-5} m/s (equivalent to k-values of fine sand) in the blocks from 1-1.5 m's depth (Table 1). The range of these values corroberate slug tests and trench-to-trench experiments performed in similar tills in Denmark (Fredericla 1990, 1991).

Based on the assumptions of Grisak et. al. (1980) the fracture porosity (θ) of the intact blocks was calculated from

$$\theta = \frac{2(2b)}{S_e}$$

where the relationship between 2b (effective fracture aperture) and S_e (effective fracture spacing), is given by

$$(2b)^3 = \frac{Q_T}{A_T} \cdot \frac{12\mu}{\rho g\ (dh/dx)} \cdot S_e$$

where

Q_T	total volumetric flow rate, L^3T^{-1};
μ	dynamic viscosity of fluid, $ML^{-1}T^{-1}$:
A_T	total cross-sectional area of sample, L^2;
ρ	fluid density, ML^{-3};
g	gravititional constant, LT^{-2};
dh/dx	hydraulic gradient , LL^{-1};

Furthermore the average hydraulic conductivity of the individual fractures (K_f) in each block was calculated from Snow (1968, 1969), given by

$$K_f = \frac{(2b)^2 \rho g}{12\mu}$$

Effective fracture spacing (half the distance between fractures) in the blocks was measured from the distribution of the illuminating traces observed in the intersected till blocks after the fluorescent dye tracer experiment (Fig.4c).

Effective fracture spacings, calculated effective apertures, fracture porosities and calculated hydraulic conductivities of fractures are displayed in Table 1.

Table 1. Structural and hydraulic parameters of intact till blocks.

Depth of blocks (m)	Effective mean fracture spacing (m)	Calculated mean fracture aperture (μm)	Total porosity	Calculated fracture porosity	Mean bulk hydraulic conductivity (m/s)	Calc. mean hydraulic conductivity of fractures (m/s)
1.0-1.5	0.03	86	0.32	0.0030	$1.3 \cdot 10^{-5}$	$4.6 \cdot 10^{-3}$
2.0-2.5	0.05	70	0.31	0.0014	$4.2 \cdot 10^{-6}$	$3.1 \cdot 10^{-3}$
4.0-4.5	0.08	32	0.25	0.0004	$2.5 \cdot 10^{-7}$	$6.4 \cdot 10^{-4}$

The indicated trend of very strongly increasing bulk hydraulic conductivity, fracture density and fracture porosity upwards in the till profile corroberate the distribution of fractures and macropores mapped in the study site as well as in other localities in Denmark (Jørgensen 1990, Fredericia 1991). However, the actual trend of hydraulic conductivity in the field will be even stronger than shown by the blocks as these were specially sampled in fracture zones. Unfractured zones are occurring in the lower weathered and unweathered zone.

On sloping land the upward increasing hydraulic conductivity has an important implication on lateral groundwater flow and associated migration of mobile pollutants. In the summer the groundwater table is about 2.5-4 m below the ground surface, at the sampling site, and in this situation only slow lateral groundwater movement occurs. However, in the autumn and winter the groundwater table quickly rises to about 1-0.5 m below ground surface, and in this situation the lateral flow is about 10^2-10^3 times more than at minimum groundwater level in the summer (Table 1). In relation to the possibility of lateral leaching of pesticides (and other pollutants) into out-cropping aquifers or streams, this mechanism has to be considered seriously.

Percolation of Intact Blocks with $CaCl_2$-Tracer and The Pesticides Mecoprop and Simazine.

To evaluate the importance of saturated fracture- and macropore flow to the hydraulic activity and leaching of pollutants, an experiment with percolation of the blocks with $CaCl_2$-tracer was performed. Normalized breakthrough curves of the tracer are shown in Fig.3. As a common feature the breakthrough curves tend to flatten after an initial rapid rise in electric conductivity. This can be interpreted on the basis of hydraulic transport of solutes through fractures, which provides the early breakthrough, while the subsequent flattening of the curves can be interpreted as being due to diffusion of the solute from the fractures into the porous matrix.

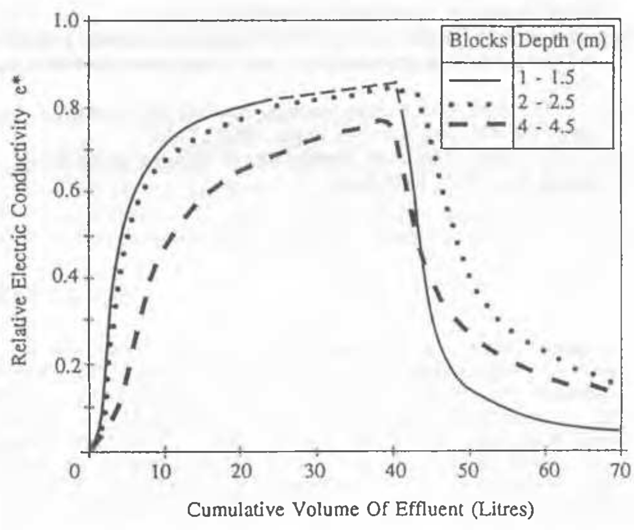

Fig.3. Breakthrough data for $CaCl_2$-tracer in individual blocks. Influent was 40 l of $CaCl_2$ percolate followed by 30 l of artificial groundwater. Hydraulic gradient of percolation dh/dx = 1, triaxial cell pressure = 60 KPa.

To visualize the actual configuration of the hydraulic activity of the blocks they were finally percolated with a fluorescent dye tracer. After percolation the blocks were intersected and illuminated with ultraviolet light. This revealed a characteristic pattern of hydraulic active fractures which coincide with the fractures observed in the blocks during the sampling in the orchard, Fig.4a,c.

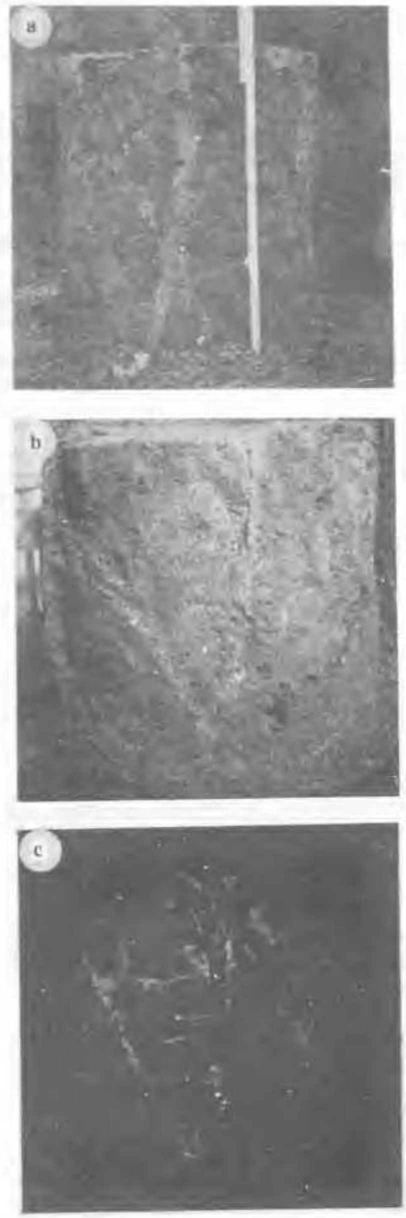

Fig.4. (a) Intact block during sampling from 2-2.5 m depth, showing displacement (arrows) of the less bleached fracture by glacial faulting (stick 0.9 m long) ; (b) and (c) The apperance of the intact block after the fluorescent dye experiment and intersection, (b): normal light, (c): ultraviolet light. The latter showing the paths of preferential flow in the fractures observed during sampling of the block.

Thus, the tracer experiments document that the advective transport of solutes in the clayey till principally occurs as macropore flow bypassing the clayey matrix.

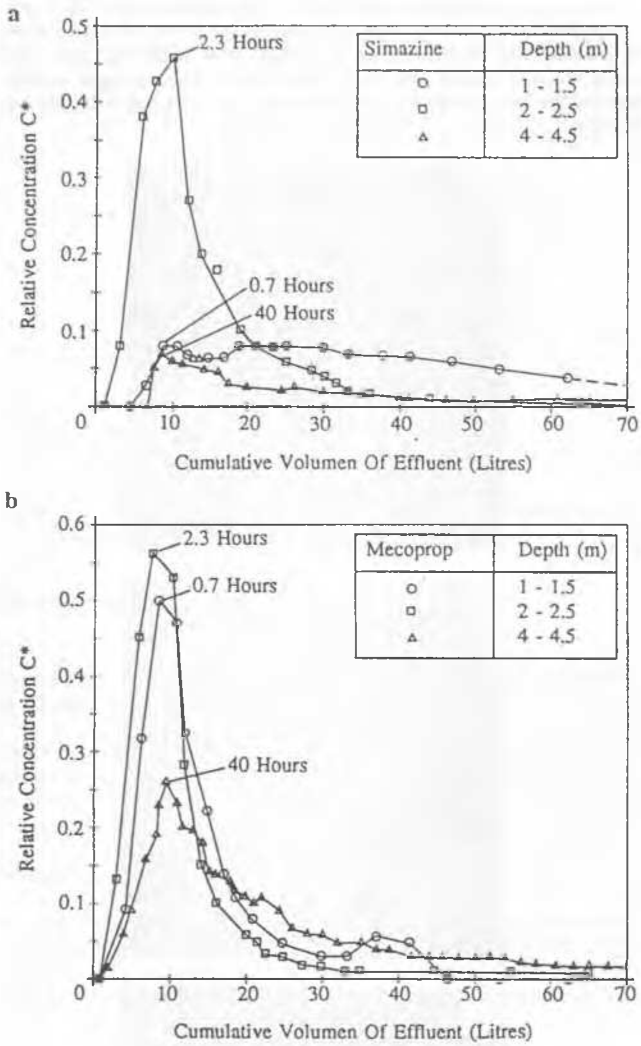

Fig. 5a,b. Breakthrough data of a 6 l mixed puls of (a) simazine and (b) mecoprop in the individual intact blocks. Hydraulic gradient of percolation dh/dx = 1, triaxial cell pressure = 60 KPa.

The effect of macropore flow on the leaching of pesticides was investigated by percolating simazine and mecoprop in a 6 litre mixed puls of simazine and mechoprop through the intact blocks. The breakthrough curves of the effluent percolate are shown in Fig.5a,b. After a small volume of effluent had been transmitted, the pesticide break-through was registered in all blocks. The breakthrough times (measured at half the maximum value of effluent concentration) were approximately 20, 1.2 and 0.3 hours decreasing upward through the successive block specimens, reflecting the parallel increase in hydraulic conductivity. Adsorption of simazine was indicated by significant tailing of the break through curve (Fig.5 a) in the block from the bottom of the soil zone (organic rich). Simazine is adsorbed moderately to organic matter. When there was no significant parallel indication of adsorption of mecoprop in this block (Fig.5b) it is probably due to the anionic properties of this pesticide. Additional retardation of solute transport was indicated in the deepest block well below the soil zone. In this block the percolate had a long residence time due to the low hydraulic conductivity of the block. Long residence is giving way for diffusion of the pesticides into the surronding clayey matrix. However, the relative importance of matrix diffusion and adsorption below the soil zone in clayey till is still a subject of investigation.

CONCLUSION

Laboratory leaching of tracers, simazine and mecoprop, through large intact till blocks, under simulated in situ conditions, showed that the advective transport of pesticides in clayey till occurs as very rapid fracture flow which was bypassing the clayey matrix. However, a significant potential for retardation of pesticide leaching was indicated by adsorption in the soil zone and by matrix diffusion in the intact block of low hydraulic conductivity. The block specimens revealed hydraulic conductivity increasing upward in the sampling profile from 10^{-7} m/s at 4.5 m's depth to 10^{-5}m/s at 1.5 m's depth.

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