

Solute movement prediction was initiated by van der Molen (1956). He derived the convective dispersive equation from chromatography theory based in the assumption that all water percolating through the soil moves at approximately with the same velocity. The solute disperses around the solute front that moves down with the average velocity and is described by a dispersion coefficient. In the early 1980's it became obvious that under field conditions ground water contamination could with pesticides could not be described by the convective dispersive equation and that some kind of preferential flow occurred. Since that preferential flow has been researched widely. The term preferential flow refers to the rapid transport of solutes and water through preferential pathways in the subsoil (Stagnitti et al 1994; Park et al, 2008). Now 30 years later Koestel et al (2011) write (page 10012 line 23) "reason for choosing the CDE (convective Dispersive Equation) and MIM (Mobile Immobile Model) parameter sets is the frequent application of these two models in the peer reviewed literature". So it seems that we have gone full circle. Unless the soil has clear distinct flow path such as with fingered flow in coarse sandy soils, the preferential flow path regime has a distinct different velocity than the matrix flow (Kung, 1990; Kung et al., 2000) and therefore violates the assumptions of the convective dispersive equation. Thus there is a basic problem in using the convective dispersive equation with a single velocity (this is the case also for the Mobile Immobile Model) and it is necessary to employ a model that has several distinct velocities for the different regions in the soil. We understand that the data are not available and one has to resort to the approach of using the convective dispersive equation as done by Koestel et al (2011). In my opinion, using the convective dispersive equation is far from ideal and one really should document in how many cases the convective dispersive equation actually can represent the break through curves and especially the early part (first 0-1%) rather than report the results of the parameters when the breakthrough curves fit.

One of the objectives of the manuscript is the find pedotransfer functions. Bouma (1989) introduced the term pedotransfer function, which he described as the translation of data (soil survey data) that is available in data (soil hydraulic data) that is needed (Iverson et al, 2011). Thus, Bouma used the pedotransfer function to describe a static property of the soil. Preferential flow this is certainly not a static property and one need to be careful applying pedotransfer to these kinds of phenomena as it might be dependent on the boundary conditions. So let's us see what the effect is of the boundary condition.

In any soil the mass balance should be met. Assuming steady state conditions and no dispersion and one set of unique flow paths, we can write that the flux q should equal the product of the downward velocity, v , the moisture content, and the area wetted up (Kim et al, 2005, Darnault et al, 2004)

$$q = v\theta A$$

Thus changing the flux q will alter either the velocity, the moisture content or the velocity. In the typical standard convective equation the area is considered constant and when the flux is increasing the moisture content and the velocity increases. Since roughly for a 1% change in moisture content the conductivity changes by a factor of 2, it is the velocity that increases faster than the moisture content, Under preferential flow conditions as shown by Koestel et al (2011), the velocity as a fraction of the imposed flux did not change significantly between the experiments, and thus the wetted area A (that takes part in the preferential flow transport) need to change as a function of the imposed flux. This was

also confirmed experimentally by Darnault et al (2004) and Kim et al (2005) for a sandy soil with fingered flow. These findings have actually interesting implications, because it would mean that the length of the storm (or the wet period) determines to what depth the preferentially moving chemical would go and not the amount of rainfall since the velocity is constant. It also shows the limitations of the pedotransfer functioning

The manuscript by Koestel et al (2011) is confusing and it might be very well that I have interpreted the text wrongly. For example the following is highly puzzling to me: On (page 10010 near line 25) the authors discuss the different flow regimes and its shortcomings for using the convective dispersive equations. In fact that the authors write "Therefore model-independent (non-parametric) PTFs for solute transport properties should be preferred to model-dependent ones (line 4 page 10011). Then later as cited above line 20 page 10012 it is stated above "we used the CDE and MIM parameter sets. Simply dividing the CDE parameters by a quantity with the same units does not make them independent of the convective dispersive equation with one average transport velocity.

It also stated that the convective dispersive equation with a Dirac pulse input (page 10013 line 13). was used to fit the breakthrough curves. When looking at almost any preferential flow picture (Figures 1 and 2) one can see that there is a layer at the surface (we called it a mixing layer) that distributes the solutes to the preferential flow paths. Therefore, a boundary condition with an exponentially decreasing concentration is more appropriate than a Dirac pulse input (details are given in Darnault et al, 2005 and Kim et al, 2004)

The authors use four dimensional numbers (or as the authors indicate, non-parametric shape parameters) to characterize the preferential flow: One of the dimensional numbers is the "normalized arrival time" that indicates the (normalized) time for 5% of the solute to leach. The 5% is arbitrary and likely too large. Let's assume that a typical pesticide application of 2 kg/ha is added to the soil and let's assume too that it dissolves in a large 10 cm rainfall that fell shortly after application without the opportunity for the pesticide to adsorb to the soil (although theoretically the same concentration is obtained when the pesticide is adsorbed, Steenhuis and Naylor, 1987). The concentration in the water will be: $2 \text{ (kg/ha)} / (0.1 \text{ m} * 10,000 \text{ m}^2) = 2 \cdot 10^{-3} \text{ kg}$ or $2,000,000 \text{ } \mu\text{g/l}$. Dissolving 5% of this high concentration in the total rainfall still gives a concentration of $100,000 \text{ } \mu\text{g/l}$!

Another way of looking at the problem is how much of the groundwater that five percent of 2kg/ha of pesticide can pollute the aquifer at the drinking water standard level. Assuming a reasonable standard of $10 \text{ } \mu\text{g/l}$, five percent of an application of 2kg (or 100 g) of pesticide can bring $10,000 \text{ m}^3/\text{ha}$ of groundwater up to a concentration of $10 \text{ } \mu\text{g/l}$. That is equivalent to 1 meter depth of water or equivalent to 2-3 years of recharge in most humid temperate climates. In reality these high groundwater concentrations are not observed, because pesticides degrade.

In order for the reader to make its own conclusion, Table 1 needs much more information on the type of data that each article provides how many breakthrough curves could be fitted to the convective dispersive equation. It also needs to state exactly how the analysis was done and for each experiment and what the calculated four non-dimensional parameters were and how many of the BTC's curves could be fit with

reasonable numbers. The authors used this information to make their figures in there is no reason not to share this information with the reader since supplementary material is now very common part of manuscripts. With this information the reader can make a guess how well the convective dispersive equation is valid for describing preferential flow conditions. Perhaps we could settle the argument with this information if the use of the convective dispersive equation is justified for describing preferential flow or not.

Finally at the end the authors suggest (page 10025 around line 10) that more breakthrough curves should be done for sandy soils. May be the authors should then consider not throwing away the data sets that are collected under suction (page 10012 line 3) because that is the only way that in sandy soils solute samples can be collected because water will bypass gravity samplers in sandy soils. Data collected for sandy soils with suction are available such as by Boll et al (1997)

In all an interesting article but it should be set better in what is known about preferential flow since and when it was first rediscovered in the 1980's after it was first mentioned by Laws et (1882) hundred years before that. In addition more information is needed on how the data were analyzed

Reviewers name: Tammo Steenhuis, Department of Biological and Environmental Engineering Cornell University Ithaca NY 14853 USA

References

- J. Boll, J.S. Selker, G. Shalit & T.S. Steenhuis. 1997. Frequency distribution of water and solute transport properties derived from pan sampler data. *Water Resources Research* 33: 2655-2664
- Darnault, C.J.D., T.S. Steenhuis, P. Garnier, Y.J. Kim, M.B. Jenkins, W.C. Ghiorse, P.C. Baveye & J.-Y. Parlange. 2004. Preferential flow and transport of *Cryptosporidium parvum* oocysts through the vadose zone: Experiments and modeling *Vadose Zone Journal* 3:262–270 -
- Iversen, B.V., C.DBørgesen, M.E., Lægdsmand, M.H. Greve. G. Heckrath, C.E. Kjærgaard. 2011. Risk Predicting of macropore flow using pedotransfer functions, textural maps, and modeling. *Vadose Zone J.* 10:1185–1195
- Kim, Y.J., C.J.G. Darnault, N.O. Bailey, J.Y. Parlange & T.S. Steenhuis. 2005. Equation for Describing Solute Transport in Field Soils With Preferential Flow Paths. *Soil Science Society of America Journal* 69: 291-300
- Koestel, J.K. J. Moeyss and N.J. Jarvis 2011. Meta-analysis of the effects of soil properties, site factors and experimental conditions on preferential solute transport. *Hydrol. Earth Syst. Sci. Discuss.*, 8, 10007–10052
- Kung, K.-J.S. 1990. Preferential flow in a sandy vadose zone: 2. Mechanisms and implications. *Geoderma* 46:59–71

Kung, K.-J.S., T.S. Steenhuis, E. Klavdivko, T.J. Gish, G. Bubenzer, and C.S. Helling. 2000. Impact of preferential flow on the transport of adsorptive and non-adsorptive tracers. *Soil Sci. Soc. Am. J.* 64:1290–1296.

Lawes, J.B., J.H. Gilbert and R. Warington 1882. On the amount and composition of the rain and drainage water collected at Rothamstead. Williams Clowes and Sons, London

Park C-H, C Beyer, S. Baue and O Kolditz 2008 A study of preferential flow in heterogeneous media using random walk particle tracking. *Geosciences Journal* 12: 285 -297, September 2008

Steenhuis T.S. and L.M. Naylor 1987. A Screening Method for Preliminary Assessment of Risk to Groundwater from Land-Applied Chemicals. *J. Contam. Hydrol.* 1: 395-406

van der Molen, W.H. 1956. Desalinisation of saline soils as a column process. *Soil Sci.* 81:19–27

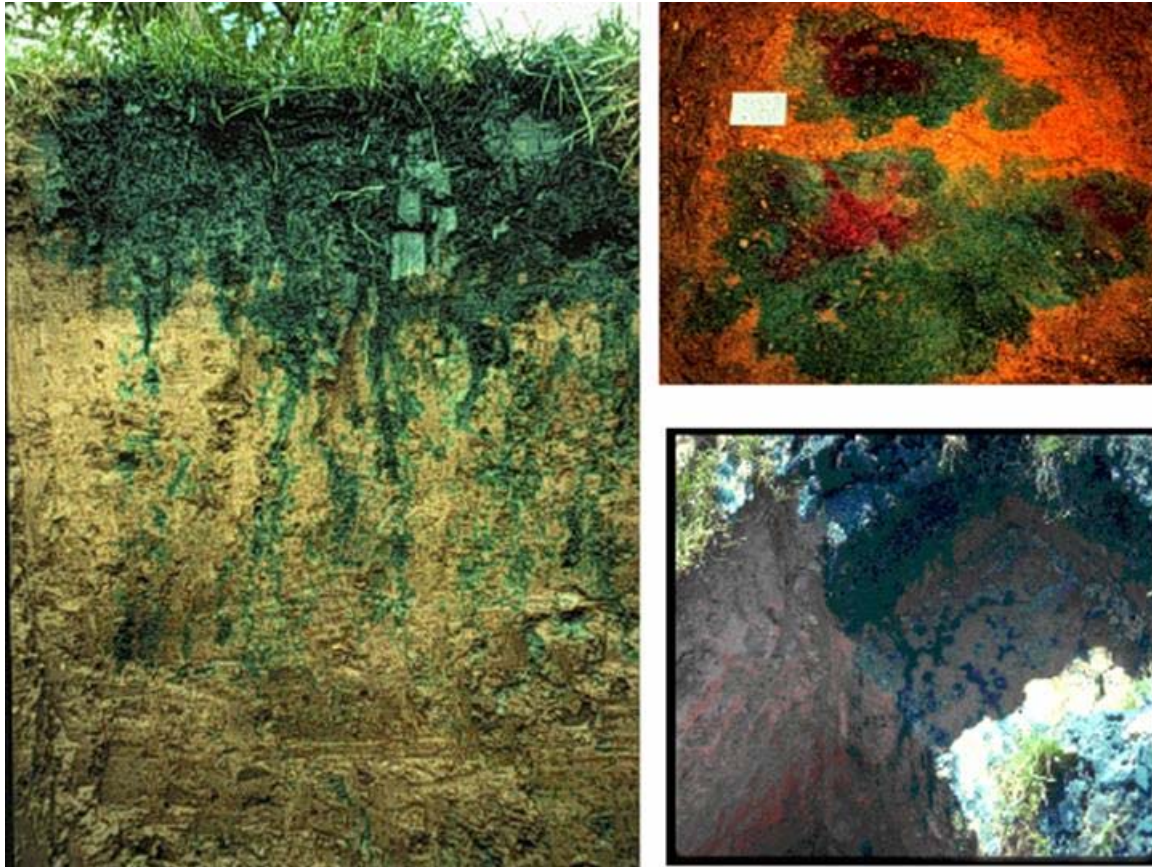


Figure 1: Examples of use of colored vegetable dyes for revealing soil structure heterogeneity on chemical transport. Left: Vertical excavation revealing a plow layer of about 10 cm. Below the plow layer, the blue dye reveals preferential paths. Top: Using red and blue dyes applied at different times provides an indication of the diffusion rate from macropores to surrounding micropores. Bottom: Deep macropores about one meter below the soil surface. (Source: Soil and Water Lab, Cornell University).

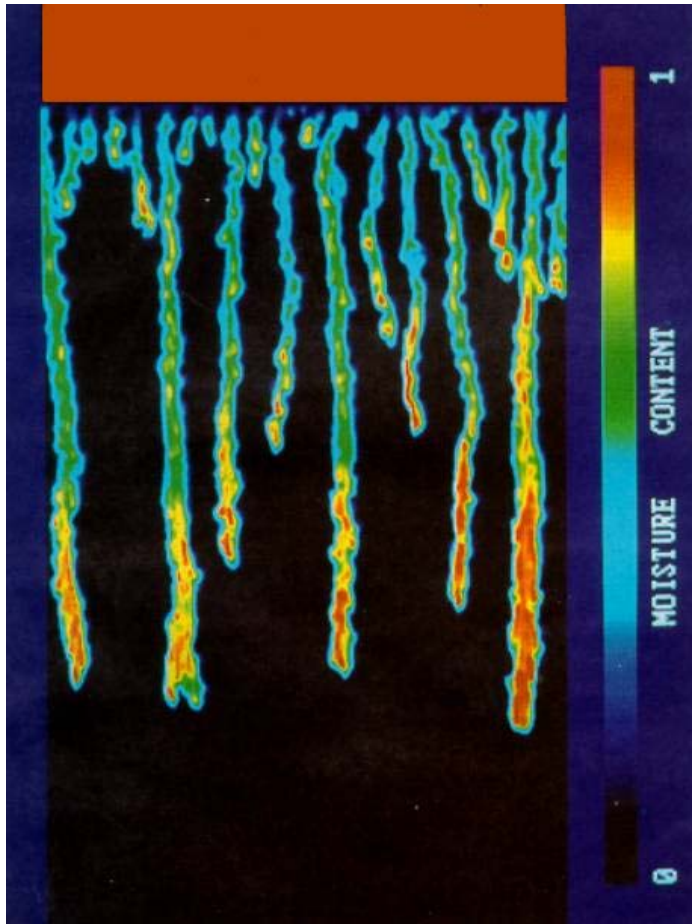


Figure 2: Fingers in homogeneous sandy soils with a mixing layer in the fine sand overlaying the coarse sand . These images are produced by passing light through a "sand sandwich" and converting the different intensities to different colors by a computer program. The black represents regions of low moisture content and red represents soil-water saturation. (The red for the mixing layer is drawn). The range of colors between black and red represent degree of moisture saturation.(Source: Soil and Water Lab, Cornell University)