

**Replies to Interactive comments of Referee #2, Tammo Steenhuis
on**

“Meta-analysis of the effects of soil properties, site factors and experimental conditions on preferential solute transport” by J. K. Koestel, J. Moeys and N. J. Jarvis.

The referee’s comments are shown in blue.

Our replies are shown in black.

Solute movement prediction was initiated by van der Molen (1956).

We agree that van der Molen (1956) was one of the first to apply a physically based model to solute transport through soils. We would like to mention that there were other similar studies published before or at the same time as the one of von der Molen (1996). For example, van der Molen (1956) cites *Rible and Davis* (1955) as having undertaken a similar approach. Other examples are the publications of Yuhara (1953) and *Day* (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.

We agree that van der Molen (1956) applied the convective dispersive equation (CDE) which had been developed for ion chromatography some years earlier (*Glueckauf*, 1949; *Lapidus and Amundson*, 1952) to soil columns. In his article van der Molen argues that the CDE should be applicable to Dutch soils because “under natural conditions the streaming velocity is extremely small ... [and therefore it] is not likely that non-equilibrium conditions will be important (page 20)”. In the next sentence he warns the reader that equilibrium conditions are not likely for heavy soils with a clay content of above 20%, because there, “large cracks are likely to be present which allow a rapid downward movement of water”. Obviously, van der Molen knew about what is today called “preferential flow”.

Based on the results of our study, we would say that preferential macropore flow paths (like large cracks) can already be present in soils with a clay content of 10% or more. In any case we thank the reviewer for mentioning von der Molen (1956), a reference which had been previously unknown to us.

Notably, the CDE was also derived by *Scheidegger* (1954) for porous media in general, independently and probably unaware of the contemporary research on ion chromatography. Some years later *Scheidegger* (1958) showed that the CDE is not valid if the transport velocities are auto-correlated as it is the case in preferential transport.

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.

(see reply above).

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”.

It is obvious that the reviewer has unfortunately misunderstood what we used the CDE and MIM parameters for. We agree that the respective passage in the original manuscript did not overly well describe that. To illustrate more clearly what we used the CDE and MIM model parameters for, we have now modified the passage:

See on p. 5, l. 127 – 153 as well as p. 6-7, l. 168-191 in the modified manuscript.

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.

We advise the reviewer that we do not assume convective dispersive transport. In fact, we do not assume any transport process at all (see reply above).

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 advise the reviewer that this article is not about model application or solute transport prediction. We do not apply any model to infer to any transport process. We use the parameters of the CDE and the MIM published in peer-reviewed literature to reconstruct BTCs. However, we do not make any assumption for the BTC reconstructions except that the model which was applied in the respective articles fitted the data well enough to describe the BTCs shape (also see replies above).

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.

The majority of the data published in peer-reviewed literature has unfortunately a rather poor record in how well the chosen transfer function fitted the first 1% of the tracer arrival. In most cases, the data sampling frequency and resolution is not even adequate enough to investigate this. In a previous study we found, however, that the differences in estimating early tracer arrival were very small between 7 different transfer functions fitting 115 BTC dataset collected from various publications (Koestel et al, 2011; the reference is given at the end of this text).

Furthermore, the relative tracer arrival-times of < 0.05 (i.e. $p0.01 \dots p0.05$) were very strongly correlated with Spearman rank correlation coefficients of larger 0.97. We therefore inferred that it suffices to just consider one relative tracer-arrival time for classifying the BTC-shape. We chose the 5%-arrival time ($p0.05$) because it had been already investigated by Knudby and Carrera (2005) who approve $p0.05$ as an indicator for preferential transport (note that Knudby and Carrera (2005) used $1/p0.05$ whereas we used $p0.05$ directly).

Besides, we have now modified Table 1 according to the reviewer's remarks. Table 1 now also contains information on the median coefficient of determination published in the source literature (if published) and an indication in which form the BTC data was used in our meta-analysis.

We furthermore have applied more stringent data selection standards which lead to reduction of the dataset from 793 to 733 (also see replies to reviewer 1). Tables, Figures, References and all numbers in the manuscript have been modified accordingly. Note that the results are only marginally affected by the reduction of the dataset size.

A detailed description about how we selected the data used in our manuscript is now given at:

See p. 5-6, l. 148-153 in the modified manuscript.

One of the objectives of the manuscript is the find pedotransfer functions.

We are afraid that the reviewer has unfortunately misunderstood what the aims of our manuscript are. It is certainly not the objective of our study to find pedotransfer functions. To point this out, we have now slightly modified the corresponding passage in the introduction:

See p. 4, l. 101-105 in the modified manuscript.

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.

We agree with the reviewer that the boundary conditions are extremely important for prediction of preferential flow in general. However, there are an infinite number of possibilities in which a time-series of precipitation, evaporation intensities and bottom BC matrix potential are compiled for real natural boundary conditions. This makes it difficult to compare results.

In theory, controlled (transient) boundary (and also initial) conditions which are more similar to natural initial and boundary conditions than steady state conditions could be simulated in the laboratory which would at least theoretically enable a comparison between different soils. But in order to simulate these boundary conditions (BC) the bottom BC should be recursively controlled from the water saturation states in the soil column. The first author of this here manuscript is not aware of any publication where this has been done so far. Even BTC

experiments where (non-periodical) transient upper boundaries were used are rare. One such example is (Zurmühl, 1998).

Our meta-analysis therefore focuses on BTC experiments under hydrologic steady state conditions. Hence, our study only covers a fraction of the relevant preferential flow processes in the environment. We give a caveat about this:

See p.17, l. 483 – 493 in the modified manuscript.

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 the not the amount of rainfall since the velocity is constant. It also shows the limitations of the pedotransfer functioning.

We completely agree. We are and were fully aware of this fact. Again we advert to the caveat given at the end of the manuscript:

(see reply above)

The manuscript by Koestel et al (2011) is confusing and it might be very well that I have interpreted the text wrongly.

We are afraid that this has been obviously the case for some critical parts of the text. We believe that the manuscript is much less misleading with the modifications introduced in its new version.

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.

We unfortunately do not understand this comment. We did not divide the CDE parameters by any 'quantity' to make them 'independent' from the CDE. Furthermore, the reviewer hopefully agrees that the average velocity is defined as the mean transport velocity of the total tracer mass, independently of the transport process or regime. How can there be more than 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 break through curves.

Again we believe that the modified manuscript explains much better that we did not fit any BTC with the CDE:

See on p. 5, l. 127 – 153 as well as p. 6-7, l. 168-191 in the modified manuscript.

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 exponential decreasing concentration is more appropriate than a Dirac pulse input (details are given in Darnault et al, 2005 and Kim et al, 2004).

We like the figures added by the reviewer but otherwise state again that it was not our aim to model preferential transport. Instead we aimed at comparing BTC-shapes, including both, BTC from preferential and non-preferential transport processes. We chose the Dirac-pulse like input for forward-modeling to standardize all BTC-data to one distinct tracer application. We advise the reviewer to the fact that not all collected BTC data resulted from preferential flow.

We have therefore removed the term "preferential" from the title of the original manuscript, admitting that using it in the title was rather misleading.

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 solutes to leach. The 5% is arbitrary

We do not agree with the reviewers comment that the 5%-arrival time is an arbitrary choice as it has been investigated by (*Knudby and Carrera, 2005*) and found to be an indicator for preferential transport.

See p.8-9, l. 218-235 in the modified manuscript (also see reply above).

and likely too large.

We agree with the reviewer that the 5%-arrival time may be too large for practical applications. However, as an indicator for early tracer arrival times, this appears not to be so important. For

the BTC data sampling frequency commonly applied in the literature. *Koestel et al.* (2011) 'Evaluation of Nonparametric Shape Measures for Solute Breakthrough Curves, *Vadose Zone J.*, 10(4), 1261-1275' showed that the 5% arrival time was highly correlated with the 1% arrival time (Spearman rank correlation coefficient of 0.97) for BTC data of 115 BTC sampled from peer-reviewed literature when using the transfer-function (out of seven) with the best Akaike information criterion to fit the BTC data (also see reply above).

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 theoretical the same concentration is obtained when the pesticide is adsorbed, Steenhuis and Naylor, 1987). The concentration in the water will be: $2 \text{ (kg/ha)} / (0.1 \text{ m} \cdot 10,000 \text{ m}^2) = 2 \cdot 10^{-3} \text{ kg}$ or 2,000,000 $\mu\text{g/l}$. Dissolving 5% of this high concentration in the total rainfall still gives a concentration of 100,000 $\mu\text{g/l}$! 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 $\mu\text{g/l}$, five percent of an application of 2kg (or 100 g) of pesticide can bring 10,000 m^3 /ha of ground water up to a concentration of 10 $\mu\text{g/l}$. 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 ground water concentration are not observed, because pesticide degrade.

(see replies above)

In order for the reader to make its own conclusion, Table 1 need much more information on the type of data that each articles provides how many breakthrough curves could be fitted to convective dispersive equation.

We added this information in Table 1 (see reply above).

It also need to state exactly how the analysis was done and for each experiment and what the calculated four non dimensional parameters were

We have modified the material and methods part which describes how the 4 non-parametric shape-measures were calculated:

See p.6-10, l. 174 – 263 in the modified manuscript

and how many of the BTC's curves could be fit with reasonable numbers.

We have included this information in Table 1 and also explained our data selection rules in more detail in the modified manuscript:

See p. 5-6, l. 148-153 in the modified manuscript.

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.

(see replies above)

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.

(see replies above)

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).

We had considered including these data. We had decided against it because there are strong indications that BTCs sampled with suction cups are not representative, especially not for heterogeneous soil where preferential flow is likely (see e.g. *Lutz Weihermueller et al.*, (2006) and *L. Weihermueller et al.* (2011).

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.

We know about (*Lawes et al.*, 1881a; b; 1882). We would like to add that had been also mentioned by (*Schumacher*, 1864) some years earlier and afterwards by (*Burger*, 1923; *Mosier and Gustafson*, 1917) and even in the 1950s, e.g. by (*van der Molen*, 1956).

In addition more information is needed on how the data were analyzed

We have added extra information on how the data were analyzed.

See p.5-10, l. 127 – 263 as well as Table 1 in the modified manuscript.

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