Interactive comment on "Quantifying heterogeneous transport of a tracer and a degradable contaminant in the field, under two infiltration rates" by D. Schotanus et al.

Anonymous Referee #1

Received and published: 23 May 2012

Comments to
Quantifying heterogeneous transport of a tracer and a degradable contaminant in the field, under two infiltration rates
by: Schotanus, D., van der Ploeg, M.J., and van der Zee S.E.A.T.M.

We thank the anonymous referee for his/her review of our manuscript. The comments helped us to improve the manuscript.

General Comments:
Schotanus et al. presented in the manuscripts (ms) a field experiment where they examined the transport behaviour of two contrasting substances (conservative tracer and degradable de-icing liquid) under natural infiltration after a snowmelt event and artificial irrigation. In general, the ms is of high interest to the community due to the analysis of the local breakthrough of the substances. Especially, the measurement of persist preferential flow patterns has not been reported under field conditions yet. Therefore, the ms fits into the scope of HESS. Nevertheless, the ms in its present form is returned to authors for major revisions for several reasons. 1. The introduction needs revision in terms of introducing the state of the art and in the line of argumentation (see also specific comments). 2. Some of the conclusions are not supported by the data presented or are not discussed properly
Considering both reviews, we appreciated the need to rewrite parts of the manuscript, including the introduction and conclusion section. In the present form, we are confident (and we have, of course checked this point by point) that all conclusions are soundly based on the (revised) manuscript.

Specific Comments:
Abstract:
In general, the abstract needs revision in terms of wording and line of argumentation.
P1 L2: "of a non-degradable tracer" – per definition a tracer should be non-degradable and should not sorb. Better use "conservative tracer"
To avoid ambiguity, we deleted the 'non-degradable' and now just refer to 'tracer'.

P1 L3: should be:..field experiments were performed
P1 L5: should be: During the second. . ..
P1 L8: should be:..correlated, indicating that. . .
We changed this in the ms.

P1 L8: here you argue that the flow path are stable between the seasons which will not be supported by the data presented. The only conclusion you can draw is that they are comparable between the two experiments. Later you argue that the preferential flow is not induced by macropores but by local differences in soil hydraulic properties. But if you assume that this will be the case, flow pattern will change in dependency of the flow rate imposed (or the actual water content). This has been already shown by Roth (1995).
The phrasing of the reviewer is more exact. Hence, we changed the comparison between seasons, in the new version we conclude that the flow paths were stable between the two experiments.

P1 P9-11: weak sentence
P1 L11: should be: Therefore, . . .
We changed this in the ms.

P1 L13: What do you mean by clustering?
By clustering we mean that several high leaching cells tend to be close to each other. We changed this sentence to: 'With higher infiltration rates, the high and low leaching cells were more clustered.'.

P1 L13: the leached mass. . . - this should be the case if the substance is degradable and if the temperature is high enough. On the other hand, the second part is highly interesting but due to the constant flow not surprising. Please discuss critically.
The flow was not constant. We clarified the discussion of this finding is in the results section, lines 429-436.

P1 L15-16: Please rewrite the sentence and do not list up some findings without giving any interpretation or discussion here.
We have rewritten these lines, while keeping in mind the abstract should be concise. The full discussion can of course be found in the result section.

P1 L18: weak sentence and line of argumentation
We changed this sentence to: "The persistence of the flow paths during the experiments suggests soil heterogeneity as the driver for heterogeneous flow and solute transport, in this soil. At the MCS scale heterogeneous snowmelt did not seem to have much influence on the flow and solute paths."

Introduction:
In general, the introduction needs strong revisions in terms of references used and the line of argumentation. For example, there is a nice review from Javis (European Journal of Soil Science, 2007) which should give some background information and should also be cited.
We included review papers from Flury (1996), Simunek et al (2003), Jarvis (2007), Feyen et al (1998), Weihermüller et al (2007), and Allaire et al (2009).
Furthermore, we streamlined the introduction for a better line of argumentation for our objectives.

P2 L22: should be core, profile, and landscape scale
We changed this in the ms.

P2 L25: when average parameters. . .- for the soil hydraulic properties this is not an average of all single properties but an effective set of parameters. For climatic parameters this is slightly different, because short time high flow will not be captured by the daily mean
We changed average in effective.

P2 L26: should be: hydrological models
We changed this in the ms.

P2 L26-27: weak sentence: maybe like this: To account for preferential flow in the models additional parameters are needed. . ..
We changed this in the ms.

P2 L28-29: weak sentence
We changed this sentence to 'It is important to identify the main process that causes preferential flow, when solute transport is modelled.'.

P2 L30: should be: . . .insight which. . .
P2 L30: should be: Based on this knowledge. . .
P2 L45: . . .is saturated. – close to saturation. Not necessarily at saturation
We changed this in the ms. Part of this paragraph has been removed for a better focus in the introduction.

P2 L50-52: what do you want to say? This seems logic if you will have a closer look at the conductivity function. In general, I do have the impression that you did not well capture the ideas presented by Roth (1995). In general, they presented a microscopic heterogeneity and not macroscopic large scale heterogeneity. Therefore, it would be would be hard to present K-values at this short distances.
It is apparent that we did not represent the Roth paper such that the content is represented well enough. However, we disagree about what he represented: hydraulic conductivity was indeed an autocorrelated lognormally distributed property at a scale larger than that of microscopic variability, hence the 'channeling' type of flow. However, this part has been removed from the ms for a better focus in the introduction.

P3 L56: What do you mean? If the soil is dry you will not have any preferential flow at all. I believe you would like to state something different such as the flow path will change due to. . .. But if you do so please provide reasons why.
We mean that preferential flow paths might originate elsewhere after saturation or drying. However, we removed this part from the introduction.

P3 L56-57: Not necessarily. It depends on the conductivity in dependence of the actual water content. Please clarify.
P3 L57-58: see comment above
P3 L61: This might be the case here but if you will have swelling or if cracking might occur this might look differently. Maybe not in effective parameters such as velocity and dispersivity but maybe in the transport pathways.
These parts have been removed from the ms for a better focus in the introduction.

P3 L67: please delete: when macropores exist than when only matrix flow occurs. Or reformulate
We deleted 'when macropores exist'. We want to compare the effects of macropore flow and matrix flow for degradable solutes. Therefore, we still include matrix flow in this sentence.

P3 L68: biodegradation – this is not only restricted to biodegradation. Also thermal degradation and photodegradation might play a role.
We agree with the reviewer and removed "bio".

P3 L74: weak sentence.
We changed this part of the sentence to 'because the biodegradation rate depends on temperature, and soil moisture content, amongst others'.

P3 L79: using wick samplers. . . or lysimeters, or porous plates as shown for example by Kasteel, R., Pütz, T., & Vereecken, H. (2006). An experimental and numerical study on flow and transport in a field soil using zero-tension lysimeters and suction plates. European Journal of Soil Science, 58(3), 632-645.)
We added this to the ms.

P3 L80: I do not see the problem here. The biggest problem is the limited range of pressure, and therefore, that water will not be sampled if the soil is not close to saturation. There is also a review available dealing with such instruments from Weihermüller (2007, JEQ)
A wick sampler applies a constant pressure. With a constant pressure, water may flow to the sampler, or around the sampler, depending on the pressure in the sampler and the soil. With a variable pressure in the sampler that is adjusted to the pressure head, this effect will be much smaller. In the reference from Weihermuller also porous plates with variable pressure are discussed. We included this review in the ms.

P3 L82: To overcome these disadvantages Bloem et al. . ..
P3 L83: Additionally, the spatial resolution of the MCS is high and fluxes through individual 3.15. . .. . .
We changed this in the ms.

P3 L85-89: What do you want to say?
P4 first paragraph: Please do not list up all information you can find. Better to put them into a nice order of argumentation.
We changed the order of the sentences in this part.

P4 L13: first point: I do not really believe that you can answer this question based on your given experimental setup. See also all detailed questions raised above.
We extended the research question, such that it can be answered with the experimental setup.

Materials and Methods:
P4 L123: Reference should be at the end of the sentence. Or better state that details can be looked up at French et al. (1994).
We changed this in the ms.

P5 L131: The pressure in the MCs was variable. – In space or time?

The pressure was variable in time, we added this to the ms.

P5 L131-134: These are general statements, and therefore, should be provided earlier.
We moved this part to the beginning of the materials and methods section.

P5 L146: should be: the infiltration per day was calculated.
We changed this in the ms.

P5 L156: Is this caused by differences in water content? This should be critically discussed.
Referring to 'the transport of bromide was found to be too slow', we believe that this was partly caused by that the evaporation was higher than expected. In the results section we discuss that the soil moisture content was higher at the beginning of the irrigation experiment than of the snowmelt experiment. We also discuss how this influences the leaching.

P5L 164: please rewrite sentence.
We changed the sentence in 'Between 31 May and 2 June, evaporation was measured in a pan under the plastic. The measured evaporation was 1mm/d.'.

P6 L166: drainage was collected in the trench. . . - I do not believe so. It was collected in some containers or bottles which might be located in the trench.
We changed this in 'Drainage was stored in the MCS, samples were taken with a frequency that depended on the amount of drainage.'.

P6 L186: all parameters should be in italic such as I and j.
We agree, as far as we could find, in the ms all parameters are in italic, and we hope that we did not miss any.

Discussion and Discussion:
P6 L197: Please explain what you mean by drainage depth.
We changed this sentence to 'Figure 4 shows the spatial distribution of the volume of drainage, and of the bromide concentration during the snowmelt experiment at five selected cumulative drainage depths (the amount of total drainage since solute application).'.

P7 L213: . . ..concentration of bromide. – But maybe largest mass was transported here. Concentration is not always the best indicator for solute translocation.
In this section only concentrations are discussed, because we study the spatial patterns in time. In section 2.3.3 we investigate the leached masses per cell.

P7 L227: or there was only local infiltration due to heterogeneous snow melt water
A preliminary experiment was done at the same field site and with the same instrumentation during the snowmelt of 2009. We compared the areas of the sampler in which most leaching occurred. These were the same for both years. However, we did not include the results from this experiment in the ms, because it was a preliminary experiment, with less elaborative measurements.
We added a sentence about the preliminary experiment to the results section: 'Results from a preliminary experiment, which was performed during the spring of 2009, confirm this conclusion. The areas with the highest leaching were similar for 2009 and 2010, which means that the effect the heterogeneous infiltration of meltwater is small compared to the effect of soil heterogeneity. '.

P7 L231ff: I would calculate a worst case scenario and see if the density differences realy play an important role.
We asked an expert on density driven flow, who said that this density difference would influence the transport. This statement is based on that.

P8 L236: . . . by micro-organisms. Or any other mechanism (see comment above)
We agree with the reviewer and removed "bio".

P8 L237-239: You did not mentioned so far how you fitted the BTCs. This should be stated somehwre in the materials section.
We used CXTFIT to fit the transport parameters for the BTCs. We added this to the materials section.

P8 L240 should be: mean soil water content
We changed this in the ms.

P8 L240-242: how much do these mean SWC differ from those calculated from the solute transport parameters?
We changed Fig. 7, now the pressure head is shown instead of the soil moisture content, as the pressure was measured (not the soil moisture content). We removed the soil hydraulic parameters from the ms. The only remaining solute transport parameters are the velocity and dispersivity.

P8 L246ff: all units are missing for the water contents. Please check entire ms carefully.
In the new version of the ms, we use pressure head instead of water content.

P8 L249: should be. . ., and therefore, . . .
P8 L249: maybe better: the coarser material is even higher conductive as during lower water contents.
P8 L257: . . . concentrations, as plotted in Fig. 5 and 6, were. . ..
We changed this in the ms.

P8 L261: in general,mixing is also time depending!
We agree. We added to the ms: 'With a high infiltration rate and a wet soil, a small part of the soil can transport solutes rapidly, while little dilution occurs due to a low exchange with the surrounding soil, as lateral mixing is time dependent.'.

P9 L276ff: does this argumentation contradict the entire argumentation you did before. Please discuss critically and consistently.
We agree, this part is confusing and does not add to the conclusions. We removed it from the ms.

P9 L280: should be times instead of time steps.
We changed this in the ms.

P9 L290: why did you assigned it to day 2 if there is no leaching at all? Logically, they should either not be included or you can also include a bar at the end indicating all no-flow cells or flow cells without complete
We agree, we changed this figure.

BTC P10 L309: The argumentation is OK but not fully explained. If you will look at classical transport theory, you will encounter that spreading generally increases with increasing travel time (or distance). Consequently, the peak decreases (if you do not lose any mass). Secondly, faster movement reduces mixing between different flow channels or flow pathways with different concentrations, leading consequently to larger concentrations in the observed single BTCs.
P10 L311: Which should be clear if you follow up the argumentation given above.
We agree, we added to the ms 'This is expected, as the spreading generally increases with time, which will lead to a lower peak concentration for a non-degradable solute.'.

P10 L312: should be: in Fig. 10, the solute transport. . ..
We changed this in the ms.

P10 L312-317: You should provide some more details in the materials section how you fitted the CDE.
We added this to the materials section. 'The velocity, dispersivity, and first order degradation constant were estimated by fitting de convection-dispersion equation to the BTCs with the parameter fitting program CXTFIT (Toride, 1995). The dispersivity that was estimated for bromide, was also used for PG. The velocity and first-order degradation constant for PG were fitted.'

P10 L317: should be: back-transformed to units of. . ..
We changed this in the ms.

P10 L318: Do not use alpha for dispersivity because you already used it as a MvG parameter.
We removed the Van Genuchten parameters from the ms. Therefore, we can keep using alpha for the dispersivity.

P10 L319: should be: As expected, for the fast. . ...

We changed this in the ms.

P10 L320ff: In general, the discussion about the dispersivities is weak. I would like to see some hypothesis and a critical discussion here.
This part is completely rewritten. We included a more extensive discussion of the dispersivities, and compared it with literature.

P10 L330: longer, and therefore, . . .
We changed this in the ms.

P10 L340ff: I would not use only the peak concentration for the estimation of the degradation. Classically, the entire mass (or mass recovery or leached mass fraction) will be used.
We agree that the degradation rate should not be estimated on the peak concentration. Here, we only use the concentrations to study the PG and bromide leaching in time. We estimated the first-order degradation constant for PG with CXTFIT, as is now described in the new ms.

P11 L350: should be K_eff in italic as all other parameters.
We changed this in the ms.

P11 L351ff: I do not understand why you need a full BTC. You can still fit the CDE to incomplete tracer breakthrough curves.
We did fit the CDE to incomplete BTCs, for both bromide and PG. As we grouped the cells in fast, average and slow leaching cells, we do not know the initial mass of bromide and PG. This mass did not infiltrate uniformly. To fit the CDE we estimated the initial mass of bromide for the fast, average and slow cells separately, based on the incomplete BTCs. This initial mass of bromide is very important to estimate the first-order degradation constant for PG. We believe the initial mass is too uncertain to estimate reliable first-order degradation constants for PG, for the separate groups. For the three groups together, the uncertainty is less, and we are confident to estimate one degradation constant for the entire sampler.

P11 L357: This has been already shown by several other authors, therefore discuss in this context. A recent numerical study by Weihermüller et al. (2012, VZJ) indicates the same features. Additionally, they introduced (or better used) the concept of leached mass fraction which might be also of interest over here for a better and quicker interpretation of the data shown.
P11 l367: see comment above how to handle this.
We discuss the spatial distribution of leaching in fast and slow transporting regions in a horizontal plane. Furthermore, we compare the leached fractions of a degradable solute to the leached fractions of a tracer. In the study of Weihermuller et al, concentrations and fractions for multiple solutes that are extracted with one suction cup are compared. The comparison between fast and slow regions is not made in that study. Therefore, we think it would not make a good contribution to our discussion to include a comparison with Weihermuller et al.

P11 L375ff: I do not fully agree. Even if you will have large heterogeneity, the flow will be homogeneous at pressure heads where all conductivity curves cross. Please check again the paper by Roth (1995).
The paper by Roth described a field with Miller Miller similarity. Real field soils might not have this similarity. Furthermore, we study a transient system, while the paper by Roth described a steady state system. Both differences, we expect, influence the results.

P12 L392: I wonder. Is there a theory for that or do you refer to a bad experiment to excuse another bad experiment. There should be always close mass balance closure. Maybe you can find some hints in the paper by Kasteel et al. EJSS (2006)
Part of the bromide might still leach, because it might be stored in less mobile regions, and the BTC was incomplete. However, a storage of more than 50% of the applied bromide in immobile regions would be too high. Part of the bromide may have been taken up by plants.

P12 L392ff: As long as you do not have mass balance close I would be careful with such statements
We include a sentence about the incomplete BTCs, and how this affects the estimated degradation. We believe that this gives a good indication that the amount of degradation should be used with care.
'As mentioned before, the BTCs of bromide were not complete before the irrigation experiment was

stopped, and thus the bromide leaching was underestimated. This implies that the PG degradation was also underestimated.'

P12 L402: should be: . . .cumulated as proposed by Quisenberry et al. (1994), Strock et al. (2001), and De Rooij and Stagnitti (2002). . .
We changed this in the ms.

P12 L402: Do not start up with Doing so. And rewrite entire sentence.
We changed this sentence in 'The sorting of the cells removed all spatial information.'.

P12L407: Please be consistent and use also percentage in the plots (Fig. 13)
We changed this, the area is now expressed as percentage and not in m^2.

P12 L409ff: I do not understand at all. Please explain. Is this caused by changes in the water storage term during drainage?
We added the sentence 'Precipitation events after the snow had melted, or the different irrigation rates may result in a varying water storage in the soil. This water storage affects the spatial distribution of the drainage. Therefore, it is more correct to use the leaching instead of the drainage, as the leaching is labelled using a pulse, and the drainage volume itself is not labelled.'.

P13 L 419ff: I would not use the term line.
We changed 'line' in 'curve' in the ms.

P13 L426-427: That's a fairly general statement. But what do you want to say here?
We changed the order of sentences in the ms. We want to make a comparison between the laboratory and a field situation.

P13 L428: The drainage during. . . What do mean? Amount, timing, special occurrence? Please specify.
We changed this in 'The total drainage that occurred during the snowmelt experiment and the irrigation experiment…'.

P13 L430 . . .bromide leaching. . . same comment as above
We changed this in 'The total bromide leaching that occurred during the snowmelt experiment and the irrigation experiment…'.

P13 L431-432: I do not understand what you want to say.
We rewrote the sentences: 'We found that the bromide leaching may be different in the exact locations (Fig. 13), even though the area of the MCS with the highest bromide leaching is similar for both experiments (Fig. 11). Thus, the area with the highest leaching is persistent in time, but the exact locations of the highest leaching might change a bit in time.'.

P13 L435ff: Which should be clear. Do you expect something different? I would say no as long as you do not have any sorption.
When transport is slow compared to the degradation rate, we would expect a different conclusion. In that case, the PG leaching would be 0 in cells with low Br leaching (slow cells). Then, the correlation between PG leaching and Br leaching would have been lower.

Conclusion:
P13 L 470: I would not talk about any seasonal effect. See also the comments raised before. In general, the conclusion should be adapted to the comments raised above.
We agree that from the results of our two consecutive experiments no conclusions can be drawn about seasonal effects. Therefore, we changed 'seasonal effects' in 'effects of snowmelt'.
We checked all conclusions, and found that they are all based on the results.

References:
In general, all Journal titles should be in capital such as: Journal of Hydrology. Please check entire reference list.

Tables:

Tab 2:Caption should be: Solute transport parameters with pore water velocity v, dispersion coefficient D, and dispersivity lambda, (equation) for fast, average, and slow cells, as well as for the entire sampler. Does it make sense to show also dispersion coefficient if you also show dispersivity? All units are missing in caption.
We changed the caption and removed the column with the dispersion coefficient.

Tab. 3: Caption should be: . . . Moran's I, A with 1 indicates perfect spatial autocorrelation, and -0.01 no spatial autocorrelation
We changed this in the ms.

Figures:
Fig. 3: should be : daily mean air temperature. All units are missing.
We changed this in the ms.

Fig. 4: delete same remarks as. . . and add full figure description because it is not the same as in Fig. 3.
We changed the captions of Figs 3 and 4.

Fig. 5: Why don't you rescale the drainage in the legend instead of suing 10 by the power of -1? Caption: should be of the cumulative drainage since solute application. Consequently the last sentence can be deleted.
We changed this in the ms.

Fig.6: same as above
We changed this in the ms.

Fig.7: I would always sue full units or at least define it as volumetric water content. Otherwise it can be also gravimetric water content
We changed Fig. 7, now the pressure head is shown instead of the soil moisture content, as the pressure was measured (not the soil moisture content).

Fig. 8: Delete "marked with ellipses"
We changed this in the ms.

Fig. 9: daynumber is somehow confusing.
We changed this in 'day since solute application'.

Fig. 12: There is some duplication in the caption: What do you mean by parallel flow? Do you mean 1-D vertical flow?
We changed parallel flow in uniform flow in the captions of Figs 14 and 12, which is generally used.

Fig.13: Should be Normalized cumulative drainage. . . Delete the unit at the end of first sentence. Maybe better to express cumulative area in percentage as you did in the text. Should be drainage, bromide, or PG leaching.
We use cumulative sampler area in percentage instead of m2.We changed the caption as suggested.

Fig. 14: indicate the 1.1 line in the plots with 1:1. Again what do you mean by parallelflow?Delet the last sentence if you add in the 1:1 in the plots.
We included '1:1' in the figure, and removed the last sentence of the caption.