Response to the comments of Anonymous Referee #2

General comments

This manuscript presents a solution of the steady state transport equation in nonstationary unsaturated flow field. In the present form, it is neither a technical note nor a full paper. In the Introduction, the authors stated that the objectives of the manuscript are to address the impact of flow non-stationarity on macrodispersion in an unsaturated flow. Given these premises, I was expecting a full paper addressing at depth and with some generality the impact of flow unsteadiness on macrodispersion. Unfortunately this is not the case. The manuscript focuses on a particular solution of the steady state version of the transport equation with a very limited discussion and no justification of the strong hypotheses introduced at the beginning of the mathematical derivations. On the other hand, this manuscript can hardly be defined a technical note because it does not address a well defined technical issue. The authors, after introducing some relevant simplifications of the transport problem, present the solution followed by a very cursory discussion and a short conclusions section. The rather strong hypotheses introduced to simplify the problem limit the applicability of the solutions, but unfortunately the scanty justification of the simplifying hypotheses and discussion of the results make almost impossible to ascertain the value of the proposed solution. From a general point of view, to be useful a technical note should address a case of wide interest providing a solution easy to use. This is not the case of this manuscript, which provides a solution of a simplified (perhaps oversimplified) transport equation, which relevance is difficult to ascertain.

Reply

(1) To avoid confusion, we have modified a part of "Introduction" on page 3 (Line 13) to explain the problem we intend to solve and specify the task we plan to perform as:

"The Eulerian analysis of field-scale solute transport in heterogeneous media is built based on the solution of the stochastic convection-dispersion perturbation equation. As such, the macrodispersive flux (Gelhar and Axness, 1983), an outcome of the correlation between the velocity field and concentration fluctuations, is introduced to quantify the spreading of the solute plume at the field scale. Comprehensive overviews of the construction of the Eulerian approach and its application to the analysis of the solute transport in heterogeneous media were given by Gelhar (1993) and Rubin (2003). It is well known that the correlation between the fluctuations of the velocity and concentration enhances the degree of spreading of solute plumes (field-scale dispersion or macrodispersion) in heterogeneous aquifers that is much greater than what would occur by local dispersion (e.g., Gelhar and Axness, 1983). The quantification of the field-scale dispersion created by the heterogeneities of the geologic formations is therefore rather important in the analysis of unsaturated solute transport.

In this note, we attempt to quantify the field-scale dispersion at large times in nonstaionary unsaturated velocity fields analytically in terms of the statistical moments of two formation parameters, i.e., the Gardner's parameter (Gardner, 1958) and the saturated hydraulic conductivity. Eulerian approaches are well suited for modeling large-time transport of solutes in heterogeneous media. Therefore, the quantification of the field-scale dispersion in this study will be carried out within the Eulerian framework. Note that in this study the nonstationarity in unsaturated velocity fields is introduced due to the presence of a fixed head boundary condition at an arbitrary depth in the flow domain and a constant flux at the land surface. The present investigation of field-scale unsaturated transport processes at large times can be considered as an extended work to the previous study by Chang and Yeh (2009) for bounded unsaturated flow processes in heterogeneous aquifers. The large-time macrodispersive flux is quantified by applying Fourier-Stieltjes integral representations of the randomly nonstationary fluctuations (Li and McLaughlin, 1991). The process of solutes' spreading in field porous formations is dominated by the spatial heterogeneity of the formation properties. We therefore conclude by analyzing the influence of the correlation scales of these two parameters on the spreading of field-scale unsaturated plume, which is the main focus of this work.

Existing stochastic studies (e.g., Rubin and Bellin, 1994; Rubin and Seong, 1994; Indelman and Rubin, 1996; Zhang, 1999; Sun and Zhang, 2000; Foussereau et al., 2000; Destouni et al., 2001; Wu et al., 2003; Hu, 2006; Dai et al., 2007; Russo and Fiori, 2009, Lu et al., 2010) on the issue of transport process of field-scale plumes in nonstationary groundwater flow fields were mostly carried out using the Lagrangian perspective. The analytical analysis of the field-scale plumes in nonstationary unsaturated groundwater flow fields using the Eulerian concept has not been presented so far. This study attempts to analyze the spreading of plumes in field-scale unsaturated formation based on the Eulerian framework and nonstationary spectral approach. The findings presented in this note may be of interest to researchers in seeking for further research."

This note initiates the use of Eulerian concept to investigate the large-time solute transport in non-stationary unsaturated flow fields. To the best of our knowledge, this task has so far not been attempted. In other words, this manuscript presents a new development in quantifying the field-scale dispersion in non-stationary unsaturated flow fields using Eulerian approach. Such a work we think is rather technical and falls within the scope of technical notes.

(2) The dependence of the moisture content and the unsaturated hydraulic conductivity on the fluid pressure leads to strong nonlinearity in flow processes. Furthermore, the parametric description of unsaturated flow properties involves many empirical parameters. Generally, the evaluation of solute transport in a partially saturated heterogeneous formation cannot be performed analytically. The analytical expression is a handy tool in predicting field-scale spreading and exploring the role of heterogeneity scales of two formation parameters in influencing the large-time behavior of macrodispersion. Taking those advantages and realizing that the effects of local heterogeneity are quite dramatic, the simplifying hypotheses are inevitable in analyzing the influence of

heterogeneity on flow processes. In the manuscript, we have used the hypotheses reported in the literature of stochastic subsurface hydrology (e.g., Russo, 1993, 1996, 1998; Harter and Zhang, 1999) to quantify the influence of heterogeneity scales of the geologic formations. All the simplifying hypotheses used in the manuscript had been referred to those existing papers.

The validity of assumptions made in developing analytical expressions in the manuscript can be checked through the method of Monte Carlo simulations, which is indeed a time-consuming work. Moreover, the introduction and result discussion for such a method may make the body of the note become too bulky to be a technical note.

Specific comments

1. Flow is two-dimensional and nonuniformity is introduced by imposing a constant flux at the surface and a constant head at a given depth. In the horizontal direction the computational domain is unbounded. However, authors did not specify at which depth the head is imposed. I am puzzled by the relevance of this type of flow nonstationarity. As commented below the steady state hypothesis for the transport equation suggests that the authors are looking for the long time behavior of macrodispersion and in this case I am wondering if another type of nonstationarity emerges: the intermittence of precipitation. However, given for granted that a constant flux at the upper BC can be accepted in some situations, the only case I can figure out for which the lower BC is of constant head is when a water table is set at a given depth and the underlying aquifer is able to convey the entire recharge flux in order to keep the water table fixed in time. All these conditions are in my view quite unrealistic, in particular if we think that the domain is unbounded in the longitudinal direction.

Reply

(1) Certainly, the intermittence of precipitation may lead to another type of nonstationarity in velocity field. The introduction of the intermittence of precipitation into the unsaturated flow system as a source may cause non-uniformity in the mean flow field and consequently, nonstationarity in the statistics of pressure head and velocity fields.

A water table boundary condition (pressure head = 0) is often specified at the bottom of the simulation domain for unsaturated formations. One often considers the bottom pressure head to be a specific value if the water table is sufficiently deep and one is interested in near-surface processes. McCord (1991) has mentioned that in general, it is appropriate bottom boundary condition for a large number of practical cases.

(3) A constant flux at the land surface and a fixed head at depth in the unsaturated zone are commonly adopted as boundary conditions (BCs) in modeling unsaturated flow in a number of papers (e.g., Indelman, 1993; Zhang and Winter, 1998; Basha, 1999; Tartakovsky et al., 1999; Lu and Zhang, 2004; Lu et al., 2007)).

2. Transport is solved in stationary conditions, i.e. concentration does not change with time. This is a very peculiar situation, which is often implemented in order to model cases in which mass flux entering the system, for example in an aquifer through a well, is consumed by some reactions modeled as a sink term. This may occur, for example, when an organic contaminant is consumed by bacteria and the equilibrium between injected and consumed mass is typically reached at late time since the beginning of injection, which is supposed to be continuous. It is unclear to me what type of steadiness is reached in the case considered by the authors, considering that they do not specify the BCs for the transport equation. I am tempted to conclude that steady state in this case implies constant mass flux entering through the upper boundary and the same constant flux exiting from the lower boundary. If this occurs isn't that the solution is simply of constant concentration through the profile? However, if any steady state condition is attained it would require that the plume had traveled many length scales. This means that the correlation length of the saturated hydraulic conductivity cannot be increased freely, while keeping constant the depth of the computational domain as done in Figure 1. In fact, as the correlation length increases a larger distance is needed in order to reach steady state conditions and when this distance becomes larger than the formation depth the proposed solution cannot be applied. This is not investigated and discussed in the manuscript.

Reply

- (1) The plume can be stabilized for two- or three-dimensional dispersion, even if there is no sink term representing pumping or biodegradation in the subsurface system. When plumes develop in the dispersive media, their mass will be transferred perpendicular to flow lines (i.e., in the transverse and/or vertical directions) for multi-dimensional dispersion. Such a process will lead the plumes with smoothly decreasing concentrations toward their steady-state stage as demonstrated in Zheng and Bennett (2002, Figure 5.6) and also shown at the end of this response. It is not uncommon that multidimensional dispersion occurs in natural earth materials, because the spreading process is influenced by the complicated paths taken by fluid moving through a heterogeneous medium.
- (2) Although the unsaturated solute transport we concern in the manuscript is time-invariant, the fluctuations in concentrations are spatially nonstationary. The stochastic analysis of field-scale steady-state unsaturated solute transport process in heterogeneous media has been performed in a number of papers (e.g., Russo, 1993, 1998; Harter and Zhang, 1999) base on the Lagrangian approach.
- (3) The asymptotic unsaturated solute transport (steady-state unsaturated transport) occurs in regions that are at least several to tens of correlation lengths away from the source (Russo, 1996, 1998), indicating that the asymptotic transport relationship is applicable only after a substantial displacement distance. As such, the BCs could be ignored when solving the transport perturbation equation (e.g., Gelhar and Axness, 1983). A brief discussion regarding to this issue can be found on page 7, line 5 in the manuscript.
- (4) Under the steady state condition, the concentration distribution is spatially dependent, but temporally independent.

(5) As mentioned in the manuscript, the mechanism of the field scale spreading of a solute by a fluid in natural porous formations is dominated by the spatial heterogeneity of the formation properties. Figure 1 is used to demonstrate simply how the heterogeneity scale (the correlation scale of formation properties) influences the large-time field-scale dispersion. In fact, "according to previous studies (Russo, 1996, 1998), the spreading of the field-scale plume would therefore reach its large-time behavior as long as the lateral length scale which is used to characterize the size of the solute body is much larger than the scale of heterogeneity", which had been stated on page 7, line 17 in the manuscript. In other words, the proposed solution of madrospersion coefficient will prevail as long as the lateral length scale of the solute body is much larger than the scale of heterogeneity.

3. Furthermore, the effect of neglecting in the transport equation the term involving the spatial derivate of soil water content is unclear. In this case referring to existing papers is not enough. A more in depth analysis and justification of the assumption, specifically referred to the case at hand, is needed in my view.

Reply

As suggested, we will add the following statements on page 4 (Line 19) as

"Note that it has been mentioned in the works of Russo (1998) and Harter and Zhang (1999) that the field-scale dispersion at the large time is insensitive to variability in water content compared to the variability of $\ln K_s$. Harter and Zhang (1999) also mentioned that the preasymptotic macrodispersion is significantly larger in soils with variable water content than in those assuming constant content. The impact of spatial variability in water content is negligible for moist soils but it could be significant for very dry soils (Harter and Zhang, 1999)."

Conclusions

For these reasons I regret to conclude that in my view this manuscript falls short both as a technical note and a full manuscript. Much more work is needed to make it suitable as a full paper, while I think that this topic, as presented, is not suitable for a technical note.

Reply

Please see the replies to "General and Specific Comments". In conclusion,

- (1) We have modified a part of manuscript so that the readers have a clearer picture of problem we intend to solve and the task we plan to perform.
- (2) This note has presented the new development and advance in quantifying the large-time, field-scale dispersion in nonstaionary unsaturated velocity fields using Eulerian methodology. This has not been reported in the stochastic subsurface hydrology literature so far.
- (3) Our analytical expressions improve the understanding of the influence of heterogeneity scales of the formation properties on the field-scale unsaturated transport processes. The present results of this study are technical issues and should be of value to researchers and practitioners interested in the solute

transport in groundwater. In addition, the length of the text of this manuscript is rather short, it is appropriate to be a technical note.

I will be happy to change my opinion expressed above if the authors are able to address coherently the following points, which in my view are the main drawbacks of this manuscript: a) a discussion of the relevance of the type of unsteadiness they consider in this manuscript. Is it possible to make the approach more general in this respect? b) transport equation: can the steady state hypothesis be removed? If not I would be glad to see an in depth discussion on the implications of this hypothesis; c) transport equation: what is the effect on the solutions of neglecting the term containing the spatial derivative of soil moisture? d) I would be happy to see a coherent and well structured discussion of the impact of flow unsteadiness on macrodispersion (possibly including early time behavior).

Reply

- (a) The analysis presented in the manuscript is valid when the mean displacement is large as compared with the scale of aquifer heterogeneity. The unsteady-state unsaturated transport occurs in regions near the source of plume where a sharp concentration gradient exists.
- (b) "According to previous studies (Russo, 1996, 1998), the spreading of the field-scale plume would reach its large-time behavior as long as the lateral length scale which is used to characterize the size of the solute body is much larger than the scale of heterogeneity." (line 17, page 7 in the manuscript) The proposed result of madrospersion coefficient in the manuscript will prevail as long as the lateral length scale of the solute body is much larger than the scale of heterogeneity.
- (c) Please see our replies in "Specific comments 2 and 3".
- (d) It is expected that at each time step the scale of aquifer heterogeneity plays the similar role in influencing the field-scale spreading of plume to that in the case of the steady-state solute transport. The result for the large-time madrospersion developed in the manuscript is an upper bound for the spreading of plume at the field scale.

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Figure



Figure 5.6. Breakthrough curves at a distance of 10 m downstream from the center of the patch source, as calculated using the analytical solution of equation (5.16).