

We thank the reviewer for his/her thoughtful and detailed comments that definitely helped us clarify the manuscript and avoid misinterpretations.

The paper deals with an inverse modelling method determining simultaneously hydraulic and transport parameters from a packed soil column. Some of the questions posed are very useful for experimental work on flow and transport and will help future work to choose efficient experimental designs to obtain parameters. Overall the paper focusses on the methodological aspects without posing a clear hypothesis. With no clear hypothesis formulated, I would expect to have a stronger statement on the benefits of the methods employed and what we should be learning from this (not just stating that the methods used in the paper are superior over the methods other researchers have used).

The modeling concepts were clearly stated in the introduction (L57-L61 of the submitted manuscript). The introduction has been improved and the different assumptions are described.

We did not claim that our methods are superior to methods used previously. We analyze the accuracy of some existing methods and we suggest an alternative one which avoids intrusive measurements of pressure and/or water content. We show that this new method provides quite good estimates of the parameters but, of course, not with the same accuracy than methods with intrusive measurements.

Even if we come up with better parameter estimation, do we have a better understanding of the physics of fluid flow in porous media? The authors should be stating what novel insights they expect from this type of numerical experiments. Furthermore, some of the findings are to be expected, for example the inclusion of both water content or outflow along with matric potential data should always provide better parameter estimation. In fact, the use of only one of those variables makes parameter estimation non-unique.

Parameter estimation through inverse modelling has a weak point: the assumption that the model is valid. Therefore, it will not provide a better understanding of the physics. It can sometimes be used to reject a model if the estimated parameters have no physical meanings.

We agree that some findings are expected. The MCMC approach allows some quantification of the uncertainties.

An interesting aspect of their work is the impact of the length of the injection of the solute pulse. Can the authors provide some kind of explanation why this occurs?

We will provide the following explanations in the discussion.

The improvement of the parameter estimation in this last scenario compared to the previous one can be explained by the fact that the injection of water and solute contaminant is stopped once the concentration reaches the column outlet. Hence, the injected volume ($0.015 \times 3000 = 45 \text{ cm}^3/\text{cm}^2$) is slightly less than the pore volume ($120 \times 0.43 = 51 \text{ cm}^3/\text{cm}^2$). Thus, when the injection is stopped, the

column is not fully saturated and the outlet flux strongly reduces (see the asymptotic behavior of the cumulative outflow when the injection is stopped). As a consequence, the concentration profile increases smoothly (see Fig. 6) until reaching its maximum value in contrast to the sharp front observed for $T_{inj} = 5000\text{min}$ in the scenario 6 (see Fig. 5). As a consequence, the breakthrough curve obtained with $T_{inj} = 3000\text{min}$ is more affected by the hydraulic parameters than the breakthrough curve obtained with $T_{inj} = 5000\text{min}$. This explains why a better estimation of the parameters is observed for the last scenario compared to the scenario 6.

Considering how fractional derivatives and continuous time random walk have been used to describe solute transport in unsaturated soil, will the parameter estimation method give hints on systematic model errors (which require real world experiments). Certainly one short coming of the approach - it is assumed that the model is indeed correct.

The modeling concepts are assumed to be valid. See our answer to your second comment.

To make this paper a value contribution I suggest the following:

(i) Include a clearer summary of what has been done on inverse modelling in the context of transient water flow and solute transport. Perhaps state the methods more explicitly that were used by other researchers.

The introduction will be rewritten with a significant number of new references as follows:

The soil parameters that influence water flow and contaminant transport in unsaturated zones are not generally known a priori and have to be estimated by fitting model responses to observed data. The unsaturated soil hydraulic parameters can be (more or less accurately) estimated from dynamic flow experiments (e.g., Hopmans et al., 2002; Vrugt et al., 2003a; Durner and Iden, 2011; Younes et al., 2013). Several authors have investigated different types of transient experiments and boundary conditions suited for a reliable estimation of soil hydraulic properties (e.g. van Dam et al., 1994; Simunek and van Genuchten, 1997; Inoue et al, 1998; Durner et al, 1999). Soil hydraulic properties are often estimated using inversion of one-step (Kool et al., 1985; van Dam et al., 1992) or multistep (Eching et al., 1994; van Dam et al., 1994) outflow experiments or controlled infiltration experiments (Hudson et al., 1996).

Kool et al. (1985) and Kool and Parker (1988) suggested that the transient experiments should cover a wide range in water contents to obtain a reliable estimation of the parameters. Van Dam et al. (1994) have shown that more reliable parameter estimates are obtained by increasing the pneumatic pressure in several steps instead of a single step. The multistep outflow experiments are the most popular laboratory methods (e.g., Eching and Hopmans, 1993; Eching et al., 1994; van Dam et al., 1994; Hopmans et al., 2002). However, their application is limited by expensive measurement equipment (Nasta et al., 2011).

Infiltration experiments have been investigated by Mishra and Parker (1989) to study the reliability of hydraulic and transport estimated parameters for a soil column of 200 cm using measurements of water content, concentration and water pressure inside the column. They showed that the simultaneous estimation of hydraulic and transport properties yields to smaller estimation errors for model parameters than the sequential inversion of hydraulic properties from the water content and/or pressure head followed by the inversion of transport properties from concentration data (Mishra and Parker, 1989).

Inoue et al. (2000) performed infiltration experiments using a soil column of 30 cm. Pressure head and solute concentration were measured at different locations. A constant infiltration rate was applied to the soil surface and a balance was used to measure the cumulative outflow. They showed that both hydraulic and transport parameters can be assessed by the combination of flow and transport experiments.

Furthermore, infiltration experiments were often conducted in lysimeters for pesticide leaching studies. Indeed, lysimeter experiments are generally used to assess the leaching risks of pesticides using soil columns of around 1.2 m depth which is the standard scale for these types of experiments (Mertens et al, 2009; Kahl et al., 2015). Before performing the column leaching experiment, several infiltration-outflow experiments are often realized to estimate the soil hydraulic parameters (Kahl et al., 2015; Dusek et al, 2015).

The key objective of this study is to evaluate the reliability of different experimental protocols for estimating hydraulic and transport parameters and their associated uncertainties for column experiments. We consider the flow and the transport of an inert solute injected into a hypothetical column filled with a homogeneous sandy clay loam soil. We assume that flow can be modelled by the Richards' equation (RE) and that the solute transport can be simulated by the classical advection-dispersion model. Furthermore, the Mualem and van Genuchten (MvG) models ([Mualem 1976](#), [van Genuchten 1980](#)) are chosen to describe the retention curve and to relate the hydraulic conductivity of the unsaturated soil to the water content. The estimation of the flow and transport parameters through flow-transport model inversion is investigated for two injection periods of the solute and different data measurement scenarios.

Inverse modelling is often performed using local search algorithms such as the Levenberg-Marquardt algorithm (Marquardt, 1963). Besides, the degree of uncertainty in the estimated parameters, expressed by their confidence intervals, is often calculated using a first-order approximation of the model near its minimum (Carrera and Neuman, 1986, Kool and parker, 1988). However, as stated by Vrugt and Bouten (2002), parameter interdependence and model nonlinearity occurring in hydrologic models violate the use of this first approximation to obtain accurate confidence intervals of each parameter. Therefore, in this work, the estimation of hydraulic and transport parameters is performed in a Bayesian framework using the Markov Chain Monte Carlo (MCMC) sampler (Vrugt and Bouten, 2002; Vrugt et al., 2008). Unlike classical parameter optimization algorithms, the MCMC approach provides parameter joint probability distributions, which are useful to assess the quality of the estimation. The MCMC samples can be used to summarize parameter uncertainties and to perform predictive uncertainty (Ades and Lu, 2003).

Hypothetical infiltration experiments are considered for a column of 120 cm depth, initially under hydrostatic conditions, free of solute and filled with a homogeneous sandy clay loam soil. Continuous

flow and solute injection are performed during a time period T_{inj} at the top of the column and with a zero pressure head at the bottom. The unknown parameters for the water flow are the hydraulic parameters: k_s [$L T^{-1}$], the saturated hydraulic conductivity; θ_s [$L^3 L^{-3}$], the saturated water content; θ_r [$L^3 L^{-3}$], the residual water content; and α [L^{-1}] and n [–], the MvG shape parameters. The only unknown parameter of the tracer transport is the longitudinal dispersivity, α_L [L].

Several scenarios corresponding to different sets of measurements are investigated to address the following questions:

- 1) Can we obtain an appropriate estimation of all flow and transport parameters from tracer-infiltration experiments, even though a limited range in water content is covered (only moderately dry conditions are used)?
- 2) What is the optimal set of measurements for the estimation of all the parameters? Can we use only non-intrusive measurements (cumulative outflow and concentration breakthrough curve) or are intrusive measurements such as the measurements pressure heads and/or water contents inside the column unavoidable,?
- 3) Similar to multistep outflow experiments, is there an optimal design for the tracer injection?

(ii) The methods sections need more precise description of numerical methods used and experimental set up. I doubt this paper is reproducible with the information provided. The language is used in such a way, that true experiments were actually done. When the authors talk about experiments they mean virtual numerical experiments. This needs to be clearly stated earlier in the paper.

The experiments are numerical experiments. This was clearly stated in the introduction (L93 of the submitted manuscript). Although we think that numerical methods for solving the flow and transport equations have to be improved, we did not address this issue here. The domain is 1D which does not require heavy computational equipment and standard numerical methods are accurate enough. Standard finite differences have been used for solving the equation.

All required data, initial and boundary conditions are described in the paper. The simulations can be reproduced.

(iii) The discussion section needs a thorough revision to address the above points – clearly relate your findings to the work of others on parameter estimation. Currently the discussion focusses only on own findings without setting a broader context.

The broader context has been described in the new introduction.

Further comments:

Lines 74-75: When stating column length, column diameter should also be mentioned if real world experiments were used.

The diameter is not a relevant characteristic for our numerical examples since we use 1D simulations.

Lines 83-92: The research questions are not logical derived from previous they were certainly retrospectively formulated based on the findings of study.

We agree and reformulate the questions in the new introduction.

Line 113: There is an issue with the van Genuchten - Mualem model near saturation (hydraulic conductivity will decrease before air entry point as been reached)- will this affect parameter estimation.

We agree. However, this effect is not taken into account in this work. An extension of our work on different kind of model (Brooks and Corey, Modified Van Genuchten) is a perspective of this work.

Lines 132-139: Be precise on what was exactly implemented. The numerical scheme should be exactly described (appendix or supplemental materials are sufficient for this purpose.

We used very standard 1D finite difference for spatial discretization. Because the method is very popular, we do not think it requires a detailed description. Details on the use of the MOL for solving RE are well described in Fahs et al. (2009). This point will be specified in the revised version.