

***Interactive comment on “A simple two layer model for simulation of adsorbing and nonadsorbing solute transport through field soils” by M. S. Akhtar et al.***

**Anonymous Referee #1**

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This paper presents a modification of a simple model that was developed to describe preferential flow. Breakthrough experiments were carried out at various flow rates in undisturbed soil columns that were taken from soils with a different structure. The breakthrough of Cl and Li were monitored in the effluent. Both the simple preferential flow model and a two region CDE model were fitted to the breakthrough curves. I think that a valuable dataset of transport experiments in undisturbed soil columns has been collected. However, I have serious questions about the paper. First, the paper requires a thorough check of the language. I have the impression that this paper was not carefully read by all co-authors before it was submitted. I would like to point at the

responsibility of all authors for the quality of the submitted paper. Second, I have questions about the method that was used to estimate the model parameters of the simple preferential flow model. It seems that these parameters were estimated by manual calibration. I think that this is not acceptable and that a more sophisticated inverse parameter estimation procedure, which also allows estimating the confidence intervals of the parameters, should be used. Third, I question the validity of the mixing layer concept and the a-priori fixing of the mixing layer depth. In the mixing layer, it is assumed that the incoming solution is instantaneously mixed with the water volume in the mixing layer and instantaneously sorbed to the soil matrix. Do the dye tracer experiments confirm that this mixing occurs? The mixing layer model also makes strong assumptions about the concentration profiles within the soil. Do the authors have evidence that these assumptions hold true? The fact that the  $K_d$  coefficient depends on the flow rate is a strong indication that there is no instantaneous sorption. I doubt strongly whether the mixing layer concept is a valid concept to describe preferential transport in soils. A good fit of the mass balance is not a sufficient argument that this concept is valid.

Detailed comments: The abstract does not contain sufficient information and can not be understood without having read the paper. Abstract: 'A modification of an existing preferential flow model . . .' In the abstract, the authors should write with modification they made to which model. Abstract: In 6: Flow rates were generated. . . From reading the abstract, it is not clear what which preferential flow model was used. The authors write: 'The existing simple preferential flow equation. . .'. It is trivial that the equation must 'exist' if one wants to fit it.

Introduction: Rapid movement of solutes to tile drains is in fact important for rapid movement of contaminants to surface water. p5633: In 27: Why 'on the other hand'? p5634: In 9: It is not clear to me which type of mixing is referred to here. Do the authors mean mixing in the top mixing layer? There could also be a mixing between the preferential flow and matrix region. I suppose that this is not the type of mixing the authors refer to here. I do not understand what is meant by simultaneous occurrence

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of mixing and leaching and why it would fail to predict transport p5634 In 13-16: I do not understand this sentence. This needs to be reformulated.

p5634-5635: Eq. 1 gives the CDE for transport and sorption in only one domain.

p 5635: In 13-14: This sentence needs to be reformulated.

p 5635: What is meant by: 'sorption and advection processes that balance'? Do the authors mean that sorption and desorption processes are always fast enough so that an equilibrium between sorbed and dissolved concentration can be assumed?

p 5635: In 16. There is a mixing layer, a conveyance layer and now a 'transport layer' pops up? What is this? What is a 'time lag to advective transport' I do not know how I have to understand this. Is this the retardation factor? Or is there a constant lag time that is added to the advective transport time? If so, why would it make sense to add a constant lag time?

P 5635: Eq. 3 is the solution of a mass balance equation in the mixing zone. It would be useful to write down this mass balance equation.

p 5638: I do not understand the function or role of Eq. 12.

p 5639: An increase in the depth of the mixing layer over #R0 results in a decrease of THE slope of leaching phase (#01, #02, #03) due to more mass stored.

p 5639 and table 1: I do not understand why the authors kept the concentration in the mixing layer at the start of flushing constant for the scenarios with different depths of the mixing layer. In this way, the amount of mass that is added to the system depends on the depth of the mixing layer. As a consequence, the amount of mass that is added depends on the thickness of the mixing layer. The same holds for the scenarios with different water contents. I think it would be better to apply the same amount of mass in all scenarios and demonstrate how the system reacts when the parameters are changed. As far as I understand the model, the size of the mixing reservoir is decisive as it determines how strongly the input concentration is reduced by mixing in

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this reservoir. The stronger the input concentration is reduced, the smaller the mass flux that leaches out of the mixing reservoir. The mixing reservoir can be increased by increasing the water content, the thickness and the sorption capacity. p 5640, ln9-10 shouldn't the equations be fitted to the measured concentrations in stead of the the concentrations fitted to the equations?

p 5640: A reference to the soil classification system that is used should be given. The World Reference Base soil classification system should be used. More information about the soils must be given in a table: % of sand, silt and clay of the different layers, % of organic matter content, dry bulk soil density. It should also be specified which pH was measured: the pH of a soil-water suspension or the pH of a soil –CaCl<sub>2</sub> solution suspension. Finally, it is important to mention the land use of the plots from which the soil columns were taken: arable land, grass land, forest? Especially the structure of the top soil layer depends strongly on the land use. '

p 5641: 'Chloride concentration ratio in the percolate AND INPUT SOLUTION approaching one...'

p 5641: Column preparation and leaching experiments. A constant pressure was applied during an experiment. Was the water flux constant with time or did it fluctuate, decrease with time?

p 5641: No information about the water content in the columns and for the different flow rates is given. This is however an important parameter for transport. Couldn't

p 5642: The authors write here that they fitted the two-region CDE model. Therefore, they should present the two region CDE equations in the modelling approaches part instead of the the equilibrium CDE equation.

p 5642: 'The boundary conditions included characteristic length of 0.48 m and total input pulse equal to the drainage depth for application phase and the initial condition that constant concentration of Chloride (equal to concentration of tap water) leaches

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through the column.' This needs to be reformulated. First, the length of the column is not a boundary condition. I have no clue what is meant with 'total input pulse equal to the drainage depth for application phase' I think the authors mean the infiltration depth rather than the drainage depth?

p 5642: '...the first pulse had the input concentration of 1 for the duration (drainage) of application phase and the second pulse had the input concentration equal to Chloride in tap water.' Reformulate. What is an 'input concentration of 1'? What are the units? Give also the concentration in the tap water.

p 5643: 'An analytical solution was accepted once the highest goodness of fit ( $r^2$ ) and dispersivity ( $D/v$ )  $2 \text{ cm} < D/v < 5 \text{ 20 cm}$ , mean square error for model  $< 0.01$ , and T-value on parameters  $> 1$ .' This needs to be reformulated.

p 5643: In 5 'The predicted parameters for Chloride..' First, the parameters were not predicted but fitted, second the parameters do not describe a specific property of Chloride but rather characterize the chloride transport.

p 5643: 'Assuming the  $v_m$  for Cl' represents the pore water velocity and given the drainage as a proxy for time, inverse of  $v_m$  represents the effective porosity  $\theta_{\text{eff}}$  or quantum of preferential flow. Mobile water fraction is then  $\theta_{\text{eff}} = \beta \times 1/v_m$  (Cl).' This sentence needs to be reformulated. The calculation of  $\theta_{\text{eff}}$  from  $\beta$  and  $v_m$  is to my opinion wrong.  $\theta_{\text{eff}}$  should be equal to  $\theta_{\text{eff}}$ , which is defined here as  $1/v_m$ .

p 5643: 'Retardation of Lithium was predicted from the pore water velocity.' This is an incorrect formulation. The retardation factor does not depend on  $v$  but it depends on the bulk density, the water content and the  $K_d$  coefficient. As a consequence,  $R$  cannot be predicted from  $v$ . I think that the authors mean that they fitted the equation to the lithium concentrations assuming  $R=1$  and derived  $R$  from the ratio of the fitted  $v$  to the Cl breakthrough and the fitted  $v$  to the Li breakthrough.

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p 5643: 'Chloride and Lithium breakthrough data were fitted to simple and modified preferential flow models, i.e., Eqs. (3) and (14), respectively.' Again, you do not fit data to a model, you fit a model to data.

p 5643: 'Chloride and Lithium breakthrough data were fitted to simple and modified preferential flow models, i.e., Eqs. (3) and (14), respectively.' I do not understand why Eq. 3 should be fitted to breakthrough data. Eq. 3 describes the concentrations in the mixing zone. Unless it is assumed that the concentrations in the effluent are instantaneously equal to the concentrations in the mixing zone, this makes sense. First, I doubt whether this assumption makes sense. Second if this assumption has been made in the past, it would be necessary to mention it in the text.

p 5643: 'In a spreadsheet, the Eq. (14) was superimposed on the scatter plot of  $\ln(1-L/M_0)$  versus  $Y$  for Chloride adjusting the analytical solution to overlay by repetitively substituting values for  $W_a$ ,  $W_d$  and  $W_2$  coefficients until the highest correlation coefficient  $r^2$  was achieved.' If I understand this correctly, the authors just used a 'manual' calibration by tuning the parameters. For inversion problems with more than 1 parameter, I think that this calibration procedure is not appropriate since a lot of parameter combinations should be tried out. The authors should definitely use more sophisticated inverse parameter estimation procedures. They should also give the confidence intervals of the estimated parameters.

p 5643: 'The depths of the mixing and distribution layers ( $d_1$  and  $d_2$ ) were the observed horizons for each soil.' This is a rather strong assumption, especially since the depth of the mixing layer is a very crucial parameter. Is there any evidence that the depth of the mixing layer corresponds with the depth of the top horizon? This assumption must be clearly mentioned in the text. Secondly, evidence must be given that this assumption makes sense. Third, the assumption must be confronted with estimates of mixing depths that are used in other model approaches. For instance, in MACRO, a fixed depth of the mixing layer is assumed and the depth of this mixing layer is only a few mm, which is much thinner than what the authors assume in their work. Finally, the

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authors should include a table in which the depths of the soil horizons of the different soil cores are included.

p 5643: 'Concentration  $C_1(T_0)$  and flow rate,  $q$  were known for each leaching experiment.' I do not understand this. The concentration  $C_1$  is the concentration in the mixing layer. This concentration wasn't measured, was it? How could it be known then? it must have been calculated by the model.

p 5643: 'and bulk density,  $\rho_{\text{ob}}$  to  $1.32 \text{Mgm}^{-3}$ .' I can hardly believe that the bulk density was the same for all soil types.

p 5643: 'Specific flow rate  $q$  does not affect since the application time was determined by ratio  $C_1/C_0$ .' Again, how can the application rate be determined by  $C_1/C_0$ ?  $C_1$  is not measured. I am completely lost.

p 5644: 'More detailed description of breakthrough and transport parameters by fitting two-domain CDE was separated previously (Akhtar et al., 2009).' This sentence does not make sense.

p 5644: 'rise in concentration with cumulative drainage was uniform' What is a 'uniform rise'?

p 5645: 'The initial drainage depth for which the data plotted nonlinear reflect interaction with the matrix.' I think this is not enough precise. It should be interaction with the pore water and sorption sites (for Li) in the transport layer.

p 5646: 'Equation (14) did not have assumption of instantaneous mixing and it fitted well in almost all cases.' I think this is not correct! Eq. 14 also assumes instantaneous mixing in the mixing layer. The difference is that Eq. 14 does not assume instantaneous transfer through the transport layer.

p 5646: In table 2, also the depths of the mixing and transport layers should be given.

p5648: 'Nevertheless, installation of a tensiometer in column could have indicated

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in vitro water potential provided contact and sufficient equilibrium time are insured (Hutchison et al., 2003).’ I do not understand what is meant by this sentence.

p 5650: If the concept of the mixing layer would be a valid concept for transport, then there should be a surface layer that is completely stained by the dye. Was such a layer observed? If not, then the whole concept is invalid.

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