The authors in this paper discuss two approximate models for preferential transport and use the models to analyze a number of breakthrough curves from undisturbed soil columns. The study is an extension of an earlier paper published several years ago in *Vadose Zone Journal* (Akhtar et al., 2003), except for the use of a modified formulation for the preferential flow model, and the use of undisturbed cored from Germany rather than New York.

My impression of the paper is not favorable. The material is presented extremely poorly in terms of English language, and a lack of precision and clarity. This is a pity since the experimental data may well be useful for testing more appropriate physics-based preferential flow models. The paper certainly should not have been submitted in its present form. This was also brought up by the first reviewer. A thorough cleaning of the paper and being more critical about many statements may well produce a useful contribution. However, the paper in its present form is unacceptable. Below are some comments. I will not further comment on the writing style since this was done very well by Reviewer 1.

The discussion of Eq. (3) is confusing. The equation appears to be a solution of the differential equation

$$d_1 \frac{d\theta RC}{dt} = -qC_1 \tag{1A}$$

subject to the initial condition

$$C(0) = C_o \quad (t=0) \tag{2A}$$

This means that all of the solute is placed instantaneously in the mixing layer. The solution for the concentration is given by

$$C = C_o \exp(-\frac{qt}{d\theta R}) \tag{3A}$$

The total amont of mass (*M*) in the mixing layer is given by

$$M = Ad\theta RC \tag{4A}$$

which, with Eq. (3A), leads to

$$M = Ad\theta RC = Ad\theta RC_o \left(\frac{C}{C_o}\right) = M_o \exp(-\frac{qt}{d\theta R}) \tag{5A}$$

where *A* is the cross-ectonal area of the column. The amount of mass (*L*) leached from the mixing layer is given by $L=M_o-M$, which shows that Eq. (3) in the paper is correct. This equation also appears in the earlier VZJ paper. To have a better connection later with Eq. (1), and also with the two-region nonequilibrium CDE used by the authors, it would seem better to use Eq. (1A) above directly, or alternatively Eq. (4) in the paper, rather than such confusing terminology *W* ("apparent water content"?), among other terms. The above derivation also shows that *L* is the integral of *C*(*t*) over time, i.e.,

$$L(t) = \int_0^t AqC(\tau)d\tau \qquad (6A)$$

Going now to Eq. (5), the first part is correct, being very much like Eq. (3A) above but for increasing concentrations in the mixing layer. Unfortunately, I have some problems with Eq. (5). How exactly are Y and Y_o defined in this equation? I think the argument of the first exp function must be positive. This depends on also how Y_o is defined in the paper (I assumed as qt_o). Also, given the above derivation, I do not understand why there is a need to integrate $C_2(t)$. Are the concentrations then not given immediately by $L(t-\theta_m x_t/q)$, where x_t is the length of the transport layer. This would be the very similar as Eq. (3A) if the filling stage were to be neglected.

There is also a conceptual problem with how the effects of chemical hysteresis is incorporated in Eq. (5) of the paper. That equation will not preserve mass if different values of K_a and K_d are used. Chemical hysteresis cannot be modeled using two linear isotherms with different distribution coefficient for adsorption and desorption. To preserve mass, the isotherms must connect at the concentration where adsorption (or desorption) reverts to desorption (or adsorption). This means that at least one of the isotherms (likely the desorption isotherm) must be nonlinear (e.g., see van Genuchten et al., Soil Sci. Soc. Am. J., 38, p. 29-35, 1974). Unless one assumes that some solute will be lost from (or created instantaneously in) the system, which I don't think the authors want.

On page 5642 the authors discuss fitting the two-region nonequilibrium CDE to the data (and not fitting the data to the model as correctly indicated by Reviewer 1). The governing equations of this model should indeed be stated somewhere, probably best as part of the introduction after Eqs. (1) and (2). Equally important is stating exactly the initial and boundary conditions used for the solutions within STANMOD. Was solute assumed to be in the applied water over a time $0 < t \le t_o$? The nonequilibrium model has four unknown parameters. Was the retardation factor R for chloride equated to 1? Some of the soils may have exhibited anion exclusion (making R less than 1 for Cl), especially since a quasi-steady state flow regime was imposed. Why not list the fitted values of θ_m/θ, D_m and ω in a table? Once the transport parameters are determined from the Cl curve, why not using those for the Li also (except for R and β)? At a minimum one could use the same values for D_m and θ_m/θ. Placing all parameters for Li and Cl transport in a table may be very educational.

I also have some questions about how the term "Retardation Factor" is used in the paper. On the one hand, R is used in terms of its formal definition given by Eq. (2). That definition applies irrespective of the model being implemented. For example, the use of R in the equilibrium and nonequilibrium models reflects the final stage of sorption processes when $t \rightarrow \infty$. On the other hand, R in the paper is mostly used only as an adjustable empirical parameter to be fitted to data. This confusion is evident on page 5635, after Eq. (2), where the authors state that the transport velocity is R times lower and the arrival time R times longer as compared to a nonadsorbing solute. The problem here is that the paper does not differentiate between the actual processes that occur, and the processes as they are modeled in the governing equations. The retardation factor based on batch studies will always hold for field conditions, but may not be applicable when used in the standard CDE model (Eq. 1). It is not the retardation factor that should be questioned

(cf. page 5649, line15), but rather the model using the retardation factor (e.g., the one or two-region CDE's; or any other model that includes provisions for preferential flow.

As such, a comparison of R values has only limited value since for the two-region CDE model R is conceptually defined, while for the proposed preferential flow model it is a fitting parameter. It would have been interesting to provide the retardation factor based on batch sorption experiments and reasonably averaged values of the water contents of the various columns. As such I wondered why not any batch studies were carried out to determine the K_d's of Li? That would have been very useful, and would have provided some means for judging the results plotted in Fig. 8. The VZJ paper showed reasonably linear sorption isotherms, and hence could have produced good estimates of R for Li (at least for the soils in that study).

Pages 5641, 5648. Do I understand that no negative pressure was applied at the bottom of the columns, and hence that a seepage face existed there. In other words the pressure head would be zero once the soil becomes there saturated (using the awkward phrasing in the paper "that the matric potential may have been prevented due to the capillary fringes"). Hence the tracer solution is allowed to drip from the column by gravity? This indeed would mean that the water content will not be constant throughout a column. It seems that this mostly affect the larger pores in the top part of the columns, which will become unsaturated when a negative pressure is applied. All this suggest that the preferential flow processes are mostly dictated by how water enters the mixing zone at the top, not the transport layer. This actually is contrary to the idea that the solutes mixing evenly and instantaneously in the mixing layer.

Figure 3. Maintaining a lower negative pressure head at the column entrance should decrease the flow rate and eliminate some of the larger pores in the mixing layer from transport. Hence, one would expect less preferential flow when the pressure head decreases from +10 to -10 to -40 to -100 mm. Some of the plots in Fig. 3 do not show this trend. For example, Fig. 3c for Cl shows earlier breakthrough for the -40 mm experiment (black symbols) as compared to the -10 mm case (red symbols). The data also suggest a lack of mass balance (areas under the curves), although this is difficult to judge because of different degrees of tailing of the curves.

Why only plot results obtained with the original and modified preferential flow models in terms of semi-log plots. Why not compare the two-region CDE with the simplified preferential flow models using plots such as those shown in Fig. 3.

Page 5636 and Fig. 5. Increased differences between the Cl and Li breakthrough curves at the lower flow rates should reflect more interaction (by diffusion or advective mixing) between the preferential flow paths and the micropores (or soil matrix). The simplified model can only account for this by empirically adjusting the parameters for each BTC. This is also the case for the modified model. Plotting predicted concentrations versus time or cumulative amount of drainage likely will show far worse comparisons, especially for Lithium.

In conclusion, this paper raises more questions than providing answers. The data may potentially be useful for testing appropriate physics-based models (e.g., MACRO, HYDRUS-1D) for preferential flow, and how preferential flow parameters are affected by the imposed inflow boundary conditions. The models presented in the present paper are probably too approximate

for application to meaningful environmental pollution problems. This may well be a judgment to be made by readers of the journal. In the end, however, the terrible wring style and the many incorrect or dubious statements are reasons of why this paper should not be accepted for publication.