General Comments

In this research, an analytical model is derived to describe the transfer of solutes from the soil surface to water during overland flow events. Several phases of this transfer process are considered in the model: the initial condition (i.e., before ponding starts), the period from the ponding start to the start of surface runoff generation, and the period between the start of surface runoff and "steady state" runoff. This model has the potential to be applicable for a broad range of solutes, even though the authors suggest that it will be most useful in predicting solutes discharge by surface runoff in agricultural fields. Here, potassium chloride is chosen to conduct lab experiments to generate data by which to validate the model.

The general utility of the model presented in this research is of concern. In order for the surface runoff concentrations to be predicted, the initial saturated chemical concentration must be known. While this is easily accomplished in a laboratory setting, this is much more difficult to do at the field or basin-scale. Solute sources are spatially heterogeneous in most catchments – either as spatially correlated random variables or in some ordered fashion - though they may be more homogeneous in the agricultural setting that the authors appear to be targeting. Regardless, in order to understand the concentration dynamics at the catchment outlet, the degree of heterogeneity of solute sources within the landscape and their mobilization dynamics during runoff must first be understood in at least a conceptual sense in order to incorporate spatial variability into the model. As it is currently written, the model is applicable only to a single solute source (or soil mixing layer, as the authors call it). Furthermore, the value of C_0 will vary over time, as the solute undergoes biogeochemical processes between rainfall events. Currently the authors do not address a way to model the temporal variability of C_0. As it is unfeasible to assume real measurements will exist for pre-event conditions in the field as opposed to the laboratory, some additional work or references to other relevant published papers that address this must be included to improve the utility of the model. Otherwise, the model is only valid for estimating concentrations for a single storm event for which the initial conditions were known. Additionally, the explicit inclusion of a surface runoff trigger in the model would enable the equations presented in this work to be included in a general solute transport model in which transport via both infiltration and surface runoff are modeled, and the equations presented in this work would be used when the surface runoff trigger is exceeded.

The authors also neglect sorption processes in Equation 3. Most contaminants of interest, particularly in agricultural settings, have moderate to high partition (sorption) coefficients and therefore sorption effects must be explicitly included in the model. If sorption and/or complexation processes are neglected, then surface runoff concentrations will be greatly overestimated for phosphorus, pesticides, hormones, and other organic contaminants. This could be corrected for by accounting for total suspended solids (TSS) and including partition (sorption) coefficients (K_D). Neglecting sorption greatly limits the contaminants whose transport processes can be appropriately modeled using the equations presented in this work. Nitrate is perhaps a good candidate (and represented by chloride as the surrogate in this study) since it does not undergo sorption processes.

The description of the experimental conditions provides inadequate explanations as to why various conditions were chosen. How did the authors decide what the initial concentrations of KCl should be in the experimental setup? What is the significance of the soil types chosen for the

experiments? The paper's introduction sets the stage for the work to be primarily applicable to surface runoff in agricultural fields, but the types of soils chosen for the experiments are not typical of soils found in the Midwestern US, which are dominated by silt loams. Discussion regarding the selection of these soil types needs to be included so that readers can better understand the experimental design. Also, the number of times the experiments were run appears to be arbitrary. Why were 3 experiments conducted for the fine loamy soil and 7 experiments conducted for the sandy soil? Furthermore, the authors appear to neglect the dissolution of KCl, as they claim that the solute concentrations they are measuring are KCl (p. 3909 line 10). Because KCl is highly soluble in water (pKa~7), it seems unlikely that the compound was being measured in the experiment. If the solute concentration being measured is stated incorrectly, then clarification is necessary in the text and throughout the table and figures.

Additionally, the authors state that measured errors are not accounted for, but given the high-resolution at which the authors attempt to prediction concentration changes (as shown in the bottom part of Figure 3), errors should be included. At minimum, errors in the analytical methodology used to measure the aqueous concentrations should be discussed (i.e., what is the accuracy of the instrument and analytical method?). Is the level of accuracy obtained by the analytical method is greater than the small changes in concentration that the authors are claiming to predict accurately? Model errors also need to be addressed. What are the errors associated with the estimates of the two model parameters, α and γ? Without this information about measured and modeling errors, the validation approach is questionable.

The results and discussion section provides a very detailed, but hard-to-follow, summary of the experiments. The main points get lost in the details and need to be emphasized. Discussion regarding the implications of the main conclusions would help to strengthen this section. The results should be put into the context of field and basin-scale solute transport dynamics so that it is clear to readers that the authors have an understanding of how their work is applicable outside of the laboratory. Currently, it is unclear how the equations and methods presented can be used outside of the laboratory because the methods used to validate the model and assess the values of the two parameters, α and γ, require that data be collected before the temporal variability of these parameters can be determined. Therefore, the authors need to develop the non-stationarity of these parameters from a theoretical basis in order for the results obtained from these experiments to be applicable outside of the specific conditions used in the experimental design.

Technical Comments
The number of grammar errors is quite large. However, given the significant amount of re-writing and revising the authors will need to do before this paper can be published, these errors are of minor concern until after the major issues with the work are adequately addressed.