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***Interactive comment on* “Modeling the impact of soil aggregate size on selenium immobilization” by M. F. Kausch and C. E. Pallud**

Anonymous Referee #2

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The authors developed a reactive transport model for describing Se(VI) reduction and retention in flow-through reactor experiments with a single artificial aggregate. They use the parameterized model to predict the effect of aggregate size on Se retention in porous media under oxic and anoxic conditions. In my opinion, this is an excellent manuscript and I enjoyed reading it. In particular, I am very enthusiastic about the experimental design and the obtained results. However, the experimental results have already been published elsewhere and the modeling approach is not innovative neither. Pallud et al. (2010a) have developed a similar reactive transport model for describing microbially induced iron mineral transformations in an identical experimental design. Consequently, the manuscript does not provide substantial new data, methods, or concepts. The evaluation of the effect of particle size on Se retention in soils is an interesting application of the parameterized model and is an innovative aspect. How-

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ever, I disagree with the conclusions the authors draw regarding the effect of particle size on Se retention in soils.

My concerns about the interpretation of the particle size effect are not severe. If the authors agree, my suggestions can be easily implemented. However, it remains questionable whether a manuscript with very limited innovative content deserves publication. I expect that many readers will read the manuscript with the same interest and pleasure as I did. It is an excellent example for the use of reactive transport modeling to interpret experimental data and to test the plausibility of hypothesis and concepts. In many studies, the importance of small scale redox heterogeneities in soils for the bioavailability and mobility of redox sensitive contaminants is emphasized but clear evidence for this statement is rare. This experimental study is a very good and illustrative example how aggregation and redox zonation in soils can control the fate and transport of redox sensitive compounds. The developed reactive transport model nicely complements the experimental data as it substantiates the interpretation of the experimental findings and helps to comprehend the underlying processes. In my opinion, these strengths of the manuscript overcompensate the lack of innovation. Hence, I recommend to accept the manuscript for publication provided that the authors revise the discussion on the aggregate size effect on Se retention.

Major Comments

I agree to the discussion on the effect of particle size on reduced selenium concentration in the aggregates but I disagree with the extrapolation of these results to natural environments. The positive effect of increasing particle size on Se retention in soils with oxic conditions in the macropores might be generally valid. That is, larger aggregates are required for the formation of anoxic microenvironments which are the prerequisite for microbial Se(VI) reduction. In my opinion, this trend does not necessarily hold when the macropores are anoxic. Based on the model calculations, the differences in Se(0) concentrations between aggregates of different size is caused by differences in diffusive loss of Se(IV). However, the diffusive flux of Se(IV) out of the aggregate is

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overemphasized in one aggregate systems compared to porous media. In a porous medium with many aggregates, the concentration gradients are expected to be much shallower because Se(IV) concentrations increase along the convective flow lines in the macropores. In this case, Se retention can be higher in soils with small aggregates, in particular when diffusive supply of electron donor and Se(VI) is limiting Se(VI) reduction rates. Furthermore, the authors should highlight that the obtained trends are only valid when Se(VI) is added in large excess and diffusive flux of Se(VI) into the aggregate is not limiting the overall reduction rates. This also holds for the provision pathway of electron donor. In the experimental study, the electron donor is provided via the inflow solution. In natural environments, the electron donor used for Se(VI) reduction might be generated from the organic matter inside the aggregates. I recommend that the authors revise section 4.3 by paying more attention to the limitations of extrapolating the calculated trends for the one aggregate systems to natural soils.

Minor Comments

P 12054 line 6: I assume that selenate and selenite principally can adsorb onto quartz surfaces by a ligand exchange mechanism as onto other metal oxides surfaces although I do not know a study on Se oxyanion adsorption onto quartz. However, I would recommend to modify the statement: Pure quartz sand was chosen as adsorption of selenium oxyanions is negligible under the experimental conditions thus...

P 12054 line 10: rate instead of rates

P 12055 section 2.4: Why did the authors not measure selenate concentrations? Did the authors verified the selenate concentrations in the inflow solutions? With the given data, the Se mass balance cannot be closed. That is a pity as the mass balance could have been very useful to constrain the parameterization of the model. I presume that Se(VI) concentrations were not measured and / or reported because only a very small fraction of the Se in the inflow was reduced in the reactor. In my opinion, this fact should be explicitly be mentioned in the results. Furthermore it should be motivated in

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the method section why Se(VI) concentrations were not monitored.

P 12056 line 16 replace 'cycling' with 'consumption or production' or 'rates of...'

P 12057 line 3 I would appreciate if the fitting of the data from Losi and Frankenberger was reported in the supplementary material section. Is it possible to produce figures with the data and the fitted model?

P 12057 line 15: simply refer to Appendix A. Appendix A is appended to the manuscript and not part of the supplementary material.

P 12059 line 18: As far as I understand, the same data as presented in Kausch et al. (2012) were used in this study. How is it possible that reduced Se concentrations did not vary significantly between different input solutions in Kausch et al. (2012) but a dependency on pyruvate concentrations was found here. Was the statistical analysis in Kausch et al. (2012) incorrect? Please clarify.

P 12063 line 26: add comma after conditions

P 12070 line 5: add 'are' after conditions

P 12073 line 9: change table S1 into table A1

I am not an expert in hydrodynamics and hence I did not assess the validity of the hydrodynamic model as described in the appendix.

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