

Interactive comment on “The role of the retention coefficient for the scavenging and redistribution of highly soluble trace gases by deep convective cloud systems: model sensitivity studies” by M. Salzmann et al.

Anonymous Referee #4

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General Comments

The model studies presented in this paper focus on the role of the retention coefficient for the scavenging of soluble tracers in convective clouds. Results from a number of simulations with a cloud resolving model are presented. The analysis of these results emphasizes differences in tracer and hydrometeor mixing ratio profiles for different model configurations, meteorological situations, and initiation of convection.

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It appears that a substantial effort was invested in this study as is evident from the number of simulations and analysis results. Some of the results are interesting. However, the scientific methods and assumptions do not appear to be appropriate for a study on the general role of the retention coefficient in convective clouds. The conclusions of the study are based on very specific model assumptions. The approach lacks realistic parameterizations and emissions for tracer processes outside convective clouds. Comparisons for the effects of different convective initiation processes (i.e. large scale forcing vs. bubble) are ambiguous because they are based on arbitrary choices for the initiation of convection and tracers, model domain, and analysis times. It is unlikely that the conclusions of this study would generally apply to tracers in the real atmosphere or other meteorological situations or models. In addition, the main conclusions appear to be based on an unrealistic simulation for the ARM LSF case. Results for this simulation do not appear to be consistent with the results from other cloud resolving models or observations for this case.

Specific Comments

The paper focuses on interactions between microphysical and chemical processes. However, the description of the microphysics scheme is not detailed enough in the paper. The authors should include an overview of the microphysical processes that are included in the model.

The description of the experiments in section 3 is confusing. The authors first introduce the concept of "large scale forcing" in this section. However, they subsequently summarize experiments that apply this concept (i.e. TOGA COARE, ARM A) and another experiment which does not use this concept (i.e. STERAO) in a single paragraph in the same section.

Page 10781: The information on the vertical resolution in the ARM and STERAO sim-

ulations is insufficient. This is particularly important because the authors specifically mention on page 10782 that some of the simulation results are sensitive to the resolution (without giving further details).

Same page: "In the multi-day runs, the tracer fields are reset to their (horizontally inhomogeneous) initial values every 24 h...". The rationale for this approach is unclear and a motivation for this approach should be presented. Presumably, the idea is to compensate for the lack of other tracer processes in the simulation (e.g. large scale forcing for tracers, chemical sources/sinks, emission, dry deposition, etc.) in order to ensure that the simulated profiles are qualitatively similar to the initial profiles over the course of the entire simulation. This is of course based on the assumption that the initial profiles are physically meaningful, i.e. that a combination of processes may exist that would be consistent with the initial profiles (which is not clear either). However, the re-initialization of the profiles is very artificial. There is no corresponding process in the real world and adjustments of the tracer fields after that strong instantaneous perturbation are almost certainly non-trivial. The results can be expected to depend on the time interval for the re-initialization of the tracer profiles. Depending on the lifetimes of the individual tracers in the simulation (e.g. from different retention coefficients), shorter or longer time intervals may be required to equally constrain different tracers by re-initialization.

Same page: The simulation periods for the bubble experiments are much shorter (2.5h) than the simulation periods for the other experiments which employ large-scale forcings (> 12 hours). Therefore, it is not obvious how to compare results from these different simulations. The different simulation and diagnostic time periods in the simulations cause qualitatively very different results as is evident from Figure 4, for example.

Same page: The Henry's law coefficient of the soluble tracers are set to 10^6 mol/l/atm. Is there a specific reason why this number has been selected? Is the idea to mimic the behaviour of a specific class of chemical tracers? If so, this should be explained.

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Page 10783: The authors note that the warm bubble in the ARM BUB run produces a relatively short lived single cell storm. Obviously, the lifetime of the storm will depend on the time of insertion of the bubble and the thermodynamic properties of the bubble. Even a relatively large perturbation may cause only weak (or none) convective activity under sufficiently stable conditions while the opposite may be true at other times. The dependency of the results on the magnitude and timing of bubble insertion is highly ambiguous and limits the usefulness of this approach in studies of tracer processes in convective clouds. In particular, a fair comparison between results from this approach with results from the large-scale forcing simulations seems to be impossible.

Page 10784, Fig. 4: This figure is confusing. Results for TOGA COARE and ARM A LSF are for 12 hours after initialization of tracer profiles and results for ARM A BUB and STERAO are for 2.5 hours after initialization. Consequently, the changes in concentration profiles in the latter simulations are much smaller than in the former ones. Furthermore, results are averaged over different regions of the simulation domain. Based on these differences and because of the ambiguous bubble insertion (see comments in previous paragraphs), an interpretation of the differences in the results is highly problematic.

Same page: "...Barth et al. (2001) have suggested that global model such as the one used by Crutzen and Lawrence (2000) may underestimate the transport of highly soluble tracers to the upper troposphere." The authors should explain Barth et al.'s hypothesis and why there may be a problem with Crutzen and Lawrence's approach. It is not clear from the description in the paper.

Same page, same paragraph: It is suggested that the use of different initial/boundary conditions in different studies can cause different efficiencies of convective tracer transport. This seems to be a rather trivial statement. There are potentially many other reasons why different studies could produce different convective tracer transports. For example, differences in washout and rainout efficiencies from differences in precipitation rates could also produce large differences in profiles of water soluble tracers.

The argumentation in this paragraph is weak and additional comparisons appear to be necessary if the purpose is indeed to study potential differences between different simulations and situations.

Page 10785, Section 5: Figure 6: Profiles of averaged simulated hydrometeor mixing ratios for ARM do not agree well with previously published results for this case for simulations with other CRM's (Xu et al., 2002). In particular, none of the CRM's in Xu et al.'s study produced any large amounts of cloud liquid water below ca. 2km. However, the simulation in the current study produces a local maximum at about this level. In addition, the simulation produces a spurious maximum in the liquid water content near the surface. Further, the graupel mixing ratio is considerably higher than in other models. These differences are worrisome since they may indicate some problem with the simulation. In particular, fog has not been observed during the ARM IOP to my knowledge.

Pages 10785, 10786: The authors interpret high cloud water liquid water mixing ratios in the convective sub-cloud layer in the ARM A LSF simulation as arcus clouds and conclude that these clouds have an important impact on the tracer transport because of the apparently efficient tracer removal in this layer. The authors further note that a similar layer is absent in the STERAO simulation and explain less efficient removal of the non-retained tracer in that simulation by the absence of that layer for STERAO. However, if low clouds are indeed important for the removal of tracers as the authors claim (which is not obvious at all from the results that are shown), one might expect the opposite conclusion if the simulation results for low clouds were in better agreement with results from other CRM's and observations for the ARM A LSF case.

Page 10789: "In Fig 8d, the signature of the three thermals which were used to initiate deep convection in the STERAO case can still be seen very clearly". This again emphasizes the problem with this approach that results are quite dependent on the assumed initial bubbles and their timing. In order to rule out that the basic conclusions of this study are affected by these assumptions, the sensitivity of the results to these

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assumptions would have to be quantified. Although this might not be easy to do in practice.

Page 10790: "This study has been limited to idealized tracers...The disadvantage of idealized tracers is, however, that they are by definition not necessarily representative of realistic tracers". If the simulations are not representative of the behaviour of any real atmospheric tracer, how general can the conclusions of this study possibly be? Interactions between tracers and atmospheric processes are not limited to convective processes in reality and a wide range of time scales is generally possible for different processes in and outside clouds, depending on the tracer and meteorological situation. Tracer removal or freezing processes inside convective clouds are not independent of other tracer processes in the real world. So what is the point of doing simulations for an idealized tracer for which no equivalent may exist in the real world?

Page 10790, conclusions: "High cloud water mixing ratios in the inflow regions were found in the cases in which LSF was applied, but not in the cases in which deep convection was initiated by bubbles." Again, these differences appear to be related to shortcomings in the LSF simulations in the study. It is very likely that these differences do not represent a robust feature of LSF simulations in general.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 10773, 2006.

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