

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.

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Received and published: 14 February 2007

We appreciate the review by this referee. The comments by this referee have led to additional insights regarding the influence of the microphysical processes. We are also very grateful for the detailed comments on technical details.

Based on the general remarks by Referee #2, the sentence (in Sect. 6):

“In Colorado, where the STERAO campaign was conducted, thunderstorms can have fairly high cloud bases above ground.”

was changed to:

“The thunderstorms in Colorado, where the STERAO campaign was conducted, have higher cloud bases relative to the mean sea level and a smaller region of liquid water, potentially increasing the importance of the retention coefficient for the transport of a boundary layer tracer. In addition, Colorado thunderstorms can have fairly high cloud bases above ground.”

Major points:

1) Section 3.2 has been divided into two sections and the discussion of the ARM A BUB case was extended. The discussion in Sect. 5 has also been extended and two figures regarding the ARM A BUB case were added (Fig. 9 and Fig. 11c of the revised manuscript).

2) All cases were re-run either in the full 3-D setup (STERAO and ARM A BUB) or in a computationally much cheaper 2-D setup (TOGA COARE, ARM A LSF) with additional diagnostics regarding microphysical processes which led to interesting new insights. Fig. 12 regarding rain formation has been added and the discussion in Sect. 5 has been extended. Furthermore, a schematic regarding the microphysical processes was added in reference to comments by Referees #4 and #5 (Fig. 1 of the revised manuscript). It turns out (see discussion in Sect. 5) that most rain is formed via the ice phase in all runs, but that cloud water plays a much bigger role in the LSF runs than in the bubble runs (panels on the left hand side in Fig. 12). Furthermore, in the LSF runs cloud water contributes to rain formation at lower levels than in the bubble runs (panels on the right hand side in Fig. 12). Figs. 9b and c suggest that in the ARM A LSF run cloud droplets coexist with rain mainly in the inflow regions of the storms. Rain formation in this region is most likely a very efficient process for the scavenging of highly soluble tracers, since direct uptake of trace gases into larger rain drops is strongly limited by gas phase diffusion, in spite of ventilation being considered in Eq. 3. (For comments regarding the ventilation coefficient see reply to Referee #1).

3) By studying deep convection under different conditions, we hoped that we would

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learn something about the effects of different types of storms. It turned, however, out that in some aspects, the effect of the initialization can be more important than that of studying maritime tropical vs. continental mid-latitude convection. The ARM A BUB run was designed to better separate the effects of the initiation method. In the revised manuscript, the discussion has been directed more towards a comparison between the ARM A LSF and the ARM A BUB case rather than towards a comparison between the ARM A LSF and the STERAO case (see below and reply to Referee #3).

4) The discussion of the ARM A BUB case was extended significantly, and two additional figures regarding this case were included as indicated above. The purpose of Fig. 9 of the original manuscript is to show that high LWP and IWP lag the occurrence of high vertical velocities in the ARM A LSF and in the STERAO run. This also holds for the TOGA COARE run. Since the focus of the discussion has been directed more towards a comparison between ARM A LSF and ARM A BUB, in the revised manuscript Fig. 9b was replaced by the corresponding figure for the ARM A BUB case (Fig. 13b of the revised manuscript). The maximum vertical velocities in cloud resolving studies depend strongly on whether a two- or three dimensional model setup is used (see Fig. 3 of Redelsperger et al., 2000). In the TOGA COARE case, the simulated maximum vertical velocities also depend on the particular input dataset used to specify the LSF. A detailed discussion of these dependencies is outside the scope of this paper. In preliminary sensitivity runs, the transport of the highly soluble tracers was found not to depend on the input dataset or model dimensionality qualitatively (regarding dimensionality see Fig. 4a to Fig. 10a of the original manuscript). Sensitivity studies were conducted for TOGA COARE in order to check whether observations from the remote tropics showing increased upper tropospheric mixing ratios of H_2O_2 after deep convection could be linked to the release of H_2O_2 from freezing hydrometeors. In polluted air masses, on the other hand, aqueous phase chemistry would have to be taken into account for H_2O_2 (see e.g. Sect. 6.2 of the supplement to Tost et al., 2007).

Specific points:

1. As indicated above, Fig. 12 regarding rain water formation has been added and the discussion in Sect. 5 has been extended. A schematic regarding the microphysical processes was added in Fig. 1 of the revised manuscript.

2. Except for Fig. 8 of the revised manuscript (Fig. 6 of the original manuscript), the mixing ratios shown are gas phase mixing ratios. This is now indicated in the revised manuscript. Fig. 4 of the original manuscript would look very similar if total mixing ratios were plotted instead of gas phase mixing ratios. A multi-day sensitivity run assuming very efficient uptake of H_2O_2 using the model setup of Salzmann (2005), which is not discussed in the manuscript, indicated that ice-uptake would have a non-negligible effect on upper tropospheric gas phase mixing ratios.

3. Fig. 5 showing radar reflectivities derived from the STERAO run was added. Note, that the radar reflectivities look reasonable in the light of Fig. 7 of Skamarock et al. (2001), although some differences exist as one might expect in such a comparison. (When comparing the Figures, note the difference in length of the cross-sections). Note also that our results regarding the role of the retention coefficient agree qualitatively with those of Barth et al. (2001), who studied the same storm. For the LSF runs, a storm to storm comparison of radar reflectivities is not straight forward, since LSF runs aim at reproducing the mean properties of the cloud systems, i.e., a statistical analysis would be required for these cases. Radar reflectivities in the literature are typically reported for single storms.

4. The high time averaged near-surface hydrometeor mixing ratio for the ARM A LSF run are an artifact due to spurious condensation occurring prior to the onset of deep convection. The first 18 h have been removed from the time average in Fig. 8b of the revised manuscript (Fig. 6b of the original manuscript) and the reason for this is discussed in Sect. 5. Radar reflectivities for TOGA COARE (Fig. 5 of Jorgensen et al., 1997) and for ARM A (Cederwall et al., 1999) show high values occurring close to the surface, where precipitation is expected to lead to be located.

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5. Regarding vertical velocities see item 4 of major points. Domain averaged profiles of “T2 released” in liquid hydrometeors have been added in Fig. 8 of the revised manuscript (Fig. 6 of the original manuscript). Including these profiles adds extra information regarding the more efficient scavenging of T2 between about 5 and 9 km in the TOGA COARE vs. the ARM A LSF case. The following sentences were added to Sect. 5:

“Mixing ratios of the non-retained tracer T2 taken up by hydrometeors are shown in Fig. 8i–l. A comparison of Fig. 8i to Fig. 8j suggests that mid-tropospheric entrainment of T2 was more efficient for TOGA COARE than for ARM A LSF. This is consistent with Figs. 6a and b suggesting more efficient mid-tropospheric scavenging of T2 in the TOGA COARE run. Mid-tropospheric entrainment also enhances the mixing ratios of “T2 released” in cloud water in the ARM A BUB run (Fig. 8k) while in the STERAO case, “T2 released” in cloud water (Fig. 8l) shows even lower mid-tropospheric mixing ratios than “T1 released” in cloud water (Fig. 8h) due to lower initial mixing ratios in the boundary layer (Fig. 6). The effect of mid-tropospheric scavenging is very small for “T2 released” in the ARM A BUB and the STERAO run (Figs. 6c and d).”

6. “Competition between storms” relates to the finding that in the TOGA COARE simulation some storms release tracer in the upper troposphere, while others do not (Fig. 12 of the original manuscript). In addition to this, there is a competition between different processes in each of the storms. In order to identify the necessary conditions for a storm to efficiently release a dissolved tracer into the upper troposphere, one would have to track storms over their entire lifetime with a fairly high temporal resolution. In the TOGA COARE cloud system resolving simulations, for example, single storms often merge into larger systems, which move relative to the smaller storms (e.g. Salzmann et al., 2004). Even at a given point in time, it is often difficult to sensibly define what counts as a single “storm”, especially since the anvils in larger systems are connected (often with a small discontinuity in anvil height). Tracking storms over their lifetime complicates sorting out the differences between individual storms in a cloud system

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resolving simulation enormously.

7. The sentence “Furthermore, it is still largely uncertain to what extent H_2O_2 is taken up on ice directly from the gas phase.” on p. 10788 was removed and the following sentences were added to the discussion (in line 19 of the original manuscript):

“The uptake of H_2O_2 onto ice directly from the gas phase was found to be small in a recent study by Clegg and Abatt (2001), while an earlier study by Conklin et al. (1993), who studied the uptake of H_2O_2 in a flow tube packed with 200 μm ice-spheres, suggested more efficient uptake (see also Meier and Hendricks, 2002). It is, however, questionable whether the laboratory data obtained for packed ice crystals is applicable to single ice crystals under upper tropospheric conditions (Meier and Hendricks, 2002).”

8. The point regarding the lack of a theoretical framework has been explained in the revised version:

“In order to describe reversible exchange of trace gases between the surfaces of frozen hydrometeors and the gas phase at the same time as the possible retention of trace gases due to “burial” or trapping (e.g. Kärcher and Basko, 2004; Stuart and Jacobson, 2006) in cloud resolving model simulations, it would be necessary to carry prognostic variables for both, the surface and the bulk concentrations in the model. In this case, one would also have to take into account that microphysical processes occurring in association with the deep convection change the surface area and the ratio of surface to bulk concentration. A theoretical framework which could easily be incorporated in the microphysics parameterizations used in cloud resolving models is currently not available. Field observations of droplets freezing on an impactation grid by Snider and Huang (1998) suggested that H_2O_2 was volatilized subsequent to droplet freezing and prior to burial by continued riming.”

Technical details:

The highly appreciated technical corrections have been included into the manuscript

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with one exception and the vertical axes for STERAO and ARM have consistently been changed from AGL to above mean sea level (MSL). The only correction which was not included was the one on page 10789, line 28 of the original manuscript, where “multiply” is used as an adverb for “nested”.

p. 10777: The sentence “Here concentrations are defined as tracer mass per grid box volume” was added below the sentence containing Eq. 1.

p. 10781: The sentence “As in Barth et al. (2001) and Skamarock et al. (2000), this run is initialized with three positively buoyant thermals (“bubbles”) with radius $r=10$ km and height $z_o=1500$ m and a maximum temperature perturbation at the center of $\Delta\theta_{\max}=3$ K ...” was added.

p. 10782: The main results of the TOGA COARE case (see also Salzmann et al., 2004) have been briefly summarized in Sect. 3.1.1 of the revised manuscript.

p. 10786: The dependence on water vapor has only briefly been indicated. It is a consequence of the reaction of $O(^1D)$ with water vapor. $O(^1D)$ is produced by ozone photolysis, and its reaction with water vapor yields two OH radicals. The OH radicals readily react with CO, hydrocarbons, or ozone, to form HO_2 radicals. Some HO_2 reacts to form H_2O_2 in the HO_2+HO_2 reaction, while most reacts with NO or ozone. These reactions produce OH, amplifying tropospheric oxidation. The production of H_2O_2 does not only depend on water vapor, but also on other factors like the ozone, NO_x , and hydrocarbon concentrations. Ozone profiles often have a minimum in the remote marine boundary layer, a local maximum in the mid-troposphere, and another local minimum in the upper troposphere, the latter being associated to deep convective transport of ozone poor air.

p. 10788: In the revised manuscript, it is indicated in the caption of Fig. 6 (Fig. 4 of the original manuscript) that gas phase mixing ratios are discussed.

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