

Interactive comment on “Redistribution of trace gases by convective clouds – mixed-phase processes” by Y. Yin et al.

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General Comments:

The manuscript, “Redistribution of trace gases by convective clouds – mixed-phase processes” by Yin et al. provides some nice illustrations of how cloud microphysics can affect soluble tracer distributions in the troposphere. The redistribution of soluble constituents is important for quantifying the effect of convection on upper tropospheric chemistry and on deposition of many soluble species.

Cloud processing is complex especially when liquid and ice particles are involved. Quantifying the retention of chemical species in frozen particles when liquid drops freeze is difficult to accomplish in either laboratory or field measurements because of the challenge in constraining parameters that impact retention (e.g., pH of the solu-

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tion and chemical reactions). Recent work (Stuart and Jacobson, 2002, submitted to *J. Geophys. Res.*) indicates that several factors likely influence the retention of chemical species, including the solubility of the species (addressed here in the Yin *et al.* study), drop size and pH, temperature, and ventilation conditions. The degree of retention may also be quite different depending on the method of freezing (e.g., contact freezing, dry-growth riming, and wet-growth riming) suggesting the importance of representing cloud microphysics well. Therefore, the use of a single value for the retention coefficient is probably not realistic. Furthermore, it is difficult to determine whether measurements from field studies of the retention coefficient are appropriate for other types of clouds, especially the cloud one wants to represent in a numerical model.

The numerical model used by Yin and co-authors nicely depicts cloud microphysics and could eventually be quite useful in relating the physical processes involved in freezing drops to the retention of chemical species. Although the study presented in the article is academic and provides extremely useful information regarding the importance of the retention of chemical species in frozen particles, its limitations must also be recognized. I suggest that the following points be addressed in the final version of the manuscript.

Specific Comments:

1. What are the limitations of this study? How realistic is the simulated cloud to nature, particularly for the marine case? An initialization procedure of applying a pulse of warm air was used. Could the authors relate the simulated cloud to what we see in nature? What are the limitations of using a 2-D axisymmetric cloud model?

2. I think that the phrasing in the paper needs to be written carefully. The authors state on p. 885, line 19 that "...the maritime case is that most of the drops were converted into graupel particles upon freezing, instead of ice crystals, as in the continental case." This phrasing leads one to believe that maritime clouds tend to produce graupel while continental clouds produce ice crystals! Is that what the authors intend to say?

An earlier paper of the authors stated that the maritime cloud is maritime only in the

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characteristic of the CCN distribution. This needs to be stated in this paper too. The authors should not use the term maritime loosely. Instead, the phrasing "the cloud with the maritime CCN distribution" would be much more accurate and would remind the readers that this is not truly a maritime case.

3. Figures 6 and 7 show the tracer mass in the gas phase integrated above 4 km altitude at the simulation time of 64 min. Figures 4 and 5 indicate that ice crystals, graupel and perhaps some liquid still exist above 4 km at 64 min. Are the tracer masses in these hydrometeors negligible compared to the tracer mass in the gas phase? In other words, if the ice and graupel sublimated, would the tracer in these hydrometeors contribute much to the total tracer mass?

4. Page 886, lines 23-26, It is stated that highly soluble gases behave similarly to insoluble gases when the retention coefficient is 0 because gas expulsion from ice particles means that most of the tracer in the cloudy region is present in the gas phase. After expulsion from the ice, does the tracer dissolve into the remaining liquid (supercooled) water drops? Barth et al. (2001) *J. Geophys. Res.*, **106**, 12,381-12,400 showed evidence of this (see their Figure 11). It is not obvious to me that the highly soluble tracer is mostly in the gas phase compared to the liquid phase.

5. Section 4.3 on the effect of gas scavenging by ice particles on gas redistribution is quite interesting. The second paragraph states that the effect of gas scavenging is "negligible", but then remarks that the gas-phase concentration of a tracer with Henry's law coefficient of 10^4 M/atm is reduced by 30%. This is not negligible. However, it may be true that the total concentration of the tracer in the outflow region changes negligibly between simulations with gas scavenging and simulations without.

The text also states that the less soluble species are more affected by gas scavenging than are the highly soluble species. Without knowing how gas scavenging is represented in the model (equations in section 2.2.3 would help), it is not clear what factors contribute to the burial of a tracer into ice hydrometeors. However, I did notice that

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the authors performed two simulations with the burial coefficient equal to 0.1(*). One simulation (C7, M7) set the retention coefficient to 1 and the other simulation (C9, M9) set the retention coefficient to 0. How do the model results differ for these simulations? I would expect that much more gas scavenging of highly soluble tracers occurs for the C9, M9 simulations.

I hoped that a figure similar to Figures 6 and 7 would have been shown for the different burial coefficient simulations. Is it possible to show contours of mass of the tracer above 4 km as a function of Henry's law coefficient and burial coefficient?

(*)In the discussion of Figure 9 (text line 18 of p. 888), the text states the burial coefficient of simulation C7 is 1.0 while Table 2 shows the burial coefficient of simulation C7 to be 0.1. This needs to be consistent.

6. Washout, Mass above 4 km, and Mass initially.

Because Figures 6 and 7 and Table 2 show spatially-integrated mass of the tracer, it is important to know a few things about how the calculations were performed. The tool used is a two-dimensional model that needs to assume a third dimension in order to calculate a mass of tracer. What is the assumed grid size in the third dimension?

Because Figures 8 and 9 show mass fraction of the initial mass of the tracer, could the authors inform the reader what the initial total mass (spatially-integrated) of the tracer is. Having this knowledge allows the reader to determine the fraction of the tracer that is transported to the upper troposphere or that is deposited on the surface. However, an even better calculation of washout efficiency would be to compare the flux onto the surface to the flux of tracer being transported into the cloud. Can this kind of calculation be done?

Figures 8 and 9 need a bit of clarification. First, the percentage of the initial mass is shown (not the fraction). Second, it would help to know what x location the results represent. After about 60 minutes, the total fraction of the tracer is less than 10% of

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the initial boundary layer mixing ratio. Where did all the tracer go? Was it deposited on the surface, advected out of the domain in the outflow region, advected out of the domain in other regions, or simply diluted when mixing occurred?

It wasn't made clear, but I assume that the boundary condition of the tracer into the domain is zero (i.e., no tracer is advected into the model domain). But I remember that open boundary conditions exist so that the tracer is advected out of the model domain. How much of the tracer is exported from the model domain at low levels (with the assumption that this air did not encounter cloud)?

7. The highest solubility tracer represented has a Henry's law coefficient of 10^9 M/atm. How would the results of the simulation differ for Henry's law coefficients higher than 10^9 (such as, for a species like HNO_3)?

Technical Corrections:

- 1) p. 878, lines 15-19, I suggest rewording the last part of the sentence to something like, "... due to the low retention during riming prescribed in their simulations."
- 2) Section 2.2.1 line 4, What time step is used for dissolution?
- 3) Section 2.2.3, How is the gas scavenging of tracers calculated? It would be nice to see equations that describe the calculations.
- 4) p. 884, line 24, How many tracers with different solubilities were represented?

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 875, 2002.

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