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Interactive comment on "Simulation of trace gas redistribution by convective clouds - Liquid phase processes" *by* "Y. Yin et al."

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The manuscript, "Simulation of trace gas redistribution by convective clouds – Liquid phase processes" by Yin et al. provides some nice illustrations of how cloud microphysics can affect chemical tracer distributions in the troposphere. Below I have made some additional comments regarding their findings.

Section 4.4 The effect of assuming gas-liquid equilibrium

If one examines the kinetic limitation of transferring a species from the gas to the drop as a function of drop size, it can be found that there is a strong dependence on drop size. Soluble species have a much longer time constant to diffuse to a large drop than to a small drop. There are two interesting points regarding the Yin et al. study that arise from the above statement.

The first is to compare the marine sheared simulation with Henry's law equilibrium

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assumed to the marine case with kinetic limitations and then to the comparison of the two continental cases (kinetic limitation and equilibrium assumption). Because marine clouds generally have larger cloud drops than continental clouds, would there be a greater difference in the vertical profile of the species for the marine case than for the continental case?

The second is to compare the equilibrium case to the kinetic case for cloud droplets versus rain drops. As can be seen by Figures 6 and 12 in Yin et al., there is a big difference in the amount of soluble tracer scavenged below cloud base when the kinetic limitation is calculate (Figure 6) and when Henry's law equilibrium is used (Figure 12). (On the side, the Figure 12 caption should refer to Figure 6 rather than Figure 8 so that the reader can more easily compare the two figures.) This has an important implication on below-cloud scavenging of tracers. These figures indicate that below-cloud scavenging of soluble tracers is much smaller than in-cloud scavenging. The simulations by Barth et al. (2001) support this finding. Shown in Figure 1 below is the rate of dissolution integrated over the horizontal domain of the midlatitude convection simulated by Barth et al. at two times in the storm, at one hour when the storm was multicellular in structure and at 2 1/2 hours when the storm was a quasi-supercell. For a highly soluble tracer, the below-cloud scavenging is 3 orders of magnitude smaller than the in-cloud scavenging. For a moderately soluble tracer, the below-cloud scavenging is 30% of the in-cloud scavenging because this tracer reaches Henry's law equilibrium between the gas and liquid phases. Also note that the moderately soluble tracer volatizes when rain drops evaporate, while the highly soluble tracer does not. This indicates that the highly soluble tracer is not in Henry's law equilibrium. Thus, I would conclude from the Barth et al. and the Yin et al. studies that wet deposition parameterizations that assume Henry's law equilibrium for soluble species in rain are in error.

So how is a soluble species scavenged by precipitation? In other words, how does the species get into the rain? Soluble species readily absorb into the small cloud drops which subsequently grow via coalescence to form rain drops. So it is the microphysical processes that transfer the species from the cloud drops to the rain. If ice is considered, then a few steps may be taken, 1) absorption into cloud drops, 2) retention in the frozen drop when the drops are rimed by snow, and 3) melting of snow to form rain. Leriche et al. (2001) who employed a quasi-spectral microphysics parcel model also found that HNO3 concentrations in rain were a result of the cloud drop growth to form rain rather than direct gas absorption of HNO3. They noted that the HNO3 in rain was far away from equilibrium with the gas phase concentration and that the rain served as a reservoir of HNO3 and ultimately NOx species.

Section 4.5 Implications for specific atmospheric gases

In this section the authors discuss the implications of their results for specific atmospheric gases. This is done by applying appropriate Henry's Law coefficients for particular species. However many of the species listed would not have an initial profile as was used in this study (i.e., large amount of species in the boundary layer, no species above the boundary layer). For example, ozone will be fairly constant in the troposphere with higher values in the upper troposphere. With entrainment included, how would the authors expect their results to differ if they included more representative initial profiles for each species?

Future work:

I am curious as to what the authors think are the next steps to improving their modeling effort. Would it be to include the calculation of pH, to include ice microphysics and interactions with chemical species, or some other process?

The results of this study and Barth et al (2001) indicate that Henry's Law equilibrium can be considered in some cases, particularly for low solubility species and when small drops are considered. To improve computational efficiency, would the authors recommend and make use of a hybrid approach for considering gas absorption (an example of such an approach is given in Barth et al, 2001)?

Comment by M. Lawrence:

M. Lawrence raises some important points regarding the results of Yin et al. I agree with Lawrence's criticism that the results should be examined over an integrated

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distance of the outflow region, rather than at one particular height. In addition, the results should to be integrated over time so that the large-scale modeler can better assess the influence of convection on upper tropospheric chemistry.

Leriche, M., Chaumerliac, N., and Monod, A., Coupling quasi-spectral microphysics with multiphase chemistry: A case study of a polluted air mass at the top of the Puy de Dome mountain (France), Atmos. Environ., in press, 2001.

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