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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, Parker, and Carslaw represents an important contribution to contemporary research on the upward transport of trace gases by convective cloud updrafts and the accompanying downward transport and loss of soluble gases in convective precipitation, extending the line of recent research in Crutzen and Lawrence (2000) and Barth et al. (2001). The paper will be a strong publication in Atmospheric Chemistry and Physics after consideration of a few points which are discussed below.

The study employs a cloud-resolving model to take a detailed look at the transport and scavenging within one cloud. The results help emphasize how difficult treatment of these processes will be in global models in the forseeable future, due to the large differences in scales. For instance, it was found that in comparing a continental and marine

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cloud case, "variations of the maximum rainfall rate [of the order of 3-4 minutes] can have an important influence on the efficiency of gas transport." Since the time steps of global models are normally 10 minutes or greater, these kinds of differences are difficult to resolve and must be parameterized. Bridging the gap between the spatial and temporal scales of convective cloud processes and large-scale effects will be a major challenge for numerical model development. An important first step in many models might be including a consideration of the kinetic uptake of gases, rather than directly assuming Henry's Law equilibrium, which is indicated in this paper to be particularly important for larger raindrops. Such a kinetic computation is only done in very few global chemistry models at present. However, this will require assumtions about cloud droplet and raindrop size spectra, as well as parameterization of the uptake which occurs during the relatively long model time steps. A short discussion of the application of these results to large scale simulations, though not necessary, would help broaden the impact of this work.

My main criticism regarding the present manuscript is the strong focus on the results at the cloud top, rather than on the integrated results throughout the cloud outflow region. At the cloud top, a considerable difference between the continental unsheared, sheared, and equilibrium simulations is seen. The authors justify focusing on this level because in the middle of the cloud, where wet deposition is prevalant, the ratios of the highly soluble tracers to the insoluble tracer are very small (down to $<$ 1%), whereas at the cloud top the soluble tracer mixing ratios are up to 50% of the insoluble tracer values. However, the mixing ratios of all the tracers fall to very low levels at the cloud top, so that even if the relative amounts of the soluble gases are larger at the cloud top, it appears that the absolute amounts are larger in the main outflow region in the middle of the cloud.

For the sake of determining the chemical impact of important gases (such as halogens) which are transported to the upper troposphere, a more relevant parameter to compare would be the integrated mass of tracer between about 4 km (start of the outflow regime) and cloud top (or alternatively between 4 km and the model top). This

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indicates how much of a real tracer with a given solubility starting in the BL has actually been able to make it to the middle and upper troposphere, and how much has been lost to precipitation scavenging on the way. Based on the plots, I suspect this number will not differ much for the three continental cases considered (Figs. 6 and 12), and will only differ modestly for the marine case (Fig. 8). It is impossible to judge this accurately from the plots, due to the small values and opposing curves, and thus it would be very interesting to see what values are found if the authors can perform this integration.

Once this is done, it may be necessary to reconsider the importance of differences in the simulations, in particular if it is found that the consideration of kinetic uptake does not make much difference to the integrated amount of tracer which is transported to above 4 km (which appears to be the case based on the figures interpreted this way). If this is the case, can it still be argued that this is important to consider at present in global models? It will also be interesting to compare the results in that light to those in Crutzen and Lawrence (2000) and Barth et al. (2001). It appears that there is less scavenging in the cloud model results presented here than in our global model runs: the amounts of the Hx = 10^3, 10^4, and 10^5 tracers remaining relative to the insoluble tracer in our global model runs were found to be about 80-90%, 40-60%, and 10-20%, while in Figures 6, 8, and 12 it appears to be 90-95%, 60-80%, and 20-30%. If this difference in the integrated transport amounts cannot be attributed to the neglect of kinetic uptake limitations in the global model, then another reason will need to be proposed and tested, either by the authors or in future studies.

In order to illustrate this point, and to enable the authors to make a clear comparison to the values in our global study (rather than just guessing from the figures), I've gone back through our old data and computed the integrated global amounts of tracer above and below 600 hPa, which is roughly the lower boundary of the outflow region of most convective clouds. These results (for our base run, with tracers fixed to a mixing ratio of 1 at the surface and given a 30-day decay lifetime, plus scavenging based on their solubility), are in the Table below. I've also listed the values for thresholds of 400 and 800 hPa to check the dependence on the chosen threshold. The results confirm the basic impression from the figures in our paper: for trace gases with a Henry's Law value (Hx) of 10^3 M/atm, about 15% is scavenged relative to the insoluble gas before reaching the model levels above 600 hPa, while for the Hx=10^4 M/atm and Hx=10^5 M/atm tracers the amount scavenged is about 50% and 85%. The results are nearly independent of what threshold for the cloud base is chosen between 800 and 400 hPa. Furthermore, the results are also almost the same for the tracers with a 10-day decay lifetime, with only slightly less being scavenged (relative to the insoluble gas) than in the 30-day decay lifetime case. A comparison of these values with integrated values above and below cloud base in the cloud model for the various cases examined by Yin and colleagues may be particularly helpful in determining where descrepancies exist, what may be causing them, and how to improve the coarse representation of convection and scavenging in global models.

The following Table shows the globally integrated tracer mass values based on Crutzen and Lawrence (2000); PSPLIT is the pressure which divides the "above" and "below" model levels, and the tracer names indicate their solubility: HLNS - not soluble; HL01 - Hx=10^1 M/atm; HL02 - Hx=10^2 M/atm; etc.

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	mass (x10^{15} kg)			ratio to insoluble tracer		
tracer	total	above	below	total	above	below
PSPLIT = 600.0 (hPa)						
HLNS	2228.6	947.4	1281.2	1.00	1.00	1.00
HL01	2225.4	945.6	1279.7	1.00	1.00	1.00
HL02	2197.2	930.5	1266.7	0.99	0.98	0.99
HL03	1975.4	812.6	1162.8	0.89	0.86	0.91
HL04	1253.5	446.3	807.1	0.56	0.47	0.63
HL05	671.8	151.7	520.1	0.30	0.16	0.41
HL06	427.7	33.9	393.8	0.19	0.04	0.31
HL07	379.0	15.5	363.4	0.17	0.02	0.28
HL08	368.8	13.1	355.7	0.17	0.01	0.28
HL09	365.3	12.7	352.7	0.16	0.01	0.28
HL10	356.9	12.2	344.8	0.16	0.01	0.27
PSPLIT = 400.0 (hPa)						
HL02	2197.2	506.8	1690.4	0.99	0.98	0.99
HL03	1975.4	444.8	1530.7	0.89	0.86	0.89
HL04	1253.5	253.2	1000.2	0.56	0.49	0.58
HL05	671.8	89.0	582.7	0.30	0.17	0.34
HL06	427.7	14.8	412.9	0.19	0.03	0.24
PSPLIT = 800.0 (hPa)						
HL02	2197.2	1422.2	775.0	0.99	0.98	0.99
HL03	1975.4	1243.7	731.7	0.89	0.86	0.94
HL04	1253.5	683.4	570.1	0.56	0.47	0.73
HL05	671.8	258.3	413.5	0.30	0.18	0.53
HL06	427.7	103.0	324.7	0.19	0.07	0.42

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A final point with respect to considering the values at the cloud top: a peak in the ratio of the soluble to insoluble tracers a km above the cloud top, where relatively little vertical transport should be occuring. Do the authors have an explanation for this peak? Could it be due to numerical diffusion of the very low tracer mixing ratios?

The point is also made that the mixing ratios of the soluble tracers below the cloud in the case assuming Henry's Law equilibrium (Fig. 12) are much lower than those in the case considering kinetic uptake (Fig. 6). In this case, integrating the amount below 4 km would also be illustrative to make clear how large this difference is. Is this difference mainly due to below-cloud scavenging? This would make sense, since raindrops falling through the layers below the cloud will not reside long, and assuming Henry's Law equilibrium would be expected to enhance the uptake. This would be interesting to quantify by performing a mixed experiment with equilibrium uptake within the cloud, but kinetic uptake below the cloud. How do the differences in integrated tracer amounts below 4 km for these two cases compare with the amounts above 4 km? Does this make sense, since the below-cloud region is the source region for the tracers entering the cloud? These additional pieces of information should not be too difficult to compute, but would be quite informative.

In comparing Figures 9, 10, and 11, I do not understand why there is generally less tracer in the cloud droplets for the Hx=10^4 case than in both the Hx=10^2 case and the Hx=10^6 case (that is, why there is a minimum in the aqueous phase mixing ratios for Hx=10^4), particularly in the upper part of the cloud. Does this have to do with the tradeoff between the amounts which are lost due to wet deposition (and thus the amount left in gas phase to be taken up into droplets later in the simulation) and the fraction of gas which is taken up, or is there another explanation?

It would be interesting to see the ratio of the curves in Figure 12 versus those in Figure 6, although comparing integrated amounts above and below 4 km would also provide the same basic information.

Why isn't point 5 (HO2) in Figure 13 exactly on the line (since it has no temperature dependence)? The difference looks larger than typical printer misplacement error.

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Figure 13 would also be interesting for the 6 km maximum in cloud outflow, rather than just at the cloud top.

The statement "Model parameterizations of gas transport through clouds (Crutzen and Lawrence, 2000) assume that the gases have temperature- independent Henry's law constants" *only* applies to the Crutzen and Lawrence (2000) study, as far as I am aware. We assumed no temperature dependence in that study because it was a first study of this nature, and we wanted to demonstrate the overall principle of dependence of trace gas transport on solubility for simplified and easy to interpret tracers. In computations with real gases, we recompute the temperature dependent solubility every model timestep (see Lawrence et al., 1999); as far as I am aware, most atmospheric chemistry models also consider the temperature dependence of the solubility. This statement should be reworded appropriately.

The comparison between HCHO and CH3OH in Figure 13 is not really sufficient to demonstrate that "for moderately soluble gases the effect increases with increasing enthalpy;" a plot of the deviation versus the enthalpy would be much more informative. Is the dependence nonlinear? CH3C(O)OOH and CH3OOH have just barely higher enthalpies than CH3OH, but appear to be notably further from the line. Also, the gases HNO2 and SO2 do not fit this at all; HNO2 has a larger enthalpy than SO2, but is much closer to the line. Can these deviations from the general statement be justified (perhaps due to subsequent dissociation in the solution, which could be tested by turning the dissociation off)? The most instructive study, if possible, would be a run with a suite of fictitious tracers with a range of solubilities and enthaplies, and to then produce a 2D contour plot of the deviation from the no-temperature-dependence-line as a function of both the solubility (at a reference temperature) and the enthalpy.

Could one interpret the differences in Figure 13 as being indicative of the "effective average cloud temperature" that a trace gas (or set of trace gases) experiences? By choosing a temperature somewhat below 0 degrees C, the points would all shift towards the right to being in better agreement with the curve. Is there a temperature at which a best fit of the points to the curve is obtained? What (if anything) does this say about

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the average path of trace gases on their way to the cloud top (where the temperature is much below 0°C)?

The second to last paragraph in the Summary ("We have found...") should be revised accordingly based on my last three comments (above). Also, in that paragraph, a space is needed between "$0°$C" and "a".

In Figure 14 more of the soluble gas comes from higher altitudes than the insoluble gas; is this mainly due to downdrafts, as indicated in section 4.6 (note that it should be "downdrafts", not "downdraft"), or is there also a significant contribution from evaporating precipitation?

In section 4.6, it is mentioned that the cloud covers 1/3 of the 2D domain, or equivalently 1/9 (10%) of a 3D domain, and that larger clouds would have a greater effect than found here. Although the mean fraction of convective clouds is poorly classified, from various satellite studies it appears likely to be around 10% or less, and thus the results here are probably fairly representative of an "average" situation.

Plus a few minor points:

In the introduction, it would be appropriate to already mention the Barth et al. (2001) study, since the two studies are in many ways comparable.

Why are the lines in Figure 1c not smoother?

I appreciate that the authors added color to Figure 2 on my recommendation, but it would be wise to also keep the readily- distiguishable linestyles from the first draft for the sake of B/W copies.

In section 3.1, it would be nice to briefly state the values of N_o and k which are used, rather than only referring to the previous literature.

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