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Atmospheric
Chemistry
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Discussions

Interactive comment on "Toward a more physical representation of precipitation scavenging in global chemistry models: cloud overlap and ice physics and their impact on tropospheric ozone" *by* J. L. Neu and M. J. Prather

Anonymous Referee #2

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General Comments: The authors present a new parameterization for washout/rainout processes in CTMs, which highlights the following improvements: a) a more physical representation of removal by ice particles, representing a realistic mean between neglect of ice processes on the one hand, and parameterization of removal by ice using a simple Henry's law approach; b) taking into account cloud overlap and its consequences on the microphysics of precipitation formation. Removal by frozen precipitation is crucial for a realistic representation of nitric acid in the mid-upper troposphere, while the treatment of cloud overlap (maximum overlap) affects primarily the tropical

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troposphere. The study also considers scavenging of H2O2 and HNO4. In particular, removal of pernitric acid could have an important effect on the ozone budget in the upper troposphere. The manuscript is well organized, and the scientific results are important. In addition, the formulation presented can be applied to other species, such as SO2 and aerosols, and can be easily modified to incorporate the cloud parameters calculated by different large-scale or cloud=resolving models. Because of this, I recommend publication. However, the manuscript has some issues with clarity of exposition at some points. As discussed below, there is little exploration of the reasons for the model results (NOx, O3) for the different cases examined. The algorithm itself is by necessity somewhat complex, but I found the appendix difficult to follow in some points. This could be improved with, for example, the use of illustrator diagrams, an overall "map" for the discussion (some things, such as the local precipitation, are introduced and then discussed below), and simplifying the variables/nomenclature as much as possible. Also, the authors seem to have made it a principle not to number equations, which makes discussion somewhat harder. More details are given below.

Specific Comments Abstract, line 11: "doubles the lifetime with respect to…" It may help if the authors are more concrete hera, ie., double with respect of treatment frozen precipitation removal with a Henry's Law mechanism? Utilization of average cloud cover? (Later in the manuscript the "doubles" applies to the inclusion of improved ice removal). Also, "doubles" is somewhat altitude dependent. Maybe "increases by factors of x to y" is better?

Page 24416, line 6: "effectively buried as the crystals continuously evaporate and regrow" this sounds ambiguous; it could be interpreted as the trace gases not being rereleased once they are "buried", but that does not seem consistent with the last section of the appendix (which in itself is not very clear).

Page 24417, line 7: Given the host of different precipitations, grid=averaged vs. local, and at top or bottom level, it may be useful to have a summary table (maybe in the appendix). Page 24418, first line: In the equation for Fscav, are we assuming that

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the parametric rate of conversion to precipitation (k) is the same for liquid water and for ice? Page 24418, last paragraph: If I read it correctly, JK 99 assumes maximum-random overlap of clouds, similar to what the authors adopted in their radiation paper. Would their current assumption of maximum overlap only make a difference in the results? Page 24421, lines 20-21. See comment re. similar statement in the abstract. However, please note that here the “doubling” of lifetime is due only to the treatment of frozen precipitation, whereas the abstract suggests that it is due to both the treatment of frozen precipitation and cloud overlap. Page 24423, section 3, simulations: I found myself constantly searching through these pages to see which case was which (UCI=GMI, UCi-HLAw, etc.). What about a summary table? Page 24424, lines 13 and following. The UCI-QUANTIFY scenario seems somewhat irrelevant other scenarios which focus on the impact of different scavenging assumptions. UCI-QUANTIFY simply changes the emission scenario. Given the uncertainties in these scenarios, one could have simply scaled the emissions if one wanted better agreement, or, alternatively, the authors can simply refer to the better agreement without introducing yet another scenario. Page 24424, line 18. If I am not mistaken, the “GMI” algorithm is really the one originally used in GEOS-CHEM, as explained in Liu et al. and other publications, so proper acknowledgment should be given. Page 24425, line 8: equation for loss rate. There are two issues here: a) please define F, and b) previously, the units of the precipitation rate have been given as kg m-2 s-1, whereas the units for k seem to assume mm for the precipitation. It will not change the equation, but might as well be consistent in the units. Page 24428, first paragraph. This is the first example of results being presented which seem “surprising”, yet no explanation or conjecture is given for the model behavior. In this case, HNO3 in UCI and GMI are very similar, despite the more than a factor of 2 difference in lifetimes. Is this due to an averaging effect (i.e., the 250% difference holds in only localized regions), or is it due to the non-linearity of the system, the impact on HOx, etc? Page 24429, line 17: “by the boxes in 3c (not b)”. Page 24430, lines 5 to 10. First, it seems that the increase in HNO3 in UCI relative to GMI is much larger than the corresponding NOx increase. Why? There is no explanation, and something should be attempted, given the importance of the NOx increase to the ozone increase. For example, is this because there are other important NOx reservoirs, such as PAN? Are we seeing a non-linear behavior in the NOx/HNO3 ratio? Also, the statement that the impact is small in the tropics because of "low concentrations of nitric acid and NOx" is not very convincing: we are looking at percent changes, not absolute differences. Page 24430, discussion of Figure 5 (and Figure 5): Another unexplained result: the ozone profile seems to increase by quite a bit (20-30%) in the middle-upper troposphere. What is the increase in NOx for the same conditions? Also, why does H2O2 decrease? These questions could be answered if the authors looked at some other diagnostics (such as HOx). Page 24431. H2O2 scavenging: Why is there such a small impact on the ozone budget? Why is the H2O2 change due to scavenging so different relative to that of HNO3 (both are very soluble)? Is it due to ice? As I mentioned earlier, it would be useful to see the impact of these processes on other contributors to the ozone budget, such as HOx. Page 24431 lines 21-22: "The reason for this can be seen in Figure 7". The figure does not give a reason for the behavior, it just illustrates it. Page 24435 line 7: "likely because H2O2 is not a major source…" I believe the statement, but why "likely"? Again, this is something that should be easily diagnosed from the model results. Appendix A. Granted that the algorithm adopted is rather complex, I found it very difficult to follow this section, and, although I am confident that everything is correct, after a while I found it very time consuming to check all the equations. There are some improvements that the authors may consider: I. Give a "map" for the logical progression. The problem is that variables are introduced, but it is not until the end that you see everything fitting together. II. It may be useful to have a sketch of cloud overlap with the different variables denoted. III. What about numbering the equations? IV. It is stated at the beginning that capitals are used for grid-averaged quantities, whereas small letters are used for "local" (subgrid) quantities. But then, we are introduced to fMC, fNC, etc. Why not FMC, since these denote fractions of the whole grid box? V. The logic becomes hard (but not impossible) to follow once we start dealing with the f(cap), p, p', etc. on page 24439 and following. As a function of L, some seem to denote quantities at the bottom of the layer, while others at the top. . . I found myself repeatedly going back to these pages every time I wanted to understand one of the subsequent equations. VI. I would encourage the use of “max(1,xxxx)” and “min(0, xxxx”) whenever is appropriate. It is used in some equations later in the Appendix, but why not in the set of equations at the top of page 24439? (I think that it is better than saying that quantities are constrained to be between 0 and 1) VII. Page 24439, line 5. This is picky, I know, but the argument of the sum should be something like fXX-,>yy where XX, YY = MC, NC, AM, and the sum should be over YY.

Page 24462, Figure 5: You cannot see the blue, black lines on left panel. It they overlap one of the lines, which one is it?

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 24413, 2011.