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Discussions

Interactive comment on "The influence of nitric acid on the cloud processing of aerosol particles" *by* S. Romakkaniemi et al.

Anonymous Referee #2

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Review of "The Influence of nitric acid on the cloud processing of aerosol particles" By Rommakkaniemi, H. Kokkola, K. E. J. Lehtinen, and A. Laaksonen

Overall impression

The paper presents some interesting calculations of the simultaneous effect of co-condensation of nitric acid and water vapor, in-cloud sulphate production, and coagulation on aerosol and cloud particle size distributions. In principle, the paper is suitable for publication, but only after major revision. I elaborate below.

Major points (not necessarily in order of importance):

1) One of the main points highlighted in the abstract is that HNO3-enhanced CDNC

will result in enhanced in-cloud coagulation rate and the number of interstitial particles reduces faster. Close observation of Figure 2 suggests that the effect of HNO3 on total number is very small compared to the overall reduction rate due to coagulation. I also see no evidence of significant changes in the slopes of the lines for interstitial particles. Thus, this conclusion seems incorrect.

2) The effect of varying HNO3 concentrations on aerosol size distributions is negligible (Figure 3). The effect of coagulation by itself seems much stronger that I would have expected, but affects the size range for D $>$ 50 nm (the typical population contributing to the droplet population) very little. The drop size distributions are affected by the HNO3 - clearly there are more, smaller drops. But I suspect there are some numerical dispersion problems (or "mapping problems" - see point 5 below) producing the multiple modes since they appear with regularity (e.g., Fig 3f).

3) The authors say that they stopped the simulation before drop collisions would be an important effect but I suspect that 1200 sec = 1200 m for Figure 2 represents conditions where there would typically be significant liquid water (in an adiabatic cloud) and at CDNC of ˜ 350 - 500/cc, I suspect that coalescence may be more significant than implied. Certainly if these conditions were applied for cleaner conditions (e.g., marine calculations; table 3), I would not conclude that drop coalescence was insignificant.

4) Results in Table 3: It is very difficult to draw conclusions from this table since the different entries represent different combinations of h and w and therefore different times for processing and different liquid water histories. I think the authors should consider some ways to compare similar conditions in a way that the differences are clearly understood to be due to w, or time, or liquid water - all of which affect processing. Without this the results don't improve understanding and don't add to what is already known about processing.

5) Coagulation calculations: I am concerned about the numerical method of treating coagulation. Every time two aerosol size classes interact they will produce a new

size class so that accurate treatment of coagulation quickly becomes computationally impossible because the number of classes increases exponentially. Therefore the authors add another 20-30 classes to accommodate these new particles. This seems like a very small number. Is it adequate? What is the sensitivity of the results to the number of additional classes? I am concerned that the redistribution of drops to neighbouring size classes will generate numerical diffusion that may give coagulation rates that are much too strong. The authors should show some results of their scheme and compare, for example to an analytical solution (e.g., for a constant coagulation kernel). I was also surprised to see such strong coagulation reduction in total particle number in Figure 2 over such a short period of time.

6) Conclusions: Opening sentence of last pgph, The increased CDNC due to HNO3 was shown to speed up the scavenging of interstitial particles. I never found any physical explanation for this in the text and without adding some depth to this discussion, it does not contribute to our understanding. If others have already explored this, please summarise their work. Otherwise, please add calculations/discussion.

7) The closing statement: In some cases it is even possible that CDNC is smaller because of [the] presence of HNO3 during the previous cycle. Statements like this can be misleading since they don't establish a clear basis for comparison. This is a very important point throughout the study. If the basis for comparison were clearly established (and there are many options) then the conclusions would be much stronger. Since they are not, I am left wondering about the significance of the results in this paper.

8) I feel it is very important that the authors broaden their thinking when writing the conclusions/abstract. I was left wondering whether the effects considered are of any significance to climate change problems. Over what parameter space (HNO3, SO2, w, cloud depth, etc) are the current results expected to be of importance? Is this range of parameter space realistic in the atmosphere?

Minor points:

1) It would be much better to plot figure 2 in mixing ratio units (i.e., number per kg of air - or equivalent) to immediately remove the effect of volume increase with increasing height.

2) Figure 3: The minor tick marks are barely visible in the top panel so it is hard to see the range of sizes over which the effect of HNO3 is significant.

3) Section 3.3.2 : The opening statement is inaccurate/misleading. The bimodality occurs due to processing but the extent of it is a strong function of the number of particles upon which mass is added as well as the amount of mass produced.

4) Table 3 would be much clearer if the values were given as a % change rather than an absolute number.

5) I was surprised at the strong increase in CDNC in Figure 2 at w=1m/s. Increases in CDNC are typically at much lower w.

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