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

S. Romakkaniemi et al.

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We thank Referee # 2 for the good comments.

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.

It is true that the effect of HNO3 on total number is clearly smaller than the overall reduction. Figure 2 will be redrawn to remove the effect of volume increase. We also add a subplot where the relative increase in the scavenged fraction due to HNO3 can be seen. The highest relative effect is at the early stage of cloud, decreasing as a function of time in cloud. From the new figure it can be seen that with 1 ppb of HNO3 a scavenged fraction of aerosol particles can be more than 1.13 times the scavenged fraction without HNO3. With 2 ppb of HNO3 the enhancement can be as high as 1.22. We feel that such a high increase in the scavenged fraction is not insignificant. We will change the wording in the abstract to be more precise.

2) and 5) 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") producing the multiple modes since they appear with regularity (e.g., Fig 3f). 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.

ACPD

5, S3987–S3993, 2005

The coagulation scheme was compared to Smoluchowski's analytical solution for a constant coagulation kernel, and it was found that analytical solution and model result were identical.

The 20-30 size classes mentioned are for droplets from collisions between cloud droplets. This is because cloud droplet distribution is very narrow and in the collisions between 2 cloud droplets the forming droplet is much more dilute than the cloud droplet of similar size formed through condensation only. If the new droplet is placed to the same bin with the cloud droplets formed through condensation only, the wet size will be correct but the dry size of particles in the size class will decrease. During the simulation the "target size class" i_{wet} is determined by the wet size of aerosol particles and cloud droplets. This is compared to "target size class" i_{dry} based on dry size. If i_{wet} is considerably bigger than i_{dry}, the formed droplet will be positioned to a new distribution, i.e. to those 20-30 classes mentioned. Otherwise the collision product is placed to the correct place in the initial distribution. Introduction of these new size classes is important only in the simulation of cloud cycles and is not essential if only individual updraft is studied like is done in Figure 2. Actually these new size classes are not included in the simulations presented in Figure 2. This part will be clarified in the manuscript.

The reason for the multiple modes seen in the cloud droplet distribution presented in Figure 3f is that the cloud droplet distribution is very narrow and so there will be peaks just like if we start the coagulation from monodisperse aerosol. This is because the size resolution in the model is better than the space between the droplets formed through coagulation. These peaks can be avoided only by decreasing the resolution.

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 condi-

ACPD

5, S3987–S3993, 2005

tions 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.

This is true, in cleaner conditions the drop-drop -collisions could be important and possibly the drizzle formation could start. To show that the application of a box model is justified, it is stated that in the conditions studied the reduction in cloud droplet number is only 6-9 per cc, and so the drizzle formation due to gravitational settling is not effective in this case. This statement is not ment to apply to other conditions.

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.

Table 3 will be corrected. The new presented results will be with similar processing times.

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.

ACPD

5, S3987–S3993, 2005

It is said in the beginning of section 3.2 that “HNO3 increases CDNC leading to a decrease in the mean size of the droplets. This also affects the coagulation scavenging of interstitial particles. The total cloud droplet surface area increases and so the Brownian coagulation rate increases. On the other hand, both gravitational and turbulence induced coagulation slow down.” So based on the simulations, the increase in the total surface area of the distribution overcomes the effect of decreased mean size. This will be stated more clearly in the revised version.

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.

What is ment with this statement is that it is possible that presence of HNO3 can actually decrease the cloud droplet number concentration after several cloud cycles, although it increases droplet number concentration during individual updraft. This kind of coupling makes it difficult to conclude what is the overall effect of condensable trace gases on global radiative forcing without proper 3D simulations.

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

ACPD

5, S3987–S3993, 2005

parameter space realistic in the atmosphere?

We are not able to produce simulations in the whole parameter space because the amount of different parameter combinations is too high and so the calculation time needed is too long for our current model. The main message of this paper is that coupling of the effect of semivolatile gases on cloud droplet formation and the cloud processing can be of importance. For example, it is known that semivolatile gases increase the cloud droplet concentration, but like presented in this study, this holds only for individual updrafts. Here it is shown that it is possible that presence of HNO3 can actually decrease the cloud droplet number concentration after several cloud cycles. This kind of coupling makes it difficult to conclude what is the overall effect of condensable trace gases on global radiative forcing without proper 3D simulations that also include sulfur chemistry.

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.

This is a very good idea. Figure will be redrawn. See point 1 above

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.

We will change the limits and add grid to subplots in Figure 3.

ACPD

5, S3987–S3993, 2005

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.

The first sentence will be replaced with the following statement. “Formation of sulfuric acid increases the water soluble mass in cloud droplets, which can lead to bimodality in aerosol particle size distribution after the evaporation of cloud.”

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

Table 3 will be corrected, so that we use more comparable conditions for all updrafts.

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.

HNO3 effect on cloud droplet number concentration depends on the temperature and distribution used. With a more acidic distribution the effect would be smaller.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 10197, 2005.