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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 # 1 for the good comments.

Page 4: "is between 20 and 30": is this not fixed, where does it depend on? And is the resolution not too coarse given the "wiggles" in the drop size distribution (Fig 3)?

It was found in the simulations that the additional number (described in advance) of size classes for the droplet-droplet collisions needed is 20-30. Higher number of bins increases accuracy but it also increases computation time. So the number of additional size classes is a kind of compromise between the accuracy and the computational

cost. If the possibility of a rain formation would be included into the model, the number of these classes should be higher. Because we use a box model, and the gravitational settling can not be modelled correctly, we study only nonprecipitating clouds in which the cloud droplet distribution does not broaden enough to produce drizzle.

"This decreases numerical diffusion..." I do not understand this remark. Is there another way to use processed aerosol as input that produces more numerical diffusion?'

What is meant here is that because the cloud droplet distribution is very narrow a collision between 2 cloud droplets leads to droplet which 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 the size class will decrease. The use of the term 'numerical diffusion' is not quite correct in this context, and is removed from the revised manuscript. 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 simulation of cloud cycles and is not essential if only individual updraft is studied like is done in Figure 2.

Page 5, section 3.2: Should be rewritten for clarity, some questions and apparent inconsistencies arise. "...uptake of HNO3 enhances the coagulation scavenging": How important is this when also the time period in-between clouds is considered? I can imagine that the fact that particles are NOT scavenged inside the cloud at 0 ppbv HNO3, is to some extent compensated by more efficient impaction after the cloud has evaporated. Since the model also accounts for coalescence between unactivated

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particles this issue could be investigated."

It is compensated to some extent, but the coagulation rate in clouds is many orders of magnitude higher than in cloud free conditions. So the time needed for compensation is very long.

the number of aerosol particles is appr. 50 cm-3 lower... " But the total amount of particles (aerosol+ cloud drops) is much closer, so how much of the decrease is due to additional activation (HNO3 effect) and how much to coalescence?

There is a difference of 50 cm-3 in the total number of particles (aerosol+ cloud drops) when 0 ppb and 4 ppb cases are compared, although it is not very clear in Figure 2. We revise Figure 2 according to guidelines of Referee #2 and we also add a subplot to this figure where it can be seen more clearly how much HNO3 enhances coagulation scavenging. Enhanced coagulation scavenging is due to increased total surface area of aerosols particles leading to a more effective Brownian coagulation.

chosen to be short enough to prevent..." why do you not want to consider that?

Simply because when the box model is used, the gravitational settling of large cloud droplets can not be modeled correctly and so we want to be sure that droplets are not big enough to form drizzle.

number of maximas... is smaller": the maxima probably indicate a numerical problem (resolution?).

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The reason is resolution, but we do not consider it as a problem. Like stated in the answer to Referee #2, 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 like if we start the coagulation from monodisperse aerosol. This is because the size resolution (after cloud formation) in the model is better than the space between the droplets formed through coagulation. These peaks can be avoided only by decreasing the resolution, not increasing.

"This implies that rain formation can be delayed due to HNO3" But you wanted to prevent efficient droplet-droplet coalescence! Either leave it at that and do not discuss any consequences on rain formation in the paper, or simulate it fully.

This discussion is removed from the revised manuscript.

Page 6, section 3.3. The different pH dependence of the O3 oxidation vs. peroxide oxidation has been discussed already many times. Not much news here.

This section is removed from the revised manuscript.

Section 3.3.1 "of HNO3 on aerosol size distributions.... " which distributions?

Continental from Table 1 at 90% RH. This will be added to text.

"Due to 1 ppb of HNO3 98 cm-3 more cloud drops form": in the previous section there is a smaller (50 cm-3) difference for a larger amount (4 ppb) of HNO3. Apparently I do not understand it correctly, please explain more clearly.

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In the previous section there is a difference of 50 cm^{-3} in the total number of aerosol particles (i.e. interstitial particles + cloud droplets). The difference of 98 cm^{-3} is in the cloud droplet number.

Section 3.3.2. Is it possible to add a plot of the supersaturation needed to activate the particles vs. radius, after each cloud cycle? That would make the results for the different cloud cycle simulations more clear.

We are not really sure what extra value this addition would provide and have decided not to add such a figure.

"On the third cycle CDNC is even higher..." please explain.

From Table 4 it can be seen that when three sequential cloud cycles are studied (with updraft velocities 1 m/s, 0.2 m/s and 0.1 m/s for first, second and third cycle, respectively), the number of cloud droplets formed during the last cycle is 245 per cc if there is 1 ppb of HNO3 present and 267 per cc without HNO3. This is rather surprising, because it is expected that (atleast for individual updrafts) HNO3 increases the number of cloud droplets.

Conclusions. What I miss is: what is the message of this study? What is the more general importance of these processes for cloud evolution and climate change? I would advise to formulate a clear research question as a basis for the study and a systematic approach to answer that question.
The second paragraph of the conclusions is not very clear. Please rewrite

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The main message is that coupling of the effect of semivolatile gases on cloud droplet formation and the cloud processing can be of importance. 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.
We have rewritten the conclusions accordingly in the revised manuscript.

Figure captions. It was unclear which distributions have been used in which simulations. This should also be mentioned in the captions. E.g., when I compare the distributions in Fig. 3 and Fig 4. I can see that the initial aerosol is different, but what is used exactly? Figure 3 a-c: Why is there such little difference in scavenging of interstitial particles between 0, 1 and 4 ppbv? Or is the figure simply too small? And why is there a larger difference between 0 and 1 ppbv in Figure 4? I assume that different aerosol distributions are used here, is the specific distribution a critical parameter for the magnitude of the effects?

Initial distributions are the same, but presented at different RH. In Figure 4 distribution is presented after cloud cycle, and in Figure 3 distribution is presented in cloud. Also the number of size classes used in calculation of Figure 4 distribution is smaller (because the inclusion of sulfur acid production slows down the model remarkably) and in this case it leads to the bigger difference in the number of cloud droplets formed and consequently to the bigger difference in the number of interstitial particles. Figure will be redrawn with better accuracy and conditions will be explained more carefully in the revised manuscript.

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The effect of HNO3 depends on the aerosol particle size distribution used and in this sence distribution is a parameter. Otherwise, the range of sizes HNO3 effect is signifigant can be seen from Figure 3 (better in the revised manuscript).

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