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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Growth of upper tropospheric aerosols due to uptake of HNO_3" *by* S. Romakkaniemi et al.

Anonymous Referee #2

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This paper describes the growth of upper tropospheric aerosols as they uptake HNO3 from the gas phase as a function of saturation ratio in the air. In addition, the impact of letovicite formation is shown to amplify the uptake process of HNO3. Overall, I find this paper somewhat interesting, but the presentation needs to significantly improve before the paper is accepted for publication. In my opinion this paper is only suitable for publication if the authors can show that adding the letovicite to the model improves the comparisons between model calculations and the observed data. At this point, I cannot see much of (if any) improvement (see below) for the comparisons shown in Figures 5 and 6 in the presence and/or absence of letovicite. If the contrast is not strong between Figures 5 and 6, then not much can be said about the role of letovicite, which seems to be the main focus of this paper.

Specific comments.

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Abstract: In the abstract the authors state that letovicite helps the growth of particles, but the explanation given for this effect is incorrect. More uptake of HNO3 occurs when letovicite is present not because of the lowering of the DRH, but because letovicite is an acidic salt. When letovicite forms in the solution, it removes some of the acidity, which can then help to take in more of an acidic molecule, like HNO3. Please rewrite the abstract to make it more clear. In fact, what is written in the conclusion section is perhaps a better abstract to use for this paper.

Last line, Page 1: I am not quite sure what the last sentence means. Letovicite has a fix solid composition. What partitioning are you referring to here?

Table 1. Describe the chemical composition of the three modes in the Table caption. If modes 1 and 2 have ammonium ion in solution, they can uptake more HNO3 (see the Karcher reference) as the air gets more humid as compared to pure sulfate. I am not sure why it is necessary to add ammonium only to the third mode. What is the mixing ratio of ammonium to sulfate used for mode 3? Since the Clegg model is used here, one can calculate the chemical composition for different mixing ratio assumptions of ammonium to sulfate. Thus, it seems quite arbitrary to fix this mole ratio to that of letovicite. Some explanation is needed as to why the composition in the modes were chosen to be H2SO4/H2O in modes 1 and 2 and just letovicite in mode 3. Will your answer change if all three modes were assumed to contain ammonium ion?

Similar is misspelled in page 2.

I am not quite sure what is plotted in Figures 1 and 2. The legends are not described properly and the purpose of these plots is not explained clearly in the text. For example, Figure 1 looks at HNO3 uptake for 0 to 2 ppb, while Figure 2 illustrates uptake only for 1 ppb of HNO3. Why?

Again the legends in Figures 3 and 4 are not clear. Also, how sensitive is your calculation to ammonium concentration in the third mode? As I mentioned above, it is quite arbitrary to fix the NH4+ to sulfate ratio to that of letovicite in mode 3. An explanation

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as to why a letovicite composition was assumed for mode 3 could be useful here.

The information plotted in the 3 panels of Figures 5 and 6 needs to be explained in the caption. What is M11, M12, M21, M3 and M5? Also, why are you changing the mixing ratios between Figures 5 and 6? I don't see much improvement in data/model comparisons between Figures 5 and 6. In addition, the use of such high values for HNO3 mixing ratio used in the calculations needs to be justified. Usually such high values in the upper troposphere are present only in polluted air masses, which are lofted by dry convection. Was this the case for the dataset used? Also, can you show a plot of HNO3 mixing ratio measured during this campaign?

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 121, 2004.