

Interactive comment on “Growth of upper tropospheric aerosols due to uptake of HNO₃” by S. Romakkaniemi et al.

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We thank referee 2 for the comments.

General: The main focus of this paper is, as the title implies, the effect of HNO₃ on the hygroscopic growth of UT aerosols. The role of letovicite is a secondary issue, although clearly an important one. Figs. 5 and 6 show clearly that there is a quantitative difference in the amplification by HNO₃ of hygroscopic growth depending on whether or not a letovicite mode is included. However, we believe that showing that model calculations can explain real observations of increases in UT haze mode is valuable in itself, regardless of whether or not the presence of letovicite is required.

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Abstract: Will be rewritten.

Last line, p. 1: The sentence referred was already changed for the ACPD version of the article. The current wording is "We also calculate the HNO₃-dependent DRH of letovicite."

Table 1: The chemical compositions will be de described in table caption. We have ammonium only in mode 3 because it is likely that the UT air considered here contains an external mixture of aerosol particles: sulfuric acid aerosols formed in situ in the UT, and letovicite containing particles originating from mixing of air that has had contact with the surface. We use a fixed letovicite composition because, as stated in the last paragraph of the Introduction, we assume the ammoniated sulfates to be completely involatile. Furthermore, Lin and Tabazadeh (2002) note in their Summary that "In addition, binary ammoniated salts containing sulfate ions are most likely converted to letovicite (NH₄)₃H(SO₄)₂ in the upper troposphere, which can then persist in aerosol solutions that are saturated with respect to ice." If all of the modes contained ammonium ions, the results would naturally change. In that case the growth of modes 1 and 2 would be strenghtened compared to letovicite mode. This would increase the number of accumulation modes particles which is already too high, if the number of haze mode particles is kept in agreement with observations. Besides, like already stated in the Petzold et al paper, the accumulation mode particles behave as if they are H₂SO₄-H₂O droplets. Changing the composition could spoil that agreement.

Page 2: "Similar" will be corrected.

Figures 1 and 2 show how HNO₃ amplifies the hygroscopic growth of UT aerosols as a function of relative humidity for particles in different size classes. It is of course

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important to show how much the particle diameters can grow in order for the reader to be able to understand what is possible and what is not possible regarding the increases in accumulation mode and haze mode concentrations. We are not quite sure why Referee 2 thinks these simple figures are unclear, but we will try to improve both their description in the figure legende and their motivation in the main text. In figure 2 we only show 1 ppb because otherwise the Fig. would become very unclear. At 0 ppf of HNO₃, the deliquescence point of letovicite is at $S = 0.8$, so there's not much growth at the range shown. We believe that by comparing the 1 ppb -curves in Figs. 1 and 2, and looking at the 2 ppb curves in Fig. 1, the reader can get a good idea of how much the letovicite containing aerosol population would grow at 2 ppb of HNO₃. This will be clarified in the text.

Figures 3 and 4: Legends will be improved. Regarding letovicite composition, see above.

Figures 5 and 6: M11 etc. refer to different POLSTAR II flights, this will be clarified. The HNO₃ mixing ratios are different because what we are showing are the best fits to the observations that we can obtain from our model for each type of aerosol. In other words, for pure sulfuric acid, we need 1-2 ppb of HNO₃ to obtain a satisfactory fit. When letovicite is included, we only need 0.5-1 ppb. Thus, there is a factor of 2 difference in the HNO₃ levels needed to explain the observations in the two cases. We think that this is a significant difference. Fig. 1 of Krämer et al. shows that HNO₃ mixing ratios between 0.2-0.9 were measured during flight M3 (green stars in our Figs. 5 and 6) of POLSTAR II. Regarding origin of the air, we have now performed trajectory analysis, which indicates that the airmasses most probably have had contact with the surface, see our second response to Referee 1. We will clarify these issues in the text.

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- [] Lin, J. S. and Tabazadeh, A.: The effect of nitric acid uptake on the deliquescence and efflorescence of binary ammoniated salts in the upper troposphere, *Geophys. Res. Lett.*, 29, 126-1-4, 2002.
- [] Petzold, A., C. Hoell, C., Kärcher, B., Beuermann, J., Schiller, C., Ziereis, H., and Schlager, H.: In situ observations of aerosol properties above ice saturation in the polar tropopause region, *J. Geophys. Res.*, 105, 29387-29395, 2000

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