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Atmospheric Chemistry and Physics Discussions

Interactive comment on "In-situ observations and modeling of nitric acid-containing particles in a cirrus cloud formation region" *by* C. Voigt et al.

Anonymous Referee #2

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General Comments:

The paper presents measurements from a 150-sec segment of a flight of the Geophysica aircraft during TROCCINOX. An NOy instrument with a forward-pointing inlet is used to infer the HNO3 content of ice particles in what may be a region of cloud formation, though how this interpretation is established merits more attention. The HNO3 content, expressed as a molar ratio to H2O, is abnormally high, when compared to prior measurements. This is explained based on the short time since formation of the small ice particles, and is shown to be consistent with prior measurements of lower molar ratios, on the basis of the modeled growth of particles. That is, it is likely the particles form with high ratios, if formed from STS aerosols, and in normal growth to larger sizes the molar ratio is expected to decrease to values seen in prior measurements, which

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were in generally more aged clouds. This is an important topic relevant to the scope of ACP, and is in a meteorological situation not typical of prior publications on this subject. It is an interesting case study, and the authors provide valuable insights.

A fundamental concern I have about the paper is the assertion that the measurements were taken in a "cloud formation region" (this phrase is in title). I do not think this is demonstrated. Indeed (see specific comments below), one-third of the 150-sec time interval under study is subsaturated. These particles will be evaporating. I think the work is valuable and merits publication in some form, but more attention needs to be paid to the meteorological (dynamical) situation, and perhaps a more complicated interpretation is in order. At a minimum, it would seem prudent to separate the regions of RH$>$1.0 from those $<$1.0. Also, can any more information be brought to bear (models?, sounding?, satellites?) on the life cycle of the cloud to determine whether it was growing, or whether it was an short-lived phenomena, particles appearing only briefly, growing only to small sizes, then evaporating?

Specific Comments:

(1) I had occasional trouble understanding definitions of variables when first encountered, especially as related to whether the enhancement is included. For example, at the beginning of section 2, it should be made clear that NOy,t is not simply "particle plus gas phase NOy", it is "enhanced particle signal plus gas phase NOy."

(2) Also, the use of "particle" is not consistent. At the end of the 1st paragraph of section 2: "r denotes the particle radius and r_i the ice crystal radius", so here "particle" is being distinguished from "ice crystal" (so particles are STS aerosols?), yet in the next sentence there is mention of "particle velocity in the inlet" and here "particle" includes ice crystals. And beginning of section 3.1: "optically thin particle layer." Here particles are really ice.

(3) The authors may wish to add a reference to the recent work of Gamblin et al., JGR, allowing that NOy species other than HNO3 may be responsible for the signals seen

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on such NOy inlet/instrument combinations, esp. in the context of the comment, "We assume that 100% of NOy in particles is HNO3."

(4) Top of page 1853: "NOy,r denotes the remaining NOy,r=NOy,g + HNO3,a in the cirrus region, and HNO3,a denotes the HNO3 contained in liquid aerosol." Here, on first reading, it is not yet clear that HNO3,a is the instrument signal, with enhancement. I came to this conclusion after further reading, but on the first pass, it is not clear that that term includes the enhanced signal. Same comments apply to ice IWC equations.

(5) It seems rather uncertain to infer H2O_r from the above- and below-cloud regions. It seems the water content of the air could be very stratified. Indeed the reason the air forms in the layer it does, may simply be that this air was more humid. It may be a different air mass to start with. I realize the authors have no better choice, but I believe the uncertainty this entails could be expressed more strongly.

(6) "The lidar data suggest that the Geophysica ascended through the boundary region of a convective cloud." I do not see how this is established. Looking at Fig. 1, I do not see how one can tell where the edge is, given the lidar data. It seems the plane could have risen in the middle of a cloud system (of the type shown in the figure). How does one know that a similar lidar picture would not have been present to "the left" of the ascent in the figure. Maybe it was just not seen since the lidar is pointing down. Perhaps there is more information available. Pilot reports? The quoted sentence suggests this inference rests on the lidar data, but I do not follow the argument. Is there more to it?

(7) The layer is described in the abstract and elsewhere as being supersaturated. It is also said to have been sampled over the interval 36,300-36,450 sec. This is not really consistent, as supersaturation only occurs in about the first 100 seconds of the 150-sec interval. Part of it is supersaturated. Part of it is sub-saturated. It seems this could have severe implications for the interpretation. Is it really a good idea to lump this whole time interval together, and model the situation as a cloud of growing particles? It seems

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some would be growing, some evaporating. I understand the authors were motivated by lack of numbers (poor statistics), but this seems an unfavorable compromise. The authors should consider treating the first 100 sec separately from the final 50 sec. The whole 150-sec interval should not really be described as "slightly ice supersaturated." This may be true for average values, but actually some particles are in a sub-saturated environment.

(8) Pp. 1856-7: "which might be explained either by a reduced sampling efficiency of the FSSP during ascent..." In the absence of any supporting arguments, this seems very speculative. Why would the FSSP have a reduced sampling efficiency during ascent. Why not increased? Why different at all? Perhaps there is a well-known possible explanation, but I am unaware.

(9) "... our working hypothesis is that the Geophysica coincidentally probed a cirrus layer in its formation stage." It is indeed a tenuous cloud, but how is it known that it is not in the dissipating stage? Or both? First 100 sec may formation (RH$>$100%), final 50 sec may be decaying (RH$<$100%). This may be a working hypothesis, but it seems rather uncertain, and there are other possibilities. In this same vein, it may be misleading to suggest, as the modeling exercise does, that these ice particles will continue to grow into larger and more dilute (HNO3/H2O getting smaller) particles. This could be a very transient phenomena, particles forming briefly, and then evaporating. Is there really dynamical support for the maintenance of an ice cloud? And even the 100 sec period with RH$>$100% could also be dissipating. The individual particles would be expected to be growing at the time of measurement, but there could be mixing occurring at the same time leading to evaporation in the very near term.

(10) Section 3.2, 2nd paragraph: "This allows us to attribute HNO3,a to NOy,r as NOy,r = HNO3,a + NOy,g for the present analysis." Is not this equation always true? If so, what is being said. (That is, if so, the preceding is not required in order to allow it.)

(11) Top, p. 1861: "mu_a is reduced by a factor 0.4." This particular value seems totally

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arbitrary. Is there any basis for this? Some kind of justification is needed. Why not 0.9? Or 0.01? Is it covered in Karcher and Voigt? If so, perhaps at least a sentence here for what the justification is based on, even if reader needs to go to the other paper for more details (if they are there).

(12) P. 1861: "... HNO3 mass accommodation coefficient on ice alpha=0.3." I believe the literature is not consistent on the value for this parameter (e.g., Hudson et al.). The choice of this particular value merits some discussion, or at least a reference, if a definitive one exists.

(13) Section 4.2, first sentence: "... the trapping process in nascent ice particles." As noted above, some of the ice particles will be evaporating.

Technical corrections:

Typo: accomodation –> accommodation

Typo: greatfully –> gratefully

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 1849, 2007.