

Interactive comment on “Hygroscopic behavior of individual NaNO₃ particles” by M.-J. Lee et al.

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First of all, I thank the commenter for his valuable suggestions, which would certainly be helpful for the improvement of our manuscript.

Please let me reply to his comments one by one.

1. comment: The literature already shows that when a crystalline aerosol is present the deliquescence point is $\sim 75\%$. The literature also shows that efflorescence is unlikely above $\sim 6\%$ RH unless an inclusion is present (or heating or extensive drying is used). There is a discussion of these results in e.g. Gysel et al. EST 2002 and it is noteworthy this is the same humidity response that has been found for other particle types such as ammonium bisulfate and nitrate.

* reply: To my best knowledge, no paper has discussed about crystalline and generated

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NaNO₃ aerosol particles including the paper of “Gysel et al. EST 2002”. Our work is the first one which clearly relates the different hygroscopic behavior of powdery and generated NaNO₃ particles to the presence and absence of crystal germs.

2. comment: The authors essential describe the aerosol humidity response within their apparatus which is attributed largely to the presence of heterogeneous surfaces and/or contamination within the particles. While of interest to the users of this apparatus I have to stress THIS IS NOT relevant to the atmosphere (aerosols do not lay on such surfaces and foreign material, while present, is not correlated with what is found in their lab) - which is the purpose of the ACP journal. Furthermore, these results have already been shown in the literature dating back to 2002 (and before). The questions that need to be answered – to which I can find no response in this manuscript – (a) what is the atmospheric relevance of the results and (b) what determined here has not previously been shown in the literature?

* reply: First, I admit that substrates have more or less rough surface. I know that there is substrate effect depending on the type of substrates used, and we are working on this issue. However, if this surface would interfere with our current measurements, we would not see the aerosols without ERH as rough surface can act like a kind of crystal germ; we observed aerosol particles without ERH and powdery particles with ERH, indicating that TEM grid substrate is not the concern in this NaNO₃ system. If the presence of the surface would not allow the hygroscopic study as the comment implies, then all the previous other studies which used the substrates in ESEM, ETEM, optical microscopy, etc. should not be appropriate for the hygroscopic study. The commenter also mentioned about “contamination within the particles”, implying that particles we dealt with are contaminated, which is not the description based on the scientific reasoning. I do not agree with the commenter’s argument that our work is not relevant to the atmosphere just as the hygroscopic behavior of NaNO₃ particles on the substrate was studied instead of floating and analyzing them in the gaseous environment. There have been many hygroscopic studies published in atmospheric environmental journals

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by other people, which use particles collected on substrates. To my best knowledge, our work includes a lot of new observation. In our manuscript we described what we newly observed and what the others have observed. If some of “these results” are repeated and/or already-known ones, please let me know specifically.

3. comment: To my knowledge sodium nitrate aerosol may be created by the interaction of sea salt aerosol with polluted conditions. It is NOT, as the introduction suggests, generated by sea spray in an externally mixed state (i.e., it is internally mixed with the other sea salt components). The term ‘secondary’ in the introduction is not used correctly and suggests there is some process whereby NaNO_3 aerosol is generated from the gas phase (this is a primary aerosol and heterogeneous chemistry). This section needs to be re-written to clarify this.

* reply: The comment must be about the following paragraph in our manuscript, “When “genuine” (or fresh) sea-salt aerosols, which are emitted by the so-called bubble bursting or sea spray process, react with various nitrogen oxides, such as HNO_3 , N_2O_5 , NO_2 , and NO_3 , in the atmosphere, sea-salts such as NaNO_3 can be formed, resulting in chlorine loss (Gibson et al., 2006). As the physicochemical properties of secondary NaNO_3 particles are different from those of NaCl particles, . . .”. As the “sea-salts such as” and “secondary” words might mislead the reader, they will be deleted without any change of the context.

4. comment: The authors introduce the term ‘growth factor’ as a change in surface area. This is due to their optical technique which looks at a drop on a surface which resembles a hemisphere. However, there are serious issues with this. First, to my knowledge, GF is defined as a change in RADIUS, not surface area. This is the terminology from e.g. HTDMA instruments. Is there any precedence for this definition regarding surface area (SA)? Second, if GF is defined in this manner (SA) it can be confused by readers since the references in this paper use the radial definition. Third, the authors are viewing a projection of a hemisphere in two dimensions, not a true ‘surface area’. Is any correction made to truly derive surface area (if not this needs to

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be explicitly stated)? I firmly believe this is an incorrect use of an existing definition that was adapted to agree with this technique and needs to be corrected. At the very least a subscript 'surface area' needs to be included in the figures and clarification of if it is a projected SA or assumptions are made of the droplet shape.

* reply: Indeed, the comment is right. In our system, "two-dimensionally projected area" is the "growth factor" data we deal with. As the "growth factor" has a general definition, the data will be represented as "2D projected area ratio" in our revised manuscript.

4. comment: The terms 'generated' and 'powdery' are highly subjective. For example an aerosol powder is still 'generated' for use in the lab and a 'powder' can result when an aqueous salt is dried. Terms such as 'wet dispersed' and 'dry dispersed' would be more correctly used and in agreement with the previous literature.

* reply: Respecting the comment, 'nebulized from aqueous solution' and 'dry deposited' terms will be used in our revised version.

5. comment: Have the authors considered their grinding of the crystalline material introduced the heterogeneous nucleus?

* reply: To check the possibility of the contamination during the grinding process, we worked with powders without grinding. As the same observation for non-ground powder particles to that for the ground powders was made, i.e., the consistent DRHs and ERHs observed for all the un-ground particles, the grinding is not the concern.

6. comment: Reiterating the point that this paper is written as a microphysics experiment and atmospheric relevance is not addressed: The conclusions discuss the purity provided by the chemical company from which the material was purchased, not an atmospheric process. Even the interesting topic of heterogeneous efflorescence is essentially ignored as the authors don't know the source, size or composition of the inclusions that presumably cause the nucleation events.

* reply: As the company did not reveal its secret for making powders and its nature of

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trace crystal germs, we could only speculate that the crystal germs might be insoluble solids. The point is that all the people working with standard powders for hygroscopic measurements have used the similar quality of chemicals purchased from the companies, the hygroscopic behavior of NaNO_3 particles has been controversial, and we believe that now it is clear why it has been controversial.

7. comment: I note there are numerous grammatical errors that need to be addressed throughout this paper.

* reply: In the revision of the manuscript, grammatical errors will be carefully checked and corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 23203, 2011.

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