Reviewer comments for manuscript acp-2011-463: *Experimental study of the role of physicochemical surface processing on the IN ability of mineral dust particles.*

General Remarks

The above article presents data on ice nucleation behaviour of chemically, thermally and physically treated Arizona Test Dust particles (ATD). In addition, the ice nucleation behaviour is compared to that of pure ATD particles and further inter-comparisons are made between the ice nucleability of variously treated particles. In particular the paper focuses on immersion freezing results on single particles from continuous flow measurement techniques. The aerosol treatment is characterised and supported from various suite of measurements, in particular from the Aerosol Mass Spectrometry instruments. The work and results presented herein are timely, of interest to the readers of Atmospheric Chemistry and Physics and are important for understanding the effect of chemical ageing of mineral dust ice nuclei in the atmosphere, especially under mixed phase cloud conditions.

I recommend this paper for publication after the comments below have been addressed.

Specific Remarks:

Abstract:

Line 2: 'influences' should be 'influence'

At the end of the abstract, the authors state the results 'are certainly very interesting'. While this may be obvious to a reader whose expertise is in ice nucleation or aerosol-cloud interactions but for the purpose of broad readership I suggest adding a brief statement here (along the lines of atmospheric implications), to suggest why the results are interesting!

Page 18560

Line 9: The references given here I don't think represent the articles that actually did the science that showed that ice particles (through mineral dust nucleation) can alter microphysical and dynamical properties of clouds, influence precipitation and cloud lifetime. The references given here also used the above as motivation for their studies. I expect the references you want to add here would likely be modelling or field observations of earlier studies that investigate the formation of precipitation (via the ice phase), collision-coalescence processes and albedo effects from liquid and/or ice clouds. I suggest, replacing with suitable references or cite a review article with the suitable references therein.

Line 15: insert comma after 'processes'

Page 18561

Line 14-16: the sentence 'With LACIS..... .....in each droplet' is poorly structured. I suggest splitting the sentence into two - something to the effect of:

'In the current work, LACIS was used to investigate the influence of quasi mono-disperse dust particles on immersion freezing. In particular, we note that in this method, only one particle is immersed in a single droplet'.

Line 21: insert 'ATD' between 'coated' and 'particles'

Line 25: replace 'Again' with 'Similar to FROST 1,'

Line 26: should read: '..submicron particles were considered for coating with different amounts...'i.e. continue the sentence.

Line 28: insert 'thus' after 'modifications'

Line 29: delete the parentheses and text within and replace text with 'compared to -34°C for FROST 1).

Page 18562

Line 1: delete the portion of text in parentheses (see comment above). Replace 'Also' with 'In addition'

Line 5: insert 'sulphuric acid' before 'coated'

Line 8-14: Here the authors say that their work is related to Sullivan et al (2010) and Reitz et al (2011). In addition to describing what these studies are about, perhaps you can also add in a few sentences and describe what the most important findings/results from these papers were, or at least the results that most pertain to the current study.

Line 26: replace 'in the course of' with 'during'

Page 18563

Line 13: delete '....of all...'

Line 21-25: Can you provide an amount or concentration of how much NH_3 gas was added to the sample line. Was all of this NH_3 consumed by the sulphuric acid coatings? Or did some of this NH_3 contaminate the lines/tubing or get sampled by the mass spectrometers?

Page 18564

Line 1-2: can you give a number of multiply charged particles that could have been sampled by LACIS and the CFDC instruments? Is the doubly charged particle concentration high enough so that perhaps the first freezing branch (low activated fractions) of pure ATD particles (at warmer temperatures), could be explained by the larger particles from multiple charges being active?

Page 18565

Line 20: ‘6 to 7’ should be 6 and 7’

Page 18566

Line 5: here the authors presumably meant that after the walls are cooled down to -40°C, ice in the inner walls of LACIS are generated by passing saturated air through the flow tube? If so, one sentence about how the ice coating is achieved (just a brief mention) is warranted here.

Line 12: replace ‘at’ with ‘coating in’ and ‘tube’ with ‘walls’

Page 18567

Line 20: replace ‘the latter’ with ‘ammonium sulphate’ given that the prior sentence is quite long with multiple objects.

Line 20 onwards: The authors mention the use of compressed air here. I am curious as to what the NH_3 signature is in the compressed air, any indications from background spectra? If the neutralisation of SA by NH_3 under dry conditions is very slow, then how much NH_3 is present in the compressed air such that it should still lead to ammonium sulphate being present on the dry SA processed particles?

Doesn’t compressed air also have trace contaminants (organics), that could also condense on the particles when they are cooled down to -30°C or so in LACIS? Do you think this influences your results, for example the pure ATD results? i.e. how do the pure ATD results compare to experiments done in high purity air or nitrogen?

If there were an effect, then it would be somewhat systematic across all the experiments. Is there any signature of organics from the coated aerosol particle mass spectra?

Page 18568

Line 1: replace ‘most presumably’ with ‘presumably mostly’

Line 11 onwards: I don’t understand how for ATD+SA(70°C)+WB+NH_3 and ATD+SA(70°C)+WB the ammonia signature is similar? Shouldn’t the ammonia signature in the former be significantly greater? Is it possible that the reaction of humidified SA with ATD is so fast that by the time the particles ‘see’ the ammonia, the acid has already been neutralised in forming metal sulphates? From Figure 1 and the text description it looks like the ammonia is added in sequence after the SA coating region.

Line 13-14: parentheses should read ‘(sulphate mass of between 2600 – 2700 ag particle^{-1}).’

Is it possible to confirm any of the coating composition data with supporting data from the A-TOF-MS that was also used in this study as mentioned in the experimental section (see later comment)?

Page 18570

Line 4: replace ‘any’ with ‘all’

Line 11: insert ‘of’ after ‘Regardless’

Page 18571

Line 20: insert ‘at -30°C’ after ‘too small’

Page 18572

Line 21: delete ‘an’

Line 24: Here the authors provide information Sullivan et al (2010) from the A-TOF-MS about how many particles remained uncoated during these experiments. However, is it possible to give more information about the coatings in support of the C-TOF-AMS results (see earlier comment)?

Page 18573

Line 8: replace ‘Now we look at’ with ‘Referring to the’ and also replace ‘i.e.’with ‘for’

Line 9: Insert a comma after ‘...ATD + SA(85C)’ and delete ‘For these cases’

Line 10-17: This explanation is suitable i.e. that the SA destroys the active sites for the first freezing branch and could be producing more efficient sites than previously existed for the second freezing branch. However, is there a possible explanation for why more effective sites are not produced on the particles that activate in the first freezing branch?

Related to this and to the comment about multiply charged particles: Is it possible that the first freezing branch (small activated fractions) is due the small number of multiply charged particles (larger than 300nm) with larger surface areas and therefore providing more area for surface reaction with SA (which destroys active sites), which would not be the case for the smaller particles (mode of 300 nm) where the surface area is smaller and therefore perhaps a smaller rate of reaction with the surface and thus an increase in the second freezing branch only with the shift towards colder temperatures (T) closer to homogeneous freezing Ts?

Page 18574

Line 12: replace ‘within’ with ‘for’

Line 12-13: insert a comma after ‘ammonia’

Line 20-21: insert a comma after ‘substance’

Line 24: ‘...should recover to its condition...’ Which condition are the authors referring to here? I find this sentence a little confusing. Some more clarity here would be nice. Also, I thought NH_3 was only added after passing through the water bath? So even in the case ATD+SA+WB+NH_3, the decrease in IN ability comes from the reaction of ATD+SA+WB and therefore I don’t understand the clause ‘prior to water vapour processing’ in line 24.

Page 18575

Line 1: replace ‘was’ with ‘were’

Line 17-18: replace ‘large IN ability reduction’ with ‘large reduction in IN ability’

Page 18576

Line 25: insert ‘we infer from C-TOF-AMS and ice nucleation measurements that’ after ‘...sum up’. I recommend this statement since the conclusions are reached by observing fragments of the reaction products and further supported by the change in ice nucleation properties.

Line 29: replace ‘highly active surface features ability’ with ‘IN ability of the highly active particles’

Page 18577

Line 2-3: should read ‘nucleating substances and/or surface features which display higher ice nucleation potential might have formed. In general...’

Line 11: insert ‘sulphuric acid’ after ‘water vapour’

Line 12: insert ‘to’ after ‘and’

Line 22: ‘completely vanished’ instead of ‘vanished completely’

Line 28: replace ‘of’ with ‘in’

Page 18578

Line 1: insert comma after parentheses and delete comma after ‘speculate’

Line 2: replace ‘having’ with ‘that have’

Line 1-2: There is no indication in the paper that the metal cations were also detected in the mass spectrometry of the aerosol particles. So I would rephrase this sentence, to say that the sulphates detected which are likely from metals and possibly NH_4^+ rather than saying metal sulphates were detected.

Line 7-9: Delete last sentence of this paragraph and replace with

‘The exposure of sulphuric acid coated particles to the water bath and ammonia seems to be of secondary importance’

I feel like it is important to add the water bath here to be clear that ammonia was only added after exposing the SA coated particles to the water vapour.

Line 12-14: the sentence that spans these lines is confusing even though I think I know what the authors are trying to say. It is important to be clear here (since it is in the conclusion) and indicate that the water vapour treated particles had SA on them. I suggest something to the effect of:

‘Significant reduction in IN ability was observed for SA-coated ATD particles that were passed through the thermodenuder, similar to the IN ability of SA-WB treated ATD particles.’

Line 15: replace ‘than the’ with ‘compared to those from’

Line 16: ‘enhance’ should be ‘enhanced’

Line 23: replace ‘processings’ with ‘treatment’

Final paragraph: It would be nice if the authors suggested why their results are interesting from an atmospheric implication perspective. Is this study relevant to the atmospheric ice nucleation because most mineral aerosol acting as IN found in the atmosphere is internally mixed with sulphuric acid or ammonium sulphate etc?

Figures:

Figure 1

Would it be possible to include in the caption what ‘other instruments’ are, relevant to the current study? In the main body of the paper, many instruments are mentioned, but perhaps those most pertinent to the data from this study can be included in the caption.

Shouldn’t there be a CPC downstream of the WELAS counter? I think this is missing from the schematic.

Figure 3

The dark yellow triangles appear to be quite close in colour to the green ones. In panel (a) the difference is colour is clear, but in panel (b) where unfilled symbols are used, it is a little harder on the eye. Perhaps use another colour other than dark yellow.

Figure 4

I think the order in which the figures are arranged is really good for easy comparison. Would it be possible to put a label on each chart with the respective particle type/treatment? If this is done, then you can also delete the panel descriptions from the text.

Figure 5.

This is a nice figure comparing CFDC and LACIS results. However, after reading the experimental section I was expecting results from FINCH and PINC as well, especially PINC, because it is also a laminar flow technique and almost identical to the CFDC. It would be nice to see results from this chamber. Or perhaps the authors can refer readers to where these other data are/will be published.