MS NO. acp-2015-385

We are grateful for Dr. Tang's comments. Those comments are all valuable and helpful for improving our paper. We answered the comments carefully and have made corrections in the submitted manuscript. The corrections and the responses are as following:

1. I fully agree with the comments made by ref. 1. Especially, nitrate has not been detected but inferred. Authors should provide a convincing explanation why nitrate has not been detected by any of these techniques they used, and justify how nitrate was inferred from their measurement.

Response 1: We appreciated Dr. Tang's comments. We made the new experiments after your comments and provided the more strong evidences. Please see our answers and new data in the response 1 of the reviewer#1's comments.

2. Page 16724, line 10-12: it is claimed that because aged SSA particles are more spherical compared to fresh ones, aged SSA particles tend to be droplets. I disagree with it. Although the conclusion can be true, the observation that aged particles are more spherical is only related to crystallization processes during dehumidification, and is not necessarily related to these phases at high RH.

Response 2: Yes, we fully agree with the reviewer. Here we only describe morphology of dry particles on the substrate. At high RH, they all become spherical shape in the atmosphere after the DRH.

We have revised the text (Page 16724, line 10-12) as follows:

"The fully aged SSA (Fig. 4) on the substrate become more round compared with fresh SSA (Fig. 2) and partially aged SSA (Fig. 3)."

3. Page 16727, line 1-3: I am not convinced that $NaNO_3$ coating totally determines the hygroscopic and optical properties of aged SSA particles. At most the measurements may suggest that aged SSA particles are more likely to be droplets in the ambient air, compared to fresh SSA particles. To make such a strong claim, the authors are

required to present data of hygroscopic and optical properties for both fresh and aged SSA particles. In addition, I think even fresh SSA particles are likely to be droplets for ambient RH of 56-94%. Though pure NaCl has a DRH of 77%, its ERH is much lower. By the way, even fresh SSA particles should not be pure NaCl, but containing other salts which are contained in sea water.

Response 3: We agree with your comments. We could not presume the particle phase in real atmosphere based on their compositions on the substrate. However, we did one hygroscopic experiment to understand how particle compositions change their hygroscopic properties.

We added new hygroscopic experimental data for three typical SSA in supporting information (Fig. S4). Hygroscopic properties of individual SSA in Figure S4 show fresh, partially aged, and fully aged SSA start to grow at 69%, 65%, and 60%, respectively. The results show that nitrate coatings in SSA can change hygroscopic properties of particle surfaces. In addition, we carefully made some corrections about the tone.

We have revised the text (Page 16716, line 17-19) as follows:

"These new findings indicate that this mixture of organic matter and $NaNO_3$ on particle surfaces likely determines their hygroscopic and optical properties."

We have revised the text (Page 16726, line 15-18) as follows:

"In the partially aged particles, the new coating containing $MgSO_4$, $Mg(NO_3)_2$, and $NaNO_3$ (Fig. 3) likely remained as a supersaturated liquid coating on the surface of the ambient SSA in the arctic area (Zhao et al., 2006;Li et al., 2008;Woods et al., 2010)."

We have revised the text (Page 16727, line 1-3) as follows:

"Hygroscopic experiments of individual SSA exhibit that the surfaces of partially and fully aged SSA have earlier deliquescence than that of fresh SSA based on their hygroscopic growth on the substrate. The results indicate that the nitrate coatings likely influence particle surface deliquescence (Fig. S4)."

We have revised the text (Page 16727, line 5-8) as follows:

"As a result, some partially and fully aged Arctic SSA had liquid surfaces in

ambient air (56~94% RH) and that this liquid amounted to certain amounts of water in these aerosol particles."

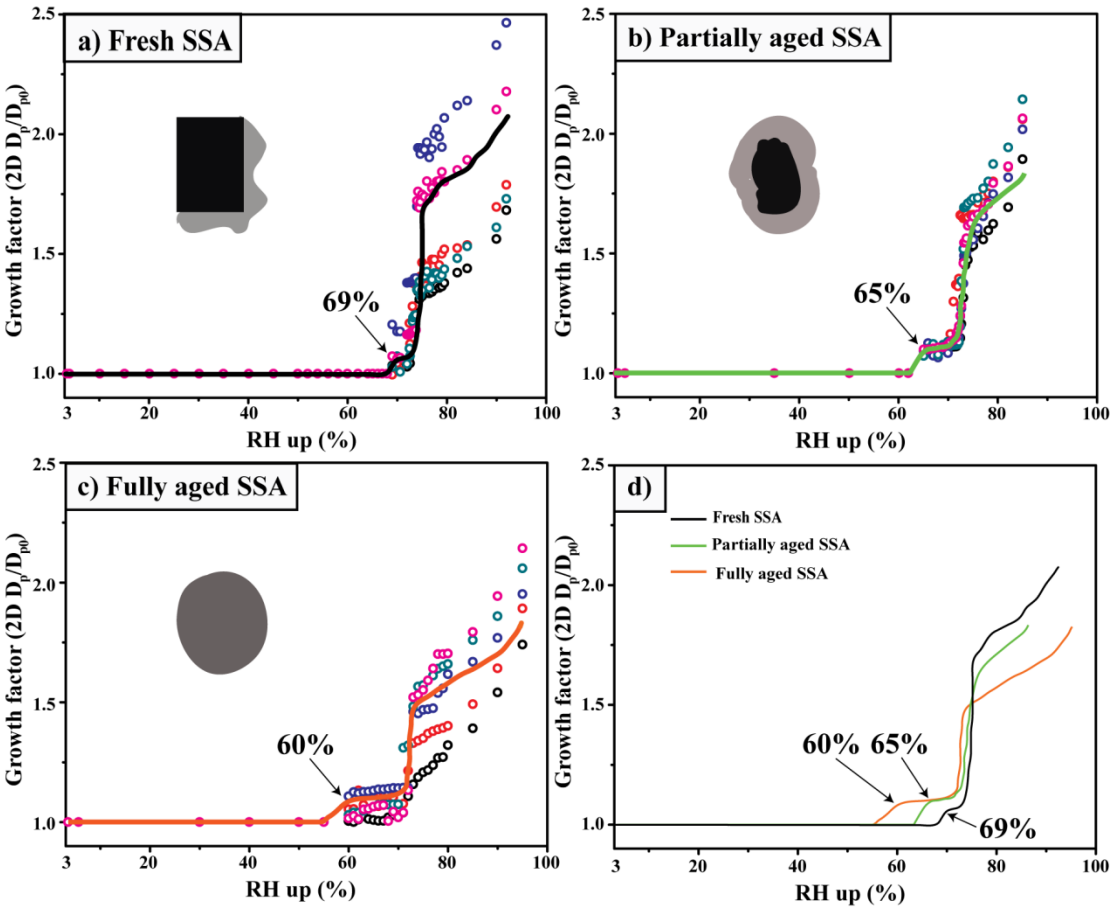

Figure S4. Deliquescence process of fresh SSA (a), partially aged SSA (b), and fully aged SSA (c) with different colors for different particles. The lines show the hygroscopic growth of individual SSA. The relative humidity for three typical SSA is 3-92%, 3-85%, and 3-95%, respectively.

4. Page 16727, line 19-24: if surface reactions produce Cl_2, the photolysis of Cl_2 will produce Cl atoms, further enhancing the photochemical reactivity. I am not sure why it will reduce the overall photochemical reactivity. More explanations should be provided.

Response 4: These reactions are beyond our study. We carefully modified it based on the references (De Haan et al., 1999;Knipping et al., 2000;von Glasow, 2008a;Thornton et al., 2010).

We have revised the text in section 4.2 as follows:

"On the other hand, these surface reactions of SSA could affect the photochemical reactivity in the Arctic air because they release gaseous halogen species (De Haan et al., 1999;Knipping et al., 2000;von Glasow, 2008b;Thornton et al., 2010)."

5. Page 16727, line 17-19: one of the four reactions will produce $ClNO_2$; instead, it is formed in the reaction of N_2O_5 with Cl^-.

Response 5: The reaction is beyond our current study. We deleted this sentence.

6. Page 16729, line 2-3: what the author found is that organic compounds are coated on aged SSA particles, and I am not sure that it is equal to phase separation used in atmospheric chemistry community. Since the editor is an expert in this field, I will leave the decision to the editor.

Response 6: From $^{12}C^-$ line scan, we did find $^{12}C^-$ signal higher in outer layer. We carefully compare it with laboratory experiment published in JACS (Ault et al., 2013) and field experiment published in JGR (Laskin et al., 2012).

7. Figure 6: I feel "volume equivalent diameter" is more proper than "equivalent spherical diameter".

Response 7: We have revised "equivalent volume diameter" to replace "equivalent spherical diameter" in the article and figures.

8. Page 16722, line 18: "don't experience" should be "have not experienced". Although the paper is not difficult to follow, it will benefit a lot if it can be revised and edited by a native speaker.

Response 8: We have revised it. The native speaker helped to smooth the context.

"The fresh SSA refer to particles which have not experienced any atmospheric chemical modification after emission."

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