

Dear Dr. Shiraiwa,

We are grateful to accept our paper.

Based on two reviewers' comments, we have revised the manuscript again. We addressed all comments as below. Our responses have been marked by red color. We are looking forward that the revised manuscript can be finally accepted and published in ACP.

Correspondence and phone calls about the paper should be directed to Weijun Li at the following address, phone, and e-mail address:

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Best regards,

Sincerely yours,

Weijun Li on behalf of the coauthors

We appreciate the reviewer's comments. Those comments are all valuable and helpful for improving our paper. We have made corrections in the submitted manuscript and addressed the comments carefully by a detailed point-by-point response as following:

**1. Page 7 line 15.**

This equation (1) still seems strange to me. Why not  $A=\pi r^2$  instead of  $A=4/3*\pi r^2$ ? Please check it again.

**Response 1:** We have revised equation (1) (Page7, Line 15) as follow:

$$A = \pi r^2 = \pi \times \left(\frac{d}{2}\right)^2 = \frac{\pi d^2}{4} \rightarrow d = \sqrt{\frac{4A}{\pi}} \quad (1)$$

**2. Fig. 5**

(1) Please specify the unit (wt%?).

(2) The distribution shows a good classification and is interesting. Could you explain why partially aged SSA have, in general, more oxygen than fully aged SSA?

**Response 2:**

(1) Triangular diagram of Na-Cl-O shows EDX data of elemental composition of the three typical SSA particles. We use the percentage for Na-Cl-O, therefore, there is no unit.

(2) For partially aged SSA, the coating mainly contained the certain amounts of  $\text{CaSO}_4$ ,  $\text{MgSO}_4$ , and  $\text{Mg}(\text{NO}_3)_2$  (e.g., Fig. 3), beside  $\text{Na}_2\text{SO}_4$  and  $\text{NaNO}_3$ . Therefore, O/Na ratio is higher in partially aged SSA than the fully aged SSA.

**3. Fig. 8**

It looks the particle in Panel (a) and that in (b) are different. The positions of Mg-rich rim and  $\text{CaSO}_4$  aggregate is upside down. The shape of  $\text{CaSO}_4$  seems different between (a) and (b). Please check if the TEM image (a) is correct one.

**Response 3:** The TEM image is correct: (a) a bright-field TEM image and (b) a

dark-field TEM image. The two TEM images were taken in different mode in the same TEM. In a word, the TEM image (a) is the mirror image of (b). Therefore, these two images seemly look different.

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We appreciate Dr. Tang's comments. Those comments are all valuable and helpful for improving our paper. We have made corrections in the submitted manuscript and addressed the comments carefully by a detailed point-by-point response as following:

1. Page 2, line 20: "absording" should be "adsording".

**Response 1:** We have revised the text (Page 2, line 20) in accordance with the requirement.

2. Page 3, line 16: "one solar absorber" should be "one solar radiation absorber".

**Response 2:** We have revised the text (Page 3, line 16) in accordance with the requirement.

3. Please cite this review paper in heterogeneous reactions: "Ammann, M., Cox, R. A., Crowley, J. N., Jenkin, M. E., Mellouki, A., Rossi, M. J., Troe, J., and Wallington, T. J.: Evaluated kinetic and photochemical data for atmospheric chemistry: Volume VI - heterogeneous reactions with liquid substrates, Atmos. Chem. Phys., 12, 8045-8228, 2013."

**Response 3:** We have cited this review paper of "(Ammann et al., 2013)" in the text (Page 3, line 22).

4. Page 7, line 16: "3" should be in the top-left of the last term.

**Response 4:** We have revised the equation (Page 7, line 16) as follows:

$$V = \frac{4}{3}\pi r^3 = \frac{4}{3} \times \frac{\pi D^3}{8} = \frac{\pi D^3}{6} \rightarrow D = \sqrt[3]{\frac{6V}{\pi}} \quad (2)$$

5. I feel a few important references are missing here, including:

(1) Osthoff, H. D., Roberts, J. M., Ravishankara, A. R., Williams, E. J., Lerner, B. M., Sommariva, R., Bates, T. S., Coffman, D., Quinn, P. K., Dibb, J. E., Stark, H.,

Burkholder, J. B., Talukdar, R. K., Meagher, J., Fehsenfeld, F. C., and Brown, S. S.: High levels of nitryl chloride in the polluted subtropical marine boundary layer, *Nature Geosci.*, 1, 324-328, 10.1038/ngeo177, 2008.

(2) Phillips, G. J., Tang, M. J., Thieser, J., Brickwedde, B., Schuster, G., Bohn, B., Lelieveld, J., and Crowley, J. N.: Significant concentrations of nitryl chloride observed in rural continental Europe associated with the influence of sea salt chloride and anthropogenic emissions, *Geophys. Res. Lett.*, 39, L10811, 10.1029/2012gl051912, 2012.

**Response 5:** We have cited the references of “(Osthoff et al., 2008;Phillips et al., 2012)” in the text (Page 15, lines 10 and 11).

6. Figure 5, “SSA” should be “SSA particles”.

**Response 6:** We have revised the text (Page 28, Fig. 5) in accordance with the requirement.

7. Figure 6, “other one” should be “the other one”.

**Response 7:** We have revised the text (Page 29, Fig. 6) in accordance with the requirement.

8. Figure 6, “showed” should be “show”.

**Response 8:** We have revised the text (Page 29, Fig. 6) in accordance with the requirement.

## References

Ammann, M., Cox, R. A., Crowley, J. N., Jenkin, M. E., Mellouki, A., Rossi, M. J., Troe, J., and Wallington, T. J.: Evaluated kinetic and photochemical data for atmospheric chemistry: volume VI – heterogeneous reactions with liquid substrates, *Atmos. Chem. Phys.*, 13, 8045-8228, doi:10.5194/acp-13-8045-2013, 2013.

Osthoff, H. D., Roberts, J. M., Ravishankara, A. R., Williams, E. J., Lerner, B. M., Sommariva, R., Bates, T. S., Coffman, D., Quinn, P. K., Dibb, J. E., Stark, H., Burkholder, J. B., Talukdar, R. K., Meagher, J., Fehsenfeld, F. C., and Brown, S. S.: High levels of nitryl chloride in the polluted subtropical marine boundary layer, *Nature Geosci.*, 1, 324-328, doi:10.1038/ngeo177, 2008.

Phillips, G., Tang, M., Thieser, J., Brickwedde, B., Schuster, G., Bohn, B., Lelieveld, J., and Crowley, J.: Significant concentrations of nitryl chloride observed in rural continental Europe associated with the influence of sea salt chloride and anthropogenic emissions, *Geophys. Res. Lett.*, 39, 2012.