Supplement of

Impact of organic acids on chloride depletion of inland transported sea spray aerosols

Bojiang Su et al.

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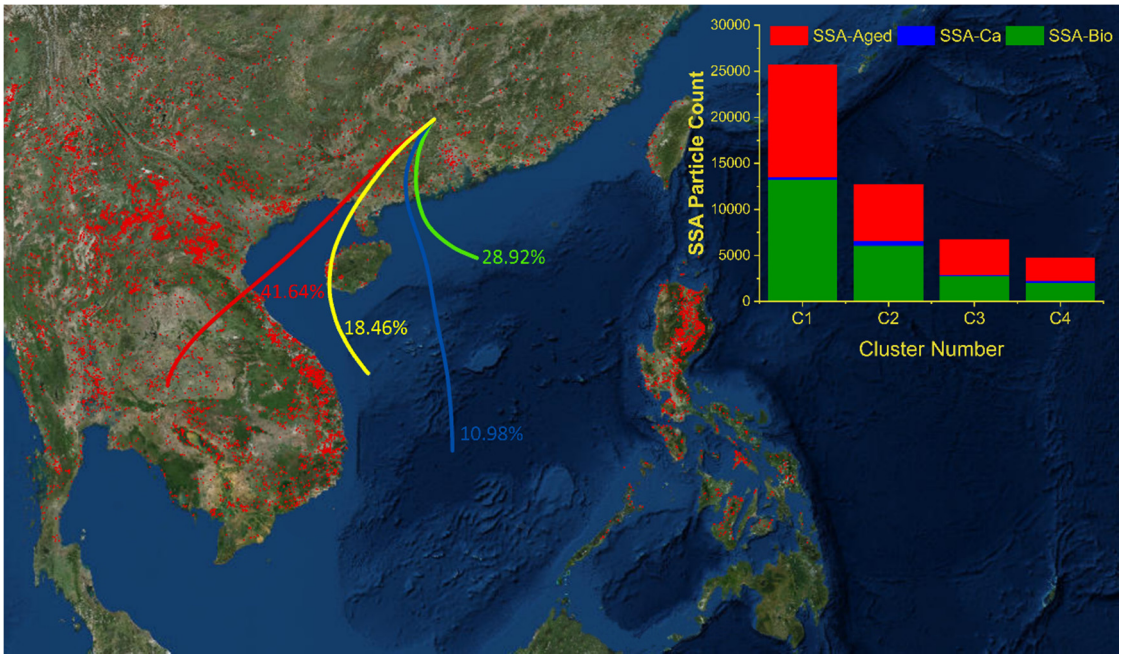
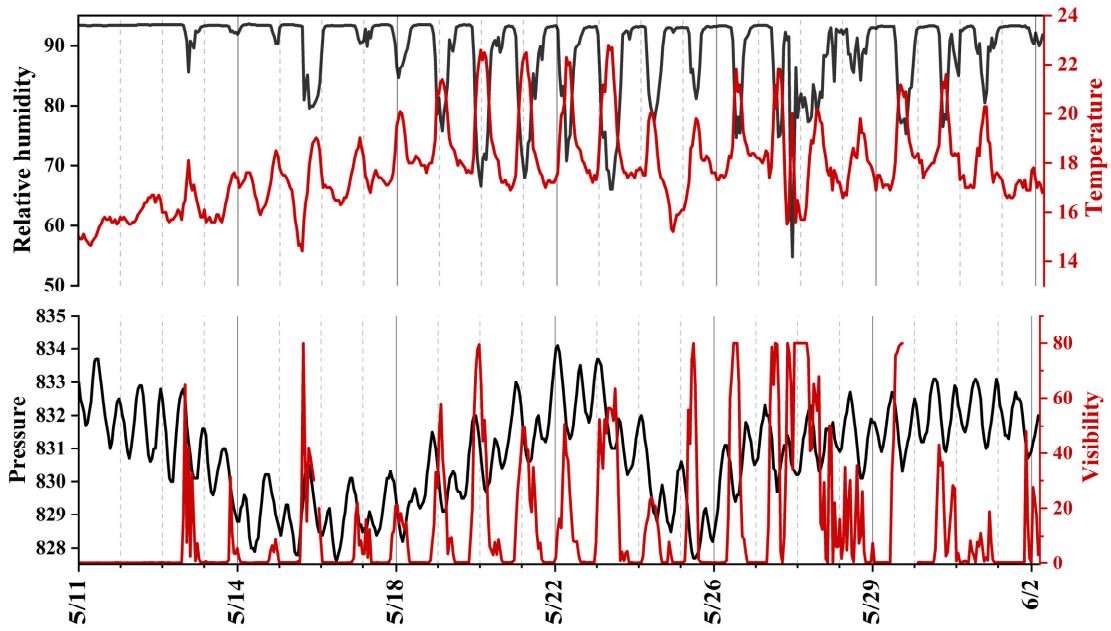


Figure S1. Quantitative distributions of SSA particles associated with clustered 72h back trajectories of air masses at 1800m above the ground during the sampling period (from 11 May to 3 June 2018). Four major cluster trajectories of air mass (namely C1, C2, C3, C4) are calculated by Meteoinfo (Wang, 2014) and plotted by Arcgis (Esri, Environmental Systems Research Institute, Inc.).



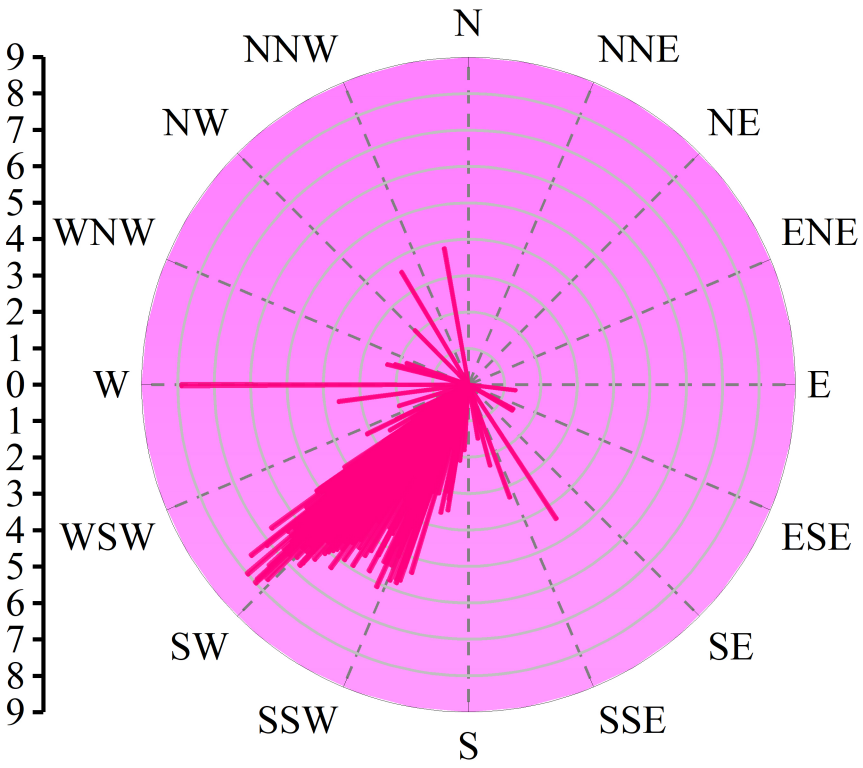


Fig S2. The hourly average variations in meteorological conditions and Wind Rose.

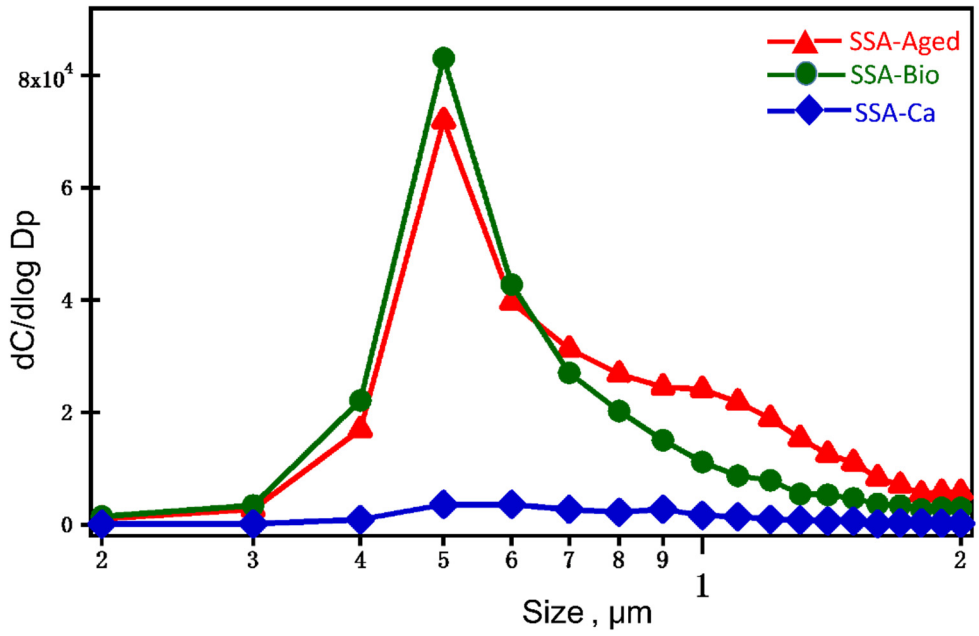


Figure S3. Unscaled size-resolved number distributions of major types of SSA particles during sampling period. SSA-Aged and SSA-Bio have similar trend in size range 0.2-2 µm, with approximately 70% of the particles concentrated in size range of 0.4-0.7 µm and peaked at size of 0.5 µm, suggesting that the both particle types may undergo the similar atmospheric chemical process. Such results indicate that SSA particles were aged during the inland transportation and exhibited in fine particles.

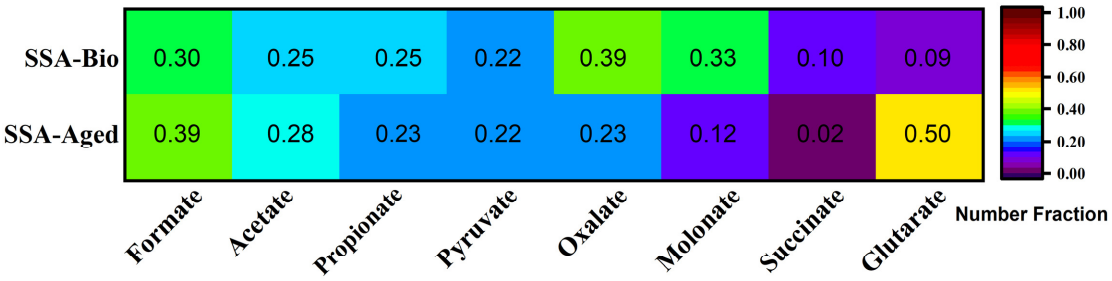


Figure S4. Number fractions (NFs) of all detected organic acids in SSA-Aged and SSA-Bio, which included formate (*m/z* -45), acetate (*m/z* -59), propionate (*m/z* -73), pyruvate (*m/z* -87), oxalate (*m/z* -89), molonate (*m/z* -103), succinate (*m/z* -117) and glutarate (*m/z* -131).

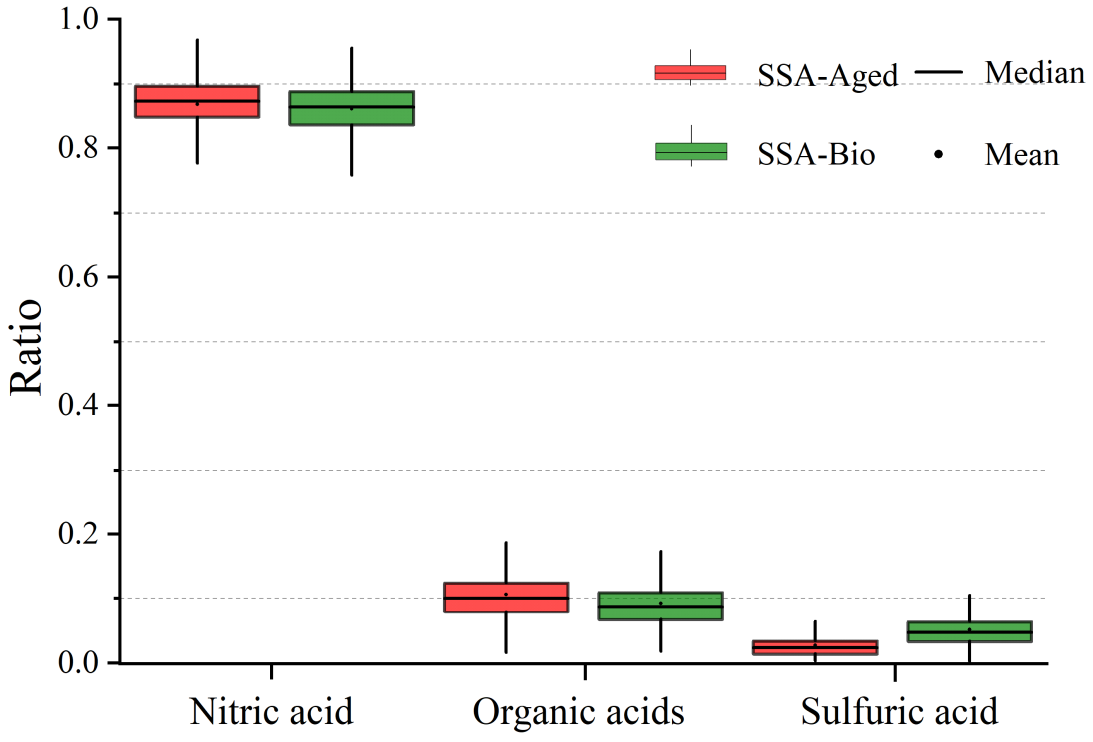


Figure S5. The relative contribution of different acids (nitric acid, sulfuric acid, and organic acids) to the chloride depletion of SSA particles. The ratio refers to the relative peak area (RPA) ratio (acids / (sulfate + nitrate + organic acids)). Sulfate, nitrate and organic acids refer to peaks at *m/z* -97, *m/z* -46 and -62, and the mentioned organic acids in Fig. S4., respectively.

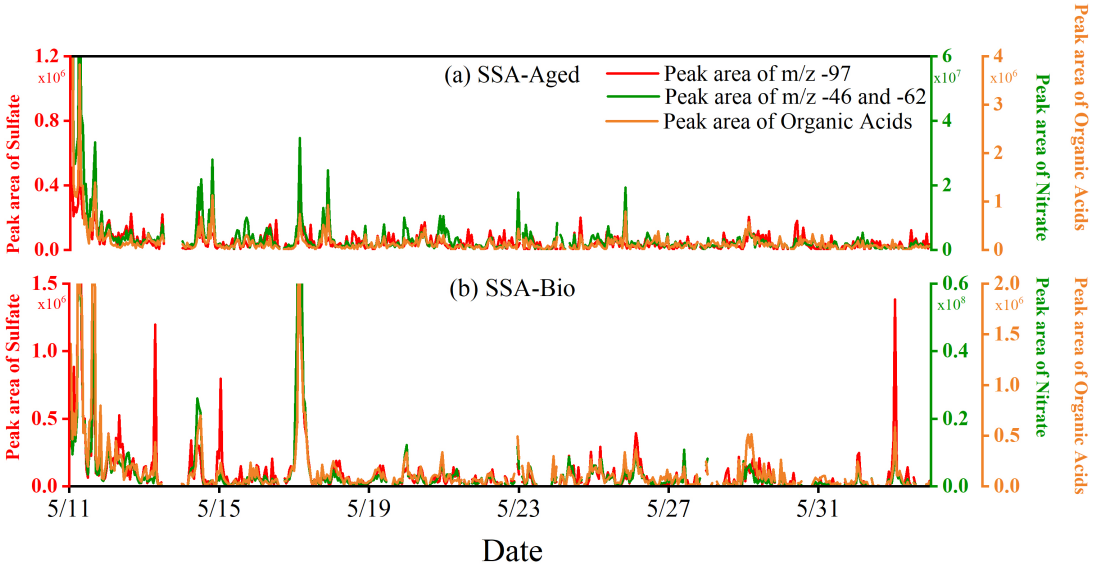


Figure S6. Hourly variations in the peak areas of sulfate (*m/z* -97), nitrate (*m/z* -46 and -62) and organic acids (sum of the mentioned organic acids in Fig. S4) in SSA-Aged and SSA-Bio during whole sampling period.

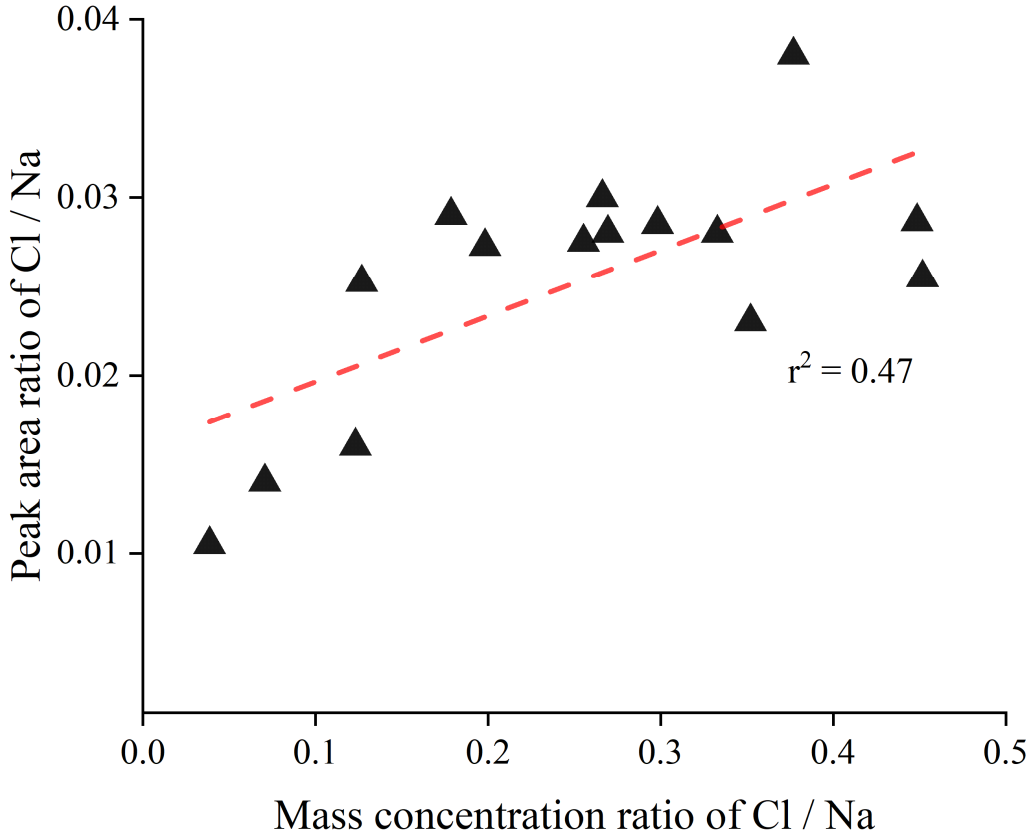


Fig S7. Correlation analysis of the both assessment methods of chloride depletion. These data were conducted significance test and $p < 0.01$. We obtained 22 set of data (detailed in Table S1), and correlation analysis was carried out on 15 set of data points due to the rest of extremum.

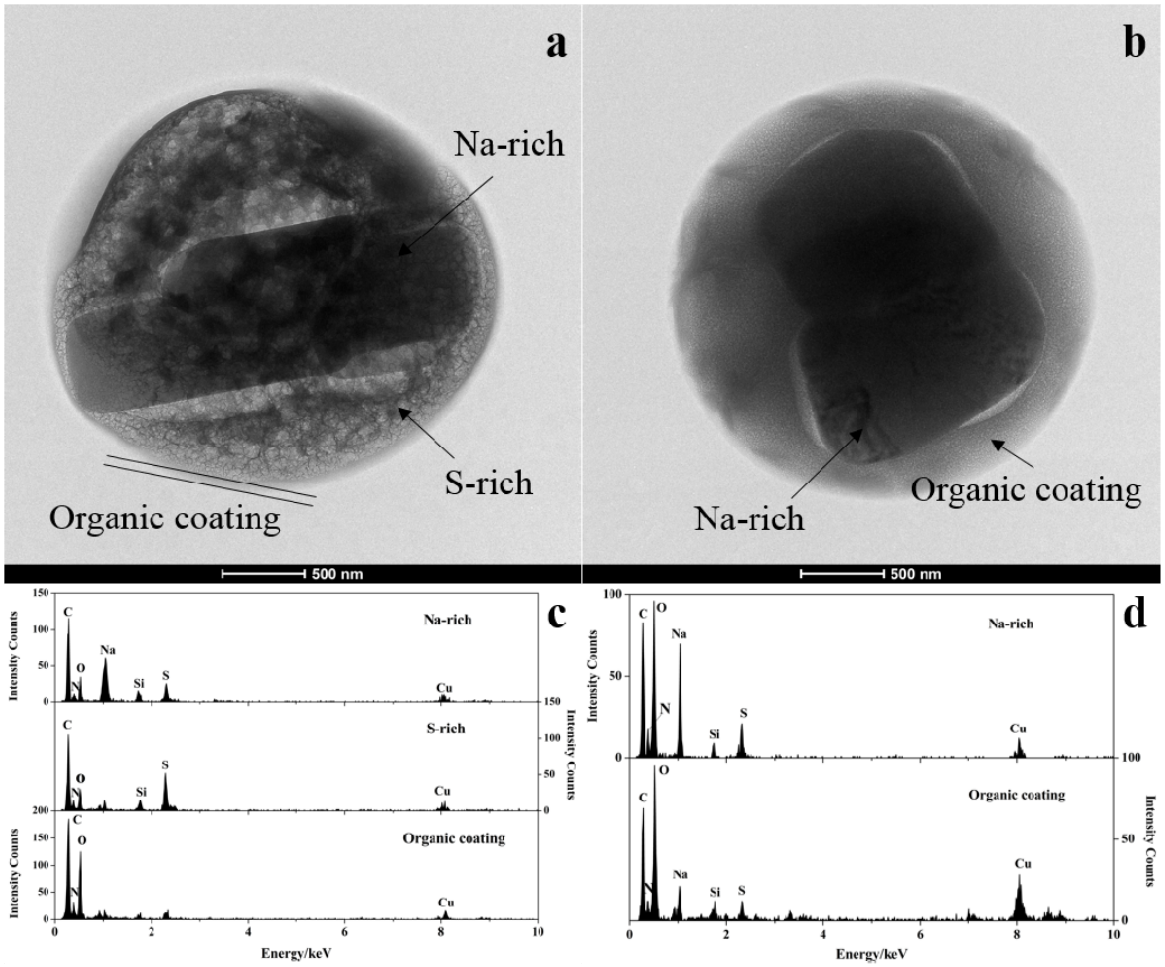


Figure S8. Transmission electron microscopy (TEM) images and energy dispersive X-ray (EDX) spectra of individual particles with coating of various thickness. These particles contained abundant Na and kept the crystalline phase with its irregular shape, which were regarded as SSA particles (Chi et al., 2015). High energy electron beams by TEM / EDS could cause the loss of volatile components, such as nitrate. This typical transparent coating with high C and O indicated a possible organic coating, and Na-rich core completely surrounded by the organic coating.

Sampling set name	Sampling dates	Duration (min)	Na^+	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	Cl depletion
Cloud Water 1	May 11	480	0.24	0.01	0.16	1.26	1.81	0.64
Cloud Water 2	May 26	150	1.31	0.74	0.58	5.59	9.32	0.65
Cloud Water 3	May 26	140	1.73	0.98	0.56	3.90	6.48	0.74
Cloud Water 4	May 26	180	5.24	1.80	0.99	6.41	9.65	0.87
Cloud Water 5	May 27	214	1.98	1.21	1.08	9.33	13.84	0.55
Cloud Water 6	May 30	270	1.85	1.35	0.53	8.57	13.08	0.73
Cloud Water 7	May 30	305	1.88	1.07	0.79	11.14	16.95	0.67
Cloud Water 8	May 30	515	1.68	1.22	0.83	11.59	19.32	0.55
Cloud Water 9	Jun 1	225	1.07	0.79	0.31	5.57	5.71	0.73
Cloud Water 10	Jun 2	230	0.99	0.42	0.05	2.10	3.84	0.96
Cloud Water 11	Jun 2	141	1.63	0.59	0.30	3.84	5.64	0.88
Cloud Water 12	Jun 2	134	1.44	0.46	0.15	1.85	2.63	0.93
Cloud Water 13	Jun 2	203	3.79	1.03	0.35	3.85	5.80	0.94
$PM_{2.5}$ 1	May 14-15	1440	1.12	1.04	0.38	1.05	2.60	0.62
$PM_{2.5}$ 2	May 18-19	1406	1.46	0.47	0.40	1.80	4.75	0.82
$PM_{2.5}$ 3	May 19-20	1423	0.45	0.22	0.07	2.07	5.09	0.88
$PM_{2.5}$ 4	May 20-21	1366	0.70	0.22	0.21	3.22	4.70	0.80
$PM_{2.5}$ 5	May 21-22	1431	0.48	0.18	0.13	2.11	3.52	0.82
$PM_{2.5}$ 6	May 22-23	1414	0.57	0.16	0.06	0.97	2.94	0.93
$PM_{2.5}$ 7	May 24-25	1419	0.72	0.21	0.17	2.59	4.22	0.80
$PM_{2.5}$ 8	May 26-27	1391	0.72	0.47	0.25	2.36	3.20	0.70
$PM_{2.5}$ 9	May 29-30	1421	0.51	0.03	0.01	0.35	1.84	0.99
Mean			1.43	0.67	0.38	4.16	6.68	0.78

Table S1. The major ions and chloride depletion (%Cl^-) in the sample of cloud water and $PM_{2.5}$ during sampling period. The mass concentrations (μg m^{-3}) of Na^+, Ca^{2+}, Cl^-, NO_3^- and SO_4^{2-} were analyzed using an ion chromatography (Metrohm, Herisau, Switzerland). The cloud water was sampled by a Caltech Active Strand Cloud Water Collector Version 2 (CASCC2) (Modini et al., 2015), when visibility was < 3 km until the volume exceeded 250 ml. The $PM_{2.5}$ was sampled using an Atmospheric particle sampler (Mingye Environmental Protection Technology Co., Ltd., China) with an inlet cyclone with a cut-off aerodynamic diameter of 2.5 μm.

References

Chi, J. W., Li, W. J., Zhang, D. Z., Zhang, J. C., Lin, Y. T., Shen, X. J., Sun, J. Y., Chen, J. M., Zhang, X. Y., Zhang, Y. M., and Wang, W. X.: Sea salt aerosols as a reactive surface for inorganic and organic acidic gases in the Arctic troposphere, Atmos. Chem. Phys., 15, 11341-11353, http://doi.org/10.5194/acp-15-11341-2015, 2015.

Modini, R. L., Frossard, A. A., Ahlm, L., Russell, L. M., Corrigan, C. E., Roberts, G. C., Hawkins, L. N., Schroder, J. C., Bertram, A. K., Zhao, R., Lee, A. K. Y., Abbatt, J. P. D., Lin, J., Nenes, A., Wang, Z., Wonaschütz, A., Sorooshian, A., Noone, K. J., Jonsson, H., Seinfeld, J. H., Toom-Sauntry, D., Macdonald, A. M.,and Leaitch, W. R.: Primary marine aerosol-cloud interactions off the coast of California, J. Geophys. Res., 120, 4282–4303, http://doi.org/10.1002/2014JD022963, 2015.

Wang, Y. Q.: MeteoInfo: GIS software for meteorological data visualization and analysis, Meteorol. Appl., 21, 360–368, https://doi.org/10.1002/met.1345, 2014.