Supplemental materials

Organic coating on sulfate and soot particles in summer Arctic atmosphere

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Elemental profiles of one individual particle were obtained by the scanning transmission electron microscopy (STEM). Figure S2 shows the spectrum of each element in individual particles including nitrogen, carbon, sulfur, and oxygen. Nitrogen spectrum displays one similar profile to carbon. The nitrogen and carbon concentrated in organic coating, suggesting that the possible nitrated organic materials occurred in organic coating. In addition, sulfur and oxygen show similar trend along with the analyzed distance. The result is consistent with the observation of TEM image that the core of individual particles is sulfate. Therefore, the elemental profiles clearly show the elemental distribution in individual particle of the core-shell structure. Figure S5 shows two low-magnification TEM images that can generally show the distribution of different types of particles on the substrate. NanoSIMS shows that these internally mixed particles containing OM and sulfate display OM coating on sulfate core (Figure S1). Intensity and distribution of oxygen in individual particles show good relationship with sulfur. Cl^- and NaO^- maps were used to exclude the aged sea salt which may contain CN and S (Figure S1).

SEM observation suggests that these particles with similar components likely have similar impaction on the TEM grid. In this study, we based on measurements of bearing area and bearing volume of individual particle from atomic force microscopy (AFM) to calculate their equivalent volume diameters. Figure 4 exhibits one higher resolution AFM image of individual secondary sulfate particle with OM coating and its cross-sectional vertical profile. In this study, a good linear correction (R^2=0.916) between equivalent circle diameter (ECD) and ESD were found (Figure 4).

Therefore, we obtained the linear relationship equations between ECD and ESD of individual particles.

y=0.38x, R_sqr=0.916 (1)

Then we can successfully apply the linear equations into all the analyzed particles from TEM analysis. As a result, if we know ECD of individual particle, ESD of the particle can be evaluated through the linear relationship equation.

Following the equations above, ESD of S-rich, soot, individual particles can be calculated. To deduct the volume of S-rich and soot from the same individual particles, equivalent volume diameters of the corresponding organics in individual particles can be known. In final, size distribution of individual particles with OM coating, soot, and sulfate region were obtained (Figure 4).

Figure S4 shows that 63% of particles were identified as the sea salt particles, 29% particles were NSS-sulfate particles, and 8% particles were unclassified particles.

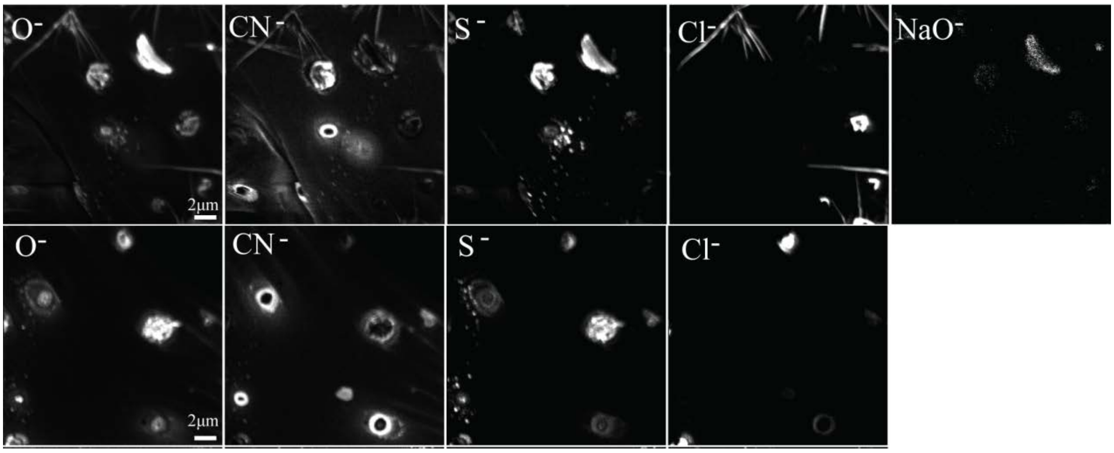


Figure S1 NanoSIMS based on intensity threshold maps of single ion in arctic aerosol particles in different samples. The mixture of OM and sulfate were identified based on CN^- and S^-. The aged sea salt particles containing CN^- and S^- were excluded based on Cl^- and NaO^-.

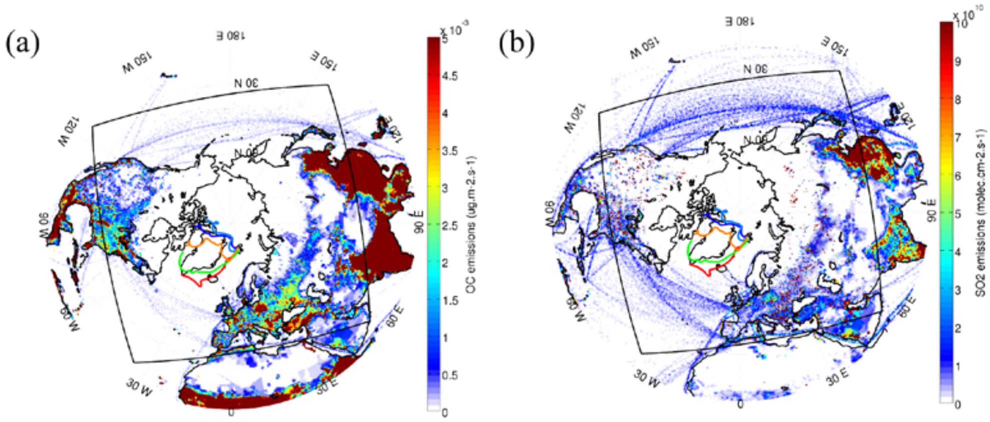
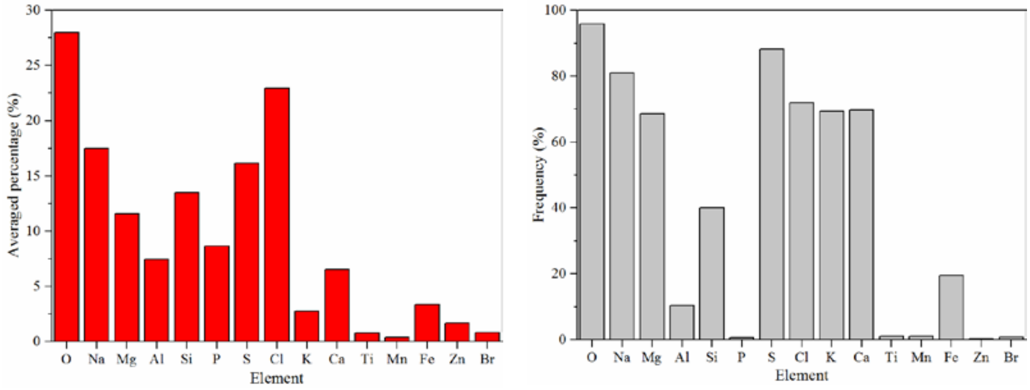


Figure S2 OC (a) and SO_2 (b) Emission intensity in Arctic area and 24h back trajectories on August 11, 12, 14, and 15, 2012.



(a) (b)

Figure S3 Elemental compositions of individual particles from EDS spectra. Left: Average weight of elemental compositions derived from the EDS spectra (b) frequency of element occurring in individual particles.

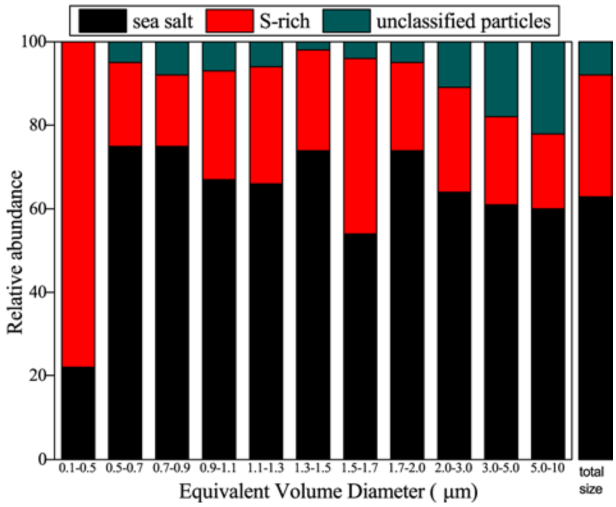


Figure S4 Morphology and relative abundances of typical individual aerosol particles in summertime Arctic samples. The Figure containing sea salts was based on our previous published paper (Chi et al., 2015). Here we added three samples containing S-rich particles on 9-15, August. Unclassified particles contain mineral dust and other particles.

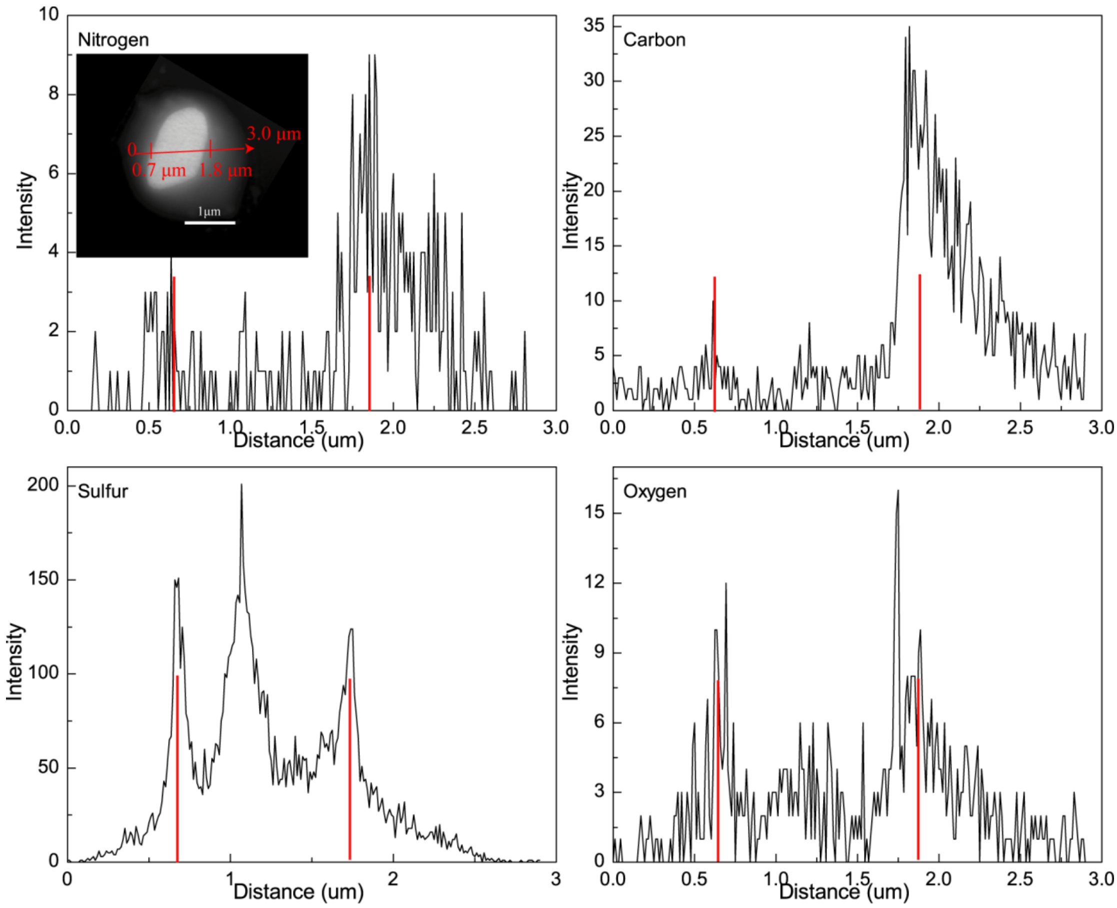


Figure S5 Elemental profile of individual particle containing sulfate with organic coating.

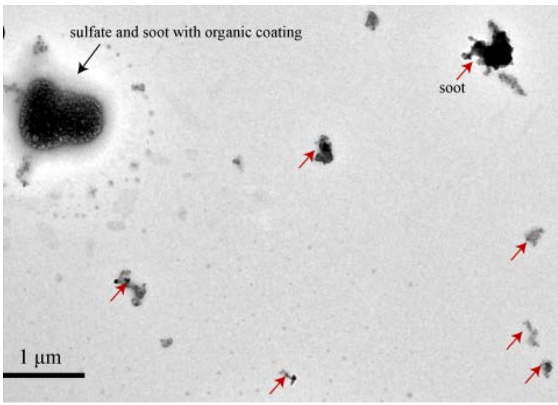


Figure S6 Low magnification TEM images of sulfate and soot particles

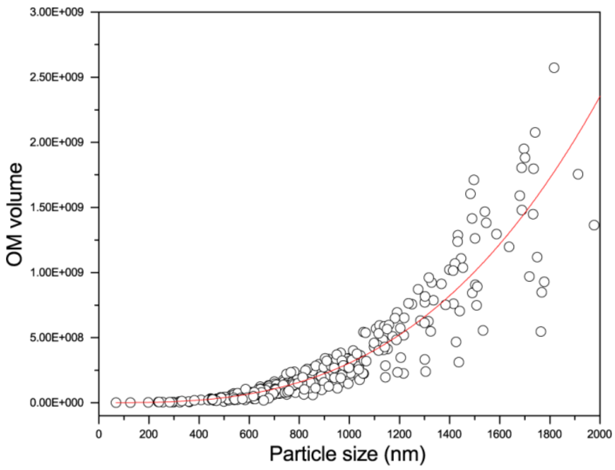


Figure S7 OM volume vs particle size in the analyzed particles. Unite of OM volume is nm^3

Table S1 Sampling information in Arctic area and their analysis

Date	Local time	T	RH	P	WD	WS	TEM	EDX	SEM	AFM	NanoSIMS
2012.8.7	20:50 -21:15	4.9	84	1009.0	296	4.1	43	10			
2012.8.8	08:23 -08:48	4.9	81	1007.6	238	2.1	38	11			
2012.8.9	14:40 -15:05	6.6	81	1003.9	129	6.5	146	50			√
	15:20 -15:49	7.0	78	1003.5	120	7.3	130	26	√		
2012.8.10	00:15 -00:40	7.3	80	998.6	135	8.9	121	23			
2012.8.11	09:10 -09:35	6.2	94	997.0	303	3.3	128	50			√
2012.8.11	16:00 -16:25	4.1	92	1002.0	327	4.6	156	55		√	
2012.8.12	15:25 -15:50	5.7	83	1006.8	132	6.9	100	15	√		
2012.8.13	08:55 -09:20	5.3	81	1009.6	91	1.1	113	16			
2012.8.13	14:15 -14:40	4.5	90	1011.4	351	2.1	136	56			√
2012.8.14	09:50 -10:20	5.0	85	1019.7	351	2.3	134	24			
2012.8.14	15:12 -15:42	4.6	88	1020.5	117	2.6	121	26			
2012.8.14	21:17 -21:47	4.8	84	1020.7	276	5.4	178	56		√	
2012.8.15	09:15 -09:45	5.8	73	1019.6	135	3.7	165	60		√	
2012.8.15	15:00 -15:33	6.8	70	1018.9	270	3.3	80	11			
2012.8.17	9:00 -10:00	3.8	86	1017.1	116	0.3	30	15			
2012.8.17	14:50 -15:20	3.7	85	1015.7	109	2.2	42	16			
2012.8.21	15:05 -15:40	1.6	87	1003.7	314	6.8	46	18			
2012.8.22	08:55 -09:30	2.8	78	999.2	331	2.8	49	19			
2012.8.23	09:00 -09:40	3.4	64	998.0	136	6.9	21	9			
2012.8.23	20:35 -21:08	3.8	59	1002.0	138	6.3	25	9			

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, 2015.

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