

Dear Dr. Wang

We really appreciated that you handle the manuscript reviewing processes.

We have carefully considered all comments and re-wrote the manuscript substantially.

- (1) We restructure the introduction section and add three new paragraphs to introduce each particle type and their impacts in the Arctic air.
- (2) We replaced the Figure 1 as the referee's comments.
- (3) We edited the English through the whole manuscript indicated by the red color.
- (4) We seriously revised the whole manuscript with the referee' comments. Our responses were uploaded with the revised the manuscript.

The point to point responses and the revised manuscript with track markers were listed as below.

We really appreciated the referee's comments which significantly improve the quality of this manuscript. We hope that the current version can be accepted and published in the ACP.

Sincerely

Weijun Li & Zongbo Shi

General comment: While there is obvious importance of conducting detailed physiochemical characterizations of Arctic aerosol in terms of their radiative impacts and subsequent indirect effects on frozen surfaces, there are several issues with the manuscript by Yu et al. that would need to be addressed prior to publication, as discussed in more detail below.

Response: We carefully revised the manuscript considering all comments.

Comment 1: Generally, the introduction could use some restructuring. The climate impacts paragraph could be expanded upon to include more details on specific aerosol types and how they affect the radiative budget and cloud microphysics, providing the motivation for why detailed characterization of aerosols in the Arctic is important – i.e., different aerosols have different effects. This is especially so since the last sentence of the introduction explicitly states that results are discussed in the context of aerosol-radiation and aerosol-cloud interactions. Thus, background material on these effects is needed. The information on lines 92-100 should be located sooner in the introduction as it provides a nice general statement of aerosol studies in the Arctic.

Response: We restructured the introduction part. We moved BC part into second paragraph and line 92-100 into the first paragraph. We also added three paragraphs to summarize the different aerosols including sea salts, mineral, and sulfate, that affect the climate and clouds. In this study, we focus on the sulfate and OM.

Based on the comments, we added more details about the motivation

Line 50-61 “Spatial and temporal variations of aerosol composition, size distribution, and sources of Arctic aerosols have been studied extensively in numerous ground-based, ship, airborne observations, and various atmospheric models (Brock et al., 2011; Burkart et al., 2017; Chang et al., 2011; Dall'Osto et al., 2017; Fu et al., 2008; Hara et al., 2003; Hegg et al., 2010; Iziomon et al., 2006; Karl et al., 2013; Latham et al., 2013; Leck and Bigg, 2008; Leck and Svensson, 2015; Moore et al., 2011; Raatikainen et al., 2015; Wörnschimmel et al., 2013; Winiger et al., 2017; Yang et al., 2018; Zangrando et al., 2013). These studies show that regional pollutants and local natural aerosol production affect sea ice albedo and the heat balance of the atmosphere, especially in the summer when mid-latitude transport is not as frequent relative to that during the winter/spring Arctic Haze season (Hansen and Nazarenko, 2004; Jacob et al., 2010; Shindell, 2007).”

Line 67-79 ” Sea salts, derived from the Arctic Ocean, are the dominant coarse particles ($>1 \mu\text{m}$) in the Arctic atmosphere (Behrenfeldt et al., 2008; Chi et al., 2015). Compared to other types of aerosols, sea salt is the largest contributor to radiative forcing in remote Ocean air (Wang et al., 2019). Natural sea salt particles can provide large surfaces for heterogeneous reaction with acidic gases in the Arctic air (Chi et al., 2015; Geng et al., 2010; Hara et al., 2003). Moreover, sea salt particles are an important source of cloud condensation nucleation (CCN) in the Arctic air (Abbatt et al., 2019). Coarse dust particles in the Svalbard region have been observed to be occasionally influenced from local (Svalbard) and/or distant (e.g., Iceland, Greenland and Siberia) sources in high latitudes (Behrenfeldt et al., 2008). Tobo et al. (2019)

showed that glacial outwash sediments in Svalbard (a proxy for glacially sourced dusts) due to the recent rapid and widespread retreat of glaciers have a remarkably high ice nucleating ability under conditions relevant for mixed-phase cloud formation.”

Line 106-115 ” Sulfate is a dominant aerosol component in the Arctic air (Quinn et al., 2007). The Community Earth System Model simulations show that sources from East Asia have the largest contribution to the Arctic sulfate column burden, with an annual mean contribution of 27%, followed by 11–13% each from South Asia, the rest of the world (including the Arctic), and Russia/Belarus/Ukraine sources and 13% from natural sources (Yang et al., 2018). Large amounts of secondary species including sulfate and OM not only change radiative forcing and number of CCN in Arctic atmosphere (Abbatt et al., 2019; Yang et al., 2018) but also influence optical, hygroscopic, and CCN activity of these internally mixed BC and mineral dust particles (Latham et al., 2013; Raatikainen et al., 2015; Zanutta et al., 2018).

”

Comment 2: There is no background on previous relevant studies conducted at the study location, even though there is a long-term monitoring station with aerosol measurements at Ny-Ålesund (<https://www.esrl.noaa.gov/psd/iasoa/stations/nyalesund>). It is not the same exact location as the Chinese site, but close enough to at least use those routine, publically-available measurements to provide some broader spatial and temporal context. Then, the discussion on the background on sources of BC, OM, BrC, and sulfate and the various mixing mechanisms can reside.

Response: We searched the data portal but there is no aerosol measurement during the year of our observation in the database. The database only has online gaseous concentration (e.g. CO₂) during our observation period. We added some background information here.

We added the background as the comments.

line 147-153: “On the west coast of the island of Spitsbergen, Ny-Ålesund is a Norwegian research and monitoring infrastructure, hosting national and international research projects and programmes. The Norwegian Polar Institute (NPI) runs the Sverdrup Research Station at the coast and Zeppelin Observatory at the Mountain 475 m asl, and Sweden, Germany, France, Italy, Japan, China, England, The Netherlands, South Korea, and India are the other countries to have established long-term programmes in Ny-Ålesund (<https://www.esrl.noaa.gov/psd/iasoa/stations/nyalesund>).”

Comment 3: Laskina et al. (2015) states, “Typically, SSA particles are deposited wet and, if possible, samples used for single-particle analysis should be stored at or near conditions at which they were collected in order to avoid dehydration.” I assume the RH was much higher during collection, so can the authors comment on what might change between collection and storage under the different RH conditions?

Response: The TEM and SEM analysis were carried out in vacuum so the particles we analysed are dry at the time of observation. It is a general practice to keep the samples in dry

conditions to prevent the sample to be exposed in humid air and phase changes during the storage.

Laskina's paper also mentioned that "...However, if samples need to be dry, as is often the case, then this study found that storing SSA particles at ambient laboratory conditions (17–23% RH and 19–21 °C) was effective at preserving them and reducing changes that would alter samples and subsequent data interpretation."

Comment 4: Also, Laskina et al. is focused on sea spray aerosol, but the authors conclude that continental sources were a major contributor (lines 377-379), so what about loss of semivolatile organics or sulfate during low RH storage? This possible caveats should be discussed in more detail than they currently are.

"Sulfate" particles is used intermittently throughout, perhaps "sulfate-containing" is more appropriate.

Response: This is now mentioned in the text:

Context in section 2.2 "In our study, the effects of water and other semi-volatile organics were not considered as they evaporate in the vacuum."

Sulfate does not evaporate so our storage does not have any impact on the sulfate aerosols.

Specific comments:

Comment 5: Title: Because this work is only conducted at one location during one month, extending it to "summer Arctic" seems like a stretch as aerosol populations can vary significantly from terrestrial to the high Arctic and can also change from early to late summer. Perhaps the authors should consider changing the title to something like, "Organic coatings on sulfate and soot particles during late summer in the Svalbard Archipelago".

Response: Yes. This is now revised title to "Organic coating on sulfate and soot particles during late summer in the Svalbard Archipelago"

Comment 6: Lines 33-34: State the motivation for the particular focus on these particle types, so that this focus is justified.

Response: We modified the sentence and made clear statement:

Line 32 to 33: determine the size and mixing state of individual sulfate and carbonaceous particles at 100 nm – 2 μm

Comment 7: Lines 34-35: State percentage of OM coated NSS-sulfate here to provide some quantification to "commonly coated".

Response: Revised

Line 33 to 34: found that 74% by number of non-sea salt sulfate particles were coated with organic matter (OM)

Comment 8: Lines 52-53: Regional pollutants and local natural aerosol production can also affect sea ice albedo, especially in the summer when midlatitude transport is not as frequent relative to the winter/spring Arctic Haze season.

Response: Revised

Line 58 to 61: “These studies show that regional pollutants and local natural aerosol production affect sea ice albedo and the heat balance of the atmosphere, especially in the summer when mid-latitude transport is not as frequent relative to that during the winter/spring Arctic Haze season (Hansen and Nazarenko, 2004; Jacob et al., 2010; Shindell, 2007). “

Comment 9: Lines 57-63: This part is very BC focused and should be included in the second paragraph.

Response: Revised. We moved the BC part into second paragraph.

Comment 10: Lines 144-146: There are no details provided on where these meteorological data came from or what instrumentation was used to measure the mentioned parameters. Although, the data are not presented anywhere so perhaps this information is not relevant in include at all.

Response: We added the details about the instrument.

In context: line 166-170 “During the sampling period, meteorological data at the sampling site including pressure (P), relative humidity (RH), temperature (T), wind speed (WS), and wind direction (WD) were measured and recorded every 5 min using a pocket weather meter (Kestrel 4500, Nielsen-Kellermann Inc., USA).”

Comment 11: Line 193: Define the size of “fine” and quantify “abundant”.

Response: We revised them.

Line 215: Here we deleted the fine and add (~30% by number) after the abundant.

Comment 12: Lines 200-201: “good consistent property” is vague.

Response: We revised them.

Section 3.2 line 221-224 “ Because all the sulfate particles in different samples collected in the Arctic had similar elemental composition and mixing state from TEM observations, we just selected three samples containing abundant sulfate particles (Table 1) for further studies.”

Comment 13: Section 2.4: Separate SEM and AFM into separate sections – not sure why these specifically are combined.

Response: We separated them into two part: section 2.4 SEM measurement and 2.5 AFM measurement.

Comment 14: Line 275: Why only 500 m?

Response: We did 50 m, 500 m, and 1000 m, these three are generally consistent. As a result, we chosen 500 m as a representation. We revised the sentence to:

Section 2.7 line 288-290 “Here we selected an altitude of 500 m as the end point in each

back trajectory (Figure 1). Back trajectories for 50 m and 1000 m above sea level are similar.”

Comment 15: Lines 276-279: This belongs in the results section.

Response: Thanks. We move this section into result in section 3.2

Comment 16: Lines 290-292: Why were different days evaluated for HYSPLIT versus FLEXPART (3 versus 10 day, respectively)?

Response: These are two different models. HYSPLIT is a meteorological model, tracing the air mass travel to a site. It does not offer conclusive information about the long term transport of particles (Stein et al., 2015). However, the FLEXPART model including WRF model uses a different method to evaluate the particle dispersion along the backward air masses. For each simulation (one per sample), 20000 pseudo-particles were released in a small volume around the station position. Each single particle position evolution backward in time was determined by Lagrangian dispersion calculation. In this study, we based on the data Figure to adjust the period. Here we found that 10 days FLEXPART data can well indicate the particle sources.

As the Figure 3 shown, 3 days back trajectories provided sufficient information to indicate the air masses movement. Furthermore, long-term time back trajectories become less certain.

Reference: Stein A. F., et al. NOAA's Hysplit Atmospheric Transport and Dispersion Modeling System. Bull. Amer. Meteorol. Soc. 96, 2059-2077 (2015).

Brioude, J., D. Arnold, A. Stohl, M. Cassiani, D. Morton, P. Seibert, W. Angevine, S. Evan, A. Dingwell, J. D. Fast, R. C. Easter, I. Pissò, J. Burkhardt, and G. Wotawa (2013), The Lagrangian particle dispersion model FLEXPART-WRF version 3.1, Geosci. Model Dev., 6(6), 1889-1904.

Comment 17: Lines 304-306: Provide a citation for this statement.

Response: added two reference here.

Moffet, R. C., T. C. Rödel, S. T. Kelly, X. Y. Yu, G. T. Carroll, J. Fast, R. A. Zaveri, A. Laskin, and M. K. Gilles (2013), Spectro-microscopic measurements of carbonaceous aerosol aging in Central California, Atmos. Chem. Phys., 13(20), 10445-10459.

Li, W., J. Sun, L. Xu, Z. Shi, N. Riemer, Y. Sun, P. Fu, J. Zhang, Y. Lin, X. Wang, L. Shao, J. Chen, X. Zhang, Z. Wang, and W. Wang (2016), A conceptual framework for mixing structures in individual aerosol particles, J. Geophys. Res., 121(22), 13,784-713,798.

Riemer, N., A. P. Ault, M. West, R. L. Craig, and J. H. Curtis (2019), Aerosol Mixing State: Measurements, Modeling, and Impacts, Rev. Geophys., 57,

<https://doi.org/10.1029/2018RG000615>.

Comment 18: Lines 310-312: 39% for the S-rich + soot + OM out of all NSS-particles? How was NSS-sulfate and OM determined?

Response: We reworded this sentence to :

Section 3.1 line 322-325 “Here, we focused on non-sea salt (NSS) particles including S-rich, soot, and OM particles NSS-particles and sulfate-containing particles account

for $39 \pm 5\%$ and $29 \pm 7\%$ by number of all the 2002 particles analysed (Figure 3).”

Comment 19: Lines 332-333: How was this documented? This statement is a bit vague.

Response: We deleted it

Comment 20: Figure 1: The date are very small and it is difficult to tell if there is any sort of time evolution in the air mass sources throughout August. Perhaps the authors could either color the lines by date or create multiple panels (e.g., 1 per week of trajectories).

Response: We revised the Figure as below. The one week is not good at all. The current data can know where these air masses were from.

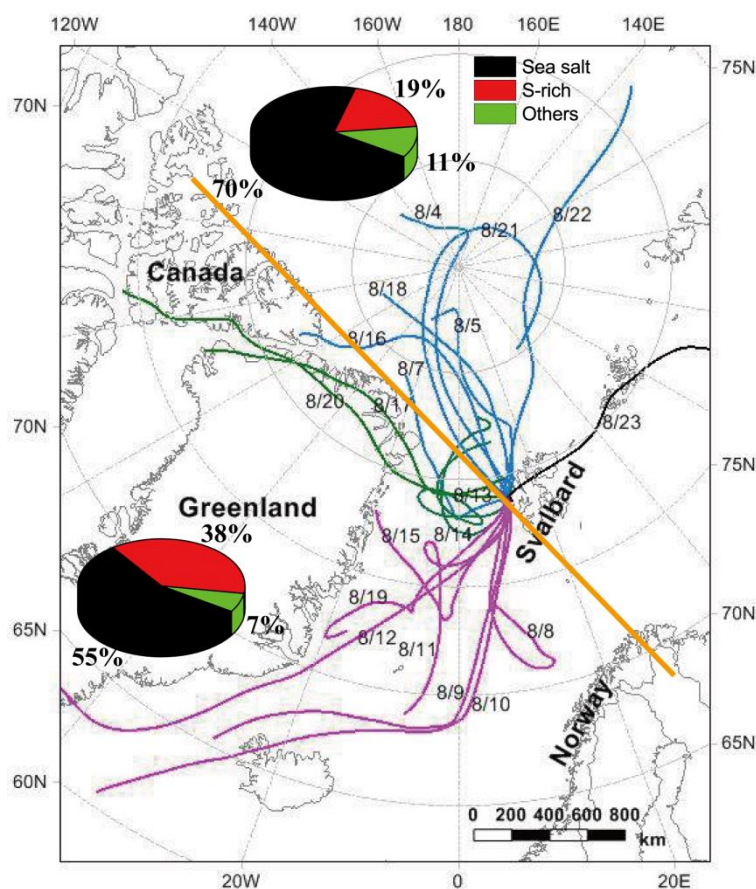


Figure 1 72 h back trajectories of air masses at 500 m over Arctic Yellow River Station in Svalbard during 3–26 August 2012, and arriving time was set according to the sampling time. Air masses were divided into two groups by the yellow line: one group from the central Arctic Ocean and the other one from North America and Greenland. Pie charts show the number fractions of sea salt, S-rich, and other particles.

Comment 21: Figure 3: I assume this is from TEM/EDX? Why are soot and OM not shown? Also, can the authors show a time series in addition to the diameter relative fraction? That might provide some insight into temporal changes due to air mass origin.

Response: We revised the Figure. More than 98% of soot and OM were internally mixed

sulfate, named as sulfate-coating particles (Figure 7). The very small number of soot and OM and some mineral particles were assigned into others.

As the reviewer's comments, air masses were divided into two groups by the yellow line: one group from the central Arctic Ocean and the other one from North America and Greenland. Pie charts show the number fractions of sea salt, S-rich, and others. Here we can find that more S-rich particles were from North America and more sea salt particles from ocean area (Figure 1).

Comment 22: Table S1: There is a substantial amount of referencing to Table S1. While I am glad to see the authors have include sample and particle numbers for each analytical technique, jumping back and forth between the manuscript and SI is tedious. I suggest the table be moved to the main manuscript.

Response: Thanks. We moved to the main manuscript.

Technical corrections:

Comment 23: There are many typos and grammatical issues that need to be fixed throughout the manuscript.

Line 32: "mixing state properties"

Response: We smooth the English writing in all the manuscript.

1 **Organic coating on sulfate and soot particles during late summer in**
2 **the Svalbard Archipelago**

3

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23

24 **Abstract**

25 Interaction of anthropogenic particles with radiation and clouds plays an important
26 role on Arctic climate change. Mixing state of aerosols is a key parameter to influence
27 **aerosol radiation** and **aerosol-cloud** interaction. However, little is known on this
28 parameter in the Arctic, preventing an accurate representation of this information in
29 global models. Here we used transmission electron microscopy with
30 energy-dispersive X-ray spectrometry, scanning electron microscopy, nanoscale
31 secondary ion mass spectrometry, and atomic forces microscopy to **determine the size**
32 **and mixing state of individual sulfate and carbonaceous particles at 100 nm – 2 μm**
33 **collected in the Svalbard Archipelago in summer. We found that 74% by number of**
34 **non-sea salt sulfate particles were coated with organic matter (OM).** 20% of sulfate
35 particles also had soot inclusions which only appeared in the OM coating. The OM
36 coating is estimated to contribute to 63% of the particle volume on average. To
37 understand how OM coating influences optical properties of sulfate particles, **a Mie**
38 **core-shell** model was applied to calculate optical properties of individual sulfate
39 particles. **Our** result shows that absorption cross section of individual OM-coated
40 particles significantly increased when assuming the OM coating as light-absorbing
41 brown carbon. **Microscopic observations here** suggest that OM modulates the mixing
42 structure of fine Arctic sulfate particles, which may determine their hygroscopicity
43 and optical properties.

44

45 **1. Introduction**

46 Surface temperatures are rising faster in the Arctic than the rest of globe (IPCC,
47 2013). Although increased human-induced emissions of long-lived greenhouse gases
48 are certainly one of the driving factors, air pollutants, such as aerosols and ozone, are
49 also important contributors to climate change in the Arctic (Law and Stohl, 2007;
50 Shindell, 2007). Spatial and temporal variations of aerosol composition, size
51 distribution, and sources of Arctic aerosols have been studied extensively in numerous
52 ground-based, ship, airborne observations, and various atmospheric models (Brock et
53 al., 2011; Burkart et al., 2017; Chang et al., 2011; Dall'Osto et al., 2017; Fu et al.,
54 2008; Hara et al., 2003; Hegg et al., 2010; Iziomon et al., 2006; Karl et al., 2013;
55 Latham et al., 2013; Leck and Bigg, 2008; Leck and Svensson, 2015; Moore et al.,
56 2011; Raatikainen et al., 2015; Wörnschimmel et al., 2013; Winiger et al., 2017;
57 Yang et al., 2018; Zangrando et al., 2013). These studies show that regional pollutants
58 and local natural aerosol production affect sea ice albedo and the heat balance of the
59 atmosphere, especially in the summer when mid-latitude transport is not as frequent
60 relative to that during the winter/spring Arctic Haze season (Hansen and Nazarenko,
61 2004; Jacob et al., 2010; Shindell, 2007).

62 Aerosol particles in the Arctic atmosphere are mainly composed of sea salt, sulfate,
63 particulate organic matter (OM) with a small amount of ammonium, nitrate, black
64 carbon (BC) (Hara et al., 2003; Quinn et al., 2007) and mineral dust particles
65 (Dagsson-Waldhauserova et al., 2013). Sea salts, derived from the Arctic Ocean, are
66 the dominant coarse particles ($>1 \mu\text{m}$) in the Arctic atmosphere (Behrenfeldt et al.,
67 2008; Chi et al., 2015). Compared to other types of aerosols, sea salt is the largest
68 contributor to radiative forcing in remote Ocean air (Wang et al., 2019). Natural sea
69 salt particles can provide large surfaces for heterogeneous reaction with acidic gases
70 in the Arctic air (Chi et al., 2015; Geng et al., 2010; Hara et al., 2003). Moreover, sea
71 salt particles are an important source of cloud condensation nucleation (CCN) in the
72 Arctic air (Abbatt et al., 2019); Coarse dust particles in the Svalbard region have been
73 observed to be occasionally influenced from local (Svalbard) and/or distant (e.g.,
74 Iceland, Greenland and Siberia) sources in high latitudes (Behrenfeldt et al., 2008).

75 Tobo et al. (2019) showed that glacial outwash sediments in Svalbard (a proxy for
76 glacially sourced dusts) due to the recent rapid and widespread retreat of glaciers have
77 a remarkably high ice nucleating ability under conditions relevant for mixed-phase
78 cloud formation.

79 BC, commonly called “soot”, is derived from the combustion sources such as
80 diesel engines, residential solid fuel, and open burning (Bond et al., 2013). Studies
81 show BC in the Arctic absorbs solar radiation in the atmosphere and when deposited
82 on snow (Iziomon et al., 2006; Koch and Hansen, 2005; Sand et al., 2013; Shindell,
83 2007). Maahn et al. (2017) found that BC concentration is enhanced below the clouds
84 in the Arctic. This influences the mean effective radii of cloud droplets which lead to
85 the suppressed drizzle production and precipitation. Possible sources of BC particles
86 in the Arctic include natural gas flaring (Qi et al., 2017), ship emissions (Browse et al.,
87 2013; Weinbruch et al., 2012) and long range transport from emissions of biomass
88 burning and fossil fuels in the northern hemisphere (Winiger et al., 2016; Xu et al.,
89 2017). Winiger et al.(2017) showed that most of the Arctic BC is from domestic
90 activities (35%) and transportation (38%), with only minor contributions from gas
91 flaring (6%), power plants (9%), and open fires (12%).

92 OM is a significant component in Arctic aerosol (Quinn et al., 2007). More than
93 100 organic species have been detected in the Arctic aerosols and polyacids are the
94 most abundant compound class, followed by phthalates, aromatic acids, fatty acids,
95 fatty alcohols, sugars/sugar alcohols, and n-alkanes (Fu et al., 2008). Recently, certain
96 organic aerosols, referred to as brown carbon (BrC), have been recognized as an
97 important light-absorbing carbonaceous aerosol in the troposphere (Alexander et al.,
98 2008; Andreae and Gelencser, 2006; Feng et al., 2013; Lack et al., 2012). BrC can be
99 directly emitted from combustion sources or form in the atmosphere via
100 photo-chemical aging (Jiang et al., 2019; Saleh et al., 2013; Updyke et al., 2012).
101 Moreover, aging of secondary organic aerosols can significantly contribute to BrC
102 during atmospheric transport (Laskin et al., 2015). Feng et al.(2013) estimated that on
103 average, BrC accounts for 66% of total OM mass globally and its light absorption is
104 about 26% of BC.

105 Sulfate is a dominant aerosol component in the Arctic air (Quinn et al., 2007). The
106 Community Earth System Model simulations show that sources from East Asia have
107 the largest contribution to the Arctic sulfate column burden, with an annual mean
108 contribution of 27%, followed by 11–13% each from South Asia, the rest of the world
109 (including the Arctic), and Russia/Belarus/Ukraine sources and 13% from natural
110 sources (Yang et al., 2018). Large amounts of secondary species including sulfate and
111 OM not only change radiative forcing and number of CCN in Arctic atmosphere
112 (Abbatt et al., 2019; Yang et al., 2018) but also influence optical, hygroscopic, and
113 CCN activity of these internally mixed BC and mineral dust particles (Latham et al.,
114 2013; Raatikainen et al., 2015; Zanatta et al., 2018).

115 BC and BrC are often internally mixed with other non-absorbing aerosols, such as
116 sulfate (Lack et al., 2012; Laskin et al., 2015). Internal mixing means that a single
117 particle simultaneously contains two or more types of aerosol components (Li et al.,
118 2016). This internal mixing can enhance BC absorption by a factor of two (Bond et al.,
119 2013) and change the activity of CCN in the Arctic atmosphere (Leck and Svensson,
120 2015; Martin et al., 2011). A few previous studies also looked at the mixing states of
121 coarse aerosol particles in Arctic troposphere (Behrenfeldt et al., 2008; Chi et al.,
122 2015; Geng et al., 2010; Hara et al., 2003; Leck and Svensson, 2015; Moroni et al.,
123 2017; Raatikainen et al., 2015; Sierau et al., 2014), but those of fine non-sea salt
124 particles, including the most important short-lived climate forcers – BC and BrC
125 (Feng et al., 2013; Fu et al., 2008; Kirpes et al., 2018; Laskin et al., 2015; Leck and
126 Svensson, 2015), are poorly characterized. A poor understanding on mixing state of
127 BC and BrC in individual particles prevents an accumulate simulation of the direct
128 aerosol forcing and aerosol-cloud interaction in the Arctic (Browse et al., 2013;
129 Samset et al., 2014; Zanatta et al., 2018).

130 In this study, individual aerosol particles were collected in Svalbard during 7-23
131 August, 2012. We combined the data from various microscopic instruments to
132 determine the size, composition, and mixing properties of individual particles, with a
133 particular focus on sulfate and carbonaceous particles. Mie theory was used to test
134 how OM coating influences optical properties of sulfate particles in the Arctic when

135 OM was assumed as BrC. The results are discussed in the context of aerosol radiation
136 and aerosol-cloud interaction.

137

138 **2. Experimental section**

139 **2.1 Field campaign**

140 The Svalbard archipelago includes all landmasses between 74 and 81 degrees
141 North and 10 and 35 degrees East (Figure 1). The islands cover 63000 km².
142 Ny-Ålesund town is situated on the west coast of the largest island, Spitsbergen and
143 1200 km from the North Pole. It is a central platform for Arctic research.

144 **The observation site is based at the Chinese Arctic Yellow River Station (78°55'N,**
145 **11°56'E) (Chi et al., 2015; Geng et al., 2010). The site is about 2 km far away from**
146 **the Zeppelin observatory station (78.9N 11.88E) run by the Ny-Ålesund Science**
147 **Managers Committee. On the west coast of the island of Spitsbergen, Ny-Ålesund is a**
148 **Norwegian research and monitoring infrastructure, hosting national and international**
149 **research projects and programmes. The Norwegian Polar Institute (NPI) runs the**
150 **Sverdrup Research Station at the coast and Zeppelin Observatory at the Mountain 475**
151 **m asl, and Sweden, Germany, France, Italy, Japan, China, England, The Netherlands,**
152 **South Korea, and India are the other countries to have established long-term**
153 **programmes in Ny-Ålesund (<https://www.esrl.noaa.gov/psd/iasoa/stations/nyalesund>).**
154 Two to three samples were regularly collected at 9:00, 16:00, 21:00 (local time) of
155 each day, with a total of 46 samples during 7-23 August, 2012.

156 A sampler containing a single-stage impactor with a 0.5-mm-diameter jet nozzle
157 (Genstar Electronic Technology, China) was used to collect individual particles by the
158 air flow rate at 1.5 l min⁻¹. Aerosol particles were collected onto copper TEM grids
159 coated with carbon film. This sampler has a collection efficiency of 31% at 100 nm
160 aerodynamic diameter and 50% at 200 nm **assuming** the density of the particles is 2 g
161 cm⁻³. Sampling times varied from twenty minutes to two hours **depending on the**
162 **loading of particles**. After collection, each sample was placed in a sealed dry plastic
163 tube and stored in a desiccator at 20 ± 3% RH for analysis. Ambient laboratory
164 conditions (17–23% RH and 19–21 °C) is effective at preserving individual

165 hygroscopic aerosol particles and reducing changes that would alter samples and
166 subsequent data interpretation (Laskina et al., 2015). During the sampling period,
167 meteorological data at the sampling site including pressure (P), relative humidity
168 (RH), temperature (T), wind speed (WS), and wind direction (WD) were recorded
169 every 5 min using a pocket weather meter (Kestrel 4500, Nielsen-Kellermann Inc.,
170 USA). Sample information including local sampling date and time and various
171 meteorological conditions are listed in Table 1.

172

173 **2.2 TEM measurement**

174 Individual particle samples were examined by a JEOL JEM-2100 transmission
175 electron microscopy operated at 200 kV with an energy-dispersive X-ray
176 spectrometry (TEM/EDS). TEM can observe the mixing structure of different aerosol
177 components within an individual particle on the substrate because electron beam
178 transmit through the specimen to form an image. EDS spectra are acquired for a
179 maximum time of 30 s to minimize potential beam damage and collect particle X-ray
180 spectra with sufficient intensity. TEM grids are made of copper (Cu) and covered by a
181 carbon-reinforced substrate, so Cu is excluded from the quantitative analyses of the
182 particles. Because of the substrate contribution, C content in TEM grid coated by
183 carbon film might be overestimated in EDS spectra of individual particles.

184 The distribution of aerosol particles on TEM grids was not uniform, with coarser
185 particles occurring near the center and finer particles on the periphery. Therefore, to
186 ensure that the analysed particles are representative, five areas were chosen from the
187 center to periphery of the sampling spot on each grid. Through a labor-intensive
188 operation, 2002 aerosol particles with diameter < 10 μm in 21 samples were analysed
189 by TEM/EDS (Table 1). To check elemental composition of individual particles, EDS
190 was manually used to obtain elemental spectra of individual particles. In the clean
191 Arctic air, there are simple particle types including sea salt, sulfate, soot, OM, and
192 mineral. Because soot particles have chain-like aggregation, it is not necessary to
193 check their elemental composition. Sea salt particles display spherical or square
194 shapes and are stable under the electron beam in TEM (Chi et al., 2015). Sulfate

195 particles are spherical but flats on the substrate and produce unstable bubble under the
196 electron beam (Buseck and Posfai, 1999). TEM observations can also identify sulfate
197 particles or sulfate with OM coating. TEM/EDS analysis is very time-consuming.
198 Thus, we did not check the composition of every single particle analysed. Instead, we
199 randomly checked the elemental composition of 20-30 particles in each sample (Table
200 1). EDS spectra of 575 particles were manually selected and saved in the computer for
201 elemental composition analysis. Particles examined by TEM were dry at the time of
202 observation in the vacuum of the electron microscope. In our study, the effects of
203 water and other semi-volatile organics were not considered as they evaporated in the
204 vacuum.

205 Elemental mapping and line profile of selected individual aerosol particles were
206 also obtained from the EDS scanning operation mode of TEM (STEM). The STEM
207 information clearly display elemental distribution in the targeted individual particles
208 which cannot be provided by the above EDS analysis. Based on preliminary
209 individual analysis, we further chose the typical samples containing abundant sulfate
210 with OM coating for the STEM analysis. High-resolution elemental distribution in
211 individual particles provides detailed mixing structure of sulfate and OM in individual
212 particles.

213 The iTEM software (Olympus soft imaging solutions GmbH, Germany) is an
214 image analysis platform for electron microscopy. In this study, it was used to
215 manually or automatically obtain area, perimeter, and equivalent circle diameter
216 (ECD) of individual particles. In these analysed samples, we found there were
217 abundant sulfate particles (~30% by number) in the 11 samples collected during 9-15
218 August, 2012. In other samples, there are more sea salt particles with few particles.
219 Based on the TEM observations, we selected the samples containing more sulfate
220 particles for further microscopic analyses (see below).

221

222 2.3 NanoSIMS measurement

223 Because the sulfate particles in different samples collected in the Arctic had
224 similar elemental composition and mixing state from the TEM observations, we

225 selected three samples (Table 1) for nanoscale secondary ion mass spectrometry
226 (NanoSIMS) analysis (CAMECA Instruments, Geneviers, France). A micro-cesium
227 source was used to generate Cs⁺ primary ions, with an impact energy of 16 kV for
228 sample interrogation. The primary beam was stepped across the sample to produce
229 element specific, quantitative digital images. The Cs⁺ primary ion beam was used to
230 obtain ¹⁶O⁻, ¹²C¹⁴N⁻, ¹⁴N¹⁶O⁻, ³²S⁻, ³⁵Cl⁻, and ¹⁶O²³Na⁻ ions in this study. The
231 NanoSIMS analysis can obtain ion mapping of particles with nanometer spatial
232 resolution over a broad range of particle sizes (Figure S1). Because the substrate of
233 TEM grid is carbon, CN⁻ is adopted to represent OM in individual particles (Chi et al.,
234 2015; Ghosal et al., 2014). S⁻ is used to infer the presence of sulfates in individual
235 particles (Li et al., 2017). A total of 32 sulfate containing particles were analysed by
236 the NanoSIMS.

237

238 2.4 SEM measurement

239 We used a Zeiss ultra 55 scanning electron microscopy (SEM) with EDS to
240 examine the vertical distribution of OM and sulfate in individual particles. TEM grids
241 were mounted onto an aluminum SEM stub and directly observed in secondary
242 electron image mode. SEM analysis was operated at 10 kv of extra high tension (EHT)
243 and 9.7 mm of work distance (WD). Processes such as sample moving, analysis
244 region selection and imaging were controlled by computer. The specimen stage in
245 SEM was tilted at the range of 0-75°, and then we vertically observed thickness of
246 OM coating and sulfate core on the substrate. Two typical samples that contain
247 abundant sulfate particles were chosen for this analysis (Table 1).

248 2.5 AFM measurement

249 AFM with a digital nanoscope IIIa instrument operating in the tapping mode was
250 used to observe surface morphology of individual aerosol particles and measure
251 particle thickness. The tapping AFM has a cantilever and conical tip of 10 nm radius.
252 By using AFM, a general image of the particles is taken at 10 μm full scan size, which
253 generally includes 1-2 particles depending on the exact location. In this study, we are
254 only interested in the sulfate-containing particles. AFM provides surface information

255 and morphology of 17 particles but no composition. Samples were firstly quickly
256 examined by the TEM under low magnification mode to find sulfate-containing
257 particles. Because TEM grids have coordinates letters, we can find the same
258 particles in the AFM. This analysis provides 3-D image of the sulfate-containing
259 particles and their volume. After we obtained AFM images of sulfate particles, the
260 NanoScope analysis software can automatically obtain bearing area (A) and bearing
261 volume (V). This can then be converted into equivalent circle diameter (d, ECD) and
262 equivalent spherical diameter (D, ESD)

$$263 \quad A = \frac{4}{3} \pi r^2 = \frac{\pi d^2}{3} \rightarrow d = \sqrt{\frac{3A}{\pi}} \quad (1)$$

$$264 \quad V = \frac{4}{3} \pi r^3 = \frac{4}{3} \times \frac{\pi D^3}{8} \rightarrow D = \sqrt[3]{\frac{6V}{\pi}} \quad (2)$$

265 ESD and ECD of sulfate-containing particles are well corrected. Using the linear
266 correlation equation (ESD=0.38ECD), we can then correct the ESD of individual
267 particles from TEM analysis to obtain the ECD (Chi et al., 2015).

268

269 **2.6 Calculation of BrC optical properties**

270 The refractive index used for the non-light-absorbing sulfate component was set to
271 $m=1.55$ at 550 nm (Seinfeld and Pandis, 2006). The refractive index of OM (as BrC)
272 is not known so we considered three scenarios: strongly absorbing ($1.65-0.03i$ at 550
273 nm), moderately absorbing ($1.65-0.003i$ at 550 nm), and non-absorbing OM (1.65 at
274 550 nm) (Feng et al., 2013). Here, we choose 550 nm as a case study to test how OM
275 coating influence sulfate particles in Arctic air.

276 BHCOAT Mie code by Bohren and Huffman (1983) was used to calculate the
277 optical properties, including scattering cross section (SCS), absorption cross section
278 (ACS), and single scattering albedo (SSA), assuming a core-shell structure. We firstly
279 calculated these parameters assuming a sulfate core and OM shell structure only
280 (ignoring some of the particles that contain soot core). Because the Mie code only can
281 calculate the core-shell structure or homogeneous model, we assume sulfate as a core
282 and OM as a shell in individual particle to build the core-shell model. Based on the

283 core-shell standard mode (Li et al., 2016), we can calculate optical properties of
284 individual internally mixed particles.

285 **2.7 Back trajectories of air masses and Lagrangian particle dispersion model**

286 Three-day (72 h) back trajectories of air masses were generated using a Hybrid
287 Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model at the Chinese
288 Arctic Yellow River Station during August 2012. Here we selected an altitude of 500
289 m as the end point in each back trajectory (Figure 1). Back trajectories for 50 m and
290 1000 m above sea level are similar.

291 A lagrangian particle dispersion model FLEXPART-WRF 3.1 (Brioude et al.,
292 2013) was also used to examine the origin of particles. The FLEXPART-WRF model
293 is using meteorological parameters from WRF dynamical simulation. The domain
294 resolution is 50×50 km with 64 vertical levels. The FLEXPART-WRF simulations
295 were launched in a backward mode over 10 days, with the Chinese Arctic Yellow
296 River Station as an origin. For each simulation (one per sample), 20000
297 pseudo-particles were released in a small volume around the station position. Each
298 single particle position evolution backward in time was determined by Lagrangian
299 dispersion calculation.

300 Based on the TEM experiments and back trajectory of air masses (Figure 1), we
301 found that there were more S-rich with OM coating particles in the samples collected
302 on August 11, 12, 14, and 15, 2012. Therefore, we further did the FLEXPART-WRF
303 simulation of these four days (Figure 2). The emission intensity in the Arctic area has
304 been also shown in Figure S2.

305 **3. Results**

306 **3.1 Composition and source of aerosol particles**

307 TEM/EDS analysis shows that O, Na, S, and Cl are dominant elements in
308 individual particles (Figure S3). On basis of the composition and morphology of
309 individual particles, we classified the particles into four major groups: Na-rich (i.e.
310 NaCl, Na₂SO₄, and NaNO₃), S-rich (i.e. ammonium sulfate and sulfuric acid),
311 carbonaceous (soot and OM), and mineral dust particles. The classification criteria of
312 different particle types and their sources have been described in a separate study (Li et

313 al., 2016). S-rich particles representing secondary inorganic particles (e.g., SO_4^{2-} ,
314 NO_3^- , and NH_4^+) are **converted** from gaseous SO_2 , NO_x , and NH_3 . OM can be divided
315 into primary organic matter (POM) and secondary organic matter (SOM). SOM is
316 produced from the chemical oxidation of volatile organic compounds (VOCs) and
317 often exhibits OM coating on S-rich particles (Li et al., 2016; Moffet et al., 2013;
318 Riemer et al., 2019). Na-rich particles in the marine air are from sea spray and have
319 typical near cubic shape. **Mineral dust particles from natural ground soil contain Si, Al,**
320 **or Ca and have irregular shape.** Soot particles, which contain C with minor O, appear
321 as a chain-like aggregate of carbon-bearing spheres. Chi et al. (2015) studied the
322 aging mechanism of sea salt particles. Here, we focused on **non-sea salt (NSS)**
323 **particles including** S-rich, soot, and OM particles **NSS-particles and**
324 **sulfate-containing particles account for $39 \pm 5\%$ and $29 \pm 7\%$ by number of all the 2002**
325 **particles analysed (Figure 3).**

326

327 **3.2 OM coating on sulfate particles**

328 TEM observations revealed a common core-shell mixing structure in fine
329 sulfate-containing particles (Figure 4a). Elemental mapping of such internally mixed
330 sulfate particles shows C signals in the coating (C map, Figure 4b) and S and O
331 signals in the center (S and O map, Figure 4c, d). The elemental line profile of a
332 sulfate particle also shows sulfate core and C coating (Figure S4). Furthermore, ion
333 maps of individual particles from the NanoSIMS further **show** $^{12}\text{C}^{14}\text{N}$ signals in the
334 coating (red color in Figure 4e, f) and ^{32}S signals in the core (green color in Figure 4e,
335 g). These results provide strong evidence that the coating is OM and the core is
336 sulfate.

337 A majority of **the 781 analysed** NSS-particles (74% by particle number) have a
338 sulfate core and OM coating (Figures 4 and 5). ~20% of them also contain small soot
339 inclusions but they only appeared in organic coating, rather than as the core (Figure
340 5b). The mixing structure is different from our previous findings in polluted air **where**
341 soot is normally mixed with sulfate instead of OM coating (Li et al., 2016). Moreover,
342 we **observed some** chain-like soot aggregates (1.3% in all **analysed** particles) (Figure

343 S5) but they only occurred in three samples during the whole sampling period (Table
344 1). Considering the remoteness of the sampling site, such fresh soot particles are
345 likely to be of local origin, for example, shipping and flaring (Gilgen et al., 2018;
346 Peters et al., 2011).

347 TEM observations showed that some sulfate-containing particles had unique
348 morphology that a sulfate particle was surrounded by some smaller particles (Figure
349 5a). They are often called “satellite” particles as they were distributed from the central
350 particles when impacted on the substrate during sample collection. Satellite particles
351 were observed around 16% of the analysed sulfate-containing particles (Figure 5a) in
352 the samples (Table 1) collected during 9-15 August. NanoSIMS analysis further
353 showed that the satellite particles (Table 1) have strong $^{32}\text{S}^-$ (Figure 6a, c) and $^{16}\text{O}^-$
354 signals (Figure 6d) as well as weak $^{12}\text{C}^{14}\text{N}^-$ signals (Figure 6a, b). Previous studies
355 showed that the similar satellite particles are normally considered as acidic sulfate
356 (Buseck and Posfai, 1999; Iwasaka et al., 1983). Our results show that these acidic
357 satellites not only contain sulfuric acid but also some OM or organic acids. Indeed, Fu
358 et al. (2008) found that polyacids are the most abundant organic compounds, followed
359 by phthalates, aromatic acids, and fatty acids in Arctic aerosol particles. Based on the
360 back trajectories of air masses and FLEXPART modelling, most air masses originate
361 in the North America and Greenland during the sampling periods (Figures 1 and 2).
362 Figure 1 shows that these air masses brought abundant sulfate-containing particles
363 into the sampling area in summertime.

364 AFM was used to obtain 3D image of individual secondary particles impacting on
365 the substrate. Figure 7a shows that the secondary particles normally have smooth
366 surface which is different from uneven surface of the Arctic fresh and aged NaCl
367 particles (Chi et al., 2015). Furthermore, we observed particle thickness through
368 tilting the specimen stage up to 75° in SEM. Figure 7a-b both shows that the
369 secondary particles look like thin pancake sticking on the substrate. Furthermore, the
370 sections of two secondary particles in the AFM images shows that the highest heights
371 of particles are only 15% (green line) and 26% (red line) of the corresponding
372 horizontal diameter (Figure 7a). These results show that the shape of individual

373 particles was modified when they impacted on the substrate following the airflow.
374 Therefore, the measured ECDs of individual particles in TEM images are much larger
375 than the real particle diameter. To calibrate the particle diameter, we obtained volume
376 of dry particles on the substrate and then calculated **their ESD** in the AFM images
377 (Figure 7c). ESD distribution of the secondary Arctic particles displayed a peak at 340
378 nm, ranging from 100 nm to 2000 nm (Figure 7d). The core particles, as sulfate or
379 soot, had a peak at 240 nm and 120 nm, respectively (Figure 7d). **It is estimated that**
380 **OM** on average accounted for $63 \pm 23\%$ of the dry sulfate-containing particle volume.
381 Our result shows that the OM volume increases following the particle size increase
382 (Figure S6).

383

384 **4. Discussion**

385 **4.1 Mixing mechanism of organic, soot, and sulfate**

386 Lagrangian particle dispersion modeling using the FLEXPART-WRF 3.1 showed
387 that air masses arriving at the sampling site during our field measurement periods
388 were likely originated from the Greenland and North America (Figure 2). Previous
389 studies reported that air masses from North America or Greenland during the summer
390 contain higher concentration of black carbon, OM, and sulfate (Burkart et al., 2017;
391 Chang et al., 2011; Fu et al., 2008; Moore et al., 2011; Park et al., 2013). Indeed, there
392 is strong emission intensity of OC and SO₂ around the Arctic area from emission
393 simulation as shown in Figure S2. However, Weinbruch et al. (2012) observed soot
394 particles when cruise ships were present in the area around Ny-Ålesund town. It is
395 possible that minor soot particles are from the ship emissions and most of them are
396 transported from **outside** Arctic area in the free troposphere (Figure S2).

397 The sulfate core-OM shell structure observed in the Arctic summer atmosphere is
398 similar to those in the background or rural air in other places (Li et al., 2016; Moffet
399 et al., 2013). Based on the images from electron microscopies, we can infer that OM
400 coating thickness in the Arctic air was comparable with them in rural places but
401 higher than them in urban places. During the transports, organic coatings on **sulfate**
402 were considered as the **SOM** and their masses increase following particle aging and

403 growth (Li et al., 2016; Moffet et al., 2013; Sierau et al., 2014). Figures 1 and 2 show
404 that most of particles in the air masses **are** transported from North American. **The**
405 sulfate/OM particles with soot inclusions are probably formed in a similar way as
406 those found elsewhere (Li et al., 2016) – e.g., soot particles may have acted as nuclei
407 for secondary sulfate or organic uptake during their transports (Riemer et al., 2009).
408 Similarly, besides the OM coating in the Arctic particles, Leck and Svensson (2015)
409 found biogenic aerosols like gel-aggregate containing bacterium in ultrafine particles.
410 However, we did **not** find any gel-like particles in the samples because our sampler
411 had very low efficiency for ultrafine particles.

412 TEM images show that most of the internally mixed sulfate-**containing** particles
413 display sulfate core and OM coating on the substrate (Figures 4a and 5b, c).
414 **Knowledge on the phase separation in individual particles is important to** understand
415 particle hygroscopic **properties**, heterogeneous reactions of reactive gases on particle
416 surface, and organic aging (You et al., 2012). **It is possible that** the thick OM coatings
417 were consistently built up during the long-range transport of sulfate-**containing**
418 particles and part of **the SOM** in the coating likely formed in Arctic area. Indeed, there
419 are various sources of organic precursors during the Arctic area, such as biogenic
420 VOCs from ice melting and open water (Dall'Osto et al., 2017) and anthropogenic
421 VOCs from shipping emissions in summertime (Gilgen et al., 2018). The dependence
422 of OM volume on particle size (Figure S6) suggests that the suspended sulfate
423 particles are initially important surface for **SOM** formation. Moreover, the presence of
424 OM coating **in 74%** sulfate particles indicates that **SOM** as the surfaces of fine
425 particles **may** govern the possible heterogeneous reactions between reactive gases and
426 sulfate-**containing** particles in the Arctic air.

427 It should be noted that most of **SOM** not only occurred on the surfaces of
428 sulfate-**containing** particles but also its mass (mean mass at $63 \pm 23\%$) dominated in
429 individual particles (Figure 7d). The OM dominating in individual particles can
430 influence the IN and CCN activities of secondary sulfate-containing particles (Latham
431 et al., 2013; Martin et al., 2011). For example, some studies found that an increase in
432 organic mass fraction in particles of a certain size would lead to a suppression of the

433 Arctic CCN activity (Leck and Svensson, 2015; Martin et al., 2011). Moreover, OM
434 as particle surfaces can significantly influence hygroscopicity and IN activity of
435 sulfate-containing particles (Wang et al., 2012).

436

437 **4.2 Potential impact of OM on optical properties of sulfate-containing particles**

438 The internal mixing of soot, sulfate, and OM can change optical properties of
439 individual particles in the atmosphere. Recent studies showed that BrC has been
440 detected in the OM in the polluted and clean air and even in upper troposphere
441 (Laskin et al., 2015; Wang et al., 2018). Feng et al. (2013) further calculated the
442 contribution up to 19% of the optical absorption of the strongly absorbing BrC in
443 global simulations which is after the absorption BC aerosols. Various colored OM (e.g.
444 nitrated/polycyclic aromatics and phenols), referred as BrC, were detected in the
445 Arctic atmosphere in different seasons (Fu et al., 2008; Währnschimmel et al., 2013;
446 Zangrando et al., 2013) and in surface ice or snowpack (Browse et al., 2013; Doherty
447 et al., 2013; Hegg et al., 2010). We also noticed that the $^{12}\text{C}^{14}\text{N}^-$ signal occurred in all
448 analysed OM coating in sulfate-containing particles (Figure 4e-f). $^{12}\text{C}^{14}\text{N}^-$ from
449 NanoSIMS is indicative of the presence of nitrogen-containing organic in the detected
450 materials (Herrmann et al., 2007). Figure 4 showed that the nitrogen-containing OM
451 was more or less homogeneously distributed in the OM coating in individual particles.
452 This suggests that some of the OM in the coating has the potential to act as BrC (Jiang
453 et al., 2019; Laskin et al., 2015).

454 To understand how OM coating influence optical properties of sulfate-containing
455 particles, we make an assumption that OM coating is strongly absorbing (case 1),
456 moderately absorbing (case 2) or non-absorbing BrC (case 3) with a refractive index
457 of 1.65-0.03i, 1.65-0.003i, and 1.65 at 550 nm according to Feng et al. (2013). Based
458 on the size measurements shown in Figure 7d, we can calculate volume of sulfate and
459 OM within each particle. We input volume of each component and the corresponding
460 refractive index into the Mie code and then calculated optical properties of individual
461 sulfate-containing particles in the samples. Based on optical data statistic of 575
462 particles, Figure 8a show that if the OM coating is strongly absorbing BrC (referred to

463 case Abs1), the average absorption cross section (ACS) of individual particles is
464 estimated to be $2.67 \times 10^{-14} \text{ m}^2$. This value is 8.30 times higher than the aerosol ACS
465 ($3.22 \times 10^{-15} \text{ m}^2$) when assuming that the BrC is moderately absorbing (referred to case
466 Abs2, Figure 8a). However, the scattering cross section (SCS) of individual particles
467 only shows a small change (Figure 8b). Figure 8c also shows that the single scattering
468 albedos (SSAs) of individual particles are 0.92, 0.99, and 1 when assuming the OM as
469 strongly, moderately and non-absorbing BrC (cases SSA1 to SSA3). These results
470 suggest that whether we consider organic coating as BrC may have a significant
471 influence on the absorption properties of individual sulfate-containing particles.

472 In this study, we explored the relationship between ACS of individual particles
473 and particle diameters. Interestingly, Figure 8d shows that ACS of individual fine
474 OM-coating sulfate particles increased with particle size. This suggests that the ACS
475 of individual particles may increase as they grow and age in the atmosphere.

476 Current climate models estimated the radiative force of Arctic BC (Sand et al.,
477 2013; Shindell, 2007; Winiger et al., 2017; Zanatta et al., 2018), but none specifically
478 considered optical properties of Arctic BrC. Our study revealed the OM coating on
479 individual sulfate particles, which should be considered in aerosol radiation effect and
480 cloud-aerosol interaction simulations in the models.

481

482 5 Summary

483 A range of individual particle observation techniques, such as TEM/EDS, STEM,
484 SEM, NanoSIMS, and AFM, were applied to study S-rich, soot, and OM particles in
485 the Arctic atmosphere in the summer 2012. Sulfate-containing particles account for
486 approximately $29 \pm 7\%$ by number of all analysed particles. TEM and NanoSIMS
487 showed that individual sulfate-containing particles have OM coating with sulfate as
488 core. The SOM on the surfaces of fine particles may affect heterogeneous reactions
489 between reactive gases and sulfate particles in the Arctic air. Furthermore, 20% of the
490 sulfate-containing particles also contain small soot inclusions but they only appeared
491 in organic coating.

492 Size distribution of the secondary Arctic particles displayed a peak at 340 nm,

493 ranging from 100 nm to 2000 nm. The core particles, as sulfate or soot, had a peak at
494 240 nm and 120 nm, respectively. OM coating on average contribute $63 \pm 23\%$ by
495 volume to the dry NSS-particles. We also found that whether the OM coating may
496 have a significant influence on the absorption properties of individual particles in the
497 Arctic air, depending on the optical properties of the OM.
498

499 **Author Contributions:** WL and ZS designed the study. YZ and XS collected aerosol
500 particles. WL, HY, and JZ contributed laboratory experiments and data analysis. HY
501 and WL performed optical calculation and wrote part of first draft. PT and MD
502 provided the online measurement data of new particle formation and growth. JS and
503 XZ coordinated the field campaign. All authors commented and edited the paper.

504

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506

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References

- Abbatt, J.P.D., Leaitch, W.R., Aliabadi, A.A., Bertram, A.K., Blanchet, J.P., Boivin-Rioux, A., Bozem, H., Burkart, J., Chang, R.Y.W., Charette, J., Chaubey, J.P., Christensen, R.J., Cirisan, A., Collins, D.B., Croft, B., Dionne, J., Evans, G.J., Fletcher, C.G., Gal í M., Ghahremaninezhad, R., Girard, E., Gong, W., Gosselin, M., Gourdal, M., Hanna, S.J., Hayashida, H., Herber, A.B., Hesaraki, S., Hoor, P., Huang, L., Hussherr, R., Irish, V.E., Keita, S.A., Kodros, J.K., Köllner, F., Kolonjari, F., Kunkel, D., Ladino, L.A., Law, K., Levasseur, M., Libois, Q., Liggio, J., Lizotte, M., Macdonald, K.M., Mahmood, R., Martin, R.V., Mason, R.H., Miller, L.A., Moravek, A., Mortenson, E., Mungall, E.L., Murphy, J.G., Namazi, M., Norman, A.L., O'Neill, N.T., Pierce, J.R., Russell, L.M., Schneider, J., Schulz, H., Sharma, S., Si, M., Staebler, R.M., Steiner, N.S., Thomas, J.L., von Salzen, K., Wentzell, J.J.B., Willis, M.D., Wentworth, G.R., Xu, J.W., Yakobi-Hancock, J.D.: Overview paper: New insights into aerosol and climate in the Arctic, *Atmos. Chem. Phys.*, 19 (4), 2527-2560, 2019.
- Alexander, D.T.L., Crozier, P.A., Anderson, J.R.: Brown Carbon Spheres in East Asian Outflow and Their Optical Properties, *Science*, 321 (5890), 833-836, 2008.
- Andreae, M.O., Gelencser, A.: Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, *Atmos. Chem. Phys.*, 6 (10), 3131-3148, 2006.
- Behrenfeldt, U., Krejci, R., Ström, J., Stohl, A.: Chemical properties of Arctic aerosol particles collected at the Zeppelin station during the aerosol transition period in May and June of 2004, *Tellus B*, 60 (3), 405-415, 2008.
- Bohren, C.F., Huffman, D.R., 1983. Absorption and scattering of light by small particles. John Wiley & Sons, Inc. , New York, USA.
- Bond, T.C., Doherty, S.J., Fahey, D.W., Forster, P.M., Berntsen, T., DeAngelo, B.J., Flanner, M.G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P.K., Sarofim, M.C., Schultz, M.G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S.K., Hopke, P.K., Jacobson, M.Z., Kaiser, J.W., Klimont, Z., Lohmann, U., Schwarz, J.P., Shindell, D., Storelvmo, T., Warren, S.G., Zender, C.S.: Bounding the role of black carbon in the climate system: A scientific assessment, *J. Geophys. Res.*, 118 (11), 5380-5552, 2013.
- Brioude, J., Arnold, D., Stohl, A., Cassiani, M., Morton, D., Seibert, P., Angevine, W., Evan, S., Dingwell, A., Fast, J.D., Easter, R.C., Pisso, I., Burkhart, J., Wotawa, G.: The Lagrangian particle dispersion model FLEXPART-WRF version 3.1, *Geosci. Model Dev.*, 6 (6), 1889-1904, 2013.
- Brock, C.A., Cozic, J., Bahreini, R., Froyd, K.D., Middlebrook, A.M., McComiskey, A., Brioude, J., Cooper, O.R., Stohl, A., Aikin, K.C., de Gouw, J.A., Fahey, D.W., Ferrare, R.A., Gao, R.S., Gore, W., Holloway, J.S., Hübler, G., Jefferson, A., Lack, D.A., Lance, S., Moore, R.H., Murphy, D.M., Nenes, A., Novelli, P.C., Nowak, J.B., Ogren, J.A., Peischl, J., Pierce, R.B., Pilewskie, P., Quinn, P.K., Ryerson, T.B., Schmidt, K.S., Schwarz, J.P., Sodemann, H., Spackman, J.R., Stark, H., Thomson, D.S., Thornberry, T., Veres, P., Watts, L.A., Warneke, C., Wollny, A.G.: Characteristics, sources, and transport of aerosols measured in spring 2008 during the aerosol, radiation, and cloud processes affecting Arctic Climate (ARCPAC) Project, *Atmos. Chem. Phys.*, 11 (6), 2423-2453, 2011.
- Browse, J., Carslaw, K.S., Schmidt, A., Corbett, J.J.: Impact of future Arctic shipping on high-latitude black carbon deposition, *Geophys. Res. Lett.*, 40 (16), 4459-4463, 2013.
- Burkart, J., Willis, M.D., Bozem, H., Thomas, J.L., Law, K., Hoor, P., Aliabadi, A.A., Köllner, F., Schneider, J., Herber, A., Abbatt, J.P.D., Leaitch, W.R.: Summertime observations of elevated levels of ultrafine particles in the high Arctic marine boundary layer, *Atmos. Chem. Phys.*, 17 (8),

- 5515-5535, 2017.
- Buseck, P.R., Posfai, M.: Airborne minerals and related aerosol particles: Effects on climate and the environment, *P. Natl. Acad. Sci. USA*, 96 (7), 3372-3379, 1999.
- Chang, R.Y.W., Leck, C., Graus, M., Müller, M., Paatero, J., Burkhardt, J.F., Stohl, A., Orr, L.H., Hayden, K., Li, S.M., Hansel, A., Tjernström, M., Leaitch, W.R., Abbatt, J.P.D.: Aerosol composition and sources in the central Arctic Ocean during ASCOS, *Atmos. Chem. Phys.*, 11 (20), 10619-10636, 2011.
- 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., Wang, W.X.: Sea salt aerosols as a reactive surface for inorganic and organic acidic gases in the Arctic troposphere, *Atmos. Chem. Phys.*, 15 (19), 11341-11353, 2015.
- Dagsson-Waldhauserova, P., Arnalds, O., Olafsson, H.: Long-term frequency and characteristics of dust storm events in Northeast Iceland (1949–2011), *Atmos. Environ.*, 77 (0), 117-127, 2013.
- Dall'Osto, M., Beddows, D.C.S., Tunved, P., Krejci, R., Ström, J., Hansson, H.C., Yoon, Y.J., Park, K.-T., Becagli, S., Udisti, R., Onasch, T., O'Dowd, C.D., Simó, R., Harrison, R.M.: Arctic sea ice melt leads to atmospheric new particle formation, *Sci. Rep.*, 7 (1), 3318, 2017.
- Doherty, S.J., Grenfell, T.C., Forsström, S., Hegg, D.L., Brandt, R.E., Warren, S.G.: Observed vertical redistribution of black carbon and other insoluble light-absorbing particles in melting snow, *J. Geophys. Res.*, 118 (11), 5553-5569, 2013.
- Feng, Y., Ramanathan, V., Kotamarthi, V.R.: Brown carbon: a significant atmospheric absorber of solar radiation?, *Atmos. Chem. Phys.*, 13 (17), 8607-8621, 2013.
- Fu, P., Kawamura, K., Barrie, L.A.: Photochemical and Other Sources of Organic Compounds in the Canadian High Arctic Aerosol Pollution during Winter–Spring, *Environ. Sci. Technol.*, 43 (2), 286-292, 2008.
- Geng, H., Ryu, J., Jung, H.-J., Chung, H., Ahn, K.-H., Ro, C.-U.: Single-Particle Characterization of Summertime Arctic Aerosols Collected at Ny-Alesund, Svalbard, *Environ. Sci. Technol.*, 44 (7), 2348-2353, 2010.
- Ghosal, S., Weber, P.K., Laskin, A.: Spatially resolved chemical imaging of individual atmospheric particles using nanoscale imaging mass spectrometry: insight into particle origin and chemistry, *Analytical Methods*, 6 (8), 2444-2451, 2014.
- Gilgen, A., Huang, W.T.K., Ickes, L., Neubauer, D., Lohmann, U.: How important are future marine and shipping aerosol emissions in a warming Arctic summer and autumn?, *Atmos. Chem. Phys.*, 18 (14), 10521-10555, 2018.
- Hansen, J., Nazarenko, L.: Soot climate forcing via snow and ice albedos, *P. Natl. Acad. Sci. USA*, 101 (2), 423-428, 2004.
- Hara, K., Yamagata, S., Yamanouchi, T., Sato, K., Herber, A., Iwasaka, Y., Nagatani, M., Nakata, H.: Mixing states of individual aerosol particles in spring Arctic troposphere during ASTAR 2000 campaign, *J. Geophys. Res.*, 108 (D7), 2003.
- Hegg, D.A., Warren, S.G., Grenfell, T.C., Sarah, J.D., Clarke, A.D.: Sources of light-absorbing aerosol in arctic snow and their seasonal variation, *Atmos. Chem. Phys.*, 10 (22), 10923-10938, 2010.
- Herrmann, A.M., Ritz, K., Nunan, N., Clode, P.L., Pett-Ridge, J., Kilburn, M.R., Murphy, D.V., O'Donnell, A.G., Stockdale, E.A.: Nano-scale secondary ion mass spectrometry — A new analytical tool in biogeochemistry and soil ecology: A review article, *Soil Biol. Biochem.*, 39 (8), 1835-1850, 2007.
- IPCC (2013), Clouds and Aerosols, in *Climate Change 2013: The Physical Science Basis. Contribution*

- of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, 571-657 pp, Cambridge, U.K. and New York, NY.
- Iwasaka, Y., Minoura, H., Nagaya, K.: The transport and spatial scale of Asian dust-storm clouds: A case study of the dust-storm event of April 1979, *Tellus, Ser. B*, 35, 189-196, 1983.
- Iziomon, M.G., Lohmann, U., Quinn, P.K.: Summertime pollution events in the Arctic and potential implications, *J. Geophys. Res.*, 111 (D12), D12206, 2006.
- Jacob, D.J., Crawford, J.H., Maring, H., Clarke, A.D., Dibb, J.E., Emmons, L.K., Ferrare, R.A., Hostetler, C.A., Russell, P.B., Singh, H.B., Thompson, A.M., Shaw, G.E., McCauley, E., Pederson, J.R., Fisher, J.A.: The Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) mission: design, execution, and first results, *Atmos. Chem. Phys.*, 10 (11), 5191-5212, 2010.
- Jiang, H., Frie, A.L., Lavi, A., Chen, J.Y., Zhang, H., Bahreini, R., Lin, Y.-H.: Brown Carbon Formation from Nighttime Chemistry of Unsaturated Heterocyclic Volatile Organic Compounds, *Environ. Sci. Techn. Lett.*, DOI: 10.1021/acs.estlett.1029b00017, 2019.
- Karl, M., Leck, C., Coz, E., Heintzenberg, J.: Marine nanogels as a source of atmospheric nanoparticles in the high Arctic, *Geophys. Res. Lett.*, 40 (14), 3738-3743, 2013.
- Kirpes, R.M., Bondy, A.L., Bonanno, D., Moffet, R.C., Wang, B., Laskin, A., Ault, A.P., Pratt, K.A.: Secondary sulfate is internally mixed with sea spray aerosol and organic aerosol in the winter Arctic, *Atmos. Chem. Phys.*, 18 (6), 3937-3949, 2018.
- Koch, D., Hansen, J.: Distant origins of Arctic black carbon: A Goddard Institute for Space Studies ModelE experiment, *J. Geophys. Res.*, 110 (D4), D04204, 2005.
- Lack, D.A., Langridge, J.M., Bahreini, R., Cappa, C.D., Middlebrook, A.M., Schwarz, J.P.: Brown carbon and internal mixing in biomass burning particles, *P. Natl. Acad. Sci. USA*, 109 (37), 14802-14807, 2012.
- Laskin, A., Laskin, J., Nizkorodov, S.A.: Chemistry of Atmospheric Brown Carbon, *Chem. Rev.*, 115 (10), 4355-4382, 2015.
- Laskina, O., Morris, H.S., Grandquist, J.R., Estillore, A.D., Stone, E.A., Grassian, V.H., Tivanski, A.V.: Substrate-Deposited Sea Spray Aerosol Particles: Influence of Analytical Method, Substrate, and Storage Conditions on Particle Size, Phase, and Morphology, *Environ. Sci. Tech.*, 49 (22), 13447-13453, 2015.
- Latham, T.L., Beyersdorf, A.J., Thornhill, K.L., Winstead, E.L., Cubison, M.J., Hecobian, A., Jimenez, J.L., Weber, R.J., Anderson, B.E., Nenes, A.: Analysis of CCN activity of Arctic aerosol and Canadian biomass burning during summer 2008, *Atmos. Chem. Phys.*, 13 (5), 2735-2756, 2013.
- Law, K.S., Stohl, A.: Arctic Air Pollution: Origins and Impacts, *Science*, 315 (5818), 1537-1540, 2007.
- Leck, C., Bigg, E.K.: Comparison of sources and nature of the tropical aerosol with the summer high Arctic aerosol, *Tellus, Ser. B*, 60 (1), 118-126, 2008.
- Leck, C., Svensson, E.: Importance of aerosol composition and mixing state for cloud droplet activation over the Arctic pack ice in summer, *Atmos. Chem. Phys.*, 15 (5), 2545-2568, 2015.
- Li, W., Sun, J., Xu, L., Shi, Z., Riemer, N., Sun, Y., Fu, P., Zhang, J., Lin, Y., Wang, X., Shao, L., Chen, J., Zhang, X., Wang, Z., Wang, W.: A conceptual framework for mixing structures in individual aerosol particles, *J. Geophys. Res.*, 121 (22), 13784-713,798, 2016.
- Li, W., Xu, L., Liu, X., Zhang, J., Lin, Y., Yao, X., Gao, H., Zhang, D., Chen, J., Wang, W., Harrison, R.M., Zhang, X., Shao, L., Fu, P., Nenes, A., Shi, Z.: Air pollution-aerosol interactions produce more bioavailable iron for ocean ecosystems, *Sci. Adv.*, 3 (3), e1601749, 2017.

- Maahn, M., de Boer, G., Creamean, J.M., Feingold, G., McFarquhar, G.M., Wu, W., Mei, F.: The observed influence of local anthropogenic pollution on northern Alaskan cloud properties, *Atmos. Chem. Phys.*, 17 (23), 14709-14726, 2017.
- Martin, M., Chang, R.Y.W., Sierau, B., Sjogren, S., Swietlicki, E., Abbatt, J.P.D., Leck, C., Lohmann, U.: Cloud condensation nuclei closure study on summer arctic aerosol, *Atmos. Chem. Phys.*, 11 (22), 11335-11350, 2011.
- Moffet, R.C., Rödel, T.C., Kelly, S.T., Yu, X.Y., Carroll, G.T., Fast, J., Zaveri, R.A., Laskin, A., Gilles, M.K.: Spectro-microscopic measurements of carbonaceous aerosol aging in Central California, *Atmos. Chem. Phys.*, 13 (20), 10445-10459, 2013.
- Moore, R.H., Bahreini, R., Brock, C.A., Froyd, K.D., Cozic, J., Holloway, J.S., Middlebrook, A.M., Murphy, D.M., Nenes, A.: Hygroscopicity and composition of Alaskan Arctic CCN during April 2008, *Atmos. Chem. Phys.*, 11 (22), 11807-11825, 2011.
- Moroni, B., Cappelletti, D., Crocchianti, S., Becagli, S., Caiazzo, L., Traversi, R., Udusti, R., Mazzola, M., Markowicz, K., Ritter, C., Zielinski, T.: Morphochemical characteristics and mixing state of long range transported wildfire particles at Ny-Ålesund (Svalbard Islands), *Atmos. Environ.*, 156, 135-145, 2017.
- Park, K., Kim, G., Kim, J.-s., Yoon, Y.-J., Cho, H.-j., Ström, J.: Mixing State of Size-Selected Submicrometer Particles in the Arctic in May and September 2012, *Environ. Sci. Technol.*, 48 (2), 909-919, 2013.
- Peters, G.P., Nilssen, T.B., Lindholt, L., Eide, M.S., Glomsrød, S., Eide, L.I., Fuglestad, J.S.: Future emissions from shipping and petroleum activities in the Arctic, *Atmos. Chem. Phys.*, 11 (11), 5305-5320, 2011.
- Qi, L., Li, Q., Li, Y., He, C.: Factors controlling black carbon distribution in the Arctic, *Atmos. Chem. Phys.*, 17 (2), 1037-1059, 2017.
- Quinn, P.K., Shaw, G., Andrews, E., Dutton, E.G., Ruoho-Airola, T., Gong, S.L.: Arctic haze: current trends and knowledge gaps, *Tellus B*, 59 (1), 99-114, 2007.
- Raatikainen, T., Brus, D., Hyvärinen, A.P., Svensson, J., Asmi, E., Lihavainen, H.: Black carbon concentrations and mixing state in the Finnish Arctic, *Atmos. Chem. Phys.*, 15 (17), 10057-10070, 2015.
- Riener, N., West, M., Zaveri, R.A., Easter, R.C.: Simulating the evolution of soot mixing state with a particle resolved aerosol model, *J. Geophys. Res.*, 114, doi:10.1029/2008JD011073, 2009.
- Riener, N., Ault, A.P., West, M., Craig, R.L., Curtis, J.H.: Aerosol Mixing State: Measurements, Modeling, and Impacts, *Rev. Geophys.*, 57, <https://doi.org/10.1029/2018RG000615>, 2019.
- Saleh, R., Hennigan, C.J., McMeeking, G.R., Chuang, W.K., Robinson, E.S., Coe, H., Donahue, N.M., Robinson, A.L.: Absorptivity of brown carbon in fresh and photo-chemically aged biomass-burning emissions, *Atmos. Chem. Phys.*, 13 (15), 7683-7693, 2013.
- Samset, B.H., Myhre, G., Herber, A., Kondo, Y., Li, S.M., Moteki, N., Koike, M., Oshima, N., Schwarz, J.P., Balkanski, Y., Bauer, S.E., Bellouin, N., Bernsten, T.K., Bian, H., Chin, M., Diehl, T., Easter, R.C., Ghan, S.J., Iversen, T., Kirkevåg, A., Lamarque, J.F., Lin, G., Liu, X., Penner, J.E., Schulz, M., Seland, Ø., Skeie, R.B., Stier, P., Takemura, T., Tsigaridis, K., Zhang, K.: Modelled black carbon radiative forcing and atmospheric lifetime in AeroCom Phase II constrained by aircraft observations, *Atmos. Chem. Phys.*, 14 (22), 12465-12477, 2014.
- Sand, M., Bernsten, T.K., Kay, J.E., Lamarque, J.F., Seland, Ø., Kirkevåg, A.: The Arctic response to remote and local forcing of black carbon, *Atmos. Chem. Phys.*, 13 (1), 211-224, 2013.

- Seinfeld, J., Pandis, S., 2006. *Atmospheric Chemistry and Physics: From air pollution to climate change* (2nd ed.). 1-1203 pp., John Wiley & Son, Inc., Hoboken, New Jersey.
- Shindell, D.: Local and remote contributions to Arctic warming, *Geophys. Res. Lett.*, 34 (14), L14704, 2007.
- Sierau, B., Chang, R.Y.W., Leck, C., Paatero, J., Lohmann, U.: Single-particle characterization of the high-Arctic summertime aerosol, *Atmos. Chem. Phys.*, 14 (14), 7409-7430, 2014.
- Tobo, Y., Adachi, K., DeMott, P.J., Hill, T.C.J., Hamilton, D.S., Mahowald, N.M., Nagatsuka, N., Ohata, S., Uetake, J., Kondo, Y., Koike, M.: Glacially sourced dust as a potentially significant source of ice nucleating particles, *Nature Geoscience*, 12 (4), 253-258, 2019.
- Updyke, K.M., Nguyen, T.B., Nizkorodov, S.A.: Formation of brown carbon via reactions of ammonia with secondary organic aerosols from biogenic and anthropogenic precursors, *Atmos. Environ.*, 63 (0), 22-31, 2012.
- Wärnschimmel, H., MacLeod, M., Hungerbühler, K.: Emissions, Fate and Transport of Persistent Organic Pollutants to the Arctic in a Changing Global Climate, *Environ. Sci. Technol.*, 47 (5), 2323-2330, 2013.
- Wang, B., Laskin, A., Roedel, T., Gilles, M.K., Moffet, R.C., Tivanski, A.V., Knopf, D.A.: Heterogeneous ice nucleation and water uptake by field-collected atmospheric particles below 273 K, *J. Geophys. Res.*, 117, 2012.
- Wang, X., Heald, C.L., Liu, J., Weber, R.J., Campuzano-Jost, P., Jimenez, J.L., Schwarz, J.P., Perring, A.E.: Exploring the observational constraints on the simulation of brown carbon, *Atmos. Chem. Phys.*, 18 (2), 635-653, 2018.
- Wang, Z., Bi, L., Yi, B., Zhang, X.: How the Inhomogeneity of Wet Sea Salt Aerosols Affects Direct Radiative Forcing, *Geophysical Research Letters*, 46 (3), 1805-1813, 2019.
- Weinbruch, S., Wiesemann, D., Ebert, M., Schütze, K., Kallenborn, R., Ström, J.: Chemical composition and sources of aerosol particles at Zeppelin Mountain (Ny Ålesund, Svalbard): An electron microscopy study, *Atmos. Environ.*, 49 (0), 142-150, 2012.
- Winiger, P., Andersson, A., Eckhardt, S., Stohl, A., Gustafsson, Ö.: The sources of atmospheric black carbon at a European gateway to the Arctic, *Nat. Commun.*, 7, 12776, 2016.
- Winiger, P., Andersson, A., Eckhardt, S., Stohl, A., Semiletov, I.P., Dudarev, O.V., Charkin, A., Shakhova, N., Klimont, Z., Heyes, C., Gustafsson, Ö.: Siberian Arctic black carbon sources constrained by model and observation, *P. Natl. Acad. Sci. USA*, 114 (7), E1054-E1061, 2017.
- Xu, J.W., Martin, R.V., Morrow, A., Sharma, S., Huang, L., Leaitch, W.R., Burkart, J., Schulz, H., Zanutta, M., Willis, M.D., Henze, D.K., Lee, C.J., Herber, A.B., Abbatt, J.P.D.: Source attribution of Arctic black carbon constrained by aircraft and surface measurements, *Atmos. Chem. Phys.*, 17 (19), 11971-11989, 2017.
- Yang, Y., Wang, H., Smith, S.J., Easter, R.C., Rasch, P.J.: Sulfate Aerosol in the Arctic: Source Attribution and Radiative Forcing, *J. Geophys. Res.*, 123 (3), 1899-1918, 2018.
- You, Y., Renbaum-Wolff, L., Carreras-Sospedra, M., Hanna, S.J., Hiranuma, N., Kamal, S., Smith, M.L., Zhang, X., Weber, R.J., Shilling, J.E., Dabdub, D., Martin, S.T., Bertram, A.K.: Images reveal that atmospheric particles can undergo liquid-liquid phase separations, *P. Natl. Acad. Sci. USA*, 109 (33), 13188-13193, 2012.
- Zanutta, M., Laj, P., Gysel, M., Baltensperger, U., Vratolis, S., Eleftheriadis, K., Kondo, Y., Dubuisson, P., Winiarek, V., Kazadzis, S., Tunved, P., Jacobi, H.W.: Effects of mixing state on optical and radiative properties of black carbon in the European Arctic, *Atmos. Chem. Phys.*, 18 (19),

14037-14057, 2018.

Zangrando, R., Barbaro, E., Zennaro, P., Rossi, S., Kehrwald, N.M., Gabrieli, J., Barbante, C., Gambaro, A.: Molecular Markers of Biomass Burning in Arctic Aerosols, *Environ. Sci. Technol.*, 47 (15), 8565-8574, 2013.

Figure Captions

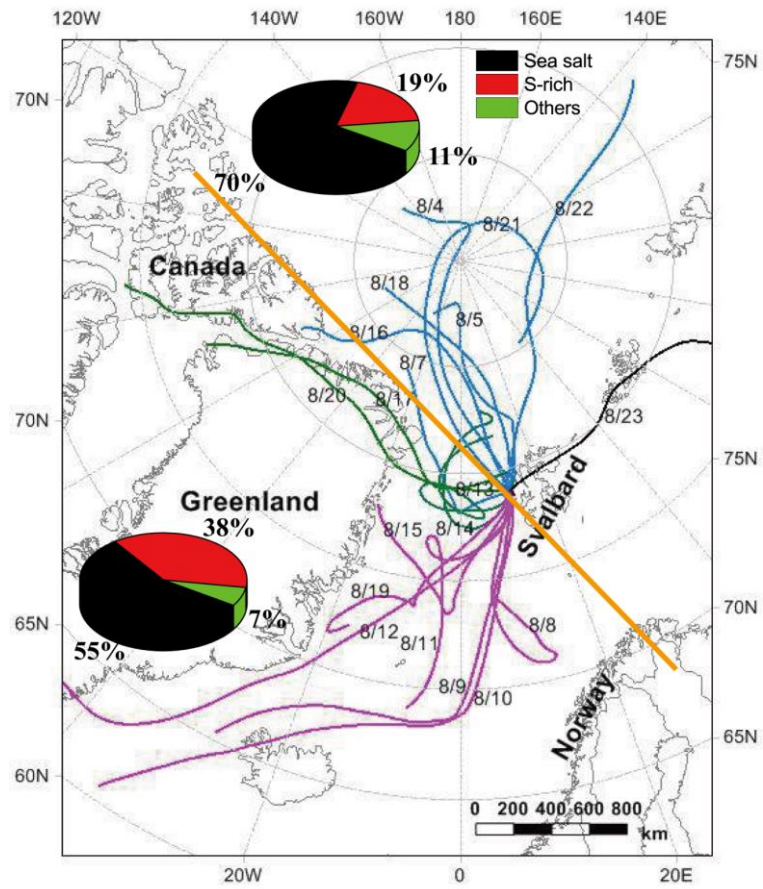


Figure 1 72 h back trajectories of air masses at 500 m over Arctic Yellow River Station in Svalbard during 3–26 August 2012, and arriving time was set according to the sampling time. Air masses were divided into two groups by the yellow line: one group from the central Arctic Ocean and the other one from North America and Greenland. Pie charts show the number fractions of sea salt, S-rich, and other particles.

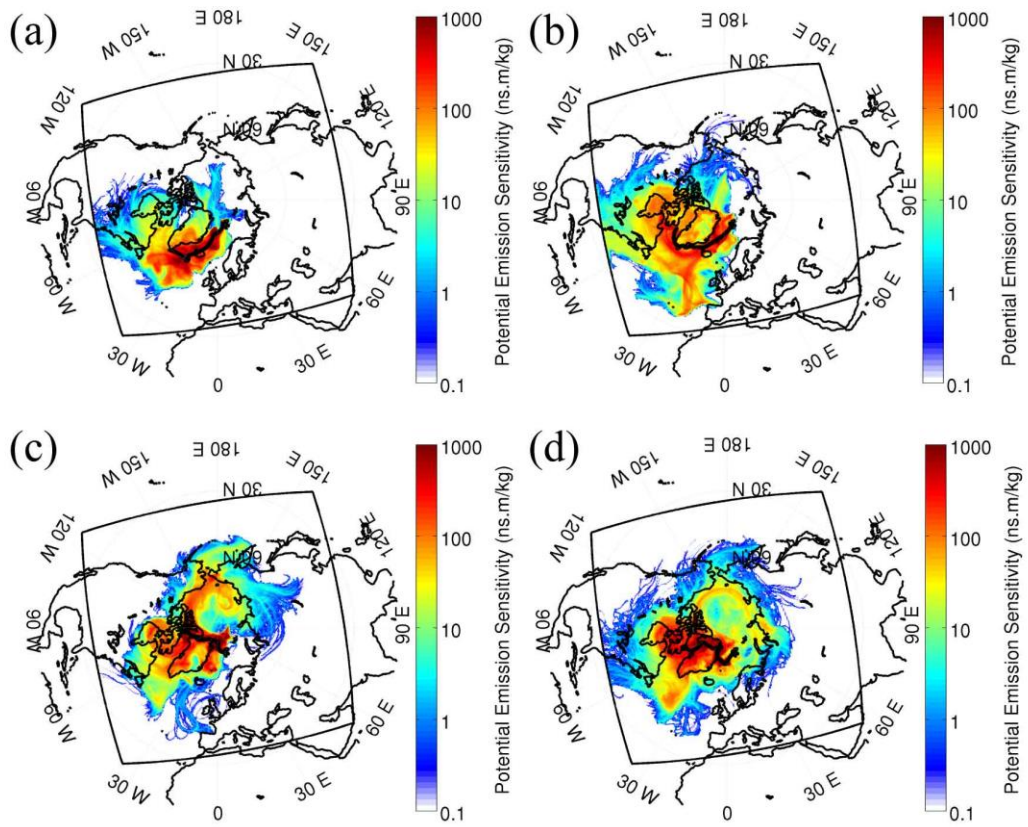


Figure 2 FLEXPART-WRF on August 11, 12, 14, and 15, 2012. Black square shows the WRF domain used to initiate the FLEXPART-WRF simulation.

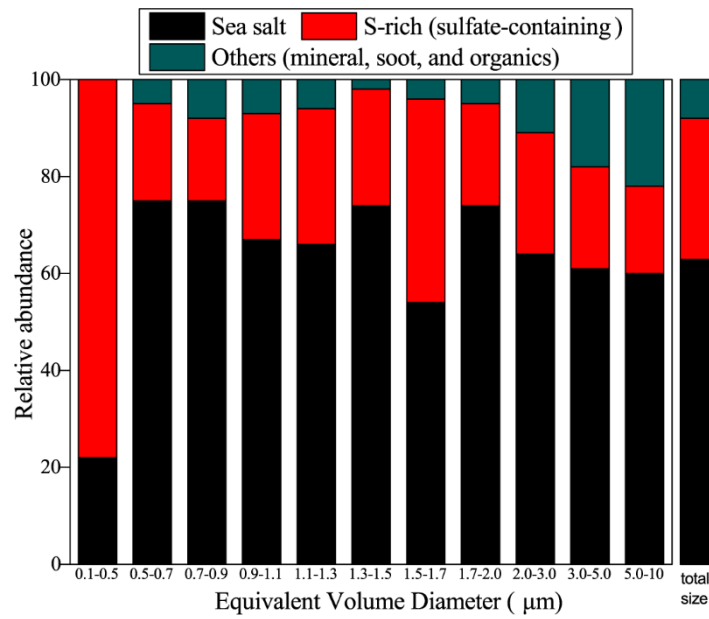


Figure 3 Relative abundances of typical individual aerosol particles in the analysed samples. Sulfate-containing particles include all particles that are internal mixture of sulfate and OM, with or without soot.

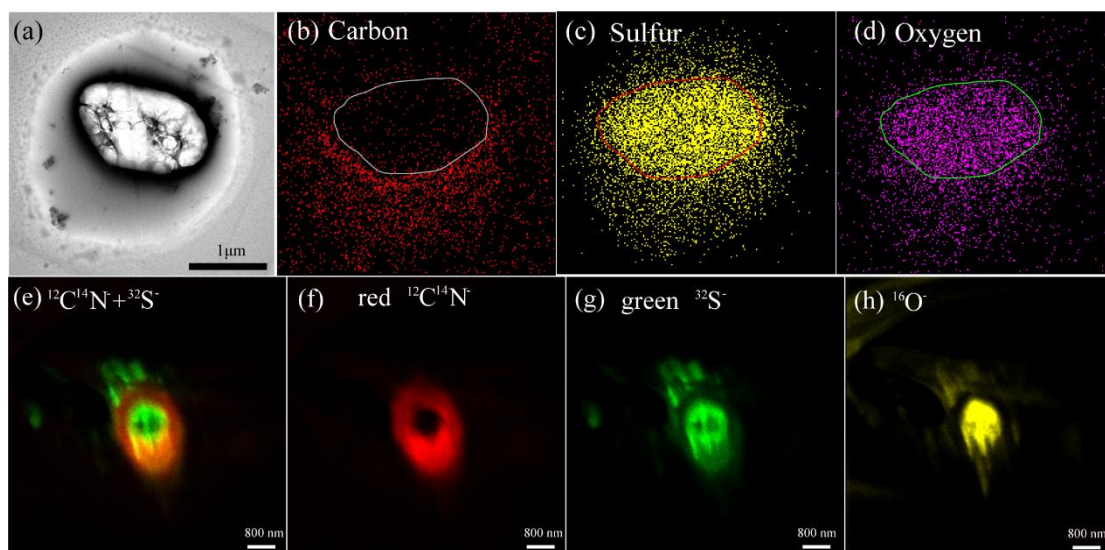


Figure 4 TEM images of a secondary particle and NanoSIMS intensity threshold maps of an aerosol particle with sulfate core and OM coating. (a) Bright-field TEM image of an internally mixed particle; (b) elemental carbon (c) sulfur and (d) oxygen maps of the internally mixed particle shown in 1(a); (e) Overlay of $^{12}\text{C}^{14}\text{N}^-$ and $^{32}\text{S}^-$ ion maps in an internally mixed particle; (f) CN^- map (g) S^- (h) O^- secondary ion maps. Ion maps with a set of aerosol particles were shown in Figure S1.

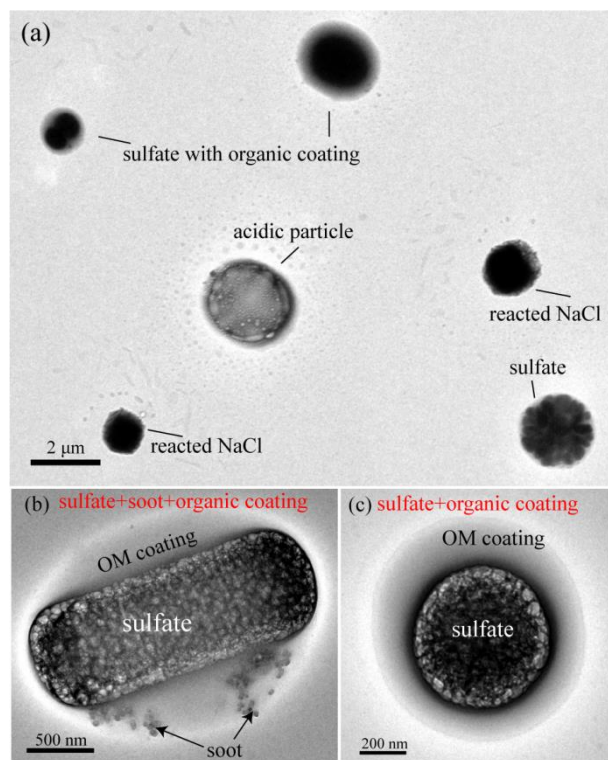


Figure 5 TEM images of individual particles containing sulfate, OM, and soot. (a) Low magnification TEM image showing sulfates, sulfate with OM coating, and reacted NaCl particles. (b) an internally mixed particle of sulfate and soot with OM coating (c) a particle with sulfate core and OM coating.

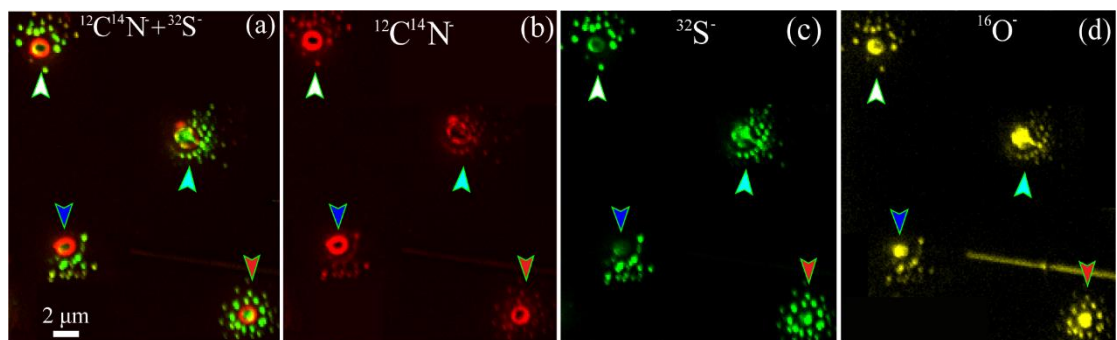


Figure 6 NanoSIMS intensity threshold maps of individual aerosol particles surrounded by satellite particles. (e) Overlay of $^{12}\text{C}^{14}\text{N}^-$ and $^{32}\text{S}^-$ ion maps of individual particles. (f) CN^- (g) S^- (h) O^- maps. Four particles were indicated by white, pink, blue, and red arrows.

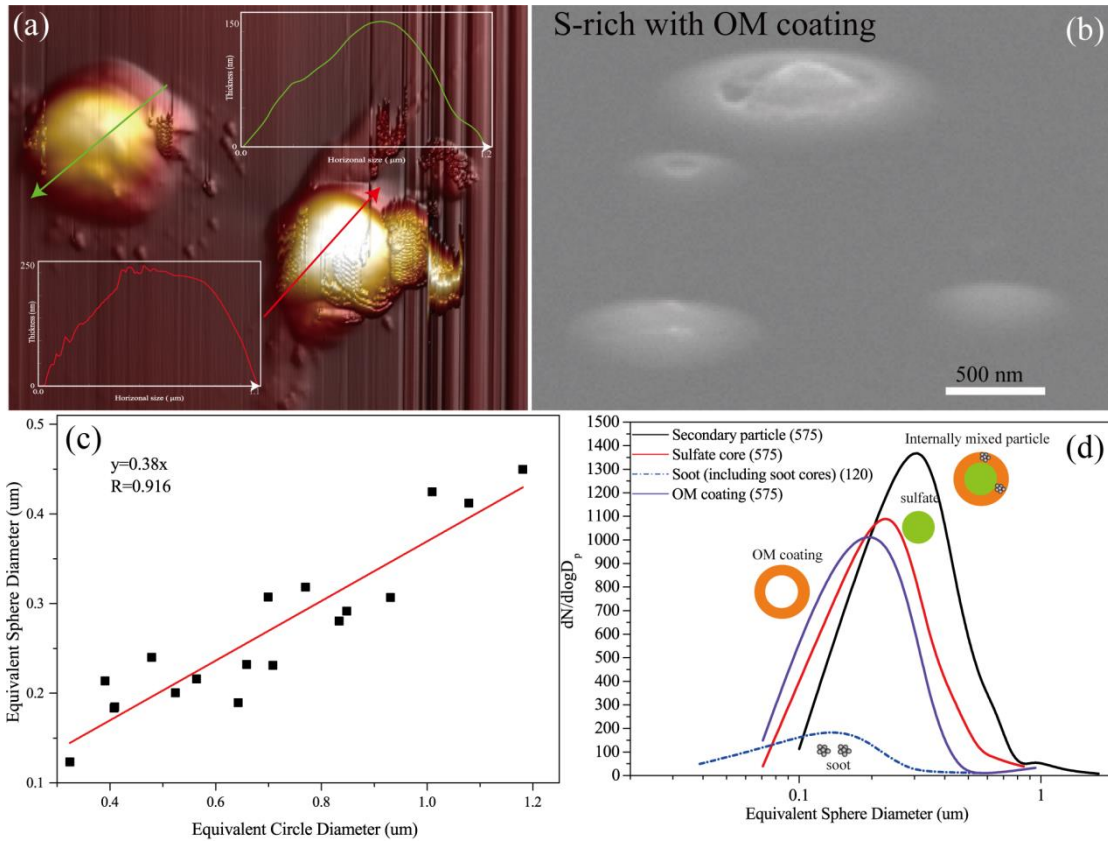


Figure 7 AFM image and calculated diameter of individual particles. (a) 3-D AFM image of sulfate-containing particles. The colored arrows represent the cross sections where the particles heights are measured (see insert figures). (b) SEM image of S-rich particles with OM coating obtained from 75° tilt of the SEM specimen stage (c) The near linear relationships between ECD and ESD based on S-rich particles with OM coating by atomic force microscopy. (d) Size distribution of individual particle with OM coating and sulfate core based on the estimated ESD diameter from TEM image.

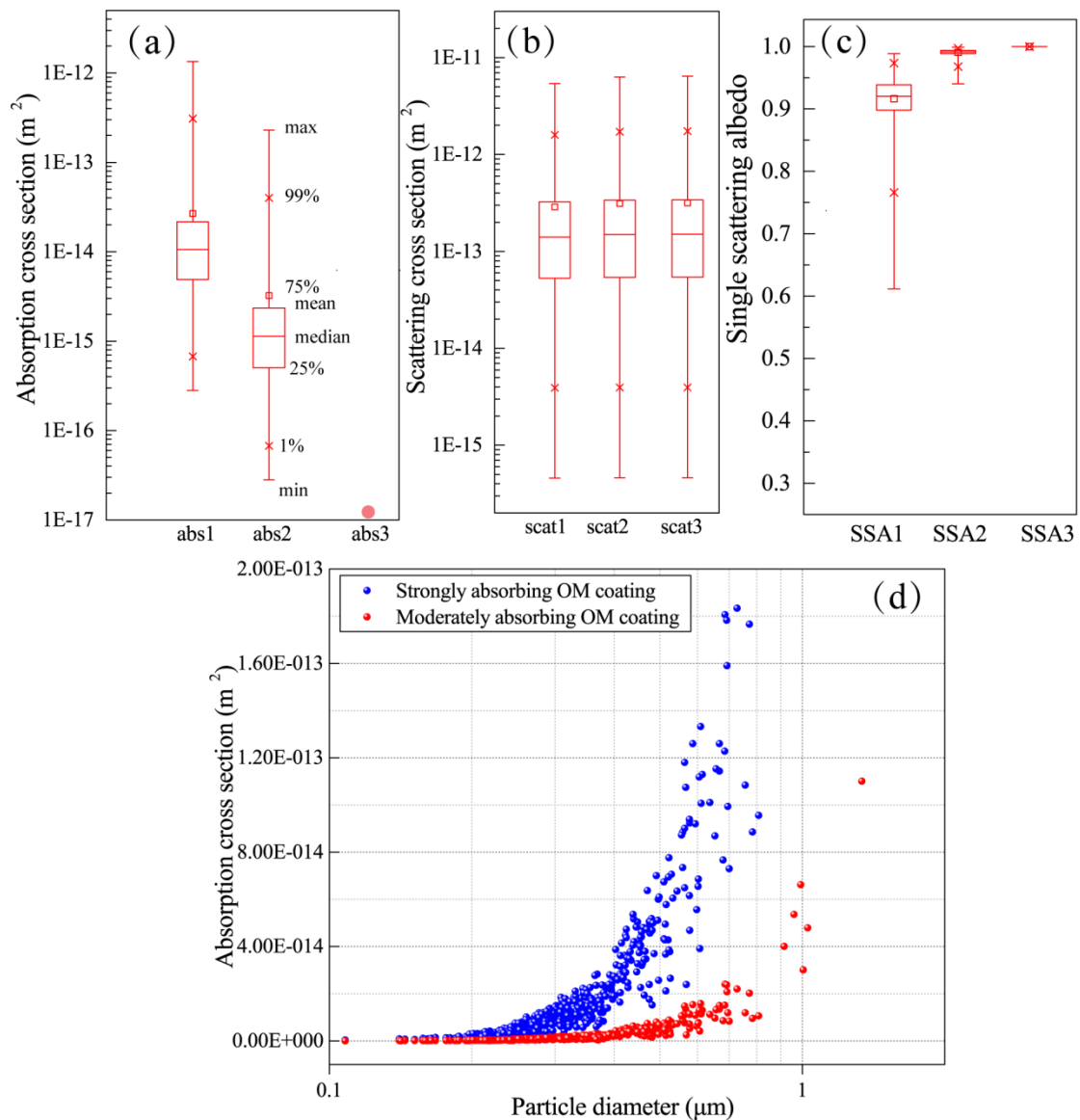


Figure 8 Optical properties of box-and-whisker plots showing optical parameters of all analysed particles assuming sulfate core and BrC shell (not considering soot cores in the particles). (a) Scattering cross section (b) Absorption cross section (c) Single scattering albedo. Top to bottom markers in the box-and-whisker represent max, 99%, 75%, mean, median, 25%, 1%, min values. (d) Absorption cross section along with particle diameter assuming strongly absorbing BrC and Moderate absorbing BrC as the particle OM coating. Abs 1, abs 2 and abs3 represent the BrC with highly, moderately and weak absorbing property.

Table 1 Sampling information in Arctic area and their analysis

Date	Local time	T	RH	P	WD	WS	TEM	EDS	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			12
	15:20 -15:49	7.0	78	1003.5	120	7.3	130	26	20		
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			10
2012.8.11	16:00 -16:25	4.1	92	1002.0	327	4.6	156	55		6	
2012.8.12	15:25 -15:50	5.7	83	1006.8	132	6.9	100	15	32		
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			10
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		5	
2012.8.15	09:15 -09:45	5.8	73	1019.6	135	3.7	165	60		6	
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			

