

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. CO<sub>2</sub>) 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

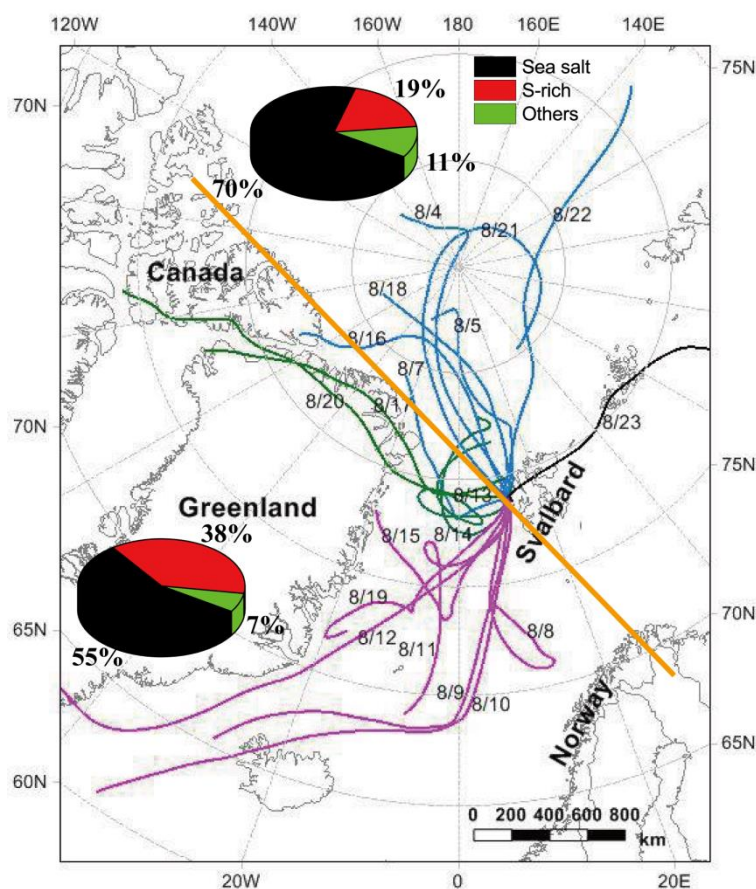
for  $39 \pm 5\%$  and  $29 \pm 7\%$  by number of all the 2002 particles analysed (Figure 3).”

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.