

General Response: We thank the reviewer for your helpful comments. We have addressed all comments and provided point by point response below. The revised manuscript is presented in below.

Response to the referee#2' comments

Overall Comments: The authors use multiple measures to investigate the physical properties of non-sea salt aerosols collected in the Arctic. They qualitatively describe the mixing state of organic, soot, and sulfate aerosols. Although the core data set of this manuscript is informative, as is, the presentation and discussion are confusing and possibly misleading. No large scale descriptive statistics are provided, the methods and presentation are often unclear and/or redundant, the environmental context is lacking, and the optical conclusions are not well explained. This manuscript would benefit from additional analysis, clarification of methods, adding proper context to results, and careful grammar review.

Response: We carefully addressed all of the questions below. We attempted to re-write the manuscript. However, we are not able to address non-specific questions.

Unclear Sample Selection: In multiple sections of this paper the authors discuss a subset of their samples without explaining why they were chosen. This detracts from their discussion and leaves the reader questioning what is not being discussed. Anytime a subset of samples is chosen or used, an explanation needs to be provided explaining why these are the best samples for that specific analysis or investigation. A few example occurrences can be found in lines 275, 281, 118, 190.

Response: Thank you for good comments. We received some comments before and revised the manuscript several rounds. After that, we added Figure S1 showing what samples were analyzed by different instruments. We removed some number which confused for the readers and rephrased some sentences in the Methods section.

To SEM and NanoSIMS analysis after TEM observations, we need to select better samples. These samples should keep good conditions of carbon film because some samples have broken carbon film during the sampling period. In the NanoSIMS and SEM analysis, these samples need to be stucked on the sample stage, they could not be recycled again.

Please see response to reviewer 1, comments 2-4.

Environmental Context: The only environmental context explored in this text is the back trajectories provided for 4 of the sampling days. Whenever field data is discussed, external variables such as temperature, humidity, and back trajectories can help explain variability observed in the data. To improve this work, the context of the sampling should be explored as a driving factor between differences in particle characteristics.

Response: We added the RH, T, P in the table S1. These meteorological data were recorded during the sampling period. Here we did back trajectories of each sampling day as shown in Figure 1.

We added the following to the revised manuscript (line 144 to 146)

“The sample information such as local sampling date and time and meteorological conditions

(e.g., temperature (T), relative humidity (RH), pressure (P), wind direction (WD), wind speed (WS)) were listed in Table S1.”

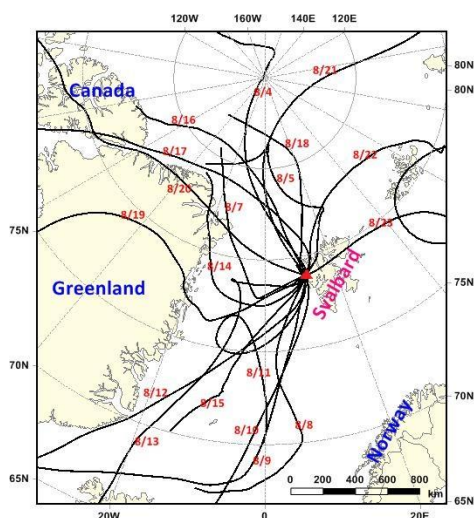


Figure 1 72 h back trajectories of air masses at 500m over Arctic Yellow River Station in Svalbard during 3–26 August 2012, and arriving time was set according to the sampling time

2.1 I am also not convinced that the daily averaged back trajectory calculations are meaningful for these samples. (I’m also not sure the trajectories were daily averaged as the methods section just said they were calculated for a given day, not the frequency of times or given time on the day.) Because the sample length was between 20 minutes and 2 hours, a back trajectory occurring during the actual sampling time for all samples would be more appropriate.

Response: We did choose the mid of the sampling time as the end point for the back trajectory analysis (see Figure 1)

3 Optical Property Calculations: The authors attempt to estimate the potential radiative forcing implications of their results using a core-shell model and Mie theory. Although this is a potentially meaningful result, the methodology needs to be discussed further and improved.

Response: We added more description in the Method section.

L263-271“BHCOAT Mie code by Bohren and Huffman (1983) was used to calculate the optical properties, including scattering cross section (SCS), absorption cross section (ACS), and single scattering albedo (SSA), assuming a core-shell structure. We firstly calculated these parameters assuming a sulfate core and OM shell structure only (ignoring some of the particles that contain soot core). Because the Mie code only can calculate the core-shell structure or homogeneous models, we assume sulfate as a core and OM as a shell in individual particle to build the core-shell model. Based on the core-shell standard mode (Li et al., 2016), we can calculate optical properties of individual internally mixed particles.”

3.1 When performing Mie theory calculations the input size distribution characteristics and wavelength dependence (angstrom exponent) have dramatic effects on the results. These data

need to be reported. The authors state that a single size distribution is used. How was this size distribution established? Could you run your analysis over all of the size distributions you observed to estimate the variability?

Response: Size distributions of particles are measured by microscopes These measurements can help us to evaluate sulfate and OM volume and then we made the Figure 8d. Once we obtained these basic data, we can input all the data to calculate optical properties of individual particles. Here the result is different from the bulk online optical properties. As the comments, we did correction between optical absorption cross section (ACS) and particle diameter assuming strongly absorbing BrC and Moderate absorbing BrC as the particle OM coating. Interestingly, we found that ACS increase along with the particle diameter increase through one nonlinearity showed as below.

In this study, it is not necessary to consider wavelength dependence because lots of work how different size of aerosol particles interacted with the different wavelength. We just did pilot cases to confirm how BrC influence the optical absorption of sulfate particles based on our TEM data (Figure 8). The purpose of this study draws attention for BrC in the Arctic air.

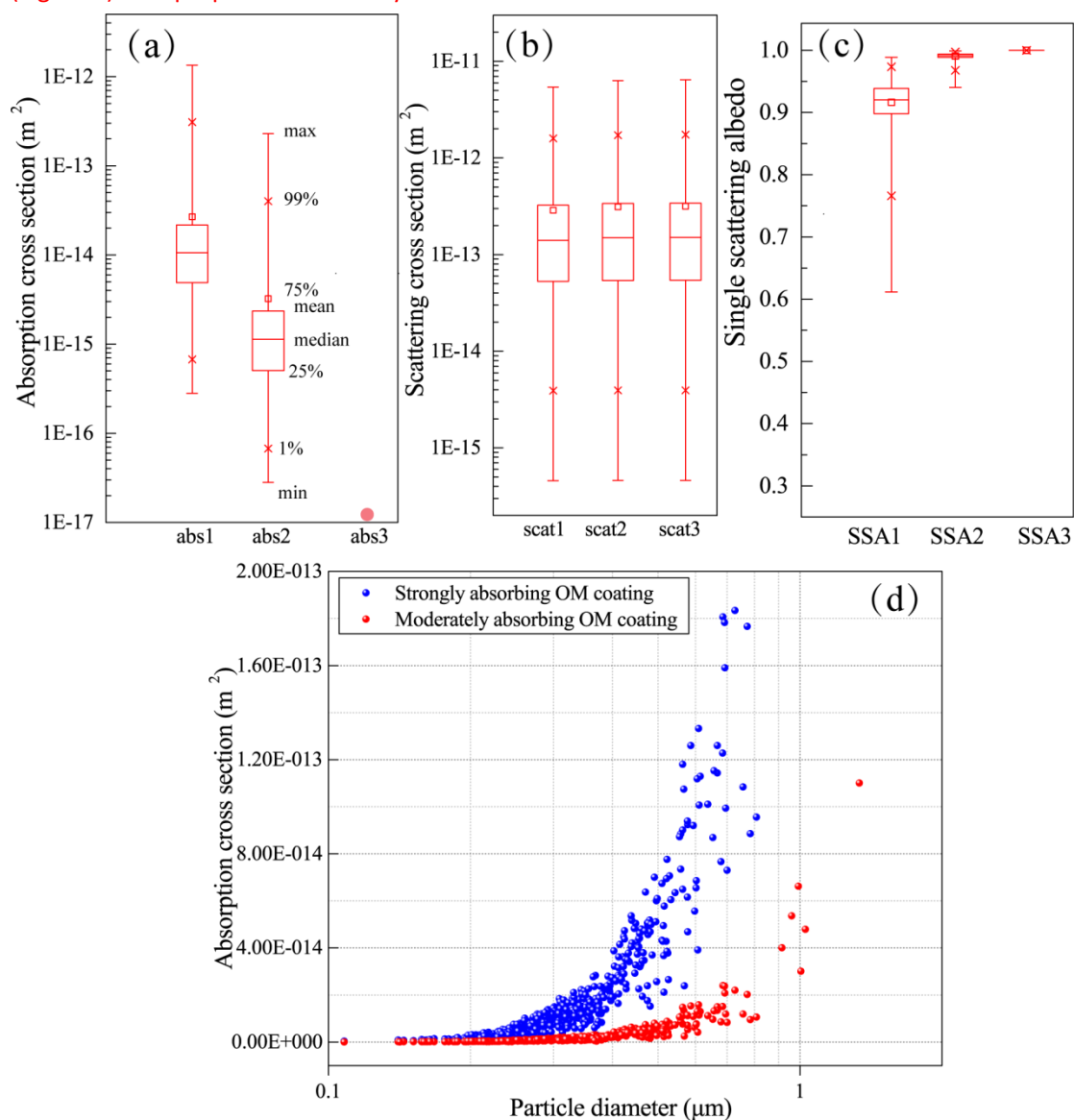


Figure 8 Optical properties of Box-and-whisker plots showing optical parameters of all

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.

3.2 The way the authors include soot in their calculations is not appropriate, as soot is treated as a core with a mixed sulfate-OM shell. This is in direct conflict with their observations that soot is only observed to be associated with the OM shell and sulfate is always the core. It would be better not to include a soot calculation than include a misleading one.

Response: We deleted the soot part here. The updated data is shown in Figure 8

3.3 The relevancy of the refractive index (RI) choice needs to be discussed further. The authors use an RI from biomass burning brown carbon as their slightly absorbing case but frequently state that they believe the OM to be secondary. If that is the case, their estimations are likely too high for all cases but the non-absorbing case. The authors should include refractive indexes from secondary organic aerosol brown carbon to get a more realistic answer. These are generally much lower than the RI values that they used. Additionally, the scattering component chosen (1.65) is relatively high and not explained. Please explain why this value was chosen as it is especially important in core-shell cases.

Response: There are many different RIs from the laboratory experiments. For example, Jiang et al., (2019) suggested the measured RIs on different organic species. The RI is dependence on the wavelength. Fortunately, we just want to test how OM coating influence the sulfate particles. Based on the relationship between RIs and wavelength (Jiang et al., 2019; Feng et al., 2013), the RI doesn't influence our conclusions in this study.

Reference added: Jiang, H., A. L. Frie, A. Lavi, J. Y. Chen, H. Zhang, R. Bahreini, and Y.-H. Lin (2019), Brown Carbon Formation from Nighttime Chemistry of Unsaturated Heterocyclic Volatile Organic Compounds, Environmental Science & Technology Letters, DOI: 10.1021/acs.estlett.1029b00017.

Here we chose the RI reported in Feng et al., 2013. Through the comparisons between strongly absorbing and moderately absorbing, non-absorbing BrC, we generally knew how BrC coating could influence optical properties of sulfate particles. Although our current study could not get a more realistic answer, but the result suggests that BrC has a potentially role to play in light absorption properties of aerosols Arctic. The conclusion warrants further study in the Arctic area. Our results from the TEM can provide solid evidence about the mixing state of sulfate and other aerosol species. As suggested by the referee, we added a sentence to explain why we selected the 550 nm:

In the context:

"Although the refractive index has dependence on the wavelength between 350-870 nm, we tried to select the 550 nm as a case to test how OM coating influence sulfate particles in Arctic air."

3.4 Stemming from the above comment, the relevancy of each refractive index case needs to be discussed in the context of the Arctic. Which case seems the most likely?

Response: We can't make this judgement because there is no data to show this. We showed that this is potentially important to consider, which warrants further study.

3.5 The authors use $m=1.55$ for sulfate. RI values have wavelength dependence, was this included in any way?

Response: Thanks. It does. We corrected the writing here.

Line 256 to 257: "The refractive index used for the non-light-absorbing sulfate component was set to $m=1.55$ at 550 nm (Seinfeld and Pandis, 2006)."

3.6 The authors average the absorption cross section on a per particle basis. This isn't meaningful since observed absorption cross sections will depend on the whole aerosol population (and the size of the particle). The authors should calculate an absorption cross section for the ambient aerosol concentration during their sampling and compare it to other absorption observations.

Response: Our methods are not able to give the ambient aerosol concentration. We tested how the absorbing OM influenced optical properties of sulfate in the Arctic based our individual particle measurements. These results showed that this is potentially important to consider, which warrants further study.

4 Redundancy and Clarity: The writing of this paper needs improvement. There are multiple times when the authors restate the same point twice or fail to introduce a topic before discussing their results. An example of redundancy can be observed when Copper TEM grids are introduced in lines 120, 140, and 156. An example of an improper discussion occurs in the discussion of satellite particles, which are introduced in line 280 with no context or explanation. This forces the reader to infer what the authors mean by satellite particles, possibly leading them to wrong conclusions. To improve the manuscript, I recommend careful reconsideration of the presentation of the data, with special consideration to avoiding redundancy and ensuring the appropriate context is present.

Response: We carefully revised them. Please see our replies as below associated with your specific comments.

4.1 There are multiple points in the methods, a few of which I've included in the specific comments, that are unclear or confusing. The reader needs to be able to understand exactly which analysis was performed on which filters for how many particles if they are to believe your result.

Response: We made one major revision in this section. We added the Table S1 to make specific information.

5 Sample Information: The only summary of the total data set is provided in figure S6 and this figure states that only 3 samples (of 46 collected and 21 analyzed). No explanation is given as to why is summary is so limited given the authors have EDS data (which they used for classification) of 20-30 particles over 21 samples (at least 400 data points). All the of following discussions only make sense if they are provided in the context of overall sample composition.

Response: The Referee is probably referring to Figure S4 rather than Figure S6. It is a spelling

mistake, which we have corrected. At the revised ms, the Figure was moved to the main ms as Figure 1.

Specific Comments:

1 There are many grammatical errors and redundancies that I have not addressed below.

Response: We carefully improved English writing in the manuscript.

2 The phrase internal mixing is used throughout the manuscript without an explicit definition. An explanation of what exactly you mean when you say something is “internally mixed” would improve the manuscript

Response: We added the following to the text

Add an explanation the first time you write it, e.g., line 88

“Internal mixing means that a single particle simultaneously contains two or more types of aerosol components (Li et al., 2016).”

3 Line 55: Change “Arctic” to “the Arctic”

Response: Corrected

4 Line 60: Change “nature” to “natural”

Response: Corrected

5 Line 64-66: Restructure this sentence for clarity. Treat the percentages in a consistent way as to not confuse the reader. For example, this sentence could be changed to:” For example, Winger et al.(2017) showed most Arctic BC is sourced from domestic activities (35%) and transportation (38%), with only minor contributions from gas flaring (6%), power plants (9%), and open fires (12%)”

Response: Corrected

6 Line 72: I’m not sure exactly how this sentence fits in with the brown carbon theme of this paragraph. Are these compounds commonly found in brown carbon or organic aerosols in general? Please add some context.

Response: Thank you very much. We move the positions of this sentence and add more description here.

L73-78: “Accumulation of secondary organic aerosol, a significant fraction of the new particles grow to sizes that are active in cloud droplet formation in the Arctic (Abbatt et al., 2019). More than 100 organic species were detected in the Arctic aerosols and polyacids were found to be the most abundant compound class, followed by phthalates, aromatic acids, fatty acids, fatty alcohols, sugars/sugar alcohols, and n-alkanes (Fu et al., 2008).”

7 Line 84: replace “were” with “have been”

Response: Corrected

8 Line 96-68: This sentence is confusing. Please rewrite it more concisely and clearly.

Response: Revised as the following (line 106 to 109)

“The poor understanding on mixing state of BC and BrC in individual particles will prevent the further simulation of atmospheric climate and aerosol-cloud interaction in the Arctic through the

current atmospheric models (Browse et al., 2013; Samset et al., 2014; Zanatta et al., 2018)."

9 Line 99-100: Change "collected on 7 to 23 August, 2012 in the Arctic." to "collected in the Arctic between August 7th and 23rd, 2012."

Response: Corrected.

10 Line 104: replace "on substrate" with "on a substrate"

Response: deleted the word here

11 Line 118: Change "samples between 7 and 23 August, 2012." to "samples collected between August 7th and 23rd, 2012."

Response: Corrected

12 Line 119: Replace "analyzed for TEM analysis" with "analyzed with TEM"

Response: Revised

13 114-131: Restructure your sampling section. As written the reader may think that you sampled with 2 separate samplers an individual particle sampler and cascade impactor. After reading the paper, there is only one sampler. This confusion can be remedied by introducing the cascade impactor earlier.

Response: Revised the part as below (line 133 to 136)

"A sampler containing a single-stage impactor with a 0.5-mm-diameter jet nozzle (Genstar Electronic Technology, China) was used to collect individual particles by the air flow rate at 1.5 l min⁻¹. Aerosol particles were collected onto copper TEM grids coated with carbon film."

14 122-123: Add the top size cutoff for this sampler.

Response: Added

15 138: replace "within a" with "for a"

Response: Corrected

16 148: 2002 particles examined over all the samples, or in a specific filter?

Response: Added the Table S1 in the sentence. Table S1 can show how many particles we analyzed in each sample.

17 151: Clarify this sentence. Do you mean to say you "we only checked elemental compositions of 20-30 particles" in each sample?

Response: We added more detailed description (line 165 to 180)

"In the clean Arctic air, there are simply particle types including sea salt, sulfate, soot, and OM. Because soot particles have chain-like aggregation, it is not necessary to check their elemental composition. Sea salt particles display spherical or square shapes and are stable under the electron beam in TEM but sulfate particles are spherical but flats on the substrate and produce unstable bubble under the electron beam (Buseck and Posfai, 1999; Chi et al., 2015). TEM observations also can clearly identify sulfate particles or sulfate with OM coating. Therefore, we

can easily identify Arctic particle types based on their morphology. Because of the time-consuming in the experiment, it is not necessary to frequently check elemental composition of the same particle type. For the data statistic in this study, we randomly checked elemental composition of 20-30 particles in each sample (Table S1). EDS spectra of 575 particles were manually selected and saved in the computer for elemental composition analysis. Particles examined by TEM were dry at the time of observation in the vacuum of the electron microscope. In our study, the effects of water and other semi-volatile organics were not considered as they evaporate in the vacuum.”

18 155-156: The statement about Cu is redundant. This has already been stated in line 120 and 140.

Response: Corrected:

19 156-157: What is the difference between what is stated here and what is stated in lines 147-149?

Response: In lines 147-149 describe how to analyze particles on the substrate using TEM/EDS. In 156-157, we used scanning TEM. The method is one other function in the TEM which can give element profile along with the one line or elemental mapping in the targeted individual particle. We added more description here and tell the reader why we did the STEM here.

The following paragraph was revised to (line 181 to 188)

“ Elemental mapping and line profile of individual aerosol particles were obtained from the EDX scanning operation mode of TEM (STEM). The STEM information can clearly display elemental distribution in the targeted individual particles which cannot be provided by the above EDS examination. Based on preliminary individual analysis, we further chose the typical samples containing abundant sulfate with OM coating for the STEM analysis. The high-resolution details of elemental distribution in individual particles can further prove the details of the mixing structure of sulfate and OM in individual particles.”

20 162-163: Replace “is the image analysis platform...” with “is an image analysis platform”.

Response: Corrected

21 177: Replace “Organic Matters” with “Organic Matter”

Response: Corrected

22 Line 184: Replace “TEM grids was” with either “The TEM grid was” or “TEM grids were”

Response: Corrected

23 Line 192-194: Incomplete sentence

Response: Corrected

“AFM with a digital nanoscope IIIa instrument operating in the tapping mode was used to observe surface morphology of individual aerosol particles and measure particle thickness.”

24 Line 194: I stopped making basic grammar and structure critiques at this point.

Response: Thank you very much. We carefully checked them and improved the English writing. Please see the red markers in the revised ms.

25 Line 222: Does treating this as a core-shell system with BC in the middle and sulfate and OM mixed on the outside have any basis? You've indicated that you have soot inclusions on the outside of predominantly sulfate particles, so why would soot be on the inside?

Response: We've deleted the part with soot – see response in comment above.

26 Line 224: This sentence reads as if you've calculated the refractive index of the particles. Did you measure the optical properties of these particles?

Response: We didn't measure their optical properties. We deleted this sentence and revised the part.

27 Line 228: "In this study". It is unclear whether this is referring to your previous work or this manuscript.

Response: corrected

28 Line 253: Is it possible that coagulation of primary organic particles and S-rich particles could have led to the formation of organic coatings? Are you sure assumption that all organic coatings are secondary valid?

Response: The possibility could happen in the polluted air due to high particle number. As our previous study, Chi et al., (2017) determined sea salt particles collected in the Arctic air. We didn't observe the association of sulfate and sea salts particles, although we found many aged sea salt particles. The observations again suggest the particle coagulation is unlikely to be important in the Arctic air.

29 Line 272: Can you say a percentage of NSS particles that are S-rich with an OM coating? Or a percentage of S-rich particles that have a coating? This would strengthen the paper if an actual number was given.

Response: No, we cannot say NSS particles are S-rich with OM coating particles. Figure S4 shows 39% by number of all the analyzed particles were NSS-particles but 29% particles contain sulfate. Here we found 73% of the analyzed NSS-particles are S-rich with OM coating. We've changed this

line 310 to 312: "Here we focused on S-rich, soot, and OM particles as the major non-sea salt particle (NSS-particle, 39±5%) in the analyzed samples, which are approximately 29±7% of 2002 particles (Figure 3). "

L323-324: "A majority of 781 analyzed NSS-particles (74% by particle number) have a sulfate core and OM coating (Figures 4 and 5)."

30 Line 274-276: Are these specific samples special or is there something that you think may have caused the low frequency of soot inclusions? If so, please explain why.

Response: We only found some fresh soot particles in three samples out of the 21 samples. We

found many sulfate with soot inclusions. Previous works showed that BC is very low in the Arctic. We've changed this (line 326 to 333).

"The mixing structure is different from our previous findings in polluted air that soot is normally mixed with sulfate instead of OM coating (Li et al., 2016). Moreover, we noticed that a few chain-like soot aggregates (1.3% in all analyzed particles) (Figure S5) only occurred in three samples during the sampling period (Table S1). Considering the remoteness of the sampling site, such fresh soot particles are likely to be of local origin, including shipping and flaring (Gilgen et al., 2018; Peters et al., 2011). Indeed, we found a few of ships moving in Arctic Ocean during these days from the Ny-Ålesund town."

31 Line 277: This statement needs to be better supported. Just because a site is remote does not mean particles are local in origin. If this is supported by your trajectory calculations, mention them here. Also, don't the soot inclusions also imply that perhaps the OM is not secondary in nature? Soot is 100% primary and often co-emitted with primary OM, so if there is soot associated with OM coatings the soot itself is primary and so possibly some of the OM is primary as well.

Response: We found fresh soot particles in Figure 6. Indeed, TEM image shows that the soot particles have very thin OM coating.

It is true that soot is often co-emitted with primary OM. However, there are plenty of evidence to show that the primary soot aggregate particles are not heavily mixed with OM and in particularly not coated with thick OM (Wang et al., 2017). Furthermore, It is well known that SOA is dominant in the remote air (Jimenez et al., 2009).

32 Line 280: You need to define what satellite particles are; you have not discussed or defined them previously. Are they simply splatter of liquid portions of the particle when the particle is collected?

Response: Thanks. We define it.

L334-337 "TEM observations showed that some sulfate particles had unique morphology that a sulfate particle was surrounded by some smaller particles (Figure 5a). They are often called "satellite" particles as they were distributed from the central particles when impacted on the substrate during sample collection."

33 Line 280-281: is there a reason why satellite particles would have been observed on these days but not other days?

Response: The samples collected during 9-15, August contained abundant sulfate particles, OM, and soot particles (Table S1). Also, Figure 2 shows that air masses from North American during these days were mainly dominant. The reasons should depend on their air masses. Although the mechanism of this point is interesting, we could not give specific evidence under the current data.

34 Line 281: This is misleading and implies you performed the NanoSIMS analysis on 11 samples. In the methods section, it says only two samples were analyzed with NanoSIMS.

Response: Corrected

L339-342: "NanoSIMS analysis further provided more information that the satellite particles

selected from the samples (Table S1) have strong 32S- (Figure 6a, c) and 16O- signals (Figure 6d) as well as weak 12C14N- signals (Figure 6a, b).”

35 Line 287-289: This is misleading. It reads as if you have done molecular characterization of the organic matter.

Response: Corrected

L346-347 “Indeed, Fu et al. (2008) found that polyacids are the most abundant organic compounds, followed by phthalates, aromatic acids, and fatty acids in Arctic aerosol particles.”

36 Line 314: You back trajectories are only for specific days, be transparent about this in the Discussion

Response: Corrected. Add one Figure S2

37 Line 317-318: This conflicts with your earlier comment that most BC should be local.

Response: The result does not conflict with earlier comments. We only pointed out the fresh soot particles (only 1.3%) may come from local shipping or combustion activities. It doesn't mean most BC were from local sources. In this study, we did not determine where the BC comes from.

38 Line 326-342: This discussion reads like a list of facts, but why they are all relevant is not always stated. Explain why each observation is important and how it adds context to your results.

Response: We carefully revised the part as below

L338-408: “The sulfate core-OM shell structure observed in the Arctic summer atmosphere is similar to those in the background or rural air in other places (Li et al., 2016; Moffet et al., 2013). Based on the images from electron microscopies, we can infer that OM coating thickness in the arctic atmosphere was comparable with them in rural places but higher than them in urban places. During the transports, organic coatings on sulfates were considered as the secondary organic aerosols and their masses increase following particle aging and growth (Li et al., 2016; Moffet et al., 2013; Sierau et al., 2014). Figures 1 and 2 show that most of particles in the air masses transported long distance from North American. The result indicates that these long-range transportation of secondary sulfate particles have enough time to experience the possible atmospheric heterogeneous reactions on particle surfaces or cloud processes in the Arctic air. Similarly, Moffet et al. (2013) found that soot inclusions occurred in OM coating when OM coating on sulfates built up through photochemical activity and pollution buildup the Sacramento urban plume aged. On the other hand, the sulfate/OM particles with soot inclusions are probably formed in a similar way as those found elsewhere (Li et al., 2016) – e.g., soot particles may have acted as nuclei for secondary sulfate or organic uptake during their transports (Riemer et al., 2009). Similarly, besides the OM coating in the Arctic particles, Leck and Svensson (2015) found some biogenic aerosols like gel-aggregate containing bacterium in ultrafine particles. However, we didn't find any gel-like particles in the samples because our sampler had very low efficiency for ultrafine particles.”

39 Line 343: Why is dry included here? Are there also wet particles that you have not discussed?

Response: Deleted the “Dry” here

40 Line 344-347: Explain LLPS in simpler terms and why it's important.

Response: Revised the part.

L409-428: “TEM images show that most of the internally mixed sulfate particles display sulfate core and OM coating on the substrate (Figures 4a and 5b, c). The sulfate and OM separation in individual particles were defined by You et al. (2012) as liquid-liquid phase separation (LLPS). Concerning the knowledges of the LLPS can better understand particle hygroscopicity, heterogeneous reactions between reactive gases on particle surface, and organic aging (You et al., 2012). They also reported that the LLPS can reflect the O:C ratio in the OM, which is roughly ≤ 0.5 . In this study, we did observe the LLPS in almost all the fine sulfate particles, which indicates that the secondary OM in the coating might be not highly aged. Therefore, we speculate that the thick OM coatings were consistently built up during the long range transport of sulfate particles and part of secondary OM in the coating likely formed in Arctic area. Indeed, some studies reported that there are various sources of organic precursors during the Arctic area, such as biogenic VOCs from ice melting and open water (Dall’Osto et al., 2017) and anthropogenic VOCs from shipping emissions in summertime (Gilgen et al., 2018). The dependence of OM volume on particle size (Figure S6) suggests that the suspended sulfate particles are initially important surface for secondary OM formation. Moreover, the common OM coating on sulfate particles indicates that secondary OM as the surfaces of fine particles might govern the possible heterogeneous reactions between reactive gases and sulfate particles in the Arctic air.”

41 Line 346: It’s unclear what 90% is referring to in this statement.

Response: We deleted this sentence here

42 Line 348: There’s likely a complex relationship between phase state, oxidation state, and humidity. This needs to be investigated and explained further if statements about aerosol age are going to be made. Additionally, shouldn’t you see a variety of ages of aerosol in your samples? Showing contrast between aged and unaged particles would be interesting and convincing.

Response: We re-wrote this part. We pointed out that the implications including LLSP in the Arctic air. Almost all particles we observed the aged. We do not have a parameter to define the age of the aerosol population and we are unable to answer the question clearly.

43 Line 376-378: This is circular reasoning because $^{12}\text{C}^{14}\text{N}^-$ was what you used to identify OM so of course it was observed in the OM coatings. I don’t think this data set is appropriate to make conclusions about the N content of OM coatings. That said, if you were able to calculate the mass concentration of N in the coatings with NanoSIMS that might give you a better indication of the BrC potential of the OM.

Response: It is impossible to calculate the N concentration in the coatings based on the NanoSIMS data here. The literature as the below proved that CN^- can indicate N-containing organic matter.

Herrmann, A. M., K. Ritz, N. Nunan, P. L. Clode, J. Pett-Ridge, M. R. Kilburn, D. V. Murphy, A. G.

O'Donnell, and E. A. Stockdale (2007), Nano-scale secondary ion mass spectrometry — A new analytical tool in biogeochemistry and soil ecology: A review article, *Soil Biology and Biochemistry*, 39(8), 1835-1850.

44 Line 390-393: The average absorption cross-section is reported on a particle basis. This would be much more meaningful if it was extrapolated to environmental conditions. Because you've sampled from the atmosphere, you should be able to approximate particle concentrations, correct? If so, you could back calculate this to an actual atmospheric absorption contribution and compare it to expected absorption from other species and measurements. This would be significantly more meaningful.

Response: Yes, we agree it would be useful to extrapolate to environmental conditions but as stated by the reviewers, we do not want to over-interpret our results. We cannot extrapolate because we do not have ambient particle concentration data. One single paper will not be able to answer all questions and we do suggest for future work to answer these questions.

45 Line 413, 260: 29% number should include a standard deviation.

Response: added

46 Line 410: The last section shouldn't simply repeat what was stated in the above sections, but instead present the data in additional context and discuss the implications.

Response: We made major revision in the section. Please see the revised red words.

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., Galí, 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., Lévassieur, 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.
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- 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.
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