Response to Referee #2

1. **General comments:**

In this study, Ye and coworkers report on the abundance of organosulfates in aerosol particles collected on filters in the Arctic, the Antarctic and at two sites in southern China (Guangzhou, rural and urban). The filter samples were extracted, and afterwards analyzed by high-resolution mass spectrometry with direct infusion electrospray ionization in negative mode. From the obtained signals, sum formulas for organic compounds were generated and assigned to certain compound classes (i.e., organosulfates (OSs), nitrooxy-organosulfates (NOSs), organonitrates (ONs), and oxygenated hydrocarbons (OxyCs)). The authors find that aerosol components from the Arctic and Antarctic exhibited rather high carbon oxidation states and a large percentage of high molecular weight (HMW) compounds, which they assign to long-range transport of such compounds from stratospheric reservoirs. In comparison to the aerosol samples from China, the authors observe larger fractions of sulfur-containing compounds and lower fractions of ONs, which is assigned to low NOx levels in the polar atmosphere. Nonetheless, the obtained number fractions of polycyclic aromatic OSs and NOSs and their oxidation states were similar to urban Guangzhou samples, indicating larger anthropogenic influences at the Arctic and Antarctic sampling sites.

Given the quite exotic sampling sites and the potential significance of such a study, I am quite disappointed from this manuscript. The way the samples were taken, processed and analyzed is not appropriate to support any of the hypotheses drawn. This is also why major parts actually remain extremely speculative, inconclusive, or sometimes even contradictory. In my view, a simple, but targeted, LC-MS analysis of the filter samples would have yielded much more (reliable) information on the abundance of OSs and NOSs in aerosol samples from the Arctic and Antarctic. Therefore, I can only recommend either to reject this manuscript or to reconsider it after substantial revisions (as detailed in my comments below).

**R:** Thanks very much for your comments and summary. We are deeply sorry for the poor manuscript of the previous version. The present manuscript is improved with major revisions. We reply to your specific comments one item by one item as follows:

1. **Major comments:**
2. My most pressing concern is the analysis procedure of the filter extracts. Electrospray ionization is known to be prone to matrix effects such as ion suppression and adduct formation. Without any pre-separation of inorganic ions, it is not possible to distinguish organic and inorganic sulfates. Thus, the assigned OSs and NOSs could just represent sulfate adducts of organic molecules. Moreover, I highly recommend including isotopic signals of sulfur in the formula assignment procedure for OSs and NOS. Besides, without any separation technique (e.g., liquid chromatography) and at least some surrogate standards, it is not possible to discuss or compare the abundance of compounds in the aerosol samples.

**R:** Thanks very much for your comments. We are sorry our mistakes in the expression. Actually, solid-phase extraction (SPE) was used in the pre-processing of samples for desalting before direct injection into Orbitrap MS, following the methods in (Lin et al., 2012a; Lin et al., 2012b). It reads “The extraction and filtration steps were performed twice and the extract was acidified to pH=2 using hydrochloric acid and then loaded on a solid-phase extraction (SPE) cartridge (Sigma-Aldrich, USA) followed by eluting using 3 mL mixture of methanol containing 2% aqueous ammonia (*w:w*) (Lin et al., 2012a; Lin et al., 2012b). The elute was then evaporated to dryness under a gentle N2 steam and immediately re-dissolved in 400 μL of 1:1 (*v:v)* mixture of MQ water and acetonitrile. To obtain an overall picture of the molecular characteristics of OSs and NOSs in the polar atmosphere, where the relevant data is still rare so far, liquid chromatography (LC) separation was not used in this study.” in line 135-141 in subsection 2.2.1. Similar experiments and analysis were presented in previous studies (e.g. Nizkorodov et al., 2011; Lin et al., 2012a; Lin et al., 2012b; Kuang et al., 2016; Bin Jiang, 2016; Wang et al., 2016; Wang et al., 2017).

The formula assignment procedure has been revised by adding isotopic signal of sulfur (34S) as stated “And 0‒1 of 34S atom was allowed in the calculation of formulas.” in line 163-164 and “For intense ions, the presence of 34S isotope peaks supports the assignment of sulfur-containing formulas (Table S1). The isotope formulas were not included in the following discussion because they shared the same elemental compositions with their corresponding 32S isotope ion peaks.” in line 170-172 in subsection 2.4.

To express more appropriately, all the inappropriate statement like “abundant” have been revised. Instead, the number of tentatively assigned formulas were compared.

1. Merely from reading the abstract, it is obvious that there are some contradicting conclusions drawn in this study. On the one hand, the authors hypothesize that aerosol components are long-range transported to the polar sites. On the other hand, they conclude that oxidation of aerosol precursors under low NOx levels at the polar sites reduce the amount of ONs. And lastly, they state that local anthropogenic emissions are the source of polycyclic aromatics at these sites. The reasoning for each of these hypotheses is very weak in the corresponding sections.

**R:** Thanks for your comments. The structure and main discussion is re-built in the present manuscript. Accordingly, our abstract has been revised as “Organosulfates (OSs) are important secondary organic aerosols (SOAs) that have been newly-identified in recent years. Due to their amphipathy and light absorptions, OSs have potential impact on the climate. Moreover, OSs can serve as molecular tracers for precursors and multiple processes leading to SOA. Therefore OSs have been investigated in many areas in the global. However, studies on OSs are rarely reported in the polar region so far, which limits our understanding of their formation pathways and influences in polar environment. Here we present the first overall observation of OSs and other organic compounds in the polar regions covering the Arctic and Antarctic. Organic compounds in aerosol samples from summer polar area during 2014/2015 Chinese National Arctic/Antarctic Research Expedition (CHINARE) were analysed by ultrahigh resolution mass spectrometry coupled with negative ion mode electrospray ionization (ESI(-)-UHRMS). OSs, nitrooxy-organosulfates (NOSs), organonitrates (ONs) and oxygenated hydrocarbons (OxyCs) were detected and tentatively determined by their formulas. Our results show that total OSs (OSs + NOSs), ONs and OxyCs accounted for 28–32%, 28–40% and 32–44% of the total detected formulas in polar areas, respectively. Large differences of the molecular characteristics were observed when aerosol samples between polar area and Guangzhou sites were compared. Total OSs featured by larger percentage of high molecular weight (HMW) formulas and higher oxidation states with wider range were found in the Arctic and Antarctic aerosol samples, which indicated the polar atmosphere is a mixture of fresh and aged organic aerosols. To better understanding the sources of total OSs in the polar regions, the relationships between total OSs and marine phytoplankton, long-range transport and local human activities were investigated. It was found that the total OSs at the Antarctic sampling sites are possibly influenced by nearby scientific stations and relevant research activities while the Arctic sampling sites were possibly influenced by long-range transport from the nearby continents. For different precursor-derived OSs, the anthropogenic precursor-derived (AP-D) OSs were detected in all the four polar sampling sites, suggesting human activities can significantly influence remote polar environment. For biogenic precursor-derived (BP-D) OSs in the polar regions, the coincidence that more BP-D OSs were tentatively identified in more acidic aerosols supports that acid-catalysed epoxy chemistry may be important for the formation of BP-D OSs in acidic atmospheric conditions. However, the precursors of a large number of OSs cannot be identified so far, which comprised more than 70% of total assigned OSs at each site. These un-identified OSs emphasized additional laboratory studies should be carried out in order to further investigate the formation mechanism of OSs and their potential contributions to OSs in the polar atmosphere.”

1. The manuscript is often difficult to read and to understand because of bad language quality. It is possible that some of my specific comments (see below) are actually related to incorrect wording. To improve readability, I would recommend the authors to look for some help either from a native speaker or from a language editing service.

**R:** Thanks a lot for your careful review. We are sorry for our poor expressions in the former manuscript. We have improved our manuscript according to your suggestions and comments.

1. **Specific comments:**
2. P3L26: Do you have any data on particle size distributions during the sampling period? This would be very interesting to get an impression of the ratio between primary and secondary particles.

**R:** Thank you for your comment. We are sorry that we do not have size fraction data during the cruise, so we are unable to discuss the ratio between primary and secondary particles in the present study. Since the information about particle size distributions is quite important, we plan to sample aerosols by size in the future cruise as you suggested.

1. P3L32: Can the authors estimate the impact from ship exhaust? In particular, regarding the source of polycyclic aromatics in the polar samples, it would be necessary to exclude any influences from the exhaust.

**R:** Thanks for your comment. We used two criteria to minimize the possible impact from ship emissions during the cruise in the Arctic: 1) “Since the air samplers were installed at the top front deck while the exhaust pipe was located in the rear of *Xuelong*, at a distance of more than 100 m from the sampler, aerosol sample is usually unaffected by the ship emission when the icebreaker navigate at normal speed (> 5 m/s)*.* However, when the icebreaker stops or navigates slowly, the collected aerosol sample may be contaminated by ship emissions.” in line 94-97. 2) “It should be noted that a constraint, that’s CO≤150 ppb, was applied in choosing aerosol samples from the Arctic sites to eliminate the possible contamination from the icebreaker itself (Stehr et al., 2002; Lohmann et al., 2004; Großmann et al., 2013; Sommariva and von Glasow, 2012; Hu et al., 2016). The average concentrations of CO in the central Arctic Ocean site and Arctic pack ice site were 93.74 ppb and 107.66 ppb, with the maximum concentrations being 118.49 ppb and 124.28 ppb, respectively (Fig. S1). In addition, no visible black carbon was found on our selected TSP filters. Therefore, the potential contamination from *Xuelong* emissions is expected to be minor for the selected samples.” In line 103-108. However, as we stated in the manuscript “To be not speculative, we cannot completely rule out the possible contribution from ship emissions to OSs for the Arctic samples.” in line 109-110.

1. P4L11: Why did the authors choose these filter samples? No information is given on the sampling dates or the selection procedure. Are there any additional data available for these samples?

**R:** Thanks for your suggestions. The reasons for choosing the two Arctic sites were: 1) we want to see whether the OSs are related to different types of sea ice cover as the Arctic pack ice site is in Arctic Ocean covered partly with pack ice while the central Arctic Ocean site is covered with dense sea ice; 2) the central Arctic Ocean site (81 °N, 157 °W) is the most northerly latitude reached by the 6th CHINARE in 2014; 3) CO during both of these two sampling period is relatively low. The information on the sampling is given in the manuscript that “TSP samples collected at the Arctic pack ice zone (68.45–75.50 °N, 165.10–169.09 °W) on September 7‒8 and the central Arctic Ocean area (81.00–81.09 °N, 157.02–157.67 °W) on August 19‒21 were used for further analysis. The Arctic pack ice zone is located in the Chukchi Sea, where nutrient is relatively rich, especially in summertime when the sea ice retreats (Connell et al., 2018; Arrigo et al., 2012). This area is also massively influenced by the air mass from nearby continents. The central Arctic Ocean site was a temporary ice station surrounded by dense sea ice. This site is the most northerly latitude reached by the 6th CHINARE in 2014, where the icebreaker *Xuelong* was anchored for 7 days. It should be noted that a constraint, that’s CO≤150 ppb, was applied in choosing aerosol samples from the Arctic sites to eliminate the possible contamination from the icebreaker itself (Stehr et al., 2002; Lohmann et al., 2004; Großmann et al., 2013; Sommariva and von Glasow, 2012; Hu et al., 2016). The average concentrations of CO in the central Arctic Ocean site and Arctic pack ice site were 93.74 ppb and 107.66 ppb, with the maximum concentrations being 118.49 ppb and 124.28 ppb, respectively (Fig. S1). In addition, no visible black carbon was found on our selected TSP filters. Therefore, the potential contamination from *Xuelong* emissions is expected to be minor for the selected samples. However, to be not speculative, we cannot completely rule out the possible contribution from ship emissions to OSs for the Arctic samples (Lohmann et al., 2004).” in line 98-110. For the Antarctic sampling sites, we want to see the characteristics of OSs at coastal Antarctic (Zhongshan Station) and inland Antarctic (Kunlun Station). The information on the sampling is given as “For the Antarctic samples, TSP were collected at Zhongshan Station (70.13°S, 76.58°E) and Kunlun Station (80.42°S, 77.12°E) from January to February 2015. Medium-volume air samplers were installed at these two stations to collect TSP samples through 82 mm QFFs. Each sampling lasted for 12–24 h. The Zhongshan and Kunlun Stations are both Chinese Antarctic scientific research stations. Zhongshan Station is located on the coastal southeast of Prydz Bay, affected by the air mass from both marine and inland ice sheet. The area around Zhongshan Station is strongly influenced by human activities. This station is equipped with dozens of buildings and vehicles, consuming 200 tons of diesel fuel each year for power generation, heating, water production, and equipment operation. An Adélie penguin colony is located ~1 km to the station and the emission of their droppings is a potential source of atmospheric sulfur (Xie et al., 2002). Kunlun Station is about 7.3 km southwest of Dome A (4087 m above sea level), which is the highest research station in Antarctica. The atmospheric condition is relatively stable in Dome A. For such a remote site, a key factor that influences the summer atmosphere is anthropogenic emissions due to research activities. Samples collected during January 12 and 15‒17, 2015 at Kunlun Station and during February 5‒6, 2015 at Zhongshan Staion were used for further analysis.” in line 111-122.

1. P4L14: Did the authors check for compositional changes of the aerosol samples from sonication? Sonication is known to produce OH radicals in aqueous solutions and might therefore induce chemical reactions. Did the authors quantify extraction efficiencies? Why was methanol replaced by acetonitrile after evaporation?

**R:** Thanks for your comments. The preparation method used in this paper followed the method stated in (Lin et al., 2012b). According to this paper, they used ultrapure water for extracting humic-like substances (HULIS), methanol as eluting solvent for SPE and water/ACN for reconstitution. SPE was used in preparation of polar samples in our analysis. The organic compounds would be retained on the catridage and be eluted by methanol containing 2% aqueous ammonia (w/w). So the initial solvent (MQ/water) was not able to affect the re-dissolution. In addition, we added methanol to extraction solvent for the purpose of dissolving more organic components. The reason for not using ACN is the membrane for filtering insoluble materials only allows passing of polar solvent. We are sorry that we did no test the efficiencies.

We are sorry we didn’t consider the extraction efficiencies at first because of our original intension of qualitative measurements is to obtain an overall picture of the molecular characteristics of organosulfates in the polar atmosphere. As for the sonication, it does have the potential to change chemical and physical composition of samples due to producing OH radical in both water and organic solutions (Riesz et al., 1985; Beckett and Hua, 2001; Miljevic et al., 2014). However, sonication has been chosen for extracting organosulfates in many prior studies (e.g. Gao et al., 2006; Surratt et al., 2007; Surratt et al., 2008; Lin et al., 2012a; Lin et al., 2012b; Zhang and Worton, 2012; Ma et al., 2014; Meade et al., 2016; Riva et al., 2016). The preparation method used in our study followed the method stated in Lin et al. (2012b), but we didn’t consider the effect of sonication, which may induce uncertainty in our study. We appreciate this suggestion and will test the extraction efficiency or choose milder extraction methods, such as orbital shaker in the future studies.

1. P4L18: Why did the sample travel for 3 min through a stainless steel capillary before ionization? This could have induced chemical reactions at the steel surface. Did the authors check such potential influences?

**R:** Thanks for your reminding. We made a mistake in our expression. 3 min is the time for analysis inside the mass spectrometer after ESI. The time for traveling through the steel capillary is about a few seconds after injecting by an auto-samlper. And we have corrected this mistake in line 142-143, it reads “Sample aliquots (70 μL) of the extracts were then injected by an auto-sampler and travelled through a steel capillary column for a few seconds.”

1. 4L23: “ppm” is not an absolute value. Moreover, Orbitrap mass spectrometers have much larger mass errors than 0.02 ppm. Typically this should be somewhere between 2 and 5 ppm.

**R:** Thanks for your reminding. Again, we made a mistake in the expression. We have corrected this mistake, it reads “The system was operated under negative ESI mode with a resolving power of 100,000 at m/z 400 and absolute mass error <2 ppm was achieved.” in line 144-145.

1. P5L13: I don’t think that the “4s+3n ≤ o” criterion is sufficient for the identification of OSs and NOSs. The compounds could also contain other functional groups, such as sulfonates, amines, etc. Moreover, as already mentioned above, the authors should at least include isotopic patterns from sulfur isotopes into their assignment procedure, to decrease the level of uncertainty. (The presence of sulfur isotopes is quite easy to detect in the high-resolution mass spectra.)

**R:** Thanks for your comment. We are sorry that we made a mistake in the expression again. The criterion should be “4s+3n ≥ o” and we have corrected it in line 167. This criterion has been widely used in prior studies (e.g., Lin et al., 2012b; O'Brien et al., 2014; Kuang et al., 2016; Wang et al., 2017). The formula assignment procedure has been revised by adding isotopic signal of sulfur (34S) as stated “And 0‒1 of 34S atom was allowed in the calculation of formulas.” In line 163-164 and “For intense ions, the presence of 34S isotope peaks supports the assignment of sulfur-containing formulas (Table S1). The isotope formulas were not included in the following discussion because they shared the same elemental compositions with their corresponding 32S isotope ion peaks.” In line 170-172.

1. P6L2: Equation 3 cannot be applied to all heteroatom-containing compounds, as the oxidation states of sulfur and nitrogen are unknown (see also comment 7).

**R:** Thanks for your important comment. The calculation of Oxidation state of carbon (OSc) has been corrected in line 189-199 in subsection 2.4, it reads“ Oxidation state of carbon (OSc) has been used as a metric for the degree of oxidation of atmospheric organic aerosols (Kroll et al., 2011). OSc can be calculated using Eq. (3):

(3)

Where OSi is the oxidation state of element i, and / is the molar ratio of element i to carbon. For organic formulas expressed as C*c*H*h*O*o*N*n*S*s* in this study, where the oxidation state of H, O, N and S is set as +1, ‒2, +5 and +6, respectively, Eq. (3) can be simplified to Eq. (4)

(4)

In which and are the elemental ratios of oxygen-to-carbon and hydrogen-to-carbon, and and are the elemental ratios of nitrogen-to-carbon and sulfur-to-carbon. Eq. (4) was applied in calculating oxidation states of tentatively assigned formulas (C*c*H*h*O*o*N*n*S*s*) on the premise that all assigned nitrogen-containing and sulfur-containing functional groups are regarded as nitrate and sulfate.”

And the corresponding figures were revised in Figure 4 for total OSs (OSs + NOSs), in Figure S5 for ONs and in Figure S6 for OxyCs.

1. P7L13: What do you mean by “aerosol formula types”?

**R:** Thanks for your comment. We have changed it to “numbers of assigned formulas” in Line 238-239.

1. P7L16: Figure S1 is only showing back trajectories for one day at each site. What about the other days?

**R:** Thank you for your suggestion. We have redrew the back trajectories at the starting time with starting location and the ending time with ending location as in Figure S2.

1. P7L20: It is trivial that the authors found compounds between 90–800 Da because this was the instrumental setting of the mass spectrometer.

**R:** Thanks for your comment. We have remove this expression.

1. P7L27: Do the authors really believe that high molecular weight compounds are preserved in the stratosphere over several days or weeks and then transported back to the ground? I don’t think that such compounds would survive such a transport mechanism. Similarly, why should LMW compounds be more reactive than HMW compounds? From the filter analysis data, none of these statements can be supported.

**R:** We are sorry for our incomprehensive discussion in former manuscript. We calculated the energy range of photons in UV light (λ=200~380 nm, excluding vacuum VU) using. The energy of photons in UV light ranges from 3.3 eV to 6.2 eV. Compared with bond dissociation energies of C‒C bond (6.29 eV) and C‒H bond (3.49 eV), organic molecules can be easily broken in the stratosphere due to the high energy of photons in UV light. So we remove the part “HMW organic molecules can be transported to polar regions through stratosphere” from the manuscript.

1. P8L1: What is the meaning of the criterion (DBE-n) < 4?

**R:** It is a criterion to identify aliphatic compounds (Lin et al., 2012b; Wang et al., 2016). However, it is not used in our data analysis, so we have deleted it.

1. P8L6: 80% of the OSs/NOSs are stated to be aliphatic and 10% to be aromatic. What about the other 10%

**R:** Thanks for your comment. In the original analysis, aliphatic structure is defined by (DBE-n) < 4, which is proposed as the maximum criterion of aliphatics, and aromatic structure is defined by Xc > 2.5, which is proposed as the minimum criterion of aromatics. Thus some compounds are in the range of (DBE-n) > 4 but Xc < 2.5. In the present manuscript, we deleted the criterion (DBE-n) < 4 and revised the corresponding content.

1. It would be very useful to have a list of the observed signals and assigned formulas for the different sampling sites.

**R:** Thanks for your suggestion. All OS and NOS peaks observed in the mass spectra and their corresponding potential precursors at the four polar sampling sites are listed in Table S1 now.

1. P8L17: This is very confusing. Do the authors want to say that the O/C ratio given in the discussed Figures is actually not the O/C ratio?

**R:** Thanks for your comment. We are sorry for our confusing expression of O/C ratio. It is described as “The *h/c* ratio is plotted versus its (*o*-2*n*-3*s*)/*c* ratio for total OSs (Fig. 4), (*o*-2*n*)/c for ONs(Fig. S3) and *o*/*c* ratio for OxyCs (Fig. S4). It should be noted that due to nitrate and sulfate groups containing more oxygen atoms than common oxygen-containing functional groups, (*o*-2*n*-3*s*)/*c* and (*o*-2*n*)/c was used to replace *o*/*c* to calculate the number of oxygen-containing functional groups of per carbon atom, which is also adopted by Lin et al. (2012b) and Wang et al. (2016).” in line 269-273.

1. P8L25: How do the authors know that biogenic emissions dominate at the rural Guangzhou site?

**R:** Thanks for your comment. This conclusion is inappropriate, and we delete it from the present manuscript.

1. P9L4: The findings for ONs and OxyCs are not discussed.

**R:** Thank you for your comment. Since our main topic is OSs, so we pay less attention to ONs and OxyCs. The results from ONs and OxyCs aromaticity has been added in line 232-238 as “However, the fraction of ONs in Guangzhou is larger than that in the polar regions. This may be due to relatively high NOX concentrations and night-time oxidation of NO3 in winter Guangzhou. NOX concentrations in near-surface atmosphere of Guangzhou is up to ~120 μg m-3 due to vehicle exhaust emissions and fossil fuel combustions (Shao et al., 2009), far more than NOx concentrations of ~2 μg m-3 in near-surface in the polar regions (Honrath and Jaffe, 1990; Stroud et al., 2003). In addition, since the Arctic and Antarctic samples were collected during the summertime with polar days, nocturnal oxidation of NO3 was not likely to be important.” And in line 265-267 as “For aromatic ONs and OxyCs, they accounted for 23‒38% and 55‒88% of total tentatively assigned ONs and OxyCs, respectively, at all the six sampling sites (Fig. 3a&b).” For oxidation conditions of ONs and OxyCs, they showed similar features with OSs and NOSs (Fig. S3‒S6).

1. P9L9 – P10L6: In my view this entire approach is invalid. Given that the number of N and S is typically for known OS and NOS, the authors assume that solely from the carbon number they can infer potential precursor. How can they distinguish anthropogenic and biogenic precursors in such an approach (in particular, as many of these precursors have the same number of carbon atoms)? This is all extremely speculative!

**R:** Thank you for your suggestions. We have revised the approach as “Tentatively assigned OSs and NOSs possessing identical carbon, hydrogen, oxygen, nitrogen and sulfur atoms with the “standard” OSs were temporarily considered to have the same precursors with the “standard” OSs in the present study. By comparing their formulas, the OSs and NOSs were classified as anthropogenic precursor-derived (AP-D), biogenic precursor-derived (BP-D), anthropogenic/biogenic precursor-derived (A/BP-D) and unknown precursor-derived (UP-D) OSs (Fig. 6 and Table 2).” in line 294-298. This approach is also adopted by Wang et al. (2016; 2017) for discussing the potential sources. To reminder the reader about the uncertainty, we use words “tentatively assigned OSs” throughout the manuscript. By using the revised method, more than 70% of total tentatively assigned OSs and NOSs were from unrecognized precursors so far at each site.

1. P10L7: How do you distinguish sea spray derived sulfate from “non-sea-salt sulfate” when you only collected TSP samples?

**R:** Thanks for your comment. The sulfate to sodium ration in seawater ((SO42-/Na‑)sea-salt=0.252) was used to calculate the non-sea-salt sulfate (nss-SO42-) concentrations as follows: (Keene et al., 1986).

1. P10L20 – P11L2: The pH values are never “extremely low” and thus do not support acid-catalyzed reactions. As noted above, it is not possible to assign AVOC and BVOC precursors for certain OSs. Moreover, abundances of single ions do not necessarily correlate with their concentrations. Therefore, they should not be compared directly.

**R:** Thanks for your comment. We have changed this part into “At Arc-CO and Ant-KLS, atmospheric aerosols were neutral/slightly alkaline (pH = 7.7 and 7.8), while aerosols from Arc-PI and Ant-ZSS were slightly acidic (pH = 3.8 and 4.8). Correspondingly, no BP-D OSs or NOSs were tentatively identified at neutral Arc-CO and Ant-KLS, while 3 and 1 BP-D total OSs were identified at acidic Arc-PI and Ant-ZSS (Table 2). The coincidence that more BP-D OSs were identified in more acidic aerosols supports that acid-catalysed epoxy chemistry may be important for the formation of BP-D OSs in acidic atmospheric conditions.” in line 367-372.

1. P11L8: RH values of 50–65% are not dry, but still quite humid and mostly above the deliquescence point of aerosol particles. Therefore, it seems not possible to draw any conclusion on RH effects here.

**R:** Thanks for your suggestion. We have changed this part into “Between the Arc-CO and Ant-KLS with similar pH conditions (pH=7.7 vs pH=7.8), more OS constituents were detected at the Ant-KLS where RH was lower. This is inconsistent with previous observations that high RH values increase OS yields at neutral pH (Barbosa, 2017, J. Li, 2013). Considering the emissions of local anthropogenic precursors from research work and domestic pollution may be higher at Ant-KLS than Arc-CO site, the higher RH at Arc-CO may not match with the possible higher precursors at Ant-KLS. Moreover, both the atmosphere are under humid conditions at these two sites, so deliquescence may not happen for most aerosols. Therefore, OSs formation during our sampling period at the polar sites may be hardly influenced by RH variations.” In line 378-384.

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