

Dear Editor

I am pleased to resubmit the revised manuscript of "Organosulfates and organic acids in Arctic aerosols: speciation, annual variation and concentration levels" for consideration to ACP.

The authors appreciate the comments and suggestions made by the referees and we have addressed each of their concerns as outlined below.

The most substantial change to the manuscript concerns the abstract. The abstract has been re-written in a more concise English language resulting in a decrease from 490 words to 352 words. Furthermore the concern of anonymous referee #2 about the two phthalic acid peaks in figure 1 has been clarified (see specific outline below).

Reply to Anonymous referee #1

General comment: This is an interesting study providing further evidence that (nitrooxy)organosulfates are ubiquitous in ambient atmospheric aerosols. What makes this study very unique is that it shows the time series of (nitrooxy)organosulfates over the course of a year in the arctic region. Overall the manuscript is well written and the results are presented in a clear manner. I have a small minor comment about the usage of the word 'tracer'; otherwise I recommend this paper be published in ACP.

1) P4751L15 The authors may want to consider a filter extraction procedure with a laboratory orbital shaker in future studies as the ultrasonication is known to degrade SOA compounds in the extract. See Mutzel et al., 2013.

A. Mutzel, M. Rodigast, Y. Iinuma, O. Böge, H. Herrmann, An improved method for the quantification of SOA bound peroxides. Atmos. Environ. 67, 365-369 (2013), Doi 10.1016/J.Atmosenv.2012.11.012.

Response: *We thank anonymous referee #1 for this excellent advice and for drawing our attention to the problem using ultrasonication in filter extraction. In future work we will consider possible alternatives.*

2) P4757L28 I feel the word 'tracer' should be reserved for those compounds that are known to originate from a certain source. Here, these organosulfates likely have multiple sources and it is more appropriate to state '...and OS 182 in the Arctic aerosols primarily originate from anthropogenic emissions. . .?'

Response: *We agree with the referee and have changed the sentences on page 4757, line 28 to "we propose that OS 140, OS 154, OS 168 and OS 182 in the Arctic aerosols primarily originate from anthropogenic emissions from combustion of fossil fuels and biomass burning."*

Reply to Anonymous referee #2

This paper reports organosulfates and organic acids in Arctic aerosols. The results are interesting to be published in ACP. However, there are several mistakes in spelling, misidentification of compound peak and unclear description. Those problems should be clarified before the consideration of acceptance.

1) Page 4746, line 4, and 2010, respectively. Add a comma.

Response: Done

2) Page 4751, line 16. Does the 90% acetonitrile solution contain 10% water or other solvent? Please clarify.

Response: Yes the 90% acetonitrile solution contains 10% milliQ water, which has now been clarified in the manuscript:

Page 4751, line 15 “The collected aerosol filters were extracted by sonication in 10 mL of a 90% acetonitrile/10% milliQ water solution in a cooled ultrasonic bath for 30 min.”

Furthermore we have also clarified the content of the solvent used for re-dissolution of the samples, by adding in MilliQ water on page 4751 line 19: “Each sample was re-dissolved twice in 0.5 mL of a solvent mixture of 0.1 % acetic acid and 3 % acetonitrile in MilliQ water.”

3) Page 4756, line 2. Pimelic acid may be of biogenic origin, which is derived from the oxidation of unsaturated fatty acids together with azelaic acid (Kawamura and Gagosian, Nature 325, 330-332, 1987).

Response: This is a very good point and we thank the referee for the comment. We have now added a sentence explain, that pimelic acid may be of biogenic origin as well as anthropogenic.

Page 4755, line 26: “In this study all organic acids are categorized into anthropogenic (benzoic acid, phthalic acid, adipic acid and pimelic acid) and biogenic (terpenylic acid, DTAA, pinic acid, pinonic acid, MBTCA, suberic acid and azelaic acid) tracers based on previous studies of their atmospheric formation and precursors (Williams et al., 2010; Claeys et al., 2009; Rybka et al., 2007; Ma et al., 2007; Mochida et al., 2003; Bunce et al., 1997; Hatakeyama et al., 1987).

However, pimelic acid, here assigned to be an anthropogenic tracer, may be of biogenic origin as well, derived from the oxidation of unsaturated fatty acids (Kawamura and Gagosian, 1987).”

4) Page 4757, lines 19-20. Glycolaldehyde, hydroxyacetone, etc. are also produced by field burning of agriculture wastes such as wheat straws (Kawamura et al., Atmos. Chem. Phys., 13, 5369-5380, 2013). This point can be added in the text.

Response: We thank the referee for pointing out additional sources to the compounds in question and they have been added to the manuscript together with the proper reference:

Page 4757, line 19: “Several studies suggest that glycolaldehyde, hydroxyacetone, methacrolein and 2-methylglyceric acid originate from biomass burning, field burning of agriculture wastes and anthropogenic sources such as automobiles and fossil fuel combustion as well as photooxidation of isoprene (...Kawamura et al., 2013).”

5) Page 4761, line13. Add a period at the end of the sentence.

Response: Done

6) Page 4761, line 19. “emissiond” should be “emission”.

Response: Done

7) Page 4761, line 21. Artic ! Arctic

Response: Done

8) Page 4764, line13. Please add the reference for Tokyo.

Response: Original references for the measurements in Alert, Tokyo and the Chinese cities have now been added to the manuscript:

Page 4764, line13: “...concentrations measured in Tokyo and in Chinese cities (Zhang et al., 2010; Ho et al., 2007; Kawamura and Yasui, 2005; Kawamura et al., 1996).”

9) Table 2. Give information for the abbreviations of DTAA and MBTCA as a footnote of the table, although they are given in the text.

Response: This is an excellent point and a footnote regarding the abbreviations has been added.

10) Fig. 1. The phrase “7 and 8. Phthalic acid” sounds strange. A single compound never shows two peaks. If peak 7 is phthalic acid, then peak 8 should be iso- or terephthalic acid. The authors need to identify the two peaks using authentic standards. If you do not have standards, the authors should at least describe that the peaks 7 and 8 are phthalic acid and its isomer (iso- or tere-phthalic acid).

Response: We agree with the referee that this point should be clarified and we have analysed authentic standards of phthalic acid, iso phthalic acid and terephthalicacid showing that peak 7 is phthalic acid and peak 8 is terephthalicacid.

This has now been explained in the figure caption and on page 4755 line 23 as well:

Fig. 1: “...7. Phthalic acid 8. Terephthalicacid”.

page 4755 line 23: “11 organic acids (terpenylic acid, benzoic acid, phthalic acid (given as the sum of phthalic acid and terephthalicacid)...”