

Interactive comment on “Organic Functional Groups in the Submicron Aerosol at 82.5° N from 2012 to 2014” by W. Richard Leaitch et al.

W. Richard Leaitch et al.

richard.leaitch@canada.ca

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Responses We are grateful to both reviewers for their generous and constructive comments as well as their time. In the revised manuscript, all revisions are highlighted. Those based on specific comments are highlighted in yellow. Revisions to improve the discussion are highlighted in blue.

Referee #1 Summary and Overall Recommendation:

Multi-year studies, such as the one presented here, on the chemical composition of submicron aerosol are highly needed in the literature and I feel are at times underappreciated by the atmospheric chemistry community. This study is highly unique in that it presents multi-year data (i.e., April 2012 to October 2014) on the organic functional

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groups (OFGs) found in submicron aerosol collected from the Arctic at the Alert Observatory. A lot of important observations are made about OFGs in submicron aerosol collected from the Arctic during this study. For example, the authors found that a secondary marine source is likely a general feature of summer OM, but during years where there is likely more combustion-related sources (such as biomass burning) the contribution of alkane groups to the OM can be higher. Overall, I think this study will be publishable in ACP. However, I do a few comments below that the authors should consider before full publication is considered.

1.) Generally, I feel at times the text in the discussion section can be a bit dense and hard to follow. I couldn't think of an easier way to reorganize the text, but I just thought to point this out to the authors, especially if this comment concerned them enough to consider reorganizing the discussion section.

Response – We appreciate that the writing is a little “dense”, and we thank both reviewers for their tolerance. We have edited some areas of the discussion (blue highlighted text) to try and make it a little more palatable, but it remains dense.

2.) Abstract, Line 12: Change "lower organic mass concentrations (OM)" to "Lower organic mass (OM) concentrations"

Response – Done.

3.) Abstract, Lines 16-17: If you are going to list the initial of the month in parentheses after each season, shouldn't you also do this for summer to be consistent?

Response – It is not given here because it was done on line 13.

4.) Abstract, Line 25: change "most persistence" to "most persistent"

Response – Corrected.

5.) 2.1 Instrumental Methods, Page 4: What was the temperature of the freezer at the Observatory? This is important to know so that readers can judge if potential changes

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in composition might have occurred.

Response – The freezer temperature (measured at -18.5C) is now given on line 35 of page 4.

6.) 2.1 Instrumental Methods, Page 5: When the authors state "Prior to OFG analysis by FTIR spectroscopy, the filters were equilibrated in a temperature and humidity controlled cleanroom environment for 24 h," what do you mean exactly? Is the temperature and RH always the same for all samples measured? What is the temperature and RH of this room? Does this change the composition since the aerosol were likely collected at much colder conditions in the Arctic?

Response - The filters are stored in sealed petri dishes inside double zip loc bags while frozen (below 0C). The closed petri dishes are moved from the freezer into the cleanroom for 24 hr prior to measurement in the FTIR spectrometer. The cleanroom is maintained at 20C and <40% relative humidity. A continuous N2 purge is used in the spectrometer. There is no evidence that the composition changes measurably during the freezer storage or cleanroom equilibration. This discussion has been added to lines 4-7 of page 5.

7.) 2.1 Instrumental Methods, Page 5: Did the authors consider conducting ammonium sulfate calibrations with the ACSM? Budisulistiorini et al. (2014, AMT) found this was necessary with the multi-year measurements of submicron aerosol in the southeastern U.S.

Response – Since we first started using the ACSM in 2009 (Takahama et al., ACP, 2011; Leaitch et al., AE, 2011), we have done spans with ammonium sulphate. We identified to Aerodyne a large difference in the measurement of sulphate by the ACSM compared with the old quad AMS and a HR AMS in 2010. The sulphate data here use the new relative ionization efficiency for sulphate. In association we have added reference to Budisulistiorini et al. (2014, AMT) on page 5, lines 35-36. We have had larger problems that have been hampered by limited hands-on attention at Alert and quite

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slow remote connections. With our limited personnel, ACSM software and hardware problems as well as upgrades, we have struggled to keep the instruments providing serviceable data; hence the limitations on the ACSM data here.

8.) Related to # 7 above, why didn't the authors consider presenting and comparing OFG data with PMF analysis of ACSM OM? Was it that the signals were too low for PMF analyses? Response – For the reasons mentioned in response to #7, our ACSM data are limited. We did consider using the few months of ACSM data in a more direct sense. However, compared with filter datasets covering approximately two and a half years, we believe it would substantially increase the complexity of the discussion without a clear advantage, and particularly considering the uncertainty in the CE.

9.) For OFG analyses by FTIR, one thing that really never comes across are the uncertainties of this technique, especially at low mass concentrations. Are the uncertainties accurately estimated? I worry that FTIR may have issues at these lower mass concentrations, which can affect all of the downstream analyses you conduct in this manuscript. This also relates to how well your peak-fitting method really works when you are limited by low amounts of OM collected on these filters. Are you missing any important functional groups? I would think offline mass spectral analyses of these filters should be something the authors consider in the future (not in this manuscript of course).

Response - Since samples were collected for multiple days in order to obtain mass loadings of 10 to 50 ug, comparable to studies in polluted areas with 4-6 hr sample times, the detection limits reported previously [Gilardoni et al.; Maria et al.] were applied here and the resulting uncertainties are similar to those previously reported ($\pm 21\%$ for OM [Russell, 2003]). Not all OFG were above detection, as reported in the manuscript. Their absence is included in the estimated uncertainty, as discussed by Russell [2003].

10.) When you say a "Mixed" factor this is very confusing to me. Is this mixed because

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PMF fails to resolve this potentially mixed statistical solution? I wonder if this is the case due to the robustness of OFG analysis by FTIR. As the authors now, this OFG analysis is not as specific as mass spec in resolving finer details in the chemistry. I guess this "mixed" factor is resulting from this underlying issue with OFG analysis by FTIR.

Response - In studies where more samples are taken at higher time resolution, more PMF factors have been resolved by FTIR [e.g. S. Liu et al. 2012], so the limitation is not the analysis technique but the number of samples. The number of samples is limited at Alert both by the clean conditions and the limited access and resources.

11.) I'm intrigued by the authors potential observation of secondary marine OM source. Do the authors think this could be BVOC-related emissions from plankton?

Response – We previously referenced papers by Willis (page 12, line 33), Mungall (page 14, line 16), Facchini (page 13, line 17) and Köllner (page 13, line 17; page 15, line 9) concerning secondary marine sources. We prefer to leave it at those references, rather than speculate here on the nature of secondary marine OM in the Arctic. Mungall et al found no evidence of significant levels of isoprene and monoterpenes in the gas-phase over the waters of Baffin Bay and the Nares Strait.

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