

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

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Received and published: 2 December 2017

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 #3

General comments This comprehensive study reports 2.5 years of weekly averaged data from the Arctic research observatory Alert. Measured species include elemental (EC) and organic carbon (OC), organic functional groups as measured by FTIR, inorganic species including oxalate and methanesulfonic acid (MSA), non-refractory

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species measured by an Aerosol Chemical Speciation Monitor (ACSM). Furthermore, particle size distributions were analysed by a Scanning Mobility Particle System (SMPS) and an Optical Particle Counter (OPC). The authors used chemical speciation in the Arctic aerosol, PMF and linear regression based on this data, and the transport model FLEXPART to associate organic aerosol components with source types and source regions. The manuscript is well written with sound discussions. The large number of figures in the manuscript could be reduced, e.g. moved to the supplementary section.

The authors state that filter-OC data will be published elsewhere, but it will add great value to the manuscript and be meaningful to compare this data with ACSM-OM concentrations and OFG-OM that has a central position in the source attribution. The authors build the factor analysis on organic functional groups, which may undergo atmospheric degradation during transport to Alert, which they correctly state could be much longer than 10 days. The authors should consider if chemical transformation of the functional groups into more oxidized in the (often acidic) arctic aerosols, could lead to erroneous conclusions in the PMF analysis.

Response 1) We do not wish to make this an intercomparison paper, but we have revised Supplement Figure S2 to include a time series comparison of OC from the Thermal Method (TM; details added to the Methods of the main text) and OC from the OFG analyses as S2a. In Figure S2c, we show a regression of the OFG-OC and TM-OC. A description of the TM-OC has been added to the Methods (page 4, lines 23-27). The TM-OC is based on OC derived at 550oC plus OC derived at 870oC. The additional OC at 870oC is based on isotopic analyses showing no significant carbonate (no reference). OC at 870oC has been shown before to correlate with OC and water soluble OC (Chan et al., ACP, 2010). For corresponding points, the mean TM-OC, OFG-OC and OFG-OM are 120 ng/m³, 64 ng/m³ and 120 ng/m³, respectively (now discussed on page 6, lines 10-14). Where the TM-OC is higher than the OFG-OM in the summer of 2013, the O/C calculated from the OFG co-varies with m_z44/m_z43 from

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the ACSM. Also, the OFG-OM, shown in Figure S2b, compares reasonably with OM estimated from the ACSM (see response 4 below). One factor that may contribute to the higher OC by the TM method is absorption of VOCs by the quartz filters used for sampling. Overall, we believe the variations in O/C, estimated from the OFG analysis, to be reasonable within defined uncertainties.

Response 2) We have added a statement (page 14, lines 7-8) to the effect that these OFG may have undergone transformation during transport, and that may impact the PMF analysis. However, whether and how this might impact the PMF analysis is unclear.

Specific comments

Line 181: How does the OFG-OM and ACSM-OM based mass concentrations compare with the offline OC measurements (estimating O/C equals that obtained from the FTIR measurements), and does the uncertainty in OFG-OM agree with that of Russel, 2003?

Response 3) – Please see Response 1 above. It seems that with this comment the reviewer is implying we can assess the uncertainty in the OFG-OM based on the ACSM-OM and the uncertainty in OFG-OC based on the TM-OC measurements. Whether that is the implication or not, the uncertainty in the ACSM-OM is relatively large due to the CE (see Response 4 below), and the TM-OC uncertainty is approximately +/-25%. Based on +/-25% for each of OFG-OC and TM-OC, their means fall within the uncertainty range. That has been added on page 7, lines 9-13.

Line 205: How is the collection efficiency (CE) justified? Based on the variation in chemical composition over the year, in particular with respect to sulphate, the acidity could vary substantially. Thus, a variable CE would probably be appropriate, e.g. based on the parameterization method. More in Middlebrook et al., 2012.

Response 4) – We calculated the CE based on equations 4 and 7 of Middlebrook et al. (2012). The resulting values of CE were relatively high (e.g. 0.75), which resulted in

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concentrations of sulphate and organics very much lower than the filter concentrations (either OFG or TM). Assuming that the organics are internally mixed with sulphate in the particles measured at Alert, we took the approach of Quinn et al. (2006) and set the CE based on a comparison of the raw ACSM SO₄ with the filter sulphate. The average of the CE values calculated that way is 0.20, but, as we show, the filter sulphate may include sulphate in particles larger than efficiently sampled by the ACSM (>500 nm). We also calculated ACSM-OM using a constant CE of 0.5 that is commonly used in many AMS publications and is consistent with the organic aerosol discussion of Middlebrook et al (2012). Time series of OFG-OM, TM-OC, ACSM-OM (CE based on comparison with filter sulphate and for a constant CE of 0.5) for the period of ACSM data are shown in Figure S2b. The statistics of linear regressions of OFG-OM with ACSM-OM and TM-OC with ACSM-OM, for the sulphate-based CE, are given in the caption, and they show that the OFG-OM and the ACSM-OM compare within viable ACSM collection efficiencies. This discussion has been added on page 6, lines 24-33.

Line 520: Does the possible marine influence of Factor 1 agree with MSA, or is this stronger in Factor 3?

Response 5) – The correlation with MSA is highest for factor 3. For factor 1, the CoD is 0.02. We have added the following sentence to the end of the paragraph discussing factor 1 (page 14, lines 19-20): “However, there are no correlations of this factor with Na⁺ or MSA.”

Line 561: Does the OFG-OM agree with filter OC derived concentrations?

Response – See response to 1.

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2017-511>, 2017.

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