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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Contributions of Transported Prudhoe Bay Oilfield Emissions to the Aerosol Population in Utqiaġvik, Alaska" *by* Matthew J. Gunsch et al.

Anonymous Referee #1

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This manuscript describes single particle characterization (ATOFMS and CCSEM-EDX) of particulate matter attributed to Arctic oil extraction activities at Prudhoe Bay, Alaska, and contrasts this with particulate matter dominated by natural emissions from the nearby Arctic Ocean for ∼1 month of measurements in late summer. This work contributes to the very few studies of local Arctic emissions of anthropogenic aerosol, and is therefore valuable in extending our understanding of local Arctic pollution sources in the context of the summertime Arctic natural background. The manuscript is overall well written and clear. It merits publication in ACP after the following comments have been addressed.

Major Comments:

In the introduction as well as in the discussion sections, the authors should make further efforts to ensure that proper, complete, and most importantly accurate, credit is given to prior related work. Specific instances are indicated in the minor comments.

Minor Comments:

Introduction: This introduction gives no context for the conditions expected in the unpolluted summer Arctic. The seasonal cycle in Arctic aerosol is very relevant to the context of these measurements, especially under the cleaner conditions of summer to autumn. Similarly, an acknowledgment of pollution influences in winter and spring is also relevant. The natural question here is how Prudhoe Bay emissions might compare to influences of long range transport in terms of aerosol loading and composition.

P3 L10-15: The discussion of BC and OC contributions from total Arctic oil and gas extraction, versus that from Prudhoe Bay is at present somewhat confusing. In addition, the methods used to arrive at BC and OC estimates might be relevant to this discussion, e.g., were in-situ measurements used to arrive at these estimates?

P3 L17: Are these US contributions from Prudhoe Bay alone or does this estimate represent expansion to other sites?

P3 L20: It is not strictly true to suggest that emission of gas phase species would lead to solely new particle formation. If the intention is to highlight the role that the very low condensation sinks of the summer Arctic could play in this respect, then the authors should state this clearly.

P3 L22: Flanner et al., 2013 does not indicate anything about the impact of BC on clouds. In abstract the authors state that the study has limitations owing to their lack of treatment of indirect effects.

P3 L25: Citation of other relevant work, such as Flanner et al, 2007 (and others) is advisable here

P3 L27: Is this result directly from Koch 2009, or is it elaborated by Bond 2013?

P3 L27-28: Did the modelling studies discussed here include local Arctic BC sources? This is worth discussing.

P4 L4: To do justice to the study of Barrett 2015 the authors should discuss the varying contribution of fossil fuels to Barrow EC over the winter period. Biomass burning is also an important source and can have local Arctic sources.

P4 L9: Do the authors suggest that due to the focus on biomass burning, the results of this study are less relevant to their discussion? The measurements of Brock 2011 likely represent some of the few more complete chemical characterizations of Prudhoe Bay aerosol emissions.

P4 L22: Cappa 2012 explicitly shows that absorption enhancement was not detectable, despite significant particle mixing, during their measurement campaign. This needs to be much more clearly stated, and the complexity of this issue need to be addressed. In particular, I do not agree with the statement that light absorption is enhanced the presence of sulfate or organic species in the same particles. Some studies have observed absorption enhancements (e.g., Knox 2009, Liu 2015) while others have not (e.g., Cappa 2012, Healy 2015). It is not clear whether this effect is significant in all ambient aerosol and this issue should not be stated as being entirely clear.

P4 L26: Is such a binary definition relevant to ambient particles? It might be more accurate to state that ambient aerosol can be found with range of mixing states approaching external or internal mixing in different cases and with influence from different source types.

Method section: Please ensure that all manufacturer information is as complete as possible with relevant names, models and locations.

P5 L21: How do back trajectories compare for the two sites?

P6 L23: Given the long sampling period and the acknowledgement of local vehicle

emission contributing to particle measurements, how might fast-varying local emissions (such as vehicles or generators) influence the particle composition interpreted from these 8h measurements?

Section 2.3: How is statistical significance determined for these measurements? What is the minimum number of particles that must be analyzed in order to to have a representative sample?

P8 L10: This is a remarkably small number of particle spectra to draw conclusions from. While I acknowledge the challenges of obtaining a large number of particle spectra in such a low concentration environment, the authors should acknowledge this limitation of the ATOFMS measurement in a more prominent manner (for example, Sierau 2014 acknowledges this challenge in their abstract). Related to this, what fraction of ATOFMS detected particles did not provide mass spectra? These limitations in no way contradict the main conclusions of the work, since the CCSEM-EDX analysis provides a complementary data set that provides similar conclusions; however, this limitation needs to be clearly stated.

P9 up to L25: Emission of small, primary particles from Prudhoe Bay emissions is likely also associated with emission of oxidizable or condensible gas phase species. Why might the Prudhoe emissions have stayed relatively small during transport to your measurement site?

P10 L8: Is comparison to Alert, Nunavut also possible?

P10 L20: However, your measurements show that the chemical composition of particles >100nm is different between the two types of air masses, and I doubt that no influence on these particle sizes occurs.

P11 L1: This work is from Summit, Greenland, which is arguably quite different from other, lower altitude, regions of the Arctic for a few reasons. First, ascent over Greenland can cause deposition of transported species leading to quite clean air masses.

Second, the lack of local sources (aside from snow pack photochemistry) compared to other Arctic regions which are directly subject to marine, coastal, and tundra influences. While some recent studies have suggested the presence of condensible material in the summer Arctic (e.g., Mungall 2017, Willis 2016), it would be very surprising if Prudhoe Bay did not also emit gas phase species. The authors might want to discuss what is known about gas phase emissions from oil and gas extraction. Some studies from lower latitudes (e.g., Liggio 2016) suggest these can be substantial.

P11 L5: A contradiction of your observations by those of Kolesar 2017 does not help to make your argument clearer here. If particle growth from Prudhoe Bay was observed previously what differences do we expect in late summer 2015? Was the time of year significant (Kolesar 2017 shows growth peaking in June to July)? What the meteorology different? This really needs further discussion.

P11 L20: Given that the ATOFMS misses sulfur-rich particles, which should be a reasonable contributor to Arctic Ocean air masses owing to DMS oxidation, how reliable are ATOFMS estimates of the fractional particle number contribution from various particle types? Is there some was to estimate the fraction of particles that are detected by the ATOFMS but not desorbed and ionized due to being sulfate rich? Table 1 would suggest a large fraction of particles contained sulfate. What fraction of particles scattered light in the ATOFMS, but did not produce mass spectra?

P11 L23: Would these sources not have been captured during the 'local' or Utqiagvik influenced periods?

P12 section 3.2.1: Besides trajectories and wind-sector analysis, what chemical characteristics do the authors have for the Arctic Ocean sector being representative of a clean marine background for the local region? The presence of BC-containing particles as well as aged SSA showing nitrate peaks (suggesting NOx chemistry), while I acknowledge that sulfate peaks could arise from interaction with DMS oxidation products, suggest a pollution influence (at least to some extent) on these air masses as

well. Do these air masses, either at the measurement site or at the Barrow Observatory, conform the the thresholds in BC (or absorbing aerosol) usually associated with clean marine conditions (e.g., < 50 ng/m3 of BC (Gantt 2013))?

P13 L10: Do these estimates still correspond to 2004, or rather a yearly average?

P13 L17: Is this single fragment really an unambiguous indicator of SOA formation? If SOA formation was occurring on Prudhoe bay emissions, why did the particles remain quite small, or put another way why do the authors suggest earlier in the text that particle growth did not occur?

P13 L27: It is difficult to draw conclusions from these differences, especially when the number of detected particles is so small. Do long term aerosol absorption data from the Barrow Observatory help with these conclusions at all?

P14 L16: Why is the main sulfate peak used to identify sulfur specifc different here compared to line 20 on the previous page?

P14 L20: Does a peak at NO2- indicate nitrite or just fragmentation of nitrate?

Figure 1: Average trajectories for the study period might be more meaningful that selected trajectories. Additionally, is the area North of Utqiagvik completely ice free during the measurement, as pictured?

Figure 2: Percentiles to illustrate the range of the data might be more appropriate here

Figure 4: Were elemental carbon peaks observed in OC particles? And similarly, was OC present on the EC particles detected? Can carbon and oxygen peaks be quantified in the CCSEM-EDX spectra?

Figure 5: The number of detected particles should be noted in this figure caption

Supplement: Is mention of the TMA containing particles warranted in the main body of the paper? It helps to show different source influences, since these were presumably detected during Arctic Ocean influence. Or, are there too few particle spectra with TMA

peaks to draw conclusions?

Specific comments:

P3 L4: natural gas P3 L7: Which types of pollutants are relevant here? Maybe list the major ones we expect, that are then discussed in following paragraphs? P4 L13: “primary aerosol can. . .” P4 L22: What is meant by “mostly” here? P9 L17: perhaps: “Arctic Ocean influenced periods” P9 L19: Prudhoe Bay air masses P15 L26: respectively P16 L5: “may contribute to further decrease”

References:

Flanner, M. G.: Arctic climate sensitivity to local black carbon, J. Geophys. Res-Atmos., 118, 1840-1851, 2013.

Flanner, M. G., Zender, C. S., Randerson, J. T., & Rasch, P. J. Present-day forcing and response from black carbon in snow. J. Geophys. Res. 112, doi:10.1029/2006JD008003, 2007.

Gantt, B. and Meskhidze, N.: The physical and chemical characteristics of marine primary organic aerosol: a review, Atmos. Chem. Phys., 13, 3979-3996, doi:10.5194/acp-13-3979-2013, 2013.

Healy, R. M., J. M. Wang, C.-H. Jeong, A. K. Y. Lee, M. D. Willis, E. Jaroudi, N. Zimmerman, N. Hilker, M. Murphy, S. Eckhardt, A. Stohl, J. P. D. Abbatt, J. C. Wenger, and G. J. Evans (2015), Light-absorbing properties of ambient black carbon and brown carbon from fossil fuel and biomass burning sources. J. Geophys. Res. Atmos., 120, 6619–6633. doi: 10.1002/2015JD023382.

Knox, A., Evans, G. J., Brook, J. R., Yao, X., Jeong, C. H., Godri, K. J., Sabaliauskas, K., and Slowik, J. G.: Mass absorption cross-section of ambient black carbon aerosol in relation to chemical age, Aerosol Sci. Tech., 43, 522–532, 2009.

Liggio, J et al., Oil sands operations as a large source of secondary organic aerosols,

Nature, doi:10.1038/nature17646, 2016

Liu, S., Aiken, A. C., Gorkowski, K., Dubey, M., Cappa, C. D., Williams, L. R., Herndon, S. C., Massoli, P., Fortner, E. C., Chhabra, P., Brooks,W., Onasch, T. B., Jayne, J. T.,Worsnop, D., China, S., Sharma, N., Mazzoleni, C., Xu, L., Ng, N., Liu, D., Allan, J. D. Lee, J., Fleming, Z. L. Mohr, C., Zotter, P., Szidat, P., and Prevot, A. S. H.: Enhanced light absorption by mixed source black and brown carbon particles in UK winter, Nature Communications, 6, 8435, doi:10.1038/ncomms9435, 2015.

Mungall, E et al., Microlayer source of oxygenated volatile organic compounds in the summertime marine Arctic boundary layer, PNAS, doi: 10.1073/pnas.1620571114, 2017.

Willis, M. D., Burkart, J., Thomas, J. L., Köllner, F., Schneider, J., Bozem, H., Hoor, P. M., Aliabadi, A. A., Schulz, H., Herber, A. B., Leaitch, W. R., and Abbatt, J. P. D.: Growth of nucleation mode particles in the summertime Arctic: a case study, Atmos. Chem. Phys., 16, 7663-7679, doi:10.5194/acp-16-7663-2016, 2016.

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