Response to Reviewer #1

> **We thank the reviewer for their helpful comments and suggestions. We provide below the original comments, shown in gray, and our responses, with specific revisions noted, in bold font.**

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.

> **We added and revised references used throughout, with specific instances requested by the reviewer addressed in the responses to the comments below.**

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.

> **We added a statement to the introduction (P3 L9-13) addressing pollution contributions from oil and gas extraction in regards to the seasonal cycle, which states: "The Arctic aerosol population is characterized by a maximum mass loading in the winter, due to transported pollutants from the mid-latitudes, and a minimum in the summer, when local sources, including sea spray aerosol, dominate (Quinn et al., 2002; Quinn et al., 2007). However, there is limited knowledge of aerosols produced within the Arctic, particularly in the context of changing emissions from both natural and anthropogenic sources (Arnold et al., 2016)."**

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?

> **We revised this paragraph to reduce confusion about what is from the overall Arctic vs. Prudhoe Bay.**

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

> **These particulate emissions represent contributions from all US Arctic oil and gas extraction sites. We clarified in this paragraph that Prudhoe Bay is the primary, but not only, contributor to the estimated US oil field emissions in the Arctic.**

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.

> **We revised this to state "…drilling operations can emit aerosol precursors (NO_x, SO_2, and VOCs) and alter oxidant levels, which can lead to the formation of secondary aerosol, as well as contribute to new particle formation…".**

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.

> **This was not clearly worded, and we clarified this to now state: "BC is estimated to have a warming effect on the Arctic atmosphere (e.g. Bond et al., 2013; Flanner, 2013; Flanner et al., 2007; Sand et al., 2013; Sharma et al., 2013)."**

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

> **References to Flanner et al. (2007), Sand et al. (2013), and Sharma et al. (2013) have been added to this paragraph.**

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

> **This is directly from Koch et al. (2009), and we updated the sentence to accurately reflect this.**

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

> **The following sentence was added on P4 L4-7 to discuss that these modeling inaccuracies may be improved by the inclusion of local Arctic BC sources. "Koch et al. (2009) suggest that Arctic BC concentrations are under-predicted by a variety of models by an average factor of 2.5, which may be improved by more accurately incorporating local BC sources (Flanner, 2013)."**

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.

> **Following the reviewer's suggestion, we expanded the discussion of Barrett et al. (2015) (P4 L9-11) to now state: "…similar to the recent results of Barrett et al. (2015) at Utqiaġvik during December 2012 – March 2013 when transported particles from Russian oil fields were observed. Barrett et al. (2015) also measured regional Arctic BC from both fossil fuel combustion and biomass burning."**

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.

> **We removed this statement; it was not our intention to appear to be overlooking the characterization efforts of Brock et al. (2011) in terms of the Prudhoe Bay aerosol.**

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.

> **We acknowledge that this is a complex topic and updated P4 L26-28 accordingly: "It is currently not clear whether light absorption by a BC particle is enhanced by sulfate or organic coatings (e.g., Cappa et al., 2012; Chung and Seinfeld, 2005; Healy et al., 2015; Jacobson, 2001; Knox et al., 2009; Liu et al., 2015; Moffet and Prather, 2009)."**

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.

> **We clarified this on P5 L3-5, which now reads: "Ambient aerosol populations typically vary between internal mixtures, with multiple chemical species contained within a single particle, and external mixtures, with chemical species present as separate particles (Prather et al., 2008)."**

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

> **This information has been added.**

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

> **These sites are separated by 5.5 km, less than the resolution of HYSPLIT (1 degree, ~100 km), with only flat tundra located between them (P6 L1). Therefore, use of HYSPLIT to differentiate between the sites is not possible, and trajectories at the two sites are expected to be similar.**

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?

> **Short particle spikes due to local emissions would have been detected as brief increases in particle number concentrations by the SMPS, operated with 5 min resolution (Figure S2). These events were only observed during the Utqiagvik influence sampling periods (e.g. Sept 7, see Figure S2); these local contributions were not observed during the clean Arctic or Prudhoe Bay influenced periods (Figure S2).**

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?

> **Significance was determined using binomial statistics, which we have clarified on P8 L5-6. The minimum number of particles for a representative sample are between ~300 and 1,000. See Willis et al. (2002), Section 4.6.2.**

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.

> **We acknowledge the limitations of having a small number of particle spectra, which is why much of the in-depth analysis focuses on CCSEM-EDX data. However, the main difference is that the ATOFMS used by Sierau et al. (2014) was operating properly, to our knowledge, and that paper relied solely on the ATOFMS data obtained. In contrast, during our study, there was an instrumental issue with the time-of-flight mass analyzer, as described on P8 L25- P9 L1. This resulted in an extremely low fraction of particles producing mass spectra. The mass analyzer was fixed following the field campaign, and laboratory tests verified that the mass spectra produced during the field campaign were accurate, with just a much lower fraction of individual particle mass spectra collected. In our more recent Arctic deployment of the ATOFMS (after fixing the mass analyzer), over 33,000 particles were chemically analyzed over a similar time frame as the present study (unpublished work). To more clearly acknowledge the limitations of the ATOFMS results in the present study, we now state the**

total number of ATOFMS particles chemically analyzed in the Figure 5 caption, in addition to the statement in the methods section (P8 L23).

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?

We reorganized this section (moved later paragraph discussing emissions of condensable gases up to P10 L13-20) and clarified the discussion. P10 L23-28 now reads "Kolesar et al. (2017) previously observed Prudhoe Bay air masses to preferentially exhibit particle growth, compared to Arctic Ocean air masses. However, particle growth was not observed to occur within all Prudhoe Bay air masses during the summer, and particle growth events were not observed in September in Utqiaġvik." Given the lack of measurements of trace gases both in Kolesar et al. (2017) and this study, it is not possible, unfortunately, to comment on the reason for the lack of observed particle growth. Note that Kolesar et al. (2017) did not observe any particle growth events in September in Utqiaġvik over multiple years.

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

We added a comparison to Alert on P11 L7. The full sentence now reads: "The median particle concentration within Arctic Ocean air masses is similar to the median particle number concentrations during August at Station Nord, Greenland (227 particles cm^{-3}, Nguyen et al., 2016) and Alert, Canada (~160 particles cm^{-3}; Croft et al., 2016), during September at Tiksi, Russia (222 particles cm^{-3}; Asmi et al., 2016), and within the range of observations onboard the Swedish icebreaker Oden from July – September during multiple central Arctic Ocean studies when the air masses were exposed to the open ocean (90-210 particles cm^{-3}; Heintzenberg et al., 2015).".

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.

While the aerosol size distributions above 100 nm were not statistically different between Prudhoe Bay and Arctic Ocean air masses, there were indeed chemical composition differences observed by ATOFMS and SEM-EDX (e.g. increased chloride depletion, coupled with nitrate and sulfate addition in sea spray aerosol). Since these differences in chemical composition are discussed in the later sections, this confusing sentence has been deleted.

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.

> **We removed the reference to Ziemba et al. (2010) for work at Summit. These sentences (moved to P10 L17-23 in response to an earlier comment) now read: "Based on the simulations by Fierce et al. (2015), particle growth during transport for particles ~30-50 nm would take ~1-7 days, if coagulation-dominated due to limited condensable material. Particle growth was not observed during this study, suggesting that sufficient condensable material was not available for an observable change in particle diameter. Therefore, particles of this size could potentially be transported from Prudhoe Bay to Utqiaġvik during the average 21 ± 7 h transit time. Given the lack of primary ultrafine aerosol sources between Utqiaġvik and Prudhoe Bay, it is suggested that these particles were likely transported from Prudhoe Bay."**

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.

> **Our results do not contradict those of Kolesar et al. (2017), as particle growth was not observed to occur within all air masses from Prudhoe Bay in the summer. As shown in Fig. 3 of Kolesar et al. (2017), particle growth events were not observed in September, with particle growth occurring only part of the time during August. We expanded on this discussion on P10 L23-27.**

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?

> **In order to further address this "missing" particle type, we added estimates of fractional particle number contributions based on the CCSEM-EDX observations of the sulfur-rich particles. The sentence on P12 L9-13 now states "Based on CCSEM-EDX analysis (Figure 6), these sulfur particles likely comprised ~10 – 30% of the 0.13 – 1 µm particle number fraction during Arctic Ocean air mass influence, and ~10 – 20% of the 0.13 – 0.3 µm particle number fraction during Prudhoe Bay air mass influence. Accounting for these sulfur**

particles would reduce the reported ATOFMS fractions by ~5 – 15% for Arctic Ocean air mass influence, and ~5 – 10% for Prudhoe Bay air mass influence.”

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

As shown in Figure 5 and now stated on P12 L19-20, the contributions from biomass burning and dust particles were greatest in the wind direction of Utqiaġvik influence.

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))?

As noted in Sec. 3.2.1 (P12 L24 – P13 L3) and shown in Fig. 6, nearly all of the supermicron particles were sea spray aerosol (SSA). As shown in Table 1, during the Arctic Ocean influence, Cl/Na ratios of the observed submicron and supermicron SSA particles were 0.81 and 0.99, respectively, indicative of fresh SSA and supporting a clean marine background; corresponding SSA S/Na ratios were also close to the expected seawater ratio (Keene et al., 1986). Previous remote marine studies have also measured some anthropogenic influences (Pósfai et al., 1999; Shank et al., 2012). Also, as discussed in Section 3.1, average particle number concentrations during the Arctic Ocean influence were comparable to other Arctic background sites, further suggesting that our measurements are representative of background Arctic conditions.

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

These estimates still correspond to 2004. The sentence has been clarified: “For air masses influenced by Prudhoe Bay emissions, increased number fractions of soot, OC, and partially aged SSA particles were measured, with increased soot and OC particles expected based on 2004 estimates of soot (1.9 kt) and OC (2.0 kt) emissions from US Arctic (primarily Prudhoe Bay) oil and gas extraction activities (Peters et al., 2011).”

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?

Previous ATOFMS studies (Qin et al., 2012) established *m/z* 43 ($C_2H_3O^+$) as a marker of oxidized organic compounds. However, since particle growth was not observed, we added the following sentence on P14 L12-13: “However, as particle growth was not observed during Prudhoe Bay air mass influence (Section 3.1), it is likely that SOA contributions to particle mass were minor.”

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?

> **We incorporated data from a co-located aethalometer and revised the sentence now on P14 L21-24 as follows: "Soot was also identified by ATOFMS during Prudhoe Bay periods by C_n^+ clusters (C^+ [*m/z* 12], C_2^+ [*m/z* 24], C_3^+ [*m/z* 36], etc). Elevated black carbon mass concentrations (up to 0.27 μg/m^3) were also measured by the aethalometer during the Prudhoe Bay air mass on August 25 (Figure S5)." Unfortunately, aerosol absorption data from the Barrow Observatory are unavailable during the Prudhoe Bay influenced periods we chemically characterized in late September.**

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

> **This is due to spectral interference between HSO_4^- and $NaCl_2^-$ in SSA. We have clarified this by adding the following sentence to P12 L3-5: "Sulfate is identified as SO_3^- (*m/z* -80) in SSA due to mass spectral interferences between HSO_4^- [*m/z* -97] and $NaCl_2^-$ [*m/z* -93,95,97] (Qin et al., 2012; Sultana et al., 2017)."**

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

> **Based on previous ATOFMS studies, this is likely a fragment of nitrate (e.g. Dall'Osto et al., 2009; Liu et al., 2003). The nitrite identification has been removed.**

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?

> **As suggested, we replaced the representative trajectories with average trajectories. The area north of Utqiagvik, shown in the Figure, was completely ice free during the study, as pictured.**

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

> **We added Figure S3, which shows the 25th, 50th and 75th percentiles for the Prudhoe Bay and Arctic Ocean particle number distributions. A note was also added to the Figure 2 caption referring to this new supplemental figure.**

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?

> **OC markers were not observed in the EC particle mass spectra. However, minor contributions from m/z 12 (C^+) were observed in the OC particle mass spectra (now noted in the Supplemental P1 L20); however, without evidence of many carbon cluster ions, this ion peak can also be attributed as an OC fragment ion. However, the EC peaks in the OC mass spectra were minor and therefore not labeled in Figure 4 due to space constraints.**

Unfortunately, there are substrate interferences with carbon and oxygen peaks in the CCSEM-EDX spectra.

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

This is now noted.

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?

Unfortunately, there were too few TMA-containing mass spectra measured to draw conclusions. We added a sentence to Supplemental P1 L26 indicating this.

Specific comments:

P3 L4: natural gas

We added this.

P3 L7: Which types of pollutants are relevant here? Maybe list the major ones we expect, that are then discussed in following paragraphs?

We now list PM, VOCs, SO_2, and NO_x here.

P4 L13: "primary aerosol can. . ."

We added this.

P4 L22: What is meant by "mostly" here?

We replaced "mostly" with "primarily".

P9 L17: perhaps: "Arctic Ocean influenced periods"

We changed this to "Arctic Ocean influenced air masses".

P9 L19: Prudhoe Bay air masses

We've changed this wording to Prudhoe Bay air masses.

P15 L26: respectively

We've corrected this word.

P16 L5: “may contribute to further decrease”

This has been added.

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