

*We thank the reviewers for their insightful feedback. We have substantially revised the manuscript as a result. Mainly, we revised and coordinated almost all of the figures and improved upon the writing quality of the text based on the suggestions provided. We also revised the source classifications and redid any calculations resulting from such changes, all of which are reflected in the manuscript.*

## **Reviewer 1**

This study analysed the airborne observations during ACME-V campaign along the North Slope of Alaska in the summer of 2015 and found that summertime Alaskan Arctic was not pristine as suggested by previous evidence, but was with higher aerosol loading and trace gas concentrations than measurements even in Arctic haze. Local oil extraction activities, central Alaskan wildfires, and to a lesser extent, longrange transport enhanced the aerosol and trace gas concentrations in Alaskan Arctic during summertime. Quantifying aerosol loading and sources in the Arctic is challenging. The aircraft observations presented in this study is therefore an important contribution to the field, but the analysis and writing quality of this manuscript is really poor. I recommend publication in ACP after major revisions and substantial improvements.

Major comments:

1. The analysis of the data was a little superficial and I suggest the authors dig deeper. For example, in Figs. 3, 7 and 9, the data were color coded by flight numbers, which does not provide any valuable information. They already classified the flights into several air mass types as shown in Table 2 and Fig. 10. I think analysis based on different air mass types would provide more information than the current flight numbers used in the manuscript. In addition, Figs. 2, 3 and 4 discussing the impacts of oil extraction was based on data during the whole campaign. I suggest select the period during which these sources dominate would be better to illustrate their contributions.

*We agree with the reviewer that the data colored by flight number is not useful and redundant to Table 2 and Figure 9 (was Figure 10). We want to note that the analysis in the manuscript is intended as an overview and presentation of the unique dataset to show the influence of the sources in the Prudhoe Bay area. More detailed studies are currently undergoing the planning phase to elucidate aerosol sources in a more specific manner. However, in an effort to conduct a deeper analysis based on air mass types, we tied in the source classifications more thoughtfully throughout the discussion and respective figures. For clarity and consistency, we revised most of the figures to show all data and those data classified by the air mass types. Much of the text for section 3.1 and some of the text in section 3.2 was updated to reflect the source-specific analysis in the figures. Specifically regarding the figures, we:*

- *Revised the color scheme in Figure 9 (was Figure 10) for each source; now all source colors are consistent throughout all of the figures.*
- *Changed the color scales in Figures 2 and 5 (was Figure 6) to reflect the approximate source colors.*
- *Removed the panels colored by flight number entirely from Figure 3 and colored the data impacted by Prudhoe Bay in blue (all other data in grey).*
- *Combined Figures 3 and 4 into the new Figure 3.*
- *Changed the color scale in Figures 4 and 7 (were Figures 5 and 8) to match fires source color.*
- *Revised Figures 6 and 8 (were Figures 7 and 9) show the vertical profiles and/or correlations for all data and those data impacted by fires and all sources, respectively.*
- *Kept the spatial averaged maps to show, qualitatively, the spatial variability in the parameters and to demonstrate the locations impacted by Prudhoe Bay and the fires. However, the color scales now reflect the assumed sources.*

*With regard to the last point (restricting the oil extraction analysis to the periods during which those sources dominate), it is important to note that this signal is continuous and was encountered by almost all flights as they traversed the Prudhoe Bay area. Therefore, we felt that it was appropriate to highlight the specific signal of these emissions in contrast to the background signal encountered during much of the rest of the flights at altitudes below 500 m.*

2. The manuscript was poorly organized, making it really hard to follow. For example, in Sect. 3.1, figures were discussed back and forth. Fig. 2 c and d were discussed after Fig. 4. In the same section, the idea that ‘high concentration of small particles are restricted within 50 km of Deadhorse’ has been discussed several times (P6,

L21–26, P7, L14–16, and P7, L24–25). In Sect. 3.2, discussions of different species were also jumped back and forth. For instance, aerosols were discussed in P8, L15–19, P8, L25–29 and P9, L21–25. Background concentrations of CO and enhanced CO were discussed back and forth in P9, L1–14. In Sect. 3.3, the second paragraph discussing air mass types along the flights does not belong to this section, which is supposed to discuss the contribution from long range transport. Long range transport deserves more analysis.

*We went through and reorganized to ensure the figures are discussed in an orderly manner and prevent redundancy in ideas presented. We also added a paragraph describing the classifications of sources in more detail, which helps elucidate the long-range transport analysis. However, we disagree that the second paragraph in section 3.3 does not belong. The focus of the paper is on the abundant local and regional sources (which may be increasingly important in a dynamic Arctic environment), while long-range transport is secondary. Our study and previous studies have indicated that this is not an important source in the summer as compared to the winter/spring. The purpose of this section is to discuss the contributions from all sources compared to one another. A more detailed analysis of long-range transport would require extensive air mass trajectory analysis in addition to other remote sensing or modelling techniques to accurately evaluate long-range sources; this is outside the scope of our manuscript. To reflect the secondary importance of long-range transport, we removed ‘long-range transport’ from the title.*

*For the figures, we revised so that they are in order when first presented, however, we do refer back to certain figures when discussing different parameters. For Prudhoe Bay, we organized the discussion such that we focus on each measurement parameter (i.e., nucleation mode aerosol and rBC) at a time, which is why we go back and forth between Figures 2 and 3. For the fires, we discussed rBC and CO back and forth because they are related, correlate strongly, and thus both used as tracers for the fires. For both sections, we now show and discuss HYSPLIT first to qualitatively provide spatial evidence of the sources, then discuss how the measurements support the source modeling.*

3. Another problem of the paper is the sloppy style of writing and the use of the English language. For instance, the tense was wrong in numerous places. To name a few, P1, L 22–44, P3, L16, and P3, L30. The references were not always written in the correct format. ‘... and colleagues (year)’ should be ‘... et al. (year)’. The acronyms were not properly used (e.g. ‘rBC’ and ‘black carbon’, ‘CO’ and ‘carbon monoxide’ were used back and forth; AMSL and MSL were not spelled out when they were used for the first time; ARM and AOD were spelled out twice). A lot of ‘and/or’ were used. Please double check and delete ‘and’ or ‘or’. I also list a few other problems in the ‘Minor comments’ section, but all these I’ve pointed out are only a few of the language problems in the manuscript. I suggest a much more careful checking of the manuscript and a substantial improvement of the language.

*We cleaned up the writing style throughout the manuscript and made sure we corrected wrong tense usage, citations, and acronym consistency and definitions.*

4. In section 3.2, please compare the fire activity in summer 2015 with climatology to illustrate how representative the summer is.

*We now state that it is the second largest number of acres burned since records began in 1940 based on the findings of Partain Jr., J. L.; Alden, S.; Strader, H.; Bhatt, U. S.; Bieniek, P. A.; Brettschneider, B. R.; Walsh, J. E.; Lader, R. T.; Olsson, P. Q.; Rupp, T. S.; R.L. Thoman, J.; York, A. D.; Ziel, R. H., An Assessment of the Role of Anthropogenic Climate Change in the Alaska Fire Season of 2015. Bulletin of the American Meteorological Society 2016, 97, (12), S14-S18.*

5. Axis labels of Fig. 7b are wrong.

*Fixed.*

Minor comments:

1. P2, L22: ‘to discover’ is inappropriate here. Revise please.

*Changed to ‘to conclude’.*

2. P2, L25: ‘during their Aug–Sep 2015 study’ -> ‘during Aug–Sep, 2015’

*Done.*

3. P2, L29: 'exists' -> 'locates'

*Changed to 'is located'.*

4. P2, L31: revise 'provides the ability to ...'

*We removed this sentence and instead combined with the second sentence in the paragraph to, "This site is located in the northwest region of oil extraction activities in Prudhoe Bay, making it an ideal location to determine the potential impacts of emissions from such activities on the relatively pristine Arctic atmosphere."*

5. P2, L33: 'long-range transported aerosol from lower latitudes' -> 'long-range transport from lower latitudes'

*Done.*

6. P3, L1: 'insight' -> 'insights'

*Done.*

7. P3, L2–3: please provide proper references

*Done.*

8. P4, L23: CO<sub>2</sub> were also discussed.

*Changed to 'aerosol, CO, and CO<sub>2</sub>'.*

9. P5, L6: particles -> particle

*This should be particles.*

10. P5, L23: 'landing the Deadhouse' -> 'landing in the Deadhouse''

*Changed to 'landing at the Deadhorse'.*

11. P5, L30–33: please show these locations in related figures.

*These are already shown in Figure 4 (was Figure 5) as indicated in the caption. However, we changed the color of the location markers to make them more evident.*

12. P6, L17–18: please clarify which data were used.

*Clarified that these are thermal anomaly data.*

13. P6, L19: '&' -> 'and'

*Done.*

14. P6, 31: those vapours does not nucleate, the secondary products are. Please clarify.

*Done.*

15. P8, L31: Please provide concentration values in standard summertime and springtime.

*We removed this part of the sentence because the SP2 measures refractory black carbon, and the values in most studies from the North Slope are either equivalent black carbon, or modeled black carbon. Thus, they may not be directly comparable due to possibly slight variations in sampling techniques.*

16. P9, L4–5: please clarify whether it is active flaming or smoldering.

*We already stated that the MCE value indicated active flaming, but changed ‘versus’ to ‘instead of’ for clarity.*

17. P10, L9: what are the tracers?

*We added ‘CO and rBC’ at the end of this sentence.*

18. P19: Figure 2. [mass m<sup>-3</sup>] is not a unit

*This is the unit defined by the HYSPLIT manual. It is an arbitrary unit. We now describe this in more detail in section 2.4 (was section 2.3).*

19. P22: Figure 5. Move the colour bar to the bottom of the figure.

*Done.*

## **Reviewer 2**

This paper presents measurements of aerosols made during the U.S. Department of Energy Atmospheric Radiation Measurement (ARM) program’s Fifth Airborne Carbon Measurements (ACME-V) campaign along the North Slope of Alaska during the summer of 2015. The paper focuses on how local oil extraction activities long-range transport influence aerosols and trace gases in the North Slope of Alaska. The authors should try to go beyond presenting the measurements and use the data in a clearer way to demonstrate the scientific conclusions that can be made using the data. This paper is within the scope of ACP and should be published following after the authors address the following comments:

1. I agree with reviewer #1 that the authors should rethink how to present the data in a less superficial way in addition to showing the data as a function of the flight number. I don’t object to showing these figures (Figs. 3, 7 and 9) as long as the data is shown again in a more synthetic way later in the paper, allowing the authors to draw more clear conclusions from the measurements.

*Please see response to major comment 1 from reviewer 1.*

2. The choice for the classification parameters and thresholds values in Table 2 should be more clearly justified. Have these been chosen using the Hysplit analysis?

*These are partially based on HYSPLIT analyses, but additionally on thresholds from previous work, and visual assessment of the proximity to known sources and vertical profiles. We added a new paragraph at the end of section 2.4 (was section 2.3) describing Table 2 classifications and how they were derived. In order to follow details on characterization of the fire locations, we moved what was section 2.3 to the end of the methods, after we discuss supporting satellite data.*

3. The way that Hysplit has been run should be more clearly described and justified. Even though the authors reference another paper for the description of the Hysplit runs, there is not enough information to fully understand how Hysplit was run. I assume this was run in backward mode from the measurement locations, but this is not clear. The reason for the choice of the five locations in the active fire region is also not clear. I also cannot fully understand Figure 2d and Figure 6d.

*The new paragraph describing the source classifications now provides justification for the HYSPLIT dispersion analysis. Dispersion simulations are automatically run in forward mode since it simulates emission and transport of particles from a point source. We added more detail in this section describing what information the HYSPLIT*

*dispersion model provides, which also clarifies what is shown in Figures 2a and 5a (were 2d and 6d), in addition to provided more detail on what the model output is. The five locations were chosen based on equal spacing within the highest density of fires determined from the satellite analyses from the entire study time period. This is now stated in the aerosol dispersion modelling section (2.4; was section 2.3).*

4. The reason for showing the data as column averaged values in Figures 2 and 6 needs to be justified. Don't we lose information by showing the data in this way? The main advantage of using aircraft data is that we know where the aerosol layers are vertically. The information we can learn from the altitude of the aerosol layers should be a clearer part of this analysis.

*The purpose of the maps in Figures 2 and 5 (was Figure 6) is to show, qualitatively, the spatial variability in the parameters and to demonstrate the locations impacted by Prudhoe Bay and the fires. Also, the column averaged data in Figure 2 is for altitudes < 500 m AMSL. For Figure 5 (was Figure 6), data are restricted to < 5000 m AMSL to show the vertical extent of the fire impacts. These parameters are shown as vertical profiles of the 1-second measurements in the following figures, thus any information that may be lost in Figures 2 and 5 are shown elsewhere. However, the conclusions discussed for these figures are supported by the vertical analyses.*

5. The MODIS detected fire hotspots should be shown on Figure 5 relative to the fire size or fire radiative power, such that more active fires can be identified vs. less active fires.

*Fire size and radiative power information is not available from the thermal anomaly data we used. Additionally, we used the fires as a qualitative approach to evaluate when and where these sources were present, and used the spatial density of the data to determine where HYSPLIT dispersion simulations should be initiated. Evaluation of fire properties is outside the scope of our manuscript.*

6. The influence of oil exploration is not clear to me. Is the location of oil exploration activities known? The discussion of oil exploration influenced air that was sampled should be clarified. The discussion of long range transport also needs to be developed, as noted by reviewer #1.

*This should be evident now given the additional paragraph describing the source classifications. Additionally, previous work by Gunsch et al. (2017) and Kolesar et al. (2017) clearly demonstrate how oil exploration from Prudhoe Bay is an influence on the North Slope. The locations of the oil activities (i.e., the active oil wells) is provided in Figure 1 and now include access date of the data. Please see response to reviewer 1's comment regarding long-range transport.*

7. The authors should review the manuscript writing to clean up the writing style and typos before resubmission.

*Done.*

### **Reviewer 3**

This study reports new aerosol observations from the summertime ARM ACME-V field campaign on the North Slope of Alaska. Their results indicate that oil exploration activities in Prudoe bay may contribute significantly to local aerosol concentrations. In particular, they observed high concentrations of nucleation mode particles in the area. These observations are novel and interesting and certainly add to the scientific discourse. However, many of the conclusions presented I feel are not fully justified by the results discussed. Additionally, the paper is difficult to read in several sections and requires clarification on several points. Please see below for specific instances.

Major comments:

Page 2 lines 3-4 This entire sentence is very confusing, what do the authors mean by 'beyond greenhouse gases' and which 'climate feedback' are they referring to? Please refer to the specific feedback (I assume ice-albedo) and rewrite the sentence to improve clarity.

*We specified that this is the ice-albedo feedback and reworded the sentence to, "In addition to the ice-albedo feedback described above, the principal atmospheric constituents that perturb the surface energy budget are clouds and aerosols."*

Page 2 line 5 'indirectly impact radiation through their role in cloud lifecycle' Aerosol can indirectly result in radiative forcing by increasing cloud lifetime, changing albedo and (in the case of INP) changing cloud phase. Please rewrite the sentence to address generalities (cloud microphysics) or specific processes.

*We rewrote the sentence so that it says 'roles in the modulation of cloud microphysics'.*

Page 3 line 3 'The Arctic atmosphere can be highly stratified' This statement is included without explanation or more (crucially) reference. Please cite a supporting reference.

*We added two key references.*

Page 3 lines 3-4 'at the height at which cloud modulation by aerosols occurs' This statement is very vague. To what height are you referring? Cloud base in the Arctic can be extremely low.

*Good point, Arctic cloud base can indeed be extremely low. We removed 'at the height at which cloud modulation by aerosols occurs' that sentence.*

Page 3 lines 4-5 'have focused in evaluating Alaskan Arctic aerosol sources' Why would focusing on aerosol sources help our understanding of higher altitude aerosol in the Arctic? I assume the authors mean specifically sources of higher altitude aerosol?

*We changed this sentence to read, "Accordingly, numerous airborne campaigns have focused on evaluating sources of mid- to upper-tropospheric aerosol and aerosol-cloud interactions."*

Page 3 lines 23-26 This statement seems to be conflating future and present sources of aerosol in the Arctic and while forest fires are indeed an important source the relative importance of local vs transported aerosol is not well understood. Thus, while 'great importance' may be justified for boreal forest fires (during some periods of the year) I would prefer more nuance when discussing local fossil fuel and BB combustions (by which I'm assuming the authors mean domestic wood burning?).

*We are referring to projected increases in forest fires due to a warming climate. Domestic wood burning should not play a dominant role in the summertime in this region, particularly when compared to the widespread forest fires. However, to make this sentence clearer, we changed to, "In the context of warming temperatures, emissions from oil extraction, added shipping routes due to a reduction in sea ice extent, and wildfires are expected to increase in sub-Arctic boreal regions (Randerson et al., 2006; Gautier et al., 2009; Harsem et al., 2011; Peters et al., 2011; de Groot et al., 2013; Roiger et al., 2015). Thus, regional fossil fuel and biomass burning combustion sources will further contribute to the aerosol population may serve as an increasingly crucial source of aerosol in the future."*

Page 4 line 7 'predominantly decoupled' Why is the summertime Arctic decoupled? And is this also true for the sub-Arctic region that this paper focuses on? If the Arctic is less polluted in the summer (because as your references suggest it is decoupled from the mid-latitudes) does this not suggest that local sources are unimportant?

*Thank you for pointing this out. Previous work does show that sub-boreal regions can still contribute. We referred to midlatitudes as those in the lower 48, but realize this is not correct. Thus, we simplified the sentence to, "The Arctic summertime atmosphere is historically less polluted as compared to the rest of the year (Quinn et al., 2002; Leitch et al., 2013; Heintzenberg et al., 2015), thus it is critical to assess the impacts of potentially important local sources of summertime aerosol on Arctic radiation and cloud microphysical processes."*

Page 6 lines 23-24 'Hotspots of larger particles.. were not observed near Prudhoe bay (not shown)' I don't understand why you include this sentence were you expecting to see larger particles?

*This sentence was removed.*

Page 7 lines 2-10 Here you suggest that predominance of larger particles above the BL is the result of growth during vertical transport and dynamical restriction of nucleation mode particles in the BL. It surely can't be the result of both?

*We changed this sentence to say 'or' to demonstrate that it could be from one or the other process.*

Page 8 lines 4-9 beginning 'in terms of indirect forcing' Is the argument here that accumulation mode aerosol derived from nucleation have an impact on cloud properties or that the nucleation mode particles affect cloud directly? If the latter, please explain why marine aerosol may be more hygroscopic than sulphate. Petter et al., (2007, ACP) suggest similar kappa values for both.

*The purpose of this paragraph is to provide broader implications for the direct and indirect impacts of aerosols from this source from previous work. It appears the beginning of that sentence caused confusion, so we removed it. We did not intend to indicate that marine aerosol may be more hygroscopic than sulfate. However, in addition to the sulfate, oil extraction and marine emissions include a host of different organic species that partition to the particle phase, which are not very hygroscopic. Thus, it is difficult to say which general emission source would produce the most hygroscopic aerosol, and is why we broadly state that their hygroscopicities could vary.*

Page 8 line 9-11 beginning 'In general, our results' Given your previous statement that the aerosol appears to have a trivial direct forcing effect and extremely uncertain indirect effect is your final statement really justified?

*This sentence was removed.*

Page 8 lines 27-29 Why would you expect to observe an 'abundance of coarse and accumulation mode particles' if the wildfires generate secondary organic aerosol?

*Although SOA is generally smaller in nature, they can age as they are transported and grow in size. We clarified this here.*

Page 8 line 31 'Which are higher than standard summertime and even springtime haze concentrations' what is the 'standard' concentration and why have you not provided citations here?

*See response to analogous comment by reviewer 1. This part of the sentence was removed.*

Page 9 lines 11-13 'Our observations are parallel to previous summertime observations from regional boreal fires in that they produce substantial quantities of aerosol...' I'm genuinely unsure what you mean by this sentence although I am confident that it is not your observations producing aerosol. Please clarify.

*We changed to 'in that such fires produce substantial quantities of aerosol' to clarify that we meant they are produced from fires.*

Page 11 lines 10-11 'but demonstrating the larger impact of nucleated aerosol in the vicinity surrounding Prudoe bay' Assuming you mean climate impacts I fail to see how this study demonstrates any impact from these aerosol. You state on page 7 (lines 31-33) that you didn't observe any direct forcing. I also see no evidence (from these observations) of the indirect impact. Please clarify whether the observations reported here do or do not suggest a significant indirect aerosol forcing from emissions in Prudoe bay and provide greater justification for this conclusion.

*We intended to suggest that this is a larger source than previously reported. To reflect this, we changed the sentence to, "Probably the most notable observation was that Prudhoe Bay is a persistent but localized source of black carbon and especially nucleated aerosol, supporting previous findings at Utqiagvik from Kolesar and colleagues (2017) and Gunsch and colleagues (2017), but demonstrating the larger influence of particle nucleation on the aerosol population in the vicinity surrounding Prudhoe Bay." Additionally, the impacts of these aerosols are detailed by Maahn et al. (2017).*

Page 11 lines 18-21 'With both fire activity and oil exploration projected to increase in a warming climate, these sources will likely continue to make significant contributions...' Previously the authors have stated that emissions in Prudoe bay have a localized impact only, with this in mind can the authors justify so strong a statement on the importance of future oil exploration to Arctic aerosol?

*Even though this source is what we call localized, the HYSPLIT dispersion analyses indicate that dispersion is highest within a 2 degree region, but lower mass concentrations are transported over the entire study area. Thus, a fraction of these particles is transported regionally and could thus have implications for regional climatic impacts. We think this statement is relevant for our results, but have clarified what we mean by 'localized' throughout the manuscript.*

Minor comments

Page 2 line 3 Missing 'the' before climate feedbacks. However, to improve clarity I would suggest replacing 'climate feedback' with 'ice-albedo feedback' (please see first major comment)

*Added 'the' and changed to 'ice-albedo feedback'.*

Page 2 line 6 Use of the phrase 'hinges' is colloquial I would suggest changing to 'depends'

*Done.*

Page 2 line 7 'inherently depends' is a redundancy please delete inherently

*Done.*

Page 2 line 7 Replace 'atmospheric processing' with age

*Changed to 'extent of aging'.*

Page 4 line 9 'important sources of aerosol' You mean important local sources of summertime aerosol?

*Yes, changed to 'local sources of summertime aerosol'.*

Page 4 line 11 To improve clarity please replace 'such' with local

*Done.*

Page 6 line 31- Page 7 line 32 Please split your citations to differentiate between those referencing flaring emissions and those referencing nucleation mechanisms.

*These are already split up.*

Page 7 line 2 Please change 'removal of particles' to 'transfer of particles' or equivalent. Removal suggests removal of the particles from the atmosphere.

*Changed to 'transition of particles'.*

Page 7 line 13 Please cite Stohl et al., 2013 (<https://www.atmos-chemphys.net/13/8833/2013/>) in reference to BC emissions from flaring.

*Done.*

Page 7 line 27 Please replace 'loss' with 'transition'. The particles aren't lost there just bigger.

*Done.*

Page 8 line 7 Are you referring to diameter or radius here in reference to CCN?

*Clarified that this is referring to diameter.*



Page 8 line 16 ‘evidenced by the elevated AOD originating from central Alaska’ The elevated AOD is the result of aerosol originating from central Alaska. I would suggest rewording to ‘the elevated AOD originating from central Alaskan wildfires’ or equivalent.

*Changed to ‘the elevated AOD originating from the central Alaskan wildfires’.*

Page 8 line 17 ‘extended until the end of Jul’ What was extended until the end of July?

*Changed sentence to, “ACME-V flights were impacted by the high AOD regions from late-Jun until end of Jul.”*

Page 10 lines1-2 I agree with this statement but both of the papers cited are concerned only with the Alaskan Arctic. I would also suggest citing Garrett et al., 2010 <http://journals.co-action.net/index.php/tellusb/article/view/16525/0>, Browse et al., 2012 <https://www.atmos-chem-phys.net/12/6775/2012/> or Eckhardt et al., 2003 <https://www.atmos-chem-phys.net/3/1769/2003/acp-3-1769-2003.html> (among others).

*Thank you for bringing the suggested references to our attention. We have added all three to this sentence.*

# The influence of local oil exploration and regional wildfires on summer 2015 aerosol over the North Slope of Alaska

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**Abstract.** The Arctic is warming at an alarming rate, yet the processes that contribute to [the](#) enhanced warming are not well understood. Arctic aerosols have been targeted in studies for decades due to their consequential impacts on the energy budget, [both](#) directly and indirectly through their ability to modulate cloud microphysics. Even with the breadth of knowledge afforded from these previous studies, aerosols and their effects remain poorly quantified, especially in the rapidly-changing Arctic. Additionally, many previous studies involved use of ground-based measurements, and due to the frequent stratified nature of the Arctic atmosphere, brings into question the representativeness of these datasets aloft. Here, we report on airborne observations from the U.S. Department of Energy Atmospheric Radiation Measurement (ARM) program's Fifth Airborne Carbon Measurements (ACME-V) [field](#) campaign along the North Slope of Alaska during the summer of 2015. Contrary to previous evidence that the Alaskan Arctic summertime air is relatively pristine, we show how local oil extraction activities, 2015's central Alaskan wildfires, and to a lesser extent, long-range transport introduce aerosols and trace gases higher in concentration than previously reported in Arctic haze measurements to the North Slope. Although these sources were either episodic or localized, they serve as abundant aerosol sources that have the potential to impact a larger spatial scale after emission.

## 1 Introduction

25 The Arctic is warming rapidly compared to other locations globally, which has implications for anomalous snow and ice melt (Jeffries et al., 2013). Replacement of highly reflective surfaces by darker, more absorbing surfaces (i.e., tundra and open ocean water) enhances Arctic warming, especially in the summer (Chapin et al., 2005). Such warming subsequently impacts the ecological cycle, socioeconomics, and mid-latitude weather patterns (Screen and Simmonds, 2010; Serreze and Barry, 2011). This warming feedback is one in a complex system of interrelated processes resulting in "Arctic amplification", the 30 observed enhanced warming seen in the Arctic to date, and in part motivates the need to improve our understanding of atmospheric processes that modulate energy reaching the Arctic surface (Serreze and Francis, 2006).

In addition to ~~the ice-albedo feedback~~ the ice-albedo feedback described above, the principal atmospheric constituents that perturb the surface energy budget are clouds and aerosols. Aerosols can directly scatter and absorb solar radiation or indirectly impact atmospheric radiation through their roles in the modulation of cloud microphysics by serving as cloud condensation nuclei (CCN) or ice nucleating particles (~~INPs~~ INPs). However, the ability of aerosols to serve as CCN or INPs depends on their composition, size, and number concentration, each of which depends on their source and extent of aging. Several previous studies have focused on examining the sources of Arctic aerosols, including ground-based and airborne research campaigns conducted in the Alaskan Arctic extending back to the mid-20<sup>th</sup> century (e.g., Schnell and Raatz, 1984; Barrie, 1986; Delene and Ogren, 2002; Quinn et al., 2002; Verlinde et al., 2007; Quinn et al., 2009; Brock et al., 2011; McFarquhar et al., 2011). To better understand aerosol properties in this environment, two atmospheric research facilities have been established on the North Slope of Alaska that encompass routine, aerosol measurements—including, but not limited to, aerosol optical, physical, and chemical properties, and CCN concentrations.

Utqiagvik, Alaska (formally Barrow) features an observatory established by the National Oceanic and Atmospheric Administration (NOAA) Earth System Research Laboratory's (ESRL) Global Monitoring Division (GMD) in 1976. Previous work at this facility involves different combinations of the long-term, ground-based aerosol optical, physical, and ~~for~~ chemical property measurements to evaluate the annual cycle of aerosol sources at Utqiagvik (e.g., Polissar et al., 2001; Delene and Ogren, 2002; Quinn et al., 2002; Quinn et al., 2009). For example, Quinn et al. (2002; 2009) used aerosol number concentrations, optical properties, and chemistry measurements to conclude that the winter and spring are impacted by aerosol transported from mid-latitudes, while summer and fall aerosols contain contributions from local biological activity, sea salt, and residual (i.e., unanalysed) aerosol mass that may represent mineral or organic species. More recently, Kolesar et al. (2017) used a 6-year time series of particle size distributions to ~~discover~~ conclude that particle growth events occurring at Utqiagvik resulted from gas-phase emissions originating from the oil ~~and gas~~ fields of the Prudhoe Bay area, approximately 300 kilometres east of Utqiagvik. Gunch et al. (2017) found submicron (i.e., < 1 µm in diameter) combustion-derived particles were transported from the Prudhoe Bay oil field to Utqiagvik 10% of the time during ~~their~~ Aug – Sep 2015 ~~study~~. In addition to Utqiagvik, another Northern Alaskan facility was recently established by the U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM; since 2013) program at Oliktok Point (<https://dis.arm.gov/sites/amf/oli/>). This site ~~exists~~ is located in the northwest region of oil extraction activities in Prudhoe Bay, making it an ideal location to determine the potential impacts of emissions from such activities on the relatively pristine Arctic atmosphere. Aerosol optical, physical, and chemical property measurements were implemented during the summer of 2016. ~~This site provides the ability to determine the potential impacts of oil extraction emissions on the relatively pristine Arctic atmosphere~~. Overall, the North Slope provides a unique opportunity to investigate aerosols and their impacts from the clean Arctic background, long-range transported ~~aerosol~~ from lower latitudes, and regional oil extraction activities.

While previous studies have provided significant insights into aerosol properties in Northern Alaska, one crucial deficiency is that most of them comprise *only* ground-based observations of aerosol (e.g., Barrie, 1986; Polissar et al., 2001; Delene and Ogren, 2002; Quinn et al., 2002; Quinn et al., 2009; Gunch et al., 2017; Kolesar et al., 2017). The Arctic

atmosphere can be highly stratified (Persson et al., 2002; Graversen et al., 2008), ~~thus disconnecting aerosol measured at the surface from those aloft thus hindering vertical transport of aerosols from their surface sources. To bridge this gap,~~ Accordingly, numerous airborne campaigns have focused on evaluating sources of mid- to upper-tropospheric aerosol and aerosol-cloud interactions. For example, during the ~~March~~ 1983 NOAA Arctic Gas and Aerosol Sampling Program (AGASP) flights over Alaska, ~~the highest aerosol concentrations were found from the mid- to upper-troposphere aerosol number concentrations were found to vary substantially over the vertical extent of the flight region~~ (Schnell and Raatz, 1984). Several airborne campaigns—including Aerosol, Radiation, and Cloud Processes affecting Arctic Climate (ARCPAC), Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS-A), and Indirect and Semi-direct Aerosol Campaign (ISDAC)—took place in the region during ~~April~~ 2008 to characterize tropospheric pollution and its ~~sources during source contributions to~~ the Arctic haze season during the International Polar Year. ~~These flights aimed to improve our knowledge on how changes in aerosol composition and concentration directly impact atmospheric radiation, while others extended beyond this to examine how aerosols influence cloud properties and their associated radiative forcing~~ (Brock et al., 2011; McFarquhar et al., 2011; Bian et al., 2013). These studies presented valuable information on the vertical structure of Arctic aerosol, and the relative contributions from Arctic background, fossil fuels, and biomass burning sources, but are limited to ~~April or March~~ only. Airborne measurements available from the Mixed-Phase Arctic Cloud Experiment (M-PACE), which took place from late ~~September~~ to late ~~October~~ 2008, ~~are were~~ predominantly focused on clouds and, with respect to aerosols, only encompassed aerosol size distribution measurements and INP concentrations (Verlinde et al., 2007; Prenni et al., 2009; Jackson et al., 2012). To our knowledge, only one study reports airborne *in situ* aerosol measurements over the Alaskan Arctic during the summer: NASA's ~~1988 Arctic Boundary Layer Experiment (ABLE 3A) during the summer of 1988~~ (Gregory et al., 1992). However, this study was limited to flights between Fairbanks and Utqiagvik, and to aerosol size distributions from 0.12 to 8 µm in diameter—no other aerosol measurements were obtained.

~~In the context of warming temperatures, emissions from oil extraction, added shipping routes due to a reduction in sea ice extent, and wildfires are expected to increase in sub-Arctic boreal regions (Randerson et al., 2006; Gautier et al., 2009; Harsem et al., 2011; Peters et al., 2011; de Groot et al., 2013; Roiger et al., 2015). Thus, regional fossil fuel and biomass burning combustion sources will further contribute to the aerosol population may serve as an increasingly crucial source of aerosol in the future.~~ With increasing prospects for (1) oil extraction, (2) added shipping routes due to a reduction in sea ice extent, and (3) warming temperatures inducing wildfires in sub-Arctic boreal regions, ~~regional fossil fuel and biomass burning combustion sources will further contribute to the aerosol population~~ (Randerson et al., 2006; Gautier et al., 2009; Harsem et al., 2011; Peters et al., 2011; de Groot et al., 2013; Roiger et al., 2015). However, local pollution and other high-latitude Eurasian resource extraction sources and their resulting impacts on clouds and radiation are poorly quantified (Arnold et al., 2016). Hobbs and Rango (1998) documented increased cloud droplet number concentrations in air masses originating around Prudhoe Bay through airborne measurements over the Beaufort Sea. In a companion paper by Maahn ~~and colleagues et al.~~ (2017), local emissions from Prudhoe Bay ~~are were~~ shown to impact cloud drop size in comparison with more pristine clouds over Utqiagvik. Such studies support the idea that emissions from oil extraction activities in this region have air quality and climatic

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implications and are important to assess. Additionally, Stohl et al. (2013) reported that gas flaring emissions are underestimated in the Arctic, further justifying the need to evaluate emissions from these sources.

In addition to industrial sources, it is recognized that Alaskan boreal fires periodically ~~impact~~ influence the aerosol population over the North Slope. Eck et al. (2009) reported high summer time (August) fire counts, impacting aerosol optical depths (AODs) over Utqiagvik. Stohl et al. (2006) reported similar findings using ground-based absorption and ~~CO<sub>2</sub>~~ (i.e., a tracer for biomass burning) measurements at Utqiagvik. Both studies concluded that individual smoke transport events resulted in short episodes of higher AOD and absorption values than typical springtime Arctic haze. Regardless of their episodic behaviour, summertime sub-Arctic boreal fires can produce substantial quantities of aerosol that can reside in the troposphere for 1 – 2 weeks (Stohl et al., 2013). The Arctic summertime atmosphere is historically less polluted as compared to the rest of the year (Quinn et al., 2002; Leitch et al., 2013; Heintzenberg et al., 2015), thus it is critical to assess the impacts of potentially important local sources of summertime aerosol on Arctic radiation and cloud microphysical processes. Here, we present ~~airborne~~ aerosol and trace gas observations from ~~the~~ ARM's Fifth Airborne Carbon Measurements (ACME-V) ACME-V field campaign ~~during the summer of 2015~~ to evaluate local sources during the summer of 2015 in the Alaskan Arctic.

## 15 2 Methods

### 2.1 Study location and dates

ACME-V flights were conducted over the North Slope of Alaska between five waypoints, including Oliktok Point (70.51°N, 149.86°W), Utqiagvik (71.29°N, 156.79°W), Atkasuk (70.48°N, 157.42°W), Ivotuk (68.49°N, 155.75°W), and Toolik Lake (68.63° N, 149.61° W) (Figure 1), all north of the Brooks Mountain Range. The campaign involved 38 research flights from 4 Jun to 9 Sep 2015, generally flying every 2 – 3 days (Table 1). The DOE ARM Gulfstream-1 (G-1; part of the ARM Aerial Facility) aircraft typically flew below 1000 m above ground level (m AGL) between the waypoints, while ~~spiralling~~ spiralling up to 8,000 m AGL above each waypoint. Data altitudes were converted to meters above mean sea level (m AMSL) for a more direct comparison between measurement locations. Flight tracks varied in the number and order of waypoints that were overflown.

### 25 2.2 Aircraft aerosol and trace gas payload

The G-1 was equipped with a suite of atmospheric state, cloud, aerosol, and trace gas instruments, (see <https://www.arm.gov/research/campaigns/aaf2014armacmev> for a complete list of instrumentation and available data) (Biraud et al., 2016), though in the current study we only focus on the aerosol, CO, and CO<sub>2</sub> and CO<sub>2</sub> measurements. Total number concentrations (CN) of aerosol particles 3 nm – 3 μm and 10 nm – 3 μm in diameter ( $D_g$ ) were measured using two Condensation Particle Counters (CPC, TSI, Inc. models 3025 and 3010, respectively). The CPC 3025 and 3010 have a 50% counting efficiency of 3-nm and 10-nm particles, respectively. Aerosol size distributions were measured using three different

instruments, including an Ultra-High Sensitivity Aerosol Sizer (UHSAS, Droplet Measurement Technologies, Inc.), a Passive Cavity Aerosol Spectrometer (PCASP, Droplet Measurement Technologies, Inc. model SPP-200), and an Optical Particle Counter (OPC, Climeo model C1-3100) in combination with a Multi Channel ~~Analyzer~~ ~~Analyser~~ (Ortec model Easy-MCA-8k), which measured ~~particle~~ particle optical diameters in the ranges of 0.06 – 1  $\mu\text{m}$ , 0.1 – 3  $\mu\text{m}$ , and 0.8 – 15  $\mu\text{m}$ , respectively. The PCASP was operated with an anti-ice heater, thus the particles measured are predominantly dry (Kassianov et al., 2015). The UHSAS experienced instrumental complications during most of the campaign, thus is not used for the current study to alleviate any limitations and skewness from operation dates. Total aerosol light scattering and absorption coefficients ( $\text{Mm}^{-1}$ ) were measured using a 3-wavelength (450 nm, 550 nm, and 700 nm) nephelometer (TSI, Inc. model 3563) and 3-wavelength (464 nm, 528 nm, and 648 nm) Particle Soot Absorption Photometer (PSAP, Radiance Research, Inc.), respectively. Refractory black carbon (rBC) concentrations were measured with the Single Particle Soot Photometer (SP2, Droplet Measurement Technologies, Inc.). ~~The SP2, which~~ measures individual rBC particles through laser-induced incandescence, making it selective for rBC (Sedlacek, 2016). Quality Assurance/Quality Control (QA/QC) checks of the SP2 data ensure that other potentially refractive particles such as mineral dust are not counted as rBC particles. ~~Carbon monoxide (CO)~~ concentrations were measured with a CO/N<sub>2</sub>O/H<sub>2</sub>O instrument (Los Gatos Integrated Cavity Output Spectroscopy, ~~instrument~~ model 907-0015-0001) and is used as a tracer for combustion sources, including both biomass burning and fossil fuel (Andreae and Merlet, 2001; Brock et al., 2011; Liu et al., 2014). ~~Carbon dioxide (CO<sub>2</sub>)~~ concentrations were measured by Cavity Ring Down Spectroscopy (Picarro model G2301) and together with the CO measurements were used to calculate ~~Modified-modified combustion-combustion efficiency-efficiency~~ (MCE) (Liu et al., 2014; Biraud and Reichl, 2016). ~~The MCE, MCE is defined as  $\Delta\text{CO}_2 / (\Delta\text{CO}_2 + \Delta\text{CO})$  where  $\Delta\text{CO}_2$  and  $\Delta\text{CO}$  indicate the increase from background  $\text{CO}_2$  and CO concentrations, respectively (Liu et al., 2014), and was calculated for data in which fires impacted the measurements (Liu et al., 2014). Background  $\text{CO}_2$  and CO concentrations of 383 and 0.054 ppmv, respectively, were defined from the current measurements. These values were derived from correlations of rBC mass versus  $\text{CO}_2$  and CO, and finding the minimum value of  $\text{CO}_2$  and CO on the rBC axis.~~

All data were collected at 1-second intervals and are publicly-available on the ARM data archive (<http://www.archive.arm.gov/armlogin/login.jsp>). ~~Unless noted, all data presented are 1-second.~~ Data quality was verified through quality assurance and data quality checks by DOE ARM. CPC, PCASP, OPC, and rBC data flagged for being in-cloud were excluded from the current analysis, since the isokinetic inlet used on the G-1 during the study does not discern between interstitial aerosols and cloud particles, and cloud and aerosol size ranges can potentially overlap. Data periods impacted by liquid and ice clouds were defined as those having  $1 \times 10^7 \text{ m}^{-3}$  droplets and  $100 \text{ m}^{-3}$  ice particles larger than 400  $\mu\text{m}$ , respectively (Lance et al., 2011). When a cloud was found (defined as at least 10 seconds of data where the cloud threshold is exceeded), aerosol observations 3 seconds before and 3 seconds after the cloud ~~are-were~~ discarded as well to avoid contamination of the aerosol probes with cloud particles (Maahn et al., 2017). CO data were used in- and out-of-cloud since there are not potential artefact issues. To minimize the influence of localized contamination from take-off and landing at the Deadhorse airport (19.5 m AMSL), all data below 20 m AMSL and within 3 km of the airport were removed. All data are presented in Coordinated Universal Time (UTC).

### 2.3 Supporting satellite data

The source of aerosols from the central Alaskan fires was determined using imagery from the Moderate Resolution Imaging Spectroradiometer (MODIS) on board the Terra satellite. MODIS Aqua looked similar, thus only Terra observations are discussed herein. ~~Aerosol optical depth (AOD)~~ data from MODIS were acquired from the Giovanni data server (<http://giovanni.gsfc.nasa.gov/giovanni/>) for daily dark target deep blue combined mean AOD at 550 nm and a 1° spatial resolution using a domain of 139°W to 169°W and 57°N to 72°N (MOD08\_D3\_6) (Ackerman et al., 1998). Fire and surface thermal anomaly data were also acquired from MODIS using brightness temperature measurements in the 4- $\mu\text{m}$  and 11- $\mu\text{m}$  channels (Giglio, 2010). The fire detection strategy is based on absolute detection of a fire (when the fire strength is sufficient to detect), and on detection relative to its background (to account for variability of the surface temperature and reflection by sunlight) (Giglio et al., 2003). The algorithms include masking of clouds, bright surfaces, glint, and other potential false alarms (Giglio et al., 2003). Swaths from overpasses over the state of Alaska were used to determine the daily locations of fires. The Alaskan fire season was relatively active (i.e., had the highest density of fires) from mid-Jun to mid-Jul 2015. Detected thermal anomalies or fires for the 4 Jun – 31 Aug period are used (thermal anomaly data ~~are~~ were not available from 1 – 9 Sep).

### 2.4 Aerosol dispersion modelling

Aerosol dispersion simulations were executed to demonstrate aerosol transport using version 4 of the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT4) model (Draxler, 1999; Stein et al., 2015) and 1° data from the NOAA/[National Centers for Environmental Prediction \(NCEP\)](#) Global Data Assimilation System (GDAS) (Kalnay et al., 1996). Simulation parameterization details are presented by Maahn et al. (2017), but are reiterated briefly here. ~~The HYSPLIT dispersion model simulates emission and subsequent transport of aerosols in forward mode from a point source, enabling qualitative assessment of the spatial extent of dispersion from a source of interest. The HYSPLIT dispersion model simulates emission and subsequent transport of aerosols in forward mode from a point source, enabling qualitative assessment of the spatial extent of emission dispersion from a source of interest.~~ Aerosol mass concentrations were evaluated qualitatively from one central Prudhoe Bay location and from five locations within the active fire region in central Alaska at 100-m intervals from 0 to 5,000 m AGL for 72 hours, a 6-hour release of particles at a default emission rate of one arbitrary mass unit for the study time period (1 Jun – 30 Sep 2015). ~~The five locations were chosen based on equal spacing within the highest density of fires determined from the satellite analyses for the entire study time period. The five locations were chosen based on equal spacing within the highest density of fires determined from the satellite analyses from the entire study time period of thermal anomaly data.~~ Other ~~set~~ input parameters include particle density ( $6 \text{ g cm}^{-3}$ ), shape factor (1.0), particle diameter ( $0.2 \mu\text{m}$ ) (Eck et al., 1999; Rissler et al., 2006; Brock et al., 2011; Sakamoto et al., 2015), dry deposition velocity ( $1 \times 10^{-4} \text{ m s}^{-1}$ ) (Warneck, 1999), in-cloud scavenging defined as a ratio of the pollutant in rain ( $\text{g L}^{-1}$ ) measured at the ground to that in air ( $\text{g L}^{-1}$  of air in the cloud layer) ( $4 \times 10^4$ ), and below-cloud scavenging ( $5 \times 10^{-6} \text{ s}^{-1}$ ). Radioactive decay and pollutant resuspension were set to the default values of zero days and  $0 \text{ m}^{-1}$ , respectively. ~~The results of the dispersion simulations provide arbitrary mass concentrations of~~

~~particles within the model grid after 72 hours of release from the five fire source locations. The results of the dispersion simulations provide arbitrary mass concentrations of total particles within the model grid after 72 hours of release from the five fire source locations.~~

Based on a combination of the HYSPLIT results, thresholds of ~~various~~ parameters from previous work, and visual assessment of the proximity to ~~known-potential~~ sources and vertical profiles, each 1-second data point was characterized as originating from the Prudhoe Bay oil extraction activities ~~(called Prudhoe Bay herein for brevity), wildfires, or neither. Remaining data were classified as~~ long-range transport, pristine ~~conditions~~, or background (see ~~Table 2 for classification parameters and thresholds used~~). Visual assessments are discussed in more detail ~~in~~ throughout section 3. Briefly, Prudhoe Bay emissions were characterized by visual assessment of high concentrations of particles with diameters between 3 and 10 nm within 50 km of the Deadhorse Airport. ~~Furthermore,~~ HYSPLIT dispersion results from Deadhorse were used to ~~qualitatively characterize~~ ~~determine~~ the spatial extent of Prudhoe Bay emissions. The boundary layer emissions from Prudhoe Bay were restricted to 500 m AMSL ~~based on Maahn et al.~~ (Maahn et al., 2017) and based on changes in vertical profiles of the concentrations of 3 – 10 nm particles. Fire data were characterized by using thresholds from Maahn et al. (2017). ~~The highest density of fires burned in central Alaska as detected by satellite, thus w~~We additionally constrained the data to ~~evaluated~~ fires south of 69°N to focus closer to the source region ~~(i.e., near the highest density of fires detected by MODIS that overlapped with the flight region)~~. It is important to note that 17 of the 1-second data points fell under both Prudhoe Bay and fires classifications, but the remaining 496430 data points were characterized under one source. Long-range transport ~~data were~~ was determined by data that were not characterized as Prudhoe Bay or fires, but had concentrations of 0.1 – 3  $\mu\text{m}$  diameter particles  $\geq 400 \text{ cm}^{-1}$  above 300 m AMSL based on visual assessment of the vertical profiles. We note that ~~a variety of sources could contribute to this source classification could be from a variety of sources~~, but are likely long-range transported due to the relative concentrations of particles, altitude, and dearth of ~~the other prolific-dominant~~ regional sources of aerosol. ~~Pristine conditions were characterized by remaining~~ data that were not classified as from Prudhoe Bay, fires, or long-range transport but had low 0.1 – 3  $\mu\text{m}$  diameter particle concentrations. Background conditions were characterized as any data ~~points~~ remaining after the aforementioned sources were determined.

### 3 Results and discussion

#### 3.1 Prudhoe Bay is a persistent local source of small particles in the boundary layer

Figures 2 and 3 show the spatial and vertical variability of select aerosol quantities from ACME-V, respectively. ~~Measurements from ACME-V below 500 m show~~ A clear source of aerosol ~~originated from Prudhoe Bay (Figure 2a, b) as suggested by the HYSPLIT dispersion model (Figure 2a) and in situ CN measurements (Figure 2b and c), which was predominantly composed of particles with diameters ( $D_p$ ) between 3 and 10 nm (CPC<sub>400</sub>; calculated from subtracting the CPC 3010 from CPC 3025 number concentrations).~~ The highest number concentrations of 3 – 10 nm particles (up to  $10^4$  particles  $\text{cm}^{-3}$ ; ~~calculated from subtracting the CPC 3010 from CPC 3025 number concentrations~~) and 10 nm – 3  $\mu\text{m}$  sized particles were observed within 50



km of the Deadhorse Airport (i.e., used here as a proxy for Prudhoe Bay; see Figure 1) in a layer from 100 to 300 m AMSL and during almost all flights (Figure 3). Particles within this the 3 – 10 nm size range are associated with nucleated aerosol (i.e., spontaneous *in situ* aerosol formation from precursor gases) (Colbeck and Lazaridis, 2014). These high number concentrations of small particles are likely formed from gas-to-particle partitioning of reactive gases from flaring and venting along the North Slope. Flaring and venting of gas, which is prominent near the surface in the Arctic near oil and gas facilities (Jaffe et al., 1995; Johnson and Coderre, 2011), could contribute the vapours—such as secondary products from secondary products from ozone, SO<sub>x</sub> and various aromatic hydrocarbons—that induce nucleation of new particles (Wilson and McMurry, 1981; Parungo et al., 1992; Kulmala et al., 2004; Laaksonen et al., 2008; Ismail and Umukoro, 2012; Emam, 2015). Additionally, a sharp decrease in the concentrations of 3 – 10 nm particles and, to a lesser extent, 10 nm – 3 μm particles was observed above 500 m AMSL (Figure 3a and b, respectively). The highest number concentrations of 3 – 10 nm particles were predominantly observed below 500 – 600 m, indicating: 1) transition of particles via growth into the accumulation mode (0.1 – 2.5 μm) as the plume evolves and disperses vertically (Colbeck and Lazaridis, 2014) or 2) dynamical restriction of these particles in the atmospheric boundary layer. When examining the ratio of the number of 3 – 10 nm particles the CPC<sub>diff</sub> (i.e., nucleation mode) to the number of PCASP 0.1 – 3 μm particles number concentrations (i.e., accumulation mode) with altitude (Figure 3c Figure 4), the ratio is was > 1 (i.e., nucleation mode particles were dominant) for 74% of the time and < 1 (i.e., accumulation mode particles were dominant) for 26% of the time below 500 m AMSL. More specifically, the ratio was > 1 for 86% of the time when only considering data points classified as Prudhoe Bay. Furthermore, this ratio decreased overall with altitude and with decreasing latitude (not shown), indicating the nucleation mode particles were formed at the lowest altitudes closest to their source, while accumulation mode particles originated from growth of the nucleation mode aerosol or a different source (see section 3.3).

Black carbon (as rBC) also had relatively high mass concentrations of rBC (up to 464 ng kg<sup>-1</sup>) were also observed in the Prudhoe Bay area below 500 m AMSL (Figures 2 Figure 2d and 3d), which likely originated from local combustion sources such as on- and off-road vehicles, facility heating, and to some extent, flaring (Bond et al., 2013; Stohl et al., 2013). Particles measured immediately near oil combustion sources—including rBC—normally have a size mode around 100 – 130 nm (Parungo et al., 1992; Chang et al., 2004). The smallest average mass equivalent modal size of the rBC from ACME V were 115 nm and 110 nm for all data and those closest to Deadhorse (below 500 m AMSL nearest to and < 50 km from Deadhorse), respectively, indicating: 1) particles were “fresher” (i.e., less coated or “aged” from heterogeneous reactions) closest to the Prudhoe Bay source (Maahn et al., 2017) and 2) modal sizes are analogous to what might be expected from oil combustion sources after slight aging due to farther proximity from sources (i.e., not direct measurement from stacks). However, larger sizes and higher mass concentrations (particularly above 500 m AMSL) of rBC were observed further south of Deadhorse (Figure 2d) and were likely influenced by biomass burning emissions as discussed in section 3.2. However, the larger rBC sizes further south of Deadhorse are likely influenced by biomass burning emissions as discussed in section 3.2. The localized nature of the smaller-sized CN and rBC to the Prudhoe Bay reflect the

Aerosols originating from the oil extraction activities were localized to the Prudhoe Bay area as suggested by the HYSPLIT dispersion model (Figure 2d). This reflects the source and physical removal processes occurring with the dominant-sized particles occurring in this region. Both nucleation and Aitken mode (i.e., 10 – 100 nm) aerosols have lifetimes on the order of minutes to hours and thus have typical travel distances of 1 – 10s of km (Wilson and Suh, 1997), corroborating our findings of such aerosols within 50 km of Deadhorse Airport. Such distances corroborate our finding that nucleation mode aerosol and rBC are restricted to < 50 km from Deadhorse below 500 m AMSL. Our results and those presented by Maahn and colleagues (2017) demonstrate an increase in aerosol size with distance from Prudhoe Bay, indicating transition to the accumulation mode.

Our results demonstrate that Prudhoe Bay is a strong and persistent source of nucleated aerosol and primary combustion aerosol, however, the high mass and number concentrations of these aerosols are restricted to the boundary layer and tend to remain localized to the Prudhoe Bay area. These aerosols may not have strong direct effects on the regional atmospheric radiation budget due to their inherently small size and low concentrations of larger accumulation mode particles (Friedlander, 2000). This is supported by the fact that no noticeable spatial patterns in absorption and scattering were observed as a function of distance from Deadhorse Airport (not shown). However, as these particles age through via atmospheric processing from co-emitted gases such as SO<sub>x</sub>/NO<sub>x</sub> and grow larger into the accumulation mode, it is possible they could have an impact after sufficient atmospheric residence time downwind. We do not have the compositional data necessary to determine the mixing state or extent of aging of these nucleation mode particles into the accumulation mode. Additionally, modelling studies have suggested that black carbon BC aerosols from Prudhoe Bay oil extraction have a positive net radiative forcing, particularly in the summer due to greater absorption of solar radiation (Ødemark et al., 2012). With regard to indirect effects, Maahn and colleagues et al. (2017) demonstrated the importance of Prudhoe Bay industrial aerosol in modulation of cloud properties over the North Slope. Further, Leitch and colleagues et al. (2016) and Burkhardt and colleagues et al. (2017) recently published observations of CCN diameters down to 20 nm in the Canadian Arctic; contrary to the conventional wisdom that 100 nm is the threshold relevant for CCN. However, these Canadian Arctic aerosols were likely compositionally different due to their marine origin, and thus could vary in hygroscopicity as compared to oil extraction emissions.

### 3.2 Regional fires impact air composition over much of Central and Northern Alaska

Another dominant aerosol source observed during the ACME-V field campaign was the central Alaskan wildfires. The summer 2015 season was particularly active, leading to the second largest number of acres burned in Alaska since records began in 1940 (Partain Jr. et al., 2016). The highest density of fires detected from satellite existed from mid-Jun to mid-Jul (Figure 4). These fires produced dense plumes of aerosol that propagated over much of the North Slope as evidenced by the high values of AOD originating from the central Alaska wildfires. Flights were impacted by the high AOD regions from late-Jun until end of Jul. The G-1 flew directly through the wildfire plumes 25 Jun – 1 Jul near Toolik Lake (Figure 4c), 9 – 15 Jul over most of the flight track (Figure 4e), and 16 – 22 Jul near Utqiagvik and Oliktok Point (Figure 4f).

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The HYSPLIT dispersion simulations from the five fire source points (Figure 4, first row) indicate larger particle mass concentrations spread over the flight region, particularly at the southern portion of the domain (Figure 5a). Analogously, *in situ* measurements show clear influence of Alaskan boreal fires (Figure 5b – d): the number of particles from 0.1 to 3  $\mu\text{m}$ , rBC mass, and CO were high in concentration, particularly at the southern portion of the flight track close to the Brooks Range.

Wildfires emit large quantities of primary organic aerosol (POA) and can generate secondary organic aerosol (SOA) that can develop coatings through aging while transported over long distances (Andreae and Merlet, 2001; Collier et al., 2016; Creamean et al., 2016). Therefore, we would expect to observe an abundance of coarse and accumulation mode aerosol and a dearth of nucleation mode aerosol, since nucleation of new particles is inhibited by precursor vapours instead condensing onto pre-existing aerosol (discussed in more detail in below). The largest impacts from the fires were observed from 400 to 7000 m AMSL (Figure 6a). MCE values during measurements impacted by fires were close to 1 (Figure 6b), indicating active flaming (i.e., “fresher” fires) instead of smouldering, particularly during flights 13, 17, and 18. Combined, these data suggest fires were recent, yet emissions from the fires were ejected high into the troposphere. CO and rBC concentrations were strongly correlated ( $R^2 = 0.83$ ) and reached 0.626 ppmv and 1490 ng  $\text{kg}^{-1}$ , respectively (Figure 6c). The MCE, defined as  $\Delta\text{CO}_2 / (\Delta\text{CO}_2 + \Delta\text{CO})$  where  $\Delta\text{CO}_2$  and  $\Delta\text{CO}$  indicate the increase from background  $\text{CO}_2$  and CO concentrations, respectively, was calculated for data in which fires impacted the measurements (Liu et al., 2014). Background  $\text{CO}_2$  and CO concentrations of 383 and 0.054 ppmv, respectively, were defined from the current measurements. These values were derived from correlations of rBC mass versus  $\text{CO}_2$  and CO, and finding the minimum value of  $\text{CO}_2$  and CO on the rBC axis. MCE values during measurements impacted by fires were close to 1, indicating active flaming (i.e., “fresher” fires) instead of smouldering, particularly during flights 13, 17, and 18. CO is a poor tracer for oil extraction since it originates from combustion, thus aside from the operational vehicles in Prudhoe Bay, we would expect the boreal fires to most strongly influence CO during the campaign (Crutzen et al., 1979; Andreae and Merlet, 2001). Background CO concentrations have been measured at 0.120 ppmv using summertime surface measurements at Utqiagvik and up to 0.4 ppmv ~~in during~~ ARCPAC airborne measurements of springtime long-range-transported biomass burning plumes (Liang et al., 2004; Brock et al., 2011). Brock ~~and colleagues et al.~~ (2011) reported springtime rBC mass concentrations of up to 1000 ng  $\text{m}^{-3}$  using the SP2 instrument also used in the current study. Although the fires were an abundant source of absorbing rBC, highly scattering aerosol originated from the fires (Figure 6d), which could be explained by previous work indicating fires produce larger quantities of organic carbon and sulphate (Penner et al., 1992; Wiedinmyer et al., 2011). ~~Although the fires were an abundant source of absorbing rBC, highly scattering aerosol originated from the fires (Figure 6d), which could be explained by previous work indicating fires produce larger quantities of organic carbon and sulphate (Penner et al., 1992; Wiedinmyer et al., 2011).~~ Our observations are parallel to previous summertime observations from regional boreal fires in that such fires produce substantial quantities of aerosol (Stohl et al., 2006; Eck et al., 2009), which is likely due to the proximity of the measurements to the source.

Notably, anomalously high rBC mass and CO concentrations were measured during a few flights (F13, F17, and F18). Almost all measurements from ~~F13 the~~ (30 Jun flight) were considerably high—including CO (maximum of 0.626 ppmv), rBC (1490 ng  $\text{kg}^{-1}$ ), aerosol number concentrations (20596  $\text{cm}^{-3}$  for CPC 3010, 11021  $\text{cm}^{-3}$  for PCASP, and 30  $\text{cm}^{-3}$  for OPC),

absorption ( $61.1 \text{ Mm}^{-1}$ ), and scattering ( $978.1 \text{ Mm}^{-1}$ )—as compared to other flights impacted by the fires (Figure 7). AOD was relatively high ( $> 0.1$ ) and a fire was detected by MODIS close to Toolik Lake (see Figure 4c) on 30 Jun, which is likely why the measurements were highest when spiralling over the waypoint and then decreased as the aircraft flew low to/from adjacent waypoints. However, the plume on 30 Jun also reached higher altitudes above Oliktok Point (as supported by MODIS), indicating the biomass burning plume ascended as it propagated northward. The only exception to the considerably high nature of the aerosol ~~measurements-concentrations~~ is nucleation mode aerosol, which was only slightly elevated in concentration (maximum of  $2500 \text{ cm}^{-3}$  as compared to a maximum of  $101940 \text{ cm}^{-3}$  for the same-sized particles from Prudhoe Bay) and was not elevated over Oliktok Point, demonstrating the short lifetimes of these small-sized particles ~~(via growth into the accumulation mode) (via growth into the accumulation mode)~~ in densely-populated biomass burning plumes.

The large quantity of aerosol observed from the lowest to highest altitudes flown by the aircraft closest to the Brooks Range indicate the thickness of the biomass burning aerosol layer. These particles can have implications both on the local energy budget and on cloud formation. We observed how aerosols from the 30 Jun fire event were highly ~~scattering-absorbing~~ and ~~absorbing-scattering~~ relative to the rest of the region (Figure 7e and f, respectively). Both organic and inorganic components of aerosols from wildfires can be highly hygroscopic and serve as efficient CCN (Novakov and Corrigan, 1996; Petters et al., 2009a; Engelhart et al., 2012), while mineral dust, carbonaceous, and biological aerosols from wildfires have been shown to increase atmospheric INP concentrations (Petters et al., 2009b; McCluskey et al., 2014). Additionally, ejection of such a large quantity of aerosol and trace gases into the atmosphere can affect air quality on the North Slope and to the Arctic beyond over the course of a couple of weeks (Stohl et al., 2013).

### 3.3 Relative contributions from regional and long-range-transported sources of aerosol to North Slope

Weaker poleward advection and strong aerosol removal via wet deposition ~~enable-make~~ the Arctic ~~to be subject to less~~ ~~subject to~~ transport from mid-latitude sources during the summer as compared to the spring Arctic haze season (Polissar et al., 2001; Eckhardt et al., 2003; Garrett et al., 2010; Browse et al., 2012; Bian et al., 2013). During the summer, aerosol production from local natural sources—from terrestrial and marine microbial processes and mechanical generation of sea salt—is dominant at the ground and aloft (Gregory et al., 1992; Quinn et al., 2002; Leaitch et al., 2013; Leaitch et al., 2016; Burkart et al., 2017). Although the presence of pollutants from mid-latitudes is typically lower during the summer ~~in the Arctic~~ (Raatz and Shaw, 1984), we observed occasional episodic increases in accumulation and coarse mode aerosol measured by the PCASP and OPC at higher altitudes (Figure 8~~Figure 9~~), without the presence of Prudhoe Bay or Alaskan fire tracers of CO and rBC ~~(see -)~~. These layers were deficient in CO, rBC, and 3 – 10 nm particles, and were present during flights where biomass burning was not detected as a dominant source. Thus, we assume these events were not a result of local or regional emissions that dominated the North Slope aerosol. These diagnosed long-range transport events were only observed during flights 1, 9, 10, and 11, thus supporting the idea that poleward advection is less frequent and wet removal processes are enhanced in the summer as compared to the Arctic haze season.

Recent studies have alluded to the possibility that the Arctic summer may not be as pristine as previously thought (Stohl et al., 2013). Modelled emissions from ARCTAS-A highlight summertime boreal fires and their impact on Arctic pollution, however, these flights targeted local fire plumes and were limited to the Canadian Arctic. Further, Bian et al. (2013) state that the ARCTAS-A measurements, “cannot provide a comprehensive and representative picture of Arctic pollution in the summer”. We evaluated all ACME-V data and classified each flight as impacted by fires, Prudhoe Bay, long-range transport, or some combination of these (Figure 9a). All flights contained at least a small segment that was classified as background, but flights where only background conditions were observed are labelled as such. Due to the aircraft being based in Deadhorse, emissions from Prudhoe Bay impacted nearly every flight (31 flights; remaining 7 flights were flagged for clouds near Prudhoe Bay, thus those data were eliminated from the analysis), while regional fires impacted 22 flights, and long-range transport impacted only 4 flights. For a more statistical representation of the sources, we classified each 1-second data point as influenced from fires at all flight locations, fires from the lowest latitude flown to 69°N (i.e., a subset of all fires), Prudhoe Bay emissions strictly near Prudhoe Bay, emissions near Prudhoe Bay in the boundary layer, long-range transport, background, and pristine conditions (Figure 9b; see [Table 2](#) for classification definitions). Background may include aerosol from Prudhoe Bay or the fires after significant atmospheric residence time, but we cannot distinguish these from local natural aerosol emission or production that is traditionally observed with the measurements obtained. This plot demonstrates the episodic behaviour of the fires and the localized behaviour of Prudhoe Bay emissions. However, what we are calling “pristine” conditions had the lowest occurrence overall (only 5%), which contrasts with previous North Slope summertime aerosol studies. It is important to note that these data are dependent on the location and height of the aircraft, thus may be biased. However, it provides a general overview of the sources of aerosols in the context of the flight locations, but may not be representative of the entire North Slope at all times.

#### 4 Summary

Results from the 2015 airborne ARM ACME-V field campaign demonstrate that the summer in the Alaskan Arctic is not necessarily characterized by clean conditions. The pristine nature of the atmosphere is dependent on the influence from episodic wildfires, localized oil extraction activities, and to a lesser extent, long-range-transport. Probably the most notable observation was that Prudhoe Bay is a persistent but localized source of rBC and especially nucleated aerosol, supporting previous findings at Utqiagvik from Kolesar et al. (2017) and Gunsch et al. (2017), but demonstrating the larger influence of particle nucleation on the aerosol population in the vicinity surrounding Prudhoe Bay. Such elevated aerosol levels have been shown to alter the microphysics of clouds in this region (Maahn et al., 2017), [potentially](#) impacting their radiative forcing on the surface. Most previous measurements along the North Slope have been conducted at the ground at a single location, thus thwarting the evaluation of the spatiotemporal heterogeneity of aerosol in the entire Alaskan Arctic. Although our results reveal that these sources are not significant on a larger scale (i.e., the entire North Slope), they yield valuable information on local and regional Arctic pollution sources, which produce substantial quantities of aerosols that may be transported downstream

and beyond. Further, although our observations are limited to the location and dynamical conditions of the North Slope, they can serve as a proxy for other parts of the Arctic subject to oil exploration or boreal fires. With both fire activity and oil exploration projected to increase in a warming climate, these sources will likely continue to make significant contributions to the aerosol population of the Arctic atmosphere. The particles emitted from these sources can impact atmospheric radiative transfer through modulation of cloud microphysics and direct radiative forcing. To fully understand the impacts of these particles and their relative frequency of occurrence, additional observational, modelling, and theoretical studies are required.

### Author contributions

J. Creamean analysed and interpreted ARM ACME-V aerosol data, MODIS AOD and thermal anomaly data, and ran HYSPLIT simulations and wrote the manuscript. M. Maahn participated in ACME-V aerosol and HYSPLIT data analyses, and ~~found~~ [handled](#) oil well data. G. de Boer and A. McComiskey helped with general interpretation of the ACME-V data. A. Sedlacek and Y. Feng helped with MCE calculations and interpretation of the SP2 measurements. All co-authors contributed to the writing of or provided comments for the manuscript.

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Table 1. Flight identification numbers, start and end times in **both**-UTC, flight duration, and waypoints flown over of each G-1 research flight during ARM-ACME-V. Waypoints O, U, A, I, and T represent Oliktok Point, Utqiagvik, Atkasuk, Ivotuk, and Toolik Lake, respectively.

| Flight ID | Start(UTC)     | End (UTC)      | Duration (hh:mm:ss) | Waypoints     |
|-----------|----------------|----------------|---------------------|---------------|
| F01       | 06/04 22:31:21 | 06/05 01:59:40 | 03:28:19            | O, U          |
| F02       | 06/07 20:08:51 | 06/08 01:16:28 | 05:07:37            | O, U, A, I, T |
| F03       | 06/08 19:56:37 | 06/09 00:52:47 | 04:56:10            | O, U, A, I, T |
| F04       | 06/10 18:39:18 | 06/10 20:30:59 | 01:51:41            | O             |
| F05       | 06/12 21:58:20 | 06/13 00:11:26 | 02:13:06            | O             |
| F06       | 06/13 18:57:18 | 06/13 23:57:32 | 05:00:14            | O, U, A, I, T |
| F07       | 06/15 21:57:26 | 06/16 00:35:49 | 02:38:23            | O, U          |
| F08       | 06/17 18:59:36 | 06/18 00:11:03 | 05:11:27            | O, U, A, I, T |
| F09       | 06/20 19:00:37 | 06/21 00:04:12 | 05:03:35            | O, U, A, I, T |
| F10       | 06/22 23:17:43 | 06/23 01:19:26 | 02:01:43            | O             |
| F11       | 06/23 19:14:17 | 06/24 00:19:21 | 05:05:04            | O, U, A, I, T |
| F12       | 06/27 21:06:18 | 06/27 23:12:53 | 02:06:35            | O, U          |
| F13       | 06/30 18:59:46 | 06/30 22:30:34 | 03:30:48            | O, I, T       |
| F14       | 07/02 19:34:03 | 07/02 23:31:36 | 03:57:33            | O, U, T       |
| F15       | 07/05 18:57:14 | 07/06 00:10:57 | 05:13:43            | O, U, A, I, T |
| F16       | 07/11 20:27:14 | 07/12 00:51:07 | 04:23:53            | O, U, A, T    |
| F17       | 07/14 19:01:15 | 07/14 21:18:25 | 02:17:10            | O, T          |
| F18       | 07/16 19:58:43 | 07/17 00:38:47 | 04:40:04            | O, U, A, I, T |
| F19       | 07/18 19:54:33 | 07/19 00:19:00 | 04:24:27            | O, U, A, I, T |
| F20       | 07/21 19:24:37 | 07/22 00:30:44 | 05:06:07            | O, U, A, I, T |
| F21       | 07/22 19:29:54 | 07/23 00:14:20 | 04:44:26            | O, U, A, I, T |
| F22       | 07/27 21:34:03 | 07/28 00:07:11 | 02:33:08            | O, U          |
| F23       | 07/30 21:18:11 | 07/31 01:03:48 | 03:45:37            | O, U, A, T    |
| F24       | 08/02 18:10:47 | 08/02 21:40:37 | 03:29:50            | O, U, T       |
| F25       | 08/06 19:01:12 | 08/06 23:34:33 | 04:33:21            | O, U, A, I, T |
| F26       | 08/07 18:37:02 | 08/07 19:46:35 | 01:09:33            | O             |
| F27       | 08/08 19:42:22 | 08/08 21:52:35 | 02:10:13            | O, U          |
| F28       | 08/14 18:49:22 | 08/14 22:17:13 | 03:27:51            | O, U          |
| F29       | 08/16 19:54:12 | 08/16 23:53:40 | 03:59:28            | O             |
| F30       | 08/20 20:47:02 | 08/21 00:09:46 | 03:22:44            | O, U, A, I, T |
| F31       | 08/25 19:18:13 | 08/25 23:52:06 | 04:33:53            | O, U, A, I, T |
| F32       | 08/27 21:29:46 | 08/28 01:37:41 | 04:07:55            | O, U, A, I, T |
| F33       | 08/28 22:30:14 | 08/29 00:51:35 | 02:21:21            | O             |
| F34       | 08/30 21:49:18 | 08/30 23:39:16 | 01:49:58            | O             |
| F35       | 09/02 19:00:45 | 09/02 23:27:41 | 04:26:56            | O, A, I, T    |
| F36       | 09/04 21:24:39 | 09/05 00:47:02 | 03:22:23            | I, T          |
| F37       | 09/07 18:28:10 | 09/07 21:23:57 | 02:55:47            | O, A          |
| F38       | 09/09 18:29:19 | 09/09 22:18:46 | 03:49:27            | O, U, A, I    |

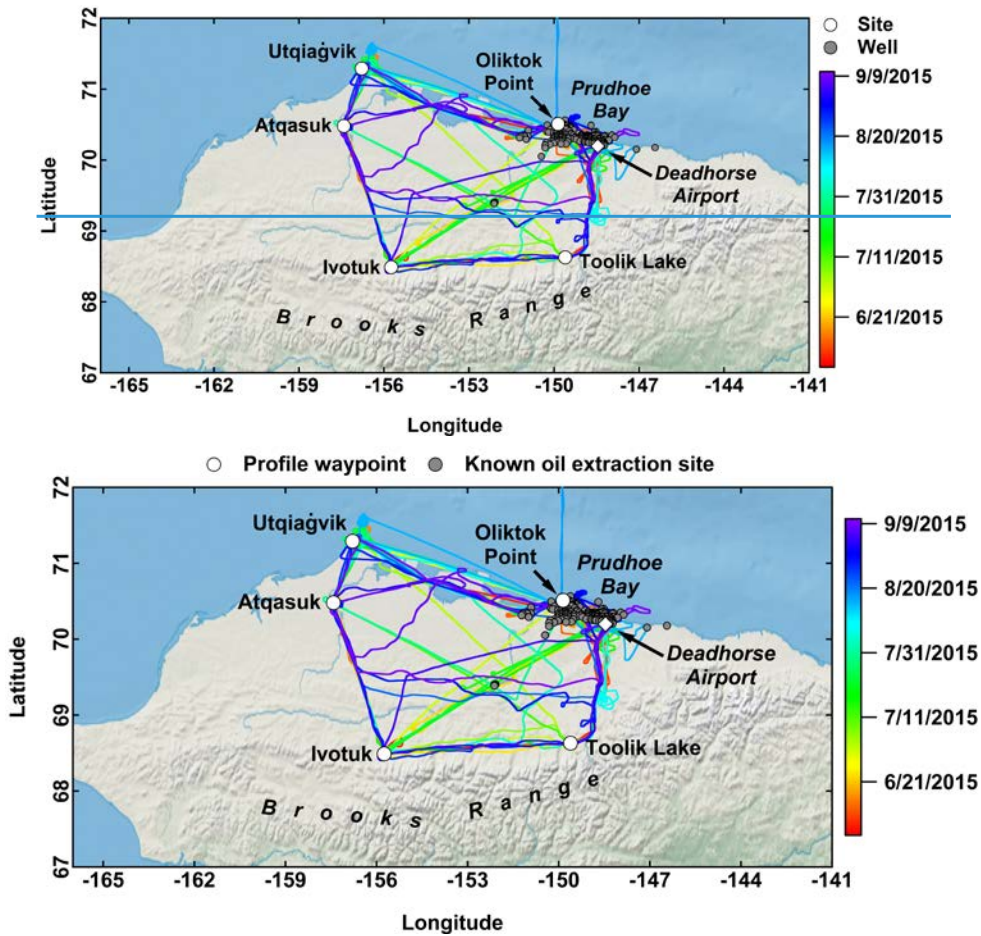
Table 2. Classification parameters and thresholds for characterization of 1-second data as being impacted by one (or more) of the sources shown in [Figure 9](#)[Figure 10b](#). “PHB”, “BL”, and “LRT” represent Prudhoe Bay, boundary layer, and long-range transport, respectively.

| Source  | Classification parameter and threshold  |
|---|---|
| <a href="#">Near Prudhoe Bay</a>                | 1) $CPC_{diff} \geq 100 \text{ cm}^{-3}$ &<br>2) Distance from Deadhorse < 50 km  |
| <a href="#">Prudhoe Bay boundary layer (BL)</a> | 1) $CPC_{diff} \geq 100 \text{ cm}^{-3}$ &<br>2) Distance from Deadhorse < 50 km &<br>3) Altitude $\leq 500 \text{ m AMSL}$                           |
| All fires                                       | 1) $rBC \geq 20 \text{ ng kg}^{-1}$ &<br>2) $CO \geq 0.1 \text{ ppmv}$  |
| Fires south                                     | 1) $rBC \geq 20 \text{ ng kg}^{-1}$ &<br>2) $CO \geq 0.1 \text{ ppmv}$ &<br>3) Latitude $\leq 69^{\circ}N$  |
| <a href="#">LRT long-range transport</a>        | 1) <u>No overlapping data with Prudhoe Bay or fires</u> &<br>+2) $PCASP \geq 4600 \text{ cm}^{-3}$ ** &<br>2)3) Altitude $\geq 300 \text{ m AMSL}$ ** |
| Pristine  | 1) <u>No overlapping data with Prudhoe Bay, fires, or LRT</u> &<br>+2) $PCASP < 4600 \text{ cm}^{-3}$ &<br>2)3) Distance from Deadhorse > 50 km       |

\*Maahn et al. (2017)

\*\*Based on threshold value selected from [Figure 9](#)[Figure 8](#)

5



5 Figure 1. Map of the North Slope of Alaska including flight tracks from the ARM-ACME-V field campaign (2015) coloured by date. Sites where the G-1 aircraft spiralled over (Oliktok Point, Utqiagvik, Atqasuk, Ivotuk are shown (profile waypoint), in addition to locations and Toolik Lake), locations of actively deployed oil wells (data obtained from <http://doa.alaska.gov/ogc/publicdb.html> in Mar 2017), the location, location of Deadhorse Airport, and approximate areas of the Brooks Mountain Range and Prudhoe Bay.

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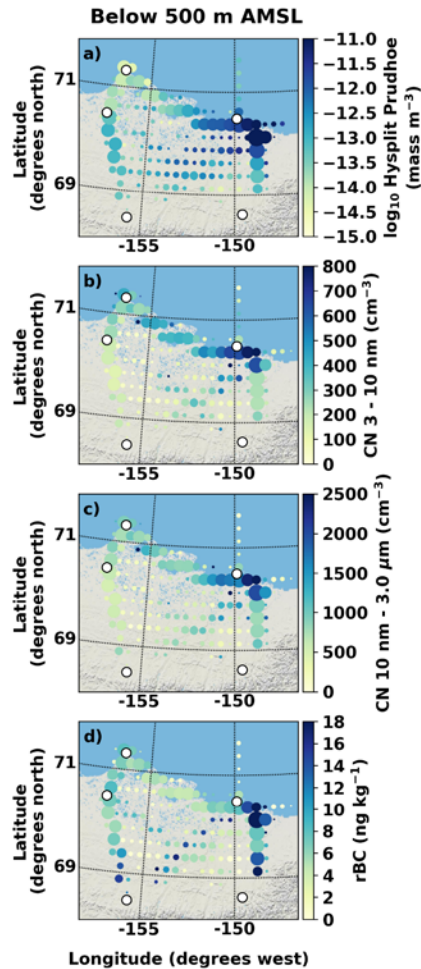
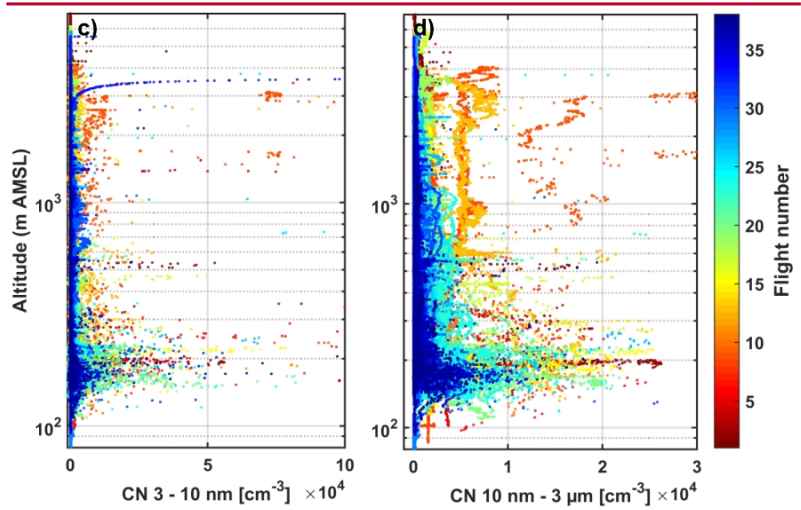
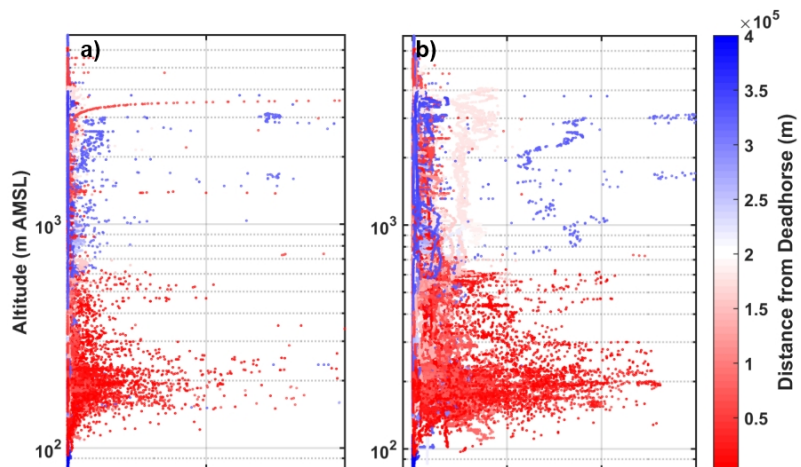


Figure 2. Maps of column average values from 20 to 500 m AMSL for a) HYSPLIT aerosol mass concentrations with from Prudhoe Bay, b) CN with  $D_p = 3 - 10 \text{ nm}$  (CPC<sub>fast</sub>), c) CN with  $D_p = 10 \text{ nm} - 3 \mu\text{m}$  (CPC-3010), and d) rBC mass (SP2) concentrations as the simulation start point for the entire ACME-V campaign from 20 to 500 m AMSL. The size of the marker equates to the number of measurements at each  $0.25^\circ$  latitude x  $0.50^\circ$  longitude grid point. The five white markers show each of the site-s where the G-1 spiralled overwaypoints.



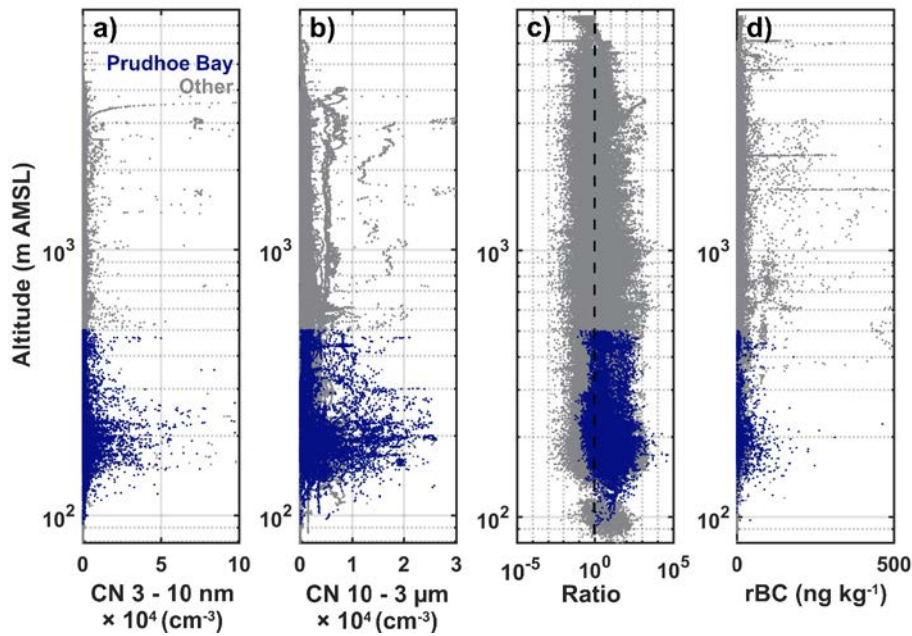


Figure 3. Vertical profiles of CN for particle-number concentrations for a) CN with  $D_p = 3 - 10$  nm (CPC<sub>air</sub>) and, b)  $D_p = 10 - 3$   $\mu\text{m}$ , c) the ratio of CN with  $D_p = 3 - 10$  nm to CN with  $D_p = 0.1 - 3$   $\mu\text{m}$ , and d) rBC (CPC 3010) coloured by distance from Prudhoe Bay (a and b, respectively) and by flight number (c and d, respectively). The dashed line in c) indicates a ratio of 1. Note that the xy scale for d) is zoomed in to show the increase below 200 m AMSL for rBC (i.e., does not show all rBC data). Data classified as originating from Prudhoe Bay is coloured blue.



-All data shown are 1-second.

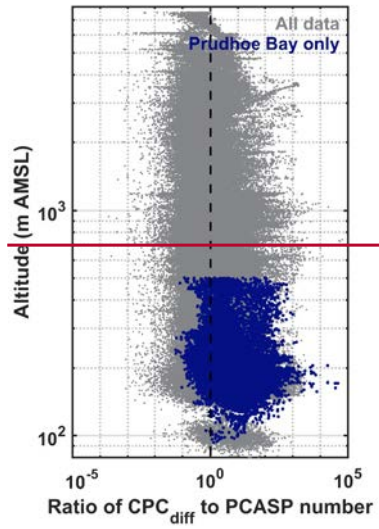


Figure 4. Ratio of the number concentrations of particles 3–10 nm in diameter  $CPC_{diff}$  to PCASP number concentrations within 50 km of Deadhorse Airport. Blue indicates a ratio  $< 1$  (i.e.,  $PCASP > CPC_{diff}$ ) and red indicates a ratio  $> 1$  (i.e.,  $PCASP < CPC_{diff}$ ).

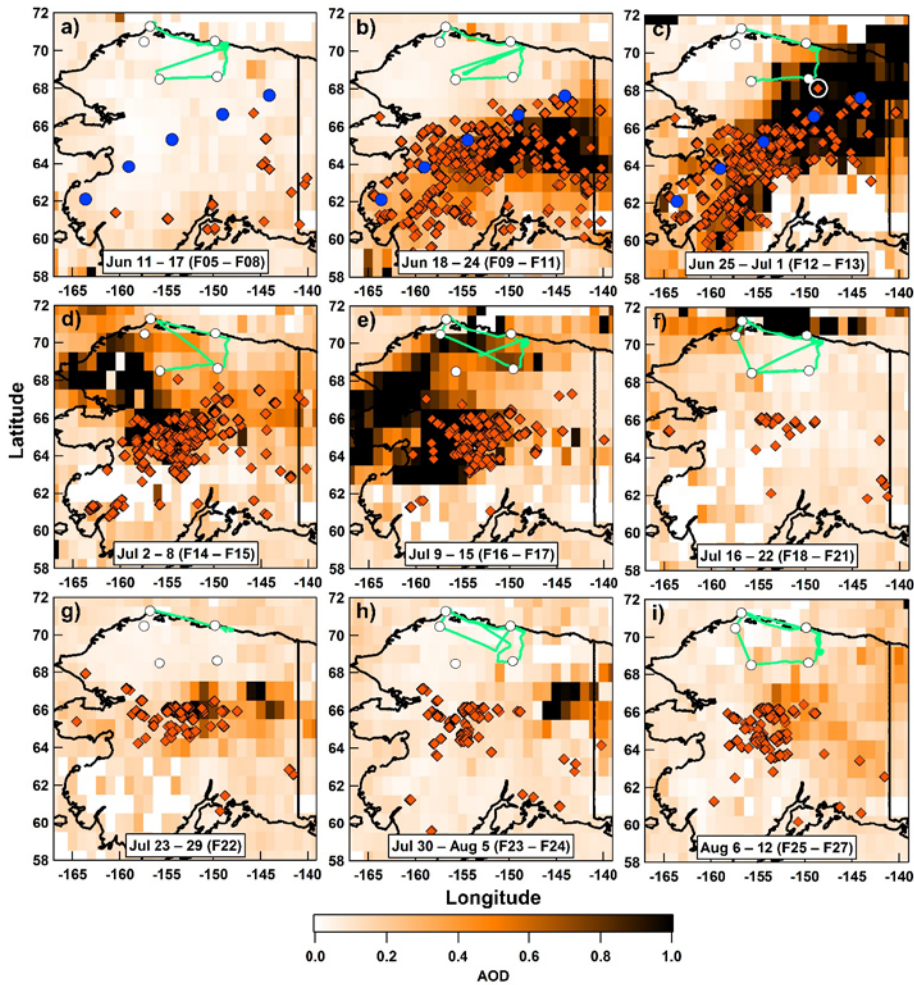


Figure 4. Maps of AOD and fires (i.e., thermal anomalies; orange diamonds) detected by MODIS for 11 Jun – 12 Aug time period, showing the transition from few fires to the highest density of fires and back. Flight tracks (green lines) and site locations (white circles) are also shown during each corresponding time period. The ~~black-blue~~ markers in the top left panel signify HYSPLIT fire start point locations. The fire location in the white circle denotes the fire closest to Toolik Lake on 30 Jun (~~top-right-panel~~panel c).

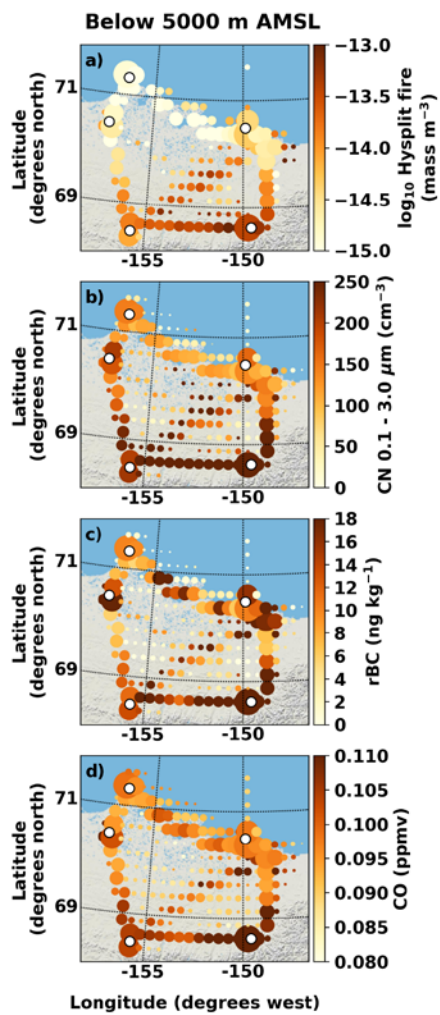
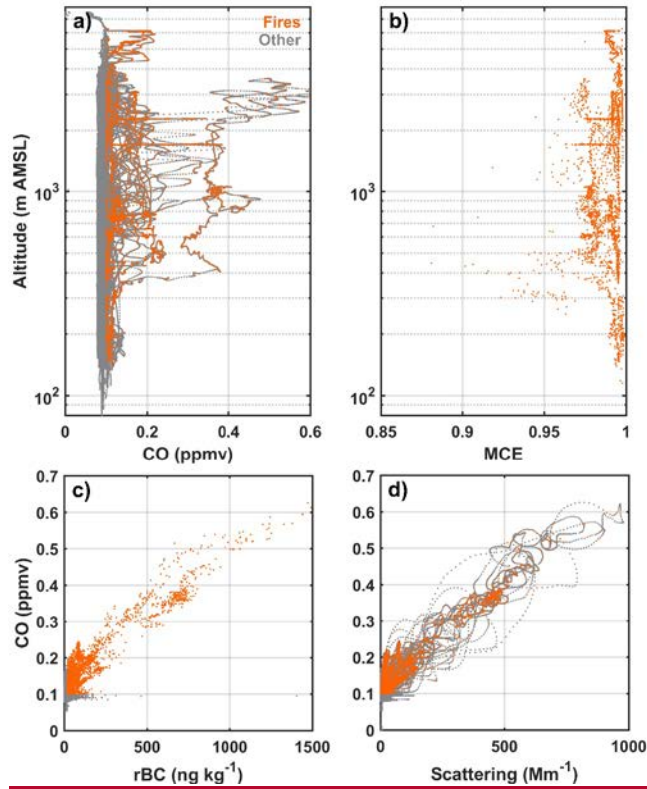


Figure 5. Same as Figure 2, but column averaged from 20 to 5,000 m AMSL and for a) HYSPLIT aerosol mass concentrations with the five fire locations as the simulation start points, b) CN with  $D_p = 0.1 - 3 \mu\text{m}$ , c) rBC mass, and d) CO.



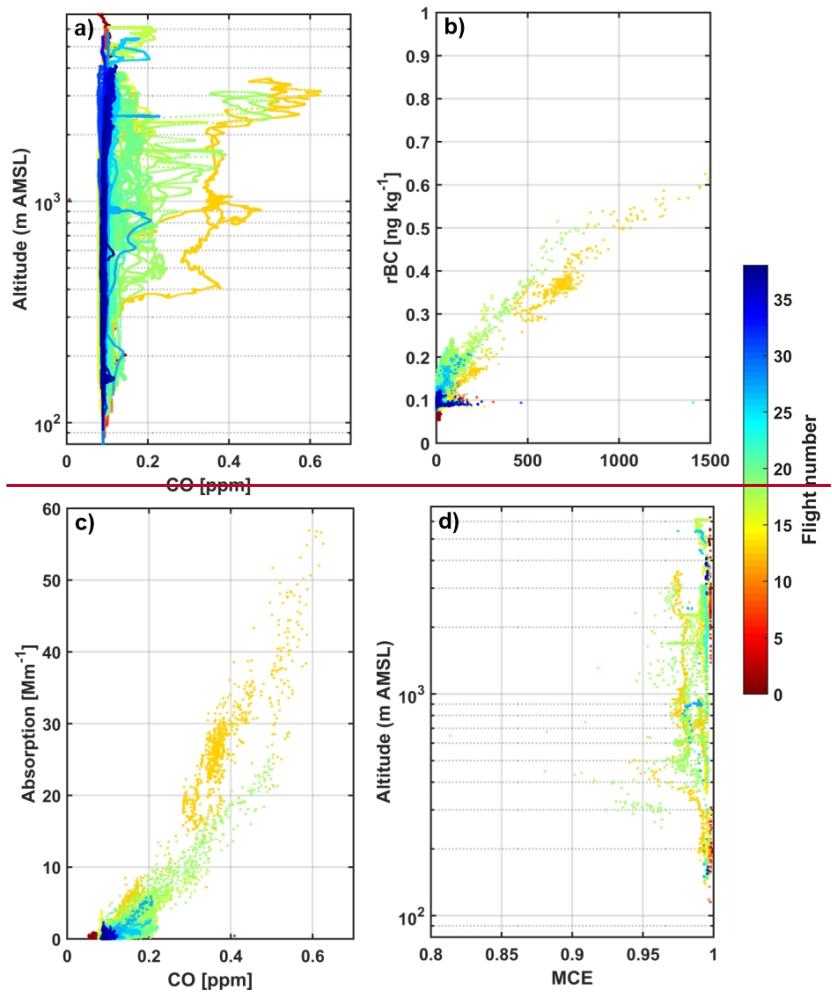


Figure 6. a) Vertical profiles of a) CO concentrations and, b) calculated MCE. MCE is only calculated for fire data. Correlations between CO and c) rBC mass and d) correlation between CO and rBC mass, and e) correlation between CO and absorption scattering coefficients at 550 nm for each flight during the entire ACME-V campaign are also shown.

~~Also shown is the calculated modified combustion efficiency (MCE) for data points classified as being impacted by the fires. Data classified as originating from the fires is coloured orange. All 1-second data are coloured by flight number.~~

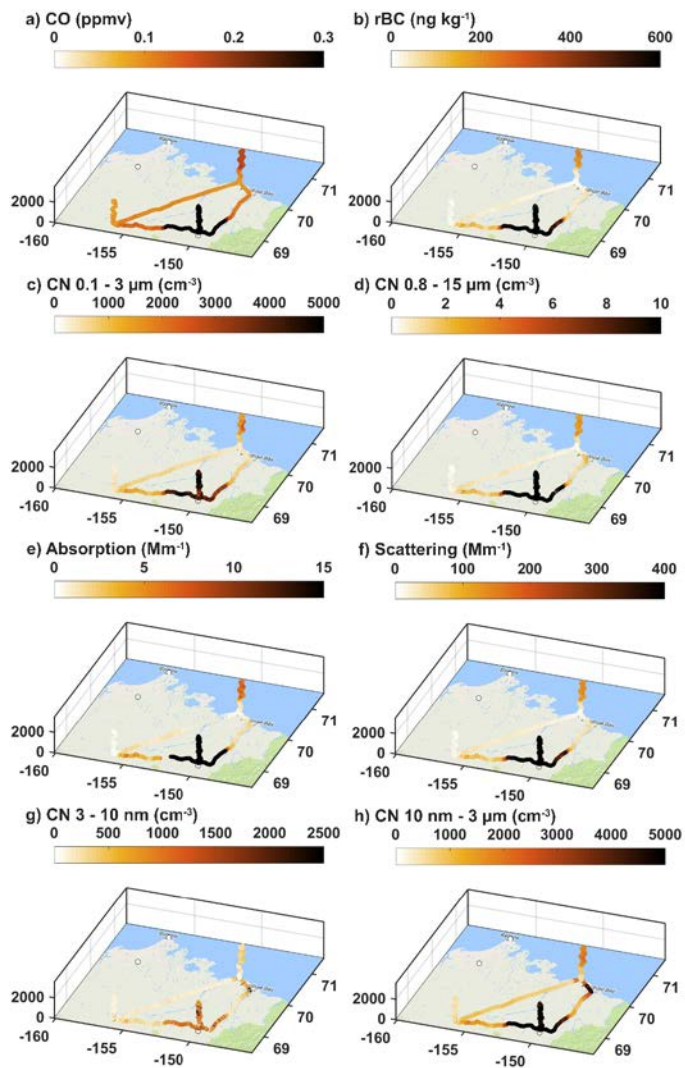
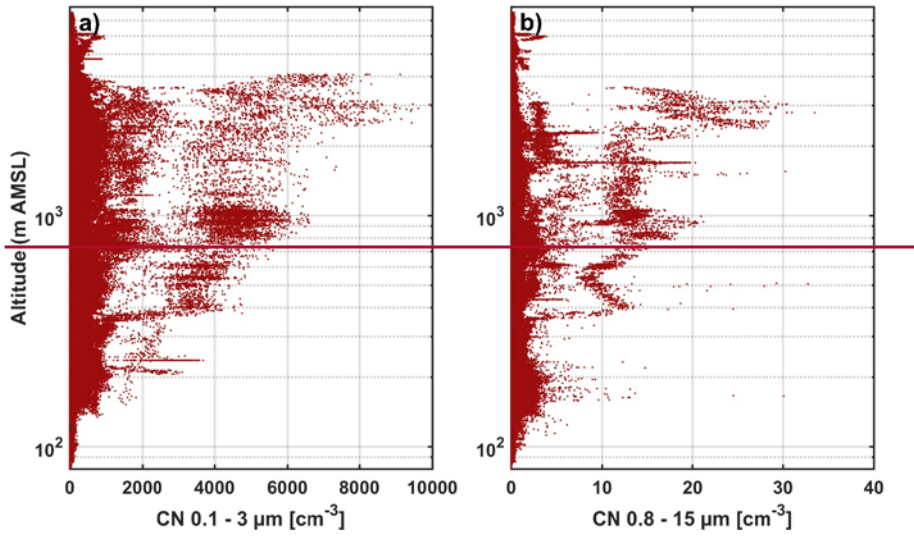


Figure 7. 4D profiles of a) CO, b) rBC mass, c) CN with  $D_p = 0.1 - 3 \mu\text{m}$ , d) CN with  $D_p = 0.8 - 15 \mu\text{m}$ , e) absorption coefficient, f) scattering coefficient, g) CN with  $D_p = 3 - 10 \text{ nm}$ , and h) CN with  $D_p = 10 \text{ nm} - 3 \mu\text{m}$  from F13 on 30 June 2015. The left, bottom, and right axes are altitude, longitude, and latitude, respectively. **Data are 1-second.**





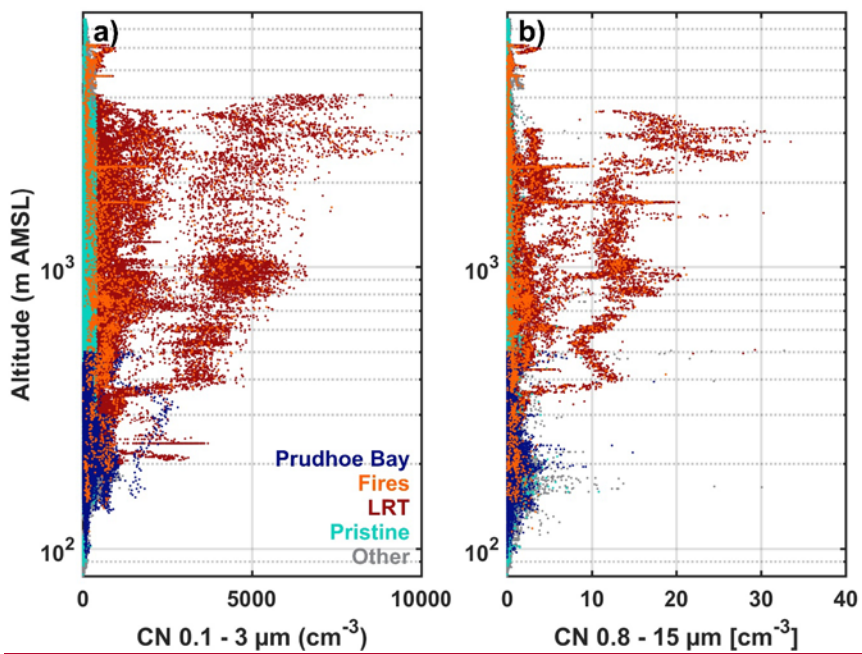


Figure 8. Vertical profiles of particle number concentrations measured from the a) CN with  $D_p = 0.1 - 3 \mu\text{m}$  and b) CN with  $D_p = 0.8 - 15 \mu\text{m}$  colored by flight number. Data classified as originating from Prudhoe Bay, fires, and long-range transported are coloured blue, orange, and red, respectively. Data shown are 1-second.

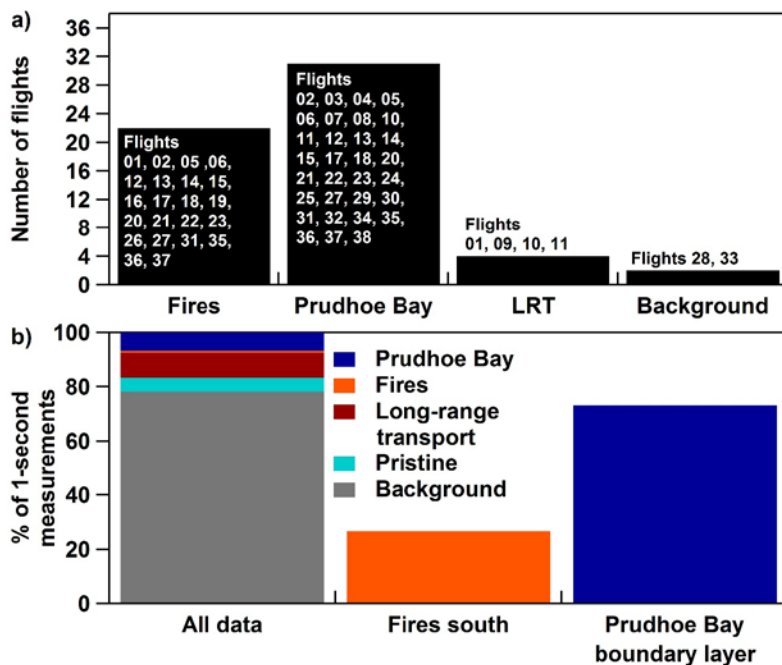


Figure 9. a) The number of flights that were classified as impacted by aerosols from the central Alaskan fires, Prudhoe Bay (PHB), long-range transport, pristine conditions, and background (BG; with no influences from fires, PHB, or LRT), and pristine conditions (based on parameter thresholds in Table 2Table 2). b) Percentage of 1-second measurements that were classified as impacted by fires, PHBPrudhoe Bay, long-range transport, BG, and pristinepristine conditions, and background. The percentage of 1-second fire data points (i.e., from the orange portion in the first bar in b), but for fires south (of 69°N) are shown by the (second bar from the left). The percentage of 1-second PHBPrudhoe Bay data points (i.e., from the grey-blue portion in the first bar in b), but for data near PHB (within a 50 km radius; third bar) and near PHB + in the boundary layer are shown by the (fourth bar last bar from the left), near PHB (within a 50 km radius and < 500 m AMSL) are also shown.