Response to Referee #1:

This paper uses estimates of cloud properties from satellite remote sensing (AIRS, CloudSat, CALIPSO, MODIS) and black carbon concentrations from the FLEXPART transport model to study cloud-aerosol microphysical effects over the Arctic Ocean. It is found that combustion aerosols are associated with large changes in surface longwave radiation over sea ice. However, up to 91% of the cloud fraction differences between all and clean conditions is due to meteorological conditions, i.e., the black carbon is essentially a passive tracer in these cases.

This paper is well-written and the results are very interesting. I think this paper is suitable for publication in ACP after addressing my concerns below.

We kindly thank this Referee for their very helpful comments. Please find our responses below. Bold lettering indicates newly added text.

Main comments:

1) There needs to be justification in the introduction for focusing on black carbon. It is not clear why other aerosol sources are excluded in this study. There needs to be an overview of previous studies of the role of black carbon in Arctic cloud-aerosol effects.

We have added the following paragraph to the introduction to a) address why the study focused on combustion aerosols (as proxied by BC), instead of on other aerosol types, and b) present an overview of previous studies relevant to combustion microphysical effects on clouds:

"**In this paper, we focus on the effects of combustion-derived (i.e., anthropogenic pollution plus smoke) aerosols on clouds. Combustion-derived aerosols are strongly impacted by anthropogenic activity, and tend to dominate columnar mass under high AOD conditions in the Arctic (Xie et al., 2018), although in spring the more well-mixed mineral dust can also contribute ~10% to total Arctic AOD levels (Breider et al., 2014; Groot Zwaaftink et al., 2016). Combustion aerosols have pronounced effects on Arctic cloud microphysical and radiative properties (e.g., Carrió et al. (2005); Coopman et al. (2016, 2018); Earle et al. (2011); Garrett et al. (2004); Jouan et al. (2014); Lubin and Vogelmann (2006); Tietze et al. (2011); Zamora et al. (2016, 2017); Zhao and Garrett (2015)). Their cloud impacts are likely to be particularly large during winter and spring, when they are transported to the Arctic most efficiently, and when precipitation is reduced, causing a peak in aerosol abundance at many remote Arctic ground stations known as "Arctic Haze" (Barrie, 1986; Croft et al., 2016; Quinn et al., 2007; Stohl, 2006). However, so far it has been challenging to assess their cloud effects on the Arctic region as a whole, due to large cloud model uncertainties, spatial/temporal observation limitations, and difficulties obtaining some remote sensing information at high latitudes.**"

We also added a clarification in section 2.1, where we explained why BC specifically is used as a proxy for combustion aerosols:

"FLEXPART BC is used in this study as a proxy for all combustion aerosols**, because they very often contain BC, although in somewhat different fractions**. The association of high levels of modeled BC with CALIPSO aerosols in general (see Zamora et al. (2017)) indicates that modeled BC is a fairly good proxy for strong (CALIPSO-detectable) aerosol layers during polar night, even though some local sources of combustion aerosols (Creamean et al., 2018; Maahn et al., 2017) might not be included in the model. Model comparisons to CALIPSO aerosol data in the study region also indicate that model-identified clean conditions (BC < 30 ng m^{-3}) are associated with significantly lower levels of CALIPSO aerosol layer presence relative to average or polluted conditions (see supplement for further details)."

2) The measurements are separated into clear and cloudy conditions as a function of height. So different days are used in each of these averages?

To clarify, we did not separate measurements into clear and cloudy conditions for the main body of work described in the paper. We identified the subset of the data present in air mass conditions determined to be clean. This subset was comprised of a reduced number of days relative to the full dataset. Because there are many clean cases, and because defining a lower bound on "polluted" cases would introduce an arbitrary threshold, we compare the clean subset to the full dataset, which includes the clean subset.

It would be very useful to have a figure showing what days were used in the different averages.

We now show what days were used in the different averages in the new Table 2. We also added some associated discussion below:

"To help better understand co-varying meteorological effects on CF specifically, we assessed a generalized additive model (GAM) (Hastie and Tibshirani, 1990) of the $\overline{dRH}$, $\overline{dT}$, and $\overline{dCF}$ data at each vertical level, season, and surface type (Table 1). **Seasonal differences in light, sea ice extent, and BC levels led to some sample number differences for sea ice and open ocean at different times of the year (Table 2). In the GAM, the seasonal values in Table 1 were weighted equally to represent the equal periods of the year being sampled.**"

Are the averages as a function of height in Figure 1 using data from different seasons?

Yes, the averages in Figure 1 are calculated from all seasons. We now state this in the text.

If so, it obscures some of the effects since, for example, if the aerosols cause increased activation of ice crystals and precipitation at high altitudes then it will not be possible to see the impact of these at lower altitudes.

That is a good point. To address this concern, we now show the data separated by season (see the new Figures S1-S3). Separating by season results in some loss of sample sizes in the individual figures, and reduced statistical significance for the individual seasonal plots. However, where sample sizes were large enough, the seasonal data show very similar vertical trends in dCF as during the annual average shown in Figure 1. To draw attention to this new information, we have added the following text in the main document:

*"*Cloud fraction substantially differed among all and clean conditions for many combinations of T, RH, altitude and surface type (Fig. 1). Estimated aerosol impacts on total CF depend on altitude and surface type**, but are fairly consistent among seasons (Figs. S1-S3)**."

Are the upper and lower quartiles used to make Figure 2 using data from different seasons? I first interpreted Figure 2f as an example of increased ice production at higher altitudes and depletion of water vapor due to deposition at lower altitudes, but this may not be the case if the values at different heights are calculated separately.

We now clarify in the text that Figure 2 does include data from all seasons, and that Fig. 2f and 2g represent median profiles (and thus the T and RH profiles were not calculated separately at different heights, unlike in Figure 1).

"To illustrate this point, Figure 2 shows the longwave CRE_{BOA} for the upper and lower quartiles of FLEXPART model column BC concentrations**, calculated** during the **entire** study period. The upper and lower quartile ranges of column BC levels are associated with very large (~10 W m^{-2}) differences in **median** longwave CRE_{BOA} over sea ice (Fig. 2). However, **when we compare the median relative humidity and temperature profiles with column BC levels in the upper quartile over sea ice (Fig. 2f, red lines) and open ocean (Fig. 2g, red lines) to the lower quartile profiles (blue lines, same figures), it is clear that** column BC levels over sea ice are also associated with noticeable differences in **median** relative humidity and temperature **profiles** (Fig. 2f). Small differences in lower tropospheric stability (Fig. 2e), defined as the difference in potential temperature between 700 and 1000 hPa, are also observed. These meteorological factors strongly affect CF and CP, which in turn help drive CRE_{BOA}. As a result, aerosol microphysical effects may contribute only a fraction of the CRE_{BOA} differences shown in Figure 2."

More investigation would be required to verify the referee's original interpretation about increased ice production. Such work is beyond the scope of this manuscript, but would certainly be interesting to pursue further.

3) Throughout the paper it is said that this study is focused on regional-scale effects. What is meant by this exactly, that sea ice and open ocean is analyzed separately? For example, page 5, line 17, what is meant by "regionally averaged"?

To avoid confusion, we have changed this wording throughout the paper. For example, see the edited specific text the referee pointed out above:

"~~Regionally averaged d~~ **D**ifferences in relative humidity, temperature, and 12.5 km^2 gridded CF ($\overline{dRH}$, $\overline{dT}$, and $\overline{dCF}$, respectively) between all ($\overline{RH}$, $\overline{T}$, and $\overline{CF}$, respectively) and clean ($\overline{RH_c}$, $\overline{T_c}$, and $\overline{CF_c}$, respectively) conditions were calculated **over sea ice and open ocean regions**…"

4) The results indicating increased ice precipitation in MPC at low altitudes and decreased precipitation at high altitudes is very interesting. It would be good to include a more detailed comparison with the results from previous studies in the discussion section.

Thank you. We have added the following new text in the Introduction:

"However, the regional-scale importance of aerosol microphysical processes on CF has been difficult to constrain from observations and models**, particularly due to uncertainties in how aerosols affect precipitation and ice nucleation rates (Gettelman, 2015; Morrison et al., 2012; Ovchinnikov et al., 2014).**"

And to the Discussion:

"There are several mechanisms by which aerosols might modify ice crystal number or size that could cause the observed changes in precipitation and CF in the 6-8.5 km range. **Although BC itself is not a good source of INPs (Vergara-Temprado et al., 2018),** combustion aerosols associated with BC might act as ice nucleating particles (INPs) (Kanji et al., 2017) at the extreme cold temperatures found at high-altitude Arctic polar night. This could potentially lead to smaller, more numerous ice particles that precipitate less (Lohmann and Feichter, 2005)**, in line with our observations**, although some models suggest that INPs may instead lead to larger ice crystals in cirrus clouds compared to homogeneous freezing (Heymsfield et al., 2016). Alternatively, combustion aerosols might reduce the efficiency of pre-existing INPs through the "deactivation effect" (Archuleta et al., 2005; Cziczo et al., 2009). Reduced ice crystal formation rates could then lead to more frequent air mass saturation with respect to liquid water, more water droplets that freeze homogeneously, and smaller, more numerous ice particles, and less precipitation (Girard et al., 2013)**, as we observe here**. This effect could lead to enhanced total CF over the Arctic (Du et al., 2011). Although absolute humidity within the different T-RH bins between 6-8.5 km is not systematically related to higher $dBC_{T,RH}$ levels as one might expect with the deactivation effect, it is possible that pre-sorting the data by 5% RH bins to reduce the impacts of meteorological co-variation could make evidence for this effect more difficult to observe. Therefore, it is difficult to say whether this study is consistent with the deactivation effect hypothesis, but it does not preclude it."

and

*"*The specific microphysical mechanisms affecting lower altitude clouds are more challenging to identify without in situ data due to the high prevalence of liquid-containing clouds (Fig. 5a). **Combustion aerosols can affect precipitation rates by changing droplet numbers and sizes, and thereby possibly collision and coalescence (Albrecht, 1989), riming (Lohmann and Feichter, 2005; Saleeby et al., 2009), or freezing (Bigg, 1953). If these aerosols affect INP levels, they could also affect ice nucleation rates and ice particle concentrations, leading to MPC and LPC glaciation, enhanced precipitation, and reduced cloud cover (the "glaciation effect"). Our observations support the possibility of a glaciation effect, because**…*"*

and

"These observations are in-line with other studies indicating that aerosols can dissipate Arctic MPCs (Fu and Xue, 2017; Norgren et al., 2018) **and increase their precipitation (Kravitz et al., 2014; Morrison et al., 2011). Assuming they act as INPs, various modelling studies and a remote sensing study also suggest that aerosols can reduce liquid water path or supercooled water frequency (Fan et al., 2009; Morrison et al., 2011; Ovchinnikov et al., 2014; Pinto, 1998; Tan et al., 2014)**. The observations over sea ice contrast with some model predictions that MPCs should increase in more polluted conditions through the deactivation effect (Du et al., 2011; Girard et al., 2005, 2013). They also contrast with a previous remote sensing study (Zamora et al., 2017) indicating that thin and predominantly liquid Arctic Ocean clouds are more likely to be the liquid phase at high BC concentrations. However, the clouds in that study may not be fully comparable, as they constitute only ~5% of the cloud types in this study. Note that shortwave processes might alter how aerosols impact mixed phase CF during daytime (Solomon et al., 2015), and any such effects would not be observed in the current, nighttime study. Changes in higher altitude clouds might also change underlying cloud properties through a seeding effect, which could impact cloud properties at lower altitudes."

Minor comments:

1) Page 5, line 4: How does focusing on relative rather than absolute differences get around the issue of misclassification of small supercooled water as ice particles?

We have re-worded for clarification:

"Previous work shows that this product can severely underestimate downwelling LW radiation due to misclassification of small supercooled water as ice particles (Van Tricht et al., 2016)**, leading to uncertainties in the absolute values of CRE_{BOA}**. **Here**, we primarily focus on **relative differences** in CRE_{BOA} **between two subsets of data: those with high and low modeled BC values. The uncertainty due to misclassification of small particle phase is similar in both subsets of data, which are collected over the same surfaces and years, allowing for meaningful comparisons to be made between the two datasets despite uncertainty in the absolute values.**"

2) Page 7, line 21-22: This is a very interesting result but why wouldn't it be the case over open ocean?

Based on comments from the other referee, we have taken this paragraph out, as it was not fully substantiated.

3) Page 7-8, lines 33-1: Why is this relationship due to microphysical effects and not meteorology? Can it be concluded that microphysical effects are stronger in stable conditions or just more observable? If it is stronger then why is there no significant difference in the LTS for high and low quartile black carbon (Figure 2e)?

Thanks for pointing this out. We have re-worded to make it clearer that the microphysical effects are more apparent under certain meteorological conditions, where their effects are less likely to be overwhelmed by other factors:

"This finding suggests that aerosol microphysical impacts on low-altitude clouds are more **observable** at the lower temperatures and/or more stable conditions over sea ice. Previous studies have also observed **more apparent** aerosol microphysical effects under more stable conditions in the Arctic (Coopman et al., 2018; Zamora et al., 2017)."

4) Page 8, lines 11-12: This is an interesting result but what would this be the case?

We have changed the text as follows:

"From Table S2, over sea ice between 1.5-2.5 km, the relative contributions of LPCs and MPCs were significantly lower at high $dBC_{T,RH}$ levels (>20 ng m^{-3}), whereas that of IPCs was significantly higher. **The reduction in liquid-containing clouds at higher $dBC_{T,RH}$ levels is consistent with a glaciation effect, whereby increased presence of aerosols leads to ice particle formation and cloud dissipation, as observed in section 3.1.**"

5) Page 10, lines 3-5: Are the results of this study consistent with the deactivation of pre-existing INP hypothesis (Archuleta et al. 2015; Cziczo et al. 2009)?

We have changed the text, as follows:

"Alternatively, combustion aerosols might reduce the efficiency of pre-existing INPs through the "deactivation effect" (Archuleta et al., 2005; Cziczo et al., 2009). Reduced ice crystal formation rates could then lead to more frequent air mass saturation with respect to liquid water, more water droplets that freeze homogeneously, and smaller, more numerous ice particles, and less precipitation (Girard et al., 2013) **as observed here**. This effect could lead to enhanced total CF over the Arctic (Du et al., 2011). Although absolute humidity within the different T-RH bins between 6-8.5 km is not systematically related to higher $dBC_{T,RH}$ levels as one might expect with the deactivation effect, it is possible that pre-sorting the data by 5% RH bins to reduce the impacts of meteorological co-variation could make evidence for this effect more difficult to observe.

Therefore, it is difficult to say whether this study is consistent with the deactivation effect hypothesis, but it does not preclude it."

6) Page 11, lines 15-16: Why would the impact on MPCs be different over sea ice and open ocean?

We took out the "relative to IPCs" text in the paragraph of reference below, since the difference relative to IPCs was not significant (see Fig. 3a), and have added the following text:

"Below 1.5 km, we also observed a 7% reduction in the LPC and MPC fractions over sea ice, but a slight increase in MPCs relative to ~~IPCs and~~ LPCs over open ocean. **The different effect on MPCs over sea ice and open ocean may be related to the higher temperatures over open ocean, leading to less efficient ice formation, or to some other, as yet unknown, factor.**"

Response to Referee #2:

Summary

The paper presents an analysis of aerosol-cloud effects using data from satellites and FLEXPART to provide first observation-based constraint on regional aerosol microphysical effects on total nighttime CF over the Arctic Ocean. An important feature is that the study accounts for the co-variation of airmass aerosol and meteorology. The topic and findings seem appropriate for the ACP readership.

Overall, the methodology seems sound and findings of interest. However, while the text is very well written from a grammatical standpoint, particularly in the presentation of the results the text is either disjointed or lacks sufficient information to follow in the figures/tables. Specific examples are given below for improving the flow of the text, along with other comments and question. The suggestions are not comprehensive so the authors are advised to please use them as a necessary-but-not-sufficient guide of how the manuscript should be modified for an outside reader to follow and absorb the content. For these reasons it is recommended that the paper be accepted but with major revision.

We appreciate the time the referee took to give such helpful and thorough comments, which have improved the quality and clarity of the manuscript. We particularly appreciate the reviewer pointing out cases where other reader might have found things confusing. Please find our responses below. Bold lettering indicates newly added text.

Major Comments

(Each specific comment is preceded by the page and line numbers. If in the supplemental material, only the line number is used).

1. In the supplement, the evaluation of FLEXPART BC concentrations is based on CALIPSO aerosol profiles. Arguments are provided as to why BC would likely be the dominant aerosol type for the location and period of study. However, since CALIPSO cannot tell the difference between BC and other aerosol types, a more direct comparison would be if FLEXPART could provide the total distributions of all relevant aerosols for the region. Is that a capability of FLEXPART? If so, then FLEXPART could also be used to assess the fraction of the total aerosols that are BC and support the arguments provided.

This is a good suggestion. However, FLEXPART is no "normal" aerosol model but rather a Lagrangian tracer model. That means that simulations are normally done for one specific aerosol type and not for a full suite of different aerosols, as is often available in other models. The advantage of FLEXPART is its higher accuracy with respect to long-range transport (notice that "normal" Eulerian transport models suffer from excessive numerical dissipation, failing to preserve chemical plumes over long transport distances, see, e.g., Eastham and Jacob (2017)). FLEXPART's disadvantage is that it is not built for simulating many species at the same time, including their interactions. For this study, we think it is more important to have the plumes of a "representative" combustion aerosol species at the right place at the right time, rather than to quantify the contributions of many different species.

Dust is definitely another important aerosol type in the Arctic, and is not well correlated with BC. Limited simulations of dust have been done with FLEXPART (Groot Zwaaftink et al., 2016). It was found that local "sharp" plumes are mainly present in the fall, whereas dust emissions in winter are limited by extensive snow cover in the Arctic. On the other hand, long-range transport of dust from low-latitude sources (e.g., from the Sahara) also occurs in winter. However, dust from these very remote sources is well mixed and, at moderate concentrations, nearly omni-present in the Arctic free troposphere, so that it would not be straightforward to distinguish "dusty" and "clean" air masses.

Based on this information, we have addressed the referee's comment above in four ways. First, we have added supporting information on dust aerosol sources:

"Mineral dust can be found throughout the Arctic atmosphere. **However, although there are some local "sharp" dust plumes at some locations in the fall, wintertime local dust emissions are limited by extensive snow cover, and long-range transport of low-latitude dust is well mixed in the winter and, at moderate concentrations, is nearly omni-present in the Arctic free troposphere (Breider et al., 2014; Groot Zwaaftink et al., 2016).**"

Second, we have clarified our focus better by changing the title:

"A satellite-based estimate of **combustion** aerosol cloud microphysical effects over the Arctic Ocean"

And we make it clearer throughout the text that we are focusing on the specific microphysical effects of combustion aerosols on clouds in this study, and not on the effects of all aerosols on clouds.

Thirdly, we better clarify how to interpret the information in the supplement. We make it clearer that our main focus in the supplement is on identifying:

1) ***false positives*** *(where BC aerosols were not present as evidenced by the lack of a CALIPSO aerosol layer, but the model said they were), and*
2) ***an upper limit on false negatives*** *(where BC aerosols could have been present based on the presence of a CALIPSO layer, but the model said they were not).*

Note that based on our method, additional information on the distributions of other aerosol types from FLEXPART would not affect the false positive rate. It is *possible that this information could help refine and lower the estimated potential false negative rate. However, because FLEXPART dust aerosols are poorly validated, especially over oceanic regions of the Arctic, we would be adding in an unknown level of uncertainty in the false negative estimates if we assume that the poorly validated distributions of dust are correct, and we use these estimates of other aerosols to lower the estimated false negative rate for combustion aerosols. Under our current assumptions, we are only able to provide an upper estimate of the false negative rate, but this upper limit is fairly well-constrained.*

Lastly, we would like to point out that although this work adds additional validation of FLEXPART BC levels by comparison to CALIPSO aerosol profiles, it is by no means the only validation of FLEXPART Arctic BC. Previous validation, with positive results, has been based on aircraft and ground measurements, and other satellite data. We cite some of this previous work in section 2.1:

"FLEXPART is widely used, and is well-validated for the purpose of studying Arctic smoke and pollution transport (Damoah et al., 2004; Eckhardt et al., 2015; Forster et al., 2001; Paris et al., 2009; Sodemann et al., 2011; Stohl et al., 2002, 2003, 2015; Zamora et al., 2017)."

Because the information in the supplementary material provides better constraints on FLEXPART BC vertical distributions over open ocean, we think it is a useful contribution. However, the previous studies cited above add much more confidence to our ability to correctly identify combustion aerosol layers with the model than the information provided in the supplement alone. Note also that there are a variety of other previous related studies that have used FLEXPART to identify combustion aerosols for aerosol-cloud interaction work, and these studies did not have the benefit of the extra

knowledge we provide in the supplementary material (e.g., Coopman et al. (2016, 2018); Tietze et al. (2011); Zamora et al. (2017)). Therefore, even though there are some uncertainties in our analysis in the supplementary material, we do not think they would preclude the greater work from being useful.

2. P3: The focus is on BC concentrations, why? Please provide a justification, which appears to have been buried in the supplemental material.

We have added the following paragraph to the introduction to address why the study focused on combustion aerosols (as proxied by BC), instead of on other aerosol sources:

"**In this paper, we focus on the effects of combustion-derived (i.e., anthropogenic pollution plus smoke) aerosols on clouds. Combustion-derived aerosols are strongly impacted by anthropogenic activity, and tend to dominate columnar mass under high AOD conditions in the Arctic (Xie et al., 2018), although in spring the more well-mixed mineral dust can also contribute ~10% to total Arctic AOD levels (Breider et al., 2014; Groot Zwaaftink et al., 2016). Combustion aerosols have pronounced effects on Arctic cloud microphysical and radiative properties (e.g., Carrió et al. (2005); Coopman et al. (2016, 2018); Earle et al. (2011); Garrett et al. (2004); Jouan et al. (2014); Lubin and Vogelmann (2006); Tietze et al. (2011); Zamora et al. (2016, 2017); Zhao and Garrett (2015)). Their cloud impacts are likely to be particularly large during winter and spring, when they are transported to the Arctic most efficiently, and when precipitation is reduced, causing a peak in aerosol abundance at many remote Arctic ground stations known as "Arctic Haze" (Barrie, 1986; Croft et al., 2016; Quinn et al., 2007; Stohl, 2006). However, so far it has been challenging to assess their cloud effects on the Arctic region as a whole, due to large cloud model uncertainties, spatial/temporal observation limitations, and difficulties obtaining some remote sensing information at high latitudes.**"

We also added a clarification in section 2.1, where we explained why BC specifically is used as a proxy for combustion aerosols:

"FLEXPART BC is used in this study as a proxy for all combustion aerosols**, because they very often contain BC, although in somewhat different fractions**. The association of high levels of modeled BC with CALIPSO aerosols in general (see Zamora et al. (2017)) indicates that modeled BC is a fairly good proxy for strong (CALIPSO-detectable) aerosol layers during polar night, even though some local sources of combustion aerosols (Creamean et al., 2018; Maahn et al., 2017) might not be included in the model. Model comparisons to CALIPSO aerosol data in the study region also indicate that model-identified clean conditions (BC < 30 ng m^{-3}) are associated with significantly lower levels of CALIPSO aerosol layer presence relative to average or polluted conditions (see supplement for further details)."

3. In the results section, there is a lot of jumping back and forth between the figures in the main text and the supplemental material. As such, some of the supplemental material did not

seem very "supplemental". Recommend moving frequently referred to figures or tables to the main text.

> *The original Table S2 was the most referenced supplemental item. This table presents the same information as in the original Figure 3, except that it also shows which cases were significantly different at high (> 20 ng m^{-3}) BC levels. We have now added this information to the new Figure 3 (as the orange triangles), and are thus able to remove the references to Table S2 in the main text. Table S1 was referenced twice, and has been moved to the main text. Figure S1 and S2 are now only referenced once each, and remain in the supplement.*

4. L94-100: The supplemental summary statement seems inconsistent. The last sentence states that "the model does represent aerosol transport over the Arctic well" but the first sentence states that "CALIPSO aerosol layers contributed significantly smaller volume than in all and model-identified polluted conditions"; if the latter is correct, how can the former be? How does this affect the results? (particularly Fig. 4)

> *Our apologies for the confusing wording that led the referee to wonder if there was an inconsistency. A low aerosol volume in clean conditions is what one would expect if the model was performing well. With re-wording of this paragraph, we hope that it is now clear that there is no inconsistency:*
>
> *"***In summary, for FLEXPART to correctly identify clean (i.e., low combustion aerosol) conditions, it needs to be able to correctly simulate the horizontal and vertical distributions of combustion aerosols**. **Previously conducted model validation studies indicate that FLEXPART has skill in simulating the horizontal locations of BC transport over the Arctic. Here, we show that the volume of CALIPSO vertical aerosol layers is significantly smaller in model-estimated clean conditions in the vertical column** than in all **conditions, or in** model-identified polluted conditions. **This result indicates that FLEXPART also has some skill in the vertical layer prediction of BC aerosols over the Arctic Ocean. Moreover, we observed** no major spatial biases in the false negative rates that would preclude the regional comparisons between sea ice and open ocean regions. **Together, these findings and previous work support the use of FLEXPART for identifying clean conditions for the purposes of this study.***"*

5. The issue with (4) might stem from confusion regarding Figure S3. The plot shows that fraction of the different altitude layers where CALIPSO detects aerosol and FLEXPART identifies (a) are clear, and (b) are polluted.

a) While (a) are false negatives (consistent with the header at the top of the plot), is seems that (b) is inconsistent with its header and it is not false positives; rather it is showing when FLEXPART accurately identifies aerosol layers (i.e., CALIPSO=yes, and Pollution=yes). Is the caption wording correct?

To be more accurate, we would slightly rephrase the referee's interpretation above. In Figure S3b we have plotted only the subset of cases with high BC levels (Pollution = yes). Figure S3b does show when FLEXPART is likely to have accurately identified aerosol layers (when the color axis values are closer to red, CALIPSO=yes). Red colors indicate that the model vertical layer was, on average, associated with a large CALIPSO aerosol layer at that location.

However, the same figure also shows the opposite (when values are closer to white, CALIPSO=no), indicating that the model vertical layer was, on average, not associated with a large CALIPSO aerosol layer at that location. With the header, we had intended to guide the reader to focus on those values that were closer to white: i.e., when false negatives are more likely to be present. We have now re-worded the caption to hopefully make this clearer.

b) Also, the caption and the headers refer to "likely at large values" and "likely at small values"; values in what, BC concentration? If so, the plot only displays part of the information, the layer fractions and, besides the "clean" and "polluted" columns, there is not information on concentration level (that is consistent with the headers at the top of the figure). Please clarify.

We were referring to values in the color axis, which show to the fraction of model vertical layer containing an observed CALIPSO aerosol layer averaged at that location. To interpret the plot, no additional BC concentration information is needed, other than the knowledge that Figure S3a only shows the clean subset (all data with BC < 30 ng m^{-3}), and Figure S3b only shows the polluted subset (all data with BC >150 ng m^{-3}). This information has now been better clarified in the caption.

6. Fig. 3: There are too many different aspects are loaded into this figure, making it very difficult to follow the discussed patterns in a single variable type with altitude (e.g., dCF). Recommend moving 3b to a new figure, and make a-c panel plots in Fig. 3 with altitude separately for (a) dCF, (b) dpptn, and (c) dCP(IPC, MPC, LPC). For the old 3b, the current overlays are too cluttered and recommend separating into (a) and (b) the pptn and CF components.

We have followed the referee's suggestions (see the new Figures 3 and 5). Note that there were two errors in the old Figure 3a. First, the labeling for the relative and absolute columns were reversed (this is now fixed in the new Figure 3). Also, we have corrected an error in the confidence interval values, which are now substantially reduced in most cases.

Secondary Comments

(Some rewordings are suggested that were easier for me to understand.)

7. P6 , L25,"very large (~25 W m^{-2})": Where does this value come from? In the plots, values range from 0 to ~70 W m^{-2}. Please explain.

We are glad you asked us to clarify our methods here, because in doing so, we found an error in how these values were calculated. The corrected median CRE_{BOA} difference over sea ice is now reduced to 10 W m^{-2} from 25 W m^{-2}. The general conclusions remain the same though, since this is still quite a large difference.

To clarify our methods, we have added the following text:

"The upper and lower quartile ranges of column BC levels are associated with very large (~**10** W m^{-2}) differences in longwave CRE_{BOA} over sea ice (Fig. 2)**. This value is estimated from the median difference in 12.5 km^2 gridded CRE_{BOA} values over sea ice regions across the Arctic Ocean during the study period, in grid cells with a minimum of at least 3 observations in the upper and lower quartile ranges of column BC levels**."

In fact, more text is needed to explain Fig. 2 which is a 7-panel plot. Currently, it seems "dropped in" without many of its aspects discussed.

In addition to the above text, we have also added in the following text:

"Systematic regional co-variability of aerosols and meteorological factors must be accounted for in order to avoid overestimating aerosol impacts on clouds (Coopman et al., 2018; Gryspeerdt et al., 2016). To illustrate this point, Figure 2 shows the longwave CRE_{BOA} for the upper and lower quartiles of FLEXPART model column BC concentrations**, calculated** during the **entire** study period. The upper and lower quartile ranges of column BC levels are associated with very large (~**10** W m^{-2}) differences in **median** longwave CRE_{BOA} over sea ice (Fig. 2). **However, when we compare the median relative humidity and temperature profiles with column BC levels in the upper quartile over sea ice (Fig. 2f, red lines) and open ocean (Fig. 2g, red lines) to the lower quartile profiles (blue lines, same figures), it is clear that** column BC levels **over sea ice** are also associated with noticeable differences in **median** relative humidity **and temperature profiles (Fig. 2f). Small differences in lower tropospheric stability (Fig. 2e), defined as the difference in potential temperature between 700 and 1000 hPa, are also observed.** These meteorological factors strongly affect CF and CP, which in turn help drive CRE_{BOA}. As a result, aerosol microphysical effects may contribute to only a fraction of the CRE_{BOA} differences shown in Figure 2."

And in section 3.1:

"Also, $dCF_{T,RH}$ changes at high $dBC_{T,RH}$/low altitude are **more observable** over sea ice (Fig. 4)**, where lower tropospheric stability was greater and temperatures were colder (Figs. 2e-g)**."

8. P7, L1, "up to 91% of the variability": Where is this value shown? I do not see any such value in Table S1.

This GAM result is a single value calculated from the data in Table S1 (now Table 1), and thus does not easily fit in that table or in any of the other figures or tables. Therefore, it is only presented in the text. To help clarify our methods regarding the GAM calculations, we have added the following text:

"To help better understand co-varying meteorological effects on CF specifically, we assessed a generalized additive model (GAM) (Hastie and Tibshirani, 1990) of the $\overline{dRH}$, $\overline{dT}$, and $\overline{dCF}$ data at each vertical level, season, and surface type (Table 1). **Seasonal differences in light, sea ice extent, and BC levels led to some sample number differences for sea ice and open ocean at different times of the year (Table 2). In the GAM, the seasonal values in Table 1 were weighted equally to represent the equal periods of the year being sampled.**

The GAM suggests that co-varying differences in $\overline{dRH}$ and $\overline{dT}$ by themselves can explain up to 91% of the variability in $\overline{dCF}$ (as measured by deviance, a statistic similar to variance (Jorgensen, 1997)). Because aerosols can co-vary with T and RH (e.g., because polluted air masses are more likely to have recently resided near the continental surface than clean air masses), aerosols could be responsible for some of this explained variability even without being explicitly included in this GAM. **For reference, a GAM based only on $\overline{dBC}$ explained up to 40% of $\overline{dCF}$ variability.** Thus, the 91% value is an upper estimate of the $\overline{dRH}$ and $\overline{dT}$ influences. Nonetheless, these findings underscore the importance of interpreting aerosol effects on clouds in the context of co-varying temperature and relative humidity. They also indicate that changes in T and RH of air masses entering the Arctic will likely have a very large influence on observed CF, to a degree that is likely to be much more regionally important than the microphysical effects of the aerosols themselves."

9. P7, L11-13, "Cloud fraction substantially differed...At the lowest levels...": At the lowest level (0.6-1.5 km) over open ocean, almost all of the grids have Xs meaning that they are not statistically significant. Is it then a correct interpretation to say that they differed substantially?

The text referred to above is:

"Cloud fraction substantially differed among all and clean conditions for many combinations of T, RH, altitude and surface type (Fig. 1). Estimated aerosol impacts on total CF depend on altitude and surface type. At the lowest levels (0.6-2.5 km over sea ice and 0.6-1.5 km over open ocean), weighted mean $dCF_{T,RH}$ ($\overline{dCF_{T,RH}}$) is negative, resulting in an ~6% reduction in CF relative to clean conditions over sea ice (-0.6% over open ocean) (Fig. 3)."

We believe the statement above is correct. From the above text, note that we do not say that there were substantial differences in all locations and at all individual T-RH grid

cells. We state that many of the grid cells differ substantially, but that the impacts depended on altitude and surface type. We also stated that the weighted mean is significantly different among all grid cells (also see our response to question 22 below).

10. P7, L20, "generally become more positive at constant RH with increasing T": This statement is not well supported given that many of the grids have white Xs preventing the "increasing with T" analysis.

To address this concern, we have taken this statement out.

11. Fig. 4: Please describe where the dots are from. Are they from the grids in the RH-T plots e.g. from Fig 1. but for each range of dBC? If so, are they only from those that are statistically different from zero?

*We now describe this better in the Figure caption: "***In order to avoid obscuring emergent properties of the full dataset, the data include all meteorological conditions, including those where $dCF_{T,RH}$ are not significantly different from zero (as noted by white Xs in Figure 1)***."*

12. P1, L19, "with implications for a warming Arctic." Such implications do not seem to have been discussed in the paper. Please add the discussion or remove this clause.

To better support this statement, we now add the following information in the "Summary and conclusions" section:

"Observations from others (e.g., Chernokulsky et al. (2017); Eastman and Warren (2010)) show that expansion of open ocean areas appears to be connected to changing Arctic Ocean cloud properties. The different cloud responses to aerosols that we observe over sea ice vs. open ocean may provide partial clues into the cause of this behaviour, and into the future impacts of combustion aerosols on the Arctic system in general."

13. P2, L22, "Tropospheric cloud data...": Please indicate earlier/here the source here (CloudSat and CALIPSO); the details of the products can remain where they are.

We have added this information, as suggested.

14. P3, L26: "in general (Zamora et al. 2017)..." → "in general. Zamora et al. (2017)..."

To clarify, the sentence now reads:

"FLEXPART BC is used in this study as a proxy for all combustion aerosols**. T**he association of high levels of modeled BC with CALIPSO aerosols in general **(see** Zamora et al. (2017)**)** indicates that modeled BC is a fairly good proxy for strong (CALIPSO-detectable) aerosol layers during polar night, even though some local sources of

combustion aerosols (Creamean et al., 2018; Maahn et al., 2017) might not be included in the model."

15. P3, L27: What is meant by "strong aerosol layers"? Also, what does "aersosols[6]" mean?

To clarify, we now state "…strong **(CALIPSO-detectable)**". "aersosols[6]" *was a typo. It now reads:* "aerosols **(Creamean et al., 2018; Maahn et al., 2017)**"

16. P3, last line, "Cloud fraction is not well defined in the literature. Here, it is...": The statement is incorrect and unnecessary: CF is defined in the literature (tho its determination can be challenging). Recommend just starting off with "Cloud fraction is operationally..."

Edited as suggested.

17. P4, L18, "and blowing snow": Your lowest altitude is 0.6 km; are you stating that blowing snow could be that high? If not, remove.

We spoke with a blowing snow expert (Y. Yang, personal communication), and were told that blowing snow up to 0.6 km in the Arctic above sea ice is not common, but is possible.

18. P4, L18-20, "Additionally, ...": As stated, why is CloudSat mistaking precipitation for clouds an issue? If the lidar signal is attenuated it is attenuated and one has no signal to work with. This is true regardless of CloudSat's potential mistake. It would only factor in the precipitation counts, which does not seem to be the topic here.

To clarify, if CloudSat wrongly misclassifies precipitation as a cloud, that not only adds error into the precipitation estimates, but also to the CF estimates that are key to this work. When available, CALIPSO lidar data add additional information to help reduce the probability of this misclassification by the CloudSat radar, but these data are not available under optically thick clouds.

19. P5, Section 2.3: Please include the local overpass time used from AIRS.

This information is now added.

20. P6, L15, "Our focus on nighttime data over the flat ocean surface likely reduces effects from large-scale vertical motion": I do not know what you intend to mean by "large-scale vertical motion" since certainly large-scale synoptic phenomena exist at nighttime (fronts, highs, lows, etc.). Recommend rephrasing.

Thank you, this now reads "Our focus on nighttime data over the flat ocean surface likely reduces effects from **solar-heating-driven** vertical motion"

21. L17, What is the meaning of "convection[36]"?

That was a typo. It now reads: "convection **(Serreze and Barry, 2005)**"

22. Fig. 1: The white Xs indicate that the grid is not significantly different from zero. In the analyses that follow, are only the non-X grids used? Please state and/or give justification for inclusion if they are.

We include all the data in the analyses, including those in gridcells marked by Xs in Figure 1. There are both technical and scientific reasons for doing so.

To clarify the technical reason (i.e., that the important thing about the white Xs in Figure 1 is their number and not their individual positions), we have added the following text to the caption of Figure 1:

"Figure 1: An example of $dCF_{T,RH}$ output at each altitude level. For illustration purposes, here each grid cell represents ≥ 7500 km^2 of gridded observations. Blue and red colors indicate negative and positive $dCF_{T,RH}$, respectively. A white X indicates that the cell value is not significantly different from zero (Wilcoxon rank test, $p < 0.05$). **Note that each underlying Wilcoxon rank test has a 5% chance of yielding a false positive indication of statistical significance or an unknown (but likely much higher) chance of yielding a false negative result. Consequently, the distribution of Xs should not be over-interpreted. The number of Xs, however, provides an objective way to test whether the evidence for an effect on the grid as a whole is significant. This is consistently the case; in all panels, individually significant cells numbered more than expected at random (binomial test, $p < 0.001$).**"

To further address the reviewer's comment, in the manuscript we have also better clarified the scientific reasons for including these cells. New text includes:

*"***Note that the $\overline{dCF_{T,RH}}$ value is based on all $dCF_{T,RH}$ data, including those from T and RH ranges where $dCF_{T,RH}$ is not significantly different from zero (i.e., as indicated by the white Xs in Figure 1). Including all data avoids biasing the results in favor of the meteorological conditions where $dCF_{T,RH}$ is most observable**."

*And in the Figure 4 caption: "***In order to avoid obscuring emergent properties of the full dataset, the data [shown in Figure 4] include all meteorological conditions, including those where $dCF_{T,RH}$ are not significantly different from zero (as noted by a white X in Figure 1)**."

Note that the cubic smoothing splines in Fig. 4 actually cross zero, so excluding those data that are not significantly different from zero would lead to less information on the system as a whole.

23. Fig. 3: Does the significance indicated by the asterisk apply to both the relative percent changes and absolute changes? Please state in text.

It does, and we now state this, as suggested.

24. P7, L17, "1.7% to 0.7%": These values for sea ice do not match what I see, which is -2% to 1% (unless you maybe meant only at higher altitudes?).

There was actually a negative sign in front of the 1.7% (it was easy to miss, being present on the line above, due to the formatting of the ACPD template). So rounded up, the -1.7% goes to -2%, and the 0.7% is rounded to 1%.

25. P8, L1, "more influential at the lower temperatures": The values are near dCF=0 for the higher altitude points so is this an accurate statement, especially given the dramatic drop off over sea ice with lower altitude (warmer temperature) suggesting the dominance of the stability criterion?

From the previous sentence, please note that we had been specifically discussing low altitude cases. To avoid confusion, we have re-worded as follows:

"Also, $dCF_{T,RH}$ changes at high $dBC_{T,RH}$/low altitude are larger over sea ice (Fig. 4). This finding suggests that aerosol microphysical impacts on **low-altitude** clouds are more influential at the lower temperatures and/or more stable conditions over sea ice."

26. P8, L6, "(Fig. 3)" → "(Fig. 3a)" for clarity.

Changed as suggested.

27. P8, L11, "Over sea ice" → "From Table S2, over sea ice..." for clarity, as otherwise it was not clear what supported the last sentence in the paragraph.

Changed as suggested.

28. P8, L18, "where LPC fractions were highest": Source for statement?

This sentence has been changed as follows: "The relative fraction of liquid clouds was reduced between 0.6-1.5 km **(Fig. 3a)**, where LPC fractions were highest **(see blue bars in Fig. 3b)**."

29. P8, L23, "An analysis..." → "We analyze the difference in precipitation frequency; however, an analysis..." (otherwise, the reader knows what you will not do, but it has not been stated what will be done).

Changed as suggested.

30. P9, L1, "~91% of the MPCs": Shown where?

The new text reads, "Over sea ice, ~94% of MPCs were present below 4 km **(Fig. 5a)**."

31. P9, L31, "night, potentially" → "night. This potentially leads to..." (break up the long sentence that also contains opposing points of view). And, which point of view does your study support?

Note that some new text was added based on a relevant paper that recently came out. The new text reads:

"**Although BC itself is not thought to be a good source of INPs (Vergara-Temprado et al., 2018), c**ombustion aerosols associated with BC might act as ice nucleating particles (INPs) (Kanji et al., 2017) at the extreme cold temperatures found at high-altitude Arctic polar night. **This could** potentially lead to smaller, more numerous ice particles that precipitate less (Lohmann and Feichter, 2005)**, in line with our observations**, although some models suggest that INPs may instead lead to larger ice crystals in cirrus clouds compared to homogeneous freezing (Heymsfield et al., 2016)."

32. P11, L12: Physically, why would one expect a larger aerosol effect for greater atmospheric stability?

The text has been modified as follows:

"In general, aerosol microphysical effects were most observable where the highest aerosol effect would be expected: at lower altitudes where aerosol concentrations are often higher (Devasthale et al., 2011b) and over sea ice, where atmospheric stability is greater**, and aerosol microphysical effects on clouds are less likely to be overwhelmed by meteorological factors such as high vertical velocity**."

33. P11, L25: Can you give a "for example" about what other cloud property relationships might exist?

The reviewer refers to the following text:

"Furthermore, these observations leave open the possibility that other cloud property relationships with $dBC_{T,RH}$ exist, but are not observable with the available data."

We are not sure we want to hypothesize in the paper, per se, but such relationships could include, for example, significant changes in cloud phase over open ocean that were just not observable in our 2-year dataset given the available sample sizes.

34. Need the "author contributions" section for ACP

These have now been added, thanks.

Supplemental material

35. L49-52: The concern about the variations in BC:OC ratios seems misplaced to me since the CALIPSO cannot tell the difference between those aerosol types.

To clarify, this uncertainty is mentioned exactly because CALIPSO cannot tell the difference between those aerosol types. The concern is that if we assume that CALIPSO can detect combustion aerosols in general (including OC and BC aerosols), and we are using model BC as a proxy for all of these combustion aerosols, we may underestimate combustion aerosol layers with very high OC:BC ratios.

36. L33-35, "it is unclear how thick an observed CALIPSO aerosol layer (measured in meters) must be to influence the average BC concentration in an altitude range...": The part "must be to influence the average BC concentration" seems odd in that there is no "influence" on the average BC concentrations. Please reword.

New text now reads: "As such, it is unclear how observed CALIPSO aerosol layer **thickness** (measured in meters) **would relate to** the average BC concentration in an altitude range equivalent to the FLEXPART model's vertical resolution (measured in kilometers)."

37. L38: "locations of false" → "locations of FLEXPART false"

Changed as suggested.

38. L63-64: Please explain a bit more about how the FLEXPART and CALIPSO data are compared. Specifically, the text refers to percentages of a "layer volume"; is that to say that the CALIPSO layering is converted into a binary present/not present mask and compared to the equivalent binary from FLEXPART? If so, is there a criterion used for the binary CALIPSO masking?

To clarify, we have added the following text:

"Vertical aerosol layer distribution was obtained from CALIPSO v. 4.10 level 2, 5-km merged aerosol and cloud layer data (CALIPSO Science Team, 2016) at 532 nm. These data are collected at 30-m vertical resolution up to 8.2 km, and at 75-m resolution between 8.2-8.5 km. Aerosol-containing profiles were required to be cloud-free and to have cloud-aerosol detection (CAD) scores > 70, indicating high confidence in cloud and aerosol separation. **For each clear-sky polar night profile during our sample period, we noted the fraction of each FLEXPART model vertical layer (0.6 to 1.5 km, 1.5 to 2.5 km, 2.5 to 4 km, 4 to 6 km, and 6 to 8.5 km) that was filled by an observed CALIPSO aerosol layer. From these fractions, ranging from 0 to 1, weighted averages were calculated on a horizontal basis at each altitude level over the Arctic Ocean region (see Figure S6).**"

"Based on the above assumptions, model false negative rates in clean conditions are likely to be highest when CALIPSO aerosol layers are observed in a large fraction of the model altitude layer. Average "clean" FLEXPART vertical layers often contain some CALIPSO-observed aerosol layers within them. **Based on a weighted-average grid analysis of data throughout the study period (Fig. S3a), CALIPSO aerosol layers are present in,** on average, ~19-27% of **FLEXPART** layer volume**s**. The actual BC concentrations of these aerosol layers are unclear."

39. L66: For clarity, recommend "so the fractions estimated" → "so the model false negative fractions estimated"

Changed as suggested.

40. L69: Remove "However". It implies a course change from the prior text but one is not present.

Changed as suggested.

41. L76: For clarity, "sea ice" → "sea ice (the pink line)"

Changed as suggested.

42. L81: For clarity, recommend "detect non-dilute aerosol" → "detect (non-dilute) aerosol"

Changed as suggested.

43. L82: "are likely to be"; shouldn't this be "will be" (?) since it is how you have defined false positives?

Yes, this is correct, we have changed the text to clarify this.

44. L86-87, "These aerosol...": I was looking for a figure to support the statement but it seems that one is not present? If there is, please indicate; if there isn't, please indicate "not shown."

We now indicate Fig. S4.

45. Figure S4: Please rescale the y-axis to cover the range of the bars plotted (i.e., most "polluted" bars exceed the plot range).

Changed as suggested.

46. Table S2: The values in the square brackets are defined in the caption, but what are the values in the parentheses that precede the square brackets? Recommend rearranging to be in column format as the font size to fit in portrait is too small to read easily.

This Table is now Table S1. We have clarified that the round brackets are the bootstrapped 95% confidence intervals for the weighted mean, and have rearranged in column format, as recommended.

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A satellite-based estimate of combustion aerosol cloud microphysical effects over the Arctic Ocean

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Abstract. Climate predictions for the rapidly changing Arctic are highly uncertain, largely due to a poor understanding of the processes driving cloud properties. In particular, cloud fraction (CF) and cloud phase (CP) have major impacts on energy budgets, but are poorly represented in most models, often because of uncertainties in aerosol-cloud interactions. Here we use over 10 million satellite observations coupled with aerosol transport model simulations to quantify large-scale microphysical effects of aerosols on CF and CP over the Arctic Ocean during polar night, when direct and semi-direct aerosol effects are minimal. Combustion aerosols over sea ice are associated with very large (~10 W m^{-2}) differences in longwave cloud radiative effects at the sea ice surface. However, co-varying meteorological changes on factors such as CF likely explain much of this signal - for example, explaining up to 91% of the CF differences between the full dataset and the clean-condition subset. After normalizing for meteorological regime, aerosol microphysical effects have small but significant impacts on CF, CP, and precipitation frequency on an Arctic-wide scale. These effects indicate that dominant aerosol-cloud microphysical mechanisms are related to the relative fraction of liquid-containing clouds, with implications for a warming Arctic.

1 Introduction

Cloud cover has a major influence on surface heating, precipitation, and future climate over the Arctic (Boucher et al., 2013), and may play a role in the enhanced warming over the Arctic compared to lower latitude regions (Södergren et al., 2017), known as Arctic amplification. Aerosols can influence a number of factors relevant to cloud fraction, including cloud droplet number, phase, lifetime, and probability of precipitation (Albrecht, 1989; Coopman et al., 2016; Girard et al., 2005; Lance et al., 2011; Lindsey and Fromm, 2008; Zamora et al., 2017). However, the regional-scale importance of aerosol microphysical processes on CF has been difficult to constrain from observations and models, particularly due to uncertainties in how aerosols affect precipitation and ice nucleation rates (Gettelman, 2015; Morrison et al., 2012; Ovchinnikov et al., 2014). This is mainly due to the complexity of the responses of different types of clouds to different aerosol types and concentrations (Fan et al., 2016; Fu and Xue, 2017; Stevens et al., 2017; Zhao et al., 2018), poorly constrained aerosol concentrations

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(particularly in winter and beneath thick cloud cover), and confounding effects from co-varying meteorology (Gryspeerdt et al., 2016). These uncertainties contribute to the large uncertainties in model CF and CP (de Boer et al., 2011; Cesana and Chepfer, 2012; Chernokulsky and Mokhov, 2012; Kay et al., 2010; Liu et al., 2011; Qian et al., 2012; Stanfield et al., 2014; Zib et al., 2012). To account for the impact of meteorological co-variability on Arctic CF, observations covering large spatial and temporal scales are required, making it difficult to estimate the regional importance of aerosol microphysical effects from *in situ* observations alone. These uncertainties have precluded better constraints on the mechanisms by which aerosols affect cloud microphysics (Morrison et al., 2012) and on general model estimates of their overall importance.

In this paper, we focus on the effects of combustion-derived (i.e., anthropogenic pollution plus smoke) aerosols on clouds. Combustion-derived aerosols are strongly impacted by anthropogenic activity, and tend to dominate columnar mass under high AOD conditions in the Arctic (Xie et al., 2018), although in spring the more well-mixed mineral dust can also contribute ~10% to total Arctic AOD levels (Breider et al., 2014; Groot Zwaaftink et al., 2016). Combustion aerosols have pronounced effects on Arctic cloud microphysical and radiative properties (e.g., Carrió et al. (2005); Coopman et al. (2016, 2018); Earle et al. (2011); Garrett et al. (2004); Jouan et al. (2014); Lubin and Vogelmann (2006); Tietze et al. (2011); Zamora et al. (2016, 2017); Zhao and Garrett (2015)). Their cloud impacts are likely to be particularly large during winter and spring, when they are transported to the Arctic most efficiently, and when precipitation is reduced, causing a peak in aerosol abundance at many remote Arctic ground stations known as "Arctic Haze" (Barrie, 1986; Croft et al., 2016; Quinn et al., 2007; Stohl, 2006). However, so far it has been challenging to assess their cloud effects on the Arctic region as a whole, due to large cloud model uncertainties, spatial/temporal observation limitations, and difficulties obtaining some remote sensing information at high latitudes.

Here we provide the first observation-based constraint on combustion aerosol microphysical effects on total nighttime CF over the Arctic Ocean region within the spectrum of current-day aerosol and meteorological conditions. Estimates of combustion aerosol microphysical effects on operational, satellite-defined CF are obtained from two years of remote sensing data and output from the FLEXPART Lagrangian particle dispersion model (Stohl et al., 1998, 2005). We use the model to provide estimates of how clean or polluted the observed air masses were. We then identify average cloud property differences over sea ice and open ocean regions between the full dataset (hereafter referred to as "all conditions") and a subset where combustion aerosols are at clean background levels ("clean conditions"). By comparing clean to all conditions, as opposed to comparing clean to non-clean or polluted-only conditions, our estimates account for the relatively high frequency of clean and low-aerosol conditions, and reflect the true microphysical effects of combustion-derived aerosols over sea ice and open ocean regions more accurately. By averaging millions of observations after stratifying them by temperature and relative humidity, we minimize confounding effects of local and large-scale meteorological co-variability. A major strength of this method, which depends primarily on direct observations, is that it requires no detailed

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parameterization of the fraction of cloud-active aerosols or underlying microphysical mechanisms to constrain the importance of large-scale aerosol microphysical effects on Arctic clouds.

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2 Methods

Tropospheric cloud data gathered from CloudSat and CALIPSO data during polar night over the Arctic Ocean were collected from 1 January 2008 to 7 December 2009, between 60 and 82° N and 0.6-8.5 km above sea level. These years had typical moisture fluxes and total cloud fractions compared to other recent years (Boisvert and Stroeve, 2015; Kay and L'Ecuyer, 2013). We focused on observations during polar night (solar zenith angle, SZA, > 90°) mainly to isolate indirect effects of aerosols on clouds as much as possible from confounding direct and semi-direct aerosol radiative effects. The nighttime focus also reduces uncertainties from any residual diurnal changes in cloud fraction, and is associated with a better lidar signal-to-noise ratio (used for aerosol transport model validation, see Supplemental Data). Data from all months meeting the above criteria were included except those between May and July. These were excluded to avoid geographic bias in the analysis, as the few nighttime data that were available during this period tended to occur mainly at the lowest latitudes. Clouds below 0.6 km were not assessed due to near-ground uncertainties in the CloudSat and CALIPSO data (de Boer et al., 2009; Liu et al., 2017).

Oceanic areas were determined by ETOPO1 Bedrock GMT4 data (Amante and Eakins, 2009). Oceanic clouds were separated into open ocean and sea ice regions following Zamora et al. (2017): for each profile, the corresponding monthly fractional sea ice cover was determined from the NOAA/NSIDC Climate Data Record of Passive Microwave Sea Ice Concentration, version 2 (Meier et al., 2013; Peng et al., 2013), and samples associated with > 80% or < 20% monthly sea ice fractions were classified as being over sea ice or open ocean, respectively.

2.1 Aerosol transport model

Passive remote sensors provide no aerosol data at night, and do not provide reliable aerosol data over bright sea ice or under clouds. Active lidar signals often get attenuated in clouds. Moreover, active sensors such as CALIPSO cannot always detect dilute aerosols, even in conditions with the highest lidar sensitivity (i.e., above clouds at night (Zamora et al., 2017)). Therefore, the presence of combustion aerosols for comparison to the satellite cloud data was estimated with vertically-resolved modeled black carbon (BC) aerosol estimates from the FLEXible PARTicle dispersion model (FLEXPART) (Stohl et al., 1998, 2005). Here, FLEXPART was driven with meteorological analysis data from the European Centre for Medium-Range Weather Forecasts (ECMWF) at a resolution of 1˚ longitude and 1˚ latitude. Anthropogenic and biomass burning BC emissions were based on the ECLIPSE (Evaluating the CLimate and Air Quality ImPacts of ShortlivEd Pollutants) (Stohl et al., 2015) and GFED (Global Fire Emission Database) (Giglio et al., 2013) inventories, respectively. Model output was produced in five vertical layers between 0.6-1.5, 1.5-2.5, 2.5-4, 4-6, and 6-8.5 km. Note that the resolution of the meteorological input data is much higher (91 levels) and, as a Lagrangian model, FLEXPART has no discrete resolution for

the particle transport. BC emissions were based on the ECLIPSE emission inventory (Stohl et al., 2015). Note that emission fluxes in the model rely on inventories of emission factor measurements that partially include thermooptical measurements, which may not always completely differentiate between BC and "brown" or organic carbon (BrC or OC) (Russell et al., 2014; Samset et al., 2018). Further details on the model and its configuration can be found in Zamora et al. (2017).

FLEXPART is widely used, and is well-validated for the purpose of studying Arctic smoke and pollution transport (Damoah et al., 2004; Eckhardt et al., 2015; Forster et al., 2001; Paris et al., 2009; Sodemann et al., 2011; Stohl et al., 2002, 2003, 2015; Zamora et al., 2017). FLEXPART BC is used in this study as a proxy for all combustion aerosols, because they very often contain BC, although in somewhat different fractions. The association of high levels of modeled BC with CALIPSO aerosols in general (see Zamora et al. (2017)) indicates that modeled BC is a fairly good proxy for strong (CALIPSO-detectable) aerosol layers during polar night, even though some local sources of combustion aerosols (Creamean et al., 2018; Maahn et al., 2017) might not be included in the model. Model comparisons to CALIPSO aerosol data in the study region also indicate that model-identified clean conditions (BC < 30 ng m^{-3}) are associated with significantly lower levels of CALIPSO aerosol layer presence relative to average or polluted conditions (see supplement for further details).

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2.2 Cloud remote sensing observations

Cloud fraction is operationally defined based on the CloudSat algorithm, in CloudSat products available at a vertical resolution of 250 m. Cloud base and top heights were originally obtained from the 2B-GEOPROF-lidar product (Mace et al., 2009; Mace and Zhang, 2014), and the resulting cloud geometric thickness information was used to obtain a profile of vertical cloud fraction at FLEXPART-vertical resolutions for each observation point. All vertical cloud fraction profiles (numbering 10,422,219 total profiles over the Arctic Ocean) that fell within 12.5 km^2 stereographic projection grid cells (Cavalieri et al., 2014) were then averaged together. The gridded observations over sea ice (open ocean) include 15,999 (31,978) grid cells from fall, 43,687 (24,008) grid cells from winter, and 38,793 (15,289) grid cells from spring, with the observation numbers being a function of sea ice extent and length of darkness periods during each season.

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Above 1 km, the 2B-GEOPROF-LIDAR product is similar to or better than ground-based observations, but cloud fraction can be underestimated by up to ~20% below ~1 km (Liu et al., 2017), indicating that cloud detection uncertainties in this study's lowest vertical bin (0.6-1.5 km) are higher there than in other altitude ranges.

CloudSat and CALIPSO do not sample north of 82°. The lack of data within this "pole hole" might mean that those sea ice regions are not well represented in this study. It is unclear how well the data outside the pole hole approximate the data inside it, as this region is the coldest part of the Arctic, and probably also contains some of the cleanest parts with respect to aerosols. Also note that thin-ice-cloud identification is particularly prone to errors over the Arctic, due in part to the widespread occurrence of sub-visible diamond dust and blowing snow. Additionally, the CloudSat radar can sometimes

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mistake precipitation for clouds (de Boer et al., 2009), which can be particularly problematic under optically thick clouds that completely attenuate the CALIPSO lidar signal, and prevent lidar data from being collected below-cloud.

Cloud precipitation presence and phase were obtained from the CloudSat 2B-CLDCLASS-LIDAR version R04 (Wang, 2013). This product captures precipitation with high confidence (Hudak et al., 2008). Phase determination has also been validated favorably at high latitudes (Barker et al., 2008), except that in some cases the radar can misclassify small liquid droplets as ice particles (Noh et al., 2011; Van Tricht et al., 2016). CloudSat may also fail to observe some ice and mixed-phase clouds below 1 km (Liu et al., 2017), suggesting higher uncertainties in cloud phase as well in the lowest vertical bin of this study. Here, cloud phase certainty values were required to be > 5, indicating a higher confidence in phase classification. If a FLEXPART-resolution vertical bin contained clouds of different phases, that bin was excluded from phase-related portions of the analysis. As with CF data, nearby cloud precipitation and phase profile data were averaged within 12.5 km^2 grids at each altitude level prior to analysis.

Estimates of the longwave cloud radiative effect at the bottom-of-the-atmosphere (CRE_{BOA}) were obtained from the CloudSat 2B-FLXHR-LIDAR product, version R04 (Henderson et al., 2012; L'Ecuyer et al., 2008). This product is based on combined CloudSat, CALIPSO, and MODIS observations and time-coincident European Centre for Medium-Range Weather Forecasts (ECMWF) of atmospheric humidity, temperature and sea surface temperature, fed into the BugsRad two-stream, plane- parallel, doubling-adding radiative transfer model, following Henderson et al. (2012). Previous work shows that this product can severely underestimate downwelling LW radiation due to misclassification of small supercooled water as ice particles (Van Tricht et al., 2016) leading to uncertainties in the absolute values of CRE_{BOA}. Here, we primarily focus on relative differences in CRE_{BOA} between two subsets of data: those with high and low modeled BC values. The uncertainty due to misclassification of small particle phase is similar in both subsets of data, which are collected over the same surfaces and years, allowing for meaningful comparisons to be made between the two datasets despite uncertainty in the absolute values.

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2.3 AIRS observations

Air mass temperature and relative humidity at pressure levels ranging from 1000-250 hPa were obtained from the Atmospheric Infrared Sounder (AIRS) level 3 version 6 dataset (Susskind et al., 2014) on the descending orbit (collected at 1:30 am local time). The AIRS instrument provides quality controlled, accurate daily observations over the full study area, including during nighttime conditions, and is validated for use over the Arctic (Boisvert et al., 2015). Data are available in most cloud conditions, although data are not available in completely cloud-covered conditions. Level 3 data, which average observations over a 1x1^o horizontal grid and report at 20 vertical pressure levels, are used instead of level 2 data to obtain closest approximate T and RH data when cloud fractions are high. Errors in this product are highest at larger cloud fractions

and below optically thick clouds. For comparison to other datasets in this study, AIRS data were averaged into the coarser FLEXPART model vertical resolution.

2.4 Data analysis

Differences in relative humidity, temperature, and 12.5 km^2 gridded CF ($\overline{\mathrm{dRH}}$, $\overline{\mathrm{dT}}$, and $\overline{\mathrm{dCF}}$, respectively) between all ($\overline{\mathrm{RH}}$, $\overline{\mathrm{T}}$, and $\overline{\mathrm{CF}}$, respectively) and clean ($\overline{\mathrm{RH_c}}$, $\overline{\mathrm{T_c}}$, and $\overline{\mathrm{CF_c}}$, respectively) conditions were calculated over sea ice and open ocean regions as follows:

(1) $\overline{dRH} = \overline{RH} - \overline{RH_c}$

(2) $\overline{dT} = \overline{T} - \overline{T_c}$

(3) $\overline{dCF} = \overline{CF} - \overline{CF_c}$

In a process conceptually fairly similar to previous work (Chen et al., 2014) (see Figure 1 as an example), spatially gridded CF and BC data in all and clean (BC < 30 ng m^{-3}) conditions were sorted into 5% relative humidity bins and 5 ^{o}C temperature bins, and then the differences in all minus clean conditions were compared within each T-RH bin ($dCF_{T,RH}$ and $dBC_{T,RH}$, respectively). We then compared the differences in all minus clean conditions within each T-RH bin for the change in cloud fraction ($dCF_{T,RH}$) black carbon ($dBC_{T,RH}$), cloud phase ($dCP_{T,RH}$) and precipitation frequency ($dpptn_{T,RH}$). Data were analyzed separately over sea ice vs. open ocean, and within different altitude layers.

Stratifying by similar T and RH conditions isolates some of the systematic BC co-variability with T and RH, helping clarify the BC aerosol impact on cloud fraction. The 5 ^{o}C T and 5% RH bin increments were chosen to balance the benefits of larger sample sizes against the drawbacks of reduced bin representativeness at smaller bin sizes. Data plotted at larger bin increments resulted in similar trends (data not shown).

The estimated microphysical impact of combustion aerosols on total CF over the Arctic Ocean during polar night is calculated from the mean $dCF_{T,RH}$, weighted by the number of 12.5 km^2 grids containing observations falling within each RH and T bin, abbreviated as $\overline{\mathrm{dCF}}_{\mathrm{T,RH}}$. Averaging over sea ice and open ocean regions helps reduce the effects of horizontal winds on factors such as aerosol advection, which can co-vary on local scales with aerosols (Engström and Ekman, 2010; Nishant and Sherwood, 2017). That, in combination with accounting for variations in the T and RH data, enables us to capture several key meteorological parameters that might influence cloud fraction. However, there are no reliable space-borne measurements for vertical velocity, which might also co-vary systematically with BC on a regional scale, and

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meteorological reanalyses of large-scale vertical motion over the wintertime Arctic are not well-validated (Jakobson et al., 2012). Our focus on nighttime data over the flat ocean surface likely reduces effects from solar-heating-driven vertical motion, but the full uncertainty from this parameter cannot be accounted for here. For example, if cold aerosol-containing continental air is routinely advected over warmer open ocean areas, that could induce systemic convection (Serreze and Barry, 2005) that might not be fully captured by T and RH. To provide at least some generalized grouping of clouds likely to be influenced by different large-scale vertical motion, we analyzed altitude layers and surface types (sea ice and open ocean) separately.

3 Results

3.1 Aerosol microphysical effects on cloud fraction

Systematic regional co-variability of aerosols and meteorological factors must be accounted for in order to avoid overestimating aerosol impacts on clouds (Coopman et al., 2018; Gryspeerdt et al., 2016). To illustrate this point, Figure 2 shows the longwave CRE_{BOA} for the upper and lower quartiles of FLEXPART model column BC concentrations, calculated during the entire study period. The upper and lower quartile ranges of column BC levels are associated with very large (~10 W m^{-2}) differences in median longwave CRE_{BOA} over sea ice (Fig. 2). However, when we compare the median relative humidity and temperature profiles with column BC levels in the upper quartile over sea ice (Fig. 2f, red lines) and open ocean (Fig. 2g, red lines) to the lower quartile profiles (blue lines, same figures), it is clear that column BC levels over sea ice are also associated with noticeable differences in median relative humidity and temperature profiles (Fig. 2f). Small differences in lower tropospheric stability (Fig. 2e), defined as the difference in potential temperature between 700 and 1000 hPa, are also observed. These meteorological factors strongly affect CF and CP, which in turn help drive CRE_{BOA}. As a result, aerosol microphysical effects may contribute to only a fraction of the CRE_{BOA} differences shown in Figure 2.

To help better understand co-varying meteorological effects on CF specifically, we assessed a generalized additive model (GAM) (Hastie and Tibshirani, 1990) of the $\overline{dRH}$, $\overline{dT}$, and $\overline{dCF}$ data at each vertical level, season, and surface type (Table 1). Seasonal differences in light, sea ice extent, and BC levels led to some sample number differences for sea ice and open ocean at different times of the year (Table 2). In the GAM, the seasonal values in Table 1 were weighted equally to represent the equal periods of the year being sampled.

The GAM suggests that co-varying differences in $\overline{dRH}$ and $\overline{dT}$ by themselves can explain up to 91% of the variability in $\overline{dCF}$ (as measured by deviance, a statistic similar to variance (Jorgensen, 1997)). Because aerosols can co-vary with T and RH (e.g., because polluted air masses are more likely to have recently resided near the continental surface than clean air masses), aerosols could be responsible for some of this explained variability even without being explicitly included in this GAM. For reference, a GAM based only on $\overline{dBC}$ explained up to 40% of $\overline{dCF}$ variability. Thus, the 91% value is an upper estimate of

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the $\overline{dRH}$ and $\overline{dT}$ influences. Nonetheless, these findings underscore the importance of interpreting aerosol effects on clouds in the context of co-varying temperature and relative humidity. They also indicate that changes in T and RH of air masses entering the Arctic will likely have a very large influence on observed CF, to a degree that is likely to be much more regionally important than the microphysical effects of the aerosols themselves.

Cloud fraction substantially differed among all and clean conditions for many combinations of T, RH, altitude and surface type (Fig. 1). Estimated aerosol impacts on total CF depend on altitude and surface type, but are fairly consistent among seasons (Figs. S1-S3). At the lowest levels (0.6-2.5 km over sea ice and 0.6-1.5 km over open ocean), weighted mean $dCF_{T,RH}$ ($\overline{dCF_{T,RH}}$) is negative, resulting in an ~6% reduction in CF relative to clean conditions over sea ice (-0.6% over open ocean) (Fig. 3). In contrast, $\overline{dCF_{T,RH}}$ between 4-8.5 km elevation increased by 3-5% over both surfaces, indicating more cloud cover at high altitudes for combustion aerosol influenced clouds compared to clean conditions. Absolute $\overline{dCF_{T,RH}}$ changes over sea ice and open ocean ranged between -1.7% to 0.7% and -0.2 to 1.4%, respectively, depending on altitude. Note that the $\overline{dCF_{T,RH}}$ value is based on all $dCF_{T,RH}$ data, including those from T and RH ranges where $dCF_{T,RH}$ is not significantly different from zero (i.e., as indicated by the white Xs in Figure 1). Including all data avoids biasing the results in favor of the meteorological conditions where $dCF_{T,RH}$ is most observable.

The $dCF_{T,RH}$ and $dBC_{T,RH}$ relationships (Fig. 4) indicate that there was more cloud cover in slightly polluted conditions but less cloud cover at higher $dBC_{T,RH}$ levels (> 10-20 ng m^{-3}) relative to clean conditions. $dCF_{T,RH}$ declined significantly at $dBC_{T,RH}$ > 20 ng m^{-3} within most single altitude layers over sea ice, and over open ocean at 1.5-2.5 km (Fig. 3).

Note that the fall period typically has cleaner and warmer conditions compared to winter and spring (Table 1), which tend to occur more heavily on opposite sides of the scatter plots for each altitude layer in Figure 4. Thus, any large, systematic differences in the vertical winds between fall and spring could influence the outermost points within individual altitude layers, and it is not easy to control for this effect. However, the trends among altitude layers show that $dCF_{T,RH}$ is essentially identical over sea ice and open ocean at low $dBC_{T,RH}$ values, which occur mostly at high altitudes. Also, $dCF_{T,RH}$ changes at high $dBC_{T,RH}$/low altitude are more observable over sea ice (Fig. 4), where lower tropospheric stability was greater and temperatures were colder (Figs. 2e-g). Previous studies have also observed more apparent aerosol microphysical effects under more stable conditions in the Arctic (Coopman et al., 2018; Zamora et al., 2017). Possible reasons for the disparate behavior at different altitudes are discussed below.

3.2 Aerosol microphysical effects on cloud phase partitioning

Weighted mean differences in CP partitioning between all minus clean conditions within the same T and RH bins ($\overline{dCP_{T,RH}}$) are discussed for clouds between 0.6-4 km, because clouds at higher altitudes occur mostly in the ice phase (Fig. 5a)

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(Devasthale et al., 2011a; Liu et al., 2017). Over sea ice between 0.6-4 km, all air masses contained a higher relative fraction of ice phase clouds (IPCs) and a lower relative fraction of liquid phase clouds (LPCs) and mixed phase clouds (MPCs) relative to all air masses (Fig. 3). This effect was significant up to 4 km (paired Wilcoxon rank test (Sokal and Rohlf, 1995), $p < 0.05$), except in LPCs between 2.5-4 km, where lower sample numbers might obscure any changes. Changes in phase partitioning over the sea ice region varied between -4.2 and 6.5%, depending on altitude and phase (Fig. 3). From Figure 3, over sea ice between 1.5-2.5 km, the relative contributions of LPCs and MPCs were significantly lower at high $dBC_{T,RH}$ levels (>20 ng m^{-3}), whereas that of IPCs was significantly higher. No significant relationships with $dBC_{T,RH}$ were observed above or below that altitude, although higher BC and CP uncertainties near the surface might mask weak signals in that altitude range. The reduction in liquid-containing clouds at higher $dBC_{T,RH}$ levels over sea ice is consistent with a "glaciation effect" (Lohmann, 2002), whereby increased presence of aerosols leads to ice particle formation and cloud dissipation, as observed in section 3.1.

Over open ocean, significant changes in $\overline{\mathbf{dCP_{T,RH}}}$ were observed less frequently (Fig. 3), and they tended to be smaller than over sea ice (absolute values < 2%). The relative fraction of liquid clouds was reduced between 0.6-1.5 km (Fig. 3), where LPC fractions were highest (Fig. 5a). However, unlike over sea ice, the relative fraction of MPCs over open ocean increased (though not significantly so between 1.5-2.5 km), and that of IPCs decreased (significant only between 2.5-4 km). The reason for the different effects on ice-containing clouds over sea ice and open ocean is unclear, although the higher temperatures may play a role.

3.3 Aerosol microphysical influences on precipitation frequency

Differences in precipitation frequency, $dpptn_{T,RH}$, reflect aerosol microphysical impacts on: 1) the frequency of precipitation within a specific air volume, and 2) the relative likelihood of individual cloud phases within that air volume to be precipitating. We analyse the difference in precipitation frequency; however, an analysis of total precipitation amounts or precipitating particle microphysics is beyond the scope of this study.

Based on weighted mean $dpptn_{T,RH}$ values ($\overline{dpptn_{T,RH}}$), estimated aerosol microphysical effects on regional precipitation frequency were small but significant at many altitudes (Fig. 3, Fig. S4). In all air mass conditions, precipitation frequency was 1.2-3.1% higher below 6 km over open ocean and below 1.5 km over sea ice relative to clean conditions, depending on altitude (Fig. 3). In contrast, clean clouds between 6-8.5 km over open ocean were slightly more likely to be precipitating ($\overline{dpptn_{T,RH}}$ ~ -1%).

Over sea ice, ~94% of MPCs were present below 4 km (Fig. 5a). In these MPCs, $\overline{dpptn_{T,RH}}$ was positive (~ +1%), indicating slightly more frequent precipitation on average in all vs. clean MPCs (Fig. 5b). Significant differences between all and clean

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conditions were not observed for $\overline{\text{dpptn}_{T,RH}}$ in IPCs or LPCs over sea ice, except for a slight (-0.4%) reduction in $\overline{\text{dpptn}_{T,RH}}$ in ice clouds at 6-8.5 km (Fig. 5b). However, significant rank correlations (Kendall's tau coefficient = 0.3, $p < 0.05$) indicate that higher $dBC_{T,RH}$ values were associated with slightly more frequent IPC precipitation over sea ice between 0.6-1.5 km (also see Fig. S5). We observed no strong link between $dBC_{T,RH}$ and $dpptn_{T,RH}$ at other locations/altitudes.

As over sea ice, MPC $\overline{\text{dpptn}_{T,RH}}$ was slightly positive (≤1%) below 4 km over open ocean (Fig. 5b), indicating slightly more MPC precipitation in all vs. clean conditions. IPC $\overline{\text{dpptn}_{T,RH}}$ was also slightly positive between 1.5-4 km, whereas liquid cloud $\overline{\text{dpptn}_{T,RH}}$ was slightly negative between 1.5-4 km. Between 6-8.5 km over open ocean, the $\overline{\text{dpptn}_{T,RH}}$ in MPCs was slightly negative at ~-1% (Fig. 5b).

Based on single or small cloud samples, others have observed decreased precipitation probability with increased aerosol concentrations in Arctic MPCs (Lance et al., 2011; Morrison et al., 2008). It is not entirely clear why the large-scale, regional trends observed here appear to be opposite these smaller-scale *in situ* observations, but recent work indicates that aerosols might influence ice content of the clouds, and thereby affect precipitation (Fu and Xue, 2017; Norgren et al., 2018; Zamora et al., 2017) and potentially CF. Perhaps lower total CF below 4 km leads to less frequent precipitation in these air volumes over the Arctic. The higher MPC precipitation probability and lower MPC cover (as indicated by reduced MPC relative fraction of total CF) at higher aerosol concentrations support this hypothesis.

4 Discussion – Potential aerosol microphysical mechanisms

Specific aerosol-cloud microphysical mechanisms are difficult to identify with confidence from space-borne measurements alone, but some possibilities can be explored. At high altitudes (6-8.5 km) over sea ice, $\overline{\text{dCF}_{T,RH}}$ was higher and $\overline{\text{dpptn}_{T,RH}}$ was lower in all vs. clean conditions (Fig. 3), supporting the hypothesis that aerosols are modifying cloud properties on a regional (i.e., sea ice and open ocean) scale at these altitudes, even though the net changes were relatively small. These modifications to predominantly IPCs at high altitude likely involve aerosol effects on ice crystal formation or properties. Oreopoulos et al. (2017) similarly reported global-scale increases in ice CF with higher aerosol concentration in their CR1 cases (typically high ice clouds of small optical thickness over the tropics), which was linked to reduced ice cloud effective radius.

There are several mechanisms by which aerosols might modify ice crystal number or size that could cause the observed changes in precipitation and CF in the 6-8.5 km range. Although BC itself is not a good source of INPs (Vergara-Temprado et al., 2018), combustion aerosols associated with BC might act as ice nucleating particles (INPs) (Kanji et al., 2017) at the extreme cold temperatures found at high-altitude Arctic polar night. This could potentially lead to smaller, more numerous ice particles that precipitate less (Lohmann and Feichter, 2005), in line with our observations, although some models suggest

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that INPs may instead lead to larger ice crystals in cirrus clouds compared to homogeneous freezing (Heymsfield et al., 2016). Alternatively, combustion aerosols might reduce the efficiency of pre-existing INPs through the "deactivation effect" (Archuleta et al., 2005; Cziczo et al., 2009). Reduced ice crystal formation rates could then lead to more frequent air mass saturation with respect to liquid water, more water droplets that freeze homogeneously, and smaller, more numerous ice particles, and less precipitation (Girard et al., 2013) as observed here. This effect could lead to enhanced total CF over the Arctic (Du et al., 2011). Although absolute humidity within the different T-RH bins between 6-8.5 km is not systematically related to higher $dBC_{T,RH}$ levels as one might expect with the deactivation effect, it is possible that pre-sorting the data by 5% RH bins to reduce the impacts of meteorological co-variation could make evidence for this effect more difficult to observe. Therefore, it is difficult to say whether this study is consistent with the deactivation effect hypothesis, but it does not preclude it.

The specific microphysical mechanisms affecting lower altitude clouds are more challenging to identify without in situ data due to the high prevalence of liquid-containing clouds (Fig. 5a). Combustion aerosols can affect precipitation rates by changing droplet numbers and sizes, and thereby possibly collision and coalescence (Albrecht, 1989), riming (Lohmann and Feichter, 2005; Saleeby et al., 2009), or freezing (Bigg, 1953). If these aerosols affect INP levels, they could also affect ice nucleation rates and ice particle concentrations, leading to MPC and LPC glaciation, enhanced precipitation, and reduced cloud cover (the "glaciation effect"). Our observations do support the possibility of a glaciation effect, because once T and RH co-variability are accounted for, all air masses at low altitudes (0.6 to 1.5-2.5 km) have lower CF compared to clean conditions. They also have more frequent precipitation in IPCs at high $dBC_{T,RH}$, and a higher relative fraction of IPCs over sea ice and MPCs over the warmer open ocean. Each of these changes is significantly different between low and high $dBC_{T,RH}$ concentrations at a variety of altitudes and surface types (Fig. 3), suggesting that aerosols may help convert liquid droplets to larger ice particles that precipitate out and reduce CF in lower altitude clouds.

These observations are in-line with other studies indicating that aerosols can dissipate Arctic MPCs (Fu and Xue, 2017; Norgren et al., 2018) and increase their precipitation (Kravitz et al., 2014; Morrison et al., 2011). Assuming they act as INPs, various modelling studies and a remote sensing study also suggest that aerosols can reduce liquid water path or supercooled water frequency (Fan et al., 2009; Morrison et al., 2011; Ovchinnikov et al., 2014; Pinto, 1998; Tan et al., 2014). The observations over sea ice contrast with some model predictions that MPCs should increase in more polluted conditions through the deactivation effect (Du et al., 2011; Girard et al., 2005, 2013). They also contrast with a previous remote sensing study (Zamora et al., 2017) indicating that thin and predominantly liquid Arctic Ocean clouds are more likely to be the liquid phase at high BC concentrations. However, the clouds in that study may not be fully comparable, as they constitute only ~5% of the cloud types in this study. Note that shortwave processes might alter how aerosols impact mixed phase CF during daytime (Solomon et al., 2015), and any such effects would not be observed in the current, nighttime study. Changes in higher altitude clouds might also change underlying cloud properties through a seeding effect, which could impact cloud

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properties at lower altitudes.

5 Summary and conclusions

Upper quartile levels of total column BC (a proxy for combustion aerosols) are associated with very large (~10 W m^{-2}) differences in longwave cloud radiative effects at the sea ice surface compared to the lower quartile column BC levels. However, relative humidity in particular over sea ice is very different in the two aerosol conditions, which likely drives much of the CRE_{BOA} differences in Figure 2. The CRE_{BOA} is impacted to a high degree by CF. We found that AIRS-derived temperature and relative humidity predicted up to 91% of the observed differences in sea ice and open ocean CF between all and clean conditions in the altitude ranges of interest in this study (0.6-8.5 km). These observations indicate that changes in T and RH of air masses entering the Arctic will likely have a very large influence on observed CF, to a degree that is much more regionally important than the microphysical effects of the aerosols themselves. In line with previous studies (e.g., Gryspeerdt et al. (2016); Coopman et al. (2018)), these results also underscore the need for large sample volumes to identify systematic air mass differences between clean and all conditions, and a way to reduce the confounding effects of meteorological co-variation on these samples. To accomplish this, we analyzed over 10 million profiles across the Arctic Ocean, which were binned into similar T and RH groups. We analyzed the data separately over sea ice vs. open ocean, and within different altitude layers.

In general, combustion aerosol microphysical effects were most observable where the highest aerosol effect would be expected: a) at lower altitudes where aerosol concentrations are often higher (Devasthale et al., 2011b) and b) over sea ice, where atmospheric stability is greater, and aerosol microphysical effects on clouds are less likely to be overwhelmed by meteorological factors such as high vertical velocity. Relative to clean conditions, low clouds over sea ice had ~6% smaller CF and 3% higher precipitation frequency, whereas at high altitudes, CF increased by 4% and precipitation was 2% less frequent. Similar but smaller trends in CF and low-altitude precipitation were observed over open ocean. Below 1.5 km, we also observed a 7% reduction in the LPC and MPC fractions over sea ice, but a slight increase in MPCs relative to LPCs over open ocean. The different effect on MPCs over sea ice and open ocean may be related to the higher temperatures over open ocean, leading to less efficient ice formation, or to some other, as yet unknown, factor. Observations from others (e.g., Chernokulsky et al. (2017); Eastman and Warren (2010)) show that expansion of open ocean areas appear to be connected to changing Arctic Ocean cloud properties. The different cloud responses to aerosols that we observe over sea ice vs. open ocean may provide partial clues into the cause of this behaviour, and into the future impacts of combustion aerosols on the Arctic system in general.

These results are subject to various uncertainties, including possible confounding influences from large-scale vertical motion that is difficult to measure in situ, any systematic model errors in identifying aerosol layers at the right altitude (see supplementary information), and the uncertain relationships between modeled BC and INP and cloud condensation nuclei

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(CCN) concentrations. Associations between $dBC_{T,RH}$ and cloud properties may be difficult to observe given the low BC concentrations at altitudes > 4 km, and may be further masked by complex aerosol-cloud relationships. Despite these uncertainties, significant differences in $dCF_{T,RH}$, $dpptn_{T,RH}$, and $dCP_{T,RH}$ at high $dBC_{T,RH}$ concentrations provide evidence that aerosol microphysical effects were driving the observed patterns, as opposed to some other factor. Furthermore, these observations leave open the possibility that other cloud property relationships with $dBC_{T,RH}$ exist, but are not observable with the available data.

The mechanisms responsible for these changes cannot be fully elucidated from modeling and remote sensing data alone. The observed increases in CF and decreases in precipitation at 6-8.5 km likely involve aerosol effects on ice crystal formation or properties, given that nearly all of these clouds are in the ice phase. These effects might include a deactivation of pre-existing INPs, or conversely an enhancement of INPs by combustion aerosols themselves at the very low temperatures observed at these high altitudes during Arctic polar night. The reduced CF, more frequent precipitation in mixed phase clouds, and reduced relative fraction of mixed (liquid) phase clouds over sea ice (open ocean) seem to point towards aerosols either participating in the conversion of liquid droplets to larger ice particles that precipitate and reduce CF, similar to a glaciation effect, or potentially to their impacts on precipitation in higher clouds, changing underlying cloud properties through a seeding effect. Further focused studies on these mechanisms would be of great interest, along with targeted aircraft measurements of the relevant aerosol and cloud properties, providing greater detail at higher spatial and temporal resolution. To improve quantification of Arctic aerosol-cloud microphysical interactions from space, two major uncertainties also require better quantification: (1) large-scale vertical motion and (2) altitude-resolved aerosol amount and type information. Obtaining more ground-based observations of clouds lower than 0.6 km, which are radiatively significant but not measured well by satellite, is also important.

Data availability. CloudSat data were obtained from http://www.cloudsat. cira.colostate.edu/order-data/, and AIRS data were obtained from https://disc.gsfc.nasa.gov/. For access to the CALIPSO, ETOPO and NSIDC data, see CALIPSO Science Team (2016), Amante and Eakins (2009), and Meier et al. (2013), respectively. The data underlying Figures 1, 2, and 4 are presented in text files in the supplementary material, and the data from Figures 3 and 5 are presented tabularly in Table S1.

Author contributions. LMZ., RAK, and KBH designed the study and the statistical analysis. LMZ processed the satellite data, and AS and SE processed FLEXPART model output. LMZ wrote the manuscript. All authors contributed to revising the manuscript.

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Competing interests. The authors declare that they have no conflict of interest.

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Tables

Table 1. The mean temperature (T, °C), relative humidity (RH, %), black carbon (BC, ng m^{-3}), and cloud fraction (CF, %) observed over the Arctic Ocean study region over sea ice and open ocean during different seasons and altitude levels. Data are shown for all conditions. Also shown are the mean differences between all minus clean conditions ("difference") for T, RH, BC, and CF, referred to as $\overline{dT}$, $\overline{dRH}$, $\overline{dBC}$, and $\overline{dCF}$ in the text.

		Sea ice						Open ocean					
	Altitude	Fall (ASO)		Winter (NDJ)		Spring (FMA)		Fall (ASO)		Winter (NDJ)		Spring (FMA)	
	levels (km)	all	difference	all	difference	all	difference	all	difference	all	difference	all	differen
Temperature (°C)	6-8.5	-48.7	0.0	-53.0	0.0	-51.9	0.1	-45.0	0.1	-49.6	0.0	-49.2	0.1
	4-6	-32.3	0.5	-38.1	0.2	-40.0	0.4	-27.7	0.3	-33.2	0.1	-35.5	0.1
	2.5-4	-21.6	0.5	-27.5	0.1	-29.5	0.1	-15.8	0.4	-21.1	0.2	-23.6	0.0
	1.5-2.5	-15.5	0.5	-21.6	-0.4	-23.4	-0.7	-9.2	0.4	-14.6	0.4	-16.6	-0.2
	0.6-1.5	-11.3	0.4	-17.7	-0.6	-19.6	-1.3	-4.5	0.3	-9.9	0.4	-11.6	-0.3
Relative Humidity (%)	6-8.5	74.6	0.0	65.6	0.4	50.4	1.0	69.0	0.1	63.8	0.2	50.9	0.4
	4-6	61.5	-0.3	62.7	0.4	59.8	-0.6	57.1	-0.1	57.6	0.6	58.5	0.9
	2.5-4	66.7	-0.6	69.3	0.2	66.2	-1.8	61.4	0.2	58.6	1.6	57.1	0.6
	1.5-2.5	76.9	-0.9	74.0	-0.4	68.2	-0.3	74.5	0.2	72.5	1.9	69.1	0.5
	0.6-1.5	85.1	-0.3	78.1	-2.2	70.6	0.2	87.2	-0.3	88.6	1.7	84.4	-0.2
BC (ng m^{-3})	6-8.5	14	1	13	2	19	5	14	2	11	1	17	3
	4-6	17	3	22	6	30	11	17	3	18	3	28	9
	2.5-4	19.8	4	31	13	41	19	19	5	28	10	38	16
	1.5-2.5	20	4	39	21	53	27	22	7	43	23	47	22
	0.6-1.5	17	3	51	31	66	32	22	9	58	34	56	27
Mean CF (%)	6-8.5	15.4	0.2	15.0	0.4	9.1	0.9	19.3	1.0	22.3	0.4	15.3	0.5
	4-6	22.7	0.6	21.8	1.3	16.8	0.1	24.3	0.8	26.0	0.8	22.7	0.7
	2.5-4	29.1	1.0	25.9	1.0	20.8	-2.1	27.9	1.0	28.8	2.2	27.4	0.5
	1.5-2.5	35.0	-0.3	30.3	-0.4	24.0	-3.5	34.3	1.4	39.4	2.1	39.5	0.3
	0.6-1.5	33.3	-0.2	26.9	-2.0	22.0	-3.0	35.0	-0.3	43.0	-0.4	43.6	-0.3

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Table 2. Total profile numbers during each season of the study over sea ice and open ocean regions. Also shown are the percent of samples determined to be clean (BC < 30 ng m^{-3}) at different altitudes. Seasonal differences in sample numbers depend on factors such as light, sea ice extent, and, for clean samples, seasonal variations in BC levels.

		Sea ice			Open ocean		
		Fall (ASO)	Winter (NDJ)	Spring (FMA)	Fall (ASO)	Winter (NDJ)	Spring (FMA)
Total samples		457,504	4,687,541	1,757,034	1,153,806	1,429,840	529,904
	Altitude levels (km)						
	6-8.5	94%	94%	85%	92%	97%	89%
% "Clean" samples	4-6	88%	81%	68%	88%	88%	69%
	2.5-4	85%	65%	54%	83%	70%	59%
	1.5-2.5	83%	51%	39%	81%	54%	50%
	0.6-1.5	88%	46%	27%	83%	50%	48%

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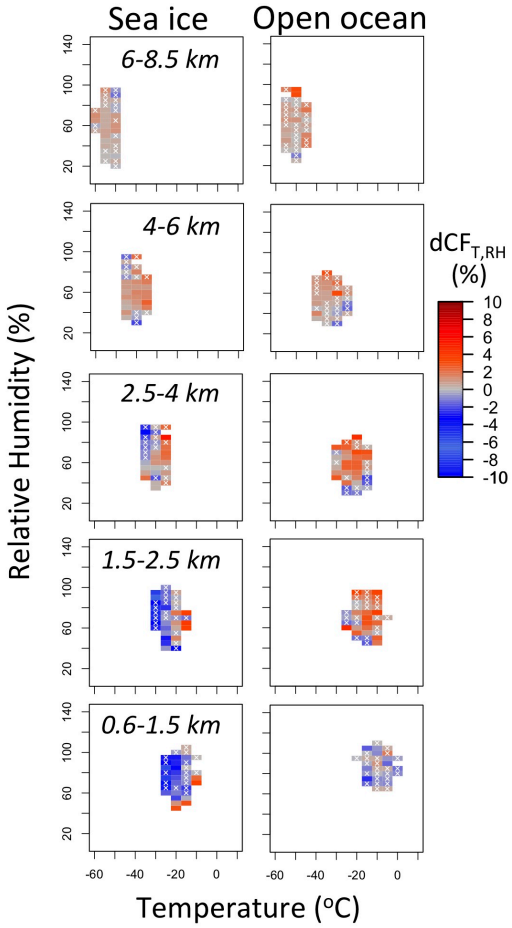

Figure 1: An example of $dCF_{T,RH}$ output at each altitude level. For illustration purposes, here each grid cell represents ≥ 7500 km^2 of gridded observations. Blue and red colors indicate negative and positive $dCF_{T,RH}$, respectively. A white X indicates that the cell value is not significantly different from zero (Wilcoxon rank test, $p < 0.05$). Note that each underlying Wilcoxon rank test has a 5% chance of yielding a false positive indication of statistical significance or an unknown (but likely much higher) chance of yielding a false negative result. Consequently, the distribution of Xs should not be over-interpreted. The number of Xs, however, provides an objective way to test whether the evidence for an effect on the grid as a whole is significant. This is consistently the case; in all panels, individually significant cells numbered more than expected at random (binomial test, $p < 0.001$).

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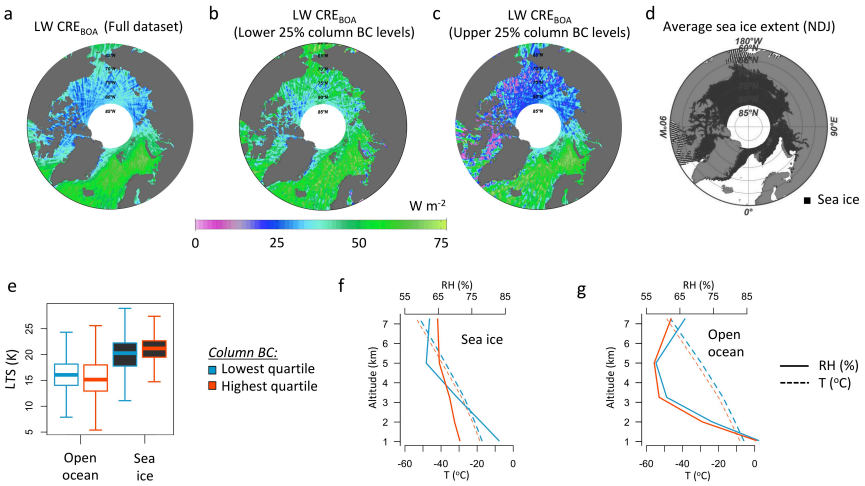
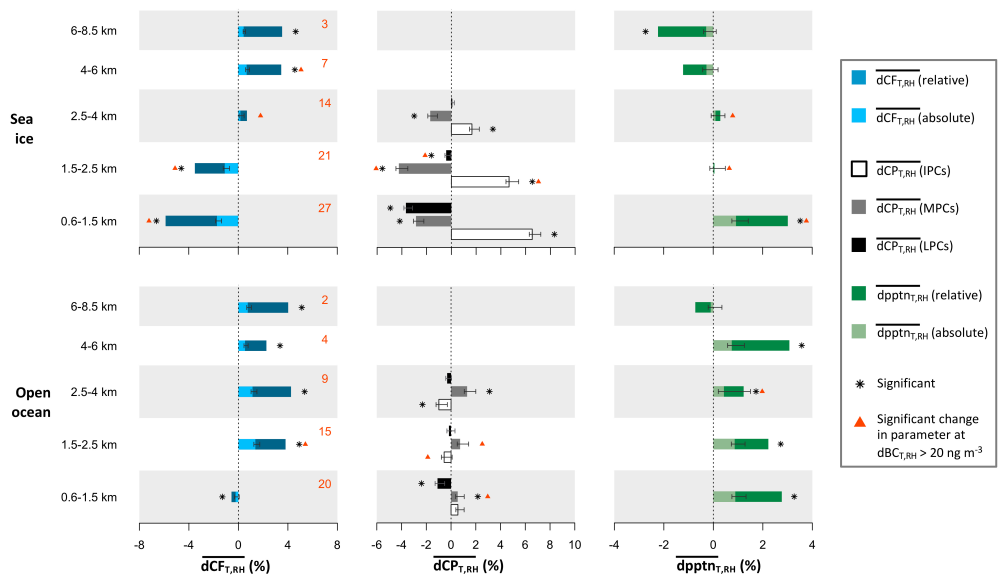


Figure 2: a-c) CloudSat FLXHR-LIDAR longwave cloud radiative effect at the bottom of atmosphere (i.e., surface) (LW CRE$_{BOA}$) during polar night for a) the full dataset, and the subsets of data containing the lower (b) and upper (c) quartiles of modeled column BC concentrations. For reference, d) shows the average winter (November to January) sea ice extent up to 82°N. Also shown are e) boxplots of the lower tropospheric stability (LTS, K), and f, g) the median temperature (T, °C) and relative humidity (RH, %) for the lower (blue) and upper (red) quartile column BC concentrations over open ocean and sea ice. All differences in e-g) are significant ($p < 0.0001$), based on a Wilcoxon rank test.



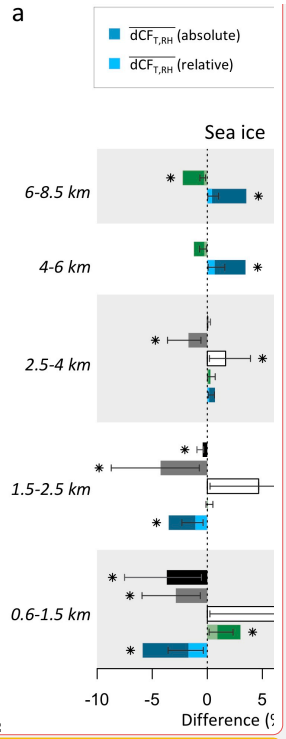


Figure 3: Summary of $\overline{dCF_{T,RH}}$, $\overline{dCP_{T,RH}}$ (up to 4 km, %), and $\overline{dpptn_{T,RH}}$ (%) $\overline{dCP_{T,RH}}$ in different altitude ranges over sea ice and open ocean. For $\overline{dCF_{T,RH}}$ and $\overline{dpptn_{T,RH}}$, different color bars (overlaid, not stacked) show the absolute change within the air volume of interest (darker colors) and the relative percent change with respect to the value found in clean conditions (lighter colors). The $\overline{dBC_{T,RH}}$ values (ng m^{-3}) are presented for each altitude (red, upper right in the left two panels). An asterisk (*) indicates that the differences between all and clean conditions were significant for both relative and absolute values, based on a paired Wilcoxon rank test, $p \leq 0.05$, using T-RH grid cells containing > 800 (400 for $\overline{dCP_{T,RH}}$) 12.5-km^2 gridded observations. Values marked by a red triangle indicate a significant change in the parameter where $dBC_{T,RH} > 20$ ng m^{-3} (Wilcoxon rank test, $p < 0.05$). Error bars show bootstrapped 95% confidence intervals for the weighted mean.

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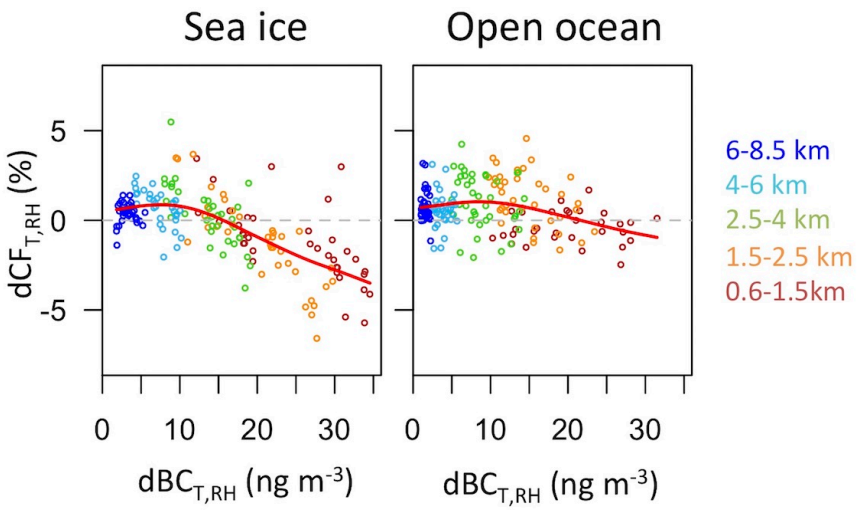

Figure 4: The relationship between dCF$_{T,RH}$ (%) and dBC$_{T,RH}$ (ng m^{-3}) over sea ice and open ocean at different altitude levels (color coded) for the points plotted in Figure 1. The red line is a cubic smoothing spline of the data among all altitudes. In order to avoid obscuring emergent properties of the full dataset, the data include all meteorological conditions, including those where dCF$_{T,RH}$ are not significantly different from zero (as noted by white Xs in Figure 1).

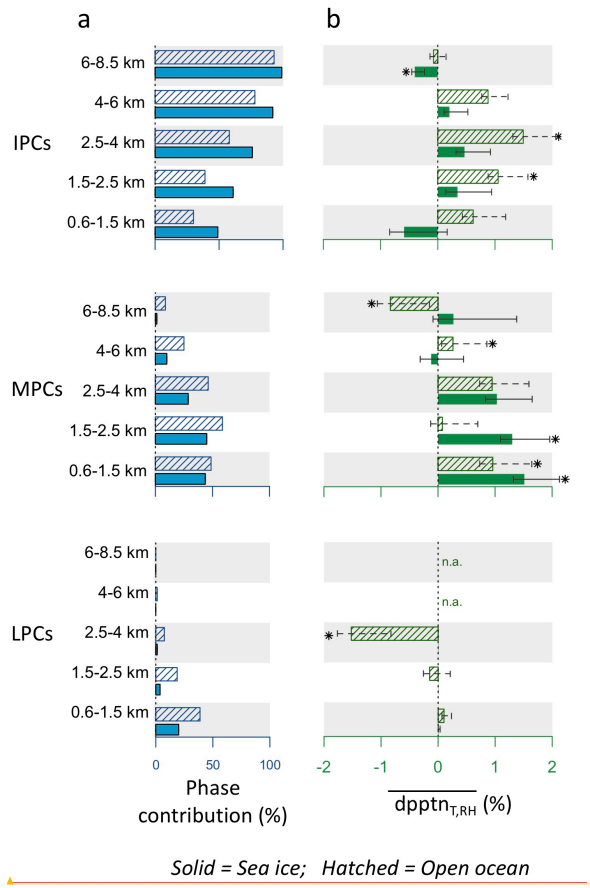

Figure 5: The absolute changes in a) CP distribution (blue), and b) $\overline{dpptn}_{T,RH}$ (green) for IPCs, MPCs, and LPCs over sea ice (solid) and open ocean (hatched) at different altitude ranges. An asterisk (*) indicates that the differences between all and clean conditions were significant for both relative and absolute values, based on a paired Wilcoxon rank test, $p < 0.05$, using T-RH grid cells containing > 800 (400 for $\overline{dCP}_{T,RH}$) 12.5-km^2 gridded observations. Error bars show bootstrapped 95% confidence intervals for the weighted mean.

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Although the focus of this work is on regional signals as a whole, it is interesting to note that in Figure 1 over sea ice up to 6 km, the $dCF_{T,RH}$ response appears to generally become more positive at constant RH with increasing T (and higher absolute humidity), hinting that combustion aerosols in that environment may be more likely to increase CF when more moisture is available, although a similar effect was not observed over open ocean.

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$\overline{dCP_{T,RH}}$ (up to 4 km, %)

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Shown in b) are the absolute changes in $\overline{\mathrm{dpptn}_{\mathrm{T,RH}}}$ (green) and the CP distribution (blue) for IPCs, MPCs, and LPCs over sea ice (solid) and open ocean (hatched) at the same altitude ranges as in a).

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The same data are presented tabularly in Table S2.

Supplemental material

Validation of the aerosol transport model

Remote-sensing and in situ aerosol observations are not available over many Arctic Ocean areas due to harsh sampling conditions, extensive cloud cover, high sea ice albedo, and the long periods of darkness during polar night. To understand as best we can how aerosol microphysics impacts clouds over the Arctic, we used the FLEXPART dispersion model with black carbon as a proxy for combustion aerosols to identify clean (i.e., non-combustion aerosol dominated) conditions in Arctic air masses, so that cloud properties in these conditions could be compared to general observations.

From the limited black carbon (BC) data that are available, the FLEXPART model appears to capture BC aerosol patterns very well over the Arctic (Eckhardt et al., 2015; Stohl et al., 2015; Zamora et al., 2017). Unfortunately, a lack of observations makes it difficult to validate model BC concentrations and spatial distributions over large swaths of the Arctic, particularly in the free troposphere and during polar night. Because spatial biases in the FLEXPART output could influence the meaningfulness of statistical comparisons between different locations, we validated FLEXPART BC output with Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) aerosol layer data, which are the only nighttime aerosol data available regionally over the Arctic.

Vertical aerosol layer distribution was obtained from CALIPSO v. 4.10 level 2, 5-km

merged aerosol and cloud layer data (CALIPSO Science Team, 2016) at 532 nm. These data are collected at 30-m vertical resolution up to 8.2 km, and at 75-m resolution between 8.2-8.5 km. Aerosol-containing profiles were required to be cloud-free and to have cloud-aerosol detection (CAD) scores > 70, indicating high confidence in cloud and aerosol separation. For each clear-sky polar night profile during our sample period, we noted the fraction of each FLEXPART model vertical layer (0.6 to 1.5 km, 1.5 to 2.5 km, 2.5 to 4 km, 4 to 6 km, and 6 to 8.5 km) that was filled by an observed CALIPSO aerosol layer. From these fractions, ranging from 0 to 1, weighted averages were calculated on a horizontal basis at each altitude level over the Arctic Ocean region (see Figure S6).

CALIPSO aerosol profiles were used here for aerosol transport model validation. FLEXPART-derived clean conditions were more frequent at higher altitudes (Fig. 3), in line with previous observations of CALIPSO aerosol distributions (Di Pierro et al., 2013). Major uncertainties arise in this comparison because of the coarse vertical resolution of the FLEXPART model output, and because CALIPSO aerosols are not necessarily equivalent to BC concentrations. As such, it is unclear how observed CALIPSO aerosol layer thickness (measured in meters) would relate to the average BC concentration in an altitude range equivalent to the FLEXPART model's vertical resolution (measured in kilometers). For these reasons, and because we use the aerosol model in this study only to identify clean (BC < 30 ng m^{-3}) air masses, our validation efforts are focused mainly on assessing the likely locations of FLEXPART false negatives (i.e., where observations indicate that aerosol layers have a large impact on model-identified "clean" air masses). Some information on false positives is provided as well.

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To estimate an upper limit of model BC false negatives from CALIPSO aerosol distributions, we make several assumptions. First, we assume that combustion aerosols are the dominant aerosol source over the Arctic during polar night at the altitudes of relevance during this study (0.6-8.5 km). Ground-based data and aerosol transport models indicate that this is a fairly reasonable assumption (Quinn et al., 2002, 2008; Stohl et al., 2002). Marine aerosols are mainly located in the shallow Arctic boundary layer. Mineral dust can be found throughout the Arctic atmosphere. However, although there are some local "sharp" dust plumes at some locations in the fall, wintertime local dust emissions are limited by extensive snow cover, and long-range transport of low-latitude dust is well mixed in the winter and, at moderate concentrations, is nearly omni-present in the Arctic free troposphere (Groot Zwaaftink et al., 2016).

We also assume that the BC contribution to combustion aerosol mass is steady, even though other studies have shown that OC:BC ratios, for example, can vary (Samset et al., 2018). In cases where the OC:BC ratio is higher than on average, FLEXPART would indicate relatively too little aerosol compared to cases with a low OC:BC ratio. Other uncertainties are introduced because CALIPSO cannot always identify dilute aerosol plumes (Kacenelenbogen et al., 2014), and it sometimes misclassifies ice clouds with very small ice particles as aerosol layers (Di Pierro, 2013). Despite these uncertainties, information from this analysis is still very useful because of how poorly the models are validated over large parts of the Arctic, particularly because these models provide the

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only regional estimates of combustion aerosol concentrations over large swaths of the Arctic.

Based on the above assumptions, model false negative rates in clean conditions are likely to be highest when CALIPSO aerosol layers are observed in a large fraction of the model altitude layer. Average "clean" FLEXPART vertical layers often contain some CALIPSO-observed aerosol layers within them. Based on a weighted-average grid analysis of data throughout the study period (Fig. S6a), CALIPSO aerosol layers are present in, on average, ~19-27% of FLEXPART layer volumes. The actual BC concentrations of these aerosol layers are unclear. Previous analysis indicates that CALIPSO misses ~33-36% of very dilute (30-50 ng BC m^{-3}) combustion aerosol layers (Zamora et al., 2017), and so the model false negative fractions estimated in Figure S6a might actually underestimate aerosol layer presence somewhat.

CALIPSO aerosol volume contributions in clean conditions were significantly less at each altitude level than those found in all or polluted (BC >150 ng m^{-3}) conditions (Wilcoxon rank test, $p < 0.05$) (Fig. S7). Most altitudes also had no major clustering of high values in Figure S6a, which provides some confidence that model-identified clean conditions are at least comparable between large regions (e.g., over sea ice and over open ocean). The largest clustering in Figure S6a occurs at the lowest altitude level (0.6-1.5 km), where there was a slightly higher likelihood of false negatives over open ocean compared to sea ice (pink line). The highest overall likelihood of false negatives occurred

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at the highest altitude level (6-8.5 km), but it was not much larger than at other altitudes (Fig. S7).

Previous analysis indicates that in nighttime clear-sky conditions, CALIPSO should detect (non-dilute) aerosol layers in profiles where FLEXPART reports median column BC concentrations greater than 150 ng m^{-3} (Zamora et al., 2017). Model false positive rates are be highest when CALIPSO aerosol layers do not fill a large fraction of the model altitude layer where modeled BC was >150 ng m^{-3} (Fig. S6b). CALIPSO detected aerosol layers nearly all the time during clear nighttime conditions in some portion of the altitude bin where high BC concentrations were predicted. These aerosol layers contributed to 0.3-2.0 times more volume on average, depending on altitude, than in clean conditions (Fig. S7). The likelihood of false positives was highest over the open ocean in the lower two altitude bins (0.6-2.5 km), which might have a small impact on our comparisons between sea ice and open ocean at these altitudes. Note that observed aerosol layers may be present in the model, but could be displaced in altitude (e.g., by a kilometer or two), which could contribute to the apparent false positives or false negatives.

In summary, for FLEXPART to correctly identify clean (i.e., low combustion aerosol) conditions, it needs to be able to correctly simulate the horizontal and vertical distributions of combustion aerosols. As mentioned in the main text, previously conducted model validation studies indicate that FLEXPART has skill in simulating the locations of BC transport over the Arctic. Here, we show that the volume of CALIPSO

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vertical aerosol layers is significantly smaller in model-estimated clean conditions in the vertical column than in all conditions, or in model-identified polluted conditions. This result indicates that FLEXPART also has some skill in the vertical layer prediction of BC aerosols over the Arctic Ocean. Moreover, we observed no major spatial biases in the false negative rates that would preclude the regional comparisons between sea ice and open ocean regions. Together, these findings and previous work support the use of FLEXPART for identifying clean conditions for the purposes of this study.

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Supplementary Figures

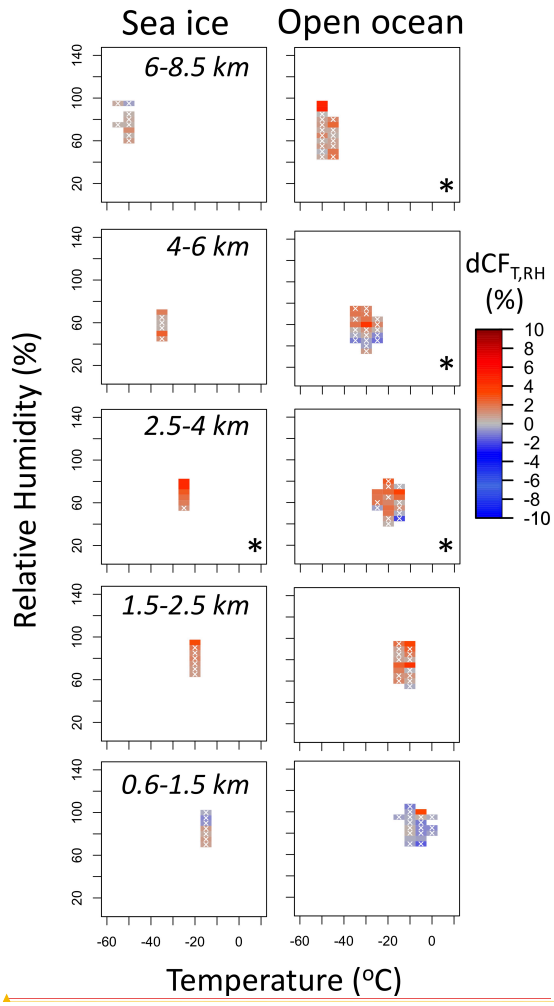

Figure S1. Same as for Figure 1, except for plotted only for Fall (September-November), with panels where individually significant cells numbered more than expected at random (binomial test, $p < 0.001$) marked with an asterisk (*). The shorter time frame results in fewer plotted cells compared to Figure 1, as plotted grid cells are required to each contain ≥ 7500 km^2 of gridded observations for illustration purposes.

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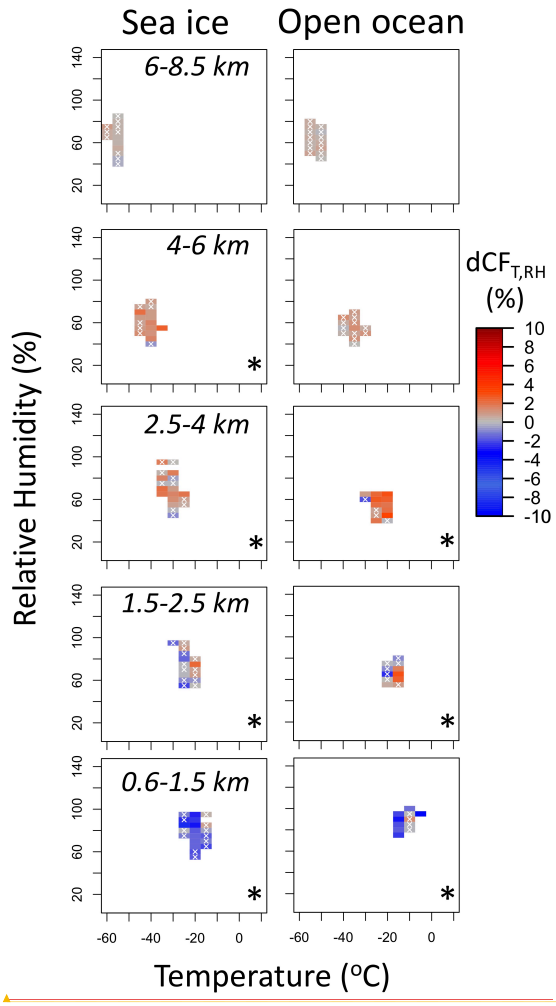

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Figure S2. Same as for Figure S1, except for plotted only for Winter (December-February).

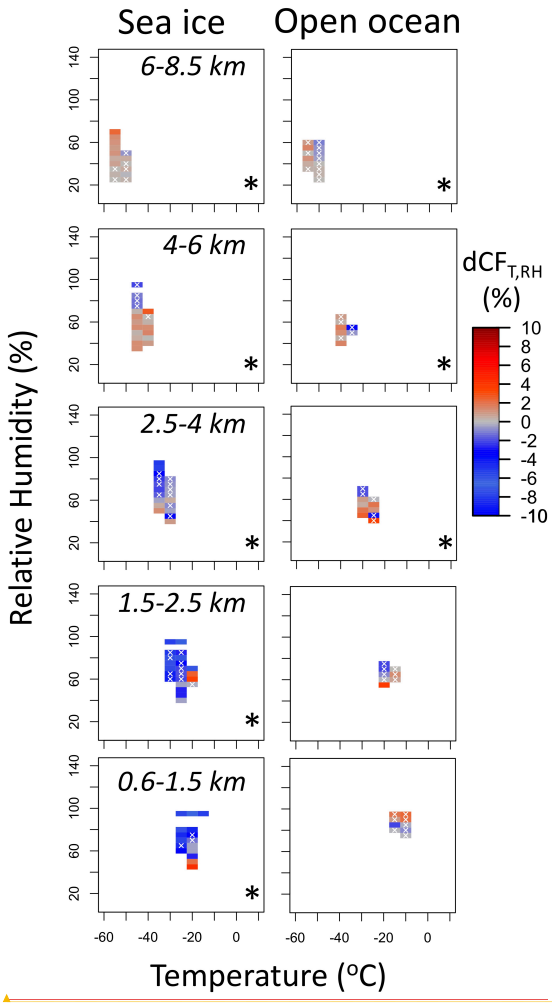

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Figure S3. Same as for Figure S1, except for plotted only for Spring (March-May), and that the open ocean cases plotted above were only required to represent ≥ 5000 km^2 of gridded observations, as opposed to ≥ 7500 km^2. This plotted sample number distinction is purely for illustrative purposes, as the spring open ocean samples contained fewer clean cases for comparison than during the other seasons.

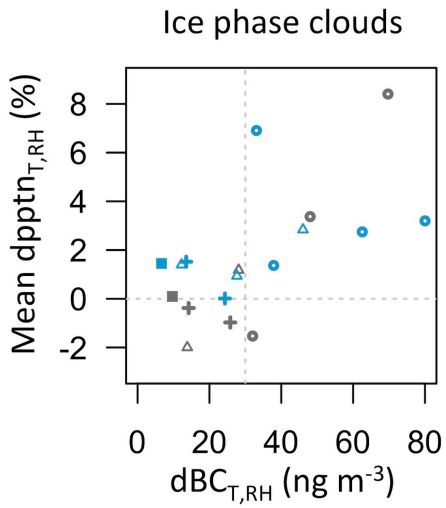

Figure S4. Mean IPC dpptn$_{T,RH}$ as a function of median dBC$_{T,RH}$ values within 10 ng m^{-3} BC increments. Values are shown for data over sea ice (grey) and open ocean (blue) at different altitudes between 0.6 and 6 km (open circles = 0.6-1.5 km, open triangles =1.5-2.5 km, crosses = 2.5-4 km, and filled squares = 4-6 km). In order to reduce the effects of outliers, each plotted data point represents at least 10 separate T/RH bins present in that altitude range, which in turn contain observations from at least 1250 km^2 over the Arctic Ocean. Thin light grey dashed lines indicate zero dpptn$_{T,RH}$ and a dBC$_{T,RH}$ value of 30 ng m^{-3}.

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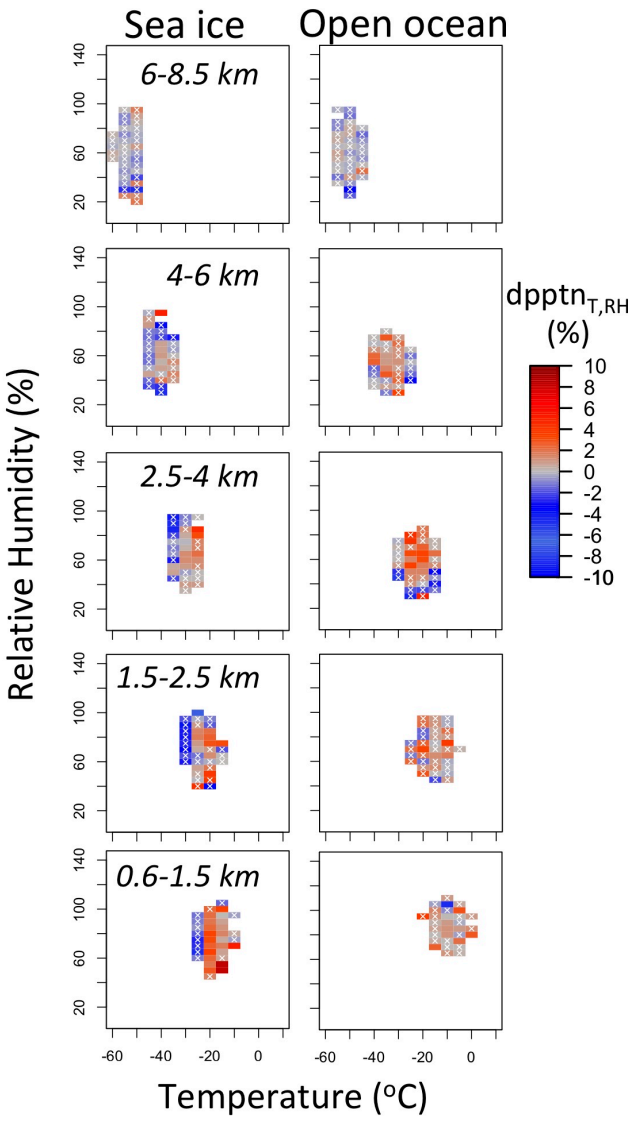

Figure S5. Same as for Figure 1, except for precipitation instead of cloud fraction. Here, individually significant cells numbered more than expected at random (binomial test, $p < 0.001$) at all levels except over open ocean between 1.5-2.5 km (significant at $p < 0.05$), and at 6-8.5 km (not significant).

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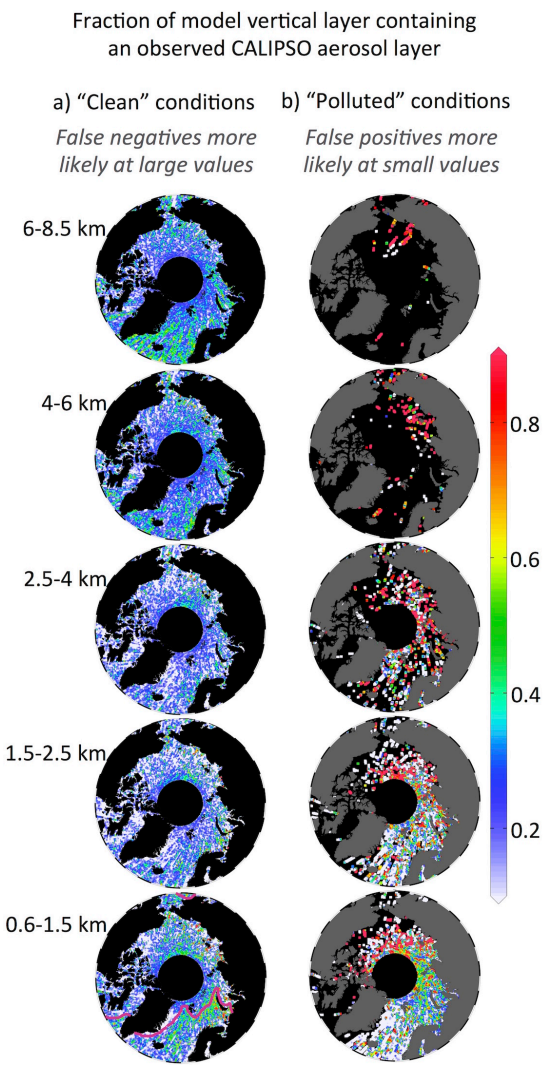

Figure S6. The fraction of different altitude layers in which CALIPSO aerosols were observed for the subsets of data with FLEXPART modeled BC concentrations a) < 30 ng m^{-3} (clean conditions), and b) > 150 ng m^{-3} (polluted conditions). Data are presented on a weighted average grid. Model false negatives are most likely to occur where fractions of the model vertical layer containing CALIPSO aerosols in (a) are high (i.e., closer to red in the color axis), because that indicates that the model predicted clean conditions, but CALIPSO aerosol layers were still frequently observed in that location. Model false positives are most likely to occur where these values in (b) are small (i.e., closer to white), because the model predicted a strong BC aerosol presence, but aerosol layers were not frequently observed by CALIPSO at that location. This interpretation is based

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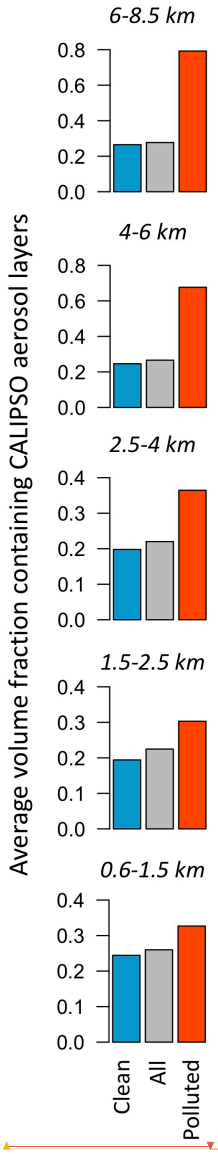
on the assumptions that combustion aerosols are associated with BC and that they are the dominant aerosol source at these altitudes during polar night. CALIPSO data were obtained in clear-sky nighttime conditions. The dark pink line on the bottom left represents sea ice extent during winter 2008-2009.



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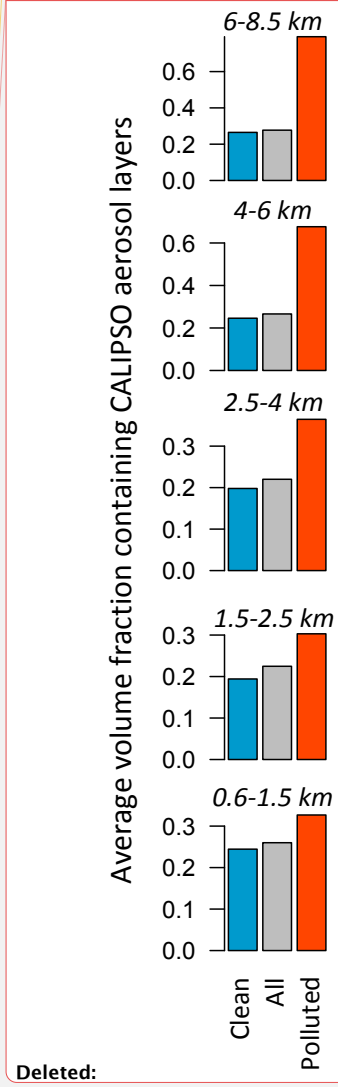

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Figure S7. The average fraction of each altitude range containing observed CALIPSO aerosol layers for FLEPXART-determined clean (BC < 30 ng m^{-3}, blue), all (grey), and polluted (BC > 150 ng m^{-3}, red) conditions from ~500,000 clear air nighttime profiles between 2008-2009. Clean conditions had significantly smaller volumes occupied by aerosol layers than all or polluted conditions at every altitude, based on a Wilcoxon rank test, $p < 0.05$. Note different x-axes ranges in the plots.

Supplementary Tables

Table S1. The data shown in Figures 3 and 5, indicating $dCF_{T,RH}$ (%), $dpptn_{T,RH}$ (%), $dCP_{T,RH}$ (%) and $dBC_{T,RH}$ (ng m^{-3}) values in different altitude ranges over sea ice and open ocean, weighted by number of cloud observations in each T/RH bin ($\overline{dCF_{T,RH}}$, $\overline{dpptn_{T,RH}}$, $\overline{dCP_{T,RH}}$, and $\overline{dBC_{T,RH}}$, respectively). Data are presented at different altitude ranges, and separately for IPCs, MPCs, and LPCs. Values of $\overline{dCF_{T,RH}}$ and $\overline{dpptn_{T,RH}}$ are expressed as the absolute change within the air volume of interest, with the bootstrapped 95% confidence intervals for the weighted mean in round brackets. The values in square brackets are the relative percent change with respect to the value found in clean conditions. An asterisk (*) indicates significant differences between all and clean conditions based on a paired Wilcoxon rank test, $p < 0.05$, using T and RH grid cells containing > 800 (400) 12.5-km^2 gridded observations for $\overline{dBC_{T,RH}}$, $\overline{dCF_{T,RH}}$, and $\overline{dpptn_{T,RH}}$ ($\overline{dCP_{T,RH}}$). Values in bold indicate a significant change in $dCF_{T,RH}$, $dpptn_{T,RH}$ or $dCP_{T,RH}$ where $dBC_{T,RH}$ > 20 ng m^{-3} (Wilcoxon rank test, $p < 0.05$). The "Total (%)" values are the CP distributions in all conditions.

	Altitude range (km)		*0.6-1.5*	*1.5-2.5*	*2.5-4*	*4-6*	*6-8.5*
Sea ice	Full dataset	$dBC_{T,RH}$ (ng m^{-3})	**27**	**21**	**14**	**7**	**3**
		$dCF_{T,RH}$ (%)	**-1.7 (-1.8 to -1.4) [-5.9]***	**-1.1 (-1.2 to -0.7) [3.5]***	**0.2 (0.1-0.5) [0.7]**	**0.7 (0.6-0.9) [3.5]***	0.4 (0.4-0.6) [3.5]*
		$dpptn_{T,RH}$ (%)	**0.9 (0.8-1.4) [3.0]***	**0.0 (-0.1-0.5) [0.1]**	**0.1 (-0.1-0.6) [0.3]**	-0.3 (-0.4-0.1) [-1.2]	-0.3 (-0.4-0.1) [-2.2]*
	Ice clouds	Total (%)	49	61	76	92	99
		$dCP_{T,RH}$ (%)	6.5 (6.3-7.3)*	**4.7 (4.4-5.4)***	1.7 (1.5-1.1)*	--	--
		$dpptn_{T,RH}$ (%)	-0.6 (-0.8 to 0.2)	0.3 (0.1-0.9)	0.5 (0.3-0.9)	0.2 (0.1-0.5)	-0.4 (-0.5 to -0.2)*
	Mixed phase clouds	Total (%)	35	36	23	8	1
		$dCP_{T,RH}$ (%)	-2.9 (-3.1 to -2.2)*	**-4.2 (-4.5 to -3.5)***	-1.7 (-1.9 to -1.1)*	--	--
		$dpptn_{T,RH}$ (%)	1.5 (1.3-2.1)*	1.3 (1.1-2.0)*	1.0 (0.8-1.7)	-0.1 (-0.3-0.4)	0.3 (-0.1-1.4)
	Liquid clouds	Total (%)	16	3	1	0	0
		$dCP_{T,RH}$ (%)	-3.7 (-3.8 to -3.1)*	**-0.4 (-0.5 to -0.1)***	0.0 (0.0-0.2)	--	--
		$dpptn_{T,RH}$ (%)	0.0 (0.0-0.0)	--	--	--	--
Open ocean	Full dataset	$dBC_{T,RH}$ (ng m^{-3})	**20**	**15**	**9**	**4**	**2**
		$dCF_{T,RH}$ (%)	-0.2 (-0.3-0.1) [-0.6]*	**1.4 (1.3-1.7) [3.8]***	1.1 (1.0-1.5) [4.2]*	0.5 (0.5-0.8) [2.3]*	0.8 (0.7-1.1) [4.0]*
		$dpptn_{T,RH}$ (%)	0.9 (0.8-1.3) [2.8]*	0.9 (0.7-1.3) [2.2]*	**0.4 (0.2-1.1) [1.2]***	0.7 (0.6-1.3) [3.1]*	-0.1 (-0.2 to 0.3) [-0.7]
	Ice clouds	Total (%)	30	39	58	78	93
		$dCP_{T,RH}$ (%)	0.6 (0.4-1.1)	**-0.6 (-0.8-0.1)**	-1.0 (-1.2 to -0.3)*	--	--
		$dpptn_{T,RH}$ (%)	0.6 (0.4-1.2)	1.1 (0.9-1.6)*	1.5 (1.3-2.1)*	0.9 (0.8-1.2)	-0.1 (-0.1-0.1)
	Mixed phase clouds	Total (%)	39	47	37	20	7
		$dCP_{T,RH}$ (%)	**0.5 (0.4-1.1)***	**0.7 (0.5-1.4)**	1.3 (1.1-2.0)*	--	--
		$dpptn_{T,RH}$ (%)	1.0 (0.7-1.6)*	0.1 (-0.1-0.7)	0.9 (0.7-1.6)	0.3 (0.1-0.9)*	-0.8 (-1.1 to -0.2)*
	Liquid clouds	Total (%)	31	15	6	1	0
		$dCP_{T,RH}$ (%)	-1.1 (-1.3 to -0.5)*	-0.2 (-0.3-0.3)	-0.3 (-0.3 to 0.0)	--	--
		$dpptn_{T,RH}$ (%)	0.1 (0.1-0.2)	-0.1 (-0.3-0.2)	-1.5 (-1.8 to -0.8)*	--	--

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	Altitude range (km)	$\overline{dBC_{T,RH}}$ (ng m^{-3})	$\overline{dCF_{T}}$
Sea ice	6-8.5	**3**	0.4 (0.4
	4-6	**7**	**0.7 (0.6**
	2.5-4	**14**	**0.2 (0.1**
	1.5-2.5	**21**	**-1.1 (-1.2 t**
	0.6-1.5	**27**	**-1.7 (-1.8 t**
Open ocean	6-8.5	**2**	0.8 (0.7
	4-6	**4**	0.5 (0.5
	2.5-4	**9**	1.1 (1.0
	1.5-2.5	**15**	**1.4 (1.3**
	0.6-1.5	**20**	-0.2 (-0.3

Table S1. The mean temperature (T, ^{o}C), relative humidity (RH, %), black carbon (BC, ng m^{-3}), and cloud fraction (CF, %) observed over the Arctic Ocean study region over sea ice and open ocean during different seasons and altitude levels. Data are shown for all conditions. Also shown are the mean differences between all minus clean conditions ("difference") for T, RH, BC, and CF, referred to as $\overline{dT}$, $\overline{dRH}$, $\overline{dBC}$, and $\overline{dCF}$ in the text.

		Sea ice						Open		
	Altitude	Fall (ASO)		Winter (NDJ)		Spring (FMA)		Fall (ASO)		Winte
	levels (km)	all	difference	all	difference	all	difference	all	difference	all
Temperature (^{o}C)	6-8.5	-48.7	0.0	-53.0	0.0	-51.9	0.1	-45.0	0.1	-49.6
	4-6	-32.3	0.5	-38.1	0.2	-40.0	0.4	-27.7	0.3	-33.2
	2.5-4	-21.6	0.5	-27.5	0.1	-29.5	0.1	-15.8	0.4	-21.1
	1.5-2.5	-15.5	0.5	-21.6	-0.4	-23.4	-0.7	-9.2	0.4	-14.6
	0.6-1.5	-11.3	0.4	-17.7	-0.6	-19.6	-1.3	-4.5	0.3	-9.9
Relative Humidity (%)	6-8.5	74.6	0.0	65.6	0.4	50.4	1.0	69.0	0.1	63.8
	4-6	61.5	-0.3	62.7	0.4	59.8	-0.6	57.1	-0.1	57.6
	2.5-4	66.7	-0.6	69.3	0.2	66.2	-1.8	61.4	0.2	58.6
	1.5-2.5	76.9	-0.9	74.0	-0.4	68.2	-0.3	74.5	0.2	72.5
	0.6-1.5	85.1	-0.3	78.1	-2.2	70.6	0.2	87.2	-0.3	88.6
BC (ng m^{-3})	6-8.5	14	1	13	2	19	5	14	2	11
	4-6	17	3	22	6	30	11	17	3	18
	2.5-4	19.8	4	31	13	41	19	19	5	28
	1.5-2.5	20	4	39	21	53	27	22	7	43
	0.6-1.5	17	3	51	31	66	32	22	9	58
Mean CF (%)	6-8.5	15.4	0.2	15.0	0.4	9.1	0.9	19.3	1.0	22.3
	4-6	22.7	0.6	21.8	1.3	16.8	0.1	24.3	0.8	26.0
	2.5-4	29.1	1.0	25.9	1.0	20.8	-2.1	27.9	1.0	28.8
	1.5-2.5	35.0	-0.3	30.3	-0.4	24.0	-3.5	34.3	1.4	39.4
	0.6-1.5	33.3	-0.2	26.9	-2.0	22.0	-3.0	35.0	-0.3	43.0