We thank the two anonymous reviewers for their careful reading and constructive suggestions on the manuscript. Below, we explain how the comments and suggestions are addressed and make note of the revisions in the revised manuscript. The reviewers' comments are in blue color. Our replies are in black, and our corresponding revisions in the manuscript are in red.

Responses to reviewer #1

In this manuscript, Shi et al. use the E3SM model nudged to MERRA-2 reanalysis to determine the relative contribution of dust from six different regions to the Arctic dust load, including the local effect. They then investigate the impact of both dust from local Arctic sources (referred to as "high-latitude dust" (HLD) and dust transported from lower latitudes (referred to collectively as "low-latitude dust" (LLD) on Arctic mixed-phase clouds and the Arctic radiative budget at the surface and top of the atmosphere (TOA). The authors find that HLD, LLD from Asia and LLD from North Africa contribute to 31%, 44% and 24% of the total dust burden in the Arctic, respectively. The influence of HLD on Arctic mixed-phase clouds was found to be limited to the surface due to frequent stable thermodynamic conditions, while LLD particularly from Asia were found to influence mixed-phase clouds at colder isotherms at higher altitudes. In terms of the seasonal variations, HLD exhibited more variation and peak concentrations at summer and autumn, whereas seasonal variability was minimal for LLD, although the largest concentrations were found in spring and winter. Overall, the HLD was found to have a net cooling cloud radiative effect (CRE) at the surface to a decrease in warm liquid clouds near the surface during autumn when sunlight is relatively weak.

HLD is currently poorly characterized yet of great importance, especially in a warming world where new sources may be emitted and is thus now becoming the focus of an increasing number of studies. The work of Shi et al. is both interesting and insightful in this regard. I recommend publication of the manuscript after the authors consider some additional suggestions below.

Reply: We thank the reviewer for the insightful comments. We have revised the manuscript following your comments. Please see our point-to-point responses to your comments below.

1. The limited vertical transport of HLD was claimed to be due to the existence of a stably stratified Arctic lower-troposphere. I would suggest to actually quantify this using the lower tropospheric stability (e.g. as the difference in potential temperature between 850 hPa and 2m). Are there differences over sea-ice and open ocean surfaces when LTS is substantially different? Does E3SM simulate LTS in reasonable agreement with observations? Please evaluate.

Reply: We show the simulated LTS (defined as the potential temperature difference between 700 and 1000 hPa) from CTRL simulation and compare it to the MERRA2 reanalysis in the revised Figure S3. It shows that the LTS from CTRL agrees well with the MERRA2 reanalysis. Also, the

LTS in the Arctic is 4 to 12 K higher than the mid- and low-latitudes, which indicates a more stratified lower troposphere in the Arctic. Moreover, since the LTS over the open water is less than that over sea ice, we expect the vertical transport of HLD to be stronger over the open waters. We add discussions about these issues in the revised Section 3.2.

Line 345-354: "However, the HLD contribution decreases rapidly with height and is less than 10% above 700 hPa. This is because the lower troposphere of the Arctic is more stratified than the mid- and low latitudes, which suppresses the vertical transport of HLD. The lower tropospheric stability (LTS) from the CTRL simulation and comparison with the MERRA2 reanalysis data are shown in Figure S3. The weak HLD vertical transport in the Arctic is also reported by previous studies (Groot Zwaaftink et al., 2016, Baddock et al., 2017; Bullard, 2017). Moreover, the LTS over the Arctic sea ice is much larger than that over open ocean surface (Schweiger et al., 2008), which may lead to a stronger vertical transport of HLD over open waters. This suggests that the vertical transport of HLD may change with the sea ice reduction in a warming future."

The new Figure S3 looks as follow.

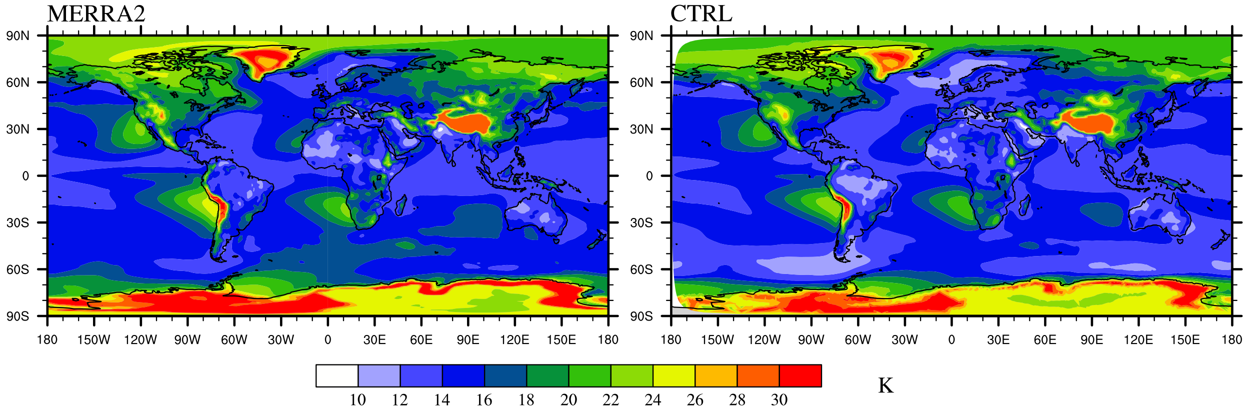


Figure S3. Annual mean (2007 to 2011) lower tropospheric stability (LTS) from MERRA2 reanalysis data and the CTRL simulation. LTS is defined as the potential temperature difference between 700 and 1000 hPa. The LTS from the CTRL simulation agrees well with the MERRA2 data.

2. Lines 38-42: The influence of these various cloud microphysical processes may also interact nonlinearly with one another and impact the phase partitioning of mixed-phase clouds as shown by Tan & Storelvmo (2016).

Reply: Thanks for pointing this out. We have included this statement and cited the paper in the revised manuscript:

Line 43-45: "All these processes can also interact with each other nonlinearly and impact the phase partitioning of mixed-phase clouds (Tan and Storelvmo, 2016)."

3. Source-tagging on lines 158-161: There is insufficient description of this technique in the manuscript itself. In addition to citing these references, please briefly describe the methodology and implementation of the technique. It seems that the six different regions are set up such that in addition to tagging them, they can also be separately tuned.

Reply: The dust source-tagging is implemented by assigning dust emitted from different sources to separate tracers. For example, in our study, there are seven tagged dust sources and thus we add seven dust tracers to the default MAM4 in E3SM. Different dust tracers are predicted independently by aerosol processes (e.g., emission, transport, and removal). Therefore, as the reviewer mentioned, each tracer can be separately tuned. We have briefly described the implementation of source-tagging in the revised manuscript:

Line 167-170: "In this method, dust emission fluxes from different sources are assigned to separate tracers and transport independently, so that dust originating from different sources can be tracked and tuned separately in a single model experiment."

4. Is aging of aerosols and the addition of coatings of pollutants that may modify the ice-nucleation efficiency of dust INPs represented in E3SM?

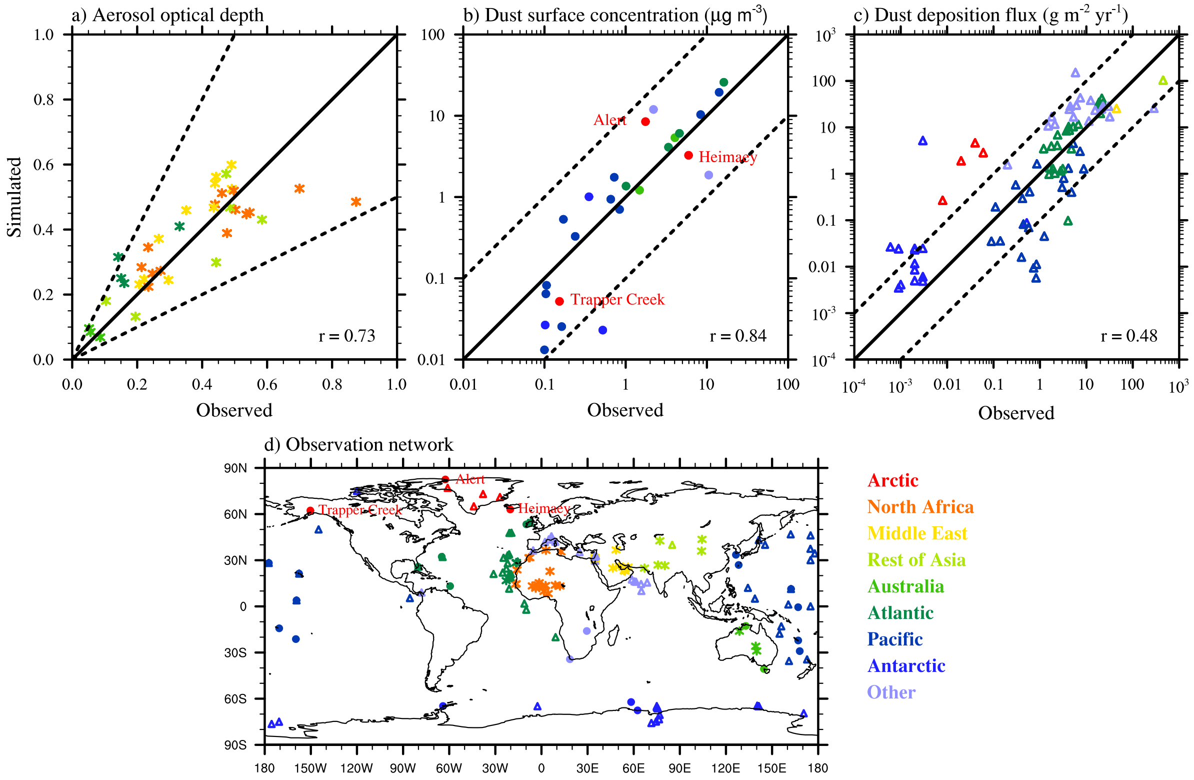
Reply: Earlier studies have found that chemical aging or coating in the atmosphere may change the ice nucleating ability of dust (Boose et al., 2016; Kulkarni et al., 2014). Our model does not consider this effect explicitly. In the CNT parameterization (please note we add the CNT scheme to the INP comparison in Section 3.3 following reviewer #2's comment), the depression of immersion freezing point by sulfate coating is considered. However, this depression effect has no differences for HLD and LLD, because dust aerosols are assumed to be internally mixed with sulfate within an aerosol mode in the MAM4 aerosol module (Liu et al., 2016) (see also in Text S2.1). The other dust ice nucleation parameterizations we used (i.e., SM20 and D15) may have already taken the aging/coating effect into account implicitly. For HLD, SM20 was derived from freshly emitted aerosol samples collected close to Iceland. For LLD, D15 included Saharan and Asian dust data collected over the Pacific Ocean basin and US Virgin Islands, respectively, which are far away from the corresponding LLD sources (we add more details about D15 and SM20 in Text S2). We have clarified this and added the discussion in the revised manuscript:

Line 436-442: "In addition, we do not explicitly represent the potential ice nucleation ability differences in freshly emitted HLD and long-range transported LLD caused by the aging and the coatings of pollutants (Kulkarni et al., 2014; Boose et al., 2016). However, D15 and SM20 may already take the aging effect into account implicitly. Because D15 is based on the Saharan and Asian dust data collected over the Pacific Ocean basin and US Virgin Islands, respectively, which are far away from the corresponding LLD sources, while SM20 is derived from the freshly emitted Icelandic HLD, which is subjected to less aging effect."

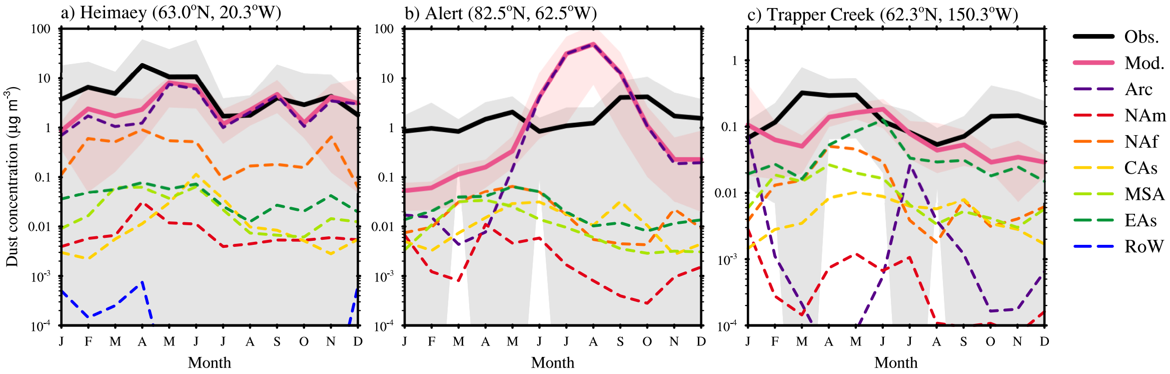
5. Given the large discrepancy between model and observations in Alert, the authors should consider utilizing long-term observations of dust available in Alert as described in Sirois and Barrie (1999).

Reply: Thanks for the suggestion. We replaced the Alert dust concentration measurements with Sirois and Barrie (1999) and updated Figures 2, 3, and previous Figure S8 (now Fig. S11). The dust concentrations from this long-term observations (1980 to 1995) are 5 to 10 times higher than those from Fan (2013), which leads to a better agreement in the comparison of annual mean dust concentrations at Alert (Figure 2b in the revised manuscript). However, the simulated results still show large high bias in the summertime (Figure 3b and S11 in the revised manuscript). So, our main conclusion related to the Alert dust comparison does not change. We still attribute the large discrepancies to the limitation of the dust emission parameterization.

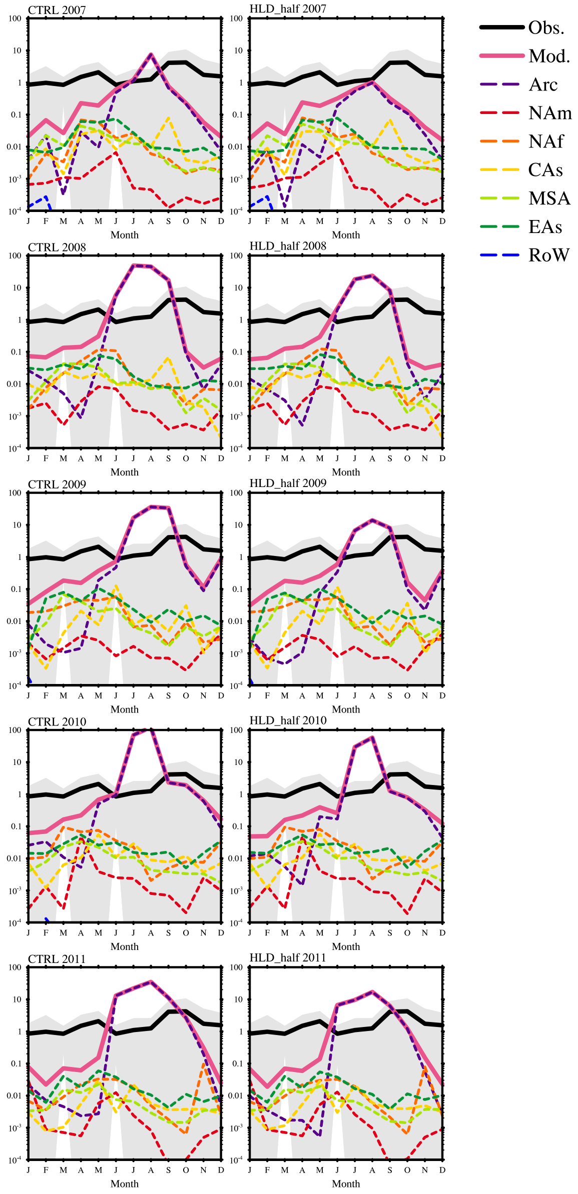
The revised Figure 2 looks as follow (only the red dot representing annual mean dust surface concentration at Alert on Figure 2b is changed).



The revised Figure 3 looks as follow (only Figure 3b is changed).



The previous Figure S8 (now Figure S11) looks as follow.



6. Are the observations and simulated AOD, dust concentrations and deposition flux directly comparable? For example, observations of cloud properties cannot be directly compared with remote sensing observations without a simulator to account for differences in the definitions of these quantities. One would expect the same for aerosol properties as well.

Reply: This is a valid point. Comparing the observations directly with simulated dust properties leads to some uncertainties. For AOD, the AERONET measurements are biased towards clear-sky conditions due to the cloud-screening procedure (Smirnov et al., 2000), while the simulated results are under all-sky conditions. For dust concentrations and dust deposition fluxes, the cut-off size of the measurements may be a source of uncertainties in the comparisons. The simulated dust particles are mainly smaller than 10 µm, while the cut-off sizes of the measurements vary from several micrometers to several tens of micrometers. We tried to address this bias for the dust concentration comparison at Trapper Creek (Figure 3c; cut-off size at 2.5 µm) and during ARCTAS flight campaign (Figure 4; cut-off size at 4 µm) by applying the same cut-off sizes to the simulated dust concentrations. However, it is hard for us to do similar things for other comparisons, because the cut-off sizes of these observations are either unclear (e.g., the dust deposition fluxes dataset) or much larger than the simulated dust size range (e.g., many dust concentration measurements have a cut-off size of 40 µm). However, the cut-off size may not be a significant source of bias in the latter situation, because most of the dust concentration data corresponds to measurements at remote stations where most of the super coarse dust particles (>10 µm) cannot reach due to the quick sedimentation. Finally, in addition to the uncertainties mentioned above, the comparisons have representative issues caused by comparing an observational station with a global model grid that has a size of ~100 km. Some of the comparisons also have systematic errors because the measurements were for a different time period than that of the model simulation. Nevertheless, despite all the uncertainties, the direct comparisons have been widely used by previous studies of dust properties (e.g. Huneeus et al., 2011; Kok et al., 2014; Albani et al., 2014).

We summarize the major biases of the comparisons in the revised manuscript:

Line 203-205: "We note that the AERONET AOD measurements are biased towards clear-sky conditions due to the cloud-screening procedure (Smirnov et al., 2000)."

Line 216-220: "We note that the comparisons are subject to representative biases caused by comparing an observational station with a global model grid point (with a horizontal resolution of ~ 100 km). The comparisons of dust concentration and deposition flux also have systematic errors because the measurements were for a different time period than that of the model simulation."

7. The discrepancy (up to almost twice) between this study and previous studies in terms of the contribution of North African dust to the Arctic dust burden is quite large. Potential reasons are listed on lines 308-310: Of these processes, which process dominates?

Reply: The wet removal may be one of the dominant processes. This process depends on the model representation of clouds and precipitation, which have large discrepancies among different models. The dust emission parameterization may be another key factor contributing to the discrepancies. For example, the fraction of total dust emission flux from each source varies if using different dust emission parameterizations. The spatial distribution of dust emission "hot spots" may also be different, which is likely to influence the transport efficiency of dust emitted from each source. Another factor that may have some impacts is the size distribution of the dust emission. In our model, we have more dust emitted in the coarse mode (according to Kok (2011)) than earlier studies, which leads to a shorter dust lifetime in our simulations. More detailed comparisons between our work and previous studies are needed for a definite answer, which is beyond the scope of this study.

8. Figures 12 and 13: It would be useful to compare how E3SM simulates Arctic CREs at the surface and TOA (e.g. comparing with the NASA CERES instrument), and how HLD vs LLD contributes to biases in the Arctic CREs in an additional column. Similarly for the LWP. The MODIS simulator can be used for the sunlit months.

Reply: Thanks for this good suggestion. We evaluate the simulated Arctic LWP and radiative fluxes with MODIS and CERES, respectively (new Figure 14 in the revised manuscript). Two MODIS datasets are used, including the standard product (Platnick et al., 2003; P03) and an improved one (Khanal et al., 2020; K20) that corrected the positive bias in the Arctic in P03. We rerun all the four simulations to turn on the MODIS simulator for the LWP comparison. Please note the reruns are only conducted for two years (2007 and 2008), due to the limit in the computer resources. The original five-year simulations are used for the CERES comparison.

According to Fig. 14a, the simulated Arctic LWP from all the simulations are lower than P03 but higher than K20. The differences among simulations are very small compared to their discrepancies with MODIS observations. But according to the numbers shown above the bar charts, including dust INPs from each of the three sources decreases the LWP (i.e., CTRL has less LWP than the other simulations), which makes comparisons slightly better as compared to K20.

The comparisons of downwelling radiative fluxes and TOA cloud radiative forcing are shown on Figs. 14b-e. Compared to CERES, all the simulations underestimate FSDS with too strong SWCF and overestimate FLDS with too strong LWCF, which likely points to the biases of modeled clouds (e.g., too much LWP as compared to K20). Similar to the LWP comparison, the differences among simulations are very small compared to the discrepancies with CERES observations. However, we do see some improvements after including dust INPs from each of the three sources (i.e., the results from CTRL are closer to the CERES results than the other three simulations).

Overall, including HLD or LLD INPs do not contribute a lot to reduction of the biases in simulating the LWP and CREs in the AMPCs. However, the representation of AMPCs in global climate models is associated with multiple cloud macro- and microphysical processes, and large-scale dynamics (Morrison et al., 2012). As mentioned by the reviewer in comment 2, these processes interact with each other non-linearly. Therefore, even though including HLD or LLD INPs do not improve the representation of AMPCs significantly in our model, a good representation of dust INPs, especially including HLD INPs, could still be of great importance for parameterizing AMPCs.

The new Figure 14 looks:

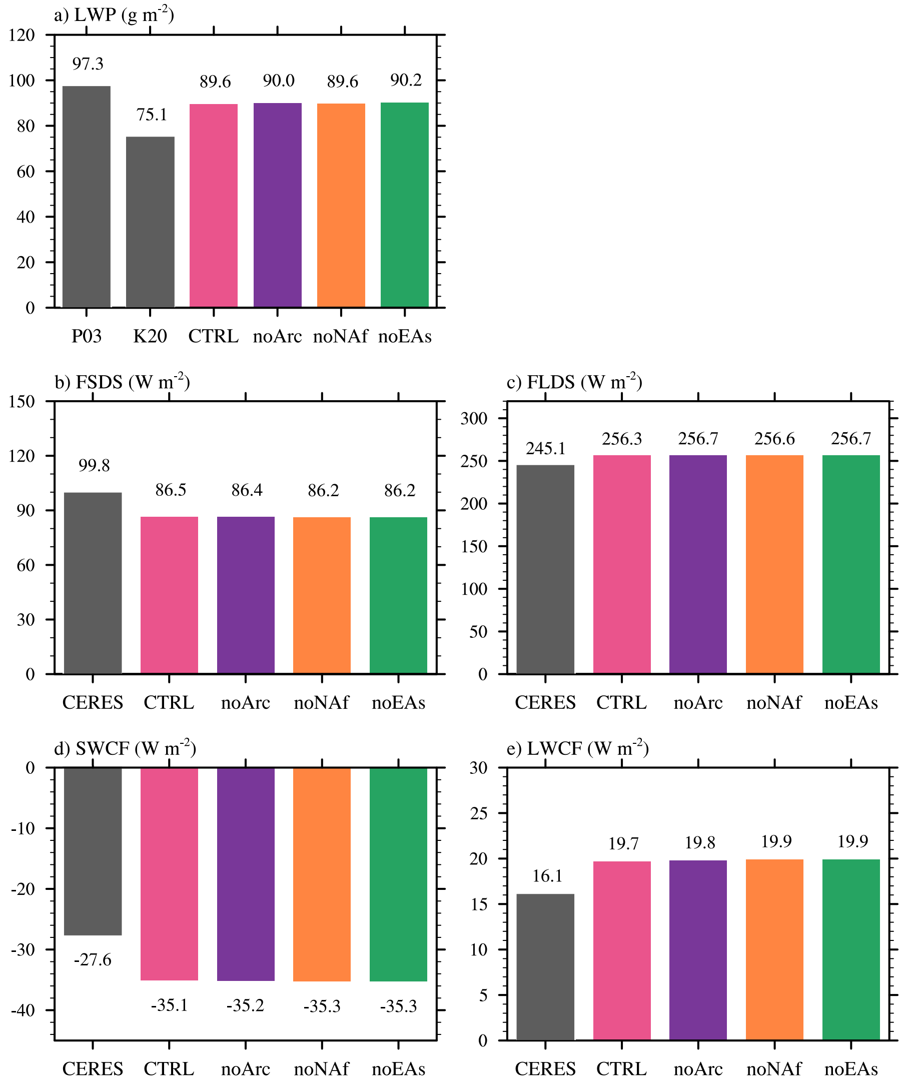


Figure 14. a) Annual mean Arctic averaged LWP over ocean for the MODIS observations (2007-2009) and the four simulations (2007-2008). Two MODIS datasets are used, including the standard product (Platnick et al., 2003; P03) and an improved one (Khanal et al., 2020; K20). The MODIS simulator is used to calculate the simulated LWP. b) - e) Annual mean Arctic averaged b) FSDS,

c) FLDS, d) SWCF, and e) LWCF for the CERES observation (2007-2011) and the four simulations (2007-2011).

We added discussions in Section 3.4 to address the reviewer's comment:

Line 534-558: "Finally, we evaluate the model performance in simulating the Arctic LWP and radiative fluxes against the Moderate Resolution Imaging Spectroradiometer (MODIS) LWP (Platnick et al., 2003) and the Cloud and the Earth's Radiant Energy System Energy Balanced and Filled Edition 4.1 (CERES-EBAF Ed4.1) products (Loeb et al., 2018; Kato et al., 2018), respectively (Figure 14). Two MODIS datasets are used, including the standard product (Platnick et al., 2003; P03) and an improved one (Khanal et al., 2020; K20) that corrected the positive bias in the Arctic in P03. The MODIS simulator is used for the LWP comparison. According to Fig. 14, the simulated LWP from the four experiments are lower than P03 but higher than K20. All the four experiments also underestimate FSDS with too strong SWCF and overestimate FLDS with too strong LWCF, which likely points to the biases of modeled clouds (e.g., too much LWP as compared to K20). The differences among the model experiments are very small compared to their discrepancies with observations. We notice including dust INPs from the three sources decreases the simulated LWP (i.e., CTRL has less LWP than the other experiments) (Figure 14a), which makes the model performance better if compared to K20. Moreover, it shows noticeable improvements in simulating both surface and TOA radiative fluxes after including dust INPs from each of the three sources (i.e., the results from CTRL are closer to the CERES results than the other three experiments) (Figure 14b-e).

Overall, including HLD or LLD INPs do not contribute a lot to the reduction of biases in simulating the LWP and radiative fluxes in the AMPCs. However, the representation of AMPCs in global climate models is associated with multiple cloud macro- and microphysical processes, and large-scale dynamics (Morrison et al., 2012) (see more discussion in Section 4), which interact with one another non-linearly. Therefore, even though including HLD or LLD INPs do not improve the representation of AMPCs significantly in our model, a good representation of dust INPs, especially including HLD INPs, could still be of great importance for parameterizing AMPCs in the model."

9. Comparison with CALIOP: Why use observations from 2007-2009? The record extends well beyond that and the 2007 observations are partially impacted by the change in the tilt of the nadir-viewing angle. Also, what CALIOP product was used and what was the version of the product? Arctic aerosol layers are frequently too tenuous to be detected by CALIOP and are also furthermore impacted by the presence of clouds that can interfere with the cloud-aerosol discrimination algorithm.

Reply: We use the CALIPSO dust extinction dataset developed by Luo et al. (2015a, 2015b). Luo et al. (2015a) developed a new method for dust separation from other aerosol types to derive the dust backscatter coefficient in the lidar equation inversion stage using CAL-L1B data, which has less uncertainties than doing the separation based on lidar inversion products (i.e., CAL-L2) in previous studies (e.g., Amiridis et al., 2013; Yu et al., 2015). Luo et al. (2015b) further developed a new dust identification method by using combined lidar-radar cloud masks from CloudSat and CALIPSO, which significantly improves the detection of optically thin dust layer, especially in the Arctic. We use both the new dust separation method (Luo et al., 2015a) and the new dust identification method (Luo et al., 2015b) to produce the nighttime dust extinction dataset.

We use the retrievals during 2007-2009 because of the data availability (lidar-radar cloud masks). Due to the battery anomaly on April 17th, 2011 the CloudSat stopped collecting data for ~1 year and since then continued to only operate during the sunlit portion of the orbit with degraded overlap with CALIPSO. Therefore, lidar-only cloud masks are needed for retrieving nighttime dust extinction, which have been in the development.

We notice that the nadir angle was increased from 0.3° to 3° to reduce specular returns from clouds containing horizontally oriented ice crystals in November 2007. According to the official document (https://asdc.larc.nasa.gov/documents/calipso/TiltModeGeometry.pdf), the optical properties reported for the measurements of aerosols and water clouds are not expected to change as a result of the change in pointing angle. However, the properties reported for individual ice clouds will change by varying amounts, which may contribute to retrieval uncertainties.

We added some descriptions about the CALIPSO data used in this study in Section 3.1:

Line 266-268: "The Luo et al. (2015a, b) data set has improvements in dust separation from other aerosol types and thin dust layer detection in the Arctic than the standard CALIPSO product (Winker et al., 2013)."

Typographical error:

- Line 137: "hour" should be "hours"

Reply: It is corrected. Thanks.

Reference

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