Reply to Referee#2

December 19, 2023

Dear Referee #2,

We greatly appreciate a number of valuable comments on our manuscript entitled "Regional variations in mineralogy of dust in ice cores obtained from northeastern and northwestern Greenland over the past 100 years" by Nagatsuka et al. submitted to the journal Climate of the Past. Please see enclosed our responses (**blue text**) to each of your comments (**black text**) describing on the following pages.

Best regards,

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General statements

This paper documents analyses of dust particles in the EGRIP Greenland ice core. The analyses consist of size information, and a set of SEM observations that have allowed mineralogical classification to be made. The work covers only the last century in 10 decade-long bins. The data are certainly worthwhile to make available, and a significant amount of work has gone into the SEM study. However it must be said that the new insights gained from this study are quite minor. I appreciate the authors' point that isotopic analyses would be demanding in terms of sample size (though not with modern instruments quite as demanding as implied); however the coarse mineralogical separation here is simply not capable of defining source areas. It might (as in the previous work from SIGMA-D) be capable of defining the appearance of local sources but in this work no evidence of any local material is presented. Additionally the time series all appear flat (within statistical variation) meaning that there is no story about changing sources here. The back trajectory work is used in a way I think is inappropriate to try to define source areas (I will elaborate later). The work about volcanic material presents nothing new – we would not expect to see tephra from most eruptions, and indeed we don't. In summary, there are data here that are worthwhile to make available, but there is no scientific story, and the data are not capable of defining source areas. It is therefore for the editor to decide: the authors could be encouraged to strip the paper down and correct/remove unsupported statements so that a correct but unexciting paper appears in CP: this would be a major revision as there are some issues that are conceptually wrong (especially regarding back trajectories) at present. Or they could recommend submission of a stripped down version to a journal such as ESSD that takes datasets without expecting too much in the way of interpretation.

We would like to thank you for all the valuable comments. We agree with your suggestions that no firm evidence of local material is presented in this study and that coarse mineralogical separation here is not enough to define source areas. Thus, we will remove Fig.11 and revise the sentences to describe there is a local supply albeit small as you suggested. We also agree that accurately 'identifying' the sources of the ice core dust is challenging based solely on trajectory results. However, our results clearly demonstrate the differences in dust sources between the EGRIP and SIGMA-D ice cores based on mineral composition. We will revise the manuscript to emphasize the distinctions between the two ice core sites and 'discuss' potential dust source areas. Consequently, one of the possible sources of the EGRIP ice core dust is likely to be Asian deserts as the other Greenland ice core dust from glacial periods and the time series of the mineral composition did not change significantly over the last 100 years. These results may not sound exciting or new. But we argue that these results are new and important since no previous study has confirmed it. Although previous studies revealed mineral dust sources of the Greenland ice cores, most of the dust were obtained from the last glacial period when the dust concentrations were high. Furthermore, we also found the new results indicating that the source areas have changed since 1970-1980, which is new insight that could improve the understanding of Greenland ice core dust. According to the suggestions from you and other referees, we will add the results of new analyses and revise our manuscript as below:

Introduction and conclusion:

First, we will clarify and emphasize new and important findings in our study, especially in introduction and conclusion sections. Our SEM-EDS analysis show the following two new results: This study is the first to demonstrate (1) continuous records of a northeast Greenland ice core dust (EGRIP) composition with a high temporal resolution during the last 100 years and (2) their significant differences from northwestern Greenland.

Although previous studies revealed mineral dust sources of the Greenland ice cores, most of the dust were obtained from the last glacial period when the dust concentrations were high. Some studies have analyzed the ice core minerals during the Holocene (8.2-11.6 ka : Han et al., 2018, 4-7 ka: Simonsen et al., 2019, 1997-2001 A.D : Bory et al., 2003a), a period of low dust concentration, however, they needed to concentrate decades to thousands of years of ice for each sample. Thus, our results can provide new and important insight to understand sources and transportation processes of ice core dust as well as climate and environmental change in recent years when global warming is progressing.

Results and Discussion:

We agree that it is difficult to "identity" the sources of the ice core dust accurately only based on our mineralogical and trajectory results as suggested by referee #2. However, our results can clearly show the differences in the dust sources between the EGRIP and SIGMA-D ice cores based on the mineral composition. Thus, we will revise the manuscript to emphasize the differences in the two ice core sites and "discuss" possible dust source areas. We will also add some new results of the mineral composition and elemental concentration of the EGRIP ice core dust obtained from the SEM-EDS and ICP-MS analyses to discuss the possible sources and their temporal variations (Figs. Reply. 1 - 4). The details are as follows.

The EGRIP dust showed mineral composition characterized in Asian-sourced dust as described in line 305-307. Thus, Asian desert can be one of possible sources of the dust at EGRIP as the other Greenland ice core sites. Comparing the mineral composition, however, the EGRIP ice core dust showed significantly lower quartz content and higher illite/micas/chlorite contents than the GRIP ice core dust originated from Chinese deserts (Fig. Reply 1 and 2). We cannot compare the EGRIP mineral composition records directly with those of previous studies because there have been no continuous records of dust composition during the last 100 years. Moreover, the periods analyzed (modern vs mostly glacial), analytical methods (X-ray diffraction analysis (XRD) vs SEM-EDS) and mineral identification methods of the ice core dust were different (XRD used in previous studies cannot separate micas from illite, whereas the elemental peak intensity ratio sorting scheme used in our study cannot separate micas from chlorite completely. We identified them as mixed layer of micas/chlorite). However, the lower quartz abundance may indicate that the EGRIP dust were contributed not only from Asia, but also from other source areas. Thus, the EGRIP ice core dust was originated from multiple sources. Air-mass trajectory results suggests that Northern Eurasia and North America are the best candidate of the other possible sources.

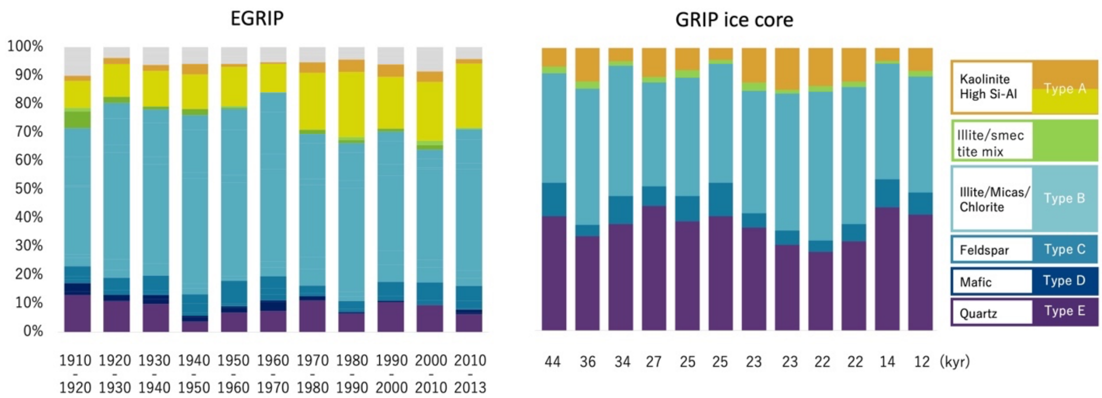

Figure Reply 1. Mineral composition of the EGRIP (this study) and GRIP ice cores (Svensson et al., 2000)

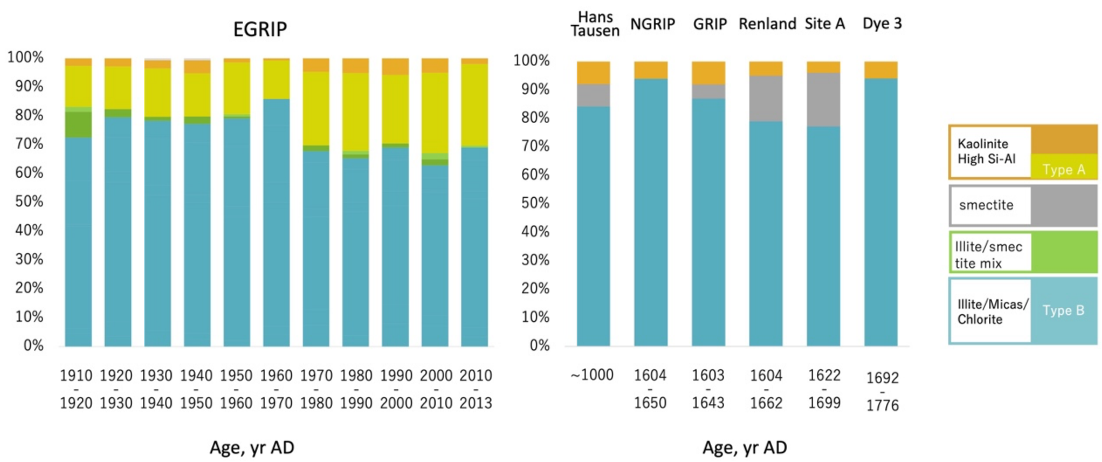

Figure Reply 2. Clay mineral composition of the EGRIP (this study) and Hans Tausen, NGRIP, GRIP, Renland, Site A, and Dye-3 ice cores (Bory et al., 2003).

Although mineral composition records indicates that the mineral sources of the EGRIP dust have not changed dramatically over the past 100 years as described in Line 26, the ternary clay mineralogy diagram (Fig. 10) and the elemental concentration ratios of the EGRIP ice core dust analyzed by the SEM-EDS and ICP-MS, respectively, suggest that there is slight difference in the dust sources in different periods. In ternary clay mineralogy diagram, the dust from 1910 to 1970 were plotted close to glacial dust from the GRIP ice core as well as the Asian dust, whereas the dust from 1970 to 2013 were not. Furthermore, the samples from 1980 to 2013 showed the steeper slope in scatter plots of elemental concentrations analyzed by ICP-MS (Fig. Reply 3). These results indicate that the contribution rate from the possible sources has changed since 1970-1980, which is new insight that could improve the understanding of Greenland ice core dust.

One of the possible causes of this change is decreasing Asian dust supply to the EGRIP. Although the number of the particles may not be sufficient to show accurate percentages, the illite content of the EGRIP showed decreasing trend since 1980 that is similar to that for dust occurrence in Asian deserts (e.g. Zhang et al., 2020). This is likely due to global warming that could have led to a decrease in atmospheric temperature gradients and decline in wind speed and then decreasing dust storm frequency/intensity. We also compared the nss Ca/Al ratios with average volume of dust particles calculated from volume concentration divided by number concentration of the EGRIP ice core, which can reflect the contribution of evaporite minerals that are abundant in deserts (Formenti et al., 2011) and the distance from the source areas, respectively (Fig. Reply 4). The nss Ca/Al ratios were slightly lower but average volume of dust particles were higher from 1970 to 1990. These results indicate that the contribution from distant deserts likely decreased during the period, which are consistent with the hypothesis that the dust contribution has changed since 1970-1980.

The higher Ca/Al ratios and lower average volume of dust particles of the EGRIP dust after 1990 imply the possibilities that the dust has originated from distant sources with higher salinity environments in this period. Previous studies have revealed that there are large variations in the Ca/Al ratios among Asian deserts and that the Taklamakan deserts has higher values than the other deserts (e.g. Formenti et al., 2011). The EGRIP ice core dust might be derived from such sources.

We will add some sentences in the manuscript to describe these points.

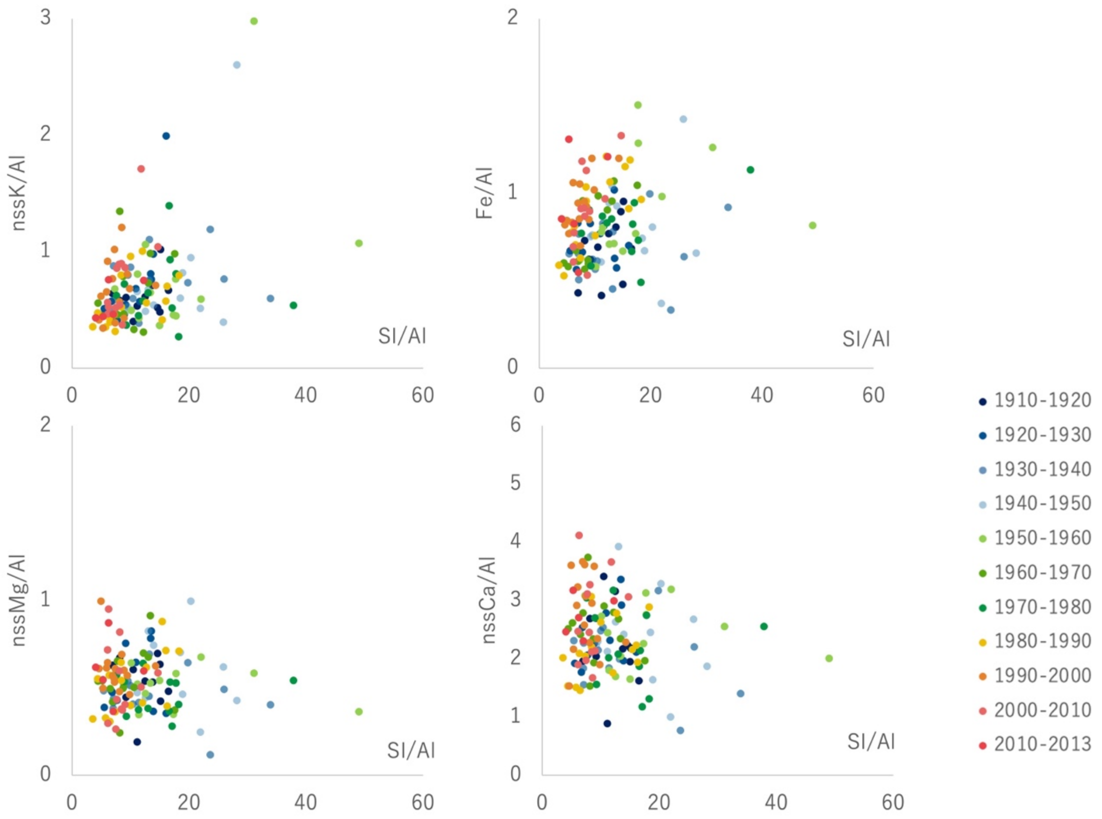

Figure Reply 3. Elemental concentration ratios of microparticles in the EGRIP ice core anayzed by ICP-MS.

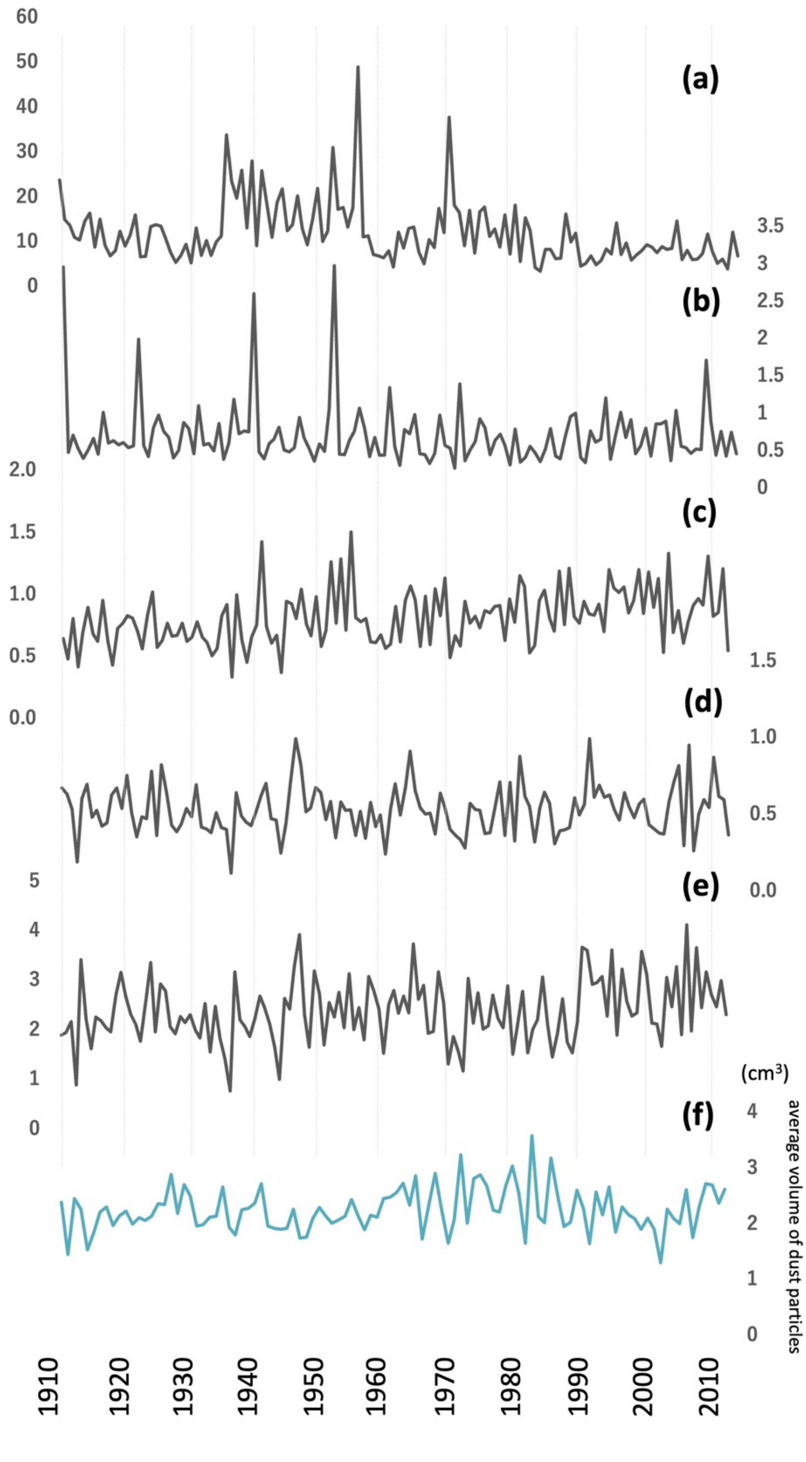

Figure Reply 4. Historical records of elemental concentration ratios ((a) Si/Al, (b) nssK/Al, (c) Fe/Al, (d) nssMg/Al, (e) nssCa/Al, and (f) average volume of dust particles in the EGRIP ice core.

Detailed comments

Abstract. There are a couple of sentences I don't think are supported by the data and text. On lines 26-27 (local dust), I don't think the case is made at all for a local dust source.

We will revise the abstract in a way that is consistent with our results and delete the sentence "The subtle variation in the EGRIP ice-core mineral composition is likely due to a minor contribution of local dust."

On lines 31-33, the back trajectories really don't make this case as presented.

As described in the response to general statement, we will revise the manuscript to emphasize the difference in dust sources between the EGRIP and SIGMA-D and tone down our description about identifying dust sources. Thus, we can use our trajectory results to indicate the significant differences in the dust contribution from possible sources between the two ice core sites in the revised manuscript although we cannot use to accurate quantitative evaluation.

And the last sentence is contradictory as written.

We will delete the last sentences.

Section 2.3: please be more precise with this explanation. As written, my reading is that you collected a total of 11 filters, and analysed 200 particles on each. However was each sample an average of all the vials from the 10 year interval or is it a spot measurement from one or more vials within the section? This is crucial to whether the samples are representative.

Many thanks for the suggestion. Each sample was an average of the 10 year-ice samples. For the SEM analysis, the ice core samples were first melted and collected in the glass vial for every 10-year and well stirred. Next, 500μL of each sample was extracted from the vial and was filtered through a polycarbonate membrane filter (So we made 11 filters). Assuming that the dust samples were distributed uniformly on a filter, we cut a filer into quarters and observed a total of 200 randomly selected particles from it. To eliminate bias for choice of mineral dust as much as possible, we observed particles in various position on a 1/4 filter over a couple of days. We will add the sentences to describe above in the Section 2.3.

And if you only analysed 200 particles per filter, then how are you later giving mineralogies to 0.1% accuracy (Table 3)? If this is really what was done (I hope not) then most of the differences between time periods would be completely statistically insignificant, so it's really important to understand this.

Fig 6. I again emphasise that if each decade is really only represented by a count of the types of 200 particles then most differences are likely to be statistically insignificant so again please clarify in methods, and if I have understood correctly please don't overinterpret counts that have large uncertainties on them.

Section 4.1 eg line 263. Again I doubt the significance of any variability here given the number of particles of each sort counted.

We agree on the referee's comments that 200 particles are likely insufficient to show accurate proportion of each mineral, and thus we will not show decimal point of the data. However, we think our results can clearly show that the size and composition and their variations of the EGRIP ice core dust differ substantially from the SIGMA-D ice core dust analyzed in the same way as this study (150 particles in every 5 years for the SIGMA-D) over the past 100 years. Thus, again, we will revise our manuscript to emphasize the differences in the sources between the EGRIP and SIGMA-D ice core dust. We will also analyze some remaining filters and confirm that their mineral composition will be the same as this study.

Back trajectories: Starting with lines 153-5. I don't understand what you are telling us about wet deposition here? Standard HYSPILT trajectories are simply that – trajectories of air masses, which take no account of the contents of the air and therefore take no account of what is lost en route or how it is deposited in Greenland. I am therefore not clear what you mean about wet deposition and precipitation. Please explain but for now I will assume you present simple back trajectories.

"was considered" (L155) may be an insufficient description. When we calculated the probability, we accumulated number of air mass within a given 1deg x 1deg grid, and then obtained the percentage against the total count at the grid. To consider the precipitation effect, we used the precipitation amount when the air mass arrived at the site. By this consideration, we can assume that the air mass without precipitation (0 mm) doesn't contribute to the probability. We will change the description L154-155 as: (Iizuka et al., 2018; Parvin et al., 2019), for which the probability was weighted by the daily precipitation at the date when the air mass arrived at the ice core site (Nagatsuka et al., 2021).

The next issue is that you describe launching trajectories from 4 different altitudes, but you don't say which is shown in your figures. I assume it's all 4 mixed together but this is an odd thing to do without first discussing what are the differences for different altitudes of launch.

We show the probability distributions of the 7-day back trajectories for the four different altitudes of the initial air mass (Fig. Reply 5). The figure shows the relatively higher contribution from distant regions (North America and Northern Eurasia) and lower contribution from Greenland coast at high altitudes. The differences in such contributions between the altitudes are smaller in the EGRIP, but larger in the SIGMA-D (the contribution from North America increase significantly at high latitudes). However, we don't think that it's necessary to change Fig. 2 to Fig. Reply 5 in the revised manuscript by the following three reasons: (1) the discussion about dust source areas will be mainly based on the SEM-EDS results as described in the response to general comments, and not quite depend on the trajectory results. (2) Fig. 2 can clearly show the differences in the airmass transportation pathways between the EGRIP and SIGMA-D. (3) previous studies have not shown nor discussed the results at different altitudes.

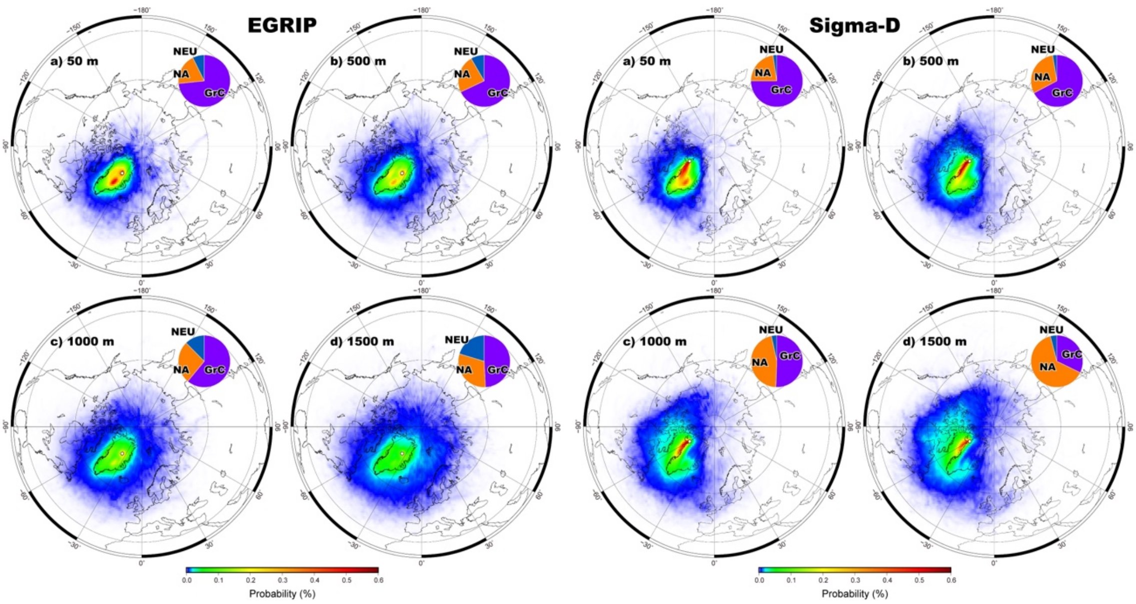

Figure Reply 5. Probability distribution for air mass at EGRIP and SIGMA-D sites in four different altitudes (50, 500, 1000, 1500m) from 7-day three-dimensional back trajectory analysis from 1958 to 2014.

Then Figure 2 is useless to the reader as the trajectory densities shown are nearly all over the ocean which cannot be a source of dust. You need to treat the trajectories in a different way: so we can judge where they might pick up dust. But there are a number of subtleties that are not well treated here. Firstly, of course every trajectory passes over the Greenland coast where there might be rock. But what matters is (a) how long the air spent over a source and (b) whether it was at low altitude where it could pick up fresh material (bearing in mind that as Schupbach discussed there could be places where air is lofted to altitude from the surface but at scales that Hysplit doesn't capture). The reason I mention this in respect of Fig 2 is that the Figure gives the impression of a high input from the Greenland coast, but the reality is that the air probably only spends an hour or less over the thin coastal strip of sediment/soil, and probably not at ground level (as the air has to have reached high altitude by the time it reaches EGRIP). The figure is therefore misleading about the potential influence of local material, and completely unsuited (because the trajectories are too short) to showing which distant continent could have contributed.

We would appreciate it if it was clear what "a different way" was. About your "matters", we cannot describe (a) how long the air mass stayed over a grid as other similar studies didn't. But the altitude for summing up air mass probability was constrained lower than 1500 a.g.l. by assuming the mixing layer with the description "within this altitude range" (L153). For clarity, we will add "(< 1500 m a.g.l.)" after "within this altitude range" (L153).

Page 7 re dating. This is OK but I'd like to have seen your assignment of the ages near the bottom of your section confirmed by deeper volcanic matches (ie ones below the ice you used). Motjabavi et al 2020 have presented a chronology for EGRIP, so it would be helpful if you compared your chronology to theirs.

Many thanks for the suggestion. We will describe deeper volcanic events to confirm the ages of our bottom section of the ice core.

Line 215. "The mode values showed an increasing trend over the 100-year period except for the 1910–1920 sample". This is not an acceptable statement. With that first sample, there is no trend, and even without it, I doubt the trend is statistically significant.

We will revise the sentences that "The mode diameter showed an almost similar values (0.37-0.48µm) during the periods, except for a large value of the 1910–1920 sample (0.57µm,), but was slightly higher of the 1960-2013 samples than the 1920-1960 samples. "

Fig 7 should be removed. It simply doesn't represent what you say for the reasons I outlined above. These are not the "contributions of air masses" unless you weight them by the length of time they spend over a location (in fact I am very unclear what is plotted here anyway but it certainly isn't what it says). Fig 8 is slightly more helpful and could possibly be used to interpret your data if we also had information about altitudes (where did trajectories last intersect with the ground?).
We will remove Fig 7. About Fig. 8, we clarify the altitude we dealt with in the reply above addressed. We show trajectory results for the four different altitudes in Fig. Reply 5.

Line 282. "The morphological properties of the EGRIP ice-core dust also suggest a small supply of minerals from local source areas". This is not correctly written. It suggests evidence that there is a local supply albeit small. What you actually have is no evidence for a local supply. Please reword.
We will reword the sentences as suggested by Referee #2.

Page 15/16. While I agree that prior data suggest an East Asian source I cannot agree that you have added new evidence for that. As your Table 1 shows, 4 of your dust types could originate from East Asia but none of them uniquely so. Indeed both Asia and North America are mentioned for types A, B, C. This method is simply incapable to differentiate the long range sources.
Although the EGRIP ice core dust was likely originated from multiple distant sources (Asia, North America and Northern Eurasia), it is impossible to identify contribution rate from each region based on the mineral composition as suggested by the referee because of the similar characteristics of the composition. However, the ternary clay mineralogy diagram (Fig. 10) can clearly distinguish Asia and North America and the EGRIP ice core dust from 1910 to 1970 were plotted close to glacial dust from the GRIP ice core as well as the Asian dust. Furthermore, the EGRIP ice core dust showed significantly lower kaolinite contents than the SIGMA-D dust that was likely derived from northern Canada, which is also supported by trajectory results (Fig. 8). Thus, we think that the contribution from North America to the EGRIP was smaller than the SIGMA-D, and that from Asia was likely larger until 1970-1980.

The discussion of trajectories on page 16 is also incapable of differentiating sources as discussed above (and also for the reasons given by Schupbach).
We agree that the back-trajectory analysis is incapable of identifying dust transport from Asia to Greenland as suggested by Schüpbach et al. (2018) and thus is not used to show the accurate contribution rate from each possible source area. However, our trajectory results show the clear difference between the EGRIP and SIGMA-D sites.

Page 17. I simply can't see the possible changes (more after 1980) in proportion of medium sized particles in Figure 11. Please either make a clear statistical case or remove this implication. Additionally changes in the proportion of 2 micron particles have not been shown to indicate a local source, but more likely a change in transport strength (if significant).

We agree with your suggestions and remove this implication and Figure 11.

Section 4.3 adds nothing and should be removed.

We will remove Section 4.3 as suggested by referee.

The conclusions need to be changed to reflect the changes in the text, eg lines 432-4 are not shown in the data.

We will revise the conclusion to reflect the discussion.

References:

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