Reply to Referee#1

December 19, 2023

Dear Referee #1,

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

In this manuscript Nagatsuka and colleagues analyze the mineral content of dust particles in a shallow ice core from central Greenland, and estimate the potential source contribution through backtrack trajectory modeling. They compare their results with a similar core drilled further west and published previously.

The main contribution of this manuscript are the detailed mineralogical analyses of this new shallow core since 1910. Although the results are new, they are rather incremental and it is not clear how these new data are improving our knowledge of Central Greenland dust advection or source contribution. That dust in central Greenland mostly originates from distant sources (mostly in East Asia) and not local ones was already known from other cores. This study mostly repeats this result at higher resolution. In addition, the authors imply links between their results with recent warming in Greenland, which is poorly supported since any kind of analyses including Atlantic and Pacific oscillations are missing. Finally, the discussion of volcanic particles is mostly a literature review without any contribution from this manuscript.

For these reasons I suggest to reject this manuscript as it does not include sufficient scientific advances for Climate of the Past. Instead, I suggest to publish these results in a more specialized journal.

We would like to thank you for all the valuable comments. We agree with your suggestions that we should discuss the link between mineral composition records and Atlantic and Pacific oscillations in much more detail. Thus, we will compare the variations in mineral composition with those in such oscillations in the revised manuscript. We also agree with the suggestion to compare the results not only with a northwest Greenland ice core (SIGMA-D) but also with other cores in Greenland. Therefore, we compared the mineral composition of the EGRIP ice core dust with that of the GRIP ice core dust. (Fig. Reply 1).

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, as suggested by you and referee #2. 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 <u>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.</u>

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, <u>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.</u>

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 core 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 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, <u>the</u> <u>EGRIP ice core dust was originated from multiple sources</u>. 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). 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.

Major Comments:

The authors mostly compare their results with an ice core from northeast Greenland (sigma-d) for which similar data are available. However, the comparison to central Greenland, east Greenland (Renland) and southeast Greenland (Dye-3) should be included in the discussion. In particular, the comparison with NGRIP should be made, as the claim that EGRIP represents Eastern Greenland and NGRIP central Greenland is a bit shaky, considering both sites are at similar altitudes and quite close to each other.

We obtained mineral composition data from Central Greenland ice cores (GRIP and GISP2; e.g., Maggi, 1997; Svensson et al., 2000; Újvári et al., 2022) and compared the results with the dust from the EGRIP ice core (Fig. Reply 1). The lower quartz abundance may indicate that the EGRIP dust was contributed not only from Asia but also from other source areas. Additionally, we found one dataset from east and southeast Greenland (Renland and Dye-3; Bory et al., 2003), providing information on clay mineral composition (Fig. Reply 2). The clay mineral composition of the EGRIP ice core dust was not similar to that of the Renland and Dye-3 ice core dust. However, we cannot discuss about the similarity of clay mineralogy between the ice cores based on only one dataset.

The authors group Europe and NorthEast Asia, as well as Africa and SouthEast Asia into single potential source areas in their analysis. Considering the long debate about Asian, European and African dust sources for Greenland, these should probably be split into four, unless the authors can justify their choice.

As shown in first response, we recalculated the back-trajectories as follows. Potential dust source (land) areas are divided into eight regions following referee #1's suggestion; the Greenland Ice Sheet (GrIS), the Greenland coast (GrC), North America (NA), Europe (EU), Russia (RUS), Central Asia (CA), Southeast Asia (SA), Middle East (ME), and Africa (AF) (Fig. Reply 5). Considering the dust sources, the ocean and GrIS were excluded from the calculation.

The authors talk about trends in the data in various sections of the manuscript, in particular comparing the last 20 years with the mid-section of the core. In particular, the authors imply that the recent warming has been responsible for various changes in dust mineralogy and concentrations. But looking at the complete record, these look more like multidecadal oscillations to me and should not be described as trends. Although recent warming in Greenland may have been responsible for some of the observed changes, such a hypothesis has to be put in context with the complete oscillations shown in the records. The authors briefly mention NAO at some point, but the link between their record and various Atlantic and Pacific oscillations should be discussed in much more details.

We will revise the sentences to describe there is a local supply albeit small as suggested by another referee. We will carry out analyses related to Atlantic and Pacific oscillations as described above.



Figure reply5 (revised from fig. 2): Map showing (a) location of EGRIP and SIGMA-D ice core sites in Greenland and five regions used for calculating regional contribution (GrIS: Greenland Ice Sheet, grey; GrC: Greenland coast, purple; NA: North America, orange; EU: Europe, blue; RUS: Russia, light blue; CA: Central Asia, yellow; SA: Southeast Asia, green; ME: Middle East, red; and AF: Africa, brown) and probability distribution for air mass at (b) EGRIP and (c) SIGMA-D sites from 7-day three-dimensional back trajectory analysis from 1958 to 2014.

Minor Comments:

Line 18: Abstract could benefit from a more general introductory phrase at the beginning. We will add a more general introductory phrase at the beginning of the abstract as suggested by the referee.

Line 33-34: Since Greenland is an island, all air masses must come from a coast. Be more precise. We will delete sentences describing air mass contribution from Greenland coast as suggested by another referee.

Line 38: 100,000 years seems a bit short for the geological timescale, although I am not a geologist and may be wrong. Maybe Milankovitch timescale?

We agree with your suggestion and will revise the sentences to read "During the glacialinterglacial climate changes (e.g., from the Eemian to the Holocene), ice-core dust records...".

Line 42: "ice-core dust shows...". Also this is only shown for Central Greenland, not the whole of Greenland.

We will revise the sentences to read "In the 20th century, the Central Greenland ice-core dust showed..." in the revised manuscript.

Line 44-45: This is not at all the message of Svensson et al., 2000. Generally, I very much doubt that the seasonal variability in dust advection to Greenland is due to climate change... We will revise the sentences to read "...climate change, environmental changes in dust sources such as their extent and aridity, and atmospheric transport"

Line 46: Not "predict". "estimate" maybe. We will change the words "predict" to "estimate" in the revised manuscript.

Line 47: "partly responsible". Grain size and partial melt is very important as well for albedo. We will add the "partly" before "responsible" as suggested by referee.

Line 53-58: The message of these phrases is unclear. Are you suggesting to collect dust from outcropped ice in the ablation zone to measure old dust? Or just dust in fresh snow on the surface? Then why talk about the movement of ice and dust through the ice sheet?

Nagatsuka et al. (2016) reported that englacial dust appear to be important for forming the dark ice surface of a glacier in northwestern Greenland. The dust was deposited from the atmosphere in the past in the upstream, traveled through the ice sheet, outcropped again in the ablation zone,

and formed cryoconite. We assume that the dust was likely deposited widely on the ice sheet in the past and thus preserved in ice cores. We will add these descriptions in the revised manuscript.

Line 60: Ujvari et al. used Hf not Pb. We will correct "Hf" to "Pb" in the revised manuscript.

Line 75: Can you give some references to support that hypothesis? We will add some references (e.g. Bory et al 2003; Kjær et al 2022) to support the hypothesis.

Line 117: Is the Beckman CC located in a normal laboratory or a clean room or a laminar flow bench? What kind of aperture tube was used?

Beckman CC is located in clean room in a Class 10000 clean room and we use $30 \,\mu$ m aperture tube.

Line 145, Table 1: Why is South America included as a possible source for Type A particles? I'm not saying it's wrong (although I do doubt it), but I wonder why it was included in the list. It was a mistake. We will delete South America from Table 1.

Lines 171-174: Snow cover fractions vary substantially from one model to another. Please provide and uncertainty estimate due to the choice of the model.

Following the suggestion, we will analyze snow cover fractions using LS3MIP multi-model results to evaluate model uncertainty.

Lines 209-216: What do the numerical ranges indicate? 1-sigma range? If so how were the mean and standard deviations calculated?

This range represents the minimum to maximum values of particle diameter. We will add a description explaining about it in the manuscript.

References:

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