Reply to Referee#3

December 19, 2023

Dear Referee #3,

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

Dear authors, the two reviews you already receive present most of my perplexities and doubts about your methodology and robusteness of conclusions.

My appreciation is that the paper presents interesting data, and could deserve publication if the treatement was better presented and justified. I only have a few additional suggestions with respect to the points rose by my colleagues.

We would like to thank you for all the valuable comments. 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 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, 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.
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 studies.

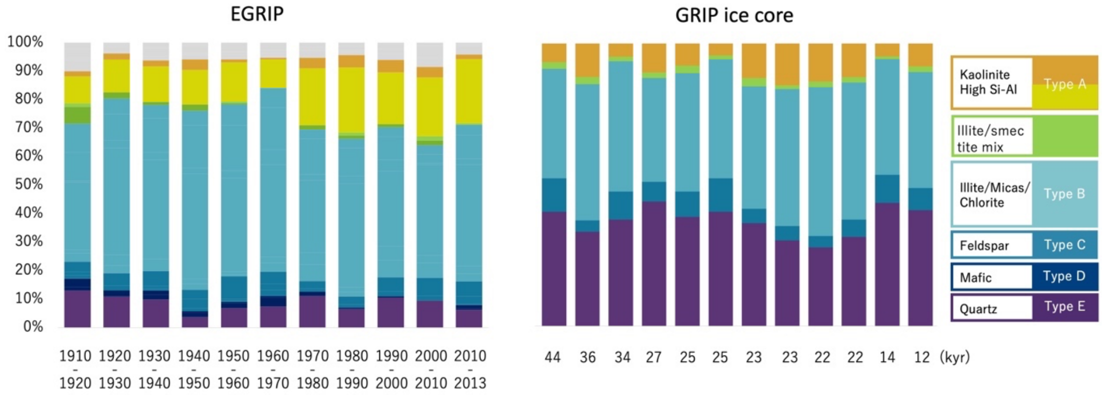

Figure Reply 1. Mineral composition of the EGRIP (this study) and GRIP ice cores (Svensson et al., 2000)

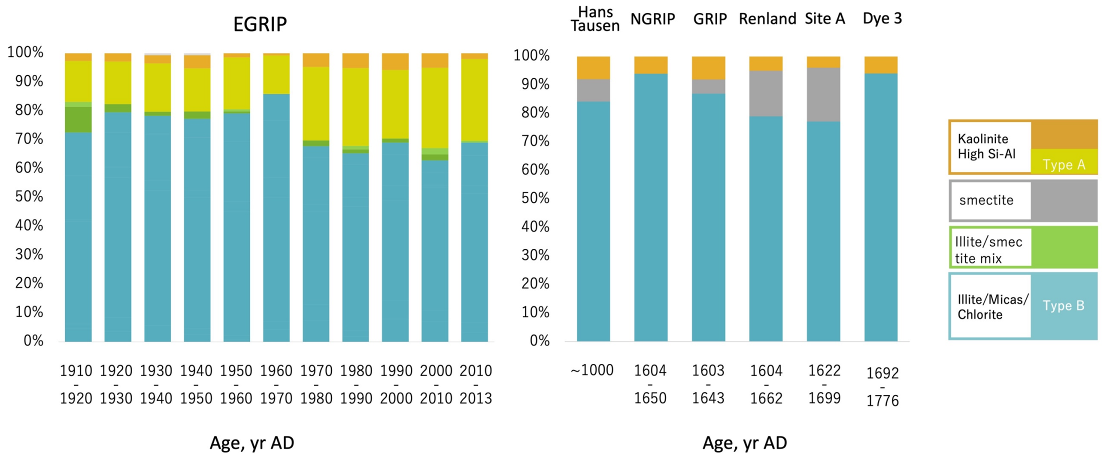

Figure Reply 2. Clay mineral composition of the EGRIP (this study) and Hans Tausen, NGRIP, GRIP, Renland, Site A, and Dye-3 ice cores (Bory et al., 2003).

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.

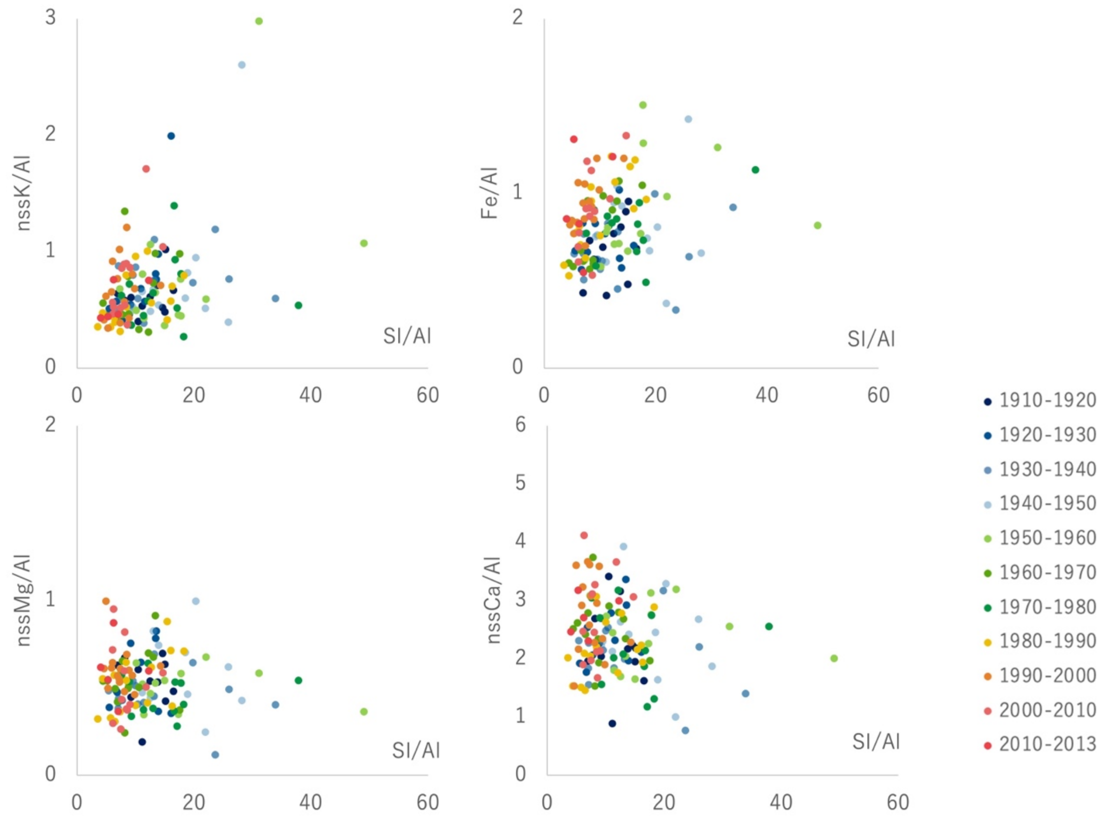

Figure Reply 3. Elemental concentration ratios of microparticles in the EGRIP ice core anayzed by ICP-MS.

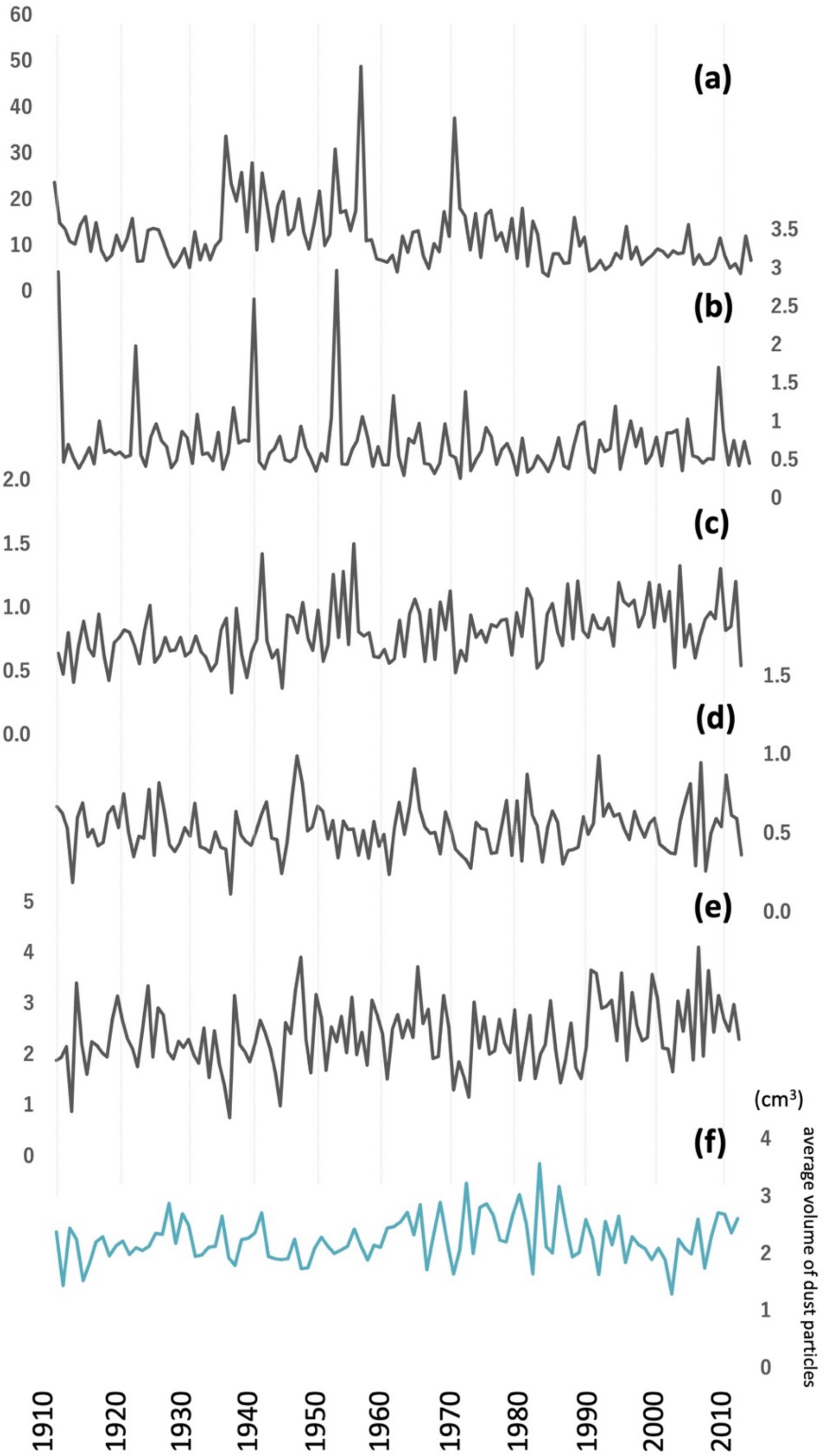

Figure Reply 4. Historical records of elemental concentration ratios ((a) Si/Al, (b) nssK/Al, (c) Fe/Al, (d) nssMg/Al, (e) nssCa/Al, and (f) average volume of dust particles in the EGRIP ice core.

Detailed comments

- is the X-ray self attenuation for light elements taken into account for the EDS analysis? this can be severe for elements such as Al when large particles are present and could false the mineralogical attribution

Yes. We consider X-ray self-attenuation using the “ZAF correction method”. The ZAF correction involves accounting for three effects on characteristic X-ray intensity during quantitative analysis: 1) the atomic number (Z) effect, 2) the absorption (A) effect, and 3) the fluorescence excitation (F) effect.

- there is too little description of the way by which the size distributions are obtained

We will add the description of how to obtain the size distribution data in the revised manuscript.

- I would suggest to present the size distribution as number of particles per unit size class and use a log scale for the x-axis

We will present figures of size distribution (Figs 4 and A2) in the description way you suggested in the revised manuscript.

- the calcium depletion could be due to size sorting and not solubility, have you considered that?

Many thanks for the suggestion. We haven’t considered the possibility of the depletion and will describe it in the revised manuscript.

- back trajectories are only showing provenance but need to be complemented by precipitation fields if you want to obtain deposition. Dry deposition can be approximated (roughly) by gravitational settling if you know the size distribution.

Does this comment mean “consider the removal effect by precipitation and by particle size dependency along the air mass pathways”? If our understanding is correct, it is difficult. The code for analysis used in the trajectory analysis can consider the precipitation only at the targeted site. We emphasize that the trajectory results show the differences of dust sources between the EGRIP and SIGMA-D site. We will not “identify” but “discuss” possible dust source areas in the revised manuscript. We think that our trajectory results are sufficient to support the differences in dust sources between the two ice core sites.

References:

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