We are grateful to Reviewer #2 for the thorough review and valuable comments on our manuscript. Our responses and the planned changes for the revision are explained below. Our replies are in blue and reviewer comments are written in black.

Comments by Reviewer #2

1. The paper by Nagatsuka and colleagues provides analysis of the size and composition of mineral dust over the last 100 years based on particles collected from the EGRIP ice core in northern eastern Greenland. The work aims to present the obtained size and compositional datasets and provides analysis to determine the sources of dust in the ice core and their temporal variation. The results are discussed in comparison to data previously published and relying on another ice core in northwestern Greenland (SIGMA-D). The main result highlighted in the paper is the lower temporal variation of dust composition in EGRIP, suggesting more local dust sources compared to SIGMA-D.

The data from this work are very interesting and deserve to be published. On the other hand, the discussion and conclusions are not supported by the analysis, and no robust evidence are provided for the source identification. Detailed reasons for this are provided in the general comments.

[**Reply 2-1**] Thank you very much for your insightful comments. We appreciate your recognition of the value of our dataset and its potential contribution to the understanding of dust provenance in Greenland. We acknowledge, however, that the discussion and conclusions in the previous version of the manuscript lacked sufficient support from the presented data, and that our interpretations regarding dust provenance were not adequately substantiated. To provide the robust evidence for the ice-core dust source identification, we will increase the number of analyzed samples and include the uncertainties in cluster composition to statistically evaluate the significance of differences in the CL data in the revised manuscript.

2. My judgment is that in the present form the paper would require major revisions to narrow the scope and/or better support conclusions. My suggestion would be to present the data (Section 3) and provide a concise and objective analysis of elements resuming the relatively long discussion now in Section 4. A "data paper" of a "measurement report" type could be more suitable for this.

[**Reply 2-2**] Thank you for your valuable comments. The issue regarding the lack of supporting evidence for our conclusion was also raised by Reviewer #1 (Comments 1-3). To address this concern, we conducted additional statistical analyses on 11 age-defined samples (1910–2013) and 13 mineral types. The results revealed statistically significant differences in mineral composition across different age groups (Chi-squared test, $p < 0.05$). Furthermore, a PERMANOVA test comparing samples from before and after 1970—a period hypothesized to reflect a shift in dust provenance—reveals a statistically significant change in mineral composition between these two periods (PERMANOVA, $p < 0.05$). These findings support the interpretation of a shift in dust source regions around 1970, which is consistent with the results suggested by SEM-CL/EDS.

Furthermore, we also conducted additional statistical analyses on elemental ratios (details provided in Reply 2-8) as well as on the relationship between dust particle size and climate oscillation indices (AMO and NAO). Regarding the latter results, we briefly note that there is a statistically significant correlation between the average volume of ice core dust and the AMO index since the 1950s, which is known to strongly influence African dust emissions. For full details, please kindly refer to our response (Reply 1-23) and Table Reply 2 to Reviewer #1 (hereafter, Reply 1-XX indicates the comment numbers in our responses to Reviewer #1). Both results further support our interpretation based on CL analysis that the primary sources of ice-core dust were likely the Asian and African deserts, and that their relative contributions may have shifted around the 1960s - 1970s. Thus, these statistically significant results provide additional support for the conclusions derived from our CL data. In the revised manuscript, we will incorporate these findings and provide a concise discussion.

To avoid long discussion in Section 4, we will remove the comparison with results obtained using different methods (see Reply 2-6).

3. I also note that in several points of the paper, including the abstract and the conclusions, the authors mention that "the findings demonstrate that the SEM-CL analysis is a valuable tool for identifying ice-core dust sources and reconstructing their variation during periods of low dust concentration". This kind of sentence mostly belongs to methodological papers dedicated to test and evaluate techniques, instead of this kind of papers that should focus on scientific interpretation of results based on well-established analysis tools. This kind of statement also question me about the most appropriate journal and targeted scope of the work.

[**Reply 2-3**] Thank you for your comment. As you correctly pointed out, such statements are indeed more appropriate for methodological papers. We included the sentence "The SEM-CL analysis is a valuable tool for identifying ice-core dust sources and reconstructing their variation during periods of low dust concentration" to highlight the novelty of our analytical approach. However, the primary aim of this study is not to establish or validate a new analytical method, but rather to identify the sources of ice-core dust and interpret their variations. Therefore, we will delete these sentences in the revised manuscript to better align with the scientific focus and scope of the paper.

We would like to emphasize that, if the reliability of the CL data can be fully demonstrated, our study would represent one of the few continuous records of Greenland ice-core dust provenance during recent decades - a period characterized by relatively low dust concentrations. These findings offer new insights into Greenland ice-core dust provenance studies and contribute to paleoenvironmental reconstruction. Furthermore, identifying not only the source regions of the ice-core dust but also its physical and chemical properties (i.e., composition and particle size), and understanding their variability is essential. This knowledge is crucial for predicting surface darkening and ice mass loss of the Greenland Ice Sheet (as discussed in Line 50-63 of the manuscript), and for advancing our understanding of ice-nucleating particles (INPs), which play an important role in cloud formation. Since the efficiency of mineral particles as INPs depends on their type (e.g., Atkinson et al., 2013), detailed mineralogical data are particularly valuable.

For these reasons, we believe that our findings provide important contributions appropriate for publication in a climate change journal. We will add this explanation to the revised manuscript to better clarify the significance and novelty of our study.

Atkinson, J., Murray, B., Woodhouse, M. et al. (2013). The importance of feldspar for ice nucleation by mineral dust in mixed-phase clouds. Nature **498**, 355–358. https://doi.org/10.1038/nature12278.

General comments

4. Section 4.1 lines 333-340 and Fig. 7: the analysis referring to Fig. 7 is not very robust. Only 5 points are considered from each decade and compared to unpublished data. Apart from the fact that unpublished datasets are considered (data which however should be made available to the scientific community prior to be used as interpretation of other data) the five points from the EGRIP core are at the edges between the West Africa and East Asia data, without – to my reading of the plot - an emerging clear pattern.

[**Reply 2-4**] [Same as Reply 1-3-1] Thank you for your valuable comments. Regarding the SEM-CL analysis results, we acknowledge that our previous manuscript did not provide sufficient detail to support our provenance conclusions. The unpublished Saharan dust data are derived from three surface marine sediment samples collected at different latitudes along the northwestern African margin: #1 (26.82°N, 15.12°W), #2 (23.21°N, 17.85°W), and #3 (19.36°N, 17.28°W). Given the prevailing trade winds that transport Saharan dust westward, these samples are considered to reliably represent the average composition of Saharan dust sources. Detailed information on the CL data of these three reference samples is presented in a separate manuscript by our co-author Dr. Nagashima, which is currently under review in *Progress in Earth and Planetary Science* (PEPS). At the time of our preprint submission, this related work had not yet been submitted, and we could only cite it as "Nagashima et al., in preparation." We apologize for the lack of clarity this may have caused. We will cite the manuscript as appropriate in the revised version, depending on its progress (e.g., Nagashima et al., submitted / Nagashima et al., 2025).

[Same as Reply 1-3-2] Regarding the limited number of data points, we initially analyzed only five samples to investigate a potential shift in dust provenance around the 1960s-1970s, given that SEM-CL analysis is highly time-consuming. However, in response to your suggestion, we will increase the number of samples subjected to CL-analysis and include estimates of uncertainties in cluster composition to statistically evaluate the significance of differences in the CL data in the revised manuscript.

5. Fig. 8 and more in general the interpretation of the clustering analysis (Sect. 4.1 and 4.2): the categorization into Types A-E described in Sect. 2.4 refers to mineral types and associate them to potential sources, but those are very broadly defined, which makes difficult to understand their usefulness for the data interpretation.

[**Reply 2-5**] [Same as Reply 1-2-1] Thank you for your valuable comments. In our previous submission (https://egusphere.copernicus.org/preprints/2023/egusphere-2023-1666/), one of our initial objectives was to compare the mineral compositions with those of the SIGMA-D ice core presented in Nagatsuka et al. (2021), and therefore we adopted a similar mineral classification (Types A-E). However, as you have pointed out, this broad classification does not allow clear differentiation of dust sources in the present study. Moreover, since this mineral classification does not significantly affect the main discussion of the manuscript, we will discontinue the classification and removed Table 1 in the revised version. Instead, to interpret the clustering analysis in Sections 4.1 and 4.2, we focus not on such categorization, but rather on changes in the chemical components that are thought to originate from minerals associated with Asian and African deserts (details in Reply 2-8).

6. Section 4.1 lines 356-367, Fig. 10: the comparison against other datasets and the similarities/differences discussed, ultimately in relation to dust origin, is difficult to evaluate due to the methodological differences mentioned, potentially affecting the reasoning and the implications of identified mineralogical patterns

[**Reply 2-6**] We fully agree with your point regarding the difficulty in evaluating the comparison due to methodological differences. In response, we will remove lines 356–367 and Fig. 10 from the revised manuscript.

7. Back trajectory analysis: I am unsure how to read and interpret the analysis of back trajectories. These are performed considering 20-days, a choice that is questionable. Indeed, this is very long, also compared to an aerosol lifetime of few days in the troposphere. Apart from the trajectory duration, I am not sure to understand how to use the information of Fig. 11, also based on the results of Fig. 2 showing that there is not a preferential direction of back trajectory air masses – which somehow can be expected as all data for more than 50 years are put together. Also attention as there is an inconsistency: in Sect. 2.6 the 20-days trajectories are mentioned, while in Fig. 11 is 25 days. Instead, Fig. 2 shows 7-days trajectories.

[**Reply 2-7**] Thank you very much for your valuable comment. We apologize for the confusion caused by inconsistencies between the figures and the methods. In our previous study (Nagatsuka et al., 2021), we conducted long-term back trajectory analyses, which initially led us to present similar results in this manuscript. However, as you correctly pointed out, 20-days back trajectories may lack reliability. In the revised manuscript, we will standardize the analysis to 7-days back trajectories.

Since the original Figure 11 was not intended to discuss daily variations in contribution rates from possible source areas, it will be removed. Instead, we will present pie charts illustrating the cumulative contribution rates from different source regions over a seven-day period in Figure 2 (we provide as Figure Reply 1 in this response letter). The contribution rates shown in these pie charts are based on the cumulative source attribution of trajectories originating from land areas other than Greenland, calculated according to the regional classifications defined in Figure 2a. The total contribution from all non-Greenland land areas is normalized to 100%, and the relative contributions of each region presented.

While the coastal regions of Greenland show the highest overall contributions, excluding these, Sigma-D exhibits a prominent contribution from North America (NA), whereas EGRIP is characterized by relatively larger contributions from Europe (EU) and Russia (RUS). Although too small to be clearly shown in the figure, the contribution from Central Asia—including China (yellow region)—was 0.03% for EGRIP and 0.0025% for Sigma-D, indicating that the contribution to EGRIP was approximately ten times greater.

8. Section 4.2 lines 40-414, Figures 12 and 13: to my reading of Fig. 12 I do not see a clear pattern in the data points, which instead seems quite dispersed. The authors should provide some statistical analysis to support their interpretation. I also do not identify an increase in Fe/Al in Fig. 13 as stated, and more statistical analysis should be provided to support this statement (line 413).

[**Reply 2-8**] [Same as Reply 1-22] Thank you for your valuable comment. In figure 13, we intend to highlight that the regression slope for samples from 1980–2013 is steeper than that for samples from 1910–1980. However, as you correctly pointed out, this trend is not immediately apparent. To clarify the temporal trend more clearly, we conducted additional linear regression analyses on elemental concentration ratios (nssMg/Al, nssK/Al, Fe/Al, and nssCa/Al), dividing the data by decade (Table Reply 1).

Regarding the relationship between Fe/Al and Si/Al, no significant correlation is observed before the 1950s (R^2 = 0.0002 – 0.2657; see Table Reply 1). However, from the 1960s to 2000s, the correlation becomes stronger, (R^2 = 0.4465 – 0.6734) and regression slopes also increase over time, suggesting a strengthening association between higher Si/Al and Fe/Al values. The results imply a growing influence of Fe-rich sources, which could include North Africa. Conversely, the nssK/Al ratio showed a generally strong positive correlation with Si/Al during 1910–1950, except in the 1930s (R^2 = 0.2385 – 0.6539; see Table Reply 1), indicating a consistent input from a common dust source. After 1960, however, this correlation weakened significantly (R^2 = 0.0029 – 0.3334), and the slope became more variable. As discussed in the manuscript, nssK in the EGRIP ice core may be derived from illite, a clay mineral typically associated with Asian dust sources (line 453-455).

Taken together, these findings suggest a shift in the dominant dust source region around the 1960s—from Asian to African origin—which is consistent with the results suggested by SEM-CL/EDS. We will add the above results and the related table to the revised manuscript.

We created Figure 12 with the same intention as Figure 13; however, we agree your comments that the increasing trend is not clearly visible. By combining Fe/Al with Si/Al, which represents a crustal origin, we believe the changes in mineral supply from different geological sources can be more clearly illustrated. Therefore, we have decided to remove Figure 12 and focus the discussion solely on Figure 13.

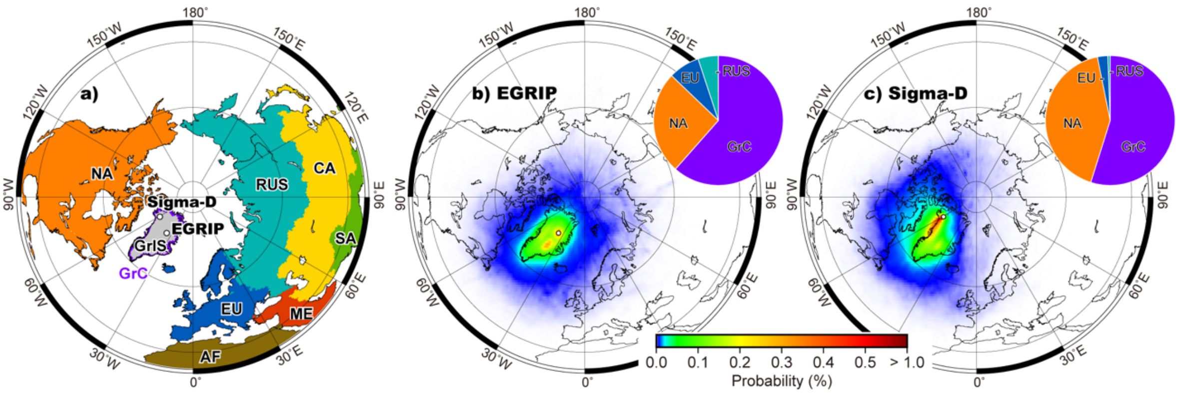

Figure Reply 1. Map showing (a) the locations of the EGRIP and SIGMA-D ice core sites in Greenland, along with nine regions used to calculate regional contributions (GrIS: Greenland Ice Sheet, grey; GrC: Greenland coast, purple; NA: North America, orange; EU: Europe, blue; RUS: Russia, light blue; CA: 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. The probability distribution was calculated by summing the air mass trajectories at 50, 500, 1000, and 1500 m above ground level. The pie charts illustrate the cumulative source contribution over the seven-day back trajectories from land regions other than Greenland, calculated according to the regional definitions shown in panel (a). The total contribution from all non-Greenland land areas is normalized to 100%, and the relative contributions of each region are presented (modified from Figure. 2).

Table Reply 1. Summary of linear regression results for the annual average elemental concentration ratios of (a) nssMg/Al, (b) nssK/Al, (c) nssCa/Al, and (d) Fe/Al versus Si/Al in the EGRIP ice core samples from 1910 to 2013. R^2 indicates the coefficient of determination for each relationship with Si/Al. Highlighted values indicate correlations with $R^2 \geq 0.5$ and statistical significance ($p < 0.05$).

Decade		nssMg/Al	nssK/Al	nssCa/Al	Fe/Al
1910s	Slope	0.0033	**0.1274**	-0.0321	0.0077
	R^2	0.0184	**0.6539**	0.0882	0.0535
1920s	Slope	-0.0016	**0.1180**	0.0355	0.0007
	R^2	0.0007	**0.5807**	0.0440	0.0002
1930s	Slope	-0.0006	0.0087	-0.0134	0.0090
	R^2	0.0013	0.0593	0.0609	0.2657
1940s	Slope	-0.0070	**0.0981**	-0.0560	-0.0168
	R^2	0.0297	**0.6148**	0.1545	0.1723
1950s	Slope	-0.0006	0.0388	0.0163	0.0132
	R^2	0.0011	0.2385	0.0408	0.0787
1960s	Slope	**0.0427**	-0.0078	0.0493	**0.0571**
	R^2	**0.6572**	0.0079	0.0876	**0.6734**
1970s	Slope	0.0024	0.0013	0.0041	**0.0147**
	R^2	0.0277	0.0029	0.0030	**0.5419**
1980s	Slope	0.0115	0.0223	0.0453	**0.0368**
	R^2	0.1196	0.2066	0.1565	**0.6098**
1990s	Slope	-0.0396	0.0661	0.0424	0.0449
	R^2	0.2525	0.3334	0.0147	0.4465
2000s	Slope	-0.0035	0.0761	0.0531	**0.0687**
	R^2	0.0024	0.3968	0.0448	**0.5208**
2010s	Slope	-0.0255	0.0208	-0.1692	-0.1032
	R^2	0.0216	0.0297	0.3336	0.1493

Detailed comments

9. Section 2.7, lines 223-226: this kind of sentences is not necessary / relevant

[**Reply 2-9**] Thank you for pointing this out. We agree with your comments and will delete the sentences.

10. Section 2.7, lines 230-235: this part can stay in supplementary information

[**Reply 2-10**] Thank you for your comment. We understand your suggestion to move the description of the CMIP6 models (lines 230–235) to the supplementary information. However, since Figure 14 - which uses snow cover fractions derived from these models—is presented in the main manuscript and discussed in the context of provenance shift of the ice-core dust, we believe a brief explanation of the models is necessary within the main manuscript. We recognize that your comment aims to reduce redundancy, so we will revise this section by removing unnecessary details to shorten the manuscript. Additionally, essential supporting information regarding the CMIP6 model data will be moved to the Data Availability section.

11. Section 2.7, lines 236-237: this is relevant to the analysis of the present paper and should be better detailed

[**Reply 2-11**] Thank you for pointing this out. We will add explanations how to calculate snow cover fraction anomalies in the northeast and southeast areas.

12. Section 3.2: the diameter is a projected-area diameter – please provide more details

[**Reply 2-12**] The projected-area diameter, referred to as the "equivalent circle diameter" in the manuscript, was measured using image-processing software (ImageJ, National Institutes of Health, USA) and represents the diameter of a circle with an area equivalent to the two-dimensional projection of the particle.

In contrast, particle size data shown in Figure 14 were obtained using a Coulter counter, which measures the equivalent spherical diameter in three dimensions. Therefore, we will also add a note explaining that the definition of particle size differs between the SEM-based measurements and those obtained using the Coulter counter.

13. Figure 4: y-axis should read “particle number concentration, x-axis should refer to “diameter” or “projected-area diameter” (if this is the case)

[**Reply 2-13**] Thank you for your comment. The Y-axis represents the number of particles (count number), not number concentration (We count 200 particles per sample, with 11 samples in total, making 2200 particles). We will revise the X-axis into “projected-area diameter”, or “equivalent circle diameter” as described in the manuscript.