Dear Referee 1,

We are grateful for your thorough and insightful review, which has substantially improved the manuscript. We have addressed each of your comments and revised the text accordingly.

Detailed responses to each point are given below, with *referee comments in italics* and our replies in plain text in blue. All revisions are also marked in blue in the attached PDF. We would be happy to provide any additional clarification you may find necessary.

Lee et al. investigated post-depositional transformations of mineral dust in the EPICA Dome C ice core using CFA–sp-ICP-TOFMS. Authors reveal depth-dependent mineral dissolution, secondary mineral formation, particle aggregation, and trace-element immobilization. Importantly, the study demonstrates that mineral dust in deep Antarctic ice is not chemically inert and may affect the preservation of paleoclimate signals. The results provide valuable insights into deep-ice geochemistry. Overall, the study is highly interesting and potentially suitable for publication after substantial revision.

1. *L6-7 "This reveals a deep-ice environment dominated by pervasive acid dissolution, leaving behind refractory mineral phases." The acidic conditions are very important. Figures 2–4 provide strong evidence for increasingly extensive mineral dissolution with depth. Some publications have already reported measuring pH in frozen samples. Can authors estimate or describe the pH variation in the ice core?*

   We thank the referee for highlighting the importance of the acidic conditions. We agree that this point deserves more attention. In the revised manuscript, we have added a short overview of previous work on acidic species within ice grains and their variability to the Introduction (lines 60-63), and we now discuss what our results imply about the variability of acidity in the deep ice in Section 3.2.2, lines 500-512. Having said that, available pH measurements in polar ice cores are most often biased due to the dissolution of CO2 and formation of carbonic acid from air bubbles in the ice or from lab air. Acidity quantification by titration has been done extremely rarely and not on the EDC samples of this study. But most importantly, the low pH values required for dissolution are only found in the grain boundaries, where the dissolution of dust occurs, and no direct pH measurements in this micro-environment exist at all. The closest we can get is a semiquantitative acidity measurement derived from the ECM measurement (solid state DC conductivity) on the ice, which is available for the core. However, even this analysis has not the high resolution required to resolve grain boundaries and it does not provide quantitative pH values.
   Accordingly, rather than measuring pH directly, we infer a plausible pH range from the formation conditions of the secondary minerals we observe: jarosite is stable only under strongly acidic conditions of approximately pH ~2–3, whereas ferric (oxyhydr)oxides form above pH ~4.

2. *In Table 1, the work analyzed 18 EDC ice-core samples, representing 10 climatic intervals. Only a single sample was available for MIS 9 or MIS 11, whereas 2 samples were available for other climatic intervals. Why?*

We thank the referee for this question. The uneven sampling reflects the limited availability of ice rather than a deliberate choice. Note that the EDC ice core has been already used for CFA of dissolved ions ("wet chemistry") in the field directly after drilling, however 20 years ago without the opportunity to measure single particles using the new ICP-TOFMS technique. Accordingly, only limited ice section still exists that provides enough ice to perform a second CFA analysis in our lab including the ICP-TOFMS. The two samples covering MIS 9 and MIS 11 (Bags 4502 and 4842) were originally selected for a detailed investigation of millimeter-scale visible dust layers, which is beyond the scope of the present study and will be reported in a separate manuscript currently in preparation. Ice adjacent to these depths could not be obtained for this study, as it had either been allocated to other projects or is archived in Antarctica. This explanation has been added to the manuscript (lines 94-97).

3. *During CFA–sp-ICP-TOFMS analysis, ice melting and sample acidification may alter particle integrity, mineral dissolution, and elemental phase partitioning. Because the mineralogical compositions of shallow and deep ice samples differ, they may respond differently to the same acidification procedure. Authors can provide more details on the sample analysis to reduce the effects of sample preparation and analytical treatment when observing depth-dependent patterns.*

   We agree that melting and acidification are unavoidable steps in the current continuous ICP setup, and that they may affect the quantification of both dissolved and particulate elemental compositions. In the revised manuscript, we therefore provide a more detailed description of the CFA system, from sample melting and injection through to acidification prior to ICP measurement (lines 128-130). In particular, we now specify: "To minimize dispersion and possible alteration of impurities (e.g., partial dissolution or adsorptive loss) during continuous sample introduction, the tubing configuration was optimized to a total transit time of only ~115 s between the melter head and the nebulizer."

   Despite these potential influences, we consider it unlikely that melting and acidification alone could generate the depth-dependent patterns we observe in elemental phase partitioning, particle size, and the composition of Al–Ca–Fe-bearing silicate particles (Fig. 2-4). The referee's concern implies that samples with different mineralogical compositions should respond differently to the same acidification procedure. We can test this expectation within the shallow section (<1100 m), where Holocene and Last Glacial samples differ substantially in mineralogy but have experienced identical treatment. The differences observed between these two intervals are considerably smaller than the systematic changes seen with increasing depth, indicating that mineralogically driven differences in the response to acidification cannot account for the observed trends. This explanation has been reflected in the manuscript (lines 242-249)

4. *L294-297 "This discrepancy is mainly attributable to two deep samples below 3000 m depth (MIS 16 and MIS 18), which deviate from the general trend (top panel, Fig. 1). Upon excluding these two samples, the correlation coefficient between the temperature*

anomaly and log-transformed optical dust PNCs strengthens to -0.85 (p = 0.007), more closely matching the elemental PNC records." However, the exclusion of the MIS 16 and MIS 18 appears to be based on their observed deviation from the regression. If possible, the authors can provide formal outlier and sensitivity analysis to demonstrate that the strengthened correlation is not specific to the selective exclusion of MIS 16 and MIS 18.

We thank the referee for this comment. We would like to clarify that our intention was not to improve the correlation by selectively removing samples, but to identify and quantify a deviation that is itself a known feature of these intervals.
Following the referee's suggestion, we have verified this quantitatively rather than visually. Examining the vertical offsets of all samples from the regression line, MIS 16 and MIS 18 are the only samples exceeding a 2.5-fold deviation (|log offset| > 0.40; +0.42 and +0.40, respectively). No sample shows a comparable negative offset, so the deviation is one-sided rather than reflecting random scatter around the temperature relationship. We have made this criterion explicit in the revised manuscript, which now describes these two samples as those "which uniquely exhibit the largest vertical offsets exceeding a 2.5-fold deviation (|log offset| > 0.40)" from the general trend (lines 352-354).

Importantly, a weakened coupling between dust particle concentration and temperature anomaly during these intervals has previously been reported (Lambert et al., 2008, and the supplement therein). Because such concentrations are derived from measured particle volumes, dust aggregates are registered as single large particles and lead to an overestimation of dust content; ultrasonication to disperse the aggregates was used in that study to help resolve this issue in their measurements. Our observation is therefore consistent with an independently documented feature of these intervals rather than representing a post-hoc exclusion specific to our dataset.

5. *To further enhance the significance of frozen reactions in ice cores, do the authors plan to analyze the species of various elements, especially iodide, iodate, arsenic, arsenate, ferrous, and ferric?*

We thank the referee for this suggestion, which we fully agree is an important direction for future work. Elemental speciation would provide direct evidence for the redox and complexation processes that we currently infer from elemental observations alone.

We plan to address this in two complementary ways. First, we intend to apply synchrotron-based X-ray techniques to obtain elemental composition together with oxidation and coordination state information at the particle scale, which would be particularly informative for the oxidation states of Fe and As. Second, if a suitable collaboration can be established, dissolved anionic species such as iodide, iodate, and arsenate could be determined by ion chromatography coupled to ICP-MS or other methods.

We specify these opportunities for future research in the Discussion part of the paper (lines 640-646).

6. *L440 "…, while oxidative conditions may further promote iodine uptake by converting iodide to triiodide and potentially to iodate (Kim et al., 2016)" In the cited ref. the triiodide was produced by oxidizing iodide in the presence of oxygen and UV irradiation, but the mineral was absent here. And iodate was not produced in Kim's ref.*

   We thank the referee for these careful observations, and we agree with both points. We have removed the reference to iodate, as Kim et al. (2016) did not report its formation, and we no longer infer oxidation of iodide beyond triiodide.
   Regarding the absence of minerals in the cited experiments, we have added Kim et al. (2019), in which iodide oxidation was examined in the presence of iron oxide minerals and is therefore more directly comparable to the mineral-bearing conditions in deep ice. Together, Kim et al. (2016, 2019) show that iodide oxidation is accelerated within the ice matrix both under irradiation and in the dark.
   The revised sentence now reads: "… , while oxidative conditions may further promote iodine uptake by converting iodide to triiodide (Kim et al., 2016, 2019)." (lines 521-522).

7. *The conclusion section should be simplified.*

   We thank the referee for this suggestion. The Conclusions have been shortened from 44 to 28 (excluding change tracking) lines by moving the implications for future work to the Discussion (lines 624-646) lines and removing explanations that are already covered in detail in the preceding sections.

   On behalf of all authors,
   Geunwoo Lee