

Replies to referees:

Referee 1

Ref1: It was my pleasure to review “Microstructure, Micro-inclusions and Mineralogy along the EGRIP ice core - Part 2: Implications for paleo-mineralogy”. This paper presents a novel dataset of impurities in a Greenland ice core. The data is interesting, and the methods properly presented. However, the introduction and discussion could be considerably improved.

The introduction brings in a great deal of previous research but doesn't make clear points about the knowledge gap that the paper is addressing. Nor does it state any hypotheses or provide much theoretical framework on which hypotheses could be made. The importance of impurities is brought up and the word important is used repeatedly. But nowhere is the link made between this particular research effort and its applications to ice mechanics, etc. Neither is the later discussions of chemistry and mineralogy at all prefigured in the introduction.

Reply: We thank the referee and value the input and ideas. We have gone through all comments and adopted most of them. Answers to the specific comments can be found below.

We changed the introduction to clarify knowledge gaps and to define our objectives more clearly. This will help to prefigure the discussion and goes as follows:

“...The impurity content plays a major role regarding the physical properties of snow, firn, and ice, such as electrical conductivity (Alley and Woods, 1996; Wolff et al., 1997), permittivity (Wilhelms et al., 1998), and mechanical properties (e.g., creep behaviour, dislocation velocity) (e.g., Jones and Glen, 1969; Dahl-Jensen and Gundestrup, 1987; Paterson, 1991; Hörhold et al., 2012). Several studies investigated the impact of impurities on the deformation of ice (e.g., Jones and Glen, 1969; Petit et al., 1987; Iliescu and Baker, 2008; Eichler et al., 2019), which in turn influences the flow of ice - a major uncertainty regarding future projections of ice sheet behaviour and solid ice discharge. This chain of processes at the micro-scale resulting eventually in the large-scale deformation behaviour of ice needs to be better understood, especially in fast flowing ice, as present in ice streams. The microstructure of ice can be impacted directly by processes related to impurities such as Zener pinning (Smith, 1948; Humphreys and Hatherly, 2004) or grain boundary drag (Alley et al., 1986, 1989). These result in changes of the energy, shape and mobility of grain boundaries. Furthermore, impurities such as micro-inclusions directly impact deformation when they form obstacles in the ice matrix and thus enhance strain localisation and the introduction of protonic defects (Glen, 1968) or the formation of dislocation lines (Weertman and Weertman, 1992). In turn, these small-scale processes can also influence climate proxies by altering the stratigraphic integrity of impurity records (Faria et al., 2010; Ng, 2021). The mineralogy of impurities, their localisation and the possible interplay between both certainly plays a role in the interaction of impurities and ice physical properties (e.g., Jones and Glen, 1969; Paterson, 1991) and ice dynamics. Reasons for the current lack in understanding of the whereabouts of impurities in polar ice are manifold, the methodology to systematically look in parallel at localisation (as a microstructural feature) and mineralogy (as a chemical feature) of micro-inclusions has just recently been developed (Eichler et al., 2019). Commonly used methods, developed to derive (paleo-) climate information on impurities such as CFA (e.g., Kaufmann et al., 2008), apply their analysis on melted sample water for the sake of decontamination and time constraints. Doing so, information on the phase and localisation of impurities are not considered. On the other hand, dedicated studies on localisation are spatially very limited and do not catch the overall chemistry of a certain sample. The mineralogy and its possible interplay for localisation was therefore not possible to investigate. However, in order to develop a profound understanding on the mineralogy and localisation of impurities in polar ice, a combination of both approaches is needed.

With our study we attempt to push forward to combine the information of both, microstructure and chemistry to elaborate on the mineralogy of micro-inclusions in polar ice, its possible sources and its interplay with microstructure. There is no investigation of the mineralogy and location of inclusions in one polar ice core with a high enough spatial resolution and reliable statistics to generalise the spatial distribution of inclusions or to investigate the effect of different minerals on ice mechanics. We aim to gain a better understanding of the mineralogical variability of micro-inclusions inside one ice core on the large (hundreds of metres) and on the small (millimetre-centimetre) scale. Was the mineralogy of the deposited impurities stable over the last 14 ka or are there changes following the evolution of the Greenland Ice Sheet? Does the mineralogy of inclusions enable a better understanding of the aerosol content of the past? Furthermore, we investigate if the location of the inclusion in the microstructure is linked to its mineralogy. Our approach is a combination of methods from microstructure and impurity research to perform a systematic high-resolution analysis along one deep ice core. The East Greenland Ice Core Project (EGRIP) ice core, the first ice core drilled through the Northeast Greenland Ice Stream (NEGIS), enables such an interdisciplinary undertaking. Stoll et al. (2021a)

investigate the location of micro-inclusions and their role regarding deformation and microstructure. In this study we investigate the mineralogy of these visible micro-inclusions. We apply bulk insoluble particle measurements from CFA and optical microscopy and Raman spectroscopy on eleven samples along the EGRIP ice core. With this combined data set covering the Holocene, the Younger Dryas, and the Bølling Allerød, i.e. the upper 1340 m, we aim to give a systematic overview of the evolution of the mineralogy of the EGRIP ice core by identifying a relevant number of micro-inclusions at equal depth intervals. We discuss the interpretation of the mineralogy of micro-inclusions in the EGRIP ice core with respect to environmental boundary conditions and survey implications for future research. Possible formation pathways of the observed minerals are briefly discussed, but are not the main focus of this study.”

We further changed the last sentence of the abstract to: “Our results show that by applying new methods the mineralogy in ice cores, its complexity and importance for localisation studies, opens new avenues for understanding the role of impurities in ice cores.”

Ref1: Without any research questions initially laid out, the discussion wallows in vague competing hypotheses without definitively defending any point of view. A great deal of the text is dedicated to hypothetical future research, methodological shortcomings, and irrelevant statements about the general state of the science. The text lacks depth of analysis in chemistry or mineralogy. Particularly, the authors need to defend a theory of how the minerals they analysed formed. The questions of whether the sulfate and nitrate minerals are atmospherically formed or formed in ice and snow is not even properly raised, much less resolved. The composition of dissolved ions should be known from the continuous flow analysis, but it isn’t included.

Reply: We understand, that the focus of the study became not clear and thus changed the introduction (see above). We clarify, that we use the dust particle concentration generated by CFA, but not CFA data (which are not the focus of this study and will be made available eventually by future studies). This is now changed throughout the text in similar ways to the abstract: “We use dust particle concentration, optical microscopy and Cryo-Raman spectroscopy to...”

We here combine different approaches from different fields, which makes it difficult to discuss all aspects in depth. We appreciate the comment and changed the text at several places to deliver more depth regarding e.g., formation of minerals. These changes are shown in detail below.

Unfortunately, it is not trivial to identify how the identified minerals formed, especially with Raman spectroscopy. We aim to identify the microstructural location and mineralogy of inclusions, deciphering the formation, possible transport and depositional histories require completely different methods and is far beyond the scope and possibilities of this study (and thus not mentioned as an objective). This led us to the discussion of the “state of science”, it is crucial to improve the systematic combination of different methods and analysed scales. The results of this study indicate the need to investigate this complex subject further, i.e. by performing dedicated studies on chemical reactions inside the ice.

As described below in more detail we enhanced the manuscript regarding the formation of minerals and added a new section on it. To clarify this we now finish the introduction as follows:

“Possible formation pathways of the observed minerals are briefly discussed, but are not the main focus of this study.”

Ref1: The text also needs to say more about the rare minerals. How confident are the authors that they really found jacobsonite, pyromorphite, kröhnkite, etc.? These are not common minerals, so this could be misidentification of the spectral patterns. If the authors are confident, then some sort of discussion is needed about whether these are detrital (i.e. you got lucky and found a rare mineral in dust) or authigenic, representing some sort of chemical pathway found specially in the ice core.

Reply: The mentioned minerals are indeed not as common as quartz or feldspar. They nevertheless have been found on all continents (see <https://www.mindat.org>). These minerals are assumed to be transported with dust and are thus detrital. This is one of the vastest studies about the mineralogy of micro-inclusions in polar ice0. We are thus confident that it is possible to find them in the EGRIP ice core, especially at the investigated depths with relatively high particle numbers.

From a mineral perspective, the occurrence of these minerals is sometimes supported by the associated mineralogy (RRUFF database, handbook of mineralogy). For example, jacobsonite is often associated with hematite, which is mirrored in sample 2 (1 jacobsonite, 11 hematite). Still, the very low absolute and relative numbers of pyromorphite, epidote, jacobsonite and prehnite (they only account for 7 of all 791 identified inclusions) do not interfere with our overall study result on the mineralogy.

In summary, we conclude that these rare minerals are transported onto the ice together with more common

dust minerals and add the following text to the discussion: “The secondary minerals pyromorphite and epidote were each only found once. It is likely that they are of detrital origin and were transported together with more common dust minerals. The micrometre size of the inclusions indicates a distant source region comparable to the source regions of dust, such as the deserts of Asia (Bory et al., 2003; Vallelonga and Svensson, 2014). Pyromorphite is usually found in the oxidised zones of lead ore deposits. Epidote is abundant in e.g., schistose rocks or marble. It can be a product of the hydrothermal alteration of minerals in igneous rocks, such as feldspar or mica. Prehnite was identified three times and is normally found in Mg- or Fe-rich (i.e. mafic) volcanic rocks and can be a product of hydrothermal alteration in chasms or veins. Jacobsite was found twice (S2 and S5) and can occur as a primary phase or as a product of the metamorphism of manganese deposits and thus, altered manganese minerals. It is typically associated with, among others, hematite, both were identified in S2.”.

Ref1: In short this is a study in search of a research question. If the authors think they have answered a scientific question, they need to state the question and state the answer. If they don't think they've learned anything from their work, they should fold this data into part one.

Reply: As from above comments we understand that the major focus of this study was not made clear. We changed the introduction (see above) and the discussion (see below).

Ref1: Line 3: “Poorly understood” is a catch-all phrase. Can you be more specific about the gap in knowledge you are trying to address?

Reply: We added the following text to be more specific:

“Impurities in polar ice do not only allow the reconstruction of past atmospheric aerosol concentration, but also influence the physical properties of the ice. However, the localisation of impurities inside the microstructure is still under debate and little is known about the mineralogy of solid inclusions. In particular, the general mineralogical diversity throughout one ice core and the specific distribution inside the microstructure is poorly investigated, the impact of the mineralogy on the localisation of inclusions and other processes is thus hardly known.”

Ref1: Line 3: Should “Continuous Flow Analysis” be capitalised?

Reply: There are publications using the capitalized version, such as McConell et al. (2006), Eichler et al. (2019), and it is thus a question of style. We can change it, but leave the final decision to the editor.

Ref1: Line 19: This sentence needs to be revised – the double usage of “chemical compounds” is awkward and confusing.

Reply: We deleted the double usage of chemical compounds.

Ref1: Line 20: “Also”? Is this different from their “atmospheric, marine, terrestrial or biological origin”?

Reply: We deleted “also”.

Ref1: Line 22: Peroxide needs a citation.

Reply: We added Legrand and Mayweski (1997).

Ref1: Line 22: This sentence is fairly tautological.

Reply: We changed the sentence to “Insoluble impurities are rejected from the ice lattice (Ashby, 1969; Alley et al., 1986) and range in size from sub-micrometre to hundreds of micrometres (Steffensen, 1997; Wegner et al., 2015; Simonsen et al., 2019).”

Ref1: Line 26: Ferrosilite, wollastonite, and troilite are not detected components of the dust!

Reply: We changed the examples to detected components of dust: “...chemical compounds, such as SiO₂ or Fe₂O₃”.

Ref1: Line 27: Are salts a subject of this study or not? In the previous sentence, you said it was mainly mineral dust.

Reply: Salts, such as sulphates or nitrates, are a subject of this study. In the previous sentence we said “mainly consist of mineral dust”, salts are thus not excluded in the addressed sentence. However, we changed the sentence to: “Insoluble impurities normally originate from terrestrial sources and mainly consist of salts (e.g.,

sulphates and nitrates) and mineral dust which is abundant in elements from the crust, such as...”
To be clearer we changed “insoluble” to “water-insoluble” throughout the text.

Ref1: Line 29: The text about the ice lattice needs to be moved upward to where you previously discuss the ice lattice.

Reply: We moved it upwards as an introductory sentence to impurities.

Ref1: Line 31: Separate the tephra and the sulfate into different sentences.

Reply: We changed it to:” Tephra layers provide a record of local and distal volcanic eruptions and offer a chronological correlation between ice cores. The sulphate record can provide time lines and information about the atmospheric impact of volcanic eruptions. Furthermore, the isotopic and chemical composition of impurities enables the investigation of climatic and atmospheric processes of the past.”

Ref1: Line 34: You already said this.

Reply: We changed the sentence to:” Impurities in polar ice can be measured with a variety of methods, each with certain limitations.”

Ref1: Line 37: What is CFA? (acronym used without definition)

Reply: We fixed this issued and define CFA now in this line.

Ref1: Line 43: What do you mean by mechanical properties?

Reply: By mechanical properties we refer to studies by e.g., Jones & Glen (1969) investigating creep behaviour of pure and “doped” ice at different temperatures, activation energy of creep, dislocation velocity, stress-strain curves etc.

To clarify this we changed the text to: “and mechanical properties (e.g., creep behaviour, dislocation velocity)”.

Ref1: Line 44: Is this another area or a consequence of the control of impurities on mechanical properties?

Reply: This is of course related, we thus changed the text to:

“Several studies investigated the impact of impurities on the deformation of ice (e.g., Jones and Glen, 1969; Petit et al., 1987; Iliescu and Baker, 2008; Eichler et al., 2019), which in turn influences the flow of ice - a major uncertainty regarding future projections of ice sheet behaviour and solid ice discharge.”

Ref1: Line 53: Say something more. Important for what?

Reply: We changed this sentence to:

The mineralogy of impurities, their localisation and the possible interplay between both certainly plays a role in the interaction of impurities and ice physical properties (e.g., Jones and Glen, 1969; Paterson, 1991) and ice dynamics. Reasons for the current lack in understanding of the whereabouts of impurities in polar ice are manifold, the methodology to systematically look in parallel at localisation (as a microstructural feature) and mineralogy (as a chemical feature) of micro-inclusions has just recently been developed (e.g., Eichler et al., 2019).

Ref1: Lines 68-72: Maybe rewrite this to be less negative about previous work.

Reply: We changed this to “All these studies are demanding in time and costly and thus often rely on a small number of samples taken at arbitrary depths. The number of identified inclusions are thus limited and the underlying processes are not fully understood yet. Developing generalisations of the mineralogy and localisation of impurities in polar ice is hence still challenging (Stoll et al., 2021).

Ref1: Line 80: What companion study? Where is the citation?

Reply: With both preprints being available we are now able to add the proper citations in both manuscripts, which was not possible at the time of submission.

“Companion paper” has been changed to Stoll et al. 2021a throughout the text.

Ref1: Line 91: Unique is a dangerous word to use.

Reply: Indeed it is, however EGRIP is the very first deep ice core from an ice stream. We changed the wording to “an exclusive possibility”.

Ref1: Lines 144-152: This should probably have some citations.

Reply: We added the following citations: "The Raman effect is the inelastic scattering of light caused by the excitation of vibrational modes of crystals or molecules. This results in the Raman shift – a loss of scattered light energy, which is specific for each vibrational mode (Raman and Krishnan, 1928). The Raman shift is used to identify chemical impurities in samples in a non-destructive way – Raman spectroscopy. It is well suited for light-transparent materials, such as ice (e.g., Fukazawa et al., 1998; Weikusat et al., 2012; Eichler et al., 2019)."

Ref1: Lines 165-176: I would move this paragraph upwards. Some of the prior details about which peaks were distinguishable etc. could be moved to results.

Reply: We agree and moved this paragraph upwards. We would like to keep the details about peaks etc. in the methods and leave this choice to the editor.

Ref1: Line 195: Can these impurity maps be made available in a supplemental file?

Reply: Impurity maps are available as original .tiff files on PANGAEA. They are also discussed in more detail in Stoll et al. (2021a).

Ref1: Line 203: Why not include all 26 species? It's not like 26 lines is too long for a table.

Reply: In this table we want to focus on the most abundant species allowing some statistics, e. g., regarding grain boundaries. A table of all 26 species is indeed interesting, but was assumed to be repetitive to Figure 4 and the text. We suggest to keep the table on the most abundant minerals and provide a table with all species in the supplement (see below).

Table A1. Raman spectra in EGRIP Holocene ice.

Mineral	Number	Formula
Gypsum	170	$CaSO_4 * 2H_2O$
Quartz	126	SiO_2
Mica undefined	42	$(K, Na, Ca, NH_4)Al_2(Si_3Al)O_{10}(OH)_2$
Feldspar	67	$(K, Na, Ca, NH_4)(Al/Si)_4O_8$
Mg-sulphate	64	$MgSO_4$
Bloedite	61	$Na_2Mg(SO_4)_2 * 4H_2O$
Nitrates	36	XNO_3
Hematite	33	Fe_2O_3
K-sulphate	31	K_2SO_4
Krohnkite	27	$Na_2Cu(SO_4)_2 * 2H_2O$
Mica (Phlogopite)	25	$KMg_3(Si_3Al)O_{10}(OH, F)_2$
Sulphate undefined	24	XSO_4
Carbonous	22	C
Dolomite	17	$CaMg(CO_3)_2$
Mica (Muscovite)	25	$KAl_2(Si_3Al)O_{10}(OH, F)_2$
Na-sulphate	9	$NaSO_4$
Anatase	8	TiO_2
Rutile	3	TiO_2
Mica (Paragonite)	3	$NaAl_2(Si_3Al)O_{10}(OH)_2$
Prehnite	3	$Ca_2Al_2Si_3O_{10}(OH)_2$
Na-nitrates	3	$NaNO_3$
Jacobsite	2	$MnFe_2O_4$
Titanite	2	$CaTiSiO_5$
Epidote	1	$Ca_2(Fe/Al)Al_2(Si_2O_7)(SiO_4)O(OH)$
Pyromorphite	1	$Pb_5(PO_4)_3Cl$
Air	1	O_2
Undefined	17	?

Ref1: Figure 4: The color scheme for this figure is strange. I would cluster the minerals by type rather than alphabetically. The color coding needs to relate to how you discuss minerals later in the text. I.e. all minerals that you presume to source from dust need to be clumped together and similarly colored. Sulfates and nitrates and black carbon should each be their own unique colors that cannot be confused with other minerals. Similar minerals should be next to each other with similar colors. I.e. rutile and anatase, "mica" and prehnite, etc.

Also, why is graph a normalised to 100% but graph b presents absolute values.

Also, might it be possible to have time on the top axis to counter depth on the bottom axis? And, I think two decimal points for depth is excessive.

Reply: We agree that Fig. 4 can be displayed in a clearer way and appreciate the specific comments. We tried out different colour codes (while keeping it colourblind-friendly) which turned out to be quite difficult due to

the amount of different mineral types.

We applied your input and changed the figure. Minerals are now grouped regarding their mineralogy/source, there is however a limitation to the possibilities in representing all mineral species while keeping a colourblind-friendly and scientifically useful (Kramer et al., 2020) colour code.

A) and B) both display information about the absolute values (n=), but there is a difference. A) shows all identified inclusions while b) only shows the identified sulphates.

Showing the absolute values in B) helps to visualize and clarify that there are other, non-sulphate minerals at these depths too. Showing two plots, each displaying 100%, could be confusing. However, we can change it to relative amounts if the editor decides this might be better.

We added a new Fig. 1, which shows time on the y-axis. The samples are now called S1-S11 increasing readability (see answer to referee 2).

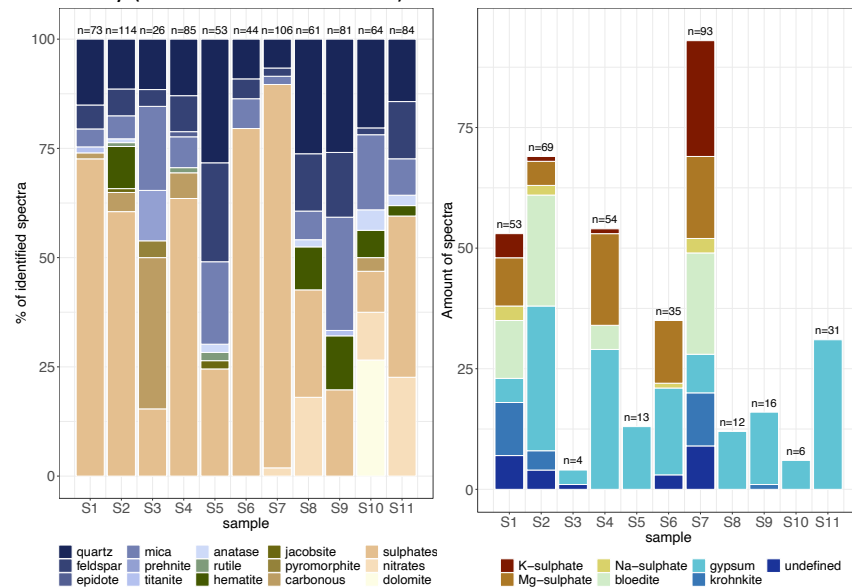


Figure 1 Identified Raman spectra of micro-inclusions in EGRIP Holocene ice, n is the total amount of identified spectra per sample. The two deepest samples are from the Younger Dryas and Bølling Allerød and have by far the highest water-insoluble particle concentrations. A) All identified Raman spectra per sample. For better visibility some Raman spectra are condensed in groups (e.g., sulphates and mica). B) Identified sulphates in detail. Sulphate diversity decreases below 900 m.

Ref1: Table 2: Please clarify why some species are identified by mineral name and others aren't. Is it unclear which magnesium sulfate mineral is present, for instance?

Reply: This is clarified in the main text, but it is a good idea to add such an explanation in the table header. The exact mineral couldn't be identified: "Mg- and K-sulphate minerals could not be identified clearly."

Ref1: Line 209 (and elsewhere): Terms like carbonate and sulfate can be confusing when applied in the fashion. It's probably better to say "the carbonate mineral ...". Though in this case, everyone should know that dolomite is a carbonate mineral, as you've included the chemical formula.

Reply: To avoid confusion we changed the text to "Sulphate minerals (from now on "sulphates") can be..." and "carbonate mineral dolomite".

Ref1: Line 210 (and elsewhere): The "might be" has me a little worried. Maybe in table 2 you can add a column for alternate possibilities for the species where identification is in doubt. Or maybe create a supplemental file that lays out the alternative possibilities.

Reply: As with many other methods there is no 100% certainty with Raman spectroscopy since spectra can be similar and the boundary conditions (especially in ice) have an impact on the clarity of the derived spectrum. We excluded all spectra of high uncertainty and "might be" should only clarify this small inevitableness. To avoid confusion, we deleted "might be".

Pyromorphite and epidote were only found once, jacobsite only twice. It is thus impossible to compare their spectra with many other examples as normally done. As discussed above, they only represent a tiny fraction of the overall mineralogy and discussing them in detail would evoke a wrong impression. Table 2 is already very wide, we thus leave it like it is.

Ref1: Line 267: In what sense is this data accompanying?

Reply: We changed the sentence to: "Applying different methods (CFA, microstructure-mapping and Raman spectroscopy) enabled us to create high-resolution impurity maps showing that sulphates and terrestrial dust are the most abundant minerals."

Ref1: Line 321: This is confusing to me. How is going from sulfates and dust to dust and sulfates (i.e. gypsum) a meaningful shift?

Reply: We find a change in mineralogy with depth. First, the occurrence of sulphates is changing: with different types of sulphates in the upper part and only gypsum in the lower. Second, we do find dolomite and nitrates only in the lower part, but not in the upper. These findings indicate a different mineralogy in these two intervals. Using the wording "shift" can be misleading, we thus replaced it with "change in mineralogy": „We have observed a pronounced change in mineralogy from a sulphate-diverse, while terrestrial dust rich, regime in the upper 900 m to a terrestrial dust-dominated regime with partially high amounts of gypsum (Fig. 5)."

"Even though we find this difference in mineralogy between the upper and lower part, this change does not correlate with a major change of the climate period, i.e. from Glacial to Holocene ice."

Ref1: Line 325: What does it mean to expose mineral diversity in detail?

Reply: The intention was to strengthen the previous sentence, stating the findings support and deepen the previous ones. We deleted this sentence.

Ref1: Line 326: You already said this in the previous paragraph.

Reply: We have restructured the discussion to clarify the different aspects of our findings. We hope this will clarify the content of each paragraph. The mentioned sentence was changed to: "The most common minerals, i.e. sulphates and terrestrial dust, are found throughout the entire ice core, but in varying abundance (Fig. 5 and simplified in Fig. 9)."

Ref1: Line 330: This makes no sense in terms of charge balance and stoichiometry. Sulfate can't just blow in without any kind of attached cation. It could be deposited as acidic aerosols of sulfuric acid (i.e. either H₂SO₄ or SO₃) and then gain its cations from other sources. Or it is deposited as a sulfate-bearing compound (gypsum, etc.). You need to explain the full chemical sequence of events that you are proposing. If it was acidic and then exchanged with chloride salts and precipitated, you would be left with a highly acidic chloride rich brine (i.e. HCl), probably in the grain boundaries of the ice crystals. If you really think that there is HCl in ice cores, by all means defend that point.

Reply: We think this is a misunderstanding, we do not state that sulphates just blow in without a cation. Ohno et al. (2005, 2006) suggest that the process you describe could occur in ice cores resulting in Cl and HCl entering the ice lattice and creating point defects. Unfortunately, Raman spectroscopy is not able to measure HCl in solid ice since HCl is expected to be a solution of monoatomic ions without vibrational spectra. Cl however was found in ice in several studies (e.g., Barnes et al. 2002a, Cullen and Baker, 2002, Obbard et al., 2003). Ohno et al. (2005, 2006) and we can thus only mention that these reactions could be possible since we (and them) found several sulphates with the cations Na⁺ or Mg⁺. This can be explained by the difference in Ca²⁺-concentration in the atmosphere during the Holocene and the Glacial as discussed by Röthlisberger et al. (2003). They state that acid-gas particles, such as H₂SO₄, reacted with sea salt aerosols during transport and in the snowpack while the acids were neutralized by terrestrial dust, i.e. Ca²⁺, in the Glacial.

We changed the paragraph to clarify this:

„The high abundance of sulphates in many samples can be largely explained by blown-in sulphate-bearing compounds, such as gypsum. However, it is suggested that sulphate minerals can also form after initial deposition when acidic aerosols of sulfuric acid, such as H₂SO₄ droplets, gain cations from other sources (e.g., Ohno et al., 2005). The main cations of the sulphate minerals in our samples are Ca²⁺, Mg²⁺, and Na⁺ (Table 2). Ohno et al. (2005) suggest that Mg²⁺, and Na⁺ could originate from blown-in sea salt (NaCl and MgCl₂), which was deposited on the ice sheet. This would result in the possibility that HCl and Cl enter the ice lattice creating point defects and thus effecting the electric conductivity (Petrenko and Whitworth, 1999; Ohno et al., 2005) and dislocation mobility (e.g., Hu et al., 1995). HCl solution is monoatomic and without a vibrational spectrum and thus not detectable with Raman spectroscopy. We are thus limited in evaluating the process of sulphate formation, but our results imply that the process proposed by Ohno et al. (2005) could occur. However, deeper samples show smaller amounts, and varieties, of sulphates, which almost completely disappear below 900 m. In the four deepest samples gypsum is the only sulphate, except for one Krohnkite micro-inclusion in S9.

Data was scarce until now, but e.g., Sakurai et al. (2009, 2011) and Eichler et al. (2019) show that sulphates are found in deeper parts of ice sheets, also consisting almost entirely of gypsum supporting our results. One possible explanation is that during the Holocene sea salt aerosols reacted with acid-gas particles during transport and in the snowpack. In the Glacial however acids were often neutralised by the higher amounts of terrestrial dust, i.e. with high concentrations of Ca²⁺, and thus reacted less with sea salt (Röthlisberger, 2003). Fittingly, our results show that high dust concentrations correlate with the dominance of gypsum over other sulphate minerals while lower dust values correlate with medium to high Na- and Mg-sulphate.“

Ref1: Line 334: Is data scarce? This paper certainly made it less scarce.

Reply: Thank you. We changed the sentence to: “Data was scarce until now, but...”

Ref1: Line 345: A closer investigation? Were they investigated at all?

Reply: We started investigating samples from the Glacial, but this is not part of this manuscript.

We thus changed the sentence to: “An investigation of EGRIP glacial samples will show if prominent changes in mineralogy also occur in deeper depth than the analysed upper 1340 m.”

Ref1: Section 4.2.2: This might benefit by some sort of figure where you plot your data against that of Sakurai et al.

Reply: This is very true and we would like to follow up on this. However, the data available online only shows the total amount of inclusions analysed with Raman spectroscopy (which sometimes differs strongly from the number of identified inclusions). Plotting our data against their data is thus unfortunately not possible.

Ref1: Line 349: Awkward writing.

Reply: We agree and changed the sentence to: “In Greenland, the GRIP ice core was analysed with Raman spectroscopy and EDS by Sakurai et al. (2009). The GRIP and EGRIP drill sites are located relatively close by, we thus compare both cores here in detail.”

Ref1: 359: Didn't you intentionally select dust rich sections? Did Sakurai et al. do the same? If not, there's your explanation.

Reply: Unfortunately, this is not mentioned in Sakurai et al. (2009). They only mention the depth and age of their samples. We are thus not able to check this hypothesis, but this could be a part of the explanation.

We added it in the following way: “The higher amount of terrestrial minerals in our samples is difficult to explain conclusively, because the GRIP and EGRIP sites are comparably close to each other and in similar distances to the coast. Intentionally choosing samples with a high insoluble particle content could partly explain this observed difference.”

Ref1: 374: I think you actually have a good number of observations. I don't think this section needs to be as self-critical.

Reply: Thank you, we changed this section to: “Some Raman spectra from a depth of 415.3 m have been difficult to identify, we thus had to exclude several spectra.”

Ref1: Line 380: What about XRD? That's usually quite a reliable way to do mineralogy.

Reply: This is true, but XRD does not provide information about the location of the inclusions, which SEM and LA-ICP-MS do. We thus suggested these two methods.

We clarified this by changing the sentence: “A partial solution would be an analysis of the same samples with a subsidiary method which identifies location and chemistry of impurities, such as SEM coupled with EDS or LA-ICP-MS.”

Ref1: Line 385: I don't understand the methodological explanation.

Reply: We deleted this part of the sentence since we discuss the issue of statistics in a different paragraph. The text is now: “This can partly be explained by different deposition conditions or by chemical reactions taking place in the ice.”

Ref1: Line 388: Iizuka et al. say that the sulfatisation is proportional the dust flux, which makes sense because it gives the atmospheric sulfuric acid something to react with.

Reply: This is true, to clarify we changed the text to:

“Iizuka et al. (2012) concluded that aerosol sulfatisation in Antarctica is proportional to the dust flux and more likely to occur in the atmosphere or during fallout than after deposition.”

Ref1: Line 394: Why would dry deposited sulfates have such diverse mineralogy? Where are the sulfates coming from? The question isn't really explained or addressed.

Reply: We here refer to the finding of high sulphate concentration in relation to wind crusts, indicating dry deposition of sulphate (Hoshina et al., 2014, Moser et al., 2020). Assuming that in periods of no accumulation (i.e. period of crust formation) sulphate aerosols are accumulated from dry deposition at the surface in layers with high concentration and get buried with time. We speculate, that these layers formed at the surface can be one source of the observed concentration of sulphates in deep ice. In the addressed sentence dry deposition is not suggested as a direct cause for mineral diversity, but as an explanation for the clustering and layering of sulphates in some samples (“The strong layering and clustering of sulphates observed in some samples could originate from dry deposition events, which form deposition crusts mainly containing sulphates.”)

Ref1: Line 398: There's quite a lot of reference to future work here. I would rather have you focus on what you can infer from this work.

Reply: We restructured the discussion to strengthen our findings as shown with examples within this reply. We merge and sharpen section 4.3 and 4.4 into one section, which enables a stronger focus on our work. With the increasing number of publications related to EGRIP, referring to findings get easier. We now e.g., can include the study by Gerber et al. (2021) in the methods and discussion. Showing the new structure is not possible within this reply, but below are some examples:

“A difference in the chemistry at the ice sheet surface at the time of deposition and different atmospheric circulation patterns, and thus varying aerosol input, could be indicated by the micro particle record (Fig. 1). Dust particle numbers generally increase with depth (Fig. 2) and especially S10 is from a dust-rich period, the Younger Dryas.”

“Accumulation rates were highest 7.8 kyr ago (0.249 ma^{-1}) and decreased towards the Last Glacial Period with a peak during the Bølling Allerød. Due to the flow of NEGIS ice from the last 8 kyr was deposited under increasingly higher accumulation rates with increasing age caused by higher precipitation closer to the ice divide (Gerber et al., 2021).”

“Gerber et al. (2021) propose that Last Glacial Period ice was deposited 197 to 332 km upstream from EastGRIP.”

“Our analysed samples were deposited within 197 km upstream from EGRIP and thus at slightly higher surface elevations ($2993 \pm 7 \text{ m a.s.l.}$ at 1400 m depth) (Gerber et al., 2021), which limits the impact on the aerosol input. Accumulation rates for ice from depths of 900 to 1400 m were low, except the peak during the Bølling Allerød. This peak coincides with the high I_{var} value in S11 displaying that a high accumulation rate enhances mineral diversity in this climatic period, contrary to the Holocene. However, it is difficult to compare the Holocene samples to the two samples from the Younger Dryas and Bølling Allerød. A systematic follow-up study on EGRIP Glacial ice is needed to investigate if the observed trends, e.g., of mineral diversity, continue with depth.”

Ref1: Line 404: What do Fe minerals have to do with anything? I don't follow the logic of this paragraph. I.e. are you claiming that Fe is a sign of authigenic mineral formation? If so, which minerals, which reaction pathways?

Reply: We here aim at discuss different studies investigating this idea and Fe-bearing minerals and their reactions inside the ice have been analysed in several publications recently. The recent works by Baccolo et al. investigate possibilities of ice as a geochemical reactor using Fe-minerals as examples (see Baccolo et al. 2018 and Baccolo et al., TCD). Baccolo et al. (2021) discuss the formation of jarosite in the Talos Dome ice core and interpret it as a product of weathering involving acidic atmospheric aerosols and dust. Since we found Fe-minerals at several depths this is of direct relevance. Contrary to Baccolo et al. (2018, TCD) our method and data do not aim to investigate reaction pathway. An in-depth discussion of the reaction pathways or mineral formation is thus not possible, but it would be wrong to intercept the on-going progress in this field. Similar in-depth studies would be needed to investigate the possible reactions occurring in ice, this goes well beyond the scope of this study.

To clarify this we changed the text to: “Fe-minerals, such as hematite and jarosite, were identified in this study and occur relatively often in polar ice (e.g., Baccolo et al., 2018; Eichler et al., 2019; Baccolo et al., 2021). Baccolo et al. (2018) showed that the deep ice sheet environment is not anoxic and dissolved oxygen and liquid

water veins might support the oxidation and dissolution of specific mineral phases. Investigating such specific chemical reactions of Fe-minerals is beyond the scope of this study, but they display the lack of knowledge regarding ice as a chemical reactor. Furthermore, Faria et al. (2010) observed the formation of solid inclusions in deep ice, which was supported by Eichler et al. (2019). A local mixing of impurities in shear bands with high strain rate and strain could explain our observations of clustering of sulphates and is not unlikely due to the dynamic conditions inside NEGIS. Local small-scale processes could lead to preferred clustering of micro-inclusions with similar chemistry. We observed preferred clustering at all depths, however samples below 900 m show significantly fewer clusters. This correlates with the depth of declining sulphate-diversity and could indicate that, around this depth, certain unknown chemical reactions occur or that large-scale boundary conditions, such as climate or ice sheet extent, changed during the time of deposition”.

Ref1: Line 418: Typography

Reply: Corrected.

Ref1: Line 431: What is the point of this whole paragraph if you only conclude that this unlikely to be relevant and untested?

Reply: In order to include the possible upstream effect on our findings we include the topic. For clarity, the section was re-arranged and work by Gerber et al. (2021) was included (see above).

Ref1: Line 435: What do you mean it is unlikely? Either it is observed, or it isn't. This constant reference to non-existent studies is really grating.

Reply: We refer to studies by eg., Svensson et al., 2000, Bory et al., 2003, Mojtabavi et al. 2020 and do a detailed comparison with the results from Sakurai et al., 2009 in Sect. 4.2.2. The recent publication of Gerber et al. (2021) addressing the upstream effect of EGRIP has been included in the text as well.

Ref1: Section 4.5: This entire section is pointless. There are no research questions addressed here.

Reply: The addressed research questions in both manuscripts lead to this section. We deleted l. 457-463 to focus on suggestions for upcoming work. We think an outlook is important in this context, but we can delete this section if demanded by the editor.

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