We thank the reviewer for their thoughtful comments on our manuscript. Please find our responses below in yellow, with changes to the manuscript indicated in **bold**.

This is an interesting effort to measure and discuss the PFAS concentrations and fluxes at a multidecadal scale from an ice core in the high arctic. The topic is of great importance, and the data set may be unique. I disagree with an important fraction of the discussion (see below). Briefly, important issues are, i) a better assessment of discussion of blanks, ii) a complete description of the data set, iii) a different discussion of air mass back trajectories, iv) a re-evaluation of the processes responsible to transfer PFAS from the air to snow, including the post depositional processes and the assessment of the role of sea-salt aerosol. Therefore, I think that this manuscript needs major revision before it can be accepted.

1. Page 4, line 5. The assessment of blanks is weak. Blanks were performed for the cartridges, but this only covers the potential contamination during analysis in the laboratory. There is no assessment of field blanks, as samples could be contaminated during sampling, handling, transport, and sectioning. This needs some comments.

Response: We thank the Reviewer for their comment concerning the assessment of blanks. We agree with the Reviewer that an ideal field blank would be able to provide background contamination contributed by the ice core extrusion and sectioning. However, the unique procedure of extruding an ice core does not provide a straightforward approach to capturing blank contamination caused by such sampling. A literature search of other ice core sampling studies confirms that field sampling blanks to encompass the sampling acquisition and handling are not typical (Table 1).

Table 1. Literature Survey on Use of Blanks in Recent Ice Core Sampling

Location	Ice Core Sampling	Blanks	Analytes	Reference
Southeast Greenland	90 m core	No field blanks; only reagent lab blanks.	$CH_3SO_3^-$, Cl^-, SO_4^{2-}, NO_3^-, Na^+, Ca^{2+}, NH_4^+, Mg^{2+}, and K^+	(Iizuka et al., 2018)
Svalbard,Norway	5 m core using a Kovacs drill	No field blanks; only reagent lab blanks.	Amino acids and chlorophyll A	(Barbaro et al., 2017)
Siberian Altai	139 m core	No field blanks; only reagent lab blanks	Mercury	(Eyrikh et al., 2017)
Svalbard, Norway	125 m core using a PICO drill	No field blanks; only reagent lab blanks	Elemental carbon	(Ruppel et al., 2017)
Akademii Nauk ice core (Siberia)	129 m	No field blanks; only reagent lab blanks	Aromatic acids (vanillic and para-hydroxybenzoic acids)	(Grieman et al., 2017)
Greenland	87 m and 213 m	No field blanks; only reagent lab blanks	Methanesulfonate, Br, Cl, Na, Ca, S, Ce, and Pb	(Maselli et al., 2017)
European Alps	10 m	No field blanks;	Polybrominated	(Kirchgeorg et

		only reagent lab blanks for sample extraction	diphenyl ethers, perfluorinated acids, polyaromatic hydrocarbons	al., 2016)
East Antarctica	1196 m core	No field blanks; only reagent lab blanks	Organochlorine pesticides	(Bigot et al., 2016)
Eastern Switzerland	101 m using electromechanical-thermal drill	No field blanks; only instrument blanks	Polychlorinated biphenyls	(Pavlova et al., 2015)
Tibetan Plateau	22 m	No field blanks; only reagent blanks for sample extraction	Perfluorinated acids	(Wang et al., 2014)

We have amassed an annual data set on field blanks (HPLC grade water) transported and exposed to the atmosphere in the Arctic location Resolute Bay, Nunavut since 2010. These samples are shipped back to the lab, extracted and analyzed with methods analogous to the ice samples and compared to the same HPLC grade water stored in the lab. These results indicate that the environmental exposure and shipping do not contribute to PFAS background contamination. We have included tables of this data in the SI (Table S4a-S4b) and added to the main text:
"Previous results from field blanks (HPLC grade water) transported and exposed to the atmosphere in the Canadian Arctic (Resolute Bay, Nunavut) have indicated that the environmental exposure and shipping do not contribute to background PFAS contamination. We have amassed an annual data set on these field blanks (Table S4a-S4b)." (P4, L4-L6, Tables S4a-S4b).

We also have some key observations that suggest PFAS background contamination is not compromising our results in this study. We have added text to the manuscript to highlight the measures taken to avoid PFAS contamination such as outfitting the sampling team in PFAS-free apparel, not using polytetrafluoroethene or any other fluorinated polymers during sampling, shipping and handling, rinsing and cleaning all sampling and sample handling equipment with methanol. We've also specified that the Kovacs ice coring drill bit is stainless steel for those unfamiliar with ice coring. For PFAS analysis, the most commonly reported blank contaminant is PFOA arising from polytetrafluoroethene (PFTE) polymers. Our analysis of PFOA in the samples have concentration that are much lower than other ice core analyses suggesting that background contamination was minimal. Any systematic bias encountered through background contamination would not impact the temporal trends.

The text we added to the manuscript now reads:
"Extensive measures were taken to avoid PFAS contamination during both sample collection and sectioning (i.e. PFAS-free apparel and equipment, methanol-rinsed sampling tools and equipment)." (P3, L4-L6)

2. Page 5, line 13. As I understand the back trajectories provided by Hysplit, there is an increasing uncertainty for longer time periods. While the 48 hours back trajectories are reliable, there is a huge uncertainty for 10 day back trajectories. I suggest to shorten the back trajectories and discuss the uncertainties.

Response: We thank the Reviewer for this important comment regarding back trajectory analysis uncertainty. It is standard to run 10-day back trajectories when subsequently running a residence time analysis, which greatly reduces the uncertainty in any one pathway (see (Criscitiello et al., 2016) and references therein). If the back trajectory run-time is shortened prior to residence time analysis, this often disregards the atmospheric residence time of the species of interest, resulting in erroneous (incomplete) scaled air mass transport densities that cluster around the trajectory endpoint (ice core site, in this case). In numerous earth science applications, 10-day back trajectories are the standard. For example; (Harris and Kahl, 1994; Huang et al., 2009; Kulshrestha and Kumar, 2014).

In addition to current peer-review literature, we base our methods on discussions contained in the following two books: (1) "Lagrangian Modeling of the Atmosphere" (Lin et al., 2013), which describes in detail why a back trajectory length of 10 days is ideal for such applications, and (2) "Intercontinental Transport of Air Pollution" (Stohl, 2004), which discusses specifically why 10-day back trajectories are used when investigating long-range transport of pollutants to the Arctic.

3. Page 6, line 5. Report and discuss the method for dating the subsamples from the ice core and determining the annual snow accumulation.

Response: We thank the Reviewer for this suggestion. We have indicated in this paragraph where more detail can be found on the ice core dating and have provided a sentence explaining how annual snow accumulation was determined. These sentences read:

"**The dating of the ice core itself is discussed in more detail in the SI (section S1). Annual snow accumulation was determined by measuring the length of the annual ice core sections.** Annual snow accumulation was **calculated as** 0.15 – 0.64 m yr^{-1}…" (P6, L7-L9)

4. Page 6, line 15. "The most: : :." This sentence needs a reference or justification.

Response: We thank the Reviewer for pointing this out. The most straightforward identification of seasonal maxima is in ice cores from the summit of the ice cap. We have added in the reference, (Legrand and Mayewski, 1997). Legrand and Mayewski say that accurate ice dating is based on visual stratigraphy, oxygen isotopic ratios of the ice, and electrical conductivity measurements in areas of high accumulation. The sentence now reads:

"The most **straightforward** temporal record of atmospheric deposition is obtained at the summit of an ice cap **(Legrand and Mayewski, 1997)**;" (P6, L22-23)

5. Page 6, line 21: : :. Compare as well with concentrations and fluxes from maritime Antarctica (Casal et al. EST 2017).

Response: As per the Reviewer's suggestion, we have added PFAA concentrations from Casal et al. (2017) to Table S13 in the SI (P22) and have referenced Casal et al. (2017) in this paragraph (P6, L27-L28).

6. Section 3.1 I suggest to include one or two figures with the patterns, concentrations or fluxes, and extend the discussion. There are some differences in the patterns reported for snow (fresh and/or accumulated) and it is not clear how the results shown here fit with

previous results. For me, the major contribution of this manuscript is the data set of the measurements, much more important than the modelling, however, the paper does not reflect this.

Response: We thank the Reviewer for this comment. Section 3.1 is meant to just be a short discussion on the PFAA concentrations detected on Devon Ice Cap for this study, whilst comparing these measurements to previous studies, particularly the previous two Devon Ice Cap studies. We believe it is important to include a section discussing what has previously been done/measured and how our current results compare. To respond to this Reviewer's comment, the rest of the sections in the paper (sections 3.3 to 3.7) do indeed discuss our measurement data set, where we include multiple figures illustrating fluxes and patterns, among other topics.

7. Page 7, line 10. Provide information for the other years.

Response: As described above and in Section 3.2, we calculated back trajectories for only 2 years to determine whether the unusual PFAA observations of 2013 were caused by a transport anomaly. Preliminary back trajectory analyses that we conducted showed minimal variation from year to year. The year 1994, modeled in Figure 1, was our typical year for comparison, which we used as our baseline, compared to the Meyer et al., 2012 paper. The back trajectory analyses between both their study and our study showed very similar results. We have added in text to section 3.2 to explicitly say this. It now reads:

"The year 1994 was selected to compare to previous trajectory analyses **conducted by Meyer et al, and showed similar results**." (P7, L10-L11)

8. Figure 1 and page 7. I don't agree with the discussion and conclusions for the results shown in figure 1. There is a very weak evidence for source regions in part of Asia, first, due to the uncertainty of the analysis, and secondly, because the signaled regions have a very low population. Figure 1 shows that the Arctic ocean (ice covered or not) may be the main source regions. I wonder if specific ice-influenced photochemistry may affect the formation of the targeted compounds.

Response: It is not clear what type of ice-mediated photochemistry the Reviewer is referring to. Perhaps the Reviewer is referring to enhanced PFAA formation from precursors? There is currently no evidence in literature for ice-mediated PFAS formation to warrant speculative discussions. However, the most thoroughly studied mechanisms for producing PFAA from atmospheric precursors, are presented in the discussion with supporting data to postulate sources of PFAAs in this remote ice cap including the measured temporal deposition profiles, known production inventories, air mass transport density analysis, PFAA molar flux ratios of homologue pairs, major ion tracers, and previous modeling studies on air mass transport (see response to comment 2). We have added a sentence to section 3.7 to warrant a focus on this subject for future work:

"**The role of ice in the formation of PFAAs from precursors is currently unknown and future work should focus on ice-mediated PFAA formation**." (P18, L3-L4)

9. I would suggest to try to correlate the concentrations and fluxes with the extension of the arctic ice cap.

We thank the Reviewer for this innovative suggestion. We have correlated the annual PFAS deposition fluxes with minimum Arctic sea ice extent and sea ice area. The results of the correlations are now mentioned in the text and reported in the SI:

“Additionally, correlations between annual PFAS deposition and Arctic sea ice minimum were calculated (NSIDC, 2017) (Table S22). If marine aerosols were a major source of PFAS to the ice cap, then a negative correlation between annual sea ice extent and deposition would be expected, but the majority (12 of 14) showed a positive correlation.” (P16, L22-L25)

Table S22. Coefficients of determination (R^2) and statistical significance (p) between PFAS deposition and Arctic sea ice extent and area. Sea ice time series are based on SMMR/SMM/I satellite observations. The slope sign is indicated as either positive (+) or negative (-) for each PFAA compound. Correlations are ranked in terms of significance by purple>blue>orange.

	Sea Ice Extent	Sea Ice Area	Slope Sign (+/-)
PFPeA	R^2= 0.047 p=0.2088	R^2= 0.050 p=0.1959	+
PFHxA	R^2= 0.217 p=0.0048	R^2= 0.215 p=0.0050	+
PFHpA	R^2= 0.295 p=0.0007	R^2= 0.295 p=0.0007	+
PFOA	R^2= 0.106 p=0.0559	R^2= 0.114 p=0.0470	+
PFNA	R^2= 0.183 p=0.0104	R^2= 0.199 p=0.0073	+
PFDA	R^2= 0.129 p=0.0339	R^2= 0.142 p=0.0258	+
PFUnDA	R^2= 0.161 p=0.0168	R^2= 0.169 p=0.0140	+
PFDoDA	R^2= 0.058 p=0.1648	R^2= 0.067 p=0.1334	+
PFTrDA	R^2= 0.030 p=0.3172	R^2= 0.033 p=0.2938	+
PFOcDA	R^2= 0.007 p=0.6437	R^2= 0.017 p=0.4618	-
PFBS	R^2= 0.113 p=0.0485	R^2= 0.116 p=0.0451	+
PFHpS	R^2= 0.055 p=0.1735	R^2= 0.049 p=0.2000	+
PFOS	R^2= 0.099 p=0.0659	R^2= 0.085 p=0.0887	+
FOSA	R^2= 0.266 p=0.0015	R^2= 0.261 p=0.0017	-

10. Page 13, lines 4-5. I don’t understand this sentence.

Response: We thank the Reviewer for mentioning this. We have clarified this sentence, which now reads:
"Indirect sources of PFAAs are contributors to the global presence of these compounds, **particularly in locations such as the Canadian Archipelago** (Benskin et al., 2012a)." (P14, L4-L5)

11. An implicit assumption in this work is that the observed pattern and concentrations are a direct consequence of atmospheric snow deposition, thus snow scavenging of atmospheric PFAS. No discussion is made of post-depositional processes affecting the concentrations and patterns of PFAS.

Response: In regards to the post-depositional processes, we do discuss this in some detail in section 3.3 (P8, L4-L24). Melting events can occur in the ice core, which can cause PFAAs to elute into the snowpack and refreeze at ice layer interfaces. MacInnis et al. (2017) discuss these post-depositional processes in more detail. To make this discussion more implicit, we have added more detail on these post-depositional processes to this paragraph, which now reads:
"One study has examined elution behaviour of PFAAs from a melting snowpack and found that elution of PFAAs is driven by **a number of factors including water solubility in the snowpack, PFAA chain length, and sorptive capacity of the snow grain surface** (Plassmann et al., 2011). **Due to melting at the surface, concentrations of PFAAs measured near the surface layer can result in inaccurate estimations whereby one year might be overestimated and another year underestimated. Melting events on the ice cap have** likely happened periodically over the last 15 – 20 years, thereby blurring to some extent, the vertical profile. However, we expect any melting that occurred to have primarily affected the seasonal trends**, as Koerner, 2005 states that the percolation of meltwater in a snowpack will refreeze within an annual layer, and so seasonal cycles of PFAA deposition will be biased, but annual interpretations should not be affected (Koerner, 2005)**." (P8, L12-L20)

12. Even If I was convinced that most PFAS in the studied arctic region come from atmospheric oxidation of neutral precursors, no discussion is made on the processes responsible for the transport of ionizable PFAS from the gas phase (I guess oxidation occurs in the gas phase) to deposited snow. Once a ionizable PFAS is formed, it may attach rapidly to aerosols. Which are the dominant aerosol types in this region? A reference is needed to support the response to this question. Furthermore, it could be that neutral precursors are the main contributors to ionizable PFAS in the surface Arctic ocean, and then these are transferred to Devon Cap by sea-salt aerosol formation and deposition.

Response: It is not clear what the Reviewer is referring to by "aerosol type" – size, inorganic composition, organic composition, nucleating ability, optical depth, water content? We feel the scope of identifying aerosol types in the Devon Ice Cap atmosphere is beyond the scope of this study. However, what is directly relevant to this paper is the differences in physical properties of the precursors (neutral and volatile) and the acids (ionic and sorptive). As we mention in the introduction, after PFAAs are formed in the gas phase, they can be deposited to the surface through both wet and dry deposition due to their physicochemical properties. We have clarified our introductory sentence and added references to a few modelling studies:
"Once these PFAAs are formed in the **gas phase**, they undergo wet or dry deposition **to the surface. The specifics of these processes have been considered in several modelling studies**

in the literature (Armitage et al., 2009a, 2009b; Schenker et al., 2008; Wallington et al., 2006; Yarwood et al., 2007)." (P2, L18-L21)

13. Page 15, line 5. I don't know any study on the occurrence of PFAS in the marine surface microlayer, but for other POPs, there is a huge variability on the enrichment factors, thus a lack of correlation does not contradict the potential role of marine aerosols. Furthermore, Na can be forced to move out from the snow/ice after deposition.

Response: The mechanisms for sea surface microlayer enrichment of PFAS are not analogous to other POPs due to the unique properties imparted by the combination of the hydrophobic and lipophobic perfluoroalkyl chain and the hydrophilic carboxylate or sulfonate moiety. A preliminary study of enrichment of PFAS in the freshwater surface microlayer has been described (Reth et al., 2011), with enrichment increasing with chain length. As discussed in a previous paper from our team (and briefly mentioned in this work), surfactant strength does not describe the differences between PFAS levels in the ice cap and ocean. To clarify this, we have added a reference to that paper at the beginning of the discussion of ice cap/ocean comparisons to make clear we are using a previously described data analysis technique:
"Another technique used to assess the influence of marine aerosol deposition of PFAAs to the Devon Ice Cap is a comparison between ocean and ice cap homologue patterns **(MacInnis et al., 2017)**." (P16, L7-L8)
We have further clarified by re-phrasing the description of the surfactant relationship:
"**Water-to-air transport of PFAS is related to surfactant strength (Reth et al., 2011). Consistent with results from MacInnis et al. (2017), differences in surfactant strength cannot account for the different homologue profiles observed on the ice cap and in the ocean.**" (P16, L10-L12)

We agree with the Reviewer that post-depositional processes may be different for PFAS and sodium, although this has not been explicitly studied. On its own, these correlations would not provide sufficient evidence to discount the role of marine aerosols. However, the combination of the lack of correlation with sodium and sea ice extent, and the differing homologue patterns support our conclusion that marine aerosols are not the dominant source of PFAS to Devon Ice Cap.

14. Page 15, line 9. This is not true because the patterns in snow are different than in seawater even for a given site, and because we do not know the patterns in the surface microlayer, nor in remote aerosols.

Response: As described above in the response to comment 13, we do have information on the patterns of enrichment for the surface microlayer and marine aerosols relative to ocean water (Reth et al., 2011). To clarify this point, we have moved our discussion of water-to-air transport to directly follow this sentence. The section now reads as follows:
"If marine aerosols were/are a major source of PFAA contamination on Devon Ice Cap, then one would expect the homologue profiles to be similar between the ocean and the ice cap. **Water-to-air transport of PFAS is related to surfactant strength (Reth et al., 2011). Consistent with results from MacInnis et al. (2017), differences in surfactant strength cannot account for the different homologue profiles observed on the ice cap and in the ocean (Fig S8)**." (P16, L8-L12)

15. I suggest to plot the ratio of concentrations between Devon cap and arctic ocean (except for PFBA which has clearly a different origin and behavior), and plot this ratio versus the number of C of the pfas chain, and then discuss taking into account the literature.

Response: We thank the Reviewer for this suggestion. We actually initially plotted the ratios of concentrations in the Devon Ice Cap compared to ocean levels, similar to what was done in MacInnis et al. (2017), but included a proportional analysis of the molar concentrations in this paper instead, as to avoid duplicating a previous paper's figure. We have added Figure S8 to the SI (P31-P32)) and included a reference to it in the main text:
"Consistent with results from MacInnis et al. (2017), differences in surfactant strength cannot account for the different homologue profiles observed on the ice cap and in the ocean **(Fig. S8).**" (P16, L10-L12)

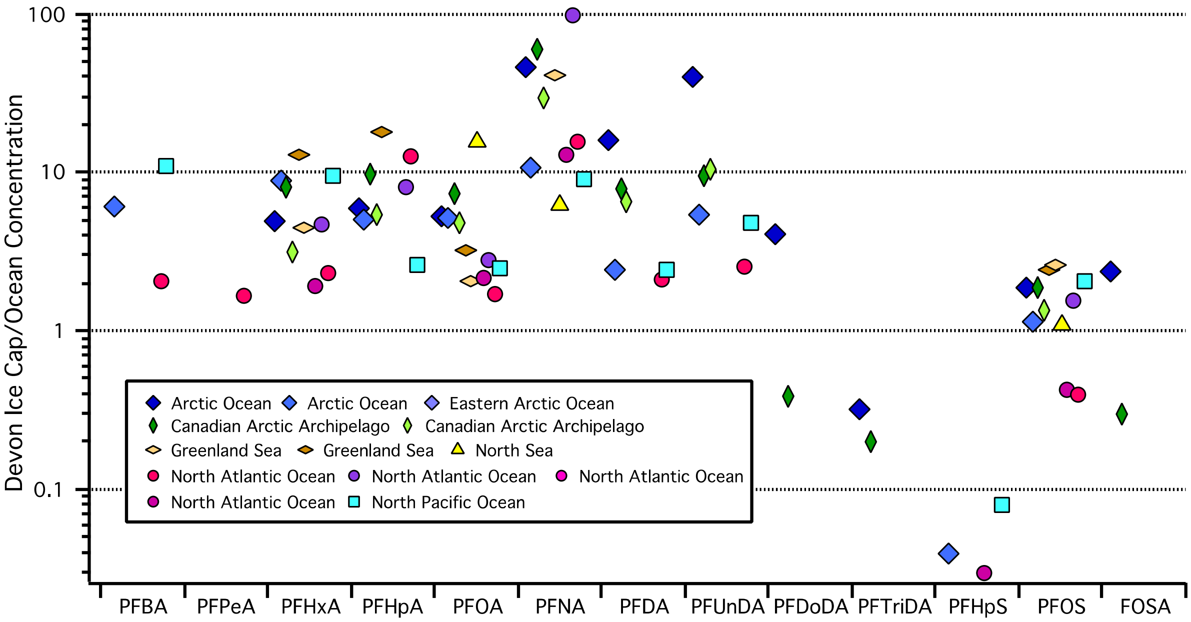

Figure S8. Ratios of observed concentrations in the Devon Ice Cap compared to levels in the Arctic Ocean (Benskin et al., 2012b)(Cai et al., 2012), Canadian Artic Archipelago (Benskin et al., 2012b), Greenland Sea (Zhao et al., 2012)(Busch et al., 2010), North Sea (Ahrens et al., 2010b), North Atlantic Ocean (Ahrens et al., 2010a)(Ahrens et al., 2009)(Zhao et al., 2012), and North Pacific Ocean (Cai et al., 2012) in 2005 and 2007 to 2010.

16. The authors insist that sea salt aerosol does not play a role, but I don't see concluding arguments. Which is the main source of aerosols in the region and how ionizable pfas behave after their formation? Ok, let's assume that sea-salt aerosols do not play an important role, then, to which aerosols are PFAS bound to? Are they in the gas phase and then scavenged by snow? Please, provide a plausible mechanistic explanation.

Response: As we have described above, PFAS formed in the gas phase will be deposited through wet and dry deposition (see response to comment 12). The identity of the aerosols with which PFAS might be associated is an open question. Our correlations with different tracers (e.g. for dust) were targeted to reduce uncertainty in this area.

17. Pages 16-17. A hypothetical dust source is commented, but this is not supported by the assessment of air mass back trajectories.

Response: Our discussion regarding dust is not based on our back trajectories, but rather on the well-established literature indicating both the presence of dust in the Arctic and that it can be coming from LRT (Darby et al., 1974; Rahn et al., 1977; Zdanowicz et al., 1998). To clarify this in the text, we have rephrased and added an additional reference:

"Mineral dust aerosols can accumulate acidic atmospheric contaminants, such as nitric and hydrochloric acids (Sullivan et al., 2007). **Dust can undergo LRT and deposit in the remote Arctic** (Rahn et al., 1977; **Zdanowicz et al., 1998**)." (P18, L6-L8)

18. Note that Ca and Mg are enriched in sea-salt aerosol coming from the sea surface microlayer (Jayarathne et al. EST 2016)! Then the correlations between Ca and PFAS would support sea salt as an important contributor to PFAS at the studied site!

Response: We thank the Reviewer for mentioning this. Although we did consider the enrichment of calcium and magnesium, we believe it has a negligible impact on our interpretation of the data. Enrichment in marine aerosols by calcium has been shown by a few studies (Jayarathne et al., 2016; Oppo et al., 1999; Salter et al., 2016). Aerosol enrichment by magnesium has been observed in a single study (Jayarathne et al., 2016), while another showed no enrichment (Oppo et al., 1999). Using established seawater cation ratios, we calculated average non-sea-salt (nss) fractions of calcium and magnesium of 99.5 % and 70.3 %, respectively (Table S19). Jayarathne et al. (2016) observed calcium enrichment factors were variable, but up to 3.38 in marine aerosols. If we assumed this maximum enrichment, nss-Ca on the ice cap would remain >98 %. It is extremely unlikely that this small change would impact the interpretation of our correlations. Given the uncertainty and observed variability in enrichment for magnesium, it is not possible at this time to determine the impact of possible magnesium marine aerosol enrichment on ice cap nss-Mg.

19. After reading this manuscript I think that the interpretation needs to be re-evaluated, and a new version prepared taking into account my comments above.

Response: We thank the Reviewer for taking the time to read our manuscript and provide important feedback. We have addressed all of the reviewer's comments and prepared a revised manuscript.

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