

Interactive comment on “Continuous Non-Marine Inputs of Per- and Polyfluoroalkyl Substances to the High Arctic: A Multi-Decadal Temporal Record” by Heidi M. Pickard et al.

Anonymous Referee #2

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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.

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- 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.
- 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.
- Page 6, line 5. Report and discuss the method for dating the subsamples from the ice core and determining the annual snow accumulation.
- Page 6, line 15. "The most. . . ." This sentence needs a reference or justification.
- Page 6, line 21. . . . Compare as well with concentrations and fluxes from maritime Antarctica (Casal et al. EST 2017).
- 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.
- Page 7, line 10. Provide information for the other years.
- 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.

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

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

- 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 wet-depositional processes affecting the concentrations and patterns of PFAS. - 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.

- 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.

- 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.

- 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.

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- 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.

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

- 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!

- 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.

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