Comments by Reviewer #2

We want to thank Reviewer #2 for their thoughtful and constructive comments that have improved the manuscript. Our responses below. We wrote our responses in the form of proposed changes to the text that we would make in a potential revised manuscript.

This manuscript presents a compilation of isotopic composition of krypton which has been obtained over the past years in different ice cores from Greenland and Antarctica.

Using the present-day understanding of the drivers of gas repartition in the firn as well as correlation with ERA products, the authors propose that the 86Kr_excess can be used for synoptic activity.

The manuscript is generally well written and well documented. The authors also explained in details the numerous limitations associated with this interpretation both in section 3.3 as well as in the supplements.

Because of the limitations in the interpretation of the 86Kr_excess, the authors should be more cautious in the proposed interpretation. While the scientists in the filed of ice cores will get the limitations and use the results with caution, it may be different for people who do not understand the complexity of processes affecting air elementar and isotopic repartition in the firn. I thus suggest to modify the abstract and the conclusion to insist on the speculative interpretation of the 86Kr_excess and on the additionnal measurements to be done to better quantify the effect of gas loss, thermal diffusion (including rectifier effect) and possible existence of a convective zone.

We of course fully agree with the reviewer on this point, and we tried to be very careful in our wording throughout. Reviewer #1 also commented we were careful in our interpretation. We see the risk that the reviewer highlights, as people tend to plot records that support their argument without always including all the caveats from the source publication. However, one can never be too cautious, and so in response to this request, we have made the following changes:

In the abstract we added:

"Limited scientific understanding of the firn physics and potential biases of $^{86}Kr_{xs}$ require caution in interpreting this proxy at present."

We further replaced the word "*tentative*" with "*speculative*".

In the conclusion, we added:

"Due to these limitations, we caution that any interpretation of temporal $^{86}Kr_{xs}$ changes remains speculative at present."

146 : it is strange to refer to Fig 6 here. Moreover, it is strange to prefer one or the other since it is shown later that gas loss and thermal effect are the most important corrections to take into account (d40Ar being sensitive to gas loss and d15N being the most sensitive to thermal fractionation). There is no obvious reason to prefer one notation compared to another.

We have removed the reference to Fig. 6 in this place in the paper.

For most cores drilled in the past 2 decades where ice samples have been stored under cold conditions, we expect that gas loss correction to be the smaller correction with the smaller uncertainty. Furthermore, the observations make it clear that the $^{86}Kr_{xs40}$ definition has less scatter than the $^{86}Kr_{xs15}$ definition – compare Figs. A3 and A4. For these reasons we prefer the $^{86}Kr_{xs40}$ definition. We have provided plots for each of the definitions, both for the WDC record and for the spatial calibration study. Therefore, readers can compare both definitions and derive their own conclusions for the comparison.

From l. 223 : the preparartion of the samples is different for DE08-OH than for the other samples. May this explain the different slopes associated with gas loss in figure A1-B.

Yes, possibly. This is a good suggestion that we had not considered. For the DE08-OH samples, the dO2/N2 and dAr/N2 were measured on smaller samples (larger surface-to-volume-ratio), which may be more affected by gas loss during pumping on the samples. We added this to the supplement A1:

"The DE08-OH samples were also analyzed differently from those at other sites, with $\delta O_2/N_2$ and $\delta Ar/N_2$ measurements performed on a separate smaller ice piece (see section 2.2); the greater surface-to-volume ratio of such small samples may result in greater gas fractionation while evacuating the sample flasks in the laboratory."

241 : How are the samples flagged for drill liquid contamination ? How is it possible to detect the drill fluid contamination ? From which measurements ?

Here we rely on Baggenstos et al. (2019), where the EDC data originate. The original study flagged the drill liquid contaminations, which can be observed in the IRMS analysis. Baggenstos et al. report in the supplementary information that drilling fluid contamination causes isobaric interference on mass 29 and 82, thereby impacting measurements of both $\delta^{15}N$ and $\delta^{86}Kr$. We have now added this to the manuscript.

245 – 247 : Can you explain the error propagation explaining why the 2 sigma is larger for 86Kr than for 86Kr_excess ?

This is a good point also brought up by reviewer #1. We copied the response to reviewer #1 here:

Good question. This depends on the value of the denominator, and for the values given in the paper we assume a $\delta^{40}Ar$ of around 1.2 permil that is typical for WAIS Divide.

Starting from the definition of Eq (2), the uncertainty in the numerator is effectively equal to the uncertainty of the $\delta^{86}Kr$ measurement. Because the value of the denominator is typically greater than 1, the uncertainty of the $^{86}Kr_{xs}$ appears smaller than that of $\delta^{86}Kr$ (in the WDC example, 26/1.2 = 22). Of course in a relative sense the $^{86}Kr_{xs}$ error is much greater than the $\delta^{86}Kr$ error.

We now clarify this in the text: *"Via standard error propagation, this results in a ~ 22 per meg ‰$^{-1}$ (2σ) analytical uncertainty for both $^{86}Kr_{xs40}$ and $^{86}Kr_{xs15}$ at a site like WDC where $\delta^{40}Ar$ ≈ 1.2 ‰."*

253 : I do not see why it is useful to present these data to remove them immediatly after. In this case, the 86_Kr data from EDC samples affected by drill fluid should also be displayed with an explanation on how they were discarded.

Yes, that is an interesting point. The data from these two sites (JRI and BRP) are not really displayed however, and only shown only in Figure A1A. The full isotope measurements are archived online for others to use, however. The samples from these two sites were provided and shipped by our international collaborators for the sole purpose of this calibration study, as we thought the Antarctic Peninsula was an important site to include. Therefore, we feel an obligation to report these failed attempts. It further provides the important lessons that melt should be avoided for Kr-86 excess (which is no surprise given the high solubility of Kr), and that refrozen meltwater can be present in the absence of visible melt features (which has not been reported before to our knowledge).

The samples from EDC were not measured for the purpose of this calibration study, however, and were previously published by Baggenstos et al. (2019). The drill fluid flagging was not done by our team, we simply rely on the original study that had flagged these samples. Therefore the choice to discard them was not ours, and justification for this choice is given in the original study (Baggenstos et al., 2019).

Section 3.1 : The Phi parameter exhibits strong seasonal and interannual variabilities and I do not understand how this variability is taken into account in the « calibration » of the 86Kr_excess. Such sensitivity should be studied or implemented in Figure 3 since this is crucial for the interpretation of the 86Kr_excess proposed here.

The seasonal and interannual variability is discussed to give the reader a better understanding of the nature of synoptic variability in Antarctica. However, neither impacts the calibration study, where we compare Kr-86 excess to the multi-decadal average Φ (1979-2017 CE). We now specify this more clearly:

"In Fig. 3A we plot the site mean 86Krxs40 (with ±1σ error bars) as a function of Φ (averaged over full 1979-2017 period)"

The samples each represent multiple years – both because of the wide age distribution of firn air, and because the large samples needed typically span multiple annual layers.

Section 3.3 : this section is interesting in providing the limitations of the interpretation of 86Kr_excess and strongly suggest that further study should be performed for a robust interpretation such as firn air pumping study at different site with a correct determination of the thermal gradient (it is really surprising to find such temperature gradient at DE08 and EDC) + analyses of ice not affected by gas loss, etc… this is the reason why the authors should be much more cautious in their conclusions and better suggest concrete perspectives on how to progress with such proxy if it is reallt promising. Actually, the concluding paragraph of section 3.3 should also be summarized in both the abstract and conclusion of the manuscript to clearly state the limit of this interpretation which is now speculative.

We agree. We are the first to admit that this proxy has challenges at present, which is why we devote an entire section of our paper to it. In response to the first reviewer #2 comment, we already added additional notes of caution to the abstract and conclusions, and specifically note that the interpretation remains speculative (using the reviewer's choice of words).

We further added to the abstract: *"A list of suggested future studies is provided."*

And to the conclusions: "*A full list of suggested follow-up studies is given in section 3.3"*

In section 4, I feel that a discussion on the seasonal variability and its possible impact is missing.

The gas age distribution at the base of the firn has a width of several years, and therefore Kr-86 excess reflects the time-averaged barometric variability over several years. For this reason, we only investigate the annual-mean pressure variance. Including an additional analysis of seasonal variability would not change any of our conclusions, and add to an already overly long article.

Section 5 : I understand that the authors do their best with the poor data quality but it would be nice to comment on the strong scattering for the data at « present-day » ? Can this scattering be used to estimate the uncertainty as the authors mention that « no true replicate to assess the reproducibility » …

This is an interesting point, and it is true that there is substantial scatter in the records. We are unclear what this scatter represents, at present. As the reviewer suggests, there may indeed be a contribution from analytical uncertainty which is not captured in our estimated precision. However, climate patterns such as the Pacific Decadal Oscillation definitely impact storminess at WDC in reanalysis data, which suggests some of this scatter probably reflects variations in WDC storminess at decadal or inter-annual time scales. Last, the DE08-OH data suggest there may be cm-scale variations in Kr-86 excess that we attribute to layering in firn microstructural properties. In our sampling we may not be fully averaging out this cm-scale variability, contributing to more scatter. We added this to the text in Section 5:

"The scatter in the late Holocene WDC $^{86}Kr_{xs}$ data exceeds the stated analytical precision. Potential explanations include (1) an underestimation of the true analytical precision; (2) interannual to decadal variations in storminess at WDC; and (3) aliasing of cm-scale variations in ice core $^{86}Kr_{xs}$ linked to layering in firn microstructural properties."

20 and 21 : the discussion is quite long for such speculative interpretation. I would suggest shorten it to stay on the safe side of the interpretation.

In response to this comment we shortened the interpretation section by removing the discussion about the split jet.

725 : I am not sure that the authors really « calibrate » the proxy – let's say that this is a first proposition of interpretation. A calibration would require more dedicated studies as mentionned in the concluding paragraphe of section 3.3.

We removed the word "calibrate" here.

Figure 3 : What is the origin of the uncertainty bars for the different sites ? Do the sites with more data have more scattering hence a larger uncertainty bar ? It would be useful to mention the number of points used for each sites in this calibration and how the error bar is calculated. A table may be useful to exactly describe the number of samples for each site, depth range, conditions of storage, etc…

The number of samples for each site in the calibration study is listed in Table 1. The number of samples and nature of the error bars is explained in the updated figure caption:

"the error bars denote the ±1σ standard deviation between samples (uncertainty in corrections and measurements not included). The number of samples at each site is listed in Table 1."

A more complete description of the sample characteristics (precise depths etc.) is provided by the data tables archived online.

Figure A2 : The displayed results show very depleted samples in dO2/N2 and dAr/N2 – are these results really relevant for this paper ? What is the origin of these samples ? core top ? Bottom ice ?

The relevance to this paper is that we use the Byrd data to estimate the impact of gas loss on δ^{40}Ar. This is described in Appendix A1. We now specify this more clearly in the figure caption:

"Argon isotopic enrichment due to gas loss in the Byrd core used to determine the δ^{40}Ar gas loss correction (appendix A1)."

Figure B1 shows that there may be a large scattering with depth of 86Kr-excess. I am sure that this is taken into account in this study but it would be nice to explain a little bit more how it is done (also for the other cores). Probably again a table explaining the number of samples considered for present-day for each core, the depth range and individual values would help.

The number of samples is listed in Table 1. We added the following statement to account for the other sites:

"At all other sites analyzed here, the sample length exceeds the annual layer thickness; this will remove some, but not all, of the effects of the sub-annual variations."

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