

Dear Editor,

Below, we address Reviewer #1 comments. First we really appreciate and thank the reviewer for diving into the data and the beneficial comments. While most of the major comments are further discussion of earlier ones, comment #1 is new and is a very important one. As detailed below, in accordance with this comment, we now revised some of the discussion and conclusions. If needed, we will be happy to further discuss this and other comments with the reviewer, as to get the best out of this manuscript.

The line numbers refer to the track changes file

Referee #1

I appreciate the authors' responses to my previous comments, and their additional explanations. Following this, I have a better understanding of their proposed hypothesis, and I think the revised text is improved. However, I still have some questions regarding their support for the hypothesis of Ra-226 diffusion from inside sediment grains, and their argument against desorption controlling the isotopic signature of ADE-2. My concerns are outlined below.

(1) Ra-228 is not discussed much in the text, but the activities between cores are similar, as is the case for Ra-226. Also, 228/223 and 228/224 ratios in ADE-17 are higher than those in ADE-2, similar to the 226/223 and 226/224 ratios. Therefore, any explanation for the 226 activities should also explain the 228 activities; is this the case with the diffusion hypothesis? Ra-228 has a much shorter half-life than Ra-226, so would Ra-228 be able to diffuse out of frozen nanopores on a ~30 year timescale before decaying? And if so, how would that change the conclusions about the relative ages of the cores?

- This is a very important comment. We have apparently forgotten this observation, and we are very thankful to the reviewer for noticing and mentioning. Although 228 data is less comprehensive, 228/223 pattern is indeed very similar to that of 226/223. Actually, this helps us with better constraining the timing of the salinization event. Namely, with 228Ra portraying the same pattern as the 226, the suggested diffusion should be on time scales of 228Ra, which is years to a few decades (if longer diffusion times, 228 will mostly decay and will not make it to the porospace). This means that diffusion must be via nano-pores, as suggested by Rama & Moore. We, accordingly, suggest that diffusion occurs through partly liquidized ice along grain boundaries, as suggested in the literature (please see revised text). This allows diffusion time scales of years (updated Figure 13), when clays and silt are considered.

We re-stress that we found no alternative explanation for the observation that in one core Ra ratios are similar to bulk sediment parent ratios, while in the nearby

core Ra ratios are in equilibrium with the (very different) parent ratios in the exchangeable fraction (please see response to other comments below).

We stress that the very young age for the fluid migration and salinization agrees well with other findings, such as the <1 year formation of ice in the adventdalem (Weinstein et al. 2019), and a recent finding (another site, 3 km away) of a brine with seawater composition that is actively salinizing the ground ice along the permafrost profile (not yet published).

Accordingly, we now made major revisions to sections 5.2.1 and 5.2.2, as well as other parts in the text, as to accommodate with the 228/223 observation (**see mainly Lines 468-665**).

(2) It is shown that Th-228 and Ac-227 (parent isotopes of Ra-224 and -223) are more concentrated on grain surfaces compared to bulk measurements. Therefore, Ra-224 and Ra-223 may be more concentrated on grain surfaces compared to the long-lived Ra-226 and Ra-228, which could explain why there is more of the short-lived isotopes released when salinity increases for ADE-2. In my previous comment about this, the authors state that the problem with this explanation is that 226 also does not increase. But my argument is that this is indeed expected because more 226 is locked inside the sediment grains (where it cannot desorb at higher salinities), unlike the short-lived isotopes, which are concentrated on the sediment surfaces and can easily desorb when the sediments come into contact with higher ionic strength waters. I understand the authors' argument for a longer timescale allowing time for Ra-226 to diffuse out of the sediment grains, but separate from that possible hypothesis, I am suggesting that this is an alternative explanation and I do not see the authors' rebuttal of this idea in the previous round of review. The explanation should also be consistent with the Ra-228 profiles.

- We certainly agree. This is not an alternative explanation, but rather agrees with what we wrote – that there is relatively high 227 and 228 (i.e. low ratios of 230/227 and 230/228) on the grain surfaces, and this is why the 226/223 and 226/224 ratios are low in ADE-2.

The problem is actually with ADE-17: why are the ratios there (incl. 228/223) higher than parent ratios on grain surfaces? The only explanation we have is that in this core there has been enough time for the long-lived Ra (now, including 228) to reach the pore space from inside the grains, where 230 and 232 are much higher, which should be by diffusion

The only alternative explanation could be that parent ratios are very different along the whole profile of the two cores (~12 m). However, this could hardly be the case, considering that the cores are from the same site (30m apart), core logs are texturally very similar and that differences persist all along the profile.

(3) If I understand correctly, the authors are stating that adsorption does not occur because the water in and around sediment grains is likely completely frozen, but that diffusion can still occur in solid ice. However, the back-of-the-envelope calculation comparing Ra-226 to Th-230 shows that Ra-226 activities are lower than expected, likely because of adsorption (line 551). Doesn't this provide evidence that liquid water is likely to be present and/or that adsorption is happening to some degree? Is the argument that there may be liquid films *around* the sediment grains, but that the nanopores *inside* the grains would stay completely frozen despite these outer liquid films? If so, this should be explicitly stated (and I apologize if I missed it somewhere in the text already).

- Thanks for this question, which allows us to re-describe not only our view but also the permafrost condition, as observed in the field.

First, we clearly wrote (**e.g., lines 48, 134-136, 161-163**), and stressed in the first Response to Reviewers, that the pore space (where the ground-ice is) often contains liquid water. This is observed during drilling, documented geophysically (e.g. Keating et al. 2018) and concluded from isotopic studies (Weinstein et al. 2019). This is why we included a discussion of the effect of K-adsorption on radium buildup in the porespace. Also, as mentioned by the reviewer, we showed that radium in ground-ice is lower than expected by ^{230}Th on grain surfaces, probably due to (partial) adsorption. As the reviewer noted, the nano-pores are a separate issue. Being an in-grain nano texture, with much lower W:R ratio, we assumed in previous version that the chances of having liquid water are much lower. But, since the nano-pore suggestion is just a conceptual model (Rama & Moore), with no experimental validation at sub-zero T, we did include adsorption considerations also for this (**Figure 11 in previous versions**).

In the current updated version, we also do check diffusion under different K-adsorption values, up to 10^6 (**Figure 13 and Lines 606-627**).

(4) The authors state that in ADE-17, there is enough time for Ra-226 to diffuse out of sediment grains, while in ADE-2 there is not enough time for this to occur. However, the Ra-226 activities in both cores are comparable. Th-230 was only measured in ADE-17 so it is difficult to assess whether we would expect a higher reservoir of Ra-226 in ADE-17 (which could support a lower percentage of Ra-226 escaping from sediment grains, despite activities being comparable), but if we assume (as the authors do) that Th-230 is similar in all cores then the observed Ra-226 activities are not consistent with the authors hypothesis of low diffusion in one core and high diffusion in another. I understand wanting to avoid over-interpretation of the absolute activities because of possible issues with incomplete Ra extraction from samples, but the fact that multiple samples from within each core have similar activities lends confidence that these measurements are fairly

reliable.

- As already noted by the reviewer, ^{226}Ra in the pore space (ground-ice) of both cores is significantly lower than expected by the ^{230}Th on grain surface (**Lines 458-459**). This means that there is adsorption going on (see comment #3). Accordingly, if the supply of 226 (whether from surface or from inside the grains) is similar in all cores, then its activities should be higher in the more saline ADE-2 (as indeed happens with the short-lived 224 & 223). Therefore, the lower than expected 226 (and 228) in ADE-2 suggests that the original supply is lower in this core, which we attribute to no time for diffusion in this core.

Minor comments (line #s reference tracked changes document):

Line 113: "...the only radioisotopes that were used.." I suggest rephrasing this as: "the only radioisotopes that have previously been used"

Thanks, it was rephrased, (Lines 111-112).

Line 438: Data from ADE-1 has been added to Figure 9, this sentence should be updated to add a summary of these newly added points. These new data also need to be added to table C2.

Thanks, the data is updated in Table C2 and in the text, (Line 335)

Figure 10: The caption is cut off: it says "Note the" but the rest of this sentence is missing (this may be an issue with printing to PDF, but should be double checked).

- Thanks. Checked and corrected.

Line 554: I agree that this sample did have high Ra activities, but there are also samples in the other cores that had similar activities; for example, DR-AD-65 had similar activities of Ra-223 and Ra-228 and much higher activities of Ra-226, and DR-AD-151 had much higher activities of Ra-224. Is there another explanation (besides salinity) that can explain these other anomalous values?

- Thanks for the question. There could be several reasons for high Ra activities. A major one is about the ice content of the sample. E.g., very low ice content and low ice: grain ratios) could result in high Ra activities.

Line 567: This sentence says that negligible diffusion is assumed in the ground ice; isn't this in contrast to the hypothesis of Ra-226 enrichment via diffusion through the ice?

- **According to other comments by the reviewer, the discussion is now revised**

Figure 11: I recommend that the authors show $K=0$, $K=10$, and $K=100$ (i.e. replace $K=6$ with $K=100$) because this will match the discussion in the text, which includes an estimate for $K=100$ but not $K=6$. This will also help address the previous comments by both reviewers that $K=100$ has been previously cited as a reasonable groundwater value.

- **Correct. Our mistake. Now it is as suggested by the reviewer.**

Line 632: I do not agree with the deletion of this calculation, I think it was helpful to provide evidence that diffusion of a small percentage of Ra-226 would be enough to support the observed activities and show that the authors' hypothesis is feasible, particularly since there is a lack of data on diffusion in low temperature cryogenic environments. I recommend this calculation be revised (with a more rigorous estimation of the likely %desorbed) and re-added.

- **Calculation text is now re-included. (Lines 609-627)**

Line 757: Citation for Zhang et al. 2025 is missing in reference list

- **Thanks, it was added**

Table C2: Please use different symbols to indicate which activities are not reported due to low adsorption efficiencies, and which were below detection limits.

- **Correct. Thanks. Now, those are below detection are marked with "ND" and those that were not analyzed marked with "NA".**

Table C3: Why are errors not shown for ^{232}Th and ^{230}Th ? Please add.

- **Thanks, the errors, although very small (0.5%-2%), are added now. Also those of ratios.**