Response to reviewers

We thank both referees for the time devoted and for their efforts to improve the manuscript. We did our best to comply with the comments and to clarify unclear issues. We believe that the attached revised version is much better.

Before we respond to comments, we wish to highlight some key issues related to the structure and conclusions of our manuscript.

1. The environment discussed in this manuscript is not the common for Ra isotope studies. While Ra isotope research mainly deals with the aquatic setup (marine and aquifers), the permafrost is predominantly frozen, where the ground rules are quite different. Also, while the few other, pioneering permafrost isotopic works mainly focused on large ice bodies (e.g. Burn and Michel, 1988; Kipp et al., 2025), the work with granular pore-space ground ice is more tricky (e.g., Ewing et al. 2014 for U isotopes), with less control on essential parameters.
2. Based on all mentioned reservations, we actually do not use the Ra data for residence time calculation. Equation 1 was used just in order to show how isotope ratios are building up to steady-state isotope ratios, and how this is affected by adsorption, if liquid is present.
3. Our main conclusions are based on several key observations:
(a) significant and consistent differences in salinity between neighboring boreholes;
(b) large differences in long-to-short lived isotope ratios;
(c) large differences in parent isotope ratios on surface versus inside grains, which were compared to (b).
We suggest that
(1) the salinity differences are due to post permafrost formation salinization;
(2) differences in Ra isotope ratios are the result of the long-term supply of the long-lived ^{226}Ra by diffusion from inside the grains.
While details and the exact mechanism, surely the times involved, are opened to discussion, the large differences are undeniable, which is very difficult to explain by differences in sediment composition (parent nuclide ratios) within <30m.
We note that our suggested maximum timing for the ground ice modification in ADE-2 (2 ka) is circumstantial (deposition time of syngenetic permafrost at the site) and not derived from Ra isotope calculations.
We also note that the discussion of the adsorption effect, which was criticized by the referees, was just for a liquid water end-member, which is clearly not the case in the permafrost, as discussed in length in the response to referees below.

Our responses follow the reviewers' comments and are indented.

Line numbers refer to those in the track changes version.

Reviewer 1

1. In section 5.2 it is argued that the difference in the long-to-short lived isotope ratios between cores ADE-2 compared to ADE-17 and ADE-1 must be related to a difference

in the release of ^{226}Ra. This is not consistent with the fact that it is the short-lived isotope activities that change between these cores; ^{226}Ra is similar in all 3 cores, while the short-lived isotopes change by 1-2 orders of magnitude. Therefore, the explanation must be related to something that changes the release of the short-lived isotopes from the sediment grains. It is shown that ^{228}Th and ^{227}Ac (the parents of the short-lived isotopes) have higher activities on grain surfaces compared to bulk sediments, thus wouldn't a simpler explanation be that in the higher salinity core (ADE-2), more of these isotopes are released into the porewaters before decaying? The authors argue that this cannot be the case because Ra-226 is not increased in ADE-2 porewaters (with the idea being that all Ra isotopes should increase with higher salinity), but they have shown that more of the ^{226}Ra parent (Th-230) is locked inside sediment grains compared to the short-lived isotopes parents, so I would expect that there would be a larger source of surface-available short-lived isotopes. In other words, there are more short-lived isotopes available at the grain surface compared to ^{226}Ra that are ready to be released into porewaters when the salinity increases.

> - We very much agree. Actually this is what we basically wrote. The parent radionuclides, 228Th and 227Ac, which are concentrated on grain surfaces, allow 223 and 224Ra to get in pore space (no chance for such short-lived to get out of the grain, see revised text, **Lines 530-537**, and with the higher salinity in ADE-2 and the existence of some liquid water next to grains, more is left in the pore space of the more saline core ADE-2. The problem is that this does not happen with the 226, and it remains low in ADE-2. Accordingly, what we suggest is that the 226 has to make its way from inside the grains (diffusion), where most of the 230 reside, a process that takes long time, in particular considering the low T. In ADE-17, there is enough time for 226 to arrive (still, ratios are lower than bulk 230/227), while in ADE-2 there is not enough time for that.

Another possible explanation could involve recoil, which would be propelling the short-lived isotopes into the pore space more often than ^{226}Ra because of their shorter half-lives.

> The quoted 230Th/227Ac ratios are not concentration ratios, but rather activity ratios. So this is actually what is recoiling (assuming similar release fractions for 226 and 223). This is now highlighted in the Results (**Line 474**). Accordingly, there should be much more recoils of the 226 compared with the 223, if bulk is considered. The only way to keep 226/223 ratios similar to the 230/227 in the CEC is by not allowing 226 from inside the grains to arrive at the pore space. On the other hand, the much higher 226/223 in ADE-17 requires contribution from inside the grains.

2. I do not agree with the argument that cold temperatures would reduce adsorption but not diffusion (which is invoked to explain how ^{226}Ra could be released from the inside of sediment grains after long time periods). I agree that a slow diffusion process could become significant over time, but if atoms are moving via diffusion what would stop them from coming into contact with grain surfaces and becoming adsorbed? Both are processes related to the movement of atoms and should therefore be affected by temperature the same way. The authors point out that higher salinities in the ground ice will further reduce adsorption, but wouldn't this also reduce diffusion because radium is a cation? The comparison between measured ^{226}Ra in porewaters and estimated ^{226}Ra

in the CEC fraction (Figure 11) also shows that most ^{226}Ra must be adsorbed onto mineral surfaces.

> Thanks for this comment. Seems like there is a confusion between frozen state and the low temperature. First, adsorption should actually increase (higher retardation) in low T. But what we wrote was that adsorption is relevant only to the liquid state (if exists) and does not practically operate in the full frozen state **(e.g. Lines 596-597, 658-9, 669-70, 682-685).** Also, the chance of occurrence of liquid water in grains' nano-pores is probably even lower than in the permafrost pore space due to the low W:R ratio **(Lines 676-677 in the revised**), so the chance of adsorption is probably very low to negligible. And of course, diffusion is very low in cold temperature and decreases with increased salinity. On the other hand, we did mention the little information there is about diffusion in ice (**Lines 663-664**; unfortunately, not Ra), which is not as low as the D's we used.
>
> And we certainly agree, diffusion should be very low in cryotic conditions (**e.g. Line 657** in the revised and simulation in figure 13), also with saline water. But, as highlighted, we have no experimental data about D-diffusion in these conditions. On the other hand, we do not put constraints on the diffusion distance, which could be from the very outer rims of the grains (<1micron; **Lines 680-681**).

3. In section 5.2.2 the authors do a simulation to estimate how much ^{226}Ra could be released from within sediment grains. Assuming an R of 10 in the equation on line 460 is on low end of what might reasonably be expected; I have more often seen R= 10^3 used for saline water (e.g. Diego-Feliu et al., 2021; Kumar et al., 2020), and because R decreases with increased salinity and the salinities in this study are generally lower than that of seawater, it would be more reasonable to assume a higher R. Also, R is linearly related to K_d, which will increase at lower temperatures (Rama & Moore, 1996), thus R in permafrost will be higher than R in temperate groundwater systems. It is noted on line 466 that a higher R would reduce effective diffusion. Would the diffusive hypothesis still hold if R is on the order of 10^3?

> Thanks for allowing us to expand on this. First, as mentioned above, the whole discussion of adsorption is about the (unlikely) end-member case, where diffusion out of the grain would be through liquid-filled nano-pores. So even if diffusion this way is assumed completely halted by adsorption, it would still be executed through solid ice, which should be the main filling of the nano-pores. We re-stressed it in the revised **(Lines 685-6.).**
>
> Specifically about the adsorption, and assuming we deal with liquid. First, as highlighted in the text, the calculation was merely a feasibility test, clearly not quantitative. This is because we have no empirical data on either diffusion or adsorption in such a low T. Also, we do not think that R in saline water is 10^3 (e.g. Kiro et al. 2012). Actually, Diego-Feliu et al. (2021) mentions that the characteristic R in saline water is 10^1 (paper's conclusions; and in Kumar et al. 2020, there is a wide Kd range of several orders of mag, both added to our Reference list). For our case, while indeed low T should increase adsorption, the soil anoxic conditions and low pH would act the other way, i.e. reduce retardation. Nevertheless, since the presented calculations use arbitrarily-chosen values, if one choose to use R=1000, then the diffusion coefficients should be higher (e.g.10^-22 instead of 10^-24) or, alternatively, the effective

diffusion would cover shorter distances (**revised version Lines 680-681**). While <<1micron distances are getting close to recoil distances, diffusion would still facilitate 226 input into pore space. We note that as shown in the paper, there is a lot of ^{230}Th in the grains, so even if effective diffusion is <1micron, it could still add a significant amount of 226 to the pore space, assuming enough time has been allowed (as happened in the two cores other than ADE-2). And again, this whole discussion is just about diffusion in liquidized nanopores.

Minor comments:

Introduction:

Line 55: Kipp et al. (2025) also used Ra & Th isotopes to provide evidence of recent ice segregation, and this citation could be added here.

Correct. We added a reference to Kipp's paper **(Line 55-56).**

Line 70: Change "has" to "had"

Thanks. Changed **(Line 73)**.

Lines 86-87: Th (thorium) should not be super script.

Thanks. Corrected **(Line 90).**

Line 89: "it will always be on the solids" – it would be better to say that thorium prefers the solid phase, or that the "majority" will be adsorbed. The way it is currently written makes it seem as if there will never be dissolved thorium, which is not the case.

Thanks, the second reviewer commented on that too. We added the following: "thorium (parent isotope) is highly particle-reactive and tends to strongly adsorb onto mineral surfaces, whereas radium is more mobile and is more common in the dissolved phase. Also, the alpha-recoil effect during radioactive decay provides the kinetic energy necessary to eject radium from the mineral lattice into the pore space" **(Lines 92-96).**

Line 90: change "^{228}Ra has 5.8 years" to "^{228}Ra has **a half life of** 5.8 years"

Changed as suggested **(**also to 5.75 years, **Line 98)**

Section 1.1: I recommend adding a summary of Ra isotopes in cryogenic environments to the end of this section, to provide a brief overview of what is already known (and/or unknown).

We added the following section **(Lines 105-114):**

Radium isotope research in the polar regions has been mostly focused on the land-ocean interface, like shelf-deep basin exchange processes (Rutgers van der Loeff et al. 1994; Kipp et al., 2019, 2023), river discharge as a carrier of carbon and nutrients to the oceans (Rutgers van der Loeff et al., 2002; Bullock et al., 2022) or lakes (Dabrowski, 2020) and direct groundwater discharge to the sea (e.g. Dimova et al. 2015; Charkin et al., 2020). While also studied in relation with soil or river contamination (e.g. Chevychelov and Sobakin, 2017), as tracers of weathering processes (Linhoff et al., 2020), or as part of geological and pedological surveys (e.g. Wojtasik et al., 2017), radium isotopes have just recently studied as a potential tracer of permafrost degradation-segregation or

as a proxy for residence time determination (Weinstein et al., 2019; Kipp et al., 2025). In fact, the only radioisotopes that were used for the determination of ground ice residence time in permafrost soil were uranium isotopes (Ewing et al., 2015) and ^{3}H (for young ice, e.g., Burn and Michel, 1988).

Methods:

Line 137: Could microwaving the samples change the adsorption/desorption of radium isotopes by increasing the temperature? This would not impact the Ra isotope ratios but could be mentioned as a possible sample processing artifact (similar to the addition of Ra-free water).

Thanks. We added a notation about it in the revised version: "We also note that centrifuging, as well as the addition of Ra-free water to samples with low ice content, could result in some desorption from grain surfaces, as well as in salt dissolution when exists. Yet, both salts and adsorbed ions are assumed part of the pore space chemistry (see a similar conception by Ewing et al., 2015, regarding U isotopes in ground ice). Nevertheless, the discussion in this study focuses on Ra isotope ratios, which is less affected by possible desorption. " **(lines 165-170).**

Line 144-145: "…both salts and adsorbed ions are assumed part of the pore space chemical composition." – It's not clear to me how this statement is related to your interpretation of the results. Are you saying that you consider both dissolved and surface adsorbed ions in the same category? Would this influence the interpretation of the low $^{224}Ra/^{228}Ra$ ratios, which assumes that most of the ^{224}Ra stays adsorbed on grain surfaces?

Thanks. First about the general chemistry. During permafrost formation and pore fluid freezing, there is a salt exclusion process, which can include salt precipitation. For the sake of our methodology and discussion, which does not include dissolution or intense leaching, we consider all extracted ions as part of pore space chemistry. About Ra isotopes. Yes, it could affect 224/228, assuming more 224Ra could be added from the adsorbed (due to its parent 228Th). This was actually considered in the relevant discussion (section 5.4). Following the comment, we stressed this point both in the Methods **(Lines 168-169**) and in Section 5.4: "… some of the samples show $(224Ra/228Ra)_{AR}$ within the 'expected' range of 1-2, although this could be affected by enhanced desorption during sample processing in the lab. This is in particular observed in the deep, epigenetic samples of ADE-2, which could be related to the higher salinity (i.e. less adsorption or enhanced desorption) in these samples." **(Lines 766-767).**

Line 153: The salinities in this study are below that of seawater (with perhaps one exception), thus high salinity would not be a reason for low Ra adsorption efficiencies on fibers. This method has been shown to quantitatively extract Ra from seawater salinities (e.g. Moore and Ried, 1973, Journal of Geophysical Research). I recommend revising this sentence to focus only on the low pH/reducing conditions as the cause for low extraction efficiencies.

Unfortunately, while we respect all tests made in the past, in particular those of Moore, our experience with secondary fibers on endless seawater and saline groundwater samples shows that efficiency is usually between 70-90%. Nevertheless, we agree that the main control is that of pH and redox. Accordingly, we changed to: "Efficiencies were very variable, from 90% down

to <50%, with the lower end being mainly due to the relatively low pH and/or reducing conditions." **(Lines 176-178).**

Line 160: Are ^{223}Ra and ^{224}Ra activities reported in excess of their parent isotopes, or are they reported as total activities? If excess, that calculation should be introduced somewhere in the methods and the activities should be listed as "excess" in Table C2.

Unlike SGD, the reported activities here are total, no excess. This is a closed system, with both short- and the long-lived isotopes being produced within the permafrost. And the discussion is about the build-up to equilibrium with parents, so no reason to subtract the supported.

Results:

Line 205: Since the ice is referred to as "saline" and "brackish", it would be helpful to list the salinities in PSU in this section in addition to providing the chloride concentration. This would more easily allow the reader to compare with typical seawater salinities.

- Thanks for the comment. Although presenting chloride concentrations is a common method in hydrology, we agree about the need to present the total salinity. However, being in the continental arena, and considering terminology in the hydrological community, we prefer to present it by TDS. Accordingly, we changed it to: "ground ice in the epigenetic section, deeper than 5-5.5m, is mostly brackish to saline, with 450-3,230 mgCl^- l^{-1} (TDS of 900-6,500 mg l^{-1}; note that HCO_3^- and Mg^{2+} were not measured in some of the samples). In ADE-2, salinity is significantly higher (up to 7,960 mg Cl^- l^{-1} and TDS of 17,190 mg l^{-1}), with one outlier (DR-AD-125) reaching >23,000 mgCl^- l^{-1} and TDS of >40,000 mg l^{-1} (Table C1). While the higher salinities were observed at epigenetic depths (>5.5m), salinities in the syngenetic section of ADE-2 were also relatively high (TDS »1000 mg l^{-1}), which reached >6000 mg l^{-1} at the base of this section (Table C1)." **(Lines 253-261).**

Line 276-278: There are some ^{224}Ra activities <1 and several ^{223}Ra activities >1 so I recommend rephrasing this sentence instead of saying that ^{223}Ra activities were an order of magnitude lower than the other isotopes.

Thanks. Rephrased to: "while ^{223}Ra presents much lower activities, …" **(Lines 377-378).**

Line 281: I disagree with the statement that there is no ^{226}Ra trend with depth- Figure 6 shows generally higher activities in the epigenetic section. I think this is also true to some extent for the short-lived isotopes, though I agree it is a weaker trend.

Thanks. Text was rephrased: " Activities of ^{224}Ra and ^{223}Ra are fairly uniform and low in ADE-17 and ADE-1 (slightly higher in the epigenetic section), while significantly higher in the more saline ground ice of ADE-2 (epigenetic section; Figs. 6a- b and 7). ^{226}Ra and ^{228}Ra also show higher activities in the epigenetic, compared with the syngenetic section, with no difference between the cores, and with the highest activities observed at the top of the section (6-10 m depth, Fig. 6c-d)." **(lines 377-385)**.

Line 285: I don't think the word "Nevertheless" is appropriate here, because this sentence supports the previous statement (that the ratios do not show a coherent trend).

Agreed. We changed it to “with marked differences between the cores” **(Line 394).**

Line 295: This should reference table C2, not C3.

Thanks, the reference was changed **(line 436)**

Line 297: An outlier of 1.0 is listed for ADE-17 but I do not see this value on Figure 9 or in Table C2.

Thank you very much for this one, we have erased the sentence. **(Line 437)**

Line 310: How does high ^{227}Ac on grain surface explain the bulk sediment ^{230}Th/^{227}Ac ratios that are significantly higher than secular equilibrium? Or is there another explanation for the difference?

The high 227Ac on surface was mentioned in order to explain the relatively low CEC 230/227 ratios. To avoid mis-understanding, we changed text to: “…while all exchangeable fraction samples show lower than equilibrium activity ratios (2-12, Table C3, and Fig. 8a). The latter is apparently due to the high concentration of ^{227}Ac on grain surfaces.” **(Lines 477-478)**

Discussion

Line 338: …”although probably also contains” should be “although **it** probably also contains”

Corrected, **(line 506)**

Line 349: What kinds of processes would introduce saline water into the permafrost at this location? It is mentioned that it could be from intrusion or intra-permafrost segregation, but what would be the driver of these mechanisms? i.e. would the region need to be inundated with seawater or receive a pulse of groundwater?

Thanks. Here we establish the observations and the generic mechanisms involved. The possible trigger(s) and specific mechanisms are discussed in Section 5.3

Line 350: The comma between “results” and “support” is not needed.

Thanks, erased, actually the whole sentence was revised. **(Line 521).**

Line 376: “triplicate” does not make sense here- should this be “triple”?

Thanks, changed to triple **(line 550).**

Line 376: This is not solely a result of recoil, but also suggests that ^{226}Ra is staying adsorbed onto mineral grains rather than desorbing into the porewaters. This is consistent with previous studies that have shown low ^{226}Ra desorption in permafrost (e.g. Kipp et al., 2025 estimated ~1-3% of ^{226}Ra desorbed from frozen sediments in samples with low salinities- these numbers agree with the values the authors have calculated).

- It seems that we did not explain it well. The presented back-of-the-envelope calculation shows that, considering the 230Th activities on grain surfaces, there should be much more 226Ra in pore space, if only recoil is considered (and assuming a frozen space, i.e. no adsorption). It is, therefore, suggested that most

226 stays attached to the grains, which could be aided by adsorption (as suggested by the reviewer and in Kipp et al.), if a liquid film is present. Accordingly, we rephrased the text to: "In fact, ^{226}Ra activities in ground ice (1-60 dpm l^{-1}) of all three cores are significantly lower than the ^{230}Th activities in the CEC (0.02-0.17 dpm g^{-1}, Table C3), which considering a dry solid density of 2.5 g/cc, a relatively high porosity of 50% (100% ice content, Table C1) and recoil release fraction of 50%, is equivalent to 25-200 dpm l^{-1}(Fig. 11; a more conservative porosity of 25% would triple these activities). The much lower ^{226}Ra activities in the ground ice are probably the result of most of recoiled radium being kept on grain surfaces (Fleischer and Raabe, 1978; Suksi and Rasilainen, 1996), either physically-attached or adsorbed, if liquid water is present." **(546-553).**

Line 386: Should this only reference Figure 8? Parent isotopes are not shown in Figure 7.

Thanks for the correction. We changed it to Fig. 8. **(Line 562).**

Line 397: The argument that the ratios depend solely on ^{226}Ra is not consistent with the data; the short-lived isotopes are what changes between the cores (by orders of magnitude!), while ^{226}Ra is similar in all samples.

Thanks for this comment. Seems like we were misunderstood. First, what we discussed here is the theory about the buildup of isotope activity from parent, not our observations. In theory, in a non-changing setting or in a steady-state hydrological system, the 223 should get to secular equilibrium or steady state activities within 5-6 half-lives, which is 2 months. For 226 it takes much mor time. So considering the permafrost setup and times involved, buildup of ratios depends solely on the 226.

But, as said in the paper and commented by the referee, the observation is that it is the 223 that changes between the cores. Accordingly, we suggest that this is about the salinity. Assuming that some liquid water is present in the pore space, we expect all Ra isotopes to show higher activities in the more saline ADE-2. This indeed happens with the short-lived 223, but not with the 226Ra. So, although the difference between cores is about the 223Ra, it is the 226Ra behavior that should be explored.

This is explained in **Lines 528-533**: "The low ratios in ADE-2 are apparently because activities of the short-lived isotopes are significantly higher than in the two other cores (Fig. 6b), while ^{226}Ra activities are quite similar (Fig. 6a). However, the significantly higher salinity in ADE-2 should result in higher activities of all radium isotopes in this core due to the reduced adsorption (e.g., Kraemer and Reid, 1984; Gonneea et al., 2008; Kiro et al., 2012; Vinson et al., 2013), and assuming a non-complete frozen nature of the permafrost (e.g., Gilbert et al., 2019; Keating et al., 2018; Weinstein et al., 2019)."

Line 416: Or this suggests that the short-lived isotopes are not being released into the pore space.

We understand that it is suggested that instead of a time-controlled mechanism of 226 arrival from inside the grains, it could be that the difference between ground ice in cores is about the efficiency of 223 release into the pore space. Accordingly, the lower 223 and the higher 226/223 in ADE-17 is due to the reduced release of 223 into the pore space.

While we agree that less 223 should be released due to the lower salinity (higher adsorption, if liquid exists), it is not clear why shouldn't this be the case also with the 226. Adsorption should work similarly on both isotopes.

Line 427: Remove "as a matter of fact" from the beginning of the sentence; it is not necessary

Thanks, we erased the whole sentence. **(Line 633)**

Line 430: Where did the estimate of 50% emanation come from? This seems quite high, especially given that Figure 11 shows high ^{226}Ra adsorption on sediment grains. Or does the 50% emanation describe release from recoil? In that case I understand the 50% estimate, but another term needs to be added to describe how much is released from sediments into the pore space via desorption.

- Correct. It was really confusing. While we could explain better this calculation, it is not so much needed, and we decided to erase this calculation as a whole in order to keep the rhythm of discussion.

Lines 429-431: I am unable to replicate the result of 100,000 dpm/L from this calculation; I get a range of 10,000 – 40,000 dpm/L when I use the provided values.

As mentioned above, this part was erased.

Line 439: Rama is spelled with only one m

Thanks, corrected **(Line 653)**

Line 458: Is there a typo in the equation? What does "erfc" mean? (this is not defined in the sentence)

No typo. "erfc" is the "complementary error function", and the equation is the common solution for diffusion from an endless reservoir (there is also one with erf, but this one is the more common).

Line 467-468: It is not internally consistent to argue that frozen/cold nanopore spaces would prevent advection but not diffusion.

- We believe that the referee meant to adsorption (no advection). Adsorption is mainly about fluids/solutions, and it hardly exists between ice and solid, usually being dependent on the existence of a thin liquid film. On the other hand, diffusion exists in all physical states. So we see no reason to compare.

Line 533: This reference does not mention Ra isotopes; should this be a different citation?

We are not sure which reference is it about. All the three mentioned references discuss 224/228 ratios.

Section 5.4: Here the authors argue that the influence of adsorption is necessary to produce the observed ratios, which is inconsistent with the argument that adsorption does not influence ^{226}Ra because of the cold temperatures.

- Thanks for this comment. As mentioned elsewhere in the manuscript, there is a distinction between in grain nano-pores, where water:rock ratios is very low, therefore the loss of energy is more effective, than in the main pore space, where we and other authors suggested that the frequent occurrence of liquid. Second, also in the pore space we claimed for a relatively low adsorption of 10 due to (1) salinity, (2) pH, (3) redox, (4) the dominance of frozen water. Last, here the adsorption is about the 228Th, which is much more particle-reactive than radium.

Following the comment, we elaborated on the text: "…this was attributed to the adsorption of ^{228}Th… onto grain surfaces or aquifer rocks, thereby retaining some of the produced ^{224}Ra on the grains and preventing it from reaching the pore space fluid/ice. This, in turn, required the presence of some non-frozen fluid in the pore space, probably as thin films around the grains, in order to allow the highly particle-reactive thorium to adsorb onto grain surfaces." **(Lines 759-762)**

Conclusion:

Line 555: "…which affects similarly both radium isotope activities and the time of reaching secular equilibrium" – I do not understand this statement.

- Basically, adsorption affects both equilibrium activities (though not 226/223 ratios) and the time of arrival at steady state (the higher the adsorption, the earlier the arrival at equilibrium is). In any case, it is clearly confusing as part of the Conclusions. Accordingly, we erased this sentence and added: "The actual activities of both are also controlled by adsorption, which in turn is dependent on the existence of liquid water in the pore space under the discussed cryotic conditions." **(Lines 780-782).**

Line 565: The final statement about permafrost vulnerability in a changing climate does not seem related to the rest of the study- is the connection to the previous sentence that there will be more fluid migration as the climate warms? If so, this should be explained more clearly (I recommend adding another sentence or two explaining this hypothesized impact.)

Thanks for the comment, we added "As Arctic temperatures continue to rise, the degradation of saline permafrost may trigger subsurface fluid mobilization and destabilization, posing significant risks to infrastructure and accelerating the release of stored carbon into the atmosphere" **(Lines 795-797).**

Figures/Tables:

Please add error bars on data figures and in data tables for Ra & Th isotopes and isotope ratios.

Errors and error bars were added to Tables C2 and C3 and to all relevant figures.

Figure 2: arrows are missing between 231Pa, 227Ac, and 227Th.

Thanks, arrows were added

Figure 6: missing key

Thanks, Key was added

Figure 7: What are the smaller inset plots showing? These data do not match the data in the larger figure panels (e.g. the ^{226}Ra inset shows ADE-2 with higher Ra activities than all but one sample from ADE-17, while in the larger panel the activities are comparable between the cores). Also, in the ^{226}Ra inset, the y-axis scale is 0-2 while the scale of the larger plot is 0-20; the 0-2 scale does not seem correct based on the reported sample activities.

Figure 7: Caption incorrectly lists the isotopes for (a) and (b)- they should be swapped.

Thanks for noticing. All noted problems in Figure 7 were corrected.

Reviewer 2

This manuscript investigates ground-ice formation and intra-permafrost fluid flow in saline permafrost from Adventdalen, Svalbard, using major-ion chemistry and Ra, Th, and Ac isotopes. The topic is relevant and potentially valuable, and I found the attempt to use short- and long-lived Ra isotopes to investigate processes within permafrost and aquifer materials particularly interesting.

However, in its current form, I think the manuscript is limited by three main issues: (1) several key radionuclide concepts are not defined or used with sufficient precision; (2) the Ra isotope model is too simplified to fully support the residence-time interpretations; and (3) the proposed conclusion that elevated long-to-short-lived Ra isotope ratios reflect ^{226}Ra diffusion from inside mineral grains is not yet sufficiently demonstrated. Overall, the study has clear potential, but substantial revision is needed before the main interpretations can be considered robust.

General Comments:

C1. Terminology and conceptual definition of radionuclide pools: Several key radionuclide concepts are not used or defined with sufficient precision. This makes parts of the interpretation difficult to follow and, in some cases, conceptually ambiguous. The manuscript should clearly distinguish between activity and concentration, bulk sediment activity, alpha-recoil-accessible activity, exchangeable or surface-associated activity, and dissolved activity in porewater or ground ice. This is especially important for the use of the “CEC fraction”. CEC is a sediment property, but the manuscript appears to use “CEC activity” as a proxy for exchangeable or surface-associated Th/Ac activity. The authors should define exactly what this fraction represents, how it was isolated analytically, and why it is the relevant pool to compare with dissolved Ra activities or activity ratios. In addition, terms such as “secular

equilibrium" and "steady state" are used several times in a way that appears inconsistent with their strict radionuclide meaning. The authors should clarify whether they refer to radioactive parent–daughter secular equilibrium, steady-state transport conditions, or an apparent balance between production, transport, adsorption/desorption, and decay. Finally, the term "diffusion" also lacks a clear conceptual framework. In some parts of the manuscript, it could be interpreted as diffusion through liquid-filled nanopores or pore films, whereas in other places the authors seem to imply solid-state diffusion from inside mineral grains. These processes are physically different and should be clearly distinguished. Without these clarifications, comparisons between bulk activity, "CEC activity", dissolved Ra activity, and inferred equilibrium ratios are difficult to evaluate.

(1) We understand about the ambiguity. For Ra isotopes, we always use activities and activity ratios. 'concentration' was mentioned in order to address readers who may not be familiar with the term 'activity.' In the revised manuscript, we changed all to 'activities'.

(2) Regarding the CEC. In the methods chapter, we described what we did, which is leaching the sediment, using a common protocol with ammonium acetate, and measuring the parent thorium and actinium isotopes (the way done with the bulk sediment). We agree that referring to it as "CEC activities" is confusing, and we changed it now to "the exchangeable fraction" throughout the paper.

(3) We thank the referee for the opportunity to elaborate about 'steady state' and 'secular equilibrium'. When 'secular equilibrium' is mentioned, it refers to equilibrium with the parent isotope in the system (i.e. similar activities), and by secular equilibrium ratios we mean assumed parent isotope activity ratios (e.g. 21.7 for 238U/235U and 226Ra/223Ra). The term 'steady state' is used twice **(Lines 574 and 756)** in order to describe the condition of constant activities or ratios, which may depend on other factors except for the secular equilibrium ones (e.g. the 224/228 ratios, which are commonly higher than 1).

(4) By "Diffusion" we mean: 'the spontaneous net movement of atoms, ions, or molecules from an area of higher concentration (e.g. inside the grains) to an area of lower concentration'. In the environment we discuss, the diffusion can be through solid mineral lattice, ice or liquid water (both in nano-pores), or a combination of all the three. This is widely discussed in the paper (e.g. section 5.2.2). In the revised, we clearly discuss it, and we also stress (section 5.2.2, Conclusions and abstract) that we believe that diffusion mainly goes through frozen nano-pores **(e.g. Lines 23, 669-70, 676-7, 777-8).** This is further discussed in the reply to C3.

C2. Limitations of the Ra isotope model and AR interpretation: The modelling framework used to interpret the Ra isotope data is not sufficiently developed to support some of the residence-time interpretations. Eq. (1) is introduced as a single-radionuclide mass-balance formulation, but the manuscript uses it to interpret activity ratios involving several Ra isotopes from different decay chains. The assumptions and limitations of this extrapolation should be made explicit. In particular, the model appears to assume homogeneous conditions, constant source terms, constant

retardation/partitioning, no changes in water chemistry, and no mixing between fluids with different histories or salinities. These assumptions are critical in this study, because the proposed interpretation depends on comparing isotope ratios among cores that differ strongly in salinity, chemistry, and inferred residence time. The authors should also explain why modelling the $^{226}Ra/^{223}Ra$ ratio provides additional information beyond modelling ^{226}Ra ingrowth alone at the timescales considered. As currently presented, the model is useful as a conceptual illustration, but it does not yet provide a sufficiently robust quantitative basis for the inferred residence times.

- Thanks for the comment. We first wish to highlight that, in practice, there is no attempt to use the equation in a quantitative way, and no ages or residence times were determined based on it. It is used only in order to describe the buildup of Ra isotope activities. Second, we do explain that in terms of isotope ratios and the relevant times (>>years), 223 (and 224) may be considered uniform (steady state activities, **Lines 574-575**), therefore it is only the long-lived 226 (and initial ratios) that matters (**Lines 576-577**).

We agree with the reviewer that things could (always) be much more complicated. But again, we treat our observations in a very robust way. Specifically, we mentioned that ratios in ADE-2 (the more saline) seem close to equilibrium with parent activity ratios in the CEC fraction (surface-bound) while in ADE-17 they are much higher, closer to parent activity ratios in bulk sediment. We note that, as highlighted widely in the paper, ground ice chemistry (ion ratios) in the more saline ADE-2 is quite similar to seawater, which calls for the dominance of binary mixing between seawater and a fresh end-member, while WRI is more significant in ADE-17 (**e.g. Lines 267-279** and Figure 4a; section 5.3).

"why modelling the $^{226}Ra/^{223}Ra$ ratio provides additional information beyond modelling ^{226}Ra ingrowth alone at the timescales considered"?

Modelling of 226 alone requires control on too many parameters, e.g. recoil efficiency, adsorption/desorption, as well as the main unknown, which is how much of the recoiled (and desorbed) radium was actually sampled, considering the limitations on ground ice extraction (**e.g. Lines 179-180**). This is at least partly solved, using isotope ratios. We highlighted the above by showing that a major part of the radium is not sampled **(e.g. Lines 547-553**), which is probably due to both adsorption (another unknown) and non-complete extraction.

In summary, we did use the equation to show how radium isotopes and ratios build-up to secular eq. with parents, but our conclusions are not based on this. Rather, it is the robust observation of low ratios in one core, which are similar to parent ratios on grain surfaces, while much higher ratios in the other (closer to bulk sediment). This drove us to the conclusion about diffusion from inside the grains (no quantitative constraints).

Last, the mentioned 2000 years is not coming from equation-based calculations, but rather from observations about the syngenetic sediments, where ages were defined mainly by OSL **(Lines 519-522;** Gilbert et al. 2018).

C3. Insufficient evidence for diffusion from inside mineral grains: A central interpretation of the manuscript is that elevated long-to-short-lived Ra isotope ratios reflect ^{226}Ra diffusion from inside mineral grains over long timescales. However, the evidence for this mechanism is not sufficiently demonstrated. The manuscript should define what is meant by "diffusion from inside grains": solid-state diffusion through the mineral lattice, diffusion through nanopores or microfractures, or release from alpha-recoil zones. These processes have very different physical meanings and expected rates, especially under low-temperature or frozen conditions. The authors should also justify why diffusion is required to explain the data, rather than alternative processes such as alpha recoil from mineral surfaces or exchange/desorption, differences in sediment texture or surface area, or heterogeneous parent-nuclide distributions. At present, the conclusion that ^{226}Ra diffusion from inside grains controls the observed activity ratios is plausible as a hypothesis, but it is not yet sufficiently supported by the presented data or model.

> - We agree that we cannot provide a quantitative modelling support for the diffusion, but we do believe that this is the most plausible explanation for our observations. First, we do not believe that there should be a major difference in sediments texture and chemistry within 30 m. If this has been the case, nobody could use representative core compositions. Also, this could be at a certain depth or stratigraphic unit, but not along the whole core, from the end of glacial period up to the Late Holocene. We show large differences between parent isotope ratios on grain surfaces and the bulk, as well as between Ra isotopes in the different cores. We, accordingly, believe that the most reasonable explanation is that in one core (ADE-2) Ra isotopes are controlled by what released from surface **(Lines 599-601**), and in others it is joined by radium (only 226) that comes from deeper in the grains **(e.g. Lines 607-609**). This has now been revised in **Lines 599-612**.
>
> As discussed above (C1(4)), diffusion could be through mineral lattice, probably mainly through nano-pores (ice or liquid), which is discussed in length in the paper (Section 5.2.2). About the recoil from mineral surfaces. This could be a source, however, this should not be different (time-wise) from surface-bound, unless time-dependent diffusion is involved. The only reasonable way we see is by import from inside the grains, where ratios are apparently much higher than 238/235 eq. ratios, and this should go by diffusion, this way or another. "While contribution from the grains could be accomplished by direct recoil from grain rims, there is no reason for this not to happen within the same time scale as that of the exchangeable fraction, which is not the case in ADE-2. Alternatively, we suggest that this contribution is via diffuse diffusion out of the mineral grains, which is time-dependent (see 5.2.2)." **(Lines 609-612)**

Specific comments:

Line 80–85: "In this work, we report the activities (i.e., concentrations) of radium isotopes in ground ice…"

The expression “activities (i.e., concentrations)” should be revised, because activity and concentration are not strictly equivalent.

> “concentrations” was mentioned for the non-familiar reader. It is now changed to “activities (disintegrations per time)” **(Line 86)**.

Line 85–90: “Specifically, all Ra isotopes are produced by the α decay of different Th (thorium) isotopes (^{227}Th, ^{228}Th, ^{230}Th and ^{227}Th, respectively…”

There seems to be a formatting issue in this sentence: the isotope mass numbers and element symbols appear entirely in superscript.

> Thanks for the comment. Corrected **(Lines 90, 91)**.

Line 85: “Specifically, all Ra isotopes are produced by the α decay of different Th (thorium) isotopes (^{227}Th, ^{228}Th, ^{230}Th and ^{227}Th, respectively), although in the case of ^{223}Ra, we mainly relate to its ‘grandparent’ ^{227}Ac…”

This sentence should be clarified. Although ^{227}Ac may be the more relevant longer-lived precursor for interpreting ^{223}Ra systematics, ^{223}Ra is still produced directly by α decay of ^{227}Th.

> Correct, we have changed the sentence to: "In the case of ^{223}Ra, we usually relate to its grandparent ^{227}Ac, with a half-life of 21.772 years, due to the short life of its direct parent 227Th (half-life: 18.7 days)". **(Lines 90-91).**

Line 90: “An important aspect is that while thorium (parent isotopes) is a particle-reactive element (i.e. in the presence of water, it will always be on the solids), radium is much more mobile in water.”

The authors should briefly explain the main chemical and physical processes that make Ra more available in dissolved form than its Th parent isotopes.

> Thanks, we added the following sentences to explain the differences: "thorium (parent isotope) is highly particle-reactive and tends to strongly adsorb onto mineral surfaces, whereas radium is more mobile and is more common in the dissolved phase. Also, the alpha-recoil effect during radioactive decay provides the kinetic energy necessary to eject radium from the mineral lattice into the pore space.. " **(Lines 92-96).**

Line 90: “The ^{226}Ra is the longest-lived radium isotope, with a half-life of 1601 years…”

The citation seems unnecessary here, as these half-lives are standard nuclear constants.

> The citation was removed as suggested. **(Line 99).**

Figure 2: “The three U-Th decay series.”

Some alpha-decay arrows appear to be missing in the ^{235}U decay chain shown in Figure 2.

> Thanks, it is now fixed

Line 145: “For Ra isotopes, the solution was run 3–5 times through manganese-coated fibers in order to adsorb the radium.”

Please specify the flow rate used during Mn-fiber extraction and the amount of Mn-coated fiber used per sample.

Thanks. We added the dry fiber weight (20 g) and flow rate (<0.5 l/min) in **Lines 172-173).**

Line 155: "…we mainly focus on isotope ratios as all isotopes are affected the same by adsorption efficiency."

There appears to be a formatting error: this sentence is underlined and should not be.

Thanks, it is now corrected

Line 160: "…while ^{223}Ra was measured after ca. 10 days, as to minimize the cross-talk between ^{224}Ra and ^{223}Ra."

Was potential cross-talk between ^{224}Ra and ^{223}Ra checked or corrected following Diego-Feliu et al. (2020)? Please clarify.

Thanks for this comment. Diego Feliu (2020) concludes by simulations that should is a significant 224-223 cross-talk, either when 224/223 > 10 or when 224 cpm > 5. While 224/223 in our case were mostly >10, 224 was <<1 (one sample showed 2 cpm). Accordingly, we checked for cross-talk (compared immediate with later measurements), and found no cross-talk in any of our samples. We changed in the revised text: "The short-lived ^{224}Ra and ^{223}Ra were measured by the RaDeCC system (Moore and Arnold, 1996) within 2-3 days after water-soil separation, while ^{223}Ra was also measured after ca. 10 days, as to check for 220-219 cross-talk interferences (e.g. Diego-Feliu et al. 2020). We note that we found no evidence for cross-talk (i.e. time-corrected activities showed no higher activities in the immediate analysis), which is probably due to the low activities of both isotopes (^{224}Ra was mostly <0.5 cpm)." **(Lines 181-185).**

Line 160: "Fibers were measured for ^{226}Ra by an emanation system and Lucas Cells… ^{228}Ra was measured by a low background well-type HPGe gamma spectrometer…"

Please clarify why ^{226}Ra was not measured by gamma spectrometry together with ^{228}Ra.

While ^{226}Ra has also been measured by gamma spectrometry, we prefer the emanation system due to its lower detection limit. Also, gamma measurements of such low activity samples take at least a week per sample, which makes it the bottle neck of the lab work. This is also why some of the samples were not measured for ^{228}Ra. In any case, we see no problem with presenting results using a very traditional method.

Line 165: "Analytical errors on ^{226}Ra, ^{228}Ra, and ^{224}Ra were commonly ≤10%, while those of the low activity ^{223}Ra were in certain cases up to >30%."

For RaDeCC-derived ^{223}Ra and ^{224}Ra activities, uncertainties should preferably be calculated following Garcia-Solsona et al. (2008), or the uncertainty calculation procedure should be explicitly described.

Garcia-Solsona, et al. (2008). Uncertainties associated with 223Ra and 224Ra measurements in water via a Delayed Coincidence Counter (RaDeCC). Marine Chemistry, 109(3–4), 198–219. https://doi.org/10.1016/j.marchem.2007.11.006

> We actually followed the exact procedure, adapted by Garcia-Solsona. We even use the same excel sheets. This is not a methodological paper, and it is not common to detail the propagated error calculation protocol. Nevertheless, following this comment, we now write more in detail about the errors and we refer to Garcia-Solsona: "Analytical errors on ^{226}Ra, ^{228}Ra were ≤7%, while on ^{224}Ra they were ≤13%, and those on the low activity ^{223}Ra averaged 28%, and in two cases exceeded 40% (errors determined, following the protocol of Garcia-Solsona et al. 2008)." **(Lines 193-196)**

Line 175: "The adsorbed fraction was determined by the CEC (Cation Exchange Capacity) measurement."

The terminology and method used for the "CEC fraction" should be clarified. CEC is a sediment property, whereas the manuscript appears to use "CEC activity" for an exchangeable or surface-associated Th/Ac activity. Please define exactly what this fraction represents and provide more methodological detail on how the ammonium acetate extraction isolates this pool. At present, it is difficult to assess whether this corresponds to truly exchangeable radionuclides, surface-associated activity, or another defined fraction. Compare also with batch desorption approaches used to quantify exchangeable radionuclide pools, such as Beck and Cochran (2013).

- Thanks for the opportunity to clarify. The use of ammonium acetate to determine the CEC (cation exchange capacity) in soils is very common, and it is undoubtedly appropriate for the ultimate and accurate determination of the concentration of an exchangeable cation. We added relevant reference in the revised text (e.g. Sumner and Miller, 1996, Line 210). On the other hand, we agree that the term "CEC" and "CEC activity" could be misleading, therefore we changed to the "exchangeable fraction" throughout the paper (e.g. in Methods: "The exchangeable fraction of thorium and actinium on grain surface was determined by the CEC (Cation Exchange Capacity) ammonium acetate leaching protocol" (Lines 208-210) and in the last paragraph of 4.3 **(Lines 475-484).**
- About Beck & Cochran (2013). In this work, the authors directly determined the adsorption-desorption of radium from sandy sediments. While this is probably not so applicable to the silty-clayey soils of the permafrost, this is not what we aimed at in the CEC measurements. We determined the activities of the parent isotopes (Th, Ac, not Ra) in the exchangeable fraction, in order to determine the potential production of Ra by recoil, not by desorption.
  This is explained in **Lines 197-200**: "The Radium isotopes accumulate in the frozen pore space following recoil from their parent nuclides Th and Ac, which are mostly located either on or within the sediment grains. Accordingly, thorium isotopes and ^{227}Ac were measured both in the bulk grains and in the

exchangeable fraction of the permafrost sediment, after thawing and completing the ground ice-soil separation." We also changed in **Lines 208-210** to: "The exchangeable fraction of thorium and actinium on grain surface was determined…"

Line 180: "^{228}Th and ^{227}Ac … were studied separately by the RaDeCC system… assuming equilibrium has been achieved with their short-lived radium isotope."

Please explain more clearly how ^{228}Th and ^{227}Ac were measured with the RaDeCC system.

> We believe all the information has been given in the text, including sample preparation and analysis. Nevertheless, we now elaborated on the text: "For the bulk sediment measurements, 0.3 g of each sample was placed in a Teflon beaker and was treated with 1 ml HF and 5 ml HNO_3…. The remaining fraction of 4 ml was buffered with NaOH to pH~7 and kept for ^{228}Th and ^{227}Ac analyses. The exchangeable fraction of thorium and actinium on grain surface was determined by the CEC (Cation Exchange Capacity) ammonium acetate leaching protocol. Five grams of sediment was shaken with 25 mL ammonium acetate overnight… All samples for ^{228}Th and ^{227}Ac were run through 20 g manganese-coated fibers and measured by the RaDeCC system (Moore and Arnold, 1996), assuming equilibrium has been achieved with their short-lived radium isotope daughters." **(Lines 201…219).**

Line 275: "Activities of ^{226}Ra, ^{228}Ra, and ^{224}Ra in ground ice are quite similar, varying between <1 to 60 dpm L^{-1} … while ^{223}Ra is an order of magnitude lower…"

The reliability of the low ^{223}Ra activities should be better documented. Most ^{223}Ra activities, especially in ADE-17, are below or around 0.5 dpm L^{-1}. Considering that the recovered ground-ice volumes ranged from only a few mL to approximately 1 L, the total ^{223}Ra activity loaded onto the Mn-fibers may have been very low. As a conservative example, a sample with 0.5 dpm L^{-1} and 1 L of recovered water would contain ~0.5 dpm of ^{223}Ra. Assuming a RaDeCC detection efficiency of ~50%, this would correspond to ~0.25 cpm at the time of extraction. However, ^{223}Ra was measured after approximately 10 days, which is close to one ^{223}Ra half-life. Therefore, the expected detector count rate would decrease by approximately a factor of two, to ~0.125 cpm, before considering background, adsorption losses, and other corrections. At such low count rates, obtaining counting uncertainties below ~10% would require on the order of at least 100 net counts, equivalent to counting times of ~800–1000 min or longer. Such long RaDeCC counting times may be problematic because instrumental limitations, including He leakage, can affect measurement stability and reliability.

- Unfortunately, it seems that the referee missed the quoted uncertainties on ^{223}Ra, which were "up to >30%", now changed to more detailed: "Analytical errors…^{223}Ra averaged 28%, and in two cases exceeded 40%" (**Lines 195**). The revised text now reads "Analytical errors on ^{226}Ra, ^{228}Ra were ≤7%, while on 224Ra they were ≤13%, and those on the low activity ^{223}Ra averaged 28%, and in two cases exceeded 40% (errors determined, following the protocol of Garcia-Solsona et al. 2008)" **(Lines 193-196).** We also wish to note several things: 1. our samples were never a few ml ("42-1391 ml", **Line 162**); 2. when low, we did measure 223Ra overnight, and indeed, whenever there was a He leak we repeated the measurements, sometimes up to 5 or more runs; (3) long runs did not affect measurement stability.

In addition, ^{224}Ra activities are generally about one order of magnitude higher than ^{223}Ra activities. A simple decay calculation indicates that, after 10 days, ^{224}Ra activities would still remain substantially higher than ^{223}Ra activities, because the initial ^{224}Ra/^{223}Ra activity ratio is large. For example, if ^{224}Ra is initially ~10 times higher than ^{223}Ra, after 10 days the remaining ^{224}Ra/^{223}Ra ratio would still be approximately 3, considering the different half-lives of ^{224}Ra and ^{223}Ra. Therefore, ^{224}Ra–^{223}Ra cross-talk may be an important additional source of uncertainty.

- Please see the reply comment on Line 160.

The authors should provide, preferably in the Supplementary Information, all relevant RaDeCC counting information, including sample volumes, counting times, detector efficiencies, background count rates, decay corrections, propagated uncertainties, and an explicit evaluation of whether the quantification limits and cross-talk criteria of Diego-Feliu et al. (2020) are fulfilled.

> First, we understand that the referee is mainly concerned about the ^{223}Ra and ^{227}Ac measurements. As already noted and highlighted in the manuscript, errors on these measurements are high. On the other hand, as explained above, cross-talk was not observed, since ^{224}Ra is relatively low (Diego-Feliu et al. 2020).
>
> Second, while we added calculated error information and error bars to tables and figures, as requested by Referee #1, we do not see any reason to provide all the detailed raw data and processing information, asked by the referee. We note that while this could possibly be a relevant request in an analytical methodology publication, this has hardly been (probably never) done in geochemistry application papers. On the other hand, we did add reference to Garcia-Solsona et al. (2008); **Line 195-196**, since this is the error calculation protocol we all use.
>
> We stress that while errors on 223 and 227 are quite high, as empahsized, this has no impact on the observational differences between the groups discussed in this manuscript, and accordingly does not affect the discussion and conclusions of this paper.

Line 280: "^{226}Ra is similar in all three cores, showing no trend with depth, although the highest activities in ADE-17 were observed at the top of the epigenetic section."

I agree that ^{226}Ra shows no clear depth trend, but the data appear more scattered in the epigenetic section. This variability should be mentioned or briefly discussed.

> This was changed, following comments of Referee #1, to "^{226}Ra and ^{228}Ra also show higher activities in the epigenetic, compared with the syngenetic section, with no difference between the cores, and with the highest activities observed at the top of the section (6-10 m depth, Fig. 6c-d)" (**Lines 381-385**)

Figure 6: "Radium isotope activities in ground ice of the three ADE cores."

The figure should include a legend identifying the cores/symbols, and uncertainty bars should be shown for the Ra isotope activities.

Thanks, legend and error bars were added.

Figure 8: "Radium isotope activities in ground ice and their parent ratios in bulk soil grains and on grain surfaces…"

Uncertainty bars should be shown for all activity ratios in Figure 8.

Error bars were added, also in other figures.

Line 300: "^{228}Th activities in the CEC are similar to those in the bulk…"

This result requires further explanation and methodological validation. The manuscript should clearly distinguish between different sedimentary radionuclide pools: (i) bulk activity, which represents the total activity in the sediment grains, including both the inner mineral lattice and the grain surface; (ii) alpha-recoil-accessible activity, which corresponds only to the fraction located within the outer mineral lattice from which daughter radionuclides may be released to porewater after alpha decay; and (iii) exchangeable or surface-associated activity (CEC for them, I guess), which represents the fraction adsorbed onto grain surfaces and potentially available for desorption or ion exchange.

Conceptually, bulk activity should be much larger than the alpha-recoil-accessible activity, because the latter is only a small fraction of the former. Similarly, the alpha-recoil-accessible activity should generally be larger than the exchangeable activity, because exchangeable radionuclides are mainly those that have already been mobilized to porewater by alpha recoil, diffusion, or advective transport and then adsorbed onto grain surfaces. Existing alpha-recoil models show that the recoil-accessible fraction is limited by the short recoil distance relative to grain size (e.g., Sun and Semkow, 1998; Diego-Feliu et al., 2021). Therefore, it is difficult to understand how the ^{228}Th activity in the "CEC fraction" can be comparable to the bulk ^{228}Th activity, especially when ^{232}Th and ^{230}Th are two to three orders of magnitude lower in the same fraction. Such a pattern would require a clear mechanism for strong ^{228}Th enrichment on exchangeable/surface sites, or an external supply of radionuclides followed by adsorption, which is not straightforward in this setting. The authors should clarify whether this reflects a real geochemical process or an artefact of the extraction/analytical procedure.

- We are very grateful to the referee for this comment. Seems like we had a mistake in the ^{228}Th activities of some of the samples (quoted activities were for the whole sample instead of per gram). After correction (see revised table and figures), 228 activities in the exchangeable fraction are on the average twice those of ^{232}Th (0.18 and 0.08 dpm/g, respectively). This seems o.k., considering the fact that ^{232}Th is almost solely part of the mineral lattice, while ^{228}Th is produced by the decay of ^{228}Ra, which on top of production from ^{232}Th in the exchangable fraction, is also produced by (1) recoil during the decay of ^{232}Th close (10's nm) to grain surface, (2) little (if any) diffusion from inside the grains (should be within years, considering ^{228}Ra half-life of 5.75 years).

  In the revised we added: "Importantly, although being progeny of ^{232}Th, ^{228}Th activities are, on the average, twice the ^{232}Th activities in the exchangeable

fraction. This is probably due to the fact that ^{228}Th is also produced from ^{228}Ra that recoiled via the decay of ^{232}Th close to grain rims." **(Lines 466-468).**

Line 385: "The similarity between (^{226}Ra/slRa)AR ratios in ADE-2 and the ^{230}Th/parent nuclide activity ratios in the CEC fraction … suggests that pore space radium isotopes are in equilibrium with parent nuclides on grain surfaces."

Demonstrating secular equilibrium would require comparing the activity of each daughter Ra isotope directly with the activity of its corresponding adsorbed/surface-associated parent, not only comparing activity ratios. The use of these ratios alone does not clearly demonstrate equilibrium between pore-space Ra and parent nuclides on grain surfaces.

- While we understand the logic in this argument, we unfortunately do not agree. As highlighted in the Methods chapter and elsewhere **(e.g. Lines 166-170)** measured Ra activities are affected both by adsorption and moreover, by the methodological efficiency of ground ice sampling after thawing in the lab. This was shown by comparing 226Ra in ground-ice to the ^{230}Th in the exchangeable fraction, which was significantly higher (**Lines 547-553**). Isotope ratios are much less affected by these; hence this is the parameter used in our discussion.

Line 385: "Equilibrium ratios are supposed to be achieved within 5–6 half-lives of the longer-lived isotope (i.e., ^{226}Ra, with a half-life of 1600 years)…"

Secular equilibrium with a parent nuclide is reached after several half-lives of the daughter radionuclide, not the parent.

- We believe that the sentence was mis-understood. It is not about the parents, but about the two daughters. We write that building a steady-state equilibrium ratios (e.g. 21.7 in the 226/223 ratio) takes 5-6 half-lives of the 226. Following the comment, we changed to: "Equilibrium activity ratios of radium isotopes are supposed to be achieved within 5-6 half-lives of the longer-lived isotope (i.e., ^{226}Ra, with a half-life of 1600 years)" **(Lines 563-564).**

Line 390: "The concentration of a certain Ra isotope in groundwater at time t is described by Equation (1), assuming negligible dissolution and diffusion…"

The assumptions behind Eq. (1) should be stated more completely. This formulation is an analytical solution of a radionuclide mass-balance model in groundwater and involves additional assumptions beyond negligible dissolution and diffusion. In particular, it assumes a simplified homogeneous system, steady production terms, constant adsorption/retardation conditions, and no temporal or spatial changes in water chemistry or aquifer properties. This is important here because the manuscript interprets activity ratios involving several Ra isotopes from different decay chains and relates them to different parent radionuclides. Treating each isotope with a single-radionuclide formulation, without explicitly accounting for coupled parent–daughter ingrowth, sequential decay, mixing, or changes in retardation, may limit the reliability of the model-based interpretation. The authors should clarify these assumptions and discuss how they affect the interpretation of Ra activity ratios.

- In a locked space like the permafrost pore space, a homogenous system, steady production **(added in Line 566**) and (if at all) constant retardation are not less (probably more) likely than in the common aquifer. Why should there be a change in retardation or mixing in standing water or ice. Moreover, all of these affect similarly all isotopes, so it is not clear why not using the same formulation, in particular that with the times involved, the short-lived is not supposed to change, so it's all about one isotope – ^{226}Ra. We do talk about mixing, which is the event of saline water introduction, discussed in this paper. Following this comment, we added in the revised (**Lines 643-645)** the possibility of a continuous process: "We note that the mentioned timing is only if one assumes a single event of ground ice formation or residence time reset. Alternatively, this could be a continuous process, which could start significantly earlier than 2000 years but its main impact would have to be relatively recent, in order to allow salinization of the syngenetic, shallow permafrost, as well as to keep the ^{226}Ra relatively low." (see also Summary and Conclusion, **Lines 773-797).**

Line 395: "K is a unitless adsorption coefficient."

I believe K should be described as a distribution coefficient, as it represents how much of the radionuclide is adsorbed versus dissolved.

- We prefer the dimension-less K, following Krishnaswami et al. 1982, which is linearly related to the retardation factor (R=K+1). We use it in other papers (e.g. Lazar et al. 2008; Kiro et al. 2012,2013; Weinstein et al. 2019,2021; Rotem et al. 2024). The thermodynamic K= k(ads)/k(des) (kinetic k's). K is related to the suggested Kd (distribution coefficient) via the equation (Krishnaswami et al. (1982):

$$K = K_d\left[\frac{\rho_s(1-\phi)}{\phi}\right]$$

where both porosity (ϕ) and solid density (ρ_s) are are taken care of.

Line 395: "The short-lived isotopes ^{224}Ra and ^{223}Ra reach steady state activities within weeks…"

Please clarify what is meant by "steady state" here. Eq. (1) already assumes steady-state conditions, so it is unclear whether the authors refer to secular equilibrium with parent nuclides, constant porewater activities, or another type of equilibrium

We agree that there is a confusion about using these terms. But we never mentioned that Eq. 1 assumes steady state. Actually, in Kiro et al. (2013), we indeed assume steady state (dC/dt=0) and solved for distance, but in Kiro et al. (2015) eq. 1 solves for both distance and time, and this is the one used here (solution for small xx). Accordingly, reference was changed to "Kiro et al. 2015" (**Line 567**).

Regarding the main question, by "steady state activities" we mean secular equilibrium with parent isotopes on the grains, but since actual activities are affected by adsorption, we prefer to use the term steady state, as in Krishnaswami et al. 1982.

Line 395: "C is the activity [dpm L^{-1}] of the isotope…"

Please also define t in the description of Eq. (1). In addition, C is usually used for concentration, whereas activity is commonly denoted by A; the notation should be revised.

> (t) was added: "the time since water (or ice) emplacement in the pore space [yr]" (**Lines 571-572**). Also, while C and C_0 were used as in previous papers (e.g. Kiro et al. 2015), we changed these in the revised to A and A_0 **(Lines 568-569).**

Line 395: "…we will make use of the average isotope activity in ADE-2 e.g. 1.4 dpm L^{-1} for ^{223}Ra…"

Please provide the standard deviation associated with this average value.

> We added: "…stdev is 1.1 dpm l^{-1}, which is mainly due to one sample of 3.9 dpm l^{-1}" **(Lines 579-580).**

Line 400: "In Fig. 12, we show the buildup of $(^{226}Ra/^{223}Ra)_{AR}$ in the ground ice with time."

Please clarify the advantage of using the $^{226}Ra/^{223}Ra$ activity ratio in this simulation. At the timescales considered, a similar ingrowth pattern would be expected for ^{226}Ra alone, so the added value of the ratio is not clear.

> This is true, and it was discussed above (comment to Line 385). For our discussion, using 226 acitivies is relevant only if no adsorption and 100% efficiency of ground ice sampling is assumed. This is why 226 is normalized to 223 and simulation is shown for this ratio.
>
> Following the comment, we added: "The actual activities of ^{226}Ra in our samples are controlled by adsorption, as well as by the efficiency of the (thawed) ground ice extraction. Accordingly, we prefer to normalize ^{226}Ra to ^{223}Ra activites." **(Lines 572-574).**

Line 400: "($^{226}Ra/^{223}Ra$)AR will reach CEC $^{230}Th/^{227}Ac$ activity ratios (secular equilibrium)…"

If the authors are using the advective transport model in Eq. (1), this should not be described as "secular equilibrium", which refers specifically to radioactive parent–daughter systems where the parent half-life is much longer than the daughter half-life. It is also not the same as steady state, which is already an assumption of the model. Here, the authors appear to refer to convergence between dissolved Ra activity ratios and parent-nuclide activity ratios in the defined CEC fraction, which is a different concept and should be named more carefully.

In addition, the expected convergence between dissolved Ra ARs and CEC parent ARs should be justified. If porewater Ra receives contributions not only from surface-associated/CEC parent nuclides but also from alpha recoil from the mineral lattice, then the dissolved Ra isotope ratios would not necessarily be expected to converge to the CEC parent ratios. This additional Ra source should be explicitly considered.

Thanks for this comment, which allows us to clarify several things and revise the text to a certain extent. First, as explained above, equation #1 relies on Kiro et al. (2015) and Weinstein et al. (2021), which solves the mass transport equation in time and space, and does not assume steady state activities. Second, we agree that this is not secular equilibrium, so we erased this. Also, we agree that "secular eq. ratio" is confusing.

Accordingly, following this comment, we changed the whole paragraph: "Assuming that ground ice radium in ADE-2 is produced only from the exchangeable fraction, the time that will take $(^{226}Ra/^{223}Ra)_{AR}$ to reach exchangeable $(^{230}Th/^{227}Ac)_{AR}$ is dependent on the adsorption. With K=0 (i.e. R-1), this will take several thousand years. On the other hand, if some liquid is present in pore space (e.g. Keating et al. 2018; Weinstein et al. 2019), this will result in adsorption joining the process, and calculated residence times will significantly shorten.. For instance, with K=10 , as is the case for seawater salinities, (e.g., Weinstein et al., 2021; Garcia-Orellana et al., 2021; Diego-Feliu et al. 2021), time to reach equilibrium will be on the order of 100's years, and with K=100 times will reduce to 10's years. We note that the cryotic conditions and the more common observation of a frozen pore space do not favor too much adsorption. Accordingly, while the Th- Ra secular equilibrium cannot be quantitatively used for residence time determination, it still indicates that the ground ice in ADE-2 has not been recently formed, and that it is probably at least decades old. On the other hand, importantly, the relatively low $(^{226}Ra/^{223}Ra)_{AR}$ in ADE-2 indicates that there is no significant contribution from inside the grains, since judging by the high $(^{230}Th/^{227}Ac)_{AR}$ of the bulk sediment, this should result in higher $(^{226}Ra/^{223}Ra)_{AR}$. Below, we use this observation and the circumstantial evidence of the high salinity in the syngenetic section of ADE-2 to infer about the mechanism that is responsible for the higher $(^{226}Ra/^{223}Ra)_{AR}$ in the other cores." **(Lines 585-603).**

Line 400: "We note that even if initial ^{226}Ra activities is an order of magnitude higher, the time to reach equilibrium values would be similar…"

This statement should be explained analytically rather than only numerically. From Eq. (1), the characteristic timescale for approaching equilibrium is controlled by the exponential term, i.e. by λ (K+1), whereas C_0 only affects the initial offset from equilibrium. Therefore, the weak dependence of equilibration time on the assumed initial ^{226}Ra activity follows directly from the equation.

This is true. Nevertheless, we changed this whole paragraph, so we also took out this argument.

Line 400: "Assuming no adsorption (K=0 for a completely frozen pore space), $(^{226}Ra/^{223}Ra)$AR will reach CEC $^{230}Th/^{227}Ac$ activity ratios…"

The analysis would be stronger if it focused more directly on parent–daughter relationships, particularly the ^{230}Th–^{226}Ra pair.

As already mentioned above, actual activities depend very much on both adsorption and extraction efficiency, not practical to us.

Line 405: "For instance, if K = 10, as is the case for seawater salinities…"

The assumed value of K = 10 for seawater appears to be at the lower end of reported Ra distribution coefficients for saline conditions. The authors may want to compare it with published Kd ranges, for example those compiled by Kumar et al. (2020) and Diego-Feliu (2022), and discuss how sensitive the model results are to this assumption. See also the attached figure.

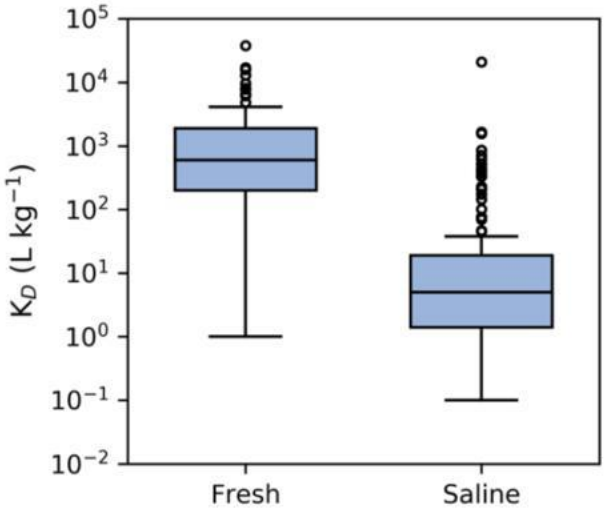

Figure 1.5. Characteristic values for Ra distribution coefficient from a compilation of ~250 values from the literature for fresh and saline groundwater (Appendix A).

First, Diego-Feliu et al. (2021) mentions that the characteristic R in saline water is 10^1 (paper's conclusions; and in Kumar et al. 2020, there is a wide Kd range of several orders of mag). Also, the attached figure 1.5 (from Diego-Feliu's thesis) shows that the median of saline water Kd's is about 2-3. Using a porosity of 0.3 and dry density of 2.5, calculated K will be 12-18, which is not very different from our 10. Also, the low oxygen and low pH should result in lower K. But mainly, as mentioned above, our conclusions are not based on the adsorption values, and this is only for the hypothetical case of a cryotic fully-liquid environment. Nevertheless, following the comment, we also related to a higher K **(Lines 595-596).**

Line 415: "…there was enough time in ADE-17 for some ^{226}Ra to diffuse out of the mineral grains."

Please define what is meant by "diffuse" in this context. This is the first time this process is introduced, and it is unclear whether the authors refer to solid-state diffusion within mineral grains, diffusion through nanopores/microfractures, or release related to alpha recoil.

- We revised this whole paragraph. Now it reads: "….there was enough time in ADE-17 for some ^{226}Ra to arrive from a different source, e.g. from inside the grains. While contribution from the grains could be accomplished by direct recoil, there is no reason for this not to happen within the same time scale as that of the exchangeable fraction, which is not the case in ADE-2. Alternatively, we suggest that this contribution is via ^{226}Ra diffusion out of the mineral grains, which is time-dependent (see 5.2.2). **(Lines 608-612).**

Line 430: "…and 50% emanation (release to pore space), ^{226}Ra in ground ice should be as high as…"

The term "emanation" should be revised or defined. It is commonly used for gaseous radionuclides such as Rn, whereas for Ra a term such as "alpha-recoil release" or "release to pore space" would be clearer.

- Thanks. This argument has been erased, following a comment from Referee #1

Line 445: "…radium diffusion from inside the grains could be significant…"

Please clarify what is meant by "diffusion from inside the grains". If the authors refer to solid-state diffusion through the mineral lattice, this mechanism should be justified, especially under low-temperature or frozen conditions.

- Thanks. This whole section is about this question. In general, and as also mentioned now in Summary and Conclusions and the abstract, we believe that it is mainly through frozen nano-pores **(e.g. Lines 23, 669-670, 778)**