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.

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.

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.

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.

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.

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.

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.

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.

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.

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.

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.

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.

Diego-Feliu et al. (2020). Guidelines and Limits for the Quantification of Ra Isotopes and Related Radionuclides With the Radium Delayed Coincidence Counter (RaDeCC). Journal of Geophysical Research: Oceans, 125(4), https://doi.org/10.1029/2019JC015544

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.

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

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).

Beck, A. J., & Cochran, M. A. (2013). Controls on solid-solution partitioning of radium in saturated marine sands. Marine Chemistry, 156, 38–48. https://doi.org/10.1016/j.marchem.2013.01.008

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.

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.

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. 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.

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.

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.

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.

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.

Sun, H., & Semkow, T. M. (1998). Mobilization of thorium, radium and radon radionuclides in ground water by successive alpha-recoils. Journal of Hydrology, 205(1–2), 126–136. https://doi.org/10.1016/S0022-1694(97)00154-6

Diego-Feliu, et al. (2021). New perspectives on the use of 224Ra/228Ra and 222Rn/226Ra activity ratios in groundwater studies. Journal of Hydrology, 596, 126043. https://doi.org/10.1016/j.jhydrol.2021.126043

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.

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.

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.

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.

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.

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.

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.

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.

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.

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 $\lambda\,(K+1\,)$, 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.

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.

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.

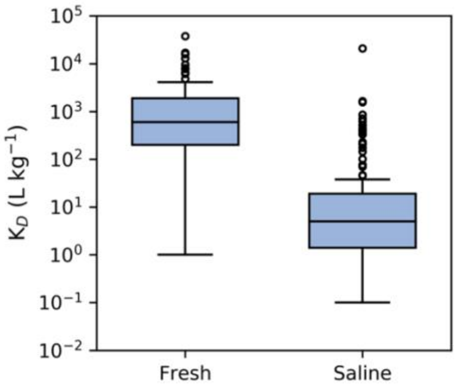

Figure 1.5. Characteristic values for Ra distribution coefficient from a compilation of ~250 values from the literature for fresh and saline groundwater (Appendix A).

Kumar, et al. (2020). A review on distribution coefficient (Kd) of some selected radionuclides in soil/sediment over the last three decades. Journal of Radioanalytical and Nuclear Chemistry, 323(1), 13–26. https://doi.org/10.1007/s10967-019-06930-x

Diego-Feliu, M. (2022). Improving the Use of Radium Isotopes and Radon As Tracers of Submarine Groundwater Discharge.

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.

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.

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.