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Reviewer 1:

This manuscript provides a nice extension of our understanding of Ra sorption to common minerals. As the authors point out a number of times, the very low environmental levels of Ra preclude spectroscopic measurements. It is certainly a weakness of the paper that none of the models are particularly satisfactory or applicable across all mineral types tested. However, the results of this study provide a substantial extension of our understanding of controls on Ra mobility in aquatic systems. I support publication of this manuscript in its current form. Two minor comments are included below.

Lines 91-100. This section states the finding of the study and should be moved to the conclusions or omitted.

In Table S1, what do the reported uncertainties represent? Replicate measurements or analytical uncertainty?

Reviewer 2:

This paper describes the results of radium adsorption experiment using different mineral matrix and solutions with different salinity in order to quantify radium adsorption equilibrium. The results show differential adsorption capacity of the different solid minerals and decrease of adsorption magnitude with increasing ionic strength/salinity. These results are consistent with previous studies and therefore there are no new breakthrough discoveries presented in this paper. Perhaps the most important contribution of this paper is that it provides systematic characterization of the adsorption capacity of the different minerals and the possible changes of Ra adsorption capacity with diagenetic modification and weathering of minerals. There is nothing wrong with this paper but in order to make it more attractive and relevant to ES&T readers I suggest some modifications that could improve the paper.

First, the paper addresses the changes in adsorption capacity between “brackish” and “seawater” salinities and the role of divalent cations over monovalent cations in completion of adsorption sites; yet there is no quantitative evaluation of the role of the chemistry on Ra adsorption. For example, the authors should include the Ca/Na ratios in the solutions to demonstrate the specific role and the thresholds from which Ca over Na affects Ra adsorption. Since there are no pure Na- or Ca-rich water in nature, quantification of the Ca/Na ratios on Ra adsorption is important for the ability to predict Ra retention to solids. It might require additional experimental work but this would provide a much better assessment of the role of the water chemistry on Ra adsorption, which is the major objective of this study.

Second, although the paper has a section on “Implications of radium mobility in soils, sediments and aquifers” – this section needs a major lift. It lacks a systematic literature review that would demonstrate actual cases of Ra occurrence and distribution in soils, sediments, and aquifers. For examples, several studies have shown elevated levels of radium in anoxic aquifers, which contradicts the high surface area and the claim that sulfur minerals like pyrite can adsorb Ra – how the authors reconcile this contradiction? Also the reference to hydraulic fracturing is misleading since hydraulic fracturing water is typically low-saline and Ra is not adsorb during hydraulic fracturing process. In short, there are many case studies that report Ra occurrence and distribution in soils and aquifers, and the authors should relate the results of their experiments to these studies. It is conducted in a very general way in the paper – it should be much more specific with actual case studies data.

The third point is related to the Introduction; the introduction section does not include detailed information on previous adsorption studies that have examined the retention of Ra to different minerals. It is important to specify the results and outcome of these experiments and to show how the new study is different from these previous experimental works. In particular, several studies have demonstrated the role of salinity and exchangeable cations on radium adsorption and thus it’s important to demonstrate how the experimental design of this current paper is different from previous studies. In addition, the authors do not include all possible processes and mechanisms that control Ra occurrence in aquatic systems (e.g., adsorption-desorption, recoil, dissolution, co-precipitation, decay). It is important to emphasis that dissolved Ra in water reflects complex processes, and adsorption is not the only mechanism of Ra retention. Finally, there is some confusion in the literature review; some of the previous studies address Ra occurrence in saline oil produced waters while other show results from low-saline groundwater in the Middle East. Many of the other studies that show the correlation of with on groundwater salinity are not cited. In short, a better and much wider literature review is needed.

Finally, some comments on the analytical section; most common Ra measurements in environmental samples are conducted by gamma spectrophotometry. The authors need to provide more information on the scintillation counting method, what nuclides were measured, the precision and detection limit of the method. Is there any a cross-method comparison that can demonstrate the capability of this method?

Reviewer 3:

Sorry, but I'm still not convinced of the section on surface complexation modeling.

The first paragraph (line 248-258) starts with "good visual fits were achieved...", but I can't find any such plots from which to evaluate this good visual match. In principle, SCM fit quality can visually be evaluated from % adsorbed vs. pH value plots together with the model curves. Fig. 1 provides the data points, but no any model curves (altough denoted "fits" in figure caption) to evaluate visually the good matching. RMSE values for logK prediction is not representative for a SCM fit quality, but for a fixed experimental condition data set (fixed pH, solid/solution ratio, etc.) only.

In the second paragraph (line259-274), the SCM approach is mixed up with ion exchange. Both processes are quite different with respect to clay minerals as can be read in any related textbook. SCM relies on the varying (pH dependent) charging on frayed edge sites, while ion exchange is due to pH independent negative charging in interlayer sites due to cation substitution in the clay lattice. Therefore, the flawed statement "Surface complexation modeling of Ra sorption to montmorillonite was achieved through inclusion of an inter-layer exchange reaction..." by which this paragraph starts, and further discussion thereafter, are highly misleading and definitely not on an EST state-of-the-art level of sorption modeling.