

# On the Influence of Ionic Strength on Radium and Strontium Sorption to Sandy Loam Soils

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Models which can estimate environmental transport of radioactive contaminants in natural and engineered systems are required to 1) deploy effective remediation strategies for contaminated sites, 2) design waste repositories for future waste streams, and 3) ensure protection of human and environmental health in all cases. These models require accurate transport parameters in order to correctly predict how these contaminants will move in the subsurface. This work aimed to determine more accurately the distribution coefficients for radium and strontium sorption to Savannah River Site (SRS) soils. Radium and strontium sorption to the soils was found to be highly dependent upon ionic strength due to competition for ion exchange sites. Radium distribution coefficients ( $K_d$ ) for the clayey soil were determined to be  $185.1 \pm 25.63 \text{ L kg}^{-1}$  and  $30.35 \pm 0.66 \text{ L kg}^{-1}$  for ionic strengths of 0.02M and 0.1M as NaCl which is the approximate ionic strength of groundwater at the SRS. Radium distribution coefficients for the sandy soil were determined to be  $24.95 \pm 2.97 \text{ L kg}^{-1}$  and  $9.05 \pm 0.36 \text{ L kg}^{-1}$  for ionic strengths of 0.02M and 0.1M as NaCl. Sorption of  $\text{Ra}^{2+}$  was generally greater than  $\text{Sr}^{2+}$ , consistent with the frequent use of higher distribution coefficients for Ra in performance assessments.

## Introduction

Radium is present in the environment as a decay product from uranium bearing ores as  $^{226}\text{Ra}$  which has a 1602 year half-life. Stable  $^{88}\text{Sr}$  is found in most rocks while  $^{90}\text{Sr}$  is present in the environment due to releases from legacy nuclear weapons wastes, nuclear reactors, or from atmospheric testing of nuclear weapons.  $^{90}\text{Sr}$  is a high yield product from the fission of  $^{235}\text{U}$ ,  $^{233}\text{U}$ , and  $^{239}\text{Pu}$ . Radium and strontium are both divalent cations existing only in the +2 oxidation state. Sposito [1] indicates that sorption affinity of the alkaline earth metals follow the trend  $\text{Ra}^{2+} > \text{Ba}^{2+} > \text{Sr}^{2+} > \text{Ca}^{2+} > \text{Mg}^{2+}$ , where increasing sorption occurs with increasing ionic radii. Since ionic potential, the ratio of the electric charge of the ion to the radius of the ion decreases with increasing ionic radius this implies that the larger ions will create a smaller electric field and be more prone to sorption. This trend also represents the increasing gibbs free energy of hydration of the divalent cations implying that ion dehydration and hydration may be coupled with sorption and desorption reactions, respectively. Chen and Hayes [2] propose that divalent cations form outer-sphere complexes which are relatively weak and can easily be displaced by other cations in solution. However, this has not been experimentally verified. The increase in  $\text{Ra}^{2+}$  sorption relative to  $\text{Sr}^{2+}$  observed in this work is consistent with the hypothesis that the hydration sphere of heavy alkali earth cations is perturbed upon sorption which may lower the free energy of the surface reaction. This is consistent with decreasing ion distances (and inferred dehydration) with increasing atomic mass of alkali and alkali earth cations monovalent alkali metals upon sorption to mica and rutile surfaces [3-9].

The Savannah River Site (SRS) located near Aiken, SC disposed of solid waste in the old radioactive waste burial

grounds (ORWBG). It has been estimated that the inventory of  $^{226}\text{Ra}/^{228}\text{Ra}$  and  $^{90}\text{Sr}$  in the ORWBG is 0.18 Ci and 54,000 Ci, respectively [10].  $^{226}\text{Ra}$  waste is primarily present as a daughter product of  $^{238}\text{U}$  disposal and there is approximately 17 Ci of  $^{238}\text{U}$  buried in the ORWBG [10]. Thus the  $^{238}\text{U}$  represents a significant source term of  $^{226}\text{Ra}$ , and based on the half-lives of  $^{238}\text{U}$  and  $^{226}\text{Ra}$ , risk from  $^{226}\text{Ra}$  disposition will still be an issue far beyond the 10,000 year assessment period of the PA.

Sorption of radium and strontium also is highly dependent upon ionic strength and the concentration of competing ions. Chen and Hayes [2] found that the sorption of  $\text{Sr}^{2+}$  increased with increasing pH and that the mineral sorption affinity increased as: montmorillonite > illite > silica. Furthermore, greater sorption was observed in systems with lower ionic strength (0.01 M NaCl compared with 0.10 M NaCl). This behavior was proposed to be due to competition between  $\text{Sr}^{2+}$  and  $\text{Na}^+$  based on the following reaction:



Where " $\equiv\text{X}^-$ " represents an ion exchange site on the soil and " $\text{--}$ " represents the electrostatic bond between the anionic site and the exchanging cation. The reaction stoichiometry maintains charge balance of the surface complex and Le Chatlier's principle indicates that higher concentrations of competing cations could prevent radium and strontium from sorbing to the soil.

Currently, SRS is using distribution coefficient ( $K_d$ ) values of  $17 \text{ mL g}^{-1}$  and  $5 \text{ mL g}^{-1}$  for the clayey and sandy soils, respectively, in the SRS PA [11] for strontium. The distribution coefficient is a linear function representing the ratio of the solid phase to aqueous phase concentration of a contaminant. These values were determined using actual SRS groundwater which had an ionic strength ranging from 0.01 to 0.1 M, but they may not be

applicable to all groundwater applications. In the absence of data describing Ra<sup>2+</sup> sorption to similar soils, the K<sub>d</sub> values for Sr<sup>2+</sup> are often used as a proxy. However, the expected stronger sorption of Ra<sup>2+</sup> relative to Sr<sup>2+</sup> results in a higher than expected potential risk from <sup>226</sup>Ra disposition. Looney et al. [12] recommended a K<sub>d</sub> value for radium sorption on SRS soils of 100 L kg<sup>-1</sup> with a range 10-1,000,000 L kg<sup>-1</sup>. These values were based on the sorption of other metals, namely strontium. Thibault et al. [13] gave radium K<sub>d</sub> values for a clay soil of 9,100 L kg<sup>-1</sup> and for a sand soil of 500 L kg<sup>-1</sup>. Nathwani and Phillips [14] were also able to show that increasing the concentrations of Ca<sup>2+</sup> resulted in decreasing the K<sub>d</sub> due to increased competition for surface sites. By generating separate K<sub>d</sub> values for Sr<sup>2+</sup> and Ra<sup>2+</sup> in this work, it may be possible to more accurately predict the risks posed by Ra which is expected to result in a lower estimated risk. As discussed above, a comparative study of these two ions may also facilitate a deeper understanding of the role of hydration on ion sorption.

The objective of this work is to directly measure <sup>226</sup>Ra K<sub>d</sub> values for two soils from the Department of Energy Savannah River Site and compare those values to <sup>90</sup>Sr sorption to the same soil. Therefore, the work proposed here may be valuable to <sup>90</sup>Sr geochemistry as well as <sup>226/228</sup>Ra geochemistry.

## Materials and Methods

### Soil Selection and Characteristics

The soils used for these experiments were obtained from the Savannah River Site. The subsurface sandy soil will be referred to as the sandy soil and the subsurface clayey soil will be referred to as the clayey soil. The clayey soil was heated in an oven at 85°C overnight to remove excess moisture. The sandy soil was already dry and did not receive any treatment. Specific characteristics of each soil are shown in Table 1. As the table indicates, both soils are very low in organic matter.

### Radium and Strontium Experiments

Radium and strontium sorption was examined on both soils. The experiments were performed with soil concentrations of 25 g L<sup>-1</sup> and pH 5.5. Two ionic strengths, 0.01 M and 0.1 M, were used to explore possible influences of ion exchange processes. Due to the requirement to adjust the pH of the samples using NaOH and HCl, the 0.01 M NaCl solutions were actually ~0.02

M NaCl and monitored for each sample. These experiments were performed using the two SRS sediments. Stable <sup>88</sup>Sr was used to spike the sediment suspensions and initial concentration ranged from 50 ppb to 1000 ppb. <sup>226</sup>Ra was also spiked into the soil suspensions with initial radioactivity concentrations ranging from 25 to 250 cpm/mL (counts per minute per mL). A summary of the experimental conditions is shown in Table 2 where each condition was performed in duplicate for each of the two soils. For comparison both stable-Sr and radioactive <sup>226</sup>Ra have been converted to units of mol/L in Table 2. <sup>226</sup>Ra sorption experiments were conducted with and without Sr<sup>2+</sup> present in order to observe potential competition between Sr<sup>2+</sup> and Ra<sup>2+</sup>. After spiking, the suspensions were mixed end over end for a week to ensure equilibrium [11]. The sample vials were then centrifuged to remove particles greater than 100 nm based on Stokes Law. Then a 1 mL aliquot of the supernatant was analyzed on the ICP-MS (Thermo XSeries II) to determine the strontium concentration.

To quantify the activity of aqueous <sup>226</sup>Ra remaining in solution, two different detection methods were employed. The first method involved pipetting approximately 4 mL of the equilibrated supernatant into a liquid scintillation vial along with 15 mL of High Safe 3 cocktail. This counting method assumes that no diffusion of <sup>222</sup>Rn out of the cocktail will occur, allowing detection of <sup>226</sup>Ra and 5 of its daughters (<sup>222</sup>Rn, <sup>218</sup>Po, <sup>214</sup>Pb, <sup>214</sup>Bi, <sup>214</sup>Po). Therefore, the activity of <sup>226</sup>Ra will be 1/6 that of the total activity measured after 30 days as the sample is permitted to reach secular equilibrium. The second detection method was performed by pipetting another 4 mL aliquot of the equilibrated solution into another liquid scintillation vial along with 10 mL of mineral oil scintillating cocktail. The mineral oil scintillating cocktail method is an ASTM standard method for radon measurements [15] and is useful because <sup>222</sup>Rn is the daughter product of <sup>226</sup>Ra. After the 30 days required to reach secular equilibrium passed, each vial was shaken to mix the immiscible fluids. Radon selectively partitions into the mineral oil phase which scintillates when radon and its daughter products decay and can be quantified. The samples were analyzed on the Quantulus Ultra Low Level Liquid Scintillation Counter (LSC) along with a set of standards prepared from a NIST traceable <sup>226</sup>Ra source to determine the <sup>226</sup>Ra concentration. All data shown in the Results section was generated using the modified ASTM standard method. The calibration curve generated using the <sup>226</sup>Ra standards is shown in Figure 1.

Table 1: Descriptions of SRS sediments used in this work

Name	Description	Sand/Silt/Clay (wt%)	Surface Area (m <sup>2</sup> /g)	pH	Organic Matter (wt-%)	Extractable Iron (mg <sub>Fe</sub> /kg <sub>soil</sub> )
Subsurface Sandy	Subsurface Yellow Sandy Sediment Low Organic Matter	97/2/1	1.27	5.1	<0.01	15.26
Subsurface Clayey	Subsurface Red Clayey Burial Ground Sediment Low Organic Matter	58/30/12	15.31	4.5	NA	7.06

Table 2: Summary of radium-strontium sorption experiments. Each condition was performed in duplicate for each of the two soils. cpm = counts per minute.

Ionic Strength as NaCl (M)	[ <sup>226</sup> Ra] (cpm mL <sup>-1</sup> )	[ <sup>226</sup> Ra] (mol L <sup>-1</sup> )	[ <sup>88</sup> Sr] (ppb)	[ <sup>88</sup> Sr] (mol L <sup>-1</sup> )
0.01	250	5.0E-10	1000	1.1E-05
0.01	185	3.7E-10	500	5.7E-06
0.01	125	2.5E-10	200	2.3E-06
0.01	60	1.2E-10	100	1.1E-06
0.01	25	5.0E-11	50	5.7E-07
0.1	250	5.0E-10	1000	1.1E-05
0.1	185	3.7E-10	500	5.7E-06
0.1	125	2.5E-10	200	2.3E-06
0.1	60	1.2E-10	100	1.1E-06
0.1	25	5.0E-11	50	5.7E-07
0.01	250	5.0E-10	-	-
0.01	185	3.7E-10	-	-
0.01	125	2.5E-10	-	-
0.01	60	1.2E-10	-	-
0.01	25	5.0E-11	-	-

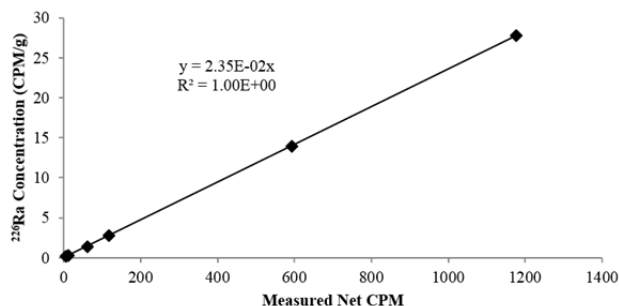


Figure 1. <sup>226</sup>Ra calibration curve for radon in mineral oil cocktail standards. LSC count time per sample was 60 minutes.

Initial experiments indicated that native strontium existed on the SRS soils and can desorb into the aqueous phase when dried sediment is suspended in 0.01 M NaCl. An experiment was performed using native strontium to determine long term  $K_d$  values for each sediment. Suspensions were made with 25 g L<sup>-1</sup> of sediment in 10 mL of water. The ionic strength was varied from 0 to 1.0 M (as NaCl) in increments (0.001, 0.005, 0.010, 0.050, 0.1, 0.5, and 1.0 M.). These suspensions mixed for 95 days. This was assumed to be a sufficient time to allow equilibrium to be reached. The vials were centrifuged to remove particles greater than 100 nm and the resulting supernatant was analyzed on the ICP-MS to determine strontium concentrations. Sediment samples then underwent microwave soil digestion using the same procedure described above. Strontium was separated

from the digested sample using a Bio-Rad poly-prep column packed with Eichrom Sr Resin. The column was first washed with distilled-deionized (DDI) H<sub>2</sub>O then glass wool was added to the top of the resin to keep it in place. The column was washed with 5 column volumes of 8 M BDH Aristar Ultra HNO<sub>3</sub>. The sample was spiked with <sup>90</sup>Sr to a concentration of 2000 cpm mL<sup>-1</sup> for use in yield calculations and acidified using BDH Aristar Ultra HNO<sub>3</sub> before being loaded onto the column. The column was washed with 5 column volumes of 8M HNO<sub>3</sub>. The <sup>88/90</sup>Sr was eluted from the column with 15 column volumes of DDI H<sub>2</sub>O into a preweighed vial. A 5 mL aliquot of the resulting sample was analyzed on the Quantalus Ultra Low Level Liquid Scintillation Counter (LSC) for <sup>90</sup>Sr analysis while the resulting solution was analyzed on the ICP-MS to determine the <sup>88</sup>Sr concentration in the sediment. Using the aqueous and sediment strontium concentrations, a  $K_d$  was obtained for each ionic strength.

## Results and Discussion

### Radium and Strontium Sorption to SRS Soils

For radium analysis, sorption studies were performed with and without strontium present (Table 1). The sorption of radium to the clayey sediment gave  $K_d$  values of 30.35 ± 0.66 mL g<sup>-1</sup> for [NaCl] = 0.1 M, 185.1 ± 25.63 mL g<sup>-1</sup> for [NaCl] = 0.02 M, and 326.2 ± 33.64 mL g<sup>-1</sup> for [NaCl] = 0.02 M and no strontium present (Figure 2). The  $K_d$  values were calculated using equation 1 based on the slope of a plot of the solid phase <sup>226</sup>Ra concentration (<sup>226</sup>[Ra]<sub>soil</sub>) versus the concentration remaining in the aqueous phase after equilibration (<sup>226</sup>[Ra]<sub>aqueous</sub>):

$$^{226}\text{[Ra]}_{\text{soil}} = K_d [^{226}\text{Ra}]_{\text{aqueous}} \quad \text{Equation 1}$$

For the highest initial radium concentration, more pH adjustment was required since the stock solutions were acidic. Therefore, the resultant ionic strength was higher than the rest of the set and these points were neglected when calculating the  $K_d$  values. There was less sorption to the sandy sediment which gave  $K_d$  values of 9.05 ± 0.36 mL g<sup>-1</sup> for [NaCl] = 0.1 M, 24.95 ± 2.97 mL g<sup>-1</sup> for [NaCl] = 0.02 M, and 34.55 ± 4.13 mL g<sup>-1</sup> for [NaCl] = 0.02 M and no strontium present (Figure 3).

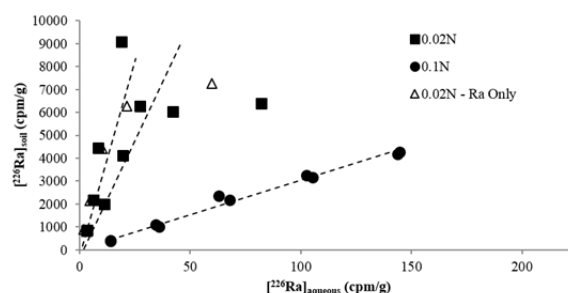


Figure 2. Radium sorption to clayey soil.  $K_d$  values of 30.35 ± 0.66 mL g<sup>-1</sup> for [NaCl] = 0.1 M, 185.1 ± 25.63 mL g<sup>-1</sup> for [NaCl] = 0.02 N, and 326.2 ± 33.64 mL g<sup>-1</sup> for [NaCl] = 0.02 N and no strontium present were reported.

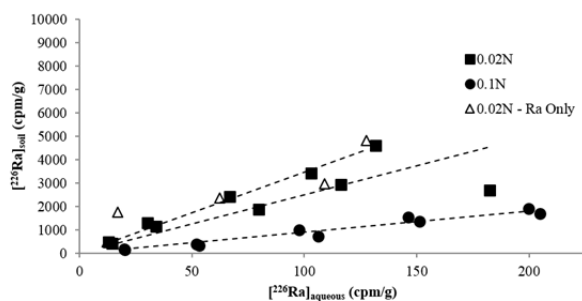
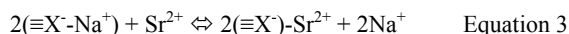
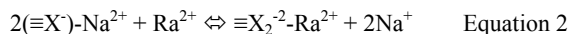


Figure 3. Radium sorption to sandy soil gave  $K_d$  values of  $9.05 \pm 0.36 \text{ mL g}^{-1}$  for  $[\text{NaCl}] = 0.1 \text{ N}$ ,  $24.95 \pm 2.97 \text{ mL g}^{-1}$  for  $[\text{NaCl}] = 0.02 \text{ N}$ , and  $34.55 \pm 4.13 \text{ mL g}^{-1}$  for  $[\text{NaCl}] = 0.02 \text{ N}$  and no strontium present were reported.

The radium  $K_d$  values for the samples with strontium added were lower than the radium only samples due to exchange site competition offered by the high mass loading of strontium compared to radium. The mass of strontium added was 6 to 7 orders of magnitude greater than the mass of radium added (Table 1). This discrepancy in masses was required to overcome the concentration of native strontium desorbing from the soils as well as to keep the activity of  $^{226}\text{Ra}$  low enough to safely work with it.

These results are in disagreement with the data currently being used in the SRS Performance Assessment (PA) of  $17 \text{ L kg}^{-1}$  and  $5 \text{ L kg}^{-1}$  for radium and strontium sorption to the clayey and sandy sediments, respectively [11]. However, the data in the SRS PA does not indicate the precise conditions under which the measurements were made. The results are consistent with the notion that sorption decreases as competing cation concentration increases. At higher ionic strengths, there is a higher ratio of competing cations to radium ions which decreases the ability for the radium to sorb to the surface sites which can be shown by the generic ionic exchange reactions:



Data describing  $\text{Sr}^{2+}$  sorption to the clayey and sandy soils are shown in Figures 4 and 5, respectively. The equation to determine the final strontium solid phase concentration is shown in Equation 4 and considers the concentration of native Sr that desorbed into the aqueous phase (Figure 6). Similar to the observed sorption behavior, the aqueous concentration of native strontium in equilibrium with the soil decreases with increasing pH for the clayey soil. However, it is noteworthy that the concentrations of aqueous Sr fall within a fairly narrow range of 2 to  $6 \mu\text{g L}^{-1}$  for solution with ionic strengths ranging from 0.001 M to 1 M. This indicates a potential solubility control on Sr partitioning. Incorporating the native strontium into the  $K_d$  calculations, the  $K_d$  values for the clayey soil were  $8.05 \pm 0.62 \text{ mL g}^{-1}$  for  $[\text{NaCl}] = 0.1 \text{ M}$  and  $32.06 \pm 3.62 \text{ mL g}^{-1}$  for  $[\text{NaCl}] = 0.02 \text{ M}$ . For the sandy soil, the  $K_d$  values were  $6.02 \pm 0.14 \text{ mL g}^{-1}$  for  $[\text{NaCl}] = 0.1 \text{ M}$  and  $5.86 \pm 0.35 \text{ mL g}^{-1}$  for  $[\text{NaCl}] = 0.02 \text{ M}$ . These experimentally determined  $K_d$  values for the clayey

sediment were lower than the values used for the SRS PA while the  $K_d$  values for the sandy soils were roughly the same [11].

$$[\text{Sr}]_{\text{soil}} = \frac{([\text{Sr}]_o - [\text{Sr}] - [\text{Sr}]_{\text{Blank}}) * V}{M_{\text{soil}}} \quad \text{Equation 4}$$

where:

- $[\text{Sr}]_{\text{soil}}$  Final concentration of strontium on soil, ppb
- $[\text{Sr}]_o$  Initial concentration of strontium in solution, ppb
- $[\text{Sr}]$  Final aqueous concentration of strontium, ppb
- $[\text{Sr}]_{\text{Blank}}$  Concentration of strontium desorbed from sediment in blank samples, ppb
- $V$  Volume of liquid (mL)
- $M_{\text{soil}}$  Mass of soil (g)

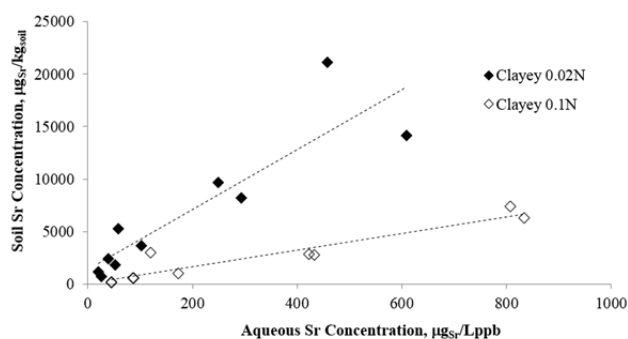


Figure 4. Sorption isotherm for strontium sorption on to SRS clayey soil. The  $K_d$  values were  $8.05 \pm 0.62 \text{ mL g}^{-1}$  for  $[\text{NaCl}] 0.1 \text{ N}$  and  $32.06 \pm 3.62 \text{ mL g}^{-1}$  for  $[\text{NaCl}] 0.02 \text{ N}$ .

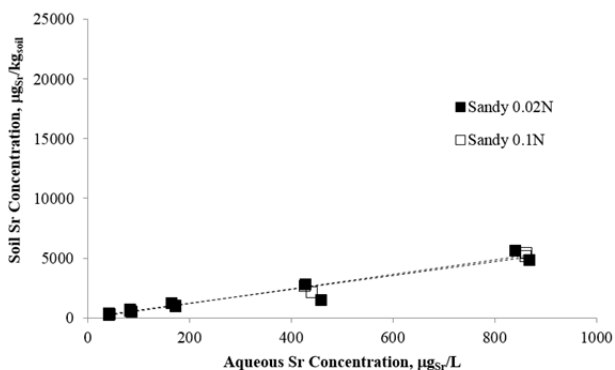


Figure 5. Sorption isotherm for strontium sorption on to SRS sandy soil. The  $K_d$  values were  $6.02 \pm 0.14 \text{ mL g}^{-1}$  for  $[\text{NaCl}] 0.1 \text{ N}$  and  $5.86 \pm 0.35 \text{ mL g}^{-1}$  for  $[\text{NaCl}] 0.02 \text{ N}$ .

An experiment was also performed to determine the  $K_d$  values for the native strontium on the SRS soils at varying ionic strengths. These measurements used the aqueous concentrations of native strontium shown in Figure 6 along with the analysis of the total Sr concentration in the soil from total acid digestion. These measurements indicated total Sr concentrations of  $3800 \pm 460 \mu\text{g/g}$  for the clayey soil and  $2110 \pm 480 \mu\text{g/g}$  for the sandy soil. The  $K_d$  values for native Sr were calculated by taking the ratio of the total Sr soil concentration to the aqueous phase

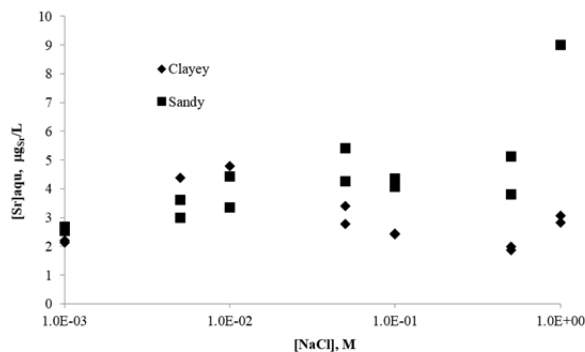


Figure 6. Native strontium dissolution concentration vs. ionic strength. Equilibration time of 95 days. Initial soil concentration  $25 \text{ g L}^{-1}$ .

concentration from the suspensions reported in Figure 6. These calculated  $K_d$  values are plotted as a function of the solution ionic strength in Figure 7. The  $K_d$  values reported here for the native strontium on the soils are approximately two orders of magnitude greater than the sorption experiments where strontium was added to the solution. This is likely due to the fact that the concentration of strontium associated with the soil that was determined by soil digestion includes strontium that is within the sediment matrix and possibly not available for dissolution/desorption. This differs from the batch sorption experiments where strontium was added to the solution and the concentration of strontium associated with the soil phase was calculated based on the difference between the initial and final aqueous strontium concentrations. As such, the release of stable strontium from these tests may in part be controlled by solubility, as opposed to the sorption based  $K_d$  construct. The batch sorption experiments move towards calculating a geologic  $K_d$  value that takes into account weathering of soils into smaller particles and possibly allowing more strontium to desorb from the soil. These  $K_d$  values may also be more representative of how a  $^{90}\text{Sr}$  release would behave after equilibrating with the subsurface sediment for hundreds of years and may be more valuable than a  $K_d$  calculated after an equilibration period of 24 hours.

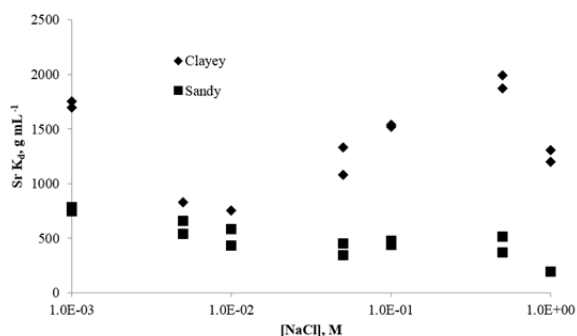


Figure 7. Native strontium  $K_d$  vs. ionic strength. Equilibration time of 95 day. Initial soil concentration  $25 \text{ g L}^{-1}$ .

## Development of Ion Exchange Conceptual and Quantitative Model

This work has shown radium and strontium sorption is highly dependent upon ionic strength which can be highly variable in subsurface environments. The  $K_d$  values reported above are only valid for the pH, ionic strength, and general conditions for which they were originally determined. Therefore, a cation exchange model would be a better predictor of sorption for these two cations than single  $K_d$  values. Using the Vanselow ion-exchange convention of representing the activity of the exchange species as a mole fraction, ion exchange constants for Ra and Sr for the chemical reactions shown in equations 2 and 3 can be determined as shown in Equation 5:

$$K_{M^{2+}} = \frac{a_{Na^+}^2 N_{M^{2+}}}{a_{M^{2+}} N_{Na^+}^2} \quad \text{Equation 5}$$

Where  $N_{M^{2+}}$  is the mole fraction of  $\text{Sr}^{2+}$  or  $\text{Ra}^{2+}$  on the soil (Equation 6),  $a_{Na^+}^2$  and  $a_{M^{2+}}$  are the activities of the aqueous cations, and  $K_{M^{2+}}$  is the ion exchange constant.

$$N_{M^{2+}} = \frac{c_{X_2M}}{c_{X_2M} + c_{XNa}} \quad \text{Equation 6}$$

The mole fractions are calculated based on the mass balance equation:

$$X_N = c_{XNa} + c_{X_2Sr} + c_{X_2Ra} \quad \text{Equation 7}$$

Where  $X_N$  is the total number of exchange sites as calculated from the cation exchange capacity and  $c_{XNa}$ ,  $c_{X_2Sr}$ , and  $c_{X_2Ra}$  are the concentrations of  $\text{Na}^+$ ,  $\text{Sr}^{2+}$ , and  $\text{Ra}^{2+}$  on exchangeable sites. Using this convention, the average ion exchange constants for  $\text{Ra}^{2+}$  and  $\text{Sr}^{2+}$  exchange are  $0.22 \pm 0.16$  and  $0.10 \pm 0.10$ , respectively. These constants use the data from both soils simultaneously indicating that similar exchange sites are active in both soils and demonstrating a universal approach for this method. The uncertainty is based up the standard deviation of all samples for both the sand and clayey soil. Separate ion exchange constants for Ra sorption to the clayey and sandy soil were  $0.26 \pm 0.16$  and  $0.18 \pm 0.16$ , respectively. It is noteworthy that this model only accounts for exchange of  $\text{Ra}^{2+}$  and  $\text{Sr}^{2+}$  for  $\text{Na}^+$ . Competition for other ions (e.g.  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ) is not considered. Expansion of this model to a more comprehensive system including all relevant ions is planned for future work. However, these constants are consistent with the greater sorption of  $\text{Ra}^{2+}$  relative to  $\text{Sr}^{2+}$  observed in Figures 2-5 as well as the relatively low influence of  $\text{Sr}^{2+}$  on  $\text{Ra}^{2+}$  sorption observed in Figures 2 and 3.

The work performed here will allow the development of more accurate models of the subsurface migration of radium and strontium. To the author's knowledge, these data represent the first data describing radium sorption to actual SRS sediments. In fact, there are few radium sorption experiments in the literature.

Previously, radium  $K_d$  values were assumed to be the same as the chemical analogue, strontium, for which there is a large body of experimental sorption data. One important practical problem with this assumption that radium and strontium will behave identically is that the radiological risk to the public associated with radium (daughter product of  $^{238}\text{U}$ ) and strontium disposition will be combined into one large risk. Thus, it is more accurate to spread this risk between strontium and radium separating their unique contributions and lowering the peak radiation does.

Radium and strontium sorption to the sediments was found to be highly dependent upon ionic strength due to competition for ion exchange sites. Radium  $K_d$  values for the clayey sediment were determined to be  $185.1 \pm 25.63 \text{ L kg}^{-1}$  and  $30.35 \pm 0.66 \text{ L kg}^{-1}$  for ionic strengths of 0.02 M and 0.1 M as NaCl. Radium  $K_d$  values for the sandy sediment were determined to be  $24.95 \pm 2.97 \text{ L kg}^{-1}$  and  $9.05 \pm 0.36 \text{ L kg}^{-1}$  for ionic strengths of 0.02M and 0.1M as NaCl. A more robust model than the  $K_d$  model is the cation exchange model, which would permit modeling strontium and radium behavior across a wider range of environmental conditions.

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