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Strontium adsorption and penetration in kaolinite at low Sr^{2+} concentration

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Abstract

Behavior of radioactive Sr^{2+} in contaminated soils is an important issue in relation to nuclear power plant accidents. The Sr^{2+} adsorption on kaolinite and its migration in a kaolinite soil were investigated because toxic effects of radioactive Sr^{2+} have been found very severe for living organisms at low Sr^{2+} concentrations. Adsorption isotherms of Sr^{2+} on kaolinite at different NaCl concentration and pH were obtained by the batch method. The calculated distribution coefficients (K_D) ranged between 600 and 40000 L kg^{-1} , which showed a strong preference for the adsorbed phase. The results were used to evaluate the ratio (r) of penetration length of Sr^{2+} relative to that of water in a model kaolinite soil. When the Sr^{2+} solution was percolated constantly into the kaolinite soil, the penetration of Sr^{2+} was delayed strongly at low Sr^{2+} concentration due to adsorption. The Sr^{2+} penetration length was only 0.001 - 0.056 % of the water penetration length at pH 6.5 (0.1-10 mmol L^{-1} NaCl). At pH 4.1 (1 mmol L^{-1} NaCl) the effect was about 17 times less than at pH 6.5 (1 mmol L^{-1} NaCl). Under all conditions, the Sr^{2+} penetration increased when the Sr^{2+} concentration increased due to the K_D decrease. The Sr^{2+} isotherms could be fitted well to the Langmuir adsorption equation, which indicates that only one site type is involved in the Sr^{2+} adsorption.

Key Words: Adsorption, distribution coefficient, kaolinite, penetration length ratio, strontium

1. Introduction

Strontium (Sr)-90 is one product of the radioactive elements of nuclear fuel materials emitting beta particles. Its half-life is 29 years, and it is harmful to the human health (Fetter *et al.* 1988). During the nuclear power plant accidents in Fukushima, Japan and Chernobyl, Ukraine, large amounts of radioactive species, including Sr-90, were released into the environment (Bondarkov *et al.* 2011; Steinhäuser *et al.* 2013). As the radioactive damage is serious, the environmental fate of Sr^{2+} must be considered, especially at low Sr^{2+} concentrations.

Kaolinite is one of the most ubiquitous phyllosilicates in soils and it has next to the basal

plane surface a relatively large edge surface (White and Dixon 2002); therefore, Sr^{2+} adsorption and transport in kaolinite soils deserve specific attention. Some reports consider Sr^{2+} adsorption on kaolinite on the basis of ion exchange (Wahlberg *et al.* 1965; Parkman *et al.* 1998; Bascetin and Atun 2006; Keçeli 2015). Sr^{2+} transport in quartz sand (Rod *et al.* 2010), sediment (Wallace *et al.* 2012) and porous material (Prigiobbe *et al.* 2012) were reported. The fact that the distribution coefficient provides information on both adsorption and migration of a component, which is the simplest parameter to characterize the influence of environmental factors on the fate of a pollutant. Thus, the distribution coefficient has been used to characterize Sr-kaolinite systems (Meyer 1979; Rafferty *et al.* 1981; Erten *et al.* 1988; Bunde *et al.* 1997; Bascetin and Atun 2006; Rani and Sasidhar 2012). The distribution coefficient of the Sr - kaolinite system under the given conditions can be determined when the adsorption isotherm of Sr^{2+} on kaolinite is known. Migration of Sr^{2+} in kaolinite soil can be conveniently characterized by using the ratio of average penetration length of Sr^{2+} relative to that of water. This ratio is determined by the distribution coefficient and the solid and solution content in the kaolinite soil (Bolt 1978). The migration of Sr^{2+} is retarded because of the adsorption of Sr^{2+} to kaolinite soil. This retardation can be quantified by using the parameter of distribution coefficient. However, there has been little research of Sr^{2+} penetration length at low Sr^{2+} concentration compared with water penetration length.

The objectives of the present study are therefore to investigate both the characteristics of Sr^{2+} adsorption on kaolinite at different NaCl concentration and pH values with emphasis to low Sr^{2+} concentrations and the migration of Sr^{2+} in a kaolinite soil under these conditions. The Sr^{2+} distribution coefficients were derived and used to describe the Sr^{2+} migration. To gain further insight in the mechanism of the adsorption, the Sr^{2+} adsorption results were analyzed with the Langmuir model. The Langmuir model assumes ideal adsorption behavior in a monolayer and considers the adsorption capacity as independent parameter and neglects valence effects (Langmuir 1918; Rani and Sasidhar 2012).

2. Materials and Experiments

The kaolinite from Iriki, Kagoshima, was suspended in deionized water and the fraction with a diameter less than $2\mu\text{m}$ was obtained by centrifugation. The obtained fraction was freeze-dried and stored in a closed container. The sample was characterized by using the X-ray diffraction apparatus (RINT1200, Rigaku Group of companies, Japan); the kaolinite content was 98.1% and the alunite content was 1.9%.

Three g of kaolinite powder was put in a 400 mL centrifuge bottle, mixed well with 300 mL

of 1 mol L⁻¹ NaCl solution and repeated for three times to reach a complete exchange. The clay was equilibrated with the prescribed NaCl solution at 0.1, 1 or 10 mmol L⁻¹. This process was repeated until the electrical conductance of the suspension equaled that of the prescribed NaCl solution at 0.1, 1 or 10 mmol L⁻¹. A suitable volume of 0.01 mol L⁻¹ HCl or NaOH solution was added to adjust the pH of the Na-Kaolinite suspension to 6.5 or 4.1. The final 10 g L⁻¹ Na-kaolinite suspension was kept for further use.

The Sr²⁺ adsorption experiments were conducted by the batch method using a series of polypropylene centrifuge tubes. Ten mL of the 10 g L⁻¹ Na-kaolinite suspension was pipetted into the centrifuge tube. Subsequently, 20 mL of SrCl₂ solution containing the prescribed NaCl concentration (0.1, 1 or 10 mmol L⁻¹) at prescribed pH (4.1 or 6.5) was added to reach an initial Sr²⁺ concentration in the range 2×10⁻³ mmol L⁻¹ to 6×10⁻² mmol L⁻¹. The tubes were shaken for 24 hours at room temperature. The pH of the equilibrium suspension was measured and recorded. Triplicate runs were performed for all batch experiments.

The equilibrium suspensions were centrifuged and the supernatants were gathered to determine the Sr²⁺ concentrations by inductively coupled plasma mass spectrometry (ICP-MS: PerkinElmer SCIEX-ELAN DRC-e). The adsorbed amount of Sr²⁺ on the Na-kaolinite was calculated by subtracting the amount of Sr²⁺ in equilibrium solution from the total amount of Sr²⁺ added initially.

3. Modeling

3.1. Distribution coefficient

The distribution coefficient, K_D (L kg⁻¹), describes the distribution of the adsorbate over the solid phase and the solution phase that can be defined as

$$K_D = \frac{Q_{Sr}}{[Sr]} \quad (1)$$

where Q_{Sr} is the adsorbed amount of Sr²⁺ on kaolinite (mmol kg⁻¹) and $[Sr]$ is the equilibrium Sr²⁺ concentration in the solution (mmol L⁻¹). The larger K_D is, the stronger is the accumulation in the adsorbed phase.

3.2. Retardation and penetration length ratio

Let a certain volume of solution containing a concentration $[Sr]$ mmol L⁻¹ of cation Sr²⁺ at the prescribed NaCl concentration and pH feed to per unit area of kaolinite. The distribution ratio, R_D , is a commonly used physical parameter for estimating the retardation of a dissolved pollutant in

the case of a solution with pollutant that permeates in a column of soil. Because the retardation is caused by adsorption, R_D is related to the partition of the contaminant between the solution and the solid (adsorbed) phase. As K_D quantifies the partition, K_D and R_D are directly related:

$$R_D = K_D \frac{\rho_b}{\theta} \quad (2)$$

where, ρ_b (kg L⁻¹) the bulk density of the kaolinite soil (mass of kaolinite in the column per unit volume of kaolinite and void) and θ the volumetric water content of the column of soil per unit soil volume in saturated condition (L L⁻¹ or m³ m⁻³). For the calculations, the soil bulk density of the hypothetical kaolinite soil has to be assumed to be homogeneous and constant. As the bulk density of natural soil usually ranges from 1.0 to 1.5 kg L⁻¹, a kaolinite soil bulk density of 1.25 kg L⁻¹ is used in this study. From this value, and the density of pure kaolinite of 2.65 kg L⁻¹ (Kuroda *et al.* 2003), the volume fraction of kaolinite in the soil can be obtained: 1.25/2.65 = 0.472. Consequently, the volume fraction of solution in the soil equals 0.528 when the kaolinite soil is saturated and this implies that under these conditions θ has reached its maximum value of 0.528. When only the saturation condition is adopted, R_D is proportional to K_D because the θ and ρ_b are constant.

In the saturated column, the average penetration length, x_p , of Sr²⁺ fed into the soil by the Sr²⁺ solution is defined by the relation (Bolt 1978):

$$x_p = \frac{V_F}{\theta} \frac{1}{1+R_D} \quad (3)$$

where V_F is the feed volume of Sr²⁺ solution per m² of soil (m³/m²) and θ is the saturated water content (m³/m³). V_F/θ is the average penetration length (m) of water in the soil. The ratio, r , of the average penetration length of Sr²⁺ to that of water is thus obtained as:

$$r = \frac{x_p}{V_F/\theta} = \frac{1}{1+R_D} \quad (4)$$

The inverse of r , $1+R_D$, is the retardation factor which is widely used for analyses of solute transport in soils.

As K_D is a positive quantity, R_D is positive and this implies that $r < 1$ and a small r implies a strong Sr²⁺ retardation. For large R_D ($R_D > 100$), i.e., large K_D , r is practically speaking inversely proportional to R_D . We can also assume that θ is a constant unsaturated volumetric water content. However, in order to obtain the largest penetration length ratio, the saturated condition is adopted here.

3.3. Langmuir model

The Langmuir model was used for the adsorption of Sr²⁺ on kaolinite as follows:

$$Q_{Sr} = \frac{Q_{Sr,m} K_L [Sr]}{1 + K_L [Sr]} \quad (5)$$

where Q_{Sr} (mmol kg⁻¹) is the Sr²⁺ adsorption, $Q_{Sr,m}$ (mmol kg⁻¹) is the maximum adsorption, K_L the Langmuir adsorption constant (L mmol⁻¹) and $[Sr]$ (mmol L⁻¹) is the equilibrium Sr²⁺ concentration. As the Langmuir model is the theoretically derived adsorption model, we use it for the theoretical consideration.

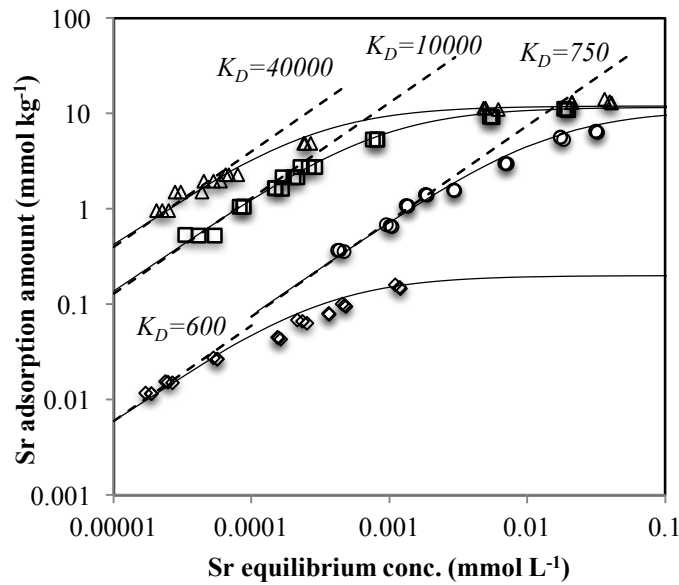
4. Results and discussion

4.1. The Sr²⁺ distribution coefficient

The adsorption isotherms of Sr²⁺ on Na-kaolinite at pH 6.5 and pH 4.1 and different NaCl concentrations are depicted in Fig. 1 as a double logarithmic plot. The measured adsorption increases linearly with slope ≈ 1 at low Sr²⁺ concentrations, but gradually levels off at higher Sr²⁺ concentrations. Slope ≈ 1 implies a constant adsorption affinity and K_D value; the low concentration ranges for which the slope ≈ 1 is the so-called ‘Henry region’; in this range the adsorption is directly proportional to the solution concentration, which can be well explained by Langmuir model as written in the later part. The dotted lines (slope =1) are lines with different K_D (L kg⁻¹) as indicated (see Eq.(1)). The constant values of K_D at low Sr²⁺ concentration are observed for each isotherm in Fig. 1. They ranged between 600 and 40000 L kg⁻¹. The magnitude of K_D clearly depends on the solution conditions: K_D strongly decreases with increasing salt concentration and decreasing pH. The K_D value at low Sr²⁺ concentration at 0.1 mmol L⁻¹ NaCl is about 53 times larger than that at 10 mmol L⁻¹ NaCl. The effect of pH is only investigated at 1 mmol L⁻¹ NaCl. K_D at pH 6.5 is about 17 times larger than K_D at pH 4.1. Similar effects for the kaolinite – Sr²⁺ system have been observed by a few researchers (Meyer 1979; Rani and Sasidhar 2012). For all solution conditions K_D decreases with increasing Sr²⁺ concentrations at Sr²⁺ concentrations beyond the Henry region, but the Sr²⁺ concentration where K_D starts to decrease depends on the solution conditions. Erten *et al.* (1988) and Bascetin and Atun (2006) also observed a decrease of K_D with the increasing Sr²⁺ concentration for kaolinite. Concluding this section, it is clear that pH and Na⁺ concentration influence the adsorption of Sr²⁺ on kaolinite and the Sr²⁺ distribution coefficient strongly.

The fittings of the Sr²⁺ adsorption data to the Langmuir equation are presented in Fig.1 as double logarithmic plot (solid lines). The measured values of Sr²⁺ adsorbed on kaolinite are quite well fitted to the Langmuir model. The adsorption affinity or Langmuir constant, K_L , and the adsorption capacity, $Q_{Sr,m}$, are collected in Table 1. The values of K_L are strongly influenced by

154 NaCl concentration because the influence of competition between Sr^{2+} and Na^+ on K_L differs with
 155 different Na^+ concentration. Thus, K_L obtained in this study should be interpreted as a *conditional*
 156 *constant*; $Q_{\text{Sr},m}$ are almost identical under different NaCl concentration.



157
 158 **Fig. 1** Sr^{2+} adsorption isotherms on kaolinite at two pH values and three salt concentrations. The dotted
 159 lines are lines of equal distribution coefficient (K_D) and the solid lines are fitting curves of Langmuir
 160 model. The symbols are measured values. At pH 6.5: $\triangle$ 0.1 mmol L⁻¹ NaCl, $\square$ 1 mmol L⁻¹ NaCl, $\circ$
 161 10 mmol L⁻¹ NaCl. At pH 4.1: $\diamond$ 1 mmol L⁻¹ NaCl

162 **Table 1** Langmuir isotherm parameters

pH	Na conc.	$Q_{\text{Sr},m}$	K_L	R^2
6.5	10	11.0	60	0.990
	1	11.6	1100	0.995
	0.1	12.0	3300	0.987
4.1	1	0.2	2500	0.934

163 By comparing K_D and K_L at given solution conditions, it should be noted that K_L is a constant
 164 for all Sr^{2+} concentrations, but that K_D is only constant in the Henry region. The difference is due
 165 to the fact that Eq.(5) takes into account the adsorption capacity, $Q_{\text{Sr},m}$. At relatively high Sr^{2+}
 166 concentration the probability of adsorption is decreased by the decrease of the vacant sites for Sr^{2+}
 167 adsorption. Eq.(5) takes this decreased probability into account, while Eq.(1) does not take the
 168 adsorption capacity into account. At sufficiently low Sr^{2+} concentration ($K_L[\text{Sr}] \ll 1$) Eq.(5)
 169 reduces to the Henry equation, $Q_{\text{Sr}} = Q_{\text{Sr},m} K_L [\text{Sr}]$, and this relation equals Eq.(1) with $Q_{\text{Sr},m} K_L = K_D$,

indicating that in the Henry region K_L and K_D are closely related.

The Sr^{2+} isotherms could be fitted well to the Langmuir adsorption equation, which indicates that only one site type is involved in the Sr^{2+} adsorption. The values of K_D keeping constant at low Sr^{2+} concentration were observed. Therefore, even if the Sr^{2+} concentration is extremely lower than those in this experiment, the values of K_D in the Henry region can be valid from the theoretical viewpoint. The value can be applicable for the Sr-90 contaminated sites where the Sr^{2+} concentration is supposed to be very low.

4.2. The Sr^{2+} penetration length ratio

The average penetration length ratio r , representing the average penetration length of Sr^{2+} relative to that of water is depicted in Fig. 2 as a function of the equilibrium Sr^{2+} concentration of the solution fed in the kaolinite soil. The four depicted data sets correspond with the four-adsorption isotherms depicted in Fig. 1. Since the ratio r is inversely proportional to $(1+R_D)$ and R_D is proportional to K_D , the large K_D values corresponding with the isotherms at low Sr^{2+} concentration induce small r values. As K_D increased with decreasing NaCl concentration and increasing pH, the reverse trend should be observed for r and this explains the decrease of r with decreasing NaCl concentration and increasing pH observed in Fig. 2. The small values of r indicate considerable retardation of Sr^{2+} upon solution penetration in the kaolinite soil. The increase of the penetration length ratio of Sr^{2+} in kaolinite with increasing Sr^{2+} concentration is due to the decrease of K_D . By considering the above trends quantitatively, the following observations can be made. The dependence of the penetration length ratio of Sr^{2+} in kaolinite on pH is considerable: at 1 mmol L⁻¹ NaCl and low Sr^{2+} concentration, the r value at pH 4.1 (7.04×10^{-4}) is 16.7 times larger than r at pH 6.5 (4.22×10^{-5}). It is favorable for Sr^{2+} adsorption at high pH, which results in a strong retardation of Sr^{2+} in sediments (Wallace *et al.* 2012) and iron oxides (Prigiobbe *et al.* 2012). The effect of the NaCl concentration at pH 6.5 and low Sr^{2+} concentration on the penetration length of Sr^{2+} is also large: at 0.1 mmol L⁻¹ NaCl r equals 1×10^{-5} which is 53.3 times smaller than r at 10 mmol L⁻¹ NaCl (5.63×10^{-4}). The trend of the present results corresponds well with literature results. Wallace *et al.* (2012) have reported that for sediments the presence of Na^+ resulted in the much lower sorption of Sr^{2+} which caused a significant migration of Sr^{2+} in the sediments. Hull *et al.* (2008) have reported that Sr^{2+} transported rapidly in the vadose zone sediment at high Na^+ concentration and the Sr^{2+} mobility decreased significantly in the absence of competing cations.

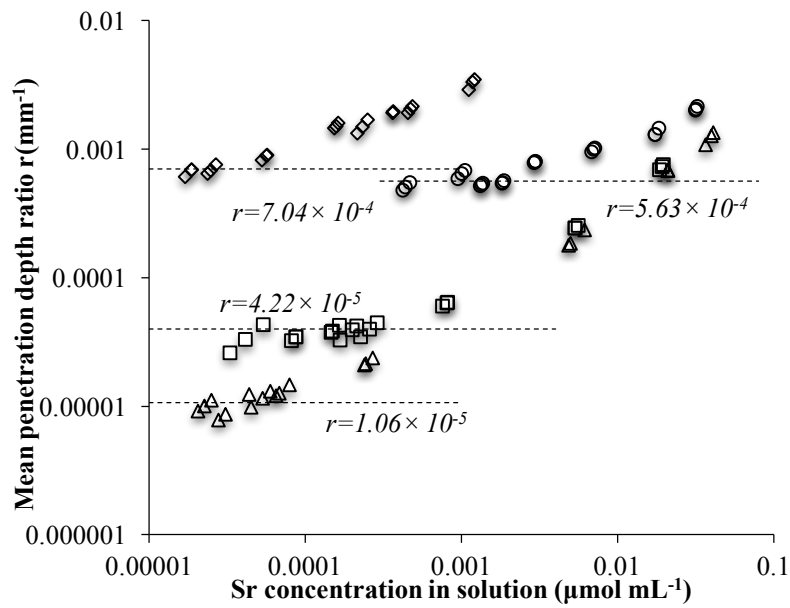


Fig.2 The penetration length ratio of Sr^{2+} versus the fed solution concentration Sr^{2+} . The dotted lines and indicated r values are the penetration ratios corresponding with the largest K_D values at low Sr^{2+} concentration for each solution condition. At pH 6.5: $\triangle$ 0.1 mmol L^{-1} NaCl, $\square$ 1 mmol L^{-1} NaCl, $\circ$ 10 mmol L^{-1} NaCl. At pH 4.1: $\diamond$ 1 mmol L^{-1} NaCl

The evaluated penetration length ratios of the Sr^{2+} at low $[\text{Sr}]$ are very small. The Sr^{2+} penetration length is about 0.001% of the water penetration length at the conditions of pH 6.5, 0.1 mmol L^{-1} NaCl and $[\text{Sr}] < 5 \times 10^{-8} \text{ mmol L}^{-1}$. At pH 6.5, 1 mmol L^{-1} NaCl and $[\text{Sr}] < 2 \times 10^{-7} \text{ mmol L}^{-1}$, the value is about 0.004%. At pH 6.5, 10 mmol L^{-1} NaCl and $[\text{Sr}] < 2 \times 10^{-6} \text{ mmol L}^{-1}$, the value is 0.056 %. At pH 4.1, 1 mmol L^{-1} NaCl and $[\text{Sr}] < 2 \times 10^{-8} \text{ mmol L}^{-1}$, the value is about 0.07 %. These values clearly indicate that Sr^{2+} is most strongly retarded by kaolinite at low NaCl concentration and relatively high pH.

The values of extremely low penetration length ratio of Sr^{2+} relative to that of water in kaolinite were calculated theoretically based on the distribution coefficient obtained from adsorption experiment. Although there are many complicated factors affecting the migration of Sr^{2+} in soils such as preferential flow, the average penetration length in the soils with uniform soil structure can be predicted.

In conclusion, because only one site type is involved in the Sr^{2+} adsorption in the kaolinite, the constant values of K_D are valid to calculate the average penetration length of Sr^{2+} even at extremely low Sr^{2+} concentration theoretically. The low values of average penetration length ratio show the strong retardation in kaolinite soils for Sr^{2+} at low Sr^{2+} concentration. Although the penetration ratio is a simple and useful parameter to estimate the mobility of Sr^{2+} , direct

observations of Sr^{2+} migration in soil are also necessary to confirm the theoretically calculated retardation.

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