Adsorption and Transport Behaviors of Potassium in Vermiculitic Soils

Shoichiro Hamamoto*, †, Tetsuya Eguchi**, Katashi Kubo**, Naoto Nihei*, Takashi Hirayama*** and Taku Nishimura*

*Graduate School of Agricultural and Life Sciences, The University of Tokyo
1–1–1 Yayoi, Bunkyo-ku, Tokyo 113–8657, Japan
** Agricultural Radiation Research Center, National Agriculture and Food Research Organization (NARO)
Tohoku Agricultural Research Center
50 Harajukuminami, Arai, Fukushima-shi, Fukushima Pref. 960–2156, Japan
*** Fukushima Agricultural Technology Centre
116 Shimonakamichi, Takakura, Hiwada-Machi, Koriyama-shi, Fukushima Pref. 963–0531, Japan

†shoichi@soil.en.a.u-tokyo.ac.jp

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It is important to understand potassium (K) behavior in soils since K fertilization is one of the most effective countermeasures to reduce the radiocaesium (RCs) uptake by crops. The K behavior in soils is highly influenced by the clay mineral composition. In this study, we investigated the K behavior for non-vermiculitic and vermiculitic nature of soils at Fukushima Prefecture in Japan. In the adsorption experiments, vermiculitic nature of soils exhibited more adsorption of K onto the soils as compared to non-vermiculitic nature of soil. One dimensional column transport experiments using 42K as a radioisotope tracer showed that almost similar K mobility for all soils as supported by adsorption isotherms of K. However, percentage of exchangeable K in the total K retained in the soil was higher in non-vermiculitic nature of soil, while the percentage of fixed K was higher in vermiculitic nature of soils. Thus, the existence of vermiculite with high K fixation capacity was suggested to reduce K retention as an exchangeable form in vermiculitic nature of soils.

Key Words: potassium, vermiculitic soils, adsorption, transport behavior, potassium-42

1. Introduction

Radioactive caesium (134Cs and 137Cs, RCs) was emitted from the disabled Tokyo Electric Power Company’s Fukushima Dai-ichi Nuclear Power Plant on March, 2011, and diffused into the environment, including agricultural area in eastern Japan. As one of most effective countermeasure to reduce RCs uptake by crops, potassium (K) fertilization when applying basal fertilizer is recommended. Effectiveness of K fertilization is evaluated by an extent of increase in exchangeable ammonium acetate-extracted K (Ex-K) content (e.g., for soybean14) since the RCs and K exhibit a great degree of physico-chemical similarity, and thus RCs complete with K in the processes of macro-elements by crops.4, 21) Therefore, knowledge of adsorption characteristics and mobility of the applied K in such soils polluted by RCs is essential for effective reduction of RCs uptakes.

Clay mineral compositions are one of important factors governing ion behaviors in soils. It is well known that the retention of alkali cations such as K, Rb, and Cs is highly associated with their adsorption on frayed-edge site (FES) located at boundary of
mica core and vermiculite structure in a particle of micaceous minerals.\textsuperscript{7} Mukai et al. (2016)\textsuperscript{15} found that partially-vermiculitized biotite (weathered biotite) sorbed RCs far more than the other clay minerals such as fresh biotite, illite, smectite, kaolinite, halloysite, allophane, and imogolite, on the same substrate. In addition, the sorbed RCs in the weathered biotite was hardly leached with 0.1 mol dm\textsuperscript{−3} hydrochloric acid. Thus, it is also expected that the vermiculitic soils have high fixation capacities for K as well as Cs, hereunder affecting Ex-K in soils after K fertilization. It has been also reported the K depletion in rhizosphere result in increase in plant uptake of RCs.\textsuperscript{10, 22, 23} It is also noted that vermiculite which originated from the weathering of granite is one of major clay minerals produced in Fukushima, Japan. Therefore, it is important to understand K behavior in such vermiculitic soils for effective countermeasure against RCs uptake by additional K fertilization. In this study, the K behavior in agricultural soils with different clay mineral compositions (non-vermiculitic and vermiculitic soils) in Fukushima were investigated based on the batch and column transport experiments.

2. Materials and methods

2.1 Soils

The soils at the depth less than 20-cm (A-horizon, plow layer) were sampled from three different agricultural fields at Fukushima Prefecture in Japan (Soil-A, -B, and -C) on Dec. of 2015. Soybean and wheat were cultivated in both Soil-A and -C and Soil-B, respectively. Soil-A (non-vermiculitic soil) was sampled at upland field located on the left bank of Abukuma river in northeastern part of Fukushima basin. Soil-A is influenced by Late Miocene dacite tuff.\textsuperscript{13, 18} Soil-B (vermiculitic soil) was sampled at the field located on Abukuma highland (eastern part of Nihonmatsu city) where geology was granodiorite.\textsuperscript{13} Soil-C (vermiculitic soil) was sampled at the field located on northeastern part of Koriyama basin where upstream geology was granodiorite.\textsuperscript{13} All fields were polluted by radioactive caesium (\textsuperscript{134}Cs and \textsuperscript{137} Cs, RCs) due to the emissions from the disabled Tokyo Electric Power Company’s Fukushima Dai-ichi Nuclear Power Plant on March, 2011.

Physical and chemical properties of each soil are shown in Table 1. Clay, silt, and sand fractions of soils were measured by a pipet method.\textsuperscript{5} Soil texture for each soil was light clay, sandy loam, and sandy clay loam, for Soil-A, -B, and -C, respectively. Total carbon was measured by CN coder (vario EL cube, Elementar, Kanagawa, Japan). Soil particle density was measured by a pycnometer method.\textsuperscript{5} Exchangeable ammonium acetate-extracted K (Ex-K) content for each soil was measured by 1:5 (by weight) mixtures of soil and 1 mol dm\textsuperscript{−3} ammonium acetate. The mixture was kept in a shaker for one hour and the supernatant was removed after centrifugation at 10,000 rpm for 30 min, and filtered through 0.45 µm filters. The K concentration in the filtered solutions were analyzed by atomic absorption spectrometry (AA-7000, Shimadzu Corp., Tokyo, Japan). As shown in Table 1, Ex-K was highest and lowest in Soil-A and Soil-C, respectively. The clay fractions for each sample were saturated with Mg or K using chloride salts\textsuperscript{11} and clay mineral compositions were assessed using X-ray diffractometer (SmartLab, Rigaku Corp., Tokyo, Japan). The Mg-saturated clay fractions were then treated with glycerol and reassessed. The K-saturated clay fractions were heated to 300 and 550°C for 1 h and reassessed. In addition, quantification of short-range ordered minerals (e.g., allophane) were achieved by combination of sodium pyrophosphate extraction and dark oxalate extraction,\textsuperscript{20} and Al, Fe, and Si in extract were determined by ICP-OES (Vista-MPX, Varian technologies Japan, Tokyo, Japan).
Batch experiments

Adsorption isotherm experiments were carried out at five different concentrations in K with triplicates under equilibrium condition. The samples were prepared in polypropylene 50 mL centrifuge tubes by mixing soil (4 g) and KCl solution (40 mL) at 3, 5, 10, 50, 100 ppm of K in 1:10 mass ratio. All samples were kept on the shaker for 24 h at 100 rpm. The centrifuge tubes were then put in the high-speed refrigerated centrifuge (SAKUMA 50A-IVD, Japan) at 10,000 rpm for 30 min. After centrifugation, the supernatant was removed and filtered through 0.45 μm filters. The equilibrium concentrations of K in the filtered solutions were analyzed by atomic absorption spectrometry (AA-7000, Shimadzu Corp., Tokyo, Japan).

Charge characteristics

Cation exchange capacity (CEC) of soils were determined by Ca-saturation at various pH.25) Soil (2 g) was placed in 50 mL pre-weighted centrifuge tube and 40 mL of 0.5 mol dm$^{-3}$ calcium chloride (CaCl$_2$) solution was added, and suspension pH was adjusted to 4.0, 5.0, 6.0, 7.0, and 8.0 by adding 1 mol dm$^{-3}$ chloric acid or saturated calcium hydroxide starring with pH electrode (9600-10D, Horiba Scientific, Kyoto, Japan) in supernatant of 0.01 mol dm$^{-3}$ CaCl$_2$ at soil solution ratio of 1:10.24) Cs-fixing capacity was determined as simple indicator of vermiculitic nature.16) A 1.0 g aliquot of each soil type was washed with 0.5 mol dm$^{-3}$ caesium chloride solution 3 times. Soils were washed once by deionized water then dried at 110°C overnight to promote fixation of Cs to vermiculite interlayer. The soils were washed with 1 mol dm$^{-3}$ ammonium acetate 5 times to remove Cs adsorbed to exchangeable sites, then digested with 10 mL of 40% hydrofluoric acid, 5 mL of 69% nitric acid, and 5 mL of 40% perchloric acid.26) Ca and Cs concentrations of solutions were determined by atomic absorption spectrophotometry (ZA-3000, Hitachi High-Technologies Corporation, Tokyo, Japan).

Column transport experiments

In the column transport experiments, the $^{42}$K (half-life of 12.36 h), a radioisotope tracer, was used as a tracer of K movement in soils. The $^{42}$K was obtained from the $^{42}$Ar–$^{42}$K generator.1) In this generator, $^{42}$Ar, whose half-life is approximately 33 years, decays to continuously produce $^{42}$K. The $^{42}$K was accumulated to a steel cathode inserted into a generator. By washing the cathode with 2 mL of KCl solution (66 g/L), the KCl solution with $^{42}$K was obtained. The KCl

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Table 1 Physical and chemical properties of soils used in this study

| Soils | Particle density g cm$^{-3}$ | Dry bulk density g cm$^{-3}$ | Gravimetric water content g g$^{-1}$ | Total porosity % | Total carbon % | Ex-K mmol kg$^{-1}$ | Clay % | Silt % | Sand % |
|-------|-----------------------------|-----------------------------|--------------------------------------|-----------------|----------------|---------------------|-------|-------|-------|
| A     | 2.51                        | 1.05                        | 0.15                                 | 58.2            | 1.41           | 5.3                 | 19    | 24    | 58    |
| B     | 2.56                        | 1.10                        | 0.11                                 | 57.0            | 0.77           | 2.0                 | 5     | 10    | 85    |
| C     | 2.80                        | 1.10                        | 0.11                                 | 60.7            | 0.66           | 0.9                 | 11    | 15    | 74    |
solution with $^{42}\text{K}$ was well mixed with each soil (wet soil corresponding to 100 g dry soil) resulting in 66 mg-K/100 g dry soil, and was incubated for around 1 hr. Each soil without K mixture was first repacked to the acrylic column (diameter: 5-cm, height: 20-cm) up to 16-cm height from the bottom with dry bulk densities from 1.05–1.10 g cm$^{-3}$ (Table 1). Each soil mixed with KCl solution with $^{42}\text{K}$ was subsequently repacked to the surface 4-cm thick layer with the same dry bulk densities (i.e., surface 4-cm thick layer contains $^{42}\text{K}$ and deeper layer is $^{42}\text{K}$ free). Rainfall was applied from the top of the column using a rainfall simulator connected to a Mariotte’s tank. During the experiments, constant rainfall intensity of 30 mm/h was kept. After the rainfall for one hour, the soil column was sectioned in 2-cm slices. Samples were taken from each slice and the total $^{42}\text{K}$ was measured using a Germanium semiconductor detector (GEM-10185-P_35-TP21068B, Seiko EG&G ORTEC, Tokyo, Japan). For each sample, 1:5 (by weight) mixtures of soil and either pure water or 1 mol dm$^{-3}$ ammonium acetate were prepared where the mixtures were kept in a shaker for one hour. The supernatant was removed after centrifugation at 10,000 rpm for 30 min, and filtered through 0.45 µm filters. The $^{42}\text{K}$ in the filtered solutions was analyzed by the Germanium semiconductor detector. In this study, the $^{42}\text{K}$ obtained from the mixtures by pure water and 1 mol dm$^{-3}$ ammonium acetate were defined as dissolved and dissolved plus ion exchangeable forms, respectively. In addition, the $^{42}\text{K}$ obtained by subtracting $^{42}\text{K}$ for 1 mol dm$^{-3}$ ammonium acetate extracts from total $^{42}\text{K}$ were defined as a fixed form.

3. Results and discussion

The XRD charts are shown in Fig. 1. Mg saturated clay from Soil-A showed 1.47, 1.01, and 0.90 nm peak indicating presence of expansible 2:1 type clay minerals, mica, and zeolite. The 1.47 nm peak shifted to around 1.8 nm by glycerol treatment indicating expansible 2:1 type clay mineral was mainly smectite. Conversely, the expansible 2:1 type clay minerals of Soil-B and -C would be mainly vermiculite because the peaks around 1.8 nm were not observed in Soil-C and very weak in Soil-B after glycerol treatment. The 1.01–1.03 nm peak and 1.25 nm peak of Mg saturated clay indicated presence of mica and mica/
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vermiculite interstratified minerals. These peaks were sharp in Soil-B whereas ill-defined or weak in Soil-C (Fig. 1). This may indicate higher degree vermiculitization of mica derived from granodiorite in Soil-C than Soil-B. Strong 1.01 nm peaks of K saturated clays from Soil-B and -C indicated that these soils contained vermiculite, and vermiculite/mica interstratified minerals with low degree of Al-interlayering as major clay minerals. The 0.50 nm peak observed in all the soils would be 002 reflection of mica. In K-saturated clay of Soil-B and Soil-C, the 0.36 nm peak was weaker than the 0.34 nm peak. However, in Mg-saturated clay, the intensity ratio of 0.36 nm peak to the 0.34 nm peak were higher than that of K-saturated clay. This would be the result of shift of 003 reflection of vermiculite by Mg-saturation. Short-range ordered minerals were not detected by chemical extraction (not shown).

Fig. 2 shows the adsorption isotherms of K for the soils. More adsorption was observed in Soil-A as compared to Soil-B and -C. The distribution coefficient (K_d, L/kg, slope of the linear adsorption isotherm) for Soil-B and -C were almost 1.7–1.8 times higher than Soil-A. Conversely, CEC values were highest in Soil-A and lowest in Soil-B overall pH conditions (Fig. 3). Consequently, CEC at the pH (CaCl_2) was also highest in Soil-A (Table 2). This could be resulted from higher clay and organic matter content of soil-A (Table 1). The pH-dependency of CEC of the soils was low suggesting negative charge of the soils was mainly composed of permanent charge (Fig. 3). Permanent charges have high affinity for K, particularly on micaceous minerals (mica, vermiculite, and their interstratified minerals) because of their high layer charge density and the presence of FES.

The FES shows remarkably high affinity for K due to the steric hindrance for large hydrated ions (e.g., Ca and Mg) and easiness for collapse. The vermiculitic nature of soils-B and -C would result in higher K_d values than soil-A.

Fig. 4 shows the 42K distributions in the soil column after the transport experiments. Although 30 mm of water was supplied, resulting increase in
water content almost up to the bottom of the column, the mobility of applied K was very low. Only a slight $^{42}$K was detected at 4 to 6-cm deep layer where $^{42}$K was initially applied to the surface 4-cm thick layer. $^{42}$K was not detected below 6-cm deep layer for all soils. This finding is supported by the results from the adsorption isotherms where the obtained K$_d$ ranging from 17.2 to 31.1 (L/kg) suggests retardation factors (R = $\rho_b$K$_d$/\theta, \theta: volumetric water content) for all soils ranging from around 30 to 60 when assuming the $\theta$ of 0.6 (cm$^3$/cm$^3$) for all soils. Therefore, the apparent moving distance of $^{42}$K is reduced to 1/30 to 1/60 of the water front due to average water flow. As shown in Fig. 4, around 75% of $^{42}$K retained in each 2-cm slice was exchangeable form for Soil-A, while marked $^{42}$K existed as a fixed form for Soil-C and more significantly Soil-B where around 60% of $^{42}$K was a fixed form for Soil-B. Again, the findings suggest the vermiculitic nature in Soil-B and -C contributed to the K fixation. It also suggests that special attentions to the K applications to reduce R Cs transfer to crops in such soils having vermiculitic nature. Additional countermeasures may be required, i.e., enhancing Ex-K by organic matter application to reduce K fixing ability of clays,$^{33}$ and intensive application of K to attain partial saturation of K fixing sites.$^{25}$ Cs fixing capacity, which is an indicator of vermiculitic nature of soil, was lowest in Soil-A (Table 2). This result was comparable to the less fixed form K in Soil-A as shown in Fig. 4. Conversely, Cs fixing capacity of Soil-B was lower than Soil-C, whereas K fixation in the column was more remarkable on Soil-B. Cs fixing capacity could have underestimated for soils having vermiculitic nature.$^{16}$ Moreover, degree of weathering of micaeous minerals (vermiculitization) may affect extent of K fixation by soils through changes in layer charge and capacity of FES.$^{19}$ Vermiculitization accompanies decrease in layer charge.$^{81}$ The FES capacity of mica first increase and then decrease during vermiculitization.$^{17}$ Again, the sharp peaks of mica and mica/vermiculite interstratified minerals in the XRD pattern clearly indicated lower degree of vermiculitization of micaeous minerals on Soil-B (Fig. 1). Lower clay content also implied the lower degree of vermiculitization on Soil-B (Table 1). Therefore, it is possible that lower degree of vermiculitization of Soil-B resulted in higher K fixation than soil-C. Since the soils selected in this study are limited, further investigations are required for characterizing

![Fig. 4  $^{42}$K distributions in the soil column.](image)
such vermiculitic soils and K and Cs behaviors in the soils.

4. Conclusions

The K behavior for non-vermiculitic and vermiculitic soils were investigated. For non-vermiculitic soils containing smectite and zeolite, batch experiments showed lower K adsorption but higher cation exchange capacity overall pH condition as compared to vermiculitic soils. In addition, column transport experiments for K showed that the retained K in soils exists as an ion exchangeable form. On the other hand, two different vermiculitic soils showed higher adsorption of K although they exhibited lower cation exchange capacity. The mobility of applied K for vermiculitic soils was similar to that for non-vermiculitic soil but larger portion of retained K was fixed form. Thus, this study suggested clay mineral composition, especially vermiculite, highly influence K adsorption and its form in soils. For effective K applications to mitigate RCs transfer to crop, further investigations about eg., RCs and K interaction (i.e., ion exchange), RCs adsorption and its mobility, and behaviors of both ions near roots, are required against soils with different clay mineral compositions.

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