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Effects of influent water quality on potassium leaching from paddy soil

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Abstract: Additional potassium (K) fertilization, a practice undertaken to reduce the absorption of radioactive cesium (Cs) by rice in paddy fields where radioactive Cs was deposited, is gradually being discontinued owing to cost issues. Understanding the dynamics of K in paddy fields and establishing a reliable and efficient K fertilization technique is necessary to continue reducing the absorption of radioactive Cs by paddy rice. Therefore, to help understand the factors that cause K leaching from paddy soils and the effect of K supply by irrigation water, we used a compact thumb-size column (diameter 2.1 cm × length 5 cm) and examined the K leaching from paddy soil following influents of different concentrations and cation composition ratios. The test soil (fine-grained ordinary lowland paddy soil) was collected from a paddy plot that received additional potassium application. The coefficients of variation during soil filling of the columns were less than 2 % on average and approximately 6 % for soil exchangeable K in the column tests, with sufficient reproducibility. The equilibrium content of soil exchangeable K formed after water percolation was determined by the concentration of K^+ and the composition ratio of major cations — K, Na, Ca, and Mg — in influent water. The potassium adsorption ratio (PAR) of influent water was a valid indicator of K leaching. The effects of ion species on K leaching were in the order $Mg^{2+} > Ca^{2+} > Na^+$ based on a 1 mmol L^{-1} change in their concentration and $Ca^{2+} > Mg^{2+} > Na^+$ based on a 10 % change in their concentration, with divalent cations having a greater effect and Na^+ having a smaller effect.

Key Words : potassium leaching, thumb-size column test, PAR (potassium adsorption ratio), irrigation water quality, paddy soil

1. Introduction

The accident at the Tokyo Electric Power Company Holdings' Fukushima Daiichi Nuclear Power Station caused by the Great East Japan Earthquake and subsequent tsunami in March 2011 resulted in the release of large amounts of radioactive materials. In farmland where absorption of radioactive materials by crops was a concern, an additional application of potassium (K) fertilizer effectively reduced radioactive Cs concentrations in rice plants (Fujimura et al., 2013) and supplemental K application was initiated in 2012. As a result, the quantity of brown rice exceeding the food standard for radioactive Cs content has greatly reduced since 2012, and a significant effect of additional K fertilization on the reduction has been confirmed. However, starting in 2015, trials began to discontinue additional K applications due to high expenditures, and since 2016, an increasing number of regions have gradually discontinued additional K applications (Fujimura, 2021). Therefore, establishing a reliable and efficient K fertilization technique is required for rice cultivation, and understanding the dynamics of K in the soil is a critical issue.

The migration of K in the soil is affected mainly by cation exchange and selective adsorption by some clay minerals. Cation exchange was identified in the late 1800s as a phenomenon in which bases of alkali salts, including K in soils, were exchanged and transferred primarily through the negative charge possessed by clay particles (Johnston et al., 2016). Thus, K migration is closely related to the dynamics of other major cations. The magnitude of the relative cation selectivity of clays is affected by the ionic radius and the amount of charge per ion, which is known experimentally to be in the order $Cs^+ > Rb^+ > K^+ \cong NH_4^+ > Na^+ > Li^+$ for the monovalent cations (Bolt et al., 1978).

Additionally, 2:1 type layered silicate minerals, such as illite and vermiculite, are highly selective for K^+, along with Cs^+, Rb^+, and NH_4^+ (Sawhney, 1972; Goulding, 1987), which also affects K mobility. These ions with small ionization and relatively large ionic radii are easily dehydrated and adsorbed as naked ions to be sandwiched

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between the unit layers of 2:1 type layered silicates (Wada, 1997). However, because soils are mostly mixtures of many clay minerals and contain many cations, it is generally difficult to describe the dynamics of ions in a rigorous theoretical or experimental manner (Evangelou and Phillips, 2005). Furthermore, the migration of K in the form of clay colloids and the ionic form has been pointed out (Brouder et al., 2021).

Some studies on the soils of upland fields have investigated the effect of the quality of irrigation water on soil K leaching (Kolahchi and Jalali, 2007; Jalali and Merrikhpour, 2007; Heidari and Jalali, 2016). However, studies on K leaching in paddy soils have been scarce until recently.

The content of exchangeable K in soil significantly affects the radioactive Cs concentration in brown rice. Therefore, among studies related to Fukushima, studies on K balance in paddy fields, including studies on K supply by irrigation water and leaching by percolation, have been conducted in recent years. Nishikiori et al. (2020) measured the K balance, including the K supply from irrigation water and K leaching to the subsurface, in a paddy field located on an alluvial river site with a considerable level of percolation. They found that K leaching was greater when there was excessive irrigation water.

In cooperation with five prefectural agricultural experiment stations in Japan, the National Agriculture and Food Research Organization (NARO) compiled guidelines (NARO, 2021) for applying K in paddy rice based on the K balance of paddy soils to promote appropriate K fertilization in paddy fields. According to the report, the presence or absence of return of rice straw has a decisive effect on the K balance. If the rice straw is returned, it almost compensates for the amount of K carried out by harvest; therefore, the K balance of the paddy field will not become negative due to fertilization and supply from irrigation water. Here, the amount of K supplied by the irrigation water was assumed to be approximately 15 kg ha^{-1} when the K^+ concentration in the irrigation water was 2.5 mg L^{-1}.

As mentioned above, the dynamics of K in agricultural lands, including the quantity of K supplied through water, has been a focus of research in recent years. It can be expected that differences in the quality of irrigation water or water infiltrating into the soil would affect K leaching. However, few studies have quantitatively evaluated the effect of water quality on the dynamics of K in paddy soils.

Therefore, in this study, we attempted to investigate the relationships between various influent water qualities and responses of K leaching by using compact thumb-size column experiments. We first examined the effects of different concentrations of major cations in the influent water on the leaching of K from paddy soils. Subsequently, we hypothesized that the K^+ concentration of influent and the composition ratio of the major cations would affect the leaching. The potassium adsorption ratio (PAR) was used as an indicator to test this hypothesis.

In addition, leaching tests were conducted using thumb-size columns. Column sizes used in similar studies varied; for example, 5 cm in diameter (D) × 20–50 cm in length (L) (Matsukawa and Nakano, 1992), 4.9 cm (D) × 14 cm (L) (Matsukawa et al., 1996), 14.6 cm (D) × 60 cm (L) (Li et al., 1994), 2 cm (D) × 15 cm (L) (Colombani et al., 2015), and 5 cm (D) × 20 cm (L) (Li et al., 2013). In recent years, the International Organization for Standardization (ISO) has promoted the international standardization of the "Up-flow percolation test" (ISO, 2019). This standard is primarily intended to evaluate the leaching of contaminants. However, inorganic ions such as Ca^{2+} and Cl^- were also evaluated by Yasutaka et al. (2017). In that standard, the column size is specified as D 5–10 cm and L 30 ± 5 cm. As described above, column diameters of approximately 5–10 cm and 15–30 cm lengths are often used. If the required results can be obtained, the smaller the column, the more labor-saving the experiment can be. In this study, we attempted to use a column with a compact size of 2.1 cm (D) × 5 cm (L) and examined the method's reproducibility.

2. Potassium adsorption ratio (PAR) and its characteristics

It is important to avoid sodium (Na) hazard that causes salt damage and reduces permeability due to soil particle dispersion in irrigated agriculture in arid and semi-arid regions. Therefore, the sodium adsorption ratio (SAR) has been defined and widely used as an indicator of the risk of irrigation water (USDA, 1954).

$$\mathrm{SAR} = \frac{[\mathrm{Na}^+]}{\sqrt{[\mathrm{Ca}^{2+}] + [\mathrm{Mg}^{2+}]}} \quad (1)$$

where $[Na^+]$, $[Ca^{2+}]$, and $[Mg^{2+}]$ are the concentrations of each ion (mol L^{-1}). Using irrigation water with high SAR promotes the leaching of Ca^{2+}, Mg^{2+}, K^+, and phosphorus from the soil and affects groundwater quality (Jalali and Merrikhpour, 2007). Sposito and Mattigod (1977) examined the theoretical chemical basis for the potential SAR of water in exchange equilibrium with soil and found that the Gapon cation exchange equilibrium constant is the chemical basis for the SAR.

Similar to Na^+, K^+, one of the major monovalent cations, can cause harmful effects when presentin excess;

however, its effects are weaker than that of Na^+. Therefore, as an indicator, PAR was defined similarly to SAR, with Na^+ replaced by K^+ (USDA, 1954).

$$\mathrm{PAR} = \frac{[K^+]}{\sqrt{[Ca^{2+}] + [Mg^{2+}]}} \quad (2)$$

The characteristics of this equation as an indicator of the effect of K in irrigation water are that it ignores the effect of Na^+, does not make a chemical distinction between Ca^{2+} and Mg^{2+}, and reflects the effect of concentration as well as composition ratio. Assuming a solution with a constant composition ratio of cations, PAR is proportional to the square root of the $[K^+]$.

The PAR of the soil solution reflects the amount of K adsorbed on the exchange surface of clay and soil organic matter, which may be an explanatory factor for aggregation and dispersion within the soil. Shainberg et al. (1987) studied the effect of PAR, among other indicators, on the decrease in hydraulic conductivity. The PAR has also been examined as a factor explaining its effectiveness as a water quality indicator for assessing the irrigation history of agricultural land and groundwater environments. Smiles and Smith (2004) examined the PAR (called KAR in the paper) of soil solutions in agricultural land irrigated with piggery effluent and showed that PAR is a good indicator of the effect of irrigation with piggery effluent. Griffioen (2001) investigated the effect of fertilizer application to agricultural land on groundwater quality and showed that the behavior of K is explained well using PAR at near neutral pH, where the major cations in the groundwater are divalent.

Parameters similar to SAR and PAR are found in many theoretical ion exchange equations (Bresler et al., 1982). Unlike those parameters, however, SAR and PAR are expressed in terms of ion concentration, not ion activity. Therefore, they do not consider the reduction of free ions due to the formation of complexes. In a case study using the K adsorption ratio of soil solution and considering the ionic activity, Ramamoorthy and Paliwal (1965) studied the availability of soil K when applied to paddy rice grown in 40 soils with different properties in India. They found a correlation between the potassium adsorption ratio and crop response in many districts, considering the ionic activity.

A parameter similar to PAR was used in the study of the Q/I relationship. The Q/I relationship quantifies the relationship between the K intensity factor (factor I) and K absorption/release by the soil (factor Q), using factor I as the activity ratio of ion pairs of K^+ and $Ca^{2+} + Mg^{2+}$, i.e., $a_{K^+}/\sqrt{(a_{Ca^{2+}} + a_{Mg^{2+}})}$ (Beckett, 1964a). Specifically, the Q/I relationship examines the change in soil potassium absorption relative to the factor I of the solution by reacting a 0.01 M $CaCl_2$ solution with several levels of K concentration and soil at a constant solid-liquid ratio and for a constant time (Eguchi et al., 2017). If the activity ratio of factor I can be regarded as proportional to the concentration ratio, it is isomorphic to PAR when replaced by the concentration ratio $[K^+]/\sqrt{([Ca^{2+}] + [Mg^{2+}])}$ (Evangelou et al., 1986).

Using the concentration of K^+ in the influent and the composition of K in the major cations as multiple explanatory variables may easily explain the K leaching. However, in this study, we would like to reduce the number of explanatory variables and concentrate on PAR as a simpler indicator to grasp the phenomenon and explain the leaching and accumulation of soil exchangeable K.

3. Materials and methods

3.1 Test soil

The test soil samples were collected from paddy field soil in Matsukawa, Fukushima City, Fukushima Prefecture, and passed through a 2 mm diameter sieve after air-drying. The soil is classified as fine-grained ordinary lowland paddy soil (NARO, 2019). The plot where the test soil was taken had been fertilized with additional K to reduce the absorption of radioactive Cs, and the soil was sampled early in the season after rice planting. In addition, rice straw return was practiced in the plot. The parent material of the soil is tuffaceous sandstone and siltstone, called the Matsukawa Formation, a late Miocene or early Pliocene terrestrial-water stratified sediment (Yoshida et al., 1983; Sakaguchi, 1995). The distribution area of the Matsukawa Formation is surrounded by granodiorite zones formed during the Paleogene, which influences its characteristics. The physicochemical properties of the test soil and its mineral composition are shown in Table 1. The mineral composition of the test soil is rich in kaolinite and contains some 2:1 type clay minerals such as smectite, vermiculite, and illite.

3.2 Overview of experimental methods

Three different experiments were conducted in this study. This section focuses on matters common to all three experiments, and the specifics of each experiment are described in Section 3.3.

The apparatus used in the experiments is shown in Fig. 1. A small thumb-sized soil column (2.1 cm in diameter, 5.0 cm in length, and 17.3 cm^3 in volume) was used for the soil column tests. The cylindrical acrylic column was carefully filled to ensure uniform filling of the test soil. The difference in mass between the empty and filled column was measured during filling, and the dry bulk density was calculated. The dry bulk density value was determined

Table 1 Physical and chemical properties of the test soil.

Attribute	Description	
pH (H_2O)	6.0	Soil-water extracts at 1:5
EC	0.09 (dS m^{-1})	Soil-water extracts at 1:5
CEC	17.6 ($cmol_c$ kg^{-1})	Shaking extraction method (10 % KCl)
Exchangeable Ca (CaO)	2.76 (g kg^{-1})	Shaking extraction method (1M CH_3COONH_4)
Exchangeable Mg (MgO)	0.50 (g kg^{-1})	Shaking extraction method (1M CH_3COONH_4)
Exchangeable K (K_2O)	0.40 (g kg^{-1})	Simplified method by using a compact K meter
Phosphate absorption coefficient (P_2O_5)	0.54 (g kg^{-1})	Orthophosphoric acid method
Humus	0.019 (kg kg^{-1})	Kumada's simplified method
Soil texture	CL	Coarse sand: 26 %, fine sand: 18 %, silt: 33 %, clay: 23 %
Clay minerals (By XRD)	Abundant	Kaolinite
	Common	Smectite, vermiculite, illite, chlorite-vermiculite
	Trace	Chlorite
	None	Halloysite

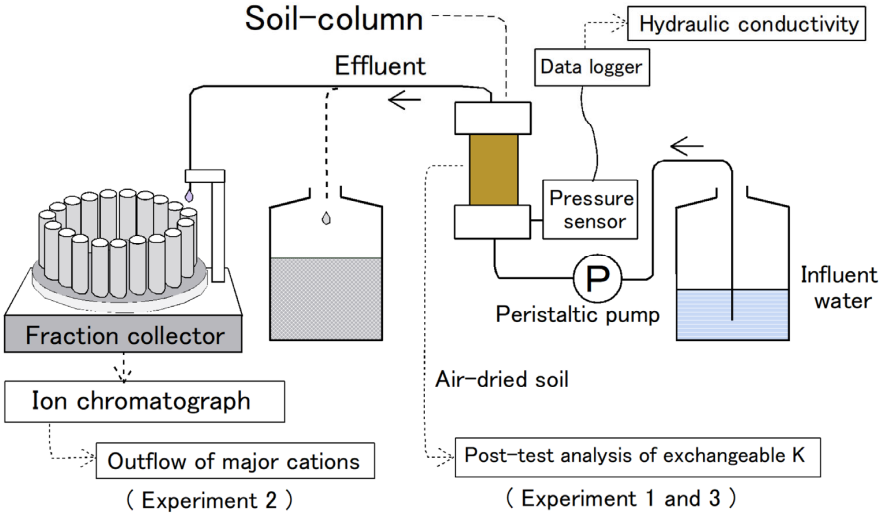


Fig. 1 Schematic of apparatus.

as approximately 1.12 g cm^{-3} using the air-dried soil, the variation of which is discussed in Section 4.1.

The quality of the influent water used for the column tests was determined based on the concentration of major cations — K, Na, Ca, and Mg — in the irrigation water of the plot from which the test soils were collected (Table 2). The influent water was prepared by dissolving reagents (KCl, NaCl, $CaCl_2$, and $MgSO_4$) in deionized water as needed to achieve the required concentrations. The specific concentration of each ion in the influent in each experiment is described in Section 3.3.

A peristaltic pump (MP-2010, Tokyo Rikakikai, Tokyo, Japan) was used to supply influent water to the soil column. First, before the start of the test, the influent water was slowly fed from the bottom of the column to saturate the soil, and the water supply was temporarily stopped until the start of the test. This may have caused air to be entrapped, but its effect was not considered. The water flow

Table 2 Cation concentration and PAR of irrigation water in test soil taken field during irrigation period in 2020.

	Cation concentration (mmol L^{-1})				PAR
	K^+	Na^+	Ca^{2+}	Mg^{2+}	
Mean*	0.036	0.19	0.29	0.13	0.055
Max.	0.052	0.31	0.37	0.21	0.079
Min.	0.023	0.095	0.23	0.091	0.037

* Samples were taken 15 times between May and August.

test was then started at a set flow rate (0.35 mL min^{-1}) and time. In the test, the water was passed through the column at a constant flow rate from the bottom to the top of the column. The tests were conducted in a laboratory at a room temperature of 21–25 °C.

The effluent from the column was collected in a polyethylene container (Experiments 1 and 3) or fraction collector (Experiment 2), and the concentrations of major cations were determined using an ion chromatography system (ICS-1500, Thermo Fisher Scientific, Waltham, MA, USA). However, this paper does not cover the analytical results of the samples in Experiments 1 and 3.

In some tests, the water pressure at the bottom of the column was measured with a pressure sensor (HTV-200KP, HTV-400KP, Sensez, Tokyo, Japan), and the saturated hydraulic conductivity K_s cm s^{-1} was calculated using the following equation

$$K_s = \frac{q}{A} \times \frac{L}{\Delta H} \times 0.0981 \quad (3)$$

where q cm^3 s^{-1} is the flow rate at the time of pressure measurement, A cm^2 is the cross-sectional area of the column, L cm is the column length, and ΔH kPa is the pressure difference between the upper and lower ends of the column. In the experiment, the water pressure at the upper end of the column was assumed to be negligibly small compared to the water pressure at the lower end, and ΔH was assumed to be equal to the pressure at the bottom of the column.

After the test, the column was dismantled, the mass of the wet soil was measured, and the volumetric water content was calculated from the wet mass of the soil and the mass of the sample when the column was filled. Since the mass of air-dried soil was used to calculate volumetric water content, there may have been a slight underestimation. After the test, the soil was air-dried and then measured for soil exchangeable K. The exchangeable K was analyzed in triplicate using a simple analytical method (Nakayama, 2013; Nakayama et al., 2018) for the exchangeable K content of soil using a compact K ion meter (LAQUA twin K-11, Horiba, Kyoto, Japan) as shown in Fig. 2.

Furthermore, to examine the reproducibility of this test using small columns, the coefficients of variation of dry bulk density and volumetric water content in Experiments 1 and 3 and water flow volume and soil exchangeable K in Experiment 3 were examined.

3.3 Specific experimental conditions

3.3.1 Experiment 1: Effect of water quality and flow volume on K leaching

Experiment 1 was designed to investigate the response of K leaching to changes in influent water quality and flow volume. The qualities of the influent water (Table 3) were set as standard water (hereinafter referred to as "1×"), low-concentration water (hereinafter referred to as "0.3×"), high-concentration water (hereinafter referred to as "3×"), and deionized water (hereinafter referred to as "DI"). The water quality of the standard water (1×) was determined with reference to the average of actual irrigation water quality. However, the range of variation of values ($0.3\times - 3\times$) was set wider than the actual range of variation in order to examine the response of K leaching to changes in influent water quality. The flow volumes were set in eight steps ranging from 120 to 7500 mm, with a flow rate of 0.35 mL min^{-1} and a flow time of approximately 2–120 h. Since the sampling plots had relatively low infiltration rates and assuming that the water flow volume for one irrigation season of paddy rice is 1200 mm, the flow volumes set in the experiment are roughly equivalent to 0.1–6 years of the flow volume. After each test, the soil was air-dried and analyzed for soil exchangeable K. Furthermore, in Experiment 1, there was basically no repetition of the same experimental conditions.

3.3.2 Experiment 2: Characteristics of major cations leaching from soil columns

In Experiment 2, we investigated changes in the effluent of major cations from the soil columns over time by collecting effluent at predetermined time intervals and an-

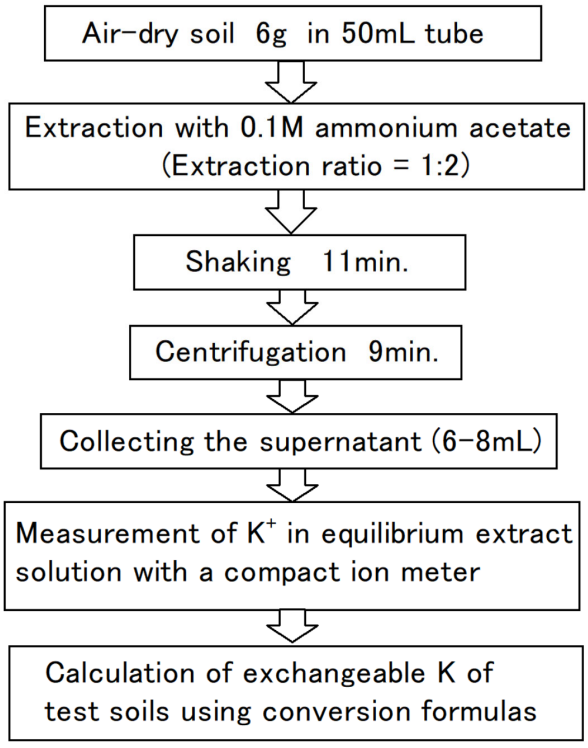


Fig. 2 Procedures for measuring soil exchangeable K using a compact ion meter (Nakayama, 2013; Nakayama et al., 2018 with some modification).

Table 3 Cation concentration of influent water in Experiments 1 and 2.

Item	Description	Cation concentration (mmol L^{-1})				PAR
		K^+	Na^+	Ca^{2+}	Mg^{2+}	
1×	Standard water	0.031	0.17	0.35	0.15	0.044
0.3×	Varying 4 cations simultaneously	0.0094	0.051	0.11	0.045	0.024
3×		0.094	0.51	1.1	0.45	0.077
DI	Deionized water	0.00	0.0	0.0	0.0	–

Table 4 Cation concentration and PAR of influent water and number of test repetitions in Experiment 3.

Description	Concentration factor	Cation concentration (mmol L^{-1})				PAR	Repetition
		K^+	Na^+	Ca^{2+}	Mg^{2+}		
Standard water	1×	0.031	0.17	0.35	0.15	0.044	4
Varying 4 cations simultaneously	0.3×	0.0094	0.051	0.11	0.045	0.024	3
	3×	0.094	0.51	1.1	0.45	0.077	3
	10×	0.31	1.7	3.5	1.5	0.14	3
Varying only K^+ conc.	0.3×	0.0094	0.17	0.35	0.15	0.013	3
	3×	0.094	0.17	0.35	0.15	0.13	3
	10×	0.31	0.17	0.35	0.15	0.44	3
Varying only Na^+ conc.	0.3×	0.031	0.051	0.35	0.15	0.044	3
	3×	0.031	0.51	0.35	0.15	0.044	3
	10×	0.031	1.7	0.35	0.15	0.044	3
Varying only Ca^{2+} conc.	0.3×	0.031	0.17	0.11	0.15	0.061	3
	3×	0.031	0.17	1.1	0.15	0.029	3
	10×	0.031	0.17	3.5	0.15	0.016	3
Varying only Mg^{2+} conc.	0.3×	0.031	0.17	0.35	0.045	0.050	3
	3×	0.031	0.17	0.35	0.45	0.035	3
	10×	0.031	0.17	0.35	1.5	0.023	3
Varying Ca^{2+} and Mg^{2+} conc.	0.3×	0.031	0.17	0.11	0.045	0.081	2
	3×	0.031	0.17	1.1	0.45	0.026	1
	10×	0.031	0.17	3.5	1.5	0.014	2

alyzing the water quality. The influent and flow rates were the same as in Experiment 1. The flow time was set to 72 h (the set flow volume was approximately 4600 mm), and the water was collected 22 times between 0.2 h and 71.9 h using a fraction collector (Model 2110, Bio-Rad, Hercules, CA, USA). The collected water was measured for pH (pH-22, Horiba, Kyoto, Japan) and analyzed for major cations. There were no repetitions in Experiment 2.

3.3.3 Experiment 3: Effects of different cation composition ratios in the influent on K leaching

In Experiment 3, the effects of different cation composition ratios in the influent on K leaching and accumulation were investigated. There were 19 patterns of influent quality settings (Table 4), and the tests were conducted in triplicate with some exceptions. The quality of the influent water consisted of standard water (1×) and its variations (0.3×, 3×, and 10×), variable concentrations (0.3×, 3×, and 10×) for each of the four major cations, and a combination of two cations (Ca^{2+}, Mg^{2+}).

These tests were conducted assuming a water flow volume of 1200 mm and a flow time of 20 h. After the tests, the soil samples were air-dried and analyzed for soil exchangeable K. In some of the tests, settings and results were shared with Experiment 1.

4. Results and discussion

4.1 Reproducibility of compact column tests

The advantage of using a smaller column is that a smaller amount of test soil to be prepared. Therefore, even when designing tests that include repetitions under various conditions, as in this study, the total amount of sample required is small. Correspondingly, the labor required to treat the sample before and after the test and the amount

Table 5 Dry bulk density, volumetric water content, and their coefficients of variation (CV) of soil columns in Experiments 1 and 3.

	Dry bulk density* ($g\ cm^{-3}$)		Volumetric water content** ($m^3\ m^{-3}$)		n***
	Mean	CV (%)	Mean	CV (%)	
Experiment 1	1.12	1.6	0.562	6.8	45
Experiment 3	1.13	1.8	0.595	1.7	64

* Dry bulk density might be slightly overestimated because the mass of air-dried soil was used in the calculation.
** Volumetric water content was measured after dismantling the column.
** Volumetric water content might be slightly underestimated because the mass of air-dried soil was used in the calculation.
*** Experiments 1 and 3 share some tests.

Table 6 Fluctuation of flow volume and soil exchangeable K content in column tests in Experiment 3.

Flow volume (n = 64) (mm)			CV (%)	Soil exchangeable K* CV (%)		
Mean	Min.	Max.		Mean	Min.	Max.
1241	1145	1339	3.2	5.8	0.9	11.8

*17 tests of three repetitions each.

of water needed are also small. In contrast, because the column is small, lower reproducibility of the test, which may be reduced by, for example, non-uniform sample filling and its effects, is a concern. Therefore, in this section, we discuss the reproducibility of the sampling method.

In total, 96 tests were conducted in the three different experiments. Using some of these data, Table 5 shows the properties and the coefficients of variation (CV) of filling the columns. The dry densities of Experiments 1 and 3 were similar, and their CVs were small, indicating good soil filling reproducibility. In contrast, there were differences in volumetric water content, and the CV was slightly larger in Experiment 1.

Table 6 shows the range and CV of measured flow volume and the CVs of the experimentally obtained soil exchangeable K values. In this study, the flow rate was controlled by a peristaltic pump, and the flow time was determined. Therefore, the flow volume varied even under the same settings owing to the fluctuation of the actual flow rates. In Experiment 3, a flow volume of 1200 mm was assumed, but the actual flow volume was 1145–1339 mm. The variation in values of soil exchangeable K content reflects not only the direct variation of factors due to the analytical method, such as the variation caused by the procedure of the simplified analytical method but also the effect of differences in the flow volume. The values of exchangeable K obtained in this experiment had an average CV of about 6 % and were sufficiently reproducible when the range of values was considered.

4.2 Effect of influent water quality and flow volume on K leaching (Experiment 1)

The results of Experiment 1 are shown in Fig. 3, indicating the effect of influent water quality and flow volume on exchangeable K of test soil. The difference between the initial value of soil exchangeable K and the value of exchangeable K at any given flow volume indicates the amount of K leaching by the flow volume. K leaching increased as the flow volume increased, and those differences in influent water quality significantly affected the K leaching. The order of leaching rates was $3\times > 1\times > 0.3\times >$ DI until the flow volume was about 500 mm; i.e., the leaching proceeded in the order of increasing concentration, after which the K leaching rate gradually decreased with increasing flow volume. If DI is excluded, the soil exchangeable K contents almost reached equilibrium in the order of increasing concentration of influent. The equilibrium exchangeable K contents were 0.17 and 0.07 g $K_2O\ kg^{-1}$ for $3\times$ and $0.3\times$ of influent water quality, respectively, a difference of approximately 0.1 g $K_2O\ kg^{-1}$.

With $0.3\times$, $1\times$, and $3\times$ influent water quality, soil exchangeable K increased slightly with increasing flow volume after the K leaching stopped. In the case of DI, the exchangeable K did not reach equilibrium until approximately 7400 mm, and the content of exchangeable K decreased gradually due to leaching.

The average pH values of the influent were 5.8, 5.6, 5.6, and 5.6 for DI, $0.3\times$, $1\times$, and $3\times$, respectively. The pH values of the effluent increased to 6.9, 6.6, 6.5, and 6.3, respectively, after more than 100 h of water flow (a flow volume of approximately 6000 mm or more). This increase

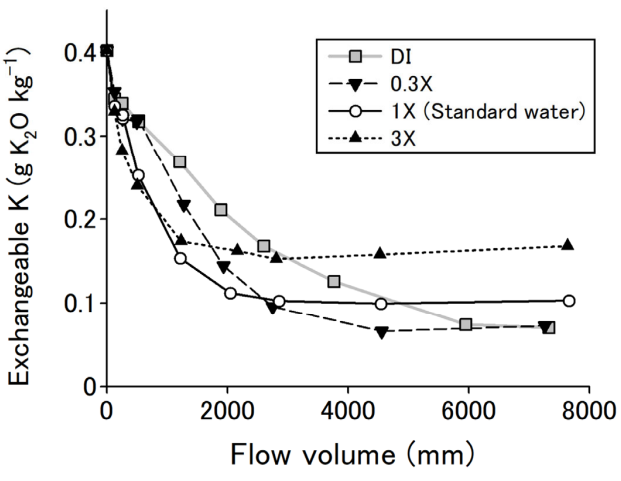


Fig. 3 Effect of influent water quality and flow volume on soil exchangeable K (Experiment 1).

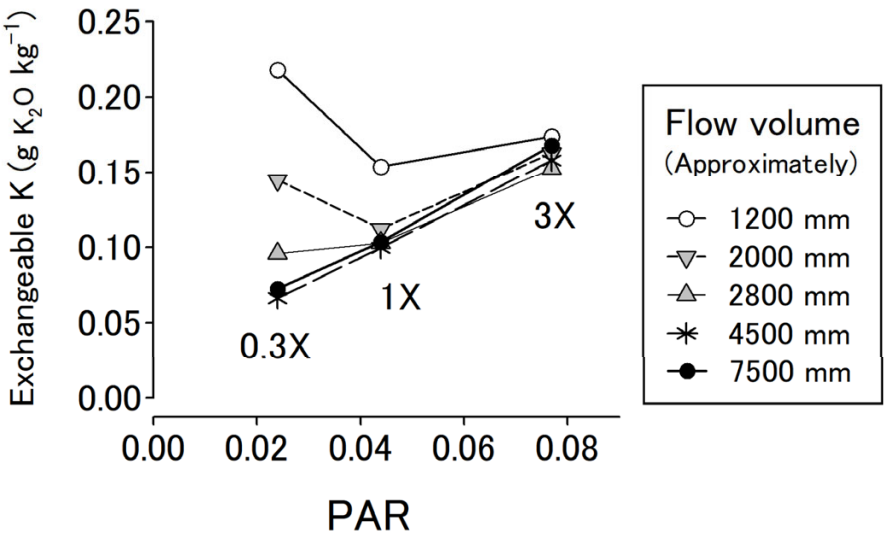


Fig. 4 Relationship between PAR in influent and soil exchangeable K with different concentrations in the same ion composition ratio (Experiment 1).

in pH during the percolation process suggests that H^+ contributes to cation exchange. In fact, the gradual progression of leaching in tests using DI is expected to include the effect of cation exchange by H^+.

The water pressures at the bottom of the column during the tests ranged from approximately 26–70 kPa, and the saturated hydraulic conductivities were generally in the range of 1.2–3.3 $\times$ 10^{-5} cm s^{-1}. However, in some cases, the hydraulic conductivity gradually decreased, and in tests in which large amounts of DI or 0.3$\times$ were passed through the column, it eventually dropped by about one order of magnitude. The pH of the effluent was higher in those cases, and the increase in pH and decrease in soil exchangeable cations might have affected the decrease in hydraulic conductivity, i.e., the dispersion of clay colloid particles and their migration, resulting in soil pore clogging. Shainberg et al. (1987) passed several dilute solutions, including distilled water, through a smectite-sand mixture and evaluated clay swelling and dispersion based on changes in permeability and the concentration of clay in the effluent. The results showed that the permeability decreased with lower concentrations of experimental water in tests with Polkville smectite (Mississippi) and SAR less than 10, with distilled water having the lowest permeability. In our study, in some of the tests in which the decrease in hydraulic conductivity was significant, the turbidity of the effluent was large, suggesting the dispersion and migration of clay colloidal particles. These details will be reported at another occasion.

Fig. 4 shows a rearrangement of the data of Fig. 3, excluding DI, in relation to the PAR of influent and soil exchangeable K. The PAR increased in response to increases in the concentration of the influent. The exchangeable K contents at equilibrium with flow volumes greater than approximately 4500 mm were correlated with PAR ($r = 0.995$; $p \leq 0.01$; $n = 6$). This means that the soil exchangeable K will reach equilibrium after approximately 4 years of infiltration, assuming 1200 mm of infiltration per irrigation season.

This result is consistent with the relationship between factor I and factor Q observed in the Q/I relationship. While the Q/I relationship indicates the relationship between factors I and Q in the equilibrium state of soil solution and soil in batch tests (Beckett 1964b; Okajima and Matsunaka 1972), the present study looks at the relationship between PAR (contrasted with factor I) and the K leaching (contrasted with factor Q) at equilibrium, which emerges from continuous percolation of influent.

4.3 Characteristics of leaching of major cations from the soil column (Experiment 2)

Fig. 5 shows the relationship between flow volume and concentrations of major cations in the effluent from soil columns for different influent water qualities. The final flow volume was approximately 4600 mm for 0.3$\times$, 1$\times$, and 3$\times$, and approximately 3900 mm for DI. In the DI test, the permeability of the soil column decreased during the test, and the flow rate decreased, resulting in approximately 15 % less flow volume than in the other tests.

The concentrations of the major cations in the effluent gradually decreased immediately after the start of the experiment. For many of the cations, the decrease in concentration stopped, and equilibrium was reached. A comparison of the concentrations for each ion species regarding the rapidity of stabilization of concentrations based on the water flow volume from the start of the experiment until

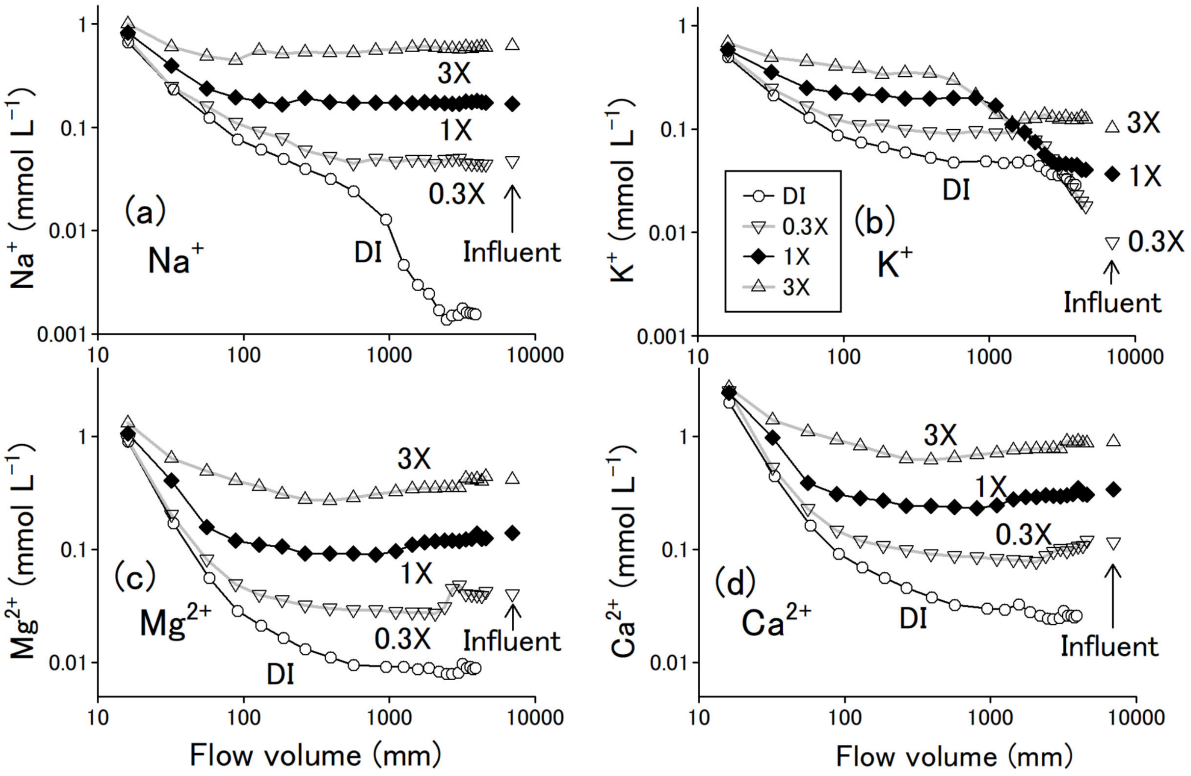


Fig. 5 Leaching of major cations from soil columns by influent water with different water quality, (a) Na^+, (b) K^+, (c) Mg^{2+}, and (d) Ca^{2+}. The vertical axis shows the concentration of each ion in effluent from soil columns (Experiment 2).

equilibrium was reached showed that the solutions stabilized faster in the order $3\times > 1\times > 0.3\times >$ DI. Also, when comparing ion species, the order of stability was faster for $Na^+ > Mg^{2+} = Ca^{2+} > K^+$ when $3\times$, $1\times$, and $0.3\times$ influents were used. For three influent types (excluding DI), the concentrations of Mg^{2+} and Ca^{2+} had decreased once to a level below those of the influent water and then gradually approached those of the influent water. When DI was used, the concentration of Na^+ showed a tendency to decrease further when the flow volume exceeded 1000 mm. Similarly, the concentration of K^+ in the effluent tended to decrease further at a flow volume of around 1000 mm for all water quality influents. On the other hand, no such two-step decrease in concentration was observed for the remaining Ca^{2+} and Mg^{2+} samples.

Fig. 5 shows that the K^+ concentration in the effluent gradually approached the K^+ concentration in the influent as the flow volume increased. However, equilibrium was not reached even after approximately 3900 mm of flow volume in DI and 4600 mm in $0.3\times$. The concentration in the effluent remained slightly higher than that in the influent at $1\times$ and $3\times$, indicating that K^+ had the slowest leaching rate of the four cations. The concentrations of the other three cations in the effluent approached those in the influent at a flow volume of a few hundred millimeters at the earliest and a thousand millimeters at the latest. Conversely, in the case of K^+, the concentration in the effluent showed a slight deviation from the K^+ concentration of the influent, even after about 4600 mm of flow volume. This may be explained by the effect of selective adsorption of K^+ by 2:1 type layered silicate minerals such as illite, vermiculite, and smectite in the test soil.

Many previous studies have investigated the mobility of exchangeable cations in soil. For example, Kozanecka et al. (2009) investigated the long-term mobility of exchangeable Ca^{2+}, Mg^{2+}, K^+, and Na^+ cations in soil cross sections to a depth of 1 m in an 85-year upland field with fixed mono-fertilization in Skiernievice, Poland. The mobility of exchangeable cations was $Ca^{2+} > K^+ > Mg^{2+} > Na^+$, indicating that the mobility of exchangeable cations differs greatly depending on the soil and other conditions.

4.4 Effect of different cation composition ratios on K leaching (Experiment 3)

Experiment 1 showed that when the concentration of influent with the same cation ratio was increased, the soil exchangeable K content equilibrated at a higher value. Therefore, the leaching of K was investigated in Experiment 3 by fixing the flow volume at approximately 1200 mm and varying the composition ratio of the major cations in the influent.

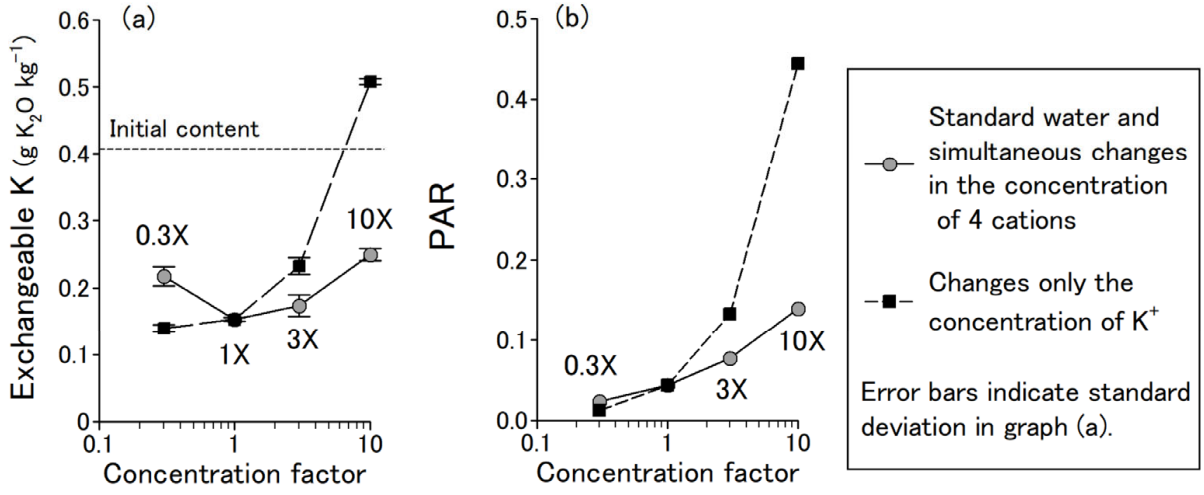


Fig. 6 Soil exchangeable K and PAR when only total major cations or K concentration in the influent is varied, (a) relationship between exchangeable K of test soil and concentration factor with three repetitions, (b) relationship between PAR and concentration factor (Experiment 3).

Fig. 6 shows a plot of soil exchangeable K content (Fig. 6 (a)), and PAR of the influent (Fig. 6 (b)) when only the K^+ concentration in the influent was varied and other cation concentrations were fixed, and when the overall concentration of major cations was varied (as in Experiment 1) as a control. When only the K^+ concentration in the influent was increased, the exchangeable K content was higher than in the control, demonstrating that the difference in cation composition ratio affected the K leaching. When only the K^+ concentration of influent was increased by a factor of 10, K accumulation occurred beyond the initial soil exchangeable K content.

The shapes of the graphs in Fig. 6 (a) and Fig. 6 (b) are alike. Comparing both graphs, soil exchangeable K was higher at 0.3× than at 1×. As can be seen from Fig. 3, this may be because the leaching rate was still high and far from equilibrium at the stage when approximately 1200 mm of the 0.3× influent had passed through the column.

Similarly, Fig. 7 (a) shows the soil exchangeable K content when the concentrations of the major cations, Na^+, Mg^{2+}, and Ca^{2+}, were increased or decreased individually or when the concentrations of Ca^{2+} and Mg^{2+} were increased or decreased simultaneously. Fig. 7 (b) shows a plot of the PAR of the influent. Interestingly, the figure in Fig. 7 (a) is quite similar to that in Fig. 7 (b). Since Na^+ is not considered in equation 2, there is no change in the value of PAR with changes in Na^+ concentration in Fig. 7 (b). The K leaching was almost the same in the influent with 3× and 1× of Na^+ but was higher in the influent with 10× of Na^+. This indicates that higher concentrations of Na^+ enhanced the K leaching.

Next, we attempted to quantitatively compare the effects of changes in each cation in the influent on the leaching and accumulation of K, using some of the results of Experiment 3. Therefore, we first calculated the sensitivity $S1^i_x$ (where x is the concentration multiplier) of the change in soil exchangeable K when the concentration of cation i changes by 1 mmol L^{-1} using the following formulas:

$$S1^i_{3X} = \frac{exK^i_{3X} - exK^i_{1X}}{C^i_{3X} - CK^i_{1X}} \tag{4}$$

$$S1^i_{10X} = \frac{exK^i_{10X} - exK^i_{1X}}{C^i_{10X} - CK^i_{1X}} \tag{5}$$

where $exK^i_x$is soil exchangeable K mg K_2O kg^{-1} and C^i_x is the influent concentration mmol L^{-1}. The results show that the increase in K^+ concentration in the influent contributes significantly to the maintenance and accumulation of soil exchangeable K (Table 7). The magnitude of the effect of a 1 mmol L^{-1} change in the concentration of each cation in the influent was found to be in the order of $S1^i_{3X}$ for Mg^{2+} >Ca^{2+} >Na^+. Furthermore, the sensitivity $S2^i_x$ of the change in soil exchangeable K for a 10 % change in the concentration of cation i based on the actual cation composition of the irrigation water (Table 2) was calculated using the following equations:

$$S2^i_{3X} = \frac{S1^i_{3X}}{0.1 \cdot C^i_{1X}} \tag{6}$$

$$S2^i_{10X} = \frac{S1^i_{10X}}{0.1 \cdot C^i_{1X}} \tag{7}$$

The magnitude of the effect of a 10 % change in the

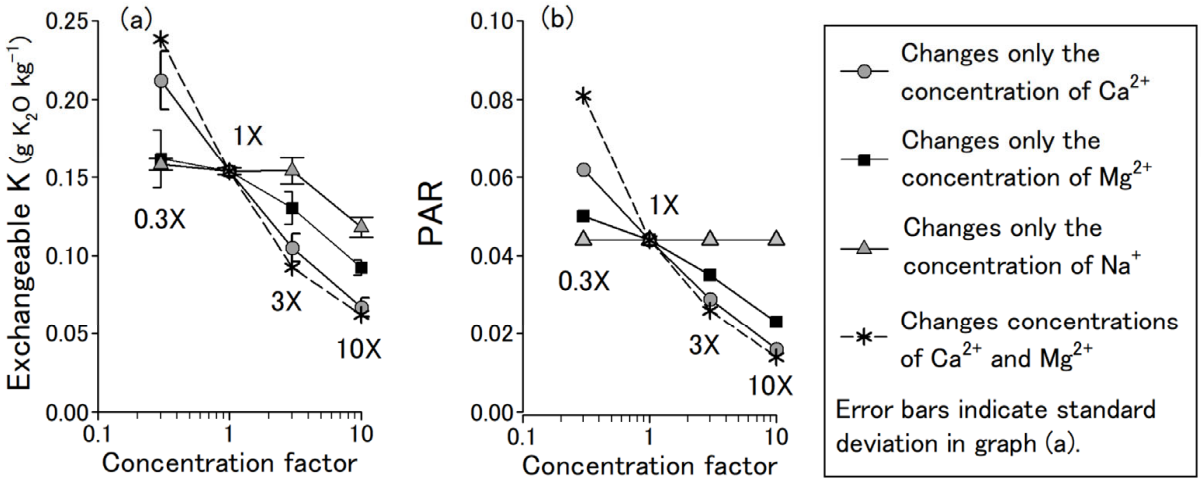


Fig. 7 Soil exchangeable K and PAR for different proportions of cations in the influent water, (a) relationship between exchangeable K of test soil and concentration factor (three repetitions with some exceptions), (b) relationship between PAR and concentration factor (Experiment 3).

Table 7 Effect of ionic species of influent water on K accumulation/leaching.

	Change in soil exchangeable K with 1 mmol L^{-1} change in concentration of each cation		Change in soil exchangeable K with 10 % change in concentration of each cation	
i	$S1^i_{3X}$	$S1^i_{10X}$	$S2^i_{3X}$	$S2^i_{10X}$
K^+	1250	1253	3.9	3.9
Na^+	1.2	−23	0.020	−0.40
Mg^{2+}	−77	−45	−1.2	−0.68
Ca^{2+}	−69	−27	−2.4	−0.96
Unit	mg K_2O kg^{-1}		mg K_2O kg^{-1}	

Note: Positive and negative values indicate accumulation and leaching, respectively.

concentration of each ion, assuming the actual cation composition of irrigation water, was $Ca^{2+} > Mg^{2+} > Na^+$ in $S2^i_{3X}$. The effect on soil exchangeable K of a 10 % increase in K^+ concentration is estimated to be equivalent to about a 16 % decrease in Ca^{2+} concentration (from $S2^{K^+}_{3X}/S2^{Ca^{2+}}_{3X}$). In all cases, the contribution of Na^+ to K leaching was small. This indicates that the exchange selectivity of Na^+ for cation exchange in the test soils is relatively low compared to Mg^{2+} and Ca^{2+}, supporting the absence of Na^+ in equation 2.

Based on the above, since there is a close relationship between K leaching and PAR of the influent, the relationship between PAR and the rate of change in soil exchangeable K and the relationship with K^+ for comparison, are shown in Fig. 8. The rate of change in soil exchangeable K (R_c) was calculated by the following equation:

$$R_c = \frac{(Exchangeable\ K\ after\ test)}{(Exchangeable\ K\ before\ test)} \times 100 \quad (\%) \quad (8)$$

The relationship between K^+ concentration and R_c (Fig. 8 (a)) showed a pattern, but K^+ concentration is not highly representative of K leaching. Conversely, except for a few data points, the relationship between PAR and R_c (Fig. 8 (b)) was almost plotted on a single curve, confirming that PAR has favorable indicator properties for R_c. The excluded points are the results of two experimental parameters, one with 0.3× of standard water and the other with 0.3× of K^+. All three repetitions for each parameter had the same trend, and the CVs were small. As seen in Fig. 3, the initial leaching was slow under low-concentration conditions, such as the excluded points. We presumed that this was affected by the fact that K was still in the mid-leaching stage at about 1200 mm of flow volume (Fig. 4).

The obtained regression curve indicated that soil exchangeable K accumulates when using influent with PAR greater than 0.3. The exchangeable K contents were estimated to be 0.161, 0.195, and 0.128 g K_2O kg^{-1}, respectively, for the mean, maximum, and minimum values of irrigation water quality at the sampling site (Table 2). This

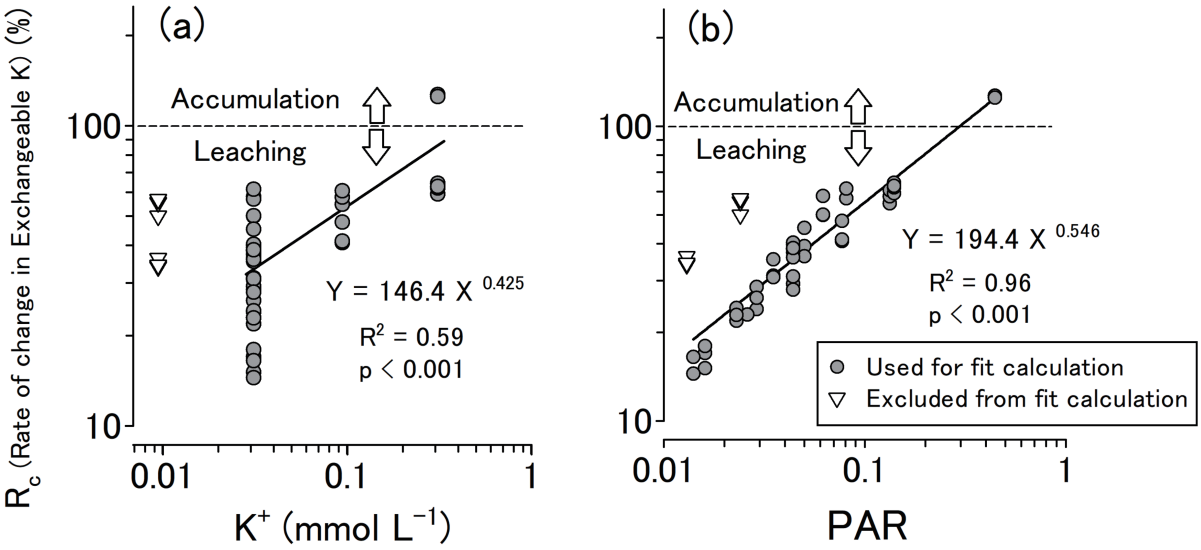


Fig. 8 K^+ and PAR as indicators of the rate of change in soil exchangeable K (Rc), (a) relationship between K^+ and Rc, (b) relationship between PAR and Rc (Experiment 3).

assumed an infiltration rate of 1200 mm and negligible effects of rainfall and fertilization.

Thus, the PAR of the influent was found to be a valid indicator that reflected the leaching and accumulation of soil exchangeable K.

4.5 Limitations of this study and future issues

This study has potential limitations. Firstly, differences in the initial values of the exchangeable K content of the test soil could significantly affect the results; the rate of change in exchangeable K used in Fig. 8 is based on the initial exchangeable K content of the test soil, and thus will have direct effects. Differences in experimental conditions, such as the flow rate or pH of the influent, might also affect the results.

Secondly, this study only examined one of the paddy soil types. It will be interesting to observe the patterns with different soil types, such as volcanic ash and peat soils. However, the exchangeable K content at equilibrium may be predicted from previous studies of the Q/I relationship. Eguchi et al. (2017) used the Q/I relationship to study the effect of clay minerals on the effect of K application on the reduction of radioactive Cs uptake by crops. They found that each soil containing characteristic clay minerals such as smectite, allophane, and vermiculite had unique Q/I relationships. This suggests that, for many soil types as well as for the test soil, a unique relationship between the PAR and the equilibrium exchangeable K content is to be expected. However, dynamic experiments such as column tests are needed to determine the rate of K leaching.

Four cations, K^+, Na^+, Ca^{2+}, and Mg^{2+}, were considered in this study. However, for greater precision, it may be necessary to consider the effect of NH_4^+, whose concentration may be temporarily increased by fertilizer application. Furthermore, several points need to be addressed as an extension of this study. For example, how do the results of the leaching test performed under pressurized conditions with a pump differ from those obtained under natural conditions, and how can the results of an accelerated test obtained with a compact column be scaled and utilized?

Based on the findings obtained in this study, ways to reduce the K leaching from the soil should be considered. If artificial inputs, such as fertilizers and soil amendments, contain unwanted Mg^{2+} or Ca^{2+}, one approach is to reduce their amounts. In the future, we would like to combine the compact column tests and field experiments to quantitatively evaluate the improvement of K retention capacity when applying soil amendment materials such as zeolite and bentonite.

5. Conclusions

In this study, as part of our efforts to understand the causes of K leaching from paddy soils and the effects of K supply by irrigation water, we investigated the characteristics of K leaching from fine-grained lowland paddy soil by using a compact thumb-size column test. The leaching/accumulation of soil exchangeable K and outflow of major cations were examined using influent with different concentrations and ion compositions. The relationship of soil exchangeable K with the PAR of the influent was also studied. The following results were obtained:

(1) The compact column test had sufficient reproducibility for fine-grained paddy soil, with an average CV of

less than 2 % for dry bulk density and approximately 6 % for soil exchangeable K obtained after the test.

(2) When infiltrating influent with the same cation composition, the lower the concentration, the slower the initial leaching; but as the flow volume increases, the leaching progresses. Conversely, the initial leaching rate was larger at higher concentrations, but equilibrium is reached faster and with a higher equilibrium exchangeable K content.

(3) The soil exchangeable K content at equilibrium is highly correlated with the PAR of the influent.

(4) When DI water was used, K leaching also occurred, albeit at a slower rate, possibly because of exchanges with protons.

(5) The major cation concentrations of eluate tended to stabilize faster in the order $Na^+ > Ca^{2+} = Mg^{2+} > K^+$, with K^+ being the slowest. This suggests that selective adsorption of K^+ by 2:1 type clay minerals may have been affected.

(6) The equilibrium content of soil exchangeable K formed after water percolation was determined by the K^+ concentration and the composition ratio of coexisting exchangeable cations in the influent.

(7) The PAR of the influent is a promising indicator to explain the leaching and accumulation of K.

(8) The PAR represents the ionic composition ratio and reflects the effect of K^+ concentration.

(9) The effects of ion species on K leaching were in the order $Mg^{2+} > Ca^{2+} > Na^+$ based on a 1 mmol L^{-1} change in their concentration and $Ca^{2+} > Mg^{2+} > Na^+$ based on a 10 % change in their concentration, with divalent cations having a greater effect and Na^+ having a smaller effect.

(10) The effect of a 10 % increase in K^+ concentration on soil exchangeable K is estimated to correspond to an approximately 16 % decrease in Ca^{2+} concentration.

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要　　旨

水田における放射性 Cs の吸収を抑制するためには，カリウム（K）の動態を把握し，信頼性の高い効率的な K の施肥技術を確立する必要がある．そこで，本研究では，水田土壌から交換性 K が溶脱する要因として浸透水の影響を調べるために，親指サイズの小型カラムを用いて，異なる陽イオン濃度や組成比を持つ流入水に対する水田土壌からの K の溶脱を調べた．カラム試験では，通水試験後の土壌の交換性 K の変動係数は約 6 % であり，十分な再現性を有していた．異なる水質を持つ流入水の浸透により生じる交換性 K の平衡含量は，流入水の K^+ 濃度と陽イオン構成比によって決定され，PAR（K 吸着比）が，K 溶脱の有効な指標となった．流入水中の各陽イオン濃度が K の溶脱に及ぼす影響は，二価のイオンで大きく，Na の影響は小さいことがわかった．

キーワード：カリウム溶脱，親指サイズカラム試験，PAR（カリウム吸着比），潅漑水質，水田土壌