

## Adsorption of arsenic on soil under different soil moisture conditions

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**ABSTRACT:** The adsorption study was conducted on three Japanese soils with different soil properties to characterize the adsorption pattern of Arsenic (As). Double tube method was used to find out the effect of soil moisture levels on As adsorption. For this study, besides double tube method, conventional batch method also was used. The As adsorption showed two phase kinetics. An initial and rapid adsorption was found at first hour and then gradually preceded before equilibrium in all the three soils in the case of double tube method and in clay loam and light clay soil in the case of batch method. Adsorption seemed to reach equilibrium at 24 h in both methods, though the initial adsorbate load was not the same at the same applied concentration in the methods. Adsorption activity differed according to soil and as well as to the methods. The highest adsorption was found in clay loam soil followed by light clay and sandy loam soil. The linear model of Freundlich adsorption was found better fitted in the case of double tube method than batch method. Suggesting that, under the experimental conditions stated here, the double tube method is more appropriate to describe the adsorption of As in the three Japanese soils under normal field condition. The concentration of As in soil water was slightly increased at different moisture levels from 50%-80% which may contribute significantly to the bioavailability of As at the moisture level below maximum field capacity.

**Keywords:** Arsenic adsorption, batch method, double tube method, Freundlich adsorption isotherm, soil moisture.

### INTRODUCTION

Adsorption is possibly the initial reaction which occurs when arsenic (As) interacts with soil (Goh and Lim, 2004). When As is mixed in to soil, a large portion of it becomes adsorbed on the soil and only a small portion which remains in soil water becomes available to plant (O'Neill, 1995). Thus, adsorption of As on soil is of particular concern. Because this process regulates the fate, mobility and bioavailability of As in soil. Arsenic

adsorption on soil constituents (i.e., clay, oxides of Al, Fe, Mn, and organic matter) is generally the most important process that immobilizes this metalloid (Pierce and Moore, 1982; Masscheleyn *et al.*, 1991; Sadiq, 1997; Matera *et al.*, 2003). It is well known that Fe and Al oxides and hydroxides in soil have a high affinity to arsenic (Raven *et al.*, 1998; Lefferty and Leopart 2005; Shipley *et al.*, 2009). Recently, P content of the soil is reported to affect the adsorption of As in soil (Zheng *et al.*, 2012; Cui and Weng, 2013). Knowledge of adsorption and desorption of

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arsenic is necessary for predicting arsenic behavior in the soil environments (Zhang and Selim, 2005).

In soil, As can exist in inorganic, organic, and gaseous states. The oxidation states of As in the natural systems are -3, 0, +3, and +5 (Sadiq, 1997). The main inorganic forms of As in contaminated soils and sediments are +5 and +3 (Harper and Haswell, 1988), but sometimes the oxidation states of -3 and 0 are expected to be found in highly reducing conditions (Mcbride, 1994). Oxidation states play a significant role in determining the potential mobility and sensitivity of As toward changes of the environmental conditions (Goh and Lim, 2004). In oxidizing condition, As (V) is the predominant species in soil (Williams *et al.*, 2003).

The overall rate of adsorption, relies on numerous factors and the adsorption study of As on soil has been mainly linked to environmental factors such as pH (Pierce and Moore, 1982; Xu *et al.*, 1988; Ticknor and McMurry, 1996; Kuan *et al.*, 1997), redox potential, reaction time (Carbonell-Barrachina *et al.*, 1996; Lo and Chen, 1997; Su and Suarez, 2000; O'Reilly *et al.*, 2001), and oxidation states of As (Sadiq, 1997). Competitive ions and soil properties are also being emphasized due to their key roles in controlling As mobility in soils (Goh and Lim, 2004). However, little is known about the role of soil moisture on As adsorption. Takahashi *et al.* (2004), examined the behavior of As under flooded and non- flooded conditions. Under upland condition, moisture contents may affect the mobility and availability of As in soil. In the soil where As is a problem for rice cultivation like Bangladesh, the fluctuation of soil moisture (during the crop growing period) might affect the adsorption of As on soil and thus, availability of As to plant. Hence, it is important to clarify As adsorption in soil, under different soil moisture conditions. However, it is impossible to measure As adsorption in

soil under different soil water conditions by conventional batch method, but possible by double tube method (Dhareensank *et al.*, 2006).

Numerous studies on As adsorption and their bioavailability have been conducted in the recent years (Manning and Goldberg, 1997; Garcia-Sanchez *et al.*, 2002; Goldberg, 2002; Prasad, 1994). However, in all of these studies, conventional batch method is used and As in the solution is being considered as As available to plant (Wauchope *et al.*, 2002; Fitz and Wenzel, 2002) and there is no report on the relationship between adsorbed As and the As in soil water that is available for plant. Although water soluble As is an indicator of bioavailability of As, it might not show the exact amount of As available to plant. In our previous experiment on phytoremediation of arsenic by barnyard grass and rice, soil water was collected and it was found that As remediation by the plant was affected by the soil water As, not by the total As in soil (Sultana and Kobayashi, 2011). In this experiment, double tube method (Kobayashi *et al.*, 1994) was used to collect the soil water directly to determine the As in soil water that is actually available to the plant.

Because adsorption regulates the bioavailability of As to the plants, even if the same level of As is applied to different soils with different physicochemical properties, the available As to the plants can be significantly different. It is reported that organic matter and clay contents are the two important soil factors that significantly affect the As adsorption in soil (Fitz and Wenzel, 2002; Alvarez-Benedi *et al.*, 2005). In the present experiment, the three soils were selected to clear out the adsorption behavior of As in soils that differs in soil properties, especially clay and organic matter under different soil moisture level at 50-80% of field capacity. This study was conducted with two objectives: (i) to characterize the

adsorption of As including a kinetic study on three different soils, using double tube method; and (ii) to find out the effect of soil moisture on As adsorption under different soil moisture conditions.

## MATERIALS AND METHODS

### Soil preparation

Three soils were collected from three different textural classes of surface and subsurface at a depth of 0-30 cm and their As-adsorption properties were examined by the double tube method (Kobayashi *et al.*, 1994) as well by conventional batch equilibrium method (Wauchope *et al.*, 2002). The soils were clay loam (Tennodai soil), light clay (Ryugasaki A soil) and

sandy loam (Ryugasaki B soil) soil. The clay loam was collected from an upland area of Tennodai, Ibaraki prefecture, Japan; the light clay soil was collected from a paddy field and the sandy loam was collected from an upland field of Ryugasaki, Ibaraki prefecture, Japan. The soils were air dried, ground and sieved through a 2 mm sieve. The pH of the soils was determined using deionized water in a 1:2 soil to water ratio (Tan, 2011). Before using in the experiments, the soils were analyzed for initial As and some other physical and chemical components. Physicochemical properties of the soils are presented in Table 1.

**Table 1. Physicochemical properties of the three selected soils used in the experiment**

| Soil                 | Clay loam           | Light clay   | Sandy loam   |
|----------------------|---------------------|--------------|--------------|
| Field condition      | Upland              | Paddy        | Upland       |
|                      | Physical properties |              |              |
| % sand               | 39.40 ± 0.58        | 31.00 ± 0.73 | 75.70 ± 1.02 |
| % silt               | 38.10 ± 0.86        | 35.70 ± 0.90 | 17.20 ± 0.35 |
| % clay               | 22.50 ± 0.60        | 33.30 ± 0.08 | 7.10 ± 0.40  |
| Field capacity (%)   | 96.50 ± 0.76        | 65.00 ± 1.1  | 43.70 ± 1.0  |
| % organic carbon     | 3.91 ± 0.81         | 2.56 ± 0.20  | 0.88 ± 0.11  |
| pH (1: 2 soil/water) | 6.30 ± 0.59         | 6.63 ± 0.51  | 6.80 ± 0.35  |
| CEC (cmol/kg)        | 17.30 ± 1.30        | 19.10 ± 2.40 | 9.10 ± 0.46  |
| Total As (mg /kg)    | 4.80 ± 0.3          | 20.60 ± 0.45 | 9.5 ± 0.8    |
| Available P (g /kg)  | 1.39 ± 0.06         | 1.41 ± 0.03  | 1.23 ± 0.04  |
| Total Fe (g /kg)     | 53.98 ± 3.38        | 37.65 ± 0.51 | 18.26 ± 0.38 |
| Total Al (g /kg)     | 84.71 ± 0.77        | 42.73 ± 1.27 | 27.08 ± 0.19 |

### Characterization of adsorption equilibrium

Adsorption equilibrium of As was characterized by both batch and double tube method. The double tube method (Fig. 1) was used to get directly the soil water that is available for plant and As in soil water can be determined (Kobayashi *et al.*, 1994; Sultana and Kobayashi, 2011). To compare the results obtained by the double tube method to other available reports, As adsorption experiment was also conducted by batch method. As (V) [disodium hydrogen arsenate heptahydrate [Na<sub>2</sub>HAsO<sub>4</sub>·7H<sub>2</sub>O (special grade)], was used for the adsorption study because it is reported that under oxidizing condition,

soil or soil at the moisture level up to the field capacity, As mainly presents as As (V) (Masscheleyn *et al.*, 1991; Williams *et al.*, 2003).

For the batch method, 1 g air-dried soil was taken in polyvinyl tube and then 25 ml of 50 mgL<sup>-1</sup> As solution was added to each tube. The suspensions were then continuously agitated at 100 oscillations per minute in a horizontal shaker (Asahi Techno-glass Corporation, Tokyo, Japan) for 0, 1, 3, 6, 12, 24 and 48 h at 25±1°C. The pH value of the solution was periodically measured, and its' changes did not exceed ±0.5 units. At the end of the desired reaction time, the suspensions in

the bottles were centrifuged for 15 min at 3000 rpm at the same temperature during agitation and the supernatants were collected. Each sample was replicated thrice. The supernatants were filtrated through a quantitative paper filter (5C, Advantech, Tokyo, Japan) followed by a disposable syringe filter, PTFE 0.20 $\mu$ m (DISMIC- 25HP, Toyo Roshi Kaisha Ltd., Tokyo, Japan) and then analyzed for As by ICP-AES (Inductively Coupled Plasma Atomic Emission Spectrophotometer) coupled with hydride generator.

For the double tube method, air dried soils were put into plastic pots. Deionized water was added to the soils to attain the moisture content at about 50% of the field capacity prior to As application. Then appropriate amounts of  $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$

was added to the soils diluted with deionized water to reach the soil As level at 50 mg  $\text{kg}^{-1}$  and the soil moisture level of approximately 80% of the field capacity. Then the soils were kept for 0, 1, 3, 6, 12, 24 and 48 h. Each sample was replicated thrice. After desired time, soil water was collected from each soil by double tube method (Kobayashi *et al.*, 1994). Briefly, the soils were centrifuged using double tubes at 16000 g for 30 min at 25°C in order to separate it into a solution which was then collected (in the bottom of the outer tubes with the soil remaining in the inner tubes) which are referred to centrifuged soils. The centrifuged soil was dried at 105°C for 24 h and the amount of moisture remained in it was determined and was added to the amount of soil water.

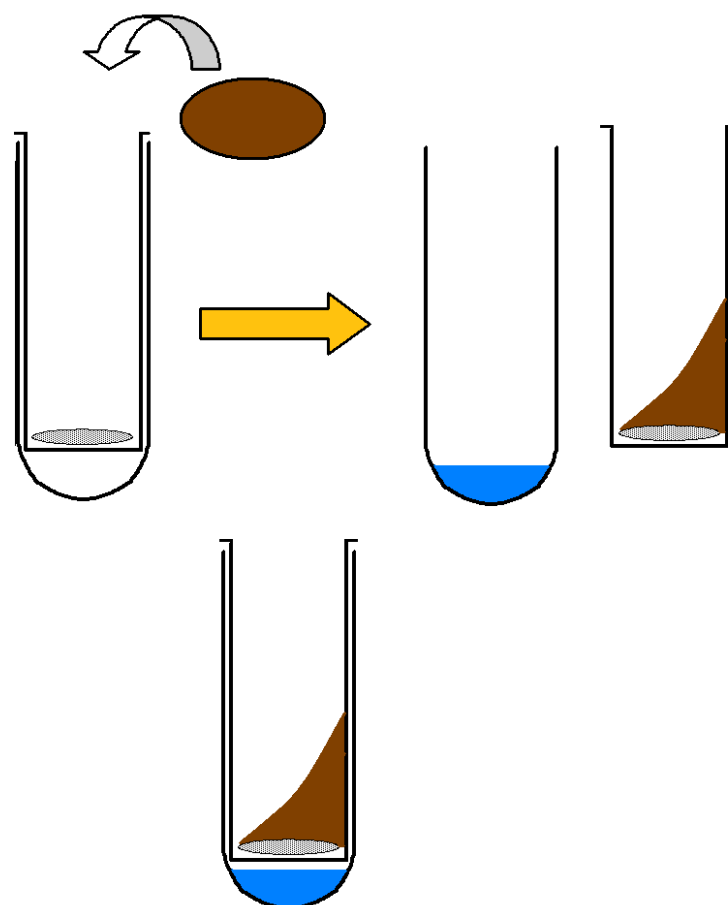


Fig. 1. Schematic representation of double tube method for collecting soil water as adopted from Kobayashi *et al.*, 1994

The amount of adsorbed As in the batch method was determined by mass balance equation modified from the calculating formula by Alvarez-Benedi *et al.* (2005):

$$qS = L (C_0 - C) \quad (1)$$

where q is the estimated amount of adsorbed As (mg/kg)

S is soil mass (kg)

L is the mass of the solution (kg)

C<sub>0</sub> is the initial concentration of As in the solution (mg/L)

C is the measured concentration of As in the solution after different time intervals (mg/L)

Same equation was used for the double tube method, where,

L is amount of soil water (kg)

C<sub>0</sub> is initial concentration of As in the soil (mg/kg)

C is measured concentration of As in the soil water after different time intervals (mg/L).

### Adsorption isotherm

Adsorption isotherms of As were obtained by carrying out adsorption experiment with a fixed amount of soil but varying initial As concentrations. In case of batch method, solutions of 0, 25, 50 and 100 mg/L were used by adding various amount of Na<sub>2</sub>HAsO<sub>4</sub>·7H<sub>2</sub>O into deionized water. Materials and experimental conditions were the same as equilibrium study described above, using the same soil: solution ratio (1:25) and performed in triplicates for each trial. The resulting suspensions were agitated for 24 h, which was the experimental time determined after the equilibrium study. After this time, the same process of centrifugation, removal of supernatant liquid, and filtration was done before ICP-AES analysis.

In the case of double tube method, appropriate amount of Na<sub>2</sub>HAsO<sub>4</sub>·7H<sub>2</sub>O were added to soils to reach the concentration of As in the soils as 0, 25, 50 and 100 mg/kg. The soils were then kept for 24 h to reach the soil water As at

equilibrium. After 24 h the soil waters were collected by the same described method. The soil waters were filtrated and the amounts of As in the soil waters were analyzed by ICP-AES.

Freundlich adsorption equation was used for modeling the adsorption isotherm (Alvarez-Benedi *et al.*, 2005). The modified linear form of Freundlich isotherm was used as:

$$q = Kfc^{1/n} \quad (2)$$

where q is the amount of As adsorbed on soil (mg/kg)

C is the concentration of As on solution at equilibrium

Kf and n are the constants.

### Effect of soil moisture on As adsorption

The effect of soil moisture on As adsorption was determined by calculating the amount of As adsorbed on soil at the moisture levels of 50%, 60% and 80% of field capacity. This range was selected to find out the adsorption within the moisture level suitable for plant growth. Adsorption at 100% moisture level was not done because at saturated soil moisture level, moisture will influence release of As on soil water, similar to aquatic or paddy condition as described by Takahashi *et al.*, 2004. The experiment was conducted only by double tube method because it is impossible to maintain the designated moisture level by batch method. The experimental method was same as mentioned above in the section of adsorption isotherm. 25, 50 and 100 mg/As kg were added to the three types of soils at the moisture levels of 50%, 60% and 80% of field capacity. After 24 h the soil waters were collected by double tube method and the amounts of As were measured.

Analysis of As was carried out by ICP-OES (Optima 7300 V ICP-OES Spectrometers) coupled with a hydride generator at a detection limit of 0.2 ppb arsenic. The relative standard deviation

(RSD) in the analysis was set at 2% prior to analysis. To get consistent measurement, the recovery was kept within 95-105% using standard (JCSS-Japan Calibration Service System) at every 10 samples interval.

All the data were analyzed using Microsoft® Excel software. Freundlich adsorption equilibriums were also done by the same software using the appropriate equations. Results were expressed as the mean of three replicates with  $\pm$  standard error.

## RESULTS AND DISCUSSION

### Adsorption equilibrium

The concentration of As in soil water - obtained by double tube method- and in soil solution -obtained by batch method- are shown in Figure 2. A rapid and high adsorption was found in all types of soil and adsorption was highest in the clay loam followed by light clay soil and sandy loam soil by both methods. Although the adsorbate load was different in two methods, adsorption seemed to reach equilibrium at 24 h in all types of soil in both the batch and the double tube methods. In both methods, there was an initial rapid adsorption in both methods. In the next phase, the adsorption was very slower. The

similar pattern of two phase adsorption kinetic obtained by batch method was reported by Goh and Lim (2004) for a clay loam soil, They stated that the adsorption rates of As(V) was rapid in the first hour and then decelerated noticeably as the reaction plateaued after 8h. The increases in adsorption beyond 8h were marginal and seemed to approach equilibrium at about 24 h. Alvarez-Benedi *et al.* (2005), also found similar adsorption equilibrium for As in different types of soil ranging from sandy to clay. Smith *et al.* (1999) showed that As (V) retention by soils attained apparent equilibrium in less than 1 h, followed by a steady, but slow rate for 72 h. Nevertheless, the effect of longer retention time of 72hr and more on As adsorption was not investigated.

It is notable that in all three soils, adsorption was rapid and followed similar pattern to reach equilibrium except sandy loam soil in batch method. In batch method, sandy loam soil took relatively higher time at initial rapid stage that might be due the lower clay content compared to other two soils. However, in double tube method, there was no significant difference in the pattern of initial rapid adsorption

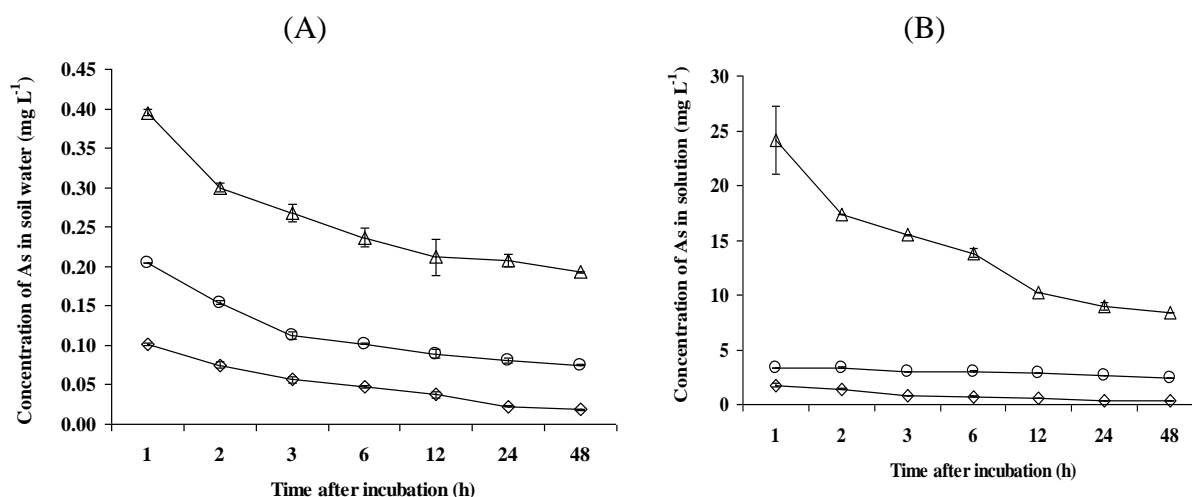


Fig. 2. Kinetics of As (V) adsorption in clay loam soil (◇), light clay soil (O), and sandy loam soil (Δ) treated with 50 mg As kg<sup>-1</sup> in the double tube method (A) and with 50 mg As L<sup>-1</sup> in the batch method (B). Bars indicate the  $\pm$  standard error of the means (n= 3).

between three soils. Suggesting that despite of lower clay content in sandy loam than the other two soils, under actual field conditions the concentration of As provided can cause sharp and rapid adsorption same as to the other two soils. Therefore, double tube method is reasonable to understand the exact feature of kinetic of As adsorption in all the three types of soils. Although the equilibrium was not reached exactly within the experimental time especially in the cases of batch methods, 24 h after As treatment was considered as equilibrium time for both methods because after 24 h the increase of adsorption was very slow.

### Adsorption isotherm

Figure 3 shows that adsorption patterns were fit to modified form of linear Freundlich adsorption isotherm. Table 2 shows the adsorption properties of As on soil described by the Freundlich equation (Feng *et al.*, 2013; Alvarez-Benedi *et al.*, 2005). In all types of soils, adsorption of As (V) was strong with the highest adsorption in clay loam soil followed by light clay and sandy loam soil. Adsorption was very sharp and rapid in the first hour and the rate was gradually decreased due to the saturation of surface sites. Although the results were not clearly linear, a linear isotherm was applied

to the model, because linear adsorption is commonly used for approximation in reactive solute transport modeling (Brusseau, 1998; Bethke and Brady, 2000). The Freundlich adsorption isotherm was better fitted in the case of double tube method than in the case of batch method. The value of  $1/n$  was lower in each soil in case of batch method than that of double method. This might be because of the higher As concentration in batch method than the other one. The ratio of adsorbed As and soil water As was found more in case of double method than the ratio of adsorbed As and solution As in the case of batch method, though the batch method allow the soil particles to be separated and expose more surface area to adsorb arsenic. It is notable that, despite the lower amount of As adsorbate load per unit soil at same concentration in the case of double method than that in the case of batch method. With the increase of concentration of the applied As from 25 to 100 mg Kg<sup>-1</sup>, the adsorption was gradually decreased and the adsorption model was fitted in the linear Freundlich isotherm. Therefore, it can be suggested that double method is the more appropriate method than the batch method to explain As adsorption pattern in soil.

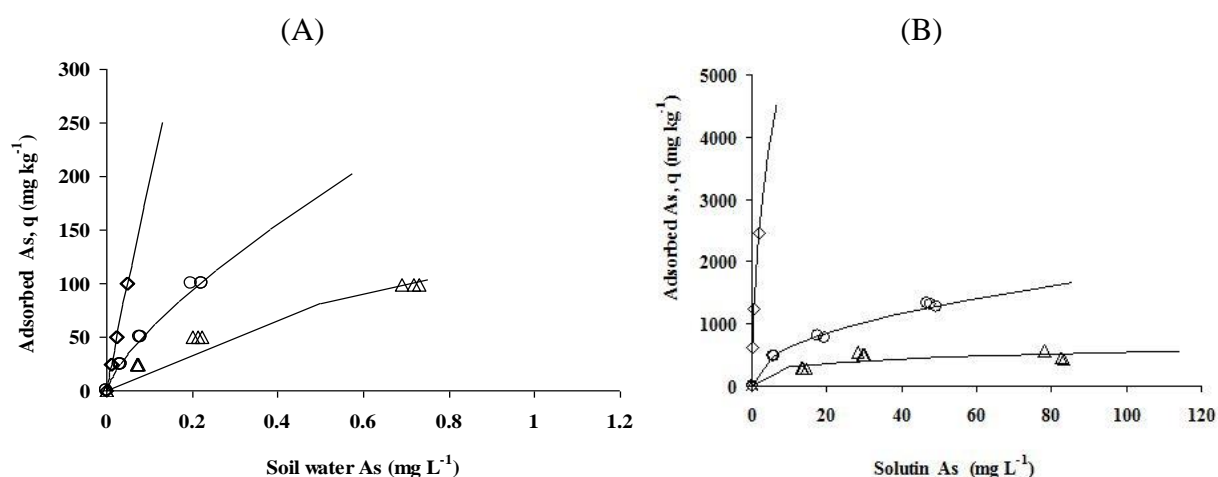


Fig.3. Freundlich adsorption isotherm of As (V) on clay loam soil (◇), light clay soil (○), and sandy loam soil (△). (A) Stands for the double tube method and (B) for the batch method

Table 2. Parameters of As adsorption isotherm in different soils

| Soil       | Double tube method |      |      | Batch equilibrium method |      |      |
|------------|--------------------|------|------|--------------------------|------|------|
|            | Kf                 | 1/n  | R2   | Kf                       | 1/n  | R2   |
| Clay loam  | 1685.11            | 0.93 | 0.98 | 1915.82                  | 0.46 | 0.99 |
| Light clay | 303.51             | 0.75 | 0.98 | 212.79                   | 0.45 | 0.98 |
| Sandy loam | 122.88             | 0.60 | 0.99 | 176.78                   | 0.26 | 0.95 |

As shown in the soil properties (Table 1), both clay and organic carbon could be the main properties influencing the As adsorption. Sandy loam soil has lower organic carbon as well as lower clay content than the other two soils. However, the highest adsorption of As in clay loam soil cannot be explained solely with organic carbon and clay content. Because there was little difference in these two properties among light clay and clay loam soil as shown in Table 1. The other factors influencing As (V) adsorption might be the Fe and Al contents of clay loam soil that is significantly higher in clay loam soil than in the other two soils. Some reports have shown that Fe hydroxide strongly interact with dissolve As and thus influence adsorption of As (V) in soil (Sadiq, 1997; Smith *et al.*, 1998). Soil colloids with different charges can also influence the adsorption of As on soil as described by Feng *et al.* (2013). The above mentioned interaction might influence the adsorption of As (V) on clay loam soil in the present experiment. These results suggested that light clay soil and sandy loam soil had relatively lower adsorption of As and thus allow more As in soil water than clay loam soil.

#### Effect of soil moisture on As adsorption

The concentration of As in soil water increased slightly with the increase of soil moisture conditions from 50-80% at all application doses (Fig. 4). The adsorption of As was found similar under all moisture conditions though there is a marginal decrease of As adsorption (was found with the increase of soil moisture). According to Takahashi *et al.* (2004), As in irrigation waters is incorporated into Fe-hydroxide in soil during the non-flooded period and

quickly released from soil to water during flooded period because of dissolution reduction of Fe (hydr) oxide phase Fe-hydroxide and reduction of As (V) to As (III). Under upland condition or normal field moisture condition Fe-hydroxide are known to adsorb arsenic (Smith *et al.*, 1998). With the increase of moisture in the soil, the release of As would increase due to the similar reason as described above. However, in the present experiment, since the adsorption of As on all the three soils were very high, the amount of As adsorbed on soil solid were significantly higher than the amount of As in soil water. That was the reason of adsorption similar its even though the As in soil water had a marginal increase with the moisture content in soil. This slight difference in concentration of As in soil water in the different soil moisture levels below maximum field capacity might contribute significantly the bioavailability of arsenic.

#### CONCLUSIONS

The adsorption study was conducted on three Japanese soils with different soil properties by double tube and batch methods. The results indicated that As adsorption showed two phase kinetics where an initial and rapid adsorption was found at the first hour and then gradually proceeded before reached to equilibrium. Adsorption seemed to reach equilibrium at 24 h in both double tube and batch methods though the adsorbate load was not same, at the same concentration in two methods. Adsorption results were fit to modified Freundlich model of adsorption isotherm. Despite the lower amount of As adsorbate load per unit soil at the same concentration in the case of double tube



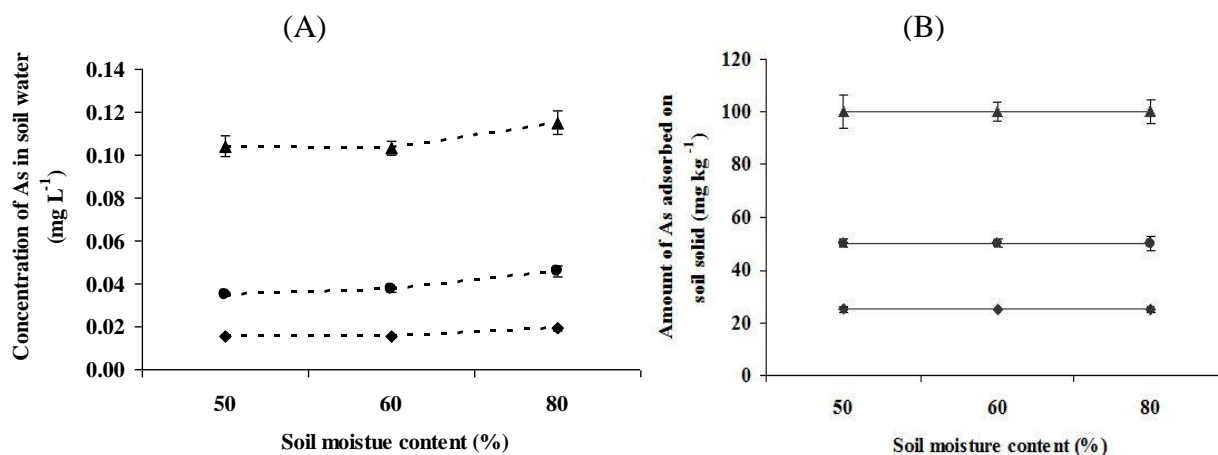


Fig.4. Amount of As in soil water (A) and on soil solid (B), under different moisture conditions. The rate of As applied was 25(■); 50(●) and 100(▲) mg As kg<sup>-1</sup>. Bars indicate the ± standard error of the means (n= 3).

method than that in the case of batch method, Freundlich adsorption isotherm was better fitted in the case of double tube method than in the case of batch method. It can be suggested that under the condition stated here, double method is better than batch method to explain the adsorption of As (V) in a soil without disturbing the normal field condition. The adsorption isotherm showed that adsorption activity differed according to soil. The highest adsorption was found in clay loam soil followed by light clay and sandy loam soil. Arsenic adsorption was studied under different moisture condition below the maximum field capacity only by double tube method because it is practically impossible to do this experiment under batch method. Arsenic concentration in soil water was slightly increased by increasing soil moisture conditions. However, due to very high As adsorption capacity of all the three soils used for the study, the increase of As in soil water was seemed to be very small and the adsorption of As on soil solid was seemed to be similar in all the soils at different moisture levels from 50%-80%. This slight increase in concentration of As in soil water under different moisture level stated here can significantly affect the bioavailability of As and phytoremediation of As from soil.

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