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Article

Uptake of uranium, thorium, radium and potassium by four kinds of dominant plants grown in uranium mill tailing soils from the southern part of China

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Abstract – The concentrations of radionuclides in soil from uranium mill tailings in South China were analyzed. The results showed that all radionuclides in soil samples exceeded the soil background value. Through the field investigation, four kinds of dominant plants were identified. It was found that the total quantity of radionuclides in the whole plant from high to low were pokeberry, Cibotium barometz, reed and garden sorrel. We also found that Cibotium barometz had the highest transfer factor (TF) for ^{238}U, garden sorrel had the highest TF for ^{232}Th and reed had the highest TF for ^{226}Ra and ^{40}K. Cibotium barometz had the highest bioconcentration factor (BF) for ^{238}U, ^{232}Th and ^{226}Ra, and pokeberry had the highest BF for ^{40}K. Pokeberry had the highest phytoremediation factor (PF) for ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K. On the basis of the above conclusions, pokeberry could be a candidate for phytoremediation of radionuclide-contaminated soils.

Keywords: radionuclide / soil / phytoremediation / uranium mill tailings

1 Introduction

With the rapid development of the nuclear industry and nuclear energy, the demand for uranium (U) mining metallurgy products keeps increasing. However, extraction of uranium and ore in milling facilities produces large amounts of uranium tailings, which result in soil radioactive contamination (Rao and Sudhakar, 2014). The uranium tailings contain a series of long-lived radionuclides, not only of uranium (^{238}U) but also of thorium (^{232}Th), radium (^{226}Ra) and potassium (^{40}K). Discharged radionuclides from uranium tailings can persist in soil for many years, travel long distances and accumulate in the food chain, causing damage to the human body not only where they are produced and used, but globally. So, investigations of soil polluted by radionuclides should receive more attention. One of the promising strategies to treat polluted soils is the use of phytoremediation techniques (Sharma *et al.*, 2015). In the southern part of China, there is a large area of radionuclide-contaminated soils in uranium mill tailings which is stalled for 20 years. As time goes on, the soils might act as a potential source of radionuclides.

The aims of this study were: (1) to select dominant plants grown in the soil from this region through field investigation; (2) to determine the concentrations of radionuclides (^{238}U, ^{232}Th, ^{226}Ra and ^{40}K) in the soils of the investigated area and identify the potential correlation among these radionuclides by the use of the Pearson coefficient; (3) to determine the concentrations of radionuclides (^{238}U, ^{232}Th, ^{226}Ra and ^{40}K) in the selected plants, and (4) to determine the soilplant factors, the TF (transfer factor), BF (bioconcentration factor) and PF (phytoremediation factor), used in the present study on the impact of releases of radionuclides into the environment, which represent the capacity of a plant species to accumulate radionuclides (IAEA, 1994). The TF is defined as the ratio of the target element concentration in the plant shoot to that in the plant root; the BF is defined as the ratio of the target element concentration in the plant to that in the soil; the PF is defined as:

PF = Target element concentration in the plant shoot × Biomass of the plant shoot/Target element concentration in the soil.

Our study will thus provide a better understanding of phytoremediation possibilities for radionuclide-contaminated soils and guide the management of this area in future.

2 Materials and methods

The uranium mill tailings are located in the southern part of China. It has a subtropical humid continental climate. The area receives regular and plentiful rainfall (1329 mm per year) during late spring and early summer. The average annual temperature is 18.1 °C, with an extreme low temperature of –5.9 °C

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Table 1. Descriptive statistics of radionuclide contents of the analyzed soil.

Parameter	Min	Max	Mean	Deviation	Skewness	Kurtosis
^{238}U (Bq kg^{-1})	717.9	945.3	841.1	57.9	–0.07	–0.60
^{232}Th (Bq kg^{-1})	41.0	70.9	54.6	6.85	0.10	–0.26
^{226}Ra (Bq kg^{-1})	1202.1	1489.3	1329.8	70.5	0.13	–0.76
^{40}K (Bq kg^{-1})	728.9	928.3	853.8	58.2	–0.43	–0.98

and extreme high temperature of 39.9 °C. The average sunshine hours per year are 1447 h and the dominant wind direction is northeast. The sampling sites extended over an area of 800 m^2.

The density of dominant plant samples which were equal to or greater than 5 strains m^{-2} were collected from the study site, and ten strains of each dominant plant were mixed to give a composite sample. All the plant samples were divided into the shoot (including seeds, leaves, stalk and stem) and the root. Soil samples were taken under each selected plant from the root zone at a depth interval of 0 m down to 0.2 m by digging profiles. Collected samples of plants and soil were packed into water-tight bags to prevent cross-contamination and shipped to the laboratory. In the laboratory, plant samples were gently washed with deionized distilled water for 3 min to remove the soil, and then the plant and soil samples were kept drying at room temperature for 4 weeks. The weight of each plant sample was precisely obtained by a scale before it was ground into powder. They were sealed in clean polyethylene containers and stored in a refrigerator for analysis. The dried powders of plant and soil samples were analyzed for the total concentrations of radionuclides at the Engineering Research Center of Biomass Materials, Ministry of Education, Southwest University of Science and Technology, China.

For each plant and soil sample, 0.1 g of the sample was taken and digested with analytical reagent-grade HNO_3 and HF, which were purified by sub-boiling distillation to give a low reagent blank and corresponding low analytic limit of detection. The concentrations of radionuclides (^{238}U, ^{232}Th, ^{226}Ra and ^{40}K) in plant and soil samples were determined by inductively coupled plasma mass spectrometry (ELEMENT 2/XR, Thermo Scientific, USA). The detection limit was 0.003 μg L^{-1} for U and Th, 0.002 μg L^{-1} for Ra and 0.02 μg L^{-1} for K, respectively. The accuracy of inductively coupled plasma mass spectrometry analyses is estimated to be better than ±3% (relative) for the elements determined.

Multivariate analysis and analysis of variance (ANOVA) were used to demonstrate differences among the samples using SPSS 19.0. Pearson's correlation coefficient (PCC) was also calculated using SPSS. $P < 0.05$ was considered to be significant.

3 Results and discussion

The concentrations of radionuclides in the uranium mill tailing soils are presented in Table 1. The arithmetic mean and the standard deviation of radionuclide concentrations for all soil samples were used to describe the central tendency and variation of the data. The mean concentrations of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K obtained in this study were 21.45, 1.12, 36.16 and 1.48 times more than the mean values of radionuclides in soils of China: 39.5 Bq kg^{-1} for ^{238}U, 49.1 Bq kg^{-1} for ^{232}Th, 36.5 Bq kg^{-1} for ^{226}Ra and 580.0 Bq kg^{-1} for ^{40}K (Table 1) (Cao *et al.*, 2012). In the light of the above data, it can be concluded that all the radionuclides in the soil samples exceeded the background values; especially ^{238}U indicated that the soil in the uranium mill tailings was severely contaminated by radionuclides. In fact, the migration of radionuclides in soils depends on many factors, such as physicochemical, biological, geochemical and microbial influences, soil and water properties, air flows and specific interactions of radionuclides with vegetation or other organisms where they accumulate (Cerne *et al.*, 2011). So, there is a need for radionuclide-contaminated soil management through phytoremediation. Also, phytoremediation technology can be used to distinguish natural uranium from releases and uranium from the radiological background. For example, Pourcelot proposed normalizing ^{238}U activity measured in plants taken in the surroundings of nuclear sites with respect to ^{232}Th, considering that the source of this latter is the background (Pourcelot *et al.*, 2015).

Table 2. The Pearson correlation matrix of the radionuclides.

	^{238}U	^{232}Th	^{226}Ra	^{40}K
^{238}U	1.00	–0.13	–0.02	0.42
^{232}Th		1.00	0.77*	0.27
^{226}Ra			1.00	0.81*
^{40}K				1.00

* $P < 0.05$.

To identify to what extent these radionuclides can exert an influence together on the study soils, correlation analyses were carried out between radionuclide pairs. It turned out that the concentration differences between ^{238}U and the other radionuclides were not significant, while the ^{232}Th concentration was significantly correlated with that of ^{226}Ra, and ^{226}Ra was related to ^{40}K because of the source of the radionuclides and the chemical properties on their own (Table 2). For example, (1) ^{226}Ra was the direct decay product of ^{230}Th, and (2) the migration patterns and dissolving conditions of ^{226}Ra and ^{40}K in soil were similar. In this case, the individual concentration of a certain radionuclide could be a good predictor for the concentration of another radionuclide.

Through on-the-spot research of the study site, we found that there are four kinds of plants widely distributed in this region. They are all perennial herbaceous plants: reed (*Phragmites australis (Cav.) Trin. exSteud.*), pokeberry (*Phytolacca americana*), garden sorrel (*Rumex acetosa Linn*) and Cibotium barometz (*Cibotium barometz (Linn.) J. Sm.*). The results indicated these four plants, reed, pokeberry, garden sorrel and Cibotium barometz, are not very sensitive to radioactive contamination stress, and they can adapt to the radionuclide-contaminated environment better. In addition, some authors have defined the hyperaccumulators for heavy metals in three ways: first, plants can grow normally in heavy metal-contaminated soil and will not show heavy metal poisoning phenomena; second, the amount of the element accumulated in an organism can be higher than that in other organisms; third, the concentration of an element accumulated in an organism can be higher than that in the soil (Rascio and Navari-Izzo, 2011; Zhang *et al.*, 2012; Rees *et al.*, 2015). Based on the standards, we further studied the potential of these four plants to become hyperaccumulators.

Among all the plant samples, Cibotium barometz accumulated the highest concentration of ^{238}U and ^{232}Th in the shoot and ^{232}Th and ^{226}Ra in the root. Pokeberry accumulated the highest concentration of ^{226}Ra and ^{40}K in the shoot and ^{238}U and ^{40}K in the root. Also, it was found that the total quantity of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K in the whole plant from high to low were pokeberry, Cibotium barometz, reed and garden sorrel, respectively. According to these results, Cibotium barometz and pokeberry were more suitable for phytoremediation than the other two plants (Table 3).

However, the potential of a plant to be used in phytoremediation does not merely depend on the concentration of the element in the plant. It also depends on the transfer and accumulation ability of radionuclides. So, we chose the TF, BF and PF to further study the capacity to absorb radionuclides of the selected plants in this study. The TF is used as an index for the accumulation of a target element in the plant and its transfer from the root to the shoot (Chang *et al.*, 2014). The factors such as the concentration of a target element, the dynamic of radionuclides in the plant and the plant type may modify the uptake and ratio of the content of the element in the plant shoot to that in the plant root (Tu *et al.*, 2002). As shown in Table 4, Cibotium barometz and pokeberry have the highest TF for ^{238}U, garden sorrel has the highest TF for ^{232}Th, and reed has the highest TF for ^{226}Ra and ^{40}K.

The concentration of a target element in the soils is another important factor that will influence the time it takes to complete the phytoremediation. The BF is used as an index for the ability to absorb and transfer radionuclides from soil to plant (Chen *et al.*, 2015). The migration of radionuclides from soil to plant is closely related to the chemical properties of radionuclides. For example, uranium in the soil is most often bioavailable for plants in the form of uranyl cations, uranyl-carbonate complexes, uranyl-phosphate complexes and uranyl-citrate complexes (Gavrilescu *et al.*, 2009). However, thorium cannot easily move or transport from soil to plant (Yan *et al.*, 2015). Radium is different; it is an easily dissolved salt and could be readily absorbed by plant roots and accumulated in the roots and shoots. Radium is chemically analogous to the essential element Ca and considered to participate in similar physiological processes in plants (Abreu *et al.*, 2014). Higher amounts of potassium in the plants are normal, because of its essential role in almost all physiological processes needed to sustain plant growth and reproduction. Potassium is a cation, with the highest concentration in the cytoplasm, and plays a vital role in photosynthesis, translocation of sugars and starches, protein synthesis, control of ionic balance, maintenance of turgor, reduction of water loss, activation of plant enzymes,

Table 3. Concentrations of radionuclides in the plant and soil samples collected from the uranium mill tailing repository.

Species	FW (kg)	^{238}U (Bq kg^{-1})			^{232}Th (Bq kg^{-1})			^{226}Ra (Bq kg^{-1})			^{40}K (Bq kg^{-1})		
		Shoot	Root	Soil	Shoot	Root	Soil	Shoot	Root	Soil	Shoot	Root	Soil
1	0.25 ± 0.02	12.38 ± 0.13	14.64 ± 0.30	847.2 ± 36.7	1.72 ± 0.03	4.54 ± 0.08	55.1 ± 4.33	149.6 ± 4.49	57.2 ± 1.55	1319.9 ± 73.6	108.2 ± 6.62	77.9 ± 2.85	857.8 ± 53.8
2	0.45 ± 0.03	64.99 ± 3.26	26.69 ± 1.23	805.0 ± 48.2	2.56 ± 0.16	9.25 ± 0.21	56.4 ± 7.21	181.4 ± 8.03	115.3 ± 6.71	1330.6 ± 76.4	176.8 ± 13.2	172.8 ± 7.19	850.0 ± 69.1
3	0.005 ± 0.0005	17.38 ± 0.94	24.23 ± 3.01	853.6 ± 72.3	0.86 ± 0.10	1.56 ± 0.13	49.6 ± 6.22	27.3 ± 2.50	76.9 ± 2.97	1339.0 ± 76.3	47.0 ± 4.03	58.7 ± 7.67	848.1 ± 63.0
4	0.003 ± 0.0003	73.9 ± 6.34	24.8 ± 3.15	858.7 ± 60.1	3.23 ± 0.36	13.4 ± 1.04	57.5 ± 7.28	160.1 ± 8.92	154.1 ± 8.74	1329.6 ± 65.0	132.6 ± 9.44	158.0 ± 10.62	859.4 ± 54.2

1: Reed 2: pokeberry, 3: garden sorrel, 4: Cibotium barometz.
FW: fresh weight.

Table 4. The TF, BF and PF of the plants collected for radionuclides in soil samples from the uranium mill tailing repository.

Species	TF				BF				PF			
	^{238}U	^{232}Th	^{226}Ra	^{40}K	^{238}U	^{232}Th	^{226}Ra	^{40}K	^{238}U	^{232}Th	^{226}Ra	^{40}K
1	0.85	0.38	2.62	1.39	0.03	0.11	0.16	0.22	0.004	0.008	0.028	0.032
2	2.43	0.28	1.57	1.02	0.114	0.21	0.22	0.41	0.036	0.02	0.061	0.094
3	0.72	0.55	0.36	0.80	0.05	0.05	0.08	0.12	1.0×10^{-4}	8.7×10^{-5}	1.0×10^{-4}	2.8×10^{-4}
4	2.98	0.24	1.02	0.84	0.115	0.30	0.24	0.34	2.6×10^{-4}	1.7×10^{-4}	3.6×10^{-4}	4.6×10^{-4}

1: Reed, 2: pokeberry, 3: garden sorrel, 4: Cibotium barometz.

resistance to plant disease and many other processes (Chérel *et al.*, 2014). In this study, Cibotium barometz has the highest BF for ^{238}U, ^{232}Th and ^{226}Ra, and pokeberry has the highest BF for ^{40}K.

Also, it has been proposed that a plant with low dry biomass would have a low resultant capability of removing an element and not be suitable for phytoremediation, though the concentration of the element would be very high in this plant (Robinson *et al.*, 1997). In this study, we found that pokeberry has the highest PF for all the radionuclides.

Synthesizing the above analysis, including the growing ability of these plants, radionuclide content in the plants, and the TF, BF and PF, pokeberry has a better performance than the other plants. So, in the present study, pokeberry has the most potential for phytoremediation among the selected plants.

4 Conclusion

All the concentrations of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K in soil samples from the uranium mill tailings exceeded the natural radionuclide content in soils of China. Based on the definition of hyperaccumulators and evaluation factors such as the TF, BF and PF, pokeberry could be a candidate for phytoremediation for radionuclide-contaminated soils. However, because pokeberry is an invasive plant in China, in-depth and careful research on this plant for phytoremediation is required.

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