Uptake of ^{137}Cs and ^{90}Sr in rice grown on soils from Qinshan and Daya Bay area

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Abstract . In order to study the transfer of water borne ^{137}Cs and ^{90}Sr into rice plants, pot experiments were done by growing rice plants on soils taken from rice beds near two nuclear power plants: Qinshan Nuclear Power Plant and Daya Bay Nuclear Power Plant in China. The vertical migration of ^{137}Cs and ^{90}Sr in the soil samples due to leaching was studied, and results show that almost all the contaminants were retained in the top 1 cm of soil, with at most a few percent down to 3 cm of soil. The transport factors (TF) of ^{137}Cs and ^{90}Sr were studied by adding various activity concentrations of the two radionuclides to the irrigation water and then measuring the activity concentration in the various rice tissues. The TF were also analysed for their dependence on soil characteristics, time and duration of the contamination.

1. INTRODUCTION

Caesium-137 and Strontium-90 are fission products that can be released in large quantities in the event of an accident involving nuclear power plant. Because of their long half-life with 28.8y for ^{90}Sr and 30y for ^{137}Cs, and their abundance and biological mobility, they are critical components in nuclear fission wastes, and are the two of the best studied artificially produced radionuclides in our environment. Owing to their chemical similarity with Ca and K respectively, ^{90}Sr and ^{137}Cs can enter into food chains easily and are thus two important contributors to internal dose of human bodies. As with most other radionuclides, Cs and Sr can enter plants by leaf surface absorption and root uptake, depending on plant species, plant growth varieties, and the ecosystem characteristics such as properties of soil, chemical and physical forms of the nuclide etc ([1], [10], [27]).

After the Chernobyl accident that occurred in 1986, numerous investigations were done to study the transfer of ^{137}Cs and ^{90}Sr in the food chains, and the migration and retention of the radionuclides in different soil types ([20]; [17]; [8]). The majority of the studies were on the air-plant and soil-plant pathways where the ^{137}Cs and ^{90}Sr enter the plant after deposition on the plant or soil. There are only a few studies on the water-soil-plant pathway for the water-cultured plants ([14]). Yet, one of these plants is the basic food for Asian people – rice. A measurement of nuclear test fallout ^{137}Cs and ^{90}Sr distribution in rice showed that ^{90}Sr concentrates more in leaves and ^{137}Cs accumulates more in the growing parts of rice plant such as hull and rachis([26]). However, the results about the transfer factors of both nuclides for edible parts of rice plant are slightly different from that reported by other organizations([26]). To improve the knowledge of Cs and Sr transfer in this principal Asian food, the transfer of water borne ^{137}Cs and ^{90}Sr into rice plant was investigated in this study.

2. METHOD

In order to study the transfer of ^{137}Cs and ^{90}Sr into rice, in the event of an accident involving a nuclear power plant, surface soil samples were taken from rice beds near two nuclear power plants (both are pressurized water reactors) in China. The soil samples were taken from 5 sites: Guantang, Futing and Changchuanba near Qinshan Nuclear Power Plant, and Shenzhen and Huiyang near Daya Bay Nuclear Power Plant. The soil characteristics were analyzed and the results are given in Table 1.

Table 1. The characterisitics of soils taken from Qinshan area and Daya Bay area*.

Area	Soil	pH	Total N (g/kg$_{soil}$)	Effective P_2O_5 (mg/kg$_{soil}$)	Exchangeable K^+ (mg/kg$_{soil}$)	Cation Exchange Capacity (nmol/100 g$_{soil}$)
Qinshan	Guantang	6.6	2.06	14.4	106	14.1
	Futing	6.6	2.13	4.1	121	18.5
	Changchuanba	6.7	1.03	34.4	102	14.5
Daya Bay	Shenzhen	5.7	1.09	11.4	56	2.7
	Huiyang	5.4	0.70	19.2	38	NA

* Data from: Office of Soil Investigation of Haiyan Government. Haiyan soil; 1986.
Office of Soil Investigation of Shenzhen Government. Shenzhen soil; 1986

2.1 Adsorption of ^{137}Cs and ^{90}Sr in soil

The adsorption and penetration of ^{137}Cs and ^{90}Sr in the soil samples were studied by leaching tests in soil columns with either $^{137}CsNO_3$ and $^{90}Sr(NO_3)_2$ solution. The topsoils taken from the five locations were air-dried, ground and sieved (0.3 mm, 60 mesh). They were filled separately into plastic tubes of 40 cm long and 1.5 cm in diameter to a height of 20 cm. $^{137}CsNO_3$ and $^{90}Sr(NO_3)_2$ solution without carrier were added respectively to the soil columns to determine the retention of ^{137}Cs and ^{90}Sr in the soil under the following conditions:

(a) 40 mL of $^{137}CsNO_3$ and $^{90}Sr(NO_3)_2$ solution having different activity concentration (3.7×10^3, 3.7×10^4 and 3.7×10^5 Bq l^{-1}), and

(b) different volumes of solution (40, 80 and 120 mL) but having the same activity concentration of 3.7×10^4 Bq l^{-1}.

The whole ^{137}Cs and ^{90}Sr solution were added to the soil column at once. It took a few hours for the solution to drain completely from the soil columns. Then kept the top of column filled water two days by adding additional tap water continually, so as to study the leach trend in column with time. After the solution has drained completely, the soil columns were air-dried and divided into sample discs of 1 cm thick. All samples were measured for the retained ^{137}Cs and ^{90}Sr by using a low level α, β proportional counter.

2.2 Transfer of ^{137}Cs and ^{90}Sr from irrigating water to rice

Jiacan 6# rice was grown in pots of 25cm diameter and 30cm height containing 3 kg of soil. Jiacan 6# has a lifetime of about 140 days. Water containing $^{137}CsNO_3$ and $^{90}Sr(NO_3)_2$ was respectively used to irrigate the rice plants and samples were taken to determine the activity of ^{137}Cs and ^{90}Sr in the plants under the following conditions.

a) ^{137}Cs solutions and ^{90}Sr solutions of four different surface activities (activity per unit surface area of soil): 2.26, 22.6, 226 and 2260 Bq cm^{-2} were irrigated to Guantang soil at the seeding stage (50 days from sowing) of the rice. Samples of stem, leaf, husk and seed were taken at harvest time.

b) ^{137}Cs solutions and ^{90}Sr solutions of 2.26 Bq cm^{-2} was irrigated to plants grown in Guantang soil at three growth stages: seeding stage, anthesis stage and milk stage. Samples were taken at harvest time.

c) ^{137}Cs solutions and ^{90}Sr solutions of 2.26 Bq cm^{-2} were irrigated to the five different soils at seeding stage of the plants. Samples were taken at harvest time.

d) ^{137}Cs solutions of 2.26 kBq cm^{-2} were irrigated to Guantang soil at seeding stage of the plants. Samples were taken at different growth stages: maximum tiller number, panicle formation, anthesis, and milk.

2.3 Sample preparation and measurement

Samples of stem and leaf were cut directly from the plants. They were washed and then air-dried and weighed. The rice was collected and removed from the plants as usual. It was then ground to remove the husk that was collected separately. In China, the rice is only lightly ground just sufficient to remove the husk, so the rice still looks light brown in color. After all the samples were weighed, ground and ashed below 400°C for 8hours, they were counted in 2.46 cm diameter planchets in a FH-1914 Low Level β Counter which has a counting efficiency of 20%. Three samples were usually used in each measurement. The counting errors were less than 5%.

3. RESULTS AND DISCUSSIONS

3.1 Adsorption of ^{137}Cs and ^{90}Sr in soil

Results of the soil column test are given in Table 2 and 3, which show that most of the ^{137}Cs and ^{90}Sr were retained in the top 1 cm of soil regardless of the soil type, the activity concentration and duration of penetration. However, the results also show that leaching ability of ^{90}Sr is higher than ^{137}Cs.

Table 2. Retention of ^{137}Cs and ^{90}Sr in soil columns with different activity concentrations

Nuclide	Soil	Depth of Soil Column (cm)	% of Retention		
			$3.7x10^3$ Bq l^{-1}	$3.7x10^4$ Bq l^{-1}	$3.7x10^5$ Bq l^{-1}
^{137}Cs	Guantang	1	98.7	98.0	99.1
		2	1.3	1.9	0.5
		3	0	0.1	0.3
		4 - 20	0	0	0.1
	Futing	1	79.4	84.6	93.9
		2	20.6	13.2	5.8
		3	0	2.2	0.2
		4 - 20	0	0	0.1
	Changchuanba	1	98.9	99.4	95.5
		2	1.1	0.3	2.4
		3	0	0.3	1.6
		4 - 20	0	0	0.5
	Shenzhen	1	99	97.8	73.8
		2	1	2.2	17.4
		3	0	0	6.9
		4 - 20	0	0	1.9
	Huiyang	1	99.3	96.3	98.7
		2	0.7	3.2	1.0
		3	0	0.5	0.3
		4 – 20	0	0	0
^{90}Sr	Guantang	1	99.5	98.7	93.1
		2	0.5	0.9	6.7
		3	0	0.2	0.1
		4	0	0.2	0.1
		5 – 20	0	0	0
	Shenzhen	1	85.4	82.0	87.7
		2	13.4	15.6	10.1
		3	0.6	2.3	1.2
		4	0.6	0.1	1.0
		5 – 20	0	0	0

Table 3 Retention of ^{137}Cs and ^{90}Sr in soil columns with different durations and volumes of penetration

Nuclide	Soil	Depth of Soil Column (cm)	% of Retention 40 mL	80 mL	120 mL
^{137}Cs					
	Guantang	1	98.0	98.8	96.9
		2	1.9	1.1	2.8
		3	0.1	0.1	0.2
		4	0	0	0.1
		5 - 20	0	0	0
	Futing	1	84.6	89.1	63.3
		2	13.2	7.8	25.0
		3	2.2	2.1	6.1
		4	0	1.0	4.6
		5 - 20	0	0	1.0
	Changchuanba	1	99.4	96.4	90.4
		2	0.3	1.5	8.0
		3	0.3	0.8	0.8
		4	0	0.7	0.4
		5 – 20	0	0.6	0.4
	Shenzhen	1	97.8	93.1	94.7
		2	2.2	4.4	3.6
		3	0	1.4	0.8
		4	0	0.7	0.5
		5 – 20	0	0.4	0.4
	Huiyang	1	96.3	94.7	92.4
		2	3.2	1.9	4.7
		3	0.5	1.6	1.2
		4	0	1.3	1.0
^{90}Sr		5 – 20	0	0.5	0.7
	Guantang	1	97.9	80.3	73.4
		2	2.0	18.2	22.2
		3	0.1	1.5	3.0
		4	0	0	1.4
		5 – 20	0	0	0
	shenzhen	1	73.5	57.9	52.7
		2	20.7	38.6	41.8
		3	4.1	2.8	4.1
		4	1.7	0.7	1.3
		5 – 20	0	0	0.1

3.2 Absorption and Distribution of ^{137}Cs and ^{90}Sr in rice tissues under the different contamination levels

From the results shown in Table 4, it is evidenced that the absorbed ^{137}Cs and ^{90}Sr is distributed throughout the plant. The absorbed ^{137}Cs distributed almost uniformly in the plant with a higher concentration in leaves, stems and husks, and a lower concentration in the seeds. In contrast to ^{137}Cs, distribution of the absorbed ^{90}Sr in rice plant varied greatly. Most of ^{90}Sr accumulated in the stem and little in seeds consistent with the fact that Cs has a higher degree of mobility than Sr in plants ([25]).

The transfer factor (TF) of ^{137}Cs and ^{90}Sr from soil to a rice tissue is defined as

$$TF = \frac{\text{Activity concentration in fresh plant tissue (Bq g}^{-1}\text{)}}{\text{Activity concentration in dry soil (Bq cm}^{-2}\text{)}}$$

Activity of ^{137}Cs and ^{90}Sr per unit surface area of soil is used because, based on the above result, all ^{137}Cs and ^{90}Sr will remain in a thin layer of topsoil. The TFs for different rice tissues are given in Table 4. It is noted that the uptake for both ^{137}Cs and ^{90}Sr by the rice plants is less dependent on the soil surface contamination up to a concentration of a few tens of Bq cm^{-2}. At these concentrations, only very little

^{137}Cs or ^{90}Sr reaches the roots of the plants, which are growing in soil down to 18cm depth, so the main absorption was through the surface of the stem immersed in 3-4cm irrigated water. At higher concentrations, the radioactivities inside the plant build up accordingly while the TFs do not vary significantly.

Table 4. Concentration of ^{137}Cs and ^{90}Sr in fresh rice tissues and the corresponding transfer factors.

Nuclide	Surface Activity in Soil* (Bq cm^{-2})	Concentration in Fresh Rice Tissue (Bq g^{-1})				Transfer Factors (cm^2 g^{-1})			
		Stem	Leaf	Husk	Seed	Stem	Leaf	Husk	Seed
^{137}Cs	2.26	1.37±0.15	1.60±0.20	2.31±0.45	0.40±0.09	0.61±0.07	0.71±0.09	1.02±0.20	0.18±0.04
	22.6	1.55±0.20	1.69±0.51	4.46±0.47	0.57±0.11	0.07±0.009	0.08±0.02	0.20±0.02	0.03±0.005
	226	3.68±0.46	5.02±0.75	6.03±0.66	2.19±0.30	0.02±0.002	0.02±0.003	0.03±0.003	0.01±0.001
	2260	142±15	155±21	117±16	71.8±8.5	0.06±0.007	0.07±0.009	0.05±0.007	0.03±0.004
^{90}Sr	2.26	4.65±0.22	1.52±1.05	0.99±0.46	0.04±0.00	2.05±0.12	0.67±0.50	0.44±0.25	0.020±0.006
	22.6	5.42±1.08	3.23±1.56	1.41±0.23	0.05±0.01	0.24±0.08	0.14±0.08	0.06±0.00	0.002±0.001
	226	40.68±9.11	12.19±6.31	1.44±0.12	1.28±0.32	0.18±0.07	0.05±0.02	0.006±0.01	0.005±0.001
	2260	452.0±86.5	35.25±11.5	11.75±8.30	4.57±1.76	0.20±0.06	0.02±0.00	0.005±0.01	0.002±0.001

* Soil from Guantang district

It is noted in the previous section (Table 3) that, compared with the same site, ^{90}Sr can leach further down the surface soil than ^{137}Cs, which implies that for the same surface contamination of ^{137}Cs and ^{90}Sr, more ^{90}Sr can reach the deeper root and be absorbed by plants. Meanwhile, distribution coefficient(Kd) value of Cs in soils is always one order of magnitude higher than of Sr, which means that Cs is more strongly bound to soil particles than Sr ([4]). This effect is apparent by comparing the difference in TFs for the two high contamination (226 and 2260 Bq cm^{-2}) for both ^{137}Cs and ^{90}Sr. Because of this change in absorption pathway with contamination levels, great care has to be taken when applying the TFs.

3.3 Absorption of ^{137}Cs and ^{90}Sr during different growth stages of the rice

A surface activity of 2.26 Bq cm^{-2} of ^{137}Cs and ^{90}Sr was used in this study. After irrigating the rice plants at different growth stages with ^{137}Cs and ^{90}Sr solution, samples were all taken at harvest time. Measurement results are given in Table 5.

It is well known that, during the seeding stage, growth is fast when compared to the anthesis stage. Anthesis stage is a relatively stable period in the growth. In milk stages, the plants already fall into shape and the root systems are flourishing. Moreover, since the stem of the plants was immersed in the irrigation water during seeding and anthesis stages, the absorption of radionuclide in these stages was mainly from the irrigation water by the stem. During milk stage, the water was dried out, the absorption was mainly by root from the topsoil, where most of the radionuclides was found because of its low leaching rate.

The result in Table 5 shows that the highest accumulation of ^{137}Cs and ^{90}Sr occur in the plants when they were contaminated during the seeding stage, while the lowest accumulation occurs during the last period – milk stage. There is no reasonable explanation for the curious result at milk stage of irrigation water containing ^{137}Cs. A possible reason would be that stems and leaves were directly contaminated during the application of radioactive water.

Table 5. Concentration of ^{137}Cs and ^{90}Sr in fresh rice tissues that were contaminated at different growing stages and their corresponding transfer factors.

Nuclide	Contamination Time	Concentration in Fresh Rice Tissue (Bq g^{-1})				Transfer Factors (cm^2 g^{-1})			
		Stem	Leaf	Husk	Seed	Stem	Leaf	Husk	Seed
^{137}Cs	Seeding stage	1.37±0.15	1.60±0.20	2.31±0.45	0.40±0.09	0.61±0.07	0.71±0.09	1.02±0.20	0.18±0.04
	Anthesis stage	0.60±0.08	0.64±0.11	1.12±0.33	0.19±0.02	0.27±0.04	0.28±0.05	0.50±0.15	0.08±0.01
	Milk stage	8.17±0.92	10.94±2.1	8.35±1.0	0.94±0.14	3.62±0.41	4.84±0.91	3.70±0.45	0.42±0.06
^{90}Sr	Seeding stage	4.65±0.22	1.52±1.05	0.99±0.46	0.04±0.00	2.05±0.12	0.67±0.50	0.44±0.25	0.020±0.006
	Anthesis stage	4.07±0.87	1.48±0.31	1.39±0.22	0.02±0.00	1.80±0.09	0.65±0.07	0.62±0.02	0.009±0.001
	Milk stage	2.50±0.64	0.88±0.05	0.90±0.07	0.01±0.00	1.11±0.07	0.39±0.03	0.40±0.00	0.004±0.001

* Soil from Guantang district
Original contamination level was 2.26 Bq cm^{-2}

3.4 Absorption of ^{137}Cs and ^{90}Sr in rice grown in different soils

Table 6 shows the ^{137}Cs and ^{90}Sr activities and TF in rice tissues grown in five different soils with a contamination level of 2.26 Bq cm^{-2} ^{137}Cs and ^{90}Sr during the seeding stage. Samples were taken at harvest time.

Table 6. Concentration of ^{137}Cs and ^{90}Sr in fresh rice tissues that were grown on different soils.

Nuclide	Soil	Concentration in Fresh Rice Tissue* (Bq g^{-1})				Transfer Factors (cm^2 g^{-1})			
		Stem	Leaf	Husk	Seed	Stem	Leaf	Husk	Seed
^{137}Cs	Guantang	1.37+0.15	1.60±0.20	2.31±0.45	0.40±0.09	0.61±0.07	0.71±0.09	1.02±0.20	0.18±0.04
	Futing	3.00±0.42	3.09±0.52	4.20±0.51	0.94±0.12	1.33±0.19	1.37±0.23	1.86±0.23	0.42±0.05
	Changchuanba	0.71±0.08	0.76±0.10	0.96±0.17	0.18±0.03	0.31±0.04	0.34±0.04	0.42±0.08	0.08±0.01
	Shenzhen	7.92±0.68	15.5±2.03	14.8±1.62	3.85±0.43	3.50±0.30	6.84±0.90	6.54±0.72	1.70±0.19
	Huiyang	2.04±0.19	2.10±0.34	4.34±0.60	1.20±0.09	0.90±0.08	0.93±0.15	1.92±0.27	0.53±0.04
^{90}Sr	Guantang	4.65±0.22	1.52±1.05	0.99±0.46	0.04±0.00	2.05±0.12	0.67±0.50	0.44±0.25	0.02±0.00
	Shenzhen	25.04±7.81	9.87±1.25	3.19±1.06	0.31±0.09	11.08±1.81	4.37±0.11	1.41±0.34	0.14±0.07

* Original contamination level was 2.26 Bq cm^{-2} at harvest

There are significant differences in the absorption of ^{137}Cs and ^{90}Sr in plants grown in different types of soil. In general, rice cultured in Daya Bay soils has a higher ability to accumulate ^{137}Cs and ^{90}Sr. The soil characteristics, especially pH, cation exchange capacity and organic matter in soil, affect the absorption of ^{137}Cs and ^{90}Sr into the plants. As shown in Table 1, pH value in Daya Bay soil is around 5.4 to 5.7 and cation exchange capacity is only about 2.7, both are smaller than in Qinshan soil. The high cation exchange capacity of Qinshan soil means that it can retain H^+ and also fix Cs^+ and Sr^{++} ions, thus increasing the pH and reducing the ^{137}Cs and ^{90}Sr transfer to plants. Moreover, the higher potassium content in Qinshan soil also inhibits the absorption of ^{137}Cs due to chemical similarity between K and Cs ([10], [16]).

3.5 Distribution of ^{137}Cs in rice tissues at different growth stages

In this experiment, ^{137}Cs solutions were added during the seeding stage and samples were taken at different growth stages of the plants. Measurement results, shown in Table 7, show that there is a re-distribution of ^{137}Cs among tissues at various growth stages. At early stages, when the plants were small and only leaves were found, the concentration of ^{137}Cs in the stems and leaves were fairly high. As the plants grew bigger and seeds were formed, the radionuclides were dispersed and the concentration became smaller. Although there is no similar study for comparison, in general, it is well known that the activities found in leaves are the highest and that in the seeds are the lowest (Fan, 1998; Wang, et al, 1998).

Table 7. Concentration of ^{137}Cs in fresh rice tissues at different growing stages.

Growing Stages	Days after Contamination	Concentration in Fresh Rice Tissue (Bq g^{-1})		
		Stem	Leaf	Husk & Seed
Maximum Tiller Number	30	246	380	--
Panicle Formation	40	105	247	--
	50	131	172	--
Anthesis	60	129	186	130
	65	114	139	97
	70	139	181	119
Milk	80	73	90	65
	87	86	104	67

* Original contamination level is 2.26 kBq cm^{-2} Soil from Guantang district.

4. CONCLUSION

From this study, the soil of Qinshan NPP were found to be less favorable for ^{137}Cs and ^{90}Sr transfer to rice plants grown in the soils than that of Daya Bay NPP. It verify once again that the soil H^+ concentration or organic matter affect the absorption of ^{137}Cs and ^{90}Sr into the plant([8]). To compare the results of the transfer factors, some data for rice, grains and crops reported by various organizations are summarized in Table 8, in which it show that the transfer from soil to rice seed is less for Sr than for Cs. The reault is similar with our this study. However, since these transfer factors in Table 8 are referred to as the ratio of the activity of plant to that of soil both in units of Bq kg^{-1} dry weight, and the data we got in this study are based on the water-soil-plant pathway for the water-cultured rice plant and referred to as the ratio of Bq g^{-1} in fresh plant to Bq cm^{-2} in dry surface soil. There is no firm conclusion of comparing could be made.

The distribution of ^{137}Cs and ^{90}Sr in rice plant can be summarized that the stem and leaf accumulate most of ^{137}Cs, and the stem concentrates most of ^{90}Sr. In general, the seeds carry the least concentration when compared to other tissues. Though people do not eat the rest of the rice plants except the seeds, the stems and leaves are commonly used to feed farm animals such as pigs and cattles in China, therefore the transfer route rice → farm animals → men can be a significant pathway.

The uptake of ^{137}Cs and ^{90}Sr at different growth stages of the rice plants was studied. Results show that the most amount of ^{137}Cs and ^{90}Sr can be absorbed if contamination was happened during the seeding stage. Therefore, from an economic point of view, the loss of crops is the least when a contamination occurs during the milk stage of rice plants.

Table 8 Soil-Plant transfer factors (Dry Weight) for rice, grains and crops published by other organization

Organization	Parts of plants	Nuclides	
		Cesium	Strontium
IUR	Rice	5.0×10^{-3}	---
IAEA	Edible parts of crops	3.0×10^{-2}	3.0×10^{-1}
NCRP	Edible parts of crops	1.5×10^{-5}- 0.29	1.6×10^{-3} – 1.7
NRPB	Edible platns	3.8×10^{-2} – 2.9×10^{-1}	1.7×10^{-2} – 3.2
US DOE	Vegetables, fruits, grains	1.1×10^{-2}	2.9×10^{-1}
US NRC	Edible parts of crops	1.0×10^{-2}	1.7×10^{-2}
CEC	Grain	6×10^{-3}	2×10^{-2}
Taiwan RMC	Rice seed	0.03 – 0.188	0.005 – 0.088
Japan RWMC	Rice seed	4×10^{-2} – 6×10^{-1}	5×10^{-3} – 3×10^{-2}

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