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ABSORPTION OF GAMMA-EMITTING FISSION PRODUCTS AND ACTIVATION PRODUCTS BY RICE UNDER FLOODED AND UNFLOODED CONDITIONS FROM TWO TROPICAL SOILS

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KEY WORDS

Activation products Antimony Black soils Cerium Cesium Cobalt Concentration ratio Fission products Flooding Iron Laterites Manganese Rice Ruthenium Uptake Zinc

SUMMARY

The absorption of gamma-emitting fission products ^{106}Ru, ^{125}Sb, ^{137}Cs and ^{144}Ce and activation products ^{59}Fe, ^{58}Co, ^{54}Mn and ^{65}Zn by rice plants grown on two contrasting tropical soils, namely, a blak soil (pellustert) and a laterite (oxisol), and the effects of flooding were studied under controlled conditions. Results indicated greater uptake of ^{106}Ru and ^{125}Sb from the black soil than from the laterite. In contrast, the uptake of ^{144}Ce and ^{137}Cs was greater in the laterite than in the black soil. Flooding treatment enhanced the uptake of all these fission products by rice plants in the laterite soil whereas this effect was observed only for ^{125}Sb and ^{137}Cs in the black soil.

The plant uptake of activation products from the two soil types showed maximum accumulation of ^{65}Zn followed by ^{54}Mn, ^{59}Fe and ^{58}Co in both soil types. Besides, uptake of these nuclides was greater from the laterite soil than from the black soil. Flooding treatment for rice while showing a reduction of ^{59}Fe uptake, showed an increase in plant uptake of ^{58}Co, ^{54}Mn and ^{65}Zn in both soil types.

INTRODUCTION

The present investigation was undertaken to elucidate the physicochemical and edaphic factors governing the transfer of longlived gamma-emitting fission products ^{106}Ru, ^{125}Sb, ^{144}Ce and ^{137}Cs and activation products ^{59}Fe, ^{58}Co, ^{54}Mn and ^{65}Zn from tropical black soils and laterites to rice plants. The influence of flooded water regime normal for rice cultivation on the plant uptake of these radionuclides from these two soils was examined. These studies formed a part of our programme of investigations[1,2,3,6,7,8,9,13] on uptake of radionuclides by crop plants from typical Indian soils aimed at evolution of guidelines for reducing radioactive contamination of crop plants.

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MATERIALS AND METHODS

The radionuclides used in the present investigation are shown in Table 1. The selection of soils was based on the location of the different nuclear installations in India and accordingly it was restricted to two major groups, namely, the black soils group (pellusterts, chromusterts and pelluderts) and the laterite soils group (plinthudults and oxisols) which represent two of the principal soil groups of India. Thus, two soils, representing the black and laterite soil types, namely, a medium black soil from Trombay and other, a laterite soil from Ratnagiri, Maharashtra were selected and bulk samples of surface (0–20 cm) soils were collected and brought to the laboratory for use in the present studies. The soil characteristics are shown in Table 2.

Five kg lots of surface soils passed through a 2 mm sieve were filled in glazed porcelain pots and were maintained at field capacity moisture status for a period of 8 days. At the end of 8 days, 5.0 μci of each of the fission products and 10.0 μci of each of the activation products in suitable volume of distilled water were applied to the soil surface to simulate conditions of contamination through irrigation waters and/or deposition of soluble global fallout. Each treatment was replicated thrice. One week after germination 12 seeds of rice (*Oryza sativa* L. var. D 622) were sown. The plants were thinned to four per pot one week after germination. Five cm of standing water was maintained in pots after 15 days growth in the flooding treatment. The control (non-flooded) pots were maintained at field capacity moisture status. The experiments were carried out in a greenhouse (35 ± 2°C temperature and 60 ± 2% relative humidity). Rice plants were harvested after 6 weeks growth in experiments with fission products and after 11 weeks (at flowering) in experiments with activation products. They were then dried at 90°C to constant weight and taken up for radioassay.

All the radionuclides were assayed through gamma-ray spectrometry using a well type 7.5 cm × 7.5 cm NaI (Tl) crystal integral line assembly and a Nuclear Data 512-Channel pulse height analyser attached to an oscilloscope and a computer readout typewriter. The following photopeaks of the individual nuclides were used for quantitative estimation: 513 keV for ^{106}Ru, 427 keV for ^{125}Sb, 134 keV for ^{144}Ce and 662 keV for ^{137}Cs for the fission products; 1290 keV for ^{59}Fe, 800 keV for ^{58}Co, 840 keV for ^{54}Mn and 1110 keV for ^{65}Zn for the activation products. Since each plant tissue sample contained only one radionuclide, no interference from other nuclides occurred during radioassay. Controls for plants were run to ascertain the absence of the radionuclides (below detectable limits). The radioassay data were corrected for detector background and processed to compute per cent uptake and concentration ratios.

RESULTS AND DISCUSSION

A. *Fission products*

The dry matter yield of rice plants grown in fission products contaminated black and laterite soils under non-flooded and flooded conditions are presented in Table 3. Data in Table 3 indicate that, in general, flooding had no significant influence on the yield of dry matter in the two soils examined. Thus, the yield data indicate the absence of carbohydrate dilution effects on radionuclide uptake by rice plants.

Data on the plant uptake of ^{106}Ru, ^{125}Sb, ^{144}Ce and ^{137}Cs by rice grown under non-flooded as well as flooded regimes in black and laterite soils are reported in Table 4. Data in Table 4 show that ^{137}Cs accumulation by non-flooded rice was much greater than that of the other gamma emitters in the black as well as the

Table 1. Radionuclides used in the present investigation

S. No.	Radio-nuclide	Chemical form	Specific activity
1.	^{106}Ru	Nitrosyl ruthenium dinitro complex	Carrier free
2.	^{125}Sb	Antimony trichloride and Antimony oxychloride	Carrier free
3.	^{144}Ce	Cerous (III) chloride	Carrier free
4.	^{137}Cs	Cesium chloride	Carrier free
5.	^{59}Fe	Ferric chloride	2.3 Ci/g Fe
6.	^{58}Co	Cobalt (II) chloride	Carrier free
7.	^{54}Mn	Manganous chloride	Carrier free
8.	^{65}Zn	Zinc chloride	363 mCi/g Zn

Table 2. Physicochemical characteristics of experimental soils

Soil characteristics	Black clay loam Maharashtra (Pellustert)	Laterite Maharashtra (Oxisol)
Clay mineral type	2:1*	1:1**
pH (1:2:5)	8.0	5.80
Moisture equivalent (%)	35.00	30.00
Total soluble salts (mmhos/cm)	0.52	1.11
Cation exchange capacity (meq%)	40.50	11.50
Exchangeable calcium (meq%)	22.10	3.86
Exchangeable potassium (meq%)	0.09	0.15
Organic carbon (%)	0.78	0.90
Available micronutrients		
Fe (ppm)	16.20	5.80
Mn (ppm)	64.50	93.60
Zn (ppm)	2.38	0.38
Texture		
Coarse sand %	3.70	9.80
Fine sand %	49.80	32.20
Silt %	24.50	25.00
Clay %	22.00	33.00

* Montmorillonite

** Kaolinite

Table 3. Dry matter yields (grams) of rice plants grown in ^{106}Ru, ^{125}Sb, ^{144}Ce and ^{137}Cs contaminated black and laterite soils. Duration of plant growth: 6 weeks

Treatment	^{106}Ru	^{125}Sb	^{144}Ce	^{137}Cs
Black soil				
Control*	1.00	1.11	1.15	1.02
Flooding (5 cm)	0.90	0.89	1.14	1.14
LSD ($p = 0.05$)	NS	NS	NS	NS
Laterite soil				
Control*	1.44	1.87	1.95	1.57
Flooding (5 cm)	1.65	1.43	1.65	1.79
LSD ($p = 0.05$)	NS	0.39	NS	NS

* Field capacity moisture status.

Table 4. Effect of flooding on the uptake of ^{106}Ru, ^{125}Sb, ^{144}Ce and ^{137}Cs by rice plants grown on a black and laterite soil. Duration of plant growth: 6 weeks

Treatment	Radionuclide uptake (% of added) ($\times 10^{-3}$)				Radionuclide concentration ratio + ($\times 10^{-3}$)			
	^{106}Ru	^{125}Sb	^{144}Ce	^{137}Cs	^{106}Ru	^{125}Sb	^{144}Ce	^{137}Cs
Black soil								
Control*	3.5	6.7	1.1	13.0	26.9	45.2	7.2	95.8
Flooding (5 cm)	3.0	13.2	0.8	35.5	24.9	114.3	5.1	241.9
LSD ($p = 0.05$)	NS	6.4	NS	13.5	NS	56.8	NS	62.2
Laterite soil								
Control*	1.7	2.2	7.1	67.3	8.8	8.8	27.3	321.3
Flooding (5 cm)	6.1	5.3	10.2	149.9	26.1	28.4	45.0	626.0
LSD ($p = 0.05$)	2.6	1.4	2.0	51.3	7.7	12.5	5.4	149.1

* Field capacity moisture status.

$^{+}$ Concentration ratio $= \frac{\text{Radionuclide content/g plant shoot}}{\text{Radionuclide content/g soil.}}$

laterite soil. Further, the uptake of ^{137}Cs and ^{144}Ce by plants growing in the laterite was considerably greater than that in the black soil. Earlier studies[10,23] had indicated that ^{137}Cs and ^{144}Ce enter plants considerably more freely from tropical laterite and acidic soils and the present data concur with the earlier findings. The data in Table 4 also indicate greater availability of ^{106}Ru and ^{125}Sb to plants in the black soil than in the laterite. While previous work has indicated that ^{106}Ru and ^{125}Sb absorption by plants varies widely between soil types, their uptake had been shown to be relatively greater from soils of high pH and exchangeable calcium status[18,22,24].

Data (Table 4) on the influence of flooding treatment on the uptake of radionuclides by rice show significant increase in ^{137}Cs and ^{125}Sb concentrations in aerial tissues of the rice plant in both soil types. While no effects of flooding were observed on the uptake of ^{106}Ru and ^{144}Ce in the tropical black soil, significantly higher plant shoot concentrations of these nuclides were obtained in the laterite soil. The greatest increase on flooding was obtained in the ^{137}Cs concentration of aerial tissues which amounted to 95 to 152 per cent of the control in the laterite and black soils, respectively. These results confirm earlier findings[14,15,16,17,29] which have suggested that entry through shoot-base of ^{137}Cs, ^{144}Ce and ^{106}Ru present in the column of standing water is, in the main, responsible for the enhanced accumulation of these nuclides in rice shoots under the flooded regime. In addition to shoot-base entry, the presence of a large number of surface roots which are likely to deplete the surface contaminated zone to a greater extent may contribute to the higher accumulation of these nuclides in the rice plants. Besides, changes in the chemical characteristics of the flooded soil[26] and the predominance of ammonium ion under submerged condition which could lead to reduction in the absorption of ^{137}Cs by soil are other factors likely to result in enhanced accumulation of ^{137}Cs by the rice plant[16].

While no previous reports are available in literature on the comparative uptake of ^{125}Sb by plants under non-flooded and flooded regimes, it is likely that the entry through the shoot base and absorption by surface roots may account for the observed greater accumulation of this nuclide under flooding treatment.

The observed lack of influence of flooded soil regime on the concentration of ^{106}Ru in the rice plants grown in the black soil may be attributed to the rapid movement of ^{106}Ru from the top contaminated zone to the lower layers in this alkaline soil[6]. The removal of ^{106}Ru from the surface layers to lower zones in the black soil may have resulted in the major fraction of ^{106}Ru becoming inaccessible to the predominantly surface roots of the rice plant; this factor is likely to offset any increase in the uptake of ^{106}Ru through shoot-base entry under flooded regime. In the laterite soil where flooding treatment resulted in marked enhancement of ^{106}Ru uptake, ^{106}Ru movement from the surface contaminated zone to the lower layers was at a very much slower rate[6]. Consequently, significant quantities of ^{106}Ru may be present in the top layers of the flooded soil and remain available for absorption by rice through the shoot-base.

As discussed above, under arable conditions relatively greater amounts of ^{144}Ce are accumulated in plants from the acidic laterite as compared to the

Table 5. Dry matter yields (grams) of rice plants grown in ^{59}Fe, ^{58}Co, ^{54}Mn and ^{65}Zn contaminated black and laterite soils. Duration of plant growth: 11 weeks

Treatment	^{59}Fe	^{58}Co	^{54}Mn	^{65}Zn
Black soil				
Control*	2.14	2.18	1.44	2.01
Flooding (5 cm)	2.34	2.28	2.10	2.17
LSD ($p = 0.05$)	NS	NS	NS	NS
Laterite soil				
Control*	7.33	6.90	8.60	7.07
Flooding (5 cm)	6.00	6.17	5.00	5.23
LSD ($p = 0.05$)	NS	NS	1.50	NS

* Field capacity moisture status.

alkaline black soil. Our present results (Table 4) demonstrate that the flooded regime further enhances the greater uptake of this radionuclide by rice plant from the laterite; no significant effects of flooding were obtained in the blak soil.

B. *Activation products*

Table 5 reports the dry matter yields of rice plants grown in black and laterite soils contaminated with activation products under non-flooded and flooded conditions. Similar to the situation in the experiments with fission products (Table 3), the yield data presented in Table 5 indicate, in general, the lack of carbohydrate dilution effects on the uptake of activation products by rice plants.

Data on the effects of non-flooding as well as flooding treatments on the plant uptake of ^{59}Fe, ^{58}Co, ^{54}Mn and ^{65}Zn by rice grown in black and laterite soils reported in Table 6 indicate that the concentration of ^{65}Zn in the rice plants was the highest followed by ^{54}Mn, ^{59}Fe and ^{58}Co. The plants accumulated greater amounts of these radionuclides from the laterite soils than black soils. It has been reported[25] that iron, cobalt, manganese and zinc tend to react in soils in a similar fashion being most soluble under acid conditions and precipitating as hydroxides under alkaline conditions. Besides, adsorption of these nuclides is greater in the 2 : 1 layer type of clay minerals which are present in the black soil compared to the 1 : 1 layer type present in the laterite soil (Table 1)[28]. These edaphic factors appear to be, in the main, responsible for the greater uptake of the activation products from the laterite soil.

Data in Table 5 further indicate that while flooding resulted in considerable reduction of ^{59}Fe concentration in the rice plant, there was an enhancement in plant uptake of ^{58}Co, ^{54}Mn and ^{65}Zn in both soil types. The reduction of ^{59}Fe on flooding was 51.6 and 32.6 per cent of controls in the black and laterite soils, respectively. Iron utilizing power of rice is reported to decrease under flooded conditions and the change in the iron-uptake power of the rice plant corresponding to the change in moisture condition is likely to be due to the increase in the oxidative power of the roots adaptively with the increase in soil moisture, and this is not beneficial for iron-uptake in a flooded soil[19]. Further, though there is increase in the solubility of iron on flooding a soil, reduction in the iron content of shoots of the rice plant may occur presumably due to the iron being oxidised and precipitated on or around the rice roots at pH values of more than 7.0[20].

The ^{58}Co concentration of the rice plant due to flooding was 10 times higher than that in the non-flooded controls in the black soil and nearly five times higher than that in controls in the laterite soil (Table 6). Kubota *et al.*[12] reported that high moisture could increase the cobalt levels in soil solution and consequently also in plants. In other studies, the absorption of ^{60}Co by rice plants has been attributed more to direct entry from irrigation water than its uptake from soil[15]. Experiments on the mobility of surface-deposited ^{58}Co in soil columns[6] indicated that ^{58}Co is not transported downwards to any appreciable extent in black and laterite soils when leached with simulated rain or irrigation waters. The present findings are, therefore, compatible with direct entry of ^{60}Co by absorption through the shoot-base as well as the possible greater absorption of ^{58}Co retained in the top layers of the soil by the highly proliferated surface roots of flooded rice.

Table 6. Effect of flooding on the uptake of ^{59}Fe, ^{58}Co, ^{54}Mn and ^{65}Zn by rice plants grown on a black and laterite soil. Duration of plant growth: 11 weeks

Treatment	Radionuclide uptake (% of added) ($\times 10^{-3}$)				Radionuclide concentration ratio ($\times 10^{-3}$)			
	^{59}Fe	^{58}Co	^{54}Mn	^{65}Zn	^{59}Fe	^{58}Co	^{54}Mn	^{65}Zn
Black soil								
Control*	3.4	0.4	23.0	174.2	31.0	4.0	314.7	1765.0
Flooding (5 cm)	1.7	5.4	138.1	378.9	15.0	39.7	1286.3	2890.0
LSD ($p = 0.05$)	1.3	0.5	85.3	144.7	8.0	6.6	356.4	968.3
Laterite soil								
Control*	16.7	6.0	219.0	1375.7	43.3	16.0	517.0	4853.7
Flooding (5 cm)	8.7	23.0	947.7	2031.3	29.0	75.0	3808.0	7756.3
LSD ($p = 0.05$	4.5	7.9	442.8	346.4	7.8	27.9	1626.7	1253.4

* Field capacity moisture status

Flooding treatment resulted an increase of ^{54}Mn content in the rice plant amounting to four times and more than seven times that in the non-flooded controls in the black and laterite soils respectively. It is well documented that under flooded conditions there is considerable reduction of the higher oxides of manganese resulting in the release of manganese into the soil solution and ultimate greater accumulation in the rice plants[5,11,20,21,26,30]. Besides, surface applied ^{54}Mn is largely retained in the top soil and is not subject to downward movement in the soil profile even on leaching with high amounts of rain or irrigation waters[6]; this situation is also likely to contribute to enhanced absorption of ^{54}Mn by flooded rice which has a well-developed surface root system.

Flooding significantly increased the ^{65}Zn content in the rice plant, the ^{65}Zn concentration ratio under flooding being 1.6 times that in the control in both the soil types. Though Giordano and Mortvedt[11] reported much greater zinc uptake under flooded condition than under arable soil condition, studies of Tiller and Wasserman[27] indicated that flooding marginally increases available zinc in the soil. Previous work[4] has demonstrated severe zinc deficiency in flooded soils attributing it to the change in soil pH. A decrease in the solubility of zinc under continuous soil submergence has been reported by Ponnamperuma[21]. Thus, though variable effects on the uptake of zinc by rice plants were reported by the above authors, the present investigation shows significant increase in zinc uptake by the rice plants in both black and laterite soil types.

In summary, results from the present study with rice plants grown in two tropical soils indicated greater uptake of ^{137}Cs, ^{144}Ce, ^{59}Fe, ^{58}Co, ^{54}Mn and ^{65}Zn from the laterite soils than from the black soils. However, the uptake of ^{106}Ru and ^{125}Sb was greater in the black soil than in the laterite. The flooding treatment normal for lowland rice cultivation enhanced the plant uptake of all fission products in the laterite soil whereas this effect was observed only for ^{125}Sb and ^{137}Cs in the black soil. In the case of activation products, the flooding treatment for rice while enhancing the plant uptake of ^{58}Co, ^{54}Mn and ^{65}Zn, resulted in a reduction of ^{59}Fe uptake.

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