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Consolidation of Radionuclides in Uranium Tailings at Jaduguda (Jharkhand): A Case Study

LAL SINGH, PRAFULLA SONI AND V.N.JHA[1]
Ecology & Environment Division, Forest Research Institute, Dehradun 248001, India
[1] *Health Physics Unit, Uranum Corporation of India Limited, Jaduguda, Jharkhand, India*
Corresponding author; Email: sonip@icfre.org

ABSTRACT

Low-grade uranium deposits discovered in Singhbhum region (Jharkhand) during early sixties have been mined and processed in several areas such as Jaduguda, Bhatin and Narwapahar since 1968. The mill tailings at Jaduguda were covered with 30 cm. thick soil layer. It is required to consolidate the radioactivity by raising plant species which do not have any socioeconomic relevance in the area and have very low uptake of radionuclides. Seven native plant species were selected for experimental trials. Distribution and concentration of U(Nat.) was evaluated in tailings pond areas at different depths in soil and tailings, and uptake of U(Nat.) by the selected plant species was estimated. The concentration of U(Nat.) followed the order: tailings > soil cover on tailings> roots > shoots. The results show that the species examined are appropriate for consolidation of radionuclide in Uranium tailings as compared to naturally occurring species which have higher radionuclide uptake and accumulation.

Key Words: U(Nat.), Concentration Ratio, Tailings, Uptake, Reclaimation, Accumulation

INTRODUCTION

India's nuclear programme had its beginning, and is still sustained, from the depths of the arid tribal belt of mineral-rich Jaduguda in Singhbhum district (Jharkhand). This is the principal and almost only major source of Uranium in India, giving it complete independence in nuclear fuel fabrication for its 10 existing nuclear reactors. Jaduguda (22^0 30' N latitude and 85^0 40' E longitude) lies 24 km from Tatanagar station. The area around the mines is mountainous. Two more mines at Bhatin and Narwapahar are also located in this area (Figure 1).

Most Uranium mines in the world usually produce low grade ores containing 0.1 to 0.3% U_3O_8. Mines in India have still lower grades of ore. Thus, the Uranium industry generates large quantities of waste. The ore excavated from Jaduguda mines is remarkably low grade, with an average 0.06% uranium content. Uranium recovery from the ore at Jaduguda involves a number of steps including change in physicochemical charac-teristics of the bulk ore and steps like oxidation, leaching, ion-exchange, and precipitation as magnesium diuranate (MDU). Though major portion of Uranium present in the ore is extracted, a quantitatively small fraction remains un-extracted and is discharged with the tailings. This fraction may be either soluble hexavalent complexes of Uranium or insoluble tetravalent form. The smelter in Jaduguda processes about 1000 Mg ore per day and therefore, the mill-tailings after Uranium removal reach 300,000 Mg per year. These are discharged into tailing ponds by pipeline in the liquid form (Jha et al 2005). Even radiological exposure in the tailing ponds is negligible as the waste had very low level of radioactivity. However, to avoid any long-term consequences, these tailings have been covered with 30 cm thick layer of soil to consolidate the radioactivity under the ground. This is to avoid any gaseous emission from tailing ponds and to avoid the direct exposure of tailings to cattle or human beings living in the vicinity.

To ensure sustainable consolidation of radioactivity in these tailings, the migration of the un-extracted fraction of uranium from tailings pile to the plants was investigated. Plant species which do not find any use by the local people for fuel, fodder, food or other requirements were selected for growing on the soil-covered tailings and the uptake of uranium by these terrestrial plants was evaluated. Concentration ratios from substrate to plants were determined for different plant species under both *in situ* (natural) and *ex situ* (experimental) conditions. The study was undertaken in the tailing pond area of Uranium Corporation of India Limited (UCIL) at Jaduguda (Figure 1).

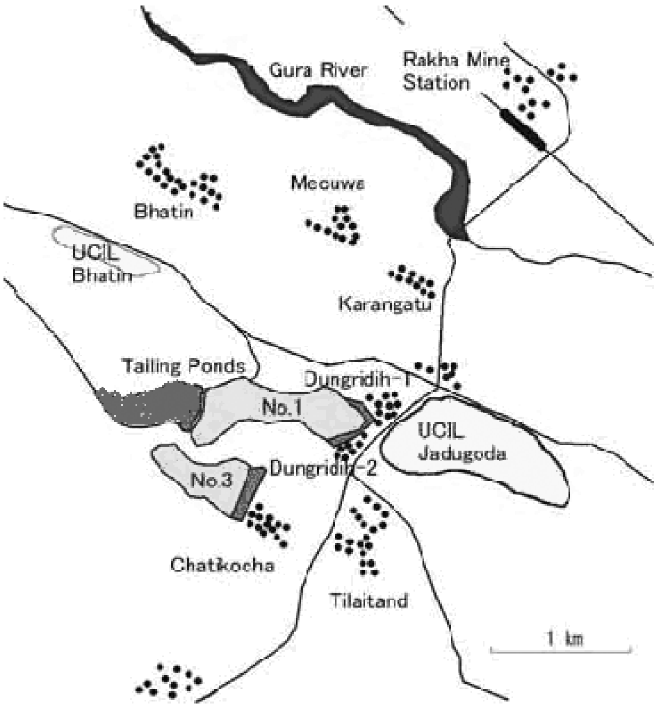

Figure 1.Location of Uranium mines, villages and tailing ponds at Jaduguda

MATERIAL AND METHODS

The work presented here is part of an on-going revegetation program of the filled up tailings pond-2 at Jaduguda. Seven plant species suitable for revegetation were selected after surveying five tribal villages surrounding the tailing pond on the basis of their growth form, low crown cover, shallow root system, non-edible nature, non-domestic use and conservation value (Table 1).

Seeds of the selected plant species were sown over the soil-covered tailings pond for the study of radionuclide uptake (*in situ* trials). Selected species were grown also under laboratory conditions (*ex situ* trials) in 200 -L containers in the Health Physics Unit (H.P.U.), Jaduguda, for comparing the uptake of uranium by these species in natural and experimental conditions. The concentration ratios of the radionuclide in roots and shoots of the selected species was computed.

Samples of tailings and tailings-covered soil were collected respectively from tailings pond and from experimental containers at different depth. Plant samples of selected species (both root and shoot) from revegetated tailing pond area as well as experimental containers were collected in two years (I^{st} age group plants and II^{nd} age group plants). The samples were collected during February, 2006 to March 2008. Five replicates were collected at random from each site.

Plants were separated into root and shoot. All the parts were thoroughly washed in running water for the removal of adhering soil particles, and then with double distilled water. Samples were air- dried and fresh weight of each plant part was recorded. Then the samples were oven-dried at 110^0C for 48 hours, cooled and dry weight was recorded.

Table 1. Selected plant species for revegetation trials.

Plant species	Family	Common name	Habit
Saccharum spontaneum Linn.	Poaceae	Kans	Grass
Imperata cylindrica (Linn.) P.Beauv.	Poaceae	Phoola	Grass
Dodonaea viscosa (Linn.) Jacq.	Sapindaceae	Wild Mehandi	Shrub
Colebrookea oppositifolia J.E.Smith	Lamiaceae	Binda	Shrub
Pogostemon benghalense (Burm.f.) O.Kuntze	Lamiaceae	Phangla, Jul-lata	Herb
Furcraea foetida Haw.	Agavaceae	Furcaria	Shrub
Jatropha gossypifolia Linn.	Euphorbiaceae	ed Jatropha	Shrub

After removal of organic matter with mixture of nitric and perchloric acid, samples were leached repeatedly with 8N HNO_3. Chemically separated uranium is fused with a fusion mixture (NaF -Na_2 CO_3) and subjected to UV radiation in a Flourimeter. A known quantity of leached sample was evaporated to near dryness and 1 mL of conc.H_2SO_4 was added and evaporated till white fumes disappear. Sample was refluxed with 20 mL 0.25 N H_2SO_4 filtered and transferred to separating funnel. After cooling, 10 mL of alamine in benzene was added (2 % alamine in 98% Benzene). The separating funnel was shaken for 1 minute and aqueous layer drained out. The organic layer (0.1 mL) was transferred on platinum planchet. 250 mg of NaF-Na_2 CO3 (15:85) fusion mixture was added and fused at 800 ^oC for 3 minutes, cooled and fluorescence intensity measured in Fluorimeter at 3650 A ^{o} and 5546 A^o (Kolthoff and Elving 1962). Standard (1 ug mL^{-1}) and blank were processed simultaneously and uranium estimated from the readings on the standard.

Data on soil (0-15, 15-30 cm) and tailing (30-45 cm) samples collected from two experimental sites in two consecutive years was subjected to factorial analysis (three-way ANOVA) using statistical pack-Gen Stat 32 version5. The CD (critical difference) was calculated using Schaeffe's method (Schaeff 1959, Snedecor 1956).

RESULTS

The soil uranium concentration differs significantly with depth. It increases with depth (Table 2). Its concentration in 31-45 cm layer was nearly ten times more than that in the upper layers. It seems to be in abundance in soil beyond 45cm.The uranium concentration in soil and tailing did not differ between the sites (Table 2). No significant change occurred in soil uranium concentrations over the two years of study.

The species differed significantly in respect of the uranium concentration in both their shoots and roots. Among the seven species, *Pogostemon benghalense* had the maximum amount of uranium in its roots (37.55 Bq kg^{-1}) while *Jatropha gossypifolia* had the lowest amount (2.69 Bq kg^{-1}) (Table 3). All species showed an increase in uranium level after one year and had higher uranium concentration in the tailing pond site (Table 3). The data show a significant difference between sites, age and species. The concentration of uranium is 46.14% higher in plants grown in the tailing pond (22.5 Bq kg^{-1}) than in experimental containers (15.4 Bq kg^{-1}) (Table 3).The level of uranium in roots increased significantly (by about 19.3%) in the second year of study to 20.6 Bq kg^{-1} (Table 3). The combined effect of age and sites shows that at both the sites, uranium concentration increased in the second year in plant roots. During both years the uranium concentration in roots of selected plant species was higher in the tailing pond than in experimental containers.

Table 2. Uranium concentration (Bq kg^{-1}) in soil and tailings of experimental containers and tailing pond.

Year	Depth (cm)			Mean
	0-15	16-30	31-45	
1st year				
Exp. Containers	62.50	77.50	65.00	67.50
Tailing pond	112.50	140.00	1080.00	445.00
Mean	*87.50*	*110.00*	*572.50*	*255.00*
2nd Year				
Exp. Containers	60.00	77.50	1092.50	410.00
Tailing pond	110.00	140.00	1067.50	440.00
Mean	*85.00*	*107.00*	*1080.00*	*425.00*
Mean of depth	86.25	108.75	826.25	340.62
Means Exp. containers 240.00				
Tailing pond 442.50				
CD				
Depth 263.25	Exp. Area 215.00	Year 215.00		
Dept*Exp. Area 372.25	Depth*Year 372.25			
Exp. Area*Year 304.00	Depth*Exp. Area*Year 526.5			

The effect of the three factors on uranium accumulation in roots, leads to the similar conclusion as discussed above. The species *Pogostemon benghalense* shows higher accumulation values of uranium in root and *Jatropha gossypifolia* shows the lowest accumulation values as compared to other species in both age groups both under *in situ* and *ex situ* trials.

Saccharum spontaneum shoots had the maximum (9.36 Bq kg^{-1}) and *Jatropha gossypifolia* shoots had the lowest uranium concentration (1.41 Bq kg^{-1}) (Table 4). The uranium concentration in plants differed signifi-

Table 3. Concentration of Uranium (Bq kg^{-1}) in roots of selected species in both *ex situ* and *in situ* experiments over two years.

Age and Sites	*Colebrookea*	*Dodonaea*	*Furcraea*	Species *Imperata*	*Jatropha*	*Pogostemon*	*Saccharum*	Mean	Mean of site
1 Year									
Exp. Container	24.125	4.975	3.125	28.250	3.625	13.650	12.525	12.896	Exp. Container =
Tailing pond	34.725	7.975	0.335	36.150	0.738	59.425	12.275	21.660	15.40
Mean	29.425	6.475	1.730	32.200	2.181	36.538	12.48	17.278	
2 Years									
Exp. Container	28.950	13.350	12.175	29.325	4.275	15.875	21.375	17.904	Tailing pond =
Tailing pond	35.975	8.625	3.475	35.625	2.125	61.250	16.375	23.350	22.50
Mean	32.463	10.988	7.825	32.475	3.200	38.563	18.875	20.627	
Mean	30.944	8.731	4.777	32.338	2.691	37.550	15.637	18.953	

CD Year 0.2265 Site 0.2265 Species 0.4237 Year*Site 0.3203 Year*Species 0.5991 Site*Species 0.5991 Year*Site*Species 0.8473

Table 4. Concentration of Uranium (Bq/kg^{-1}) in shoots of selected species under *ex situ* and *in situ* experimental trials in both ages

Age and Sites	*Colebrookea*	*Dodonaea*	*Furcraea*	Species *Imperata*	*Jatropha*	*Pogostemon*	*Saccharum*	Mean	Mean of site
1 year									
Exp. Container	6.075	3.125	1.575	2.075	2.125	6.475	8.975	4.346	Exp. Container =
Tailing pond	7.425	2.725	0.025	11.0725	0.166	6.175	7.300	5.077	5.836
Mean	6.750	2.925	0.800	6.900	1.146	6.325	8.137	4.712	
2 years									
Exp. Container	8.125	5.625	6.625	10.625	3.125	7.525	9.625	7.325	Tailing pond =
Tailing pond	8.150	3.475	1.075	12.425	0.225	12.525	11.525	7.057	6.067
Mean	8.138	4.550	3.850	11.525	1.675	10.025	10.575	7.191	
Mean	7.444	3.738	2.325	9.213	1.410	8.175	9.356	5.951	

CD Year 0.0687 Site 0.0687 Species 0.1285 Age*Site 0.0971 Age*Species 0.1817 Site*Species 0.1817 Age*Site*Species 0.2570

cantly between the two sites (Table 4). It was significantly lower in experimental containers (5.84 Bq kg^{-1}) than in the tailing pond (6.07Bq kg^{-1}). During the second year, the mean plant concentration increased by 52.6% (from 4.7 Bq kg^{-1} to 7.2 Bq kg^{-1}).

The uranium concentration increased in all the species during the second year. Shoots of almost all the species contained lesser amount of uranium in experimental containers.

DISCUSSION

Uranium is regarded as the heaviest trace element found in nature and occurs (Krick-Ottmer 1969) in numerous minerals. Uranium was reported to be the most frequent radionuclide contaminant in ground water and surface soils in more than 50% of DOE facilities involved in nuclear operations (Riley et al. 1992). Generally the tailings are covered with a relatively impermeable layer (hydraulic conductivity of 10^{-9} to 10^{-8} m s^{-1}) waste rocks, overburden, clay and soil cover. In many countries the total thickness of the cover material is of the order of 1 to 2 metres. This may, however, be site specific but should aim at reducing the gamma radiation to 0.2 Gy.h^{-1} above background and radon emanation rate to 0.74 Bq.m^{-2} s^{-1} (IAEA 1987) or as approved by the regulatory body. For the low grade ores mined and processed at Jaduguda, it has been theoretically and experimentally evaluated that a cover of 30 cm soil may reduce the gamma radiation to acceptable levels, while the thickness of limiting radon emanation to natural background level is about 75 cm (Jha et al. 2001).Uranium concentration in plants grown on tailings is mostly affected by the concentration of uranium in the soil (Stephen et al. 1998). In the present study, after covering the soil (30 cm), the concentration of natural uranium in soil was very low. It was also revealed that factor-depths have marked influence on the soil uranium and the variation among three depths was highly significant. Sarangi and Kundu (2004) have reported in whole Nagpur plateau, the concentration of uranium in the depth of 0-50cm to be 0.5 to 4 ppm. It means 12.5 Bq kg^{-1} to 100 Bq kg^{-1} was recorded. In this study also uranium up to the depths of 30cm.was low in quantity and consistent but thereafter beyond 30 cm (tailings) it was high.

According to Jha et al. (2007) the concentration of uranium in the roots of *Typha latifolia* (cattail) varied within a narrow range (57-222.5 Bq kg^{-1}). Shoots of the same species exhibited wide variation (0.05 to 22.75 Bq kg^{-1}). It appears that the transfer of uranium is a gradual process depending on the maturity status of the plant and that the mature plants retain the accumulated uranium in the shoot. However, as reported elsewhere, fruits and seeds have the lowest uranium content. Retention capacity of shoot appears to be poor and is evident from the analysis of other species as well.

In *Capparis* sp. The concentration in roots was significantly greater than in the aboveground parts (384.75 and 22.5 Bq kg^{-1}). In *Cyperus rotundas* highest concentrations among all the samples analyzed were in both roots and aboveground parts (317.25 Bq kg^{-1}) of the species (Jha et al. 2001).

In the present study, the total amount of uranium in roots (Table 3) was in the order:
Pogostemon benghalense>Imperata cylindrica> Colebrookea oppositifolia>Saccharum spontaneum> Dodonaea viscosa> Furcraea foetida>Jatropha gossypifolia.

Jha et al. (2007) observed a relatively low concentration in *Ipomoea carnea* roots (2.25-80.0 Bq kg^{-1}) but higher concentration in leaves and very low in the stems (0.675-9.0 Bq kg^{-1}). Jha at al. (2001) recorded highest concentration of uranium (44.5 to 1002.25 Bq kg^{-1}) in the roots of *Saccharum spontaneum* from tailings pond. The elevated concentration of U(nat) in roots was probably due to the presence of small particle of soil and presumably uranium adhering to roots. It can be further speculated that uranium colloid may have been sorbed on the root surface.

A comparative study on the uptake of uranium by Rumble and Bjugstad (1986), at the uranium mill tailings disposal site and control areas of South Dakota.showed higher concentration of uranium in plants growing on uranium mill tailings than on the control sites. Rickard et al. (1977) suggested that plant uptake of uranium is independent of soil concentration. They reported that the concentration of uranium in soil greater than 200 ppm or 5000 Bq kg^{-1} are toxic to some plants, and uptake of uranium from the soil by plants is in the range 10-15 ppm or 250-375 Bq kg^{-1} on fresh weight basis. In *Lantana camara* (Putus) which was collected at the edge of the tailings pond-I, Jha et al. (2007).reportd the uranium concentration in the roots to vary from 3.9 to 78.0 Bq kg^{-1} and in the shoots, it varied from 5.0 to 18.0 Bq kg^{-1}. An analysis of other less abundant species not normally found in the concerned environment, such as *Ageratum conyzoides* from the nearby hill side (Jha et al. 2007), showed nearly identical and low concentrations in the roots (4.0 Bq kg^{-1}) and shoots (5.75 Bq kg^{-1}). Natural weathering process enhances the sorption capability of substrate due to accumulation of weathered organic matter. More equal distribution of uranium across stem and leaves may lead to a slower return of uranium in the litter and/or long term sequestration of uranium in woody debris.

Miera et al. (1980) while discussing the mobility of elevated levels of uranium in the environment has discussed the difficulty in interpreting the analytical results of root concentration. Whether the sorbed uranium is penetrated inside the root or it is merely present at the outer surface, is however not clear.

The higher concentration ratios for uranium on the mill tailings further suggest that there is some dependence of uptake of these radio nuclides on initial soil concentration. The long-term implication of radionuclide uptake involves the accumulation of uranium on the soil surface from many years of the litter fall and decomposition, and off-site movement by wind and water.

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