# Concentration of radionuclides in uranium tailings and its uptake by plants at Jaduguda, Jharkhand, India

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Mining and processing of uranium ore was started in several parts of eastern Singhbhum, viz. Jaduguda, Bhatin and Narwapahar (Jharkhand) in 1968. Radioactivity in the mine tailings has to be consolidated so that it does not emanate in the atmosphere or enter the food chain. Hence, the area has been covered with 30 cm thick soil cover followed by development of plant species that do not have any socioeconomic relevance in the area. Seven native plant species of forestry origin, viz. Colebrookea oppositifolia, Dodonaea viscosa, Furcraea foetida, Imperata cylindrica, Jatropha gossypifolia, Pogostemon benghalense and Saccharum spontaneum have been selected for experimental trials. Distribution and concentration of radionuclides have been evaluated in a tailing pond at different depths in soil and tailings. Radionuclide uptake in each of the selected plant species has been evaluated and discussed in this article. The highest concentration of radionuclides has been found in tailings > soil cover on tailings > roots of selected plant species > shoots of all the selected species. These results show that among the seven species tried, J. gossypifolia and F. foetida have the lowest uptake (below detectable limits), while S. spontaneum and P. benghalense have comparatively higher uptake. However, radionuclide concentration in all the tried species is significantly low compared to species of natural occurrence which have higher radionuclides uptake and accumulation.

**Keywords:** Accumulation, mining, radionuclides, uptake, uranium tailings.

Most uranium mines in the world usually produce low grade ores containing 0.1–0.3% U<sub>3</sub>O<sub>8</sub>; mines in India having still lower grades. Thus the uranium industry generates large quantities of waste. Almost the entire mined ore comes out as waste after recovery. Waste from uranium mines and mills are in solid, liquid and gaseous forms. Australia has large reserves of uranium with significant deposits in Northern Australia. Abandonment of uranium mines which were developed in the 1950s and 1960s are examples of major environmental insults<sup>1</sup>. Another abandoned uranium mine at Rum Jungle in the

Northern Territory was the costliest rehabilitation programme ever undertaken in an Australian mine (approximately A\$ 20 m). Uranium and copper ore concentrates were produced during 1954-71 from five open-cuts in close proximity. After mine closure, no rehabilitation was required or undertaken<sup>2</sup>. The mining and processing operations resulted in pollution of the nearby East Fitness River with acid mine drainage. This was caused by oxidation of pyrites in the mine waste and led to elevated levels of aluminum iron, copper, zinc, cobalt and manganese. During 1982–86, the overburden heaps and tailings dam were rehabilitated, and the 4 million m<sup>3</sup> of acid water in pits was treated. Pollution resulting from the tailings was removed by excavating the material (approximately 330,000 m<sup>3</sup>) and dumping into one of the existing open-cuts. The floor of the original tailing dam was treated with hydrated lime, covered with 330 mm of topsoil, fertilized and a mixture of tropical grasses (Chloris gayana, Cynodon dactylon, Brachiaria decumbens, Urochloa mozambicensis and Paspalum notatum), legumes (Stylosanthes hamata and S. guianensis), native shrubs and tree species (Acacia and Eucalyptus) were planted.

It is from the depths of the arid tribal belt of mineralrich Singhbhum district of east Bihar, Jaduguda that India's nuclear programme had its beginning. 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. Uranium recovery from the ore at Jaduguda involves a number of steps including change in physicochemical characteristics of the bulk ore and steps like oxidation, leaching, ionexchange and precipitation as MDU (magnesium diuranate). Though major portion of uranium present in the ore is extracted, a fraction though quantitatively small, remains unextracted and is finally discharged with the tailings. The unextracted fraction may be either soluble hexavalent complexes of uranium or its insoluble tetravalent form. Even radiological exposure in the tailing ponds is negligible as the waste has very low level of activity concentration. However to avoid any long-term consequences of exposure to radioactivity, these tailings have been covered with 30 cm of soil to consolidate the radioactivity under the ground. This is to avoid any gaseous

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emission from tailing ponds and to avoid the direct exposure of tailings to cattle or human beings living in the vicinity. Sustainable consolidation of radioactivity in these tailings and migration of the unextracted fraction of uranium from tailings pile to the plants is the subject matter of this article. Species of plants that do not find any use by the local people for their fuel, fodder, food or other requirements have been selected to study uranium uptake and concentration ratios from substrate to plants were determined both under *in situ* and *ex situ* conditions.

#### Material and methods

#### Site description

Jaduguda is located between lat. 22°30′N and long. 85°40′E and the distance from the Tatanagar station to Jaduguda is 24 km in a straight line. The area around the mines is mountainous. Two more mines being operated at Bhatin and Narwapahar are located in this area. These uranium mines are managed by Uranium Corporation of India Limited (UCIL), a government enterprise. The uranium ore from these three mines is processed in a smelter and the grade of uranium ore is remarkably low. The average grade is about 0.06%. 300,000 tonnes per year of mill tailings after uranium extraction is released into tailing ponds by pipeline in liquid form<sup>3</sup>.

The study was undertaken on the tailing pond area of UCIL at Jaduguda Singhbhum district, located in Chota Nagpur Plateau, Jharkhand (Figure 1).

The work presented and discussed here is part of an ongoing project to consolidate the radioactivity in the

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Figure 1. Uranium mines, villages and tailing ponds at Jaduguda experimental trial sites.

tailings using native primary colonizing plant species. Under this programme, the tailing pond has been covered with 30 cm layer of soil. Seeds of the selected plant species were sown over the soil covered tailing pond for the study of radionuclide uptake (in situ trials). The size of the experimental plots was 20 m<sup>2</sup>. An attempt was also made to grow the same species under ex situ conditions in 200 litre drums of 60 cm diameter in the Health Physics Unit (HPU), Jaduguda. These drums were filled with tailings (up to 50 cm) and then covered with 30 cm soil as in the field trials and then planted. These drums were not perforated at the base. Except for sprinkling water immediately after sowing in drums, no further irrigation was applied. Under in situ conditions also no irrigation was applied. These experimental trials have been conducted to compare the uptake of uranium by these species in both ex situ as well as in situ conditions. Evaluation of concentration ratio of radionuclides in experimental species (root and shoot) was an important part of this study. Plant species for revegetation have been selected after surveying five tribal village areas surrounding the tailing pond on the basis of their growth form, low crown cover, shallow root system, non-edible nature, non-domestic use and their conservation value. Seven species have been selected and tried on both sites (Table 1).

Samples of tailings and tailings covered soil were collected from tailings pond and from experimental containers at different depths respectively. Plant samples of selected species (both root and shoot) from revegetated tailing pond area as well as experimental containers have been collected consecutively for two years (1st age group plants and 2nd age group plants). The samples were collected during February 2006. Five replicates were collected at random from each site.

*Uranium estimation:* For natural uranium estimation, collected plants were separated into root and shoot. U(nat.) was evaluated by flourimetric method<sup>4</sup>.

Estimation of  $Po^{210}$ : Estimation of polonium was carried out by the standard method of electrochemical exchange<sup>5</sup>.

 $Ra^{226}$  estimation: Ra<sup>226</sup> is estimated by allowing build up of its daughter Rn<sup>222</sup> for a known period (~2 weeks). The in-built radon was collected in a scintillation cell and counted after equilibrium (between radon and its progeny) is attended<sup>6</sup>.

The data obtained/recorded for the various parameters was analysed by adopting three-way ANOVA. The data was under the influence of three factors, viz. sites, years and depths. The individual factors as well as interaction effects were also explored. For more insights into the different treatments, critical difference (CD) was calculated using Schaeffe's method<sup>7.8</sup>.

Table 1. Selected plant species for revegetation trials

Plant species	Family	Common name	Habit
Saccharum spontaneum	Poaceae	Kans	Grass
Imperata cylindrica	Poaceae	Phoola	Grass
Dodonaea viscosa	Sapindaceae	Wild Mehandi	Shrub
Colebrookea oppositifolia	Lamiaceae	Binda	Shrub
Pogostemon benghalense	Lamiaceae	Phangla, Jul-lata	Shrub
Furcraea foetida	Agavaceae	Furcaria	Shrub
Jatropha gossypifolia	Euphorbiaceae	Red Jatropha	Shrub

Table 2. Growth parameter (height in cm) of various species in different sites

Sources of variation	Mean	Significance	CD	
Sites	One-year-old in situ trials (57.71)	***	19.58	
	Two-year-old in situ trials (86.00)			
	One-year-old ex situ trials (74.00)			
	Two-year-old ex situ trials (104.57)			
Species	Colebrookea oppositifolia (112.00)	***	14.80	
•	Dodonaea viscosa (90.50)			
	Furcraea foetida (56.00)			
	Imperata cylindrica (26.50)			
	Jatropha gossypifolia (110.00)			
	Pogostemon benghalense (127.50)			
	Saccharum spontaneum (41.50)			

Data is average of five replicates. \*\*\*Significant at 0.01% level; CD, Critical difference.

Table 3. Growth parameter (diameter in cm) of various species in different sites

Sources of variation	Mean	Significance	CD
Sites	One-year-old in situ trials (0.86)	**	0.19
	Two-year-old in situ trials (1.09)		
	One-year-old ex situ trials (1.00)		
	Two-year-old ex situ trials (1.17)		
Species	Colebrookea oppositifolia (1.55)	***	0.15
_	Dodonaea viscosa (1.25)		
	Furcraea foetida (1.28)		
	Imperata cylindrica (0.27)		
	Jatropha gossypifolia (1.42)		
	Pogostemon benghalense (1.27)		
	Saccharum spontaneum (0.20)		

Data is average of five replicates. \*\*Significant at 1% level; \*\*\*Significant at 0.01% level; CD, Critical difference

#### Results and discussion

The average canopy height, diameter, basal area and annual increment in height, diameter and basal area of all the tried species in the two experiments were recorded (Tables 2–4). The data on growth performance of these species shows considerable differences, which allowed comparisons of growth within and between both experimental trials. The stratification of canopy appears to have become more or less distinct. The maximum average height was recorded for *Pogostemon benghalense* (127.50 cm), *Colebrookea oppositifolia* (112.00 cm) and *Jatropha gossypifolia* (110.00 cm) followed by *Dodonaea viscosa* (90.50 cm), *Furcraea foetida* (56.00 cm), *Saccharum spontaneum* (41.50 cm) and *Imperata* 

cylindrica (26.50 cm). Height, diameter and basal area were increasing annually in both the experimental trials.

Although planted species formed a major component of plant community in *in situ* experiments and favoured rapid initial establishment of cover, these species were found to be more encouraging in stabilizing experimental area with their ability to regenerate and grow at a faster rate<sup>9</sup>. Work on current reclamation approach<sup>9</sup> at Syncrude oil sand mining in Canada suggests that indigenous species should be preferred as they are better adapted to the harsh local climate. It is easier to construct a stable self-sustaining ecosystem on mine lands by using local natural biological resources<sup>10,11</sup>. The results of the present study (Tables 2–4) using native species are in conformity with these workers.

Table 4. Plant growth (basal area in cm<sup>2</sup>) of various species in different sites

Sources of variation	Mean	Significance	CD
Sites	One-year-old <i>in situ</i> trials (0.74)	***	0.51
	Two-year-old in situ trials (1.15)		
	One-year-old ex situ trials (0.99)		
	Two-year-old ex situ trials (1.37)		
Species	Colebrookea oppositifolia (1.95)	**	0.39
	Dodonaea viscosa (1.25)		
	Furcraea foetida (1.29)		
	Imperata cylindrica (0.05)		
	Jatropha gossypifolia (1.60)		
	Pogostemon benghalense (1.27)		
	Saccharum spontaneum (0.03)		

Data is average of five replicates. \*\*Significant at 1% level; \*\*\*Significant at 0.01% level; CD, Critical difference.

Table 5. Concentration of uranium (Bq kg<sup>-1</sup>) of soil and tailing in both ex situ (experimental containers) and in situ (tailing pond) experimental trials

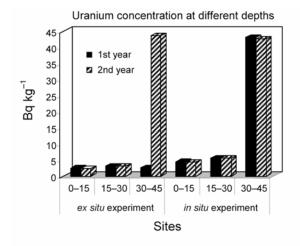
					Mean		
Year		Sites	0–15	15-30	30–45	•	
1st year	Exp.	containers	62.50	77.50	65.00	67.50	
	Tailiı	ng pond	112.50	140.00	1080.00	445.00	
	Mear	1	87.50	110.00	572.50	255.00	
2nd year	Exp.	60.00	77.50	1092.50	410.00		
	Tailii	ng pond	110.00	140.00	1067.50	440.00	
	Mear	1	85.00	107.00	1080.00	425.00	
Mean of depth			86.25	108.75	826.25	340.625	
Mean of exp. site			Exp. containers 24	10.00	Tailing pond 442	.50	
CD	Depth 263.25	63.25 Exp. site 215		Year 215		372.25	
	Depth * Year 372.25	1		1 1			

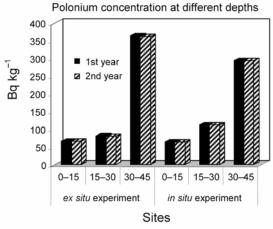
Data is average of five replicates.

Plant samples from the revegetated tailings pond (in situ trial) and from the experimental trials under ex situ conditions were analysed for estimation of natural uranium. Concentrations of U(nat) in different parts of the selected plant species were evaluated. Soil and tailings samples at different depths adhering to the plant roots were also analysed for estimation of concentration ratio. In certain cases, entire above ground parts were analysed and reported as rest parts/shoot. Analytical results of vegetation samples were reported on dry weight basis. The order of selectivity was much more for root and least accumulation was observed in the shoot. Data pertaining to uranium concentration in soil (0-15, 15-30 cm) and tailing (30-45 cm) samples were collected from two sites/experimental areas from three depths and for two consecutive years. Factorial analysis of this data was done using statistical pack – Gen Stat 32 version 5. Soil uranium differs significantly with depths. Depth of the soil is an influencing factor for concentration of uranium. It is observed that uranium increases with increase in depth. Its quantity at the upper surface up to 0-15 and 15-30 cm is very low  $(86.25 \text{ Bq kg}^{-1}, 108.75 \text{ Bq kg}^{-1}; \text{ Table 5})$  but after this depth, it significantly increases. Its quantity in tailings, i.e. 30-45 cm depth is nearly 10 times more than that of the upper strata of soil, i.e. up to the depth of 30 cm. No significant change in soil uranium concentrations over the span of two years has been seen which means that the soil in both the experimental trials over a period of two years has remained the same in respect of soil uranium levels (Table 5). The estimation of uranium concentration in soil and tailings does not show any significant difference between the sites. Uranium concentration in both the sites is statistically the same (Table 5). The mobility of weathered uranium in the soil profile is dependent upon the affinity of the soil for uranium and properties of the soil, such as its acidity or alkalinity and water content. Thus, where soil strongly binds uranium (typically soils high in organic matter) have a high affinity for binding uranium and its release into soil, water should be minimal. Correspondingly mobility is likely to be greater in soils that bind uranium less strongly, which includes soils of semiarid climate where neutral to alkaline soil pH is combined with low organic carbon content<sup>9</sup>. Plant uranium concentration was mostly affected by concentration of uranium in the soil. In the present investigation, since the area is low in organic carbon with pH range of 6-7, mobility of uranium is likely to be greater. Results further reveal that factor-depths have marked influence on soil uranium. The

variation among three depths is highly significant. It is noted that concentration of uranium increases with depth of the soil in both the sites (Table 5).

The analysis of soil polonium shows that it has positive correlation with soil organic carbon and phosphorus. Like uranium, soil polonium also does not show any significant difference between the sites. However, depths show





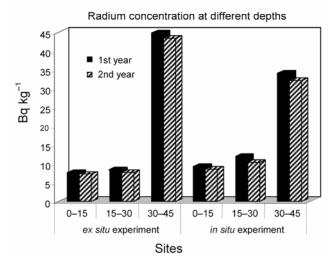


Figure 2. Concentration of radionuclides in soil and tailings.

a marked relationship with the soil polonium. It is seen that the variation among three depths is highly significant, that the soil polonium concentration increases with the depth of the soil in both the sites. Concentration in tailings, i.e. 30–45 cm is nearly five times more (329 Bq kg<sup>-1</sup>) than that of the surface soil layer (65.0 Bq kg<sup>-1</sup>), i.e. up to the depth of 15 cm. It is also noted that variation in soil polonium values over the span of two years does not show any significant difference (Table 6).

Estimation of radium concentration in soil shows that its concentration is similar to other radionuclides discussed here. It is also found that depth is an influencing factor for the concentration of radium. It is seen that the variation between three depths is highly significant. It is noted that the value of soil radium increases with the depth of the soil in both the sites. Its quantity in deeper soil, i.e. 30–45 cm is found to be nearly four times more (38.70 Bq kg<sup>-1</sup>) than that of the upper soil layers (8.20 Bq kg<sup>-1</sup>), i.e. up to 15 cm. It is also observed that variation in soil radium values over a span of two years does not show any significant change after revegetation (Table 7).

# Concentration of natural uranium in roots

The analysis of uranium in roots indicates that there is a significant difference between sites, ages and species. It has been found that the amount of uranium is significantly higher in plants grown under in situ conditions  $(22.505 \text{ Bq kg}^{-1})$  than in ex situ conditions  $(15.40 \text{ Bq kg}^{-1})$ , i.e. 46.14% higher in tailing pond area (Table 3). The average value for both the ages of experimentation shows that the level of uranium in root has increased significantly in the second age level of experimentation (20.627 Bq kg<sup>-1</sup>) in comparison to the first age level (17.278 Bq kg<sup>-1</sup>), i.e. an increase of 19.3% (Table 8). The amount of uranium in roots among the seven species has been found to vary significantly. P. benghalense contains the maximum amount of uranium in its roots (37.55 Bq kg<sup>-1</sup>, dry weight) whereas *J. gossypifolia* contains the least amount of uranium (2.691 Bq kg<sup>-1</sup>) among the seven species. The total amount of uranium in roots is in the order P. benghalense  $(37.55 \text{ Bq kg}^{-1}) > I$ . cylindrica  $(32.338 \text{ Bq kg}^{-1}) > C$ . oppositifolia  $(30.944 \text{ Bq kg}^{-1}) > S$ . spontaneum (15.637 Bq kg<sup>-1</sup>) > D. viscosa (8.731 Bq kg<sup>-1</sup>) > F. foetida  $(4.777 \text{ Bq kg}^{-1}) > J$ . gossypifolia  $(2.691 \text{ Bq kg}^{-1})$ (Table 8). The combined effect of age and sites shows that uranium in both the sites has increased in the second age group in plant roots as compared to the first age. During both ages, the uranium concentration in roots of selected plant species in tailing pond remains higher than experimental containers. Statistical analysis of data from both the sites and species interactions, leads to the conclusion that all the species (in spite of variation among themselves), contain higher uranium when grown on

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Table 6. Concentration of polonium (Bq kg<sup>-1</sup>) of soil and tailing in both ex situ and in situ experimental trials

Year		Sites	0–15	15-30	30–45	Mean	
1st year		Exp. containers	66.0	81.0	366.0	171.0	
		Tailing pond	64.0	112.0	295.0	157.0	
		Mean	65.0	96.0	331.0	164.0	
2nd year		Exp. containers	66.0	80.0	364.0	170.0	
·		Tailing pond	64.0	112.0	294.0	156.0	
		Mean	65.0	96.0	329.0	163.0	
Mean of depth			65.0	96.0	330.0		
Mean of exp. site			Exp. containers 1'	71.0	Tailing pond 157	.0	
CD	Depth 53.7	Exp. site 43.9	Year 6	52.0	Depth * Exp. site 76.0		
	Depth * Year 76	Exp. site * Year 62.0	Depth	* Exp. site * Yea			

Data is average of five replicates. CD, Critical difference.

**Table 7.** Results of analysis of concentration of radium (Bq kg<sup>-1</sup>) of soil and tailing in both ex situ and in situ experimental trials

				Depth (cm)		
Year		Sites	0–15	15–30	30–45	Mean
1st year	Exp.	containers	7.70	8.40	44.90	20.30
	Taili	ing pond	9.30	11.90	34.20	18.50
	Mea	n	8.50	10.20	39.60	19.40
2nd year	Exp.	containers	7.20	7.70	43.40	19.40
	Taili	ing pond	8.50	10.40	32.20	17.10
	Mea	n	7.90	9.10	37.80	18.30
Mean of depth			8.20	9.60	38.70	18.80
Mean of exp. site			Exp. containers 19	9.90	Tailing pond 17.8	30
CD	Depth 5.44	Exp. site 4.44	Year 4	.44	Depth * Exp. site	7.70
	Depth * Year 7.70	Exp. Site * Year 6.28	Depth	* Exp. site * Yea	nr 10.88	

Data is average of five replicates. CD, Critical difference.

**Table 8.** Concentration of uranium (Bq kg<sup>-1</sup>) in roots of selected species in both ex situ and in situ experimental trials in both ages

	Sites				Species					
Age		Colebrookea oppositifolia	Dodonaea viscosa	Furcraea foetida	Imperata cylindrica	Jatropha gossypifolia	Pogostemon benghalense	Saccharum spontaneum	Mean	Mean site
1st age (one year old)	Exp. container	24.125	4.975	3.125	28.250	3.625	13.650	12.525	12.896	Exp. container 15.40
` '	Tailing pond	34.725	7.975	0.335	36.150	0.738	59.425	12.275	21.660	
	Mean	29.425	6.475	1.730	32.200	2.181	36.538	12.48	17.278	
2nd age (two year old)	Exp. container	28.950	13.350	12.175	29.325	4.275	15.875	21.375	17.904	Tailing pond 22.505
( , , , , , , , , , , , , , , , , , , ,	Tailing pond	35.975	8.625	3.475	35.625	2.125	61.250	16.375	23.350	
	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	Age*	= 0.2265 Site = 0.320 Site*Specie			Site = 0.2 Age*Spec	2265 cies = 0.59		ecies = 0.42 e*Species =		

Data is average of five replicates. CD, Critical difference.

Table 9. Concentration of uranium (Bq kg<sup>-1</sup>) in shoot of selected species under ex situ and in situ experimental trials in both ages

					Species	3			-		
Age	Sites	Colebrookea oppositifolia	<i>Dodonaea</i> viscose	Furcraea foetida	Imperata cylindrica	Jatropha gossypifolia	Pogostemon benghalense	Saccharum spontaneum	Mean	Mean site	
1st age (one year old)	Exp. container	6.075	3.125	1.575	2.075	2.125	6.475	8.975	4.346	Exp. container 5.836	
(one year ora)	Tailing pond	7.425	2.725	0.025	11.0725	0.166	6.175	7.300	5.077	2.020	
	Mean	6.750	2.925	0.800	6.900	1.146	6.325	8.137	4.712		
2nd age (two year old)	Exp. container	8.125	5.625	6.625	10.625	3.125	7.525	9.625	7.325	Tailing pond 6.067	
(	Tailing pond	8.150	3.475	1.075	12.425	0.225	12.525	11.525	7.057		
	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	_	ge = 0.0687			Site = 0.0687 Species			ecies = 0.12			
	_	e*Site = 0. e*Site * Sp			Age*Species	= 0.1817	Sit	e*Species =	0.1817		

tailing pond. The interaction values differ highly among themselves. The effect of the three factors on uranium accumulation in roots, leads to similar conclusions as already discussed (Table 8, Figure 3).

### Concentration of natural uranium in shoots

The statistical analysis of uranium levels in the shoots indicates that uranium behaves in different patterns in different species. It has been found that a significant difference in uranium levels exists between two sites; it is significantly low in experimental containers (5.836 Bq kg<sup>-1</sup>) when compared to that in tailing pond (6.067 Bq kg<sup>-1</sup>), i.e. a decrease of 4% (Table 7). The age-wise records show that uranium has increased in the second age of experimentation. It was 4.712 Bq kg<sup>-1</sup> in the first age while in the second age it has reached the level of 7.191 Bq kg<sup>-1</sup>. It can be seen that there has been a rise of 52.61% in uranium level in the second year (Figure 3).

The difference is highly significant (Table 9). Also the seven species have been found to vary significantly in respect of uranium in their shoots. It is found that species *S. spontaneum* possesses the maximum amount of uranium in their shoots (9.356 Bq kg<sup>-1</sup>), while the species *J. gossypifolia* contains the least amount of uranium (1.410 Bq kg<sup>-1</sup>) in their shoots. It is further seen that *S. spontaneum* contains 6.64 times more uranium compared to that of *J. gossypifolia* (Table 9, Figure 3).

The interaction results of age and site show that uranium in experimental containers is very low compared to that of tailing pond area in the first age group but in second age group uranium is higher in experimental containers. The age-wise performance of the species indicates that the uranium in all the species shows an increase in the second age group. The interaction values vary significantly. A look at the site-wise performance of species in respect of uranium in the shoots reveals that almost all the species contain lesser amount of uranium in experimental containers. It does well in the tailing pond. The combination values differ significantly. The overview of three-dimensional results lead to the similar conclusion already mentioned here. The shoot of species *S. spontaneum* contains the highest level of uranium in both the sites during both the years among the seven species (Table 9).

A comparative study of uranium and radium uptake was carried out at the uranium mill tailings disposal site and control areas of South Dakota<sup>10</sup>. Higher concentration of uranium was observed in plants growing on uranium mill tailings than in those from the control sites. Plant uptake of uranium is independent of soil concentration and the concentration of uranium in soil greater than 200 ppm is toxic to some plants, and uptake of uranium from the soil by plants is in the range  $10^{-1}$ – $10^{-2}$  µgg<sup>-1</sup> calculated on fresh weight basis<sup>11</sup>. The higher concentration ratios for uranium on the mill tailings would 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 litter fall and decomposition, and off-site movement by wind and water.

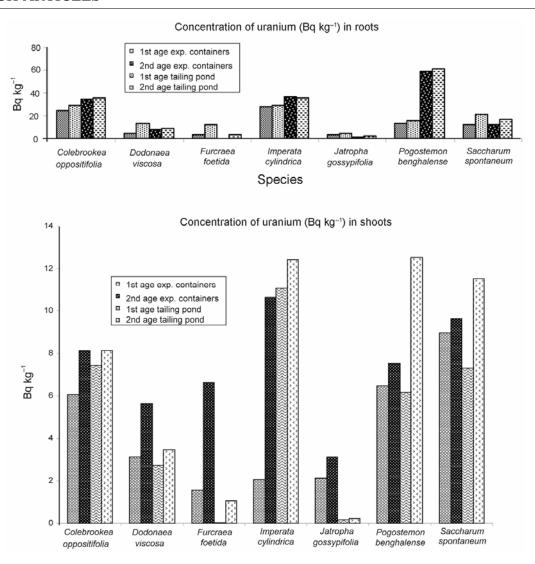


Figure 3. Concentration of uranium in roots and shoots of selected species under ex situ and in situ experimental trials in both ages.

#### Concentration of polonium in roots

It is found from the statistical analysis that there is a significant difference among sites, ages and species. The data reveals that the amount of polonium is significantly higher in tailing pond (20.381 Bq kg<sup>-1</sup>) than the experimental containers (11.14 Bq kg<sup>-1</sup>), i.e. 82.87% higher in tailing pond (Table 10). The averages of both ages of experimentation show that level of polonium in root has increased significantly in the second age of experimentation (15.86 Bq kg<sup>-1</sup>) in comparison to the first age level (15.65 Bq kg<sup>-1</sup>), i.e. a nominal but statistically significant increase. The amounts of polonium in roots have been found to vary significantly. *S. spontaneum* contains the maximum amount of polonium in its roots (36.62 Bq kg<sup>-1</sup>), whereas *J. gossypifolia* contains the least amount of polonium (4.37 Bq kg<sup>-1</sup>) among the seven species. The

combined effects of age and sites show that concentration of polonium in roots in both the sites has increased in the second age group as compared to the first age group. In both the ages, concentration of polonium in roots of selected species in *in situ* experimental site remains higher than *ex situ*. It confirms the overall findings of the sites, i.e. *in situ* (tailing pond) is higher than the *ex situ* (experimental containers). The interaction values differ significantly among themselves.

Viewing the effect of the three factors – age, site and species on polonium concentration in roots leads to the same conclusion as discussed already. *P. benghalense* shows higher values of polonium and *F. foetida* the lowest values compared to other species in both age groups in *in situ* experimental sites. All the species show an increase in polonium level after first age, i.e. in second age (Table 10, Figure 4).

Table 10. Concentration of polonium (Bq kg<sup>-1</sup>) in root of selected species in both ex situ and in situ experimental trials in both ages

				_						
Age	Sites	Colebrookea oppositifolia	Dodonaea viscosa	Furcraea foetida	Imperata cylindrica	Jatropha gossypifolia	Pogostemon benghalense	Saccharum spontaneum	Mean	Mean site
1st age	Exp. container	10.140	18.355	6.455	8.680	0.855	14.235	18.455	11.0250	Exp. con-
(one year old)	Tailing pond	12.235	22.025	8.890	13.340	7.760	23.340	54.445	20.2907	tainer 1.1454
	Mean	11.1875	20.1900	7.6725	11.0100	4.3075	18.7875	36.4500	15.6579	
2nd age	Exp. container	10.165	18.495	7.020	8.715	1.035	14.480	18.950	11.2657	Tailing pond
(two year old)	Tailing pond	12.345	22.145	8.925	13.425	7.855	23.950	54.655	20.4714	20.3811
•	Mean	11.255	20.3200	7.9725	11.0700	4.4450	19.2150	36.8025	15.8686	
	Mean	11.2213	20.2550	7.8225	11.0400	4.3763	19.0013	36.6263	15.7632	
CD		_	01905 = 0.02694 **Species = 0	0.07127	Site = 0. Age*Spe	01905 ecies = 0.05		pecies = 0.03 te*Species =		

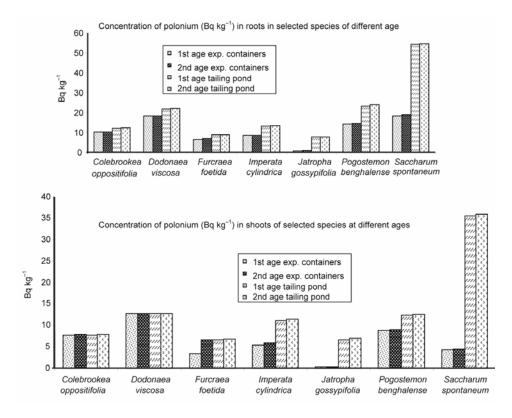


Figure 4. Concentration of polonium in roots and shoots.

## Concentration of polonium in shoots

The statistical analysis for the levels of polonium in the shoots of different plant species reveals that there exists significant difference between the age as well as sites, viz. tailing pond and the experimental containers and the species. The comparison of the average polonium level shows that there is a significant increase in the second age of experimentation.

During the first age, the amount of polonium in shoots is found to be  $9.684~Bq~kg^{-1}$  whereas in the second age it has reached  $10.095~Bq~kg^{-1}$  (Table 8). Further, it shows that

Table 11. Concentration of polonium (Bq kg<sup>-1</sup>) in shoots of selected species in both ex situ and in situ experimental trials in both ages

					Specie	S			-	Mean site
Age	Sites	Colebrookea oppositifolia	Dodonaea viscosa	Furcraea foetida	Imperata cylindrica	Jatropha gossypifolia	Pogostemon benghalense	Saccharum spontaneum	Mean	
1st age	Exp. container	7.760	12.655	3.455	5.455	0.300	8.845	4.255	6.1036	Exp.
(one year old)	Tailing pond	7.760	12.655	6.605	11.130	6.6555	12.445	35.600	13.2643	container
•	Mean	7.760	12.655	5.0300	8.2925	3.4775	10.6450	19.9275	9.6839	6.4200
2nd age	Exp. container	7.880	12.790	6.705	5.900	0.2900	9.045	4.545	6.7364	Tailing pond
(two year old)	Tailing pond	7.855	12.725	6.875	11.425	6.950	12.490	35.855	13.4536	13.3589
	Mean	7.8675	12.7575	6.7900	8.6625	3.6200	10.7675	20.200	10.0950	
	Mean	7.8137	12.7062	5.9100	8.4775	3.5487	10.7062	20.0638	9.8895	
CD	A	Age = 0.030 Age*Site = 0	0.04292		Site = 0.03 Age*Speci	035 es = $0.08030$		s = 0.05678 becies = 0.080	030	
	A	Age*Site*S <sub>1</sub>	pecies $= 0.11$	1356						

the polonium in shoots in tailing pond (13.359 Bq kg<sup>-1</sup>) is significantly higher than that of experimental containers (6.420 Bq kg<sup>-1</sup>). It can be observed that level of polonium in tailing pond is more than twice that of the experimental containers (Table 11). The species differ significantly among themselves in respect of polonium content in shoots. *S. spontaneum* possesses the highest amount of polonium (20.0638 Bq kg<sup>-1</sup>). *J. gossypifolia* lags behind all the species as it is found to contain the lowest amount of polonium (3.5487 Bq kg<sup>-1</sup>). From the interaction results of age and site, it may be seen that polonium in experimental containers as well as tailing pond area show no significant change for second age. The polonium in all the species shows a small but insignificant increase in the second age.

Site-wise performance of species in respect of polonium content in the shoots shows that most of the species contain higher amount of polonium in tailing pond. The combination values differ significantly (Table 11, Figure 4).

#### Concentration of radium in roots

The analysis of radium found in roots reveals that there is significant variation between sites, ages and the species. It is observed that amount of radium is significantly higher in *in situ* (tailing pond) experimental sites (1.07 Bq kg<sup>-1</sup>) than in *ex situ* (experimental containers) (0.82 Bq kg<sup>-1</sup>), i.e. 30.68% higher. The age-wise averages show that level of radium in root has increased significantly in the second age of experimentation (0.97 Bq kg<sup>-1</sup>) in comparison to the first age level (0.92 Bq kg<sup>-1</sup>), i.e. an increase of 5.69% (Table 12, Figure 5).

The amount of radium in the root among the seven species has been found to vary significantly. *F. foetida* possesses the least amount of radium in its roots

(0.15 Bq kg<sup>-1</sup>), whereas *I. cylindrica* contains the maximum radium concentration in roots (1.02 Bq kg<sup>-1</sup>) among the seven species. The joint effects of ages and sites show that radium in both the sites has increased in the second age as compared to the first age. During both the ages radium in roots of selected species in in situ remains higher than ex situ. It conforms the overall findings of the sites, i.e.  $in \ situ > ex \ situ$  (Table 12). From the site-wise performance of species in respect of radium in the roots, it can be inferred that all the species contain higher percentage of radium in tailing pond. The interaction values differ significantly. An overall view on the effect of the three factors leads to the same conclusion as discussed here. F. foetida shows higher values of radium and I. cylindrica the lowest values when compared to other species in both ages in both sites. All the species show a rise in radium level after one age group, i.e. in the second age group. The species possesses higher values of radium in in situ (tailing pond) experimental site (Table 12).

#### Concentration of radium in shoots

The analysis of radium in the shoot reveals that a significant difference in radium exists between two sites. Radium is significantly more in tailing pond (0.225 Bq kg<sup>-1</sup>) than in experimental containers (0.138 Bq kg<sup>-1</sup>), i.e. 63% more in tailing pond compared to experimental containers (Table 13, Figure 5).

The averages of the ages show that amount of radium decreased in the second age of experimentation as compared to the first age although the difference is statistically non-significant. It was  $0.193 \text{ Bq kg}^{-1}$  in the first age while in the second age it reduced to the level of  $0.170 \text{ Bq kg}^{-1}$ .

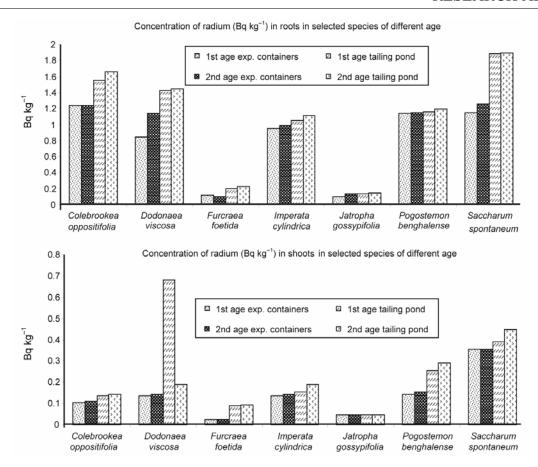


Figure 5. Concentration of radium in roots and shoots.

Table 12. Concentration of radium (Bq kg<sup>-1</sup>) in root of selected species in both ex situ and in situ experimental trials in both ages

Age	Sites	Colebrookea oppositifolia	Dodonaea viscosa	Furcraea foetida	Imperata cylindrica	Jatropha gossypifolia	Pogostemon benghalense	Saccharum spontaneum	Mean	Mean site	
1st age (one year old)	Exp. container Tailing pond	1.235 1.555	0.8450 1.425	0.1150 0.195	0.955 1.045	0.1000 0.1350	1.135 1.155	1.145 1.880	0.7900 1.0557	Exp. con- tainer	
	Mean	1.395	1.135	0.155	1.000	0.1175	1.145	1.5125	0.9229	0.8229	
2nd age	Exp. container	1.235	1.135	0.0950	0.990	0.1350	1.145	1.255	0.8557	Tailing	
(two year old)	Tailing pond	1.655	1.445	0.2250	1.1150	0.1450	1.1900	1.8900	1.0950	pond	
	Mean	1.445	1.2900	0.1600	1.0525	0.1400	1.1675	1.5725	0.9754	1.0754	
	Mean	1.4200	1.2125	0.1575	1.0262	0.1287	1.1562	1.5425	0.9491		
CD		Age = $0.0116$	Age = $0.01168$			Site = $0.01168$			Species = 0.02186		
		Age*Site = 0	.01652		Age*Speci	es = 0.03091		Site*Specie	es = 0.0309	1	
		Age*Site * S	pecies = 0.0	14371							

The recorded interaction values vary significantly. This is in contrast to polonium and uranium where a significant increase has been recorded in the second age. The

amount of radium in shoots of different species has been found to vary significantly. It is found that *S. spontaneum* contains the maximum amount of radium in

Table 13. Concentration of radium (Bq kg<sup>-1</sup>) in shoot of selected species in both ex situ and in situ experimental trials in both ages

		Species								
Age	Sites	Colebrookea oppositifolia	Dodonaea viscosa	Furcraea foetida	Imperata cylindrica	Jatropha gossypifolia	Pogostemon benghalense	Saccharum spontaneum	Mean	Mean site
1st age	Exp. container	0.105	0.135	0.025	0.135	0.045	0.145	0.355	0.135	Exp. container
(one year old)	Tailing pond	0.135	0.680	0.090	0.155	0.045	0.255	0.390	0.250	0.138
	Mean	0.120	0.407	0.057	0.145	0.045	0.200	0.373	0.193	
2nd age	Exp. container	0.110	0.145	0.025	0.145	0.045	0.155	0.355	0.140	Tailing pond
(two year old)	Tailing pond	0.145	0.190	0.095	0.190	0.045	0.290	0.450	0.201	0.225
	Mean	0.128	0.168	0.060	0.168	0.045	0.223	0.403	0.170	
	Mean	0.124	0.287	0.059	0.156	0.045	0.211	0.388	0.181	
CD	Age = $0.0767$			Site = 0.0767 Species				= 0.1434		
	Age * Site = $0.1084$				Age * Species = $0.2029$ Site * S				Species = $0.2029$	
	Age * Site * Species = $0.2869$									

its shoot (0.388 Bq kg<sup>-1</sup>), while J. gossypifolia contains the least amount of radium (0.045 Bq kg<sup>-1</sup>) among all the tried species. It is seen that S. spontaneum holds 8.62 times more radium when compared to J. gossypifolia (Table 13). However, results indicate that all the species have higher amount of radium in the tailing pond when compared to that of experimental containers. Filamentous algae were used for monitoring the Animas radioactivity in the vicinity of Uranium Mills at Colorado, USA. <sup>226</sup>Ra concentration of 148 Bq kg<sup>-1</sup> of algal ash was recorded for uncontaminated streams, whereas concentration as high as  $129.5 \times 10^3$  Bq kg<sup>-1</sup> of algal ash was recorded for the polluted areas of the river, which showed concentration potential of algae of a high order<sup>12</sup> for <sup>226</sup>Ra. A new trend has developed to carry out environmental impact assessment following uranium mill effluent discharge through the study of radioactivity associated with the algal bodies growing in the habitat<sup>12</sup>. From the above analyses of uptake and accumulation of radionuclides in the seven species, it is seen that J. gossypifolia and F. foetida have the lowest uptake and accumulation of radionuclides while S. spontaneum and P. benghalense have highest accumulation of radionuclide substances.

#### **Conclusions**

The data on growth performance of the species in terms of average canopy height, diameter, basal area and annual increment in height, diameter and basal area shows considerable differences, which allowed comparisons of growth within and between both experimental trials. The maximum average height for different species was in the order of P. benghalense > C. oppositifolia > J.

gossypifolia > D. viscosa > F. foetida > S. spontaneum > I. cylindrica.

Results of the present study led to the conclusion that all the seven species tried in the experiments, show either very low concentration or below detective limit for uptake of radionuclides. This is because of the fact that the roots of these species are confined to the top 30 cm layer of soil only and do not actually penetrate the tailings. These results are based on the preliminary investigations of one- and two-year-old plant trials, however further comprehensive studies are required to finally conclude on the basis of estimates of the uptake of radionuclides by plants and their consolidation in the tailings for further two years.

- Bastias, J. G., Retreatment of radioactive gold bearing tailings and rehabilitation of mill and tailings dump sites at Rockhole and Moline, Northern Territory (a personal view). Australian Mining Industry Council Environment Workshop Papers (Adelaide, 21–25 September 1987), Australian Mining Industry Council, Canberra. Proceedings of Conference 'Reclamation, A Global Perspective', Calgary, Alberta, 27–31 August 1989, pp. 319–346.
- Ryan, P., Run jungle mine rehabilitation Northern Territory. J. Soil Conserv., 1987, 43, 18–27; Proceedings of Conference 'Reclamation, A Global Perspective', Calgary, Alberta, 27–31 August 1989.
- Jha, V. N., Sethy, N. K., Sahoo, S. K., Shukla, A. K., Tripathi, R. M. and Khan, A. H., A comparison of radioactivity level in discharge waste and natural sources in uranium mineralized area of Singhbhum, Jharkhand. Proceedings of 27th IARP National Conference of Occupational and Environmental Radiation Protection, BARC, Mumbai, 2005, pp. 284–286.
- Kolthoff, I. M. and Elving, P. J., Treatise of Analytical Chemistry, 1962, vol. 9, part II.
- Figgins, P. E., Radiochemistry of Polonium, National Academy of Science NAS – NS series, 1961, pp. 30–37.
- Raghavayya, M., Iyengar, M. A. R. and Markose, P. M., Estimation of Radium-226 by emanometry. *Bull. Indian Assoc. Radiat. Protect.*, 1980, 3, 11–15.

- Schaeff, H., Analysis of Variance, John Wiley and Sons, New York, 1959.
- 8. Snedecor, G. W., Statistical Method, Iowa State University Press, Iowa, 1956.
- Tony, S. D. and Fung, M. V. P., Current reclamation approach at the syncrude oil sand plants. In Proceedings of the Reclamation: A Global Perspective, Calgary, Alberta II, 1989, pp. 575–578.
- Kilmartin, M. P. and Haigh, M. J., Land reclamation policies and practices. In *Mining and Environment in India* (eds Joshi, S. C. and Bhattacharya, G.), Himalayan Research Group, Nainital, India, 1988, pp. 441–467.
- Masoodi, T. H., Evaluation of role of native plant species in restoration of mined land ecosystem. Ph D thesis, FRI University, Dehradun, 1999.
- 12. Ebbs, S. D., Brady, D. J. and Leon, K. V., Role of uranium speciation in the uptake and translocation of uranium by plant. *J. Exp. Bot.*, 1998, **49**, 1183–1190.
- 13. Rumble, M. A. and Bjugstad, A. J., Uranium and radium concentrations in plant growing on uranium mill tailings in South Dakota. *Reclam. Reveg. Res.*, 1986, **4**, 271–277.

- Rickard, W. H. et al., Ecology of Waste Management Areas: Radioecology of Uranium Pacific Northwest Laboratory Annual Report 1976, Richland, WA, BN WL-2100, 1997, pp. 4.28-4.41.
- 15. Tsivoglue, E. C., Environmental monitoring in the vicinity of uranium mill. Proceedings of IAEA symposium on Radiological Health and Safety in Mining and Milling of Nuclear Materials 1963, Vienna, 1964, vol. 2, pp. 231–245.

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