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Article

Risk assessment (chemical and radiological) due to intake of uranium through the ingestion of drinking water around two proposed uranium mining areas, Jharkhand, India

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ABSTRACT **Uranium is known for both chemical and radiological toxicity. East Singhbhum is known for uranium mining, and radionuclides can be expected in its groundwater. Groundwater was collected around two proposed sites of Bagjata and Banduhurang and analysed for U(nat). The study reveals that the U(nat) varied from <0.5–11.2 and <0.5–27.5 µg.L^{-1} for the Bagjata and Banduhurang mining areas, respectively. The excess lifetime cancer risk due to the consumption of uranium in water was calculated to be in the range of 8.81 × 10^{-6} to 4.34 × 10^{-5} and 3.36 × 10^{-6} to 9.55 × 10^{-5} for the two study areas, which are within the acceptable cancer risk value of 1 × 10^{-4}. However, the risk at a few locations is very close to the threshold value. The chemical risk evaluated by the hazard quotient was found to be within 0.05–0.23 and 0.02–0.6 for the two study areas and did not exceed the limit of 1. Thus, the concentration of U(nat) in the groundwater presently does not pose any serious threat to local people but must be monitored periodically and adequate actions must be taken in the few areas with elevated levels of uranium in the groundwater.**

Keywords: groundwater / India / uranium / risk assessment / hazard quotient / cancer risk

1. Introduction

Radionuclides can come into contact with water in several ways. They may be deposited from the air or may be released to the water from the ground through erosion, seepage, or human activities such as mining. Some radionuclides that reach either groundwater or surface water move with the water. Others deposit on the surrounding soil or rocks. Ingestion of radionuclides through drinking water and food intake accounts for a substantial part of the average radiation doses to various organs of the body and also represents one of the important pathways for long-term health considerations (UNSCEAR, 2000). Radioactive materials occur

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naturally everywhere in the environment (*e.g.*, uranium, thorium and potassium-40). Some sources (*e.g.*, uranium) can be concentrated during extraction by mining and other industrial activities (WHO, 2004).

It is important to investigate the natural radiological situation of the sites and the environment before the commencement of uranium mining to be able to assess the possible radiological impact of these activities in the future. As part of a baseline study, the concentration of radionuclides was determined in the groundwater around two proposed uranium mines in Bagjata and Banduhurang (Giri *et al.*, 2011). In the present study, the chemical and radiological risk due to intake of uranium through the ingestion of drinking water was investigated.

2. Materials and methods

Natural uranium {U(nat)} was evaluated by a fluorometric method (Kolthoff and Elving, 1962). The details of the study area, sampling and analysis are provided elsewhere (Giri *et al.*, 2011).

2.1. Risk from the intake of uranium through ingestion

Uranium is a radionuclide which is known for both its chemical toxicity and radiological toxicity. It has been established that high concentrations of uranium greater than 15 $\mu g.L^{-1}$ in domestic water may present harmful biological effects in humans (WHO, 2008). The main health threat related to uranium use is chemical toxicity (Keith *et al.*, 1999). Like many other heavy metals, uranium is toxic to humans and animals, with the kidney being the target organ (Leggett, 1989). The toxic effects of uranium compounds have been extensively studied in the kidney (Guglielmotti *et al.*, 1989; Kurttio *et al.*, 2002) and bones of laboratory animals (Larivière *et al.*, 2007). The chemical toxicity effects on the human kidney by chronic ingestion of uranium through drinking water in the range of 0.004 to 9 $\mu g.L^{-1}$ per body weight per day may produce interference with kidney functions (Zamora *et al.*, 1998). In a more recent study on humans by Kurttio *et al.* (2005), nephrotoxic effects of uranium in drinking water were found even for low concentrations – without a clear threshold. Most results from uranium studies in drinking water suggest that the safe concentration of uranium in drinking water may be within the range of proposed guideline values of 2-30 $\mu g.L^{-1}$ (Kurttio *et al.*, 2002; WHO, 2008). Because uranium is a predominantly alpha-emitting radionuclide, there is concern about the potential DNA damage if the emitted alpha particles reach the cell nuclei of the body; for instance, through water ingestion. Attempts by cells to repair this damage, if it occurs, may result in repair errors, producing gene mutations or chromosomal aberrations. These effects, when

sufficiently severe, may be manifested as cancer and possibly as developmental malformations in children and developing foetuses (Amakom and Jibiri, 2010). In this view, the assessment of radiological and chemical toxicity is essential. For the radiological toxicity, the excess lifetime cancer risk is estimated, while for the chemical toxicity the hazard quotient is calculated. International regulatory agencies such as the WHO and ICRP do not provide the values for slope factors and reference doses for radionuclides. Moreover, the most widely used values for SFo and RfD are given by US-EPA. So, the US-EPA values were used for the calculations.

2.2. Excess lifetime cancer risk (absolute)

Risk factors for different tissues were based upon the estimated likelihood of inducing fatal malignant disease, nonstochastic changes, or substantial hereditary disorders expressed in live-born descendents. The risk coefficients may be used to estimate the probability of radiogenic cancer mortality or morbidity per unit intake for a given radionuclide for internal exposure or per unit dose for external exposure. Federal guidance report No. 13 provides risk coefficents for specific radionuclides (US-EPA, 1999). The slope factors or risk coefficients are estimated using state-of-the-art methods and models for estimating the risks to health from internal or external exposure. These methods and models take into account a comprehensive compilation, and the age- and gender-specific aspects of radiation risk. The risk coefficient for exposure to a given radionuclide through a given environmental medium is expressed as the probability of radiogenic cancer mortality or morbidity per unit activity inhaled or ingested, for internal exposure, or per unit time-integrated activity concentration in air or soil, for external exposure. The risk coefficients are applicable to either chronic or acute exposure to a radionuclide. For purposes of computing the factors, it was assumed that the concentration of the radionuclide in the environmental medium remains constant and that all persons in the population are exposed to that environmental medium throughout their lifetimes.

A morbidity risk coefficient or slope factor is a comparable estimate of the average total risk of experiencing a radiogenic cancer, whether or not the cancer is fatal. For each of the internal exposure modes, the risk coefficient for a radionuclide includes the contribution to dose from production of decay chain members in the body after intake of the parent radionuclide, regardless of the half-lives of the decay chain members.

The excess lifetime carcinogenic risk can be estimated by multiplying the average daily dose (*ADD*) with the slope factor (*SF*) and the duration of life (75.2 years). In the case of uranium the slope factor is 6.4×10^{-11} (US-EPA, 1989).

In the 1970s, the US Food and Drug Agency (FDA) was the first agency to address the issue, adopting a risk level of 1-in-1-million (10^{-6}) as the incremental cancer risk for carcinogenic residues in foods that was considered to be "essentially zero" (Kelly, 1991). The origin of this "essentially zero" risk level was purely arbitrary. Since then, the 10^{-6} risk level has become commonplace in the regulation and management of environmental contaminants, with the strongest endorsement coming from the US-EPA, which employs 10^{-6} as its primary risk benchmark for "acceptable" exposure to carcinogens within the general population. Although a 1-in-1-million (10^{-6}) cancer risk is the most frequently used risk level for the management of risks posed by environmental (including soil) contamination, many agencies and provinces, including the US-EPA, identify a range of increased cancer incidence risks; generally, from 1-in-10 000 (or 1×10^{-4}) to 1-in-1 000 000 (or 1×10^{-6}) is considered an acceptable risk range depending on the situation and circumstances of exposure (Graham, 1993; Kelly, 1991; Lohner, 1997; Travis *et al.*, 1987; US-EPA, 1991). Thus, in general, the US-EPA considers excess cancer risks that are below about 1 chance in 1 000 000 (1×10^{-6}) to be so small as to be negligible, and risks above 1×10^{-4} to be sufficiently large that some sort of remediation is desirable. The upper boundary of the risk range is not a discrete line at 1×10^{-4}, although the EPA generally uses 1×10^{-4} in making risk management decisions. A specific risk estimate around 10^{-4} may be considered acceptable if justified based on site-specific conditions. Some studies have even reported 10^{-3} as the acceptable level for the radiological risk (Ye-shin *et al.*, 2004).

The basic equation for calculating excess individual lifetime cancer risk is:

$$Risk = ADD \times SF_o \times 27\ 448 \text{ days (75.2 years)}$$

where:

- *Risk* = a unitless probability of an individual developing cancer over a lifetime;
- *ADD* = average daily dose (pCi);
- SF_o = slope factor, expressed in (risk/pCi).

The risk is related to doses but since the US-EPA - integrated risk information system (IRIS) database has provided slope factors (SF_o) in unit activity/risk which is specific for each radionuclide, the same procedure is adopted for the assessment (US-EPA, 1993). The slope factors are given in the units of risk/pCi so the activities are converted from Bq to pCi. The conversion factor used is 1 Bq = 27 pCi.

2.3. Hazard quotient

The EPA uses the "Reference Dose" approach to evaluate chronic (long-term) exposure to systemic toxicants. The reference dose (*RfD*) is defined as "an estimate

(with uncertainty spanning perhaps an order of magnitude) of a daily exposure to the human population (including sensitive subgroups) that is likely to be without appreciable risk of deleterious effects during a lifetime" and is expressed as a mg/kg/day dose. The *RfD* is usually based on the most sensitive known effect; that is, the effect that occurs at the lowest dose. Although some *RfDs* are based on actual human data, they are most often calculated from results obtained in chronic or subchronic animal studies. The basic approach for deriving an *RfD* or *RfC* involves determining a "No-Observed-Adverse-Effect Level (NOAEL)" or "Lowest-Observed-Adverse-Effect Level (LOAEL)" from an appropriate toxicological or epidemiological study and then applying various uncertainty factors and modifying factors to arrive at the *RfD*. *RfDs* can be used to evaluate risks from chronic exposure to systemic toxicants. In the case of uranium also, the values were developed by analysing the biological effects of test animals given relatively large amounts of uranium, then adjusting and normalizing the results to a mg/kg-day basis for humans (US-EPA, 1994a).

The EPA defines an expression of risk called a "hazard quotient" by which the risk of the chemical toxicant may be characterized. This is the ratio of the Average Daily Dose (*ADD*; milligrams per kilogram body weight per day) of a chemical to a reference dose (*RfD*, milligrams per kilogram per day). Hazard quotient values below unity imply that adverse effects are very unlikely to occur. The RfD_o of uranium (0.003) was considered from US-EPA (1993).

$$HQ = \frac{ADD}{RfD}$$

If $HQ > 1.00$, then the *ADD* of a particular metal exceeds the *RfD*, indicating that there is a potential risk associated with that metal.

For the calculation of the excess lifetime cancer risk and hazard quotient, the average water consumption of 1.48 $m^3.y^{-1}$ (4 L/day) by an Indian adult (Dang *et al*., 1994) was used. The average daily dose (*ADD*) was calculated by dividing the intake by the body weight of an average Indian man, *i.e*. 52 kg (Jain *et al*., 1995; Dang *et al*., 1996). The values are different from a Caucasian reference man (body weight = 60 kg and average water consumption = 2 L/day). As the study is from India, the values from the Indian reference were taken for better approximation of the risk.

3. Results and discussion

The overall concentration range of uranium, considering all the seasons, varied from <0.5 μgL^{-1} to 11.2 $\mu g.L^{-1}$ in the Bagjata area, whereas for the Banduhurang area it ranged from <0.5 $\mu g.L^{-1}$ to 27.5 $\mu g.L^{-1}$ (Tab. I). 0.5 $\mu g.L^{-1}$ was the method

TABLE I

Concentration of U(nat) in groundwater of Bagjata and Banduhurang mining areas.

S. No.	Location (Bagjata)	U(nat) in µg.L^{-1}				Location (Banduhurang)	U(nat) in µg.L^{-1}			
		Jun. 06	Sept. 06	Jan. 07	May 07		Jun. 06	Sept. 06	Jan. 07	May 07
1	Bagjata	7.8	<0.5	2.1	2.7	Keoradungri	1.2	<0.5	0.6	<0.5
2	Bhaduya	10.6	1.2	5.2	10.1	Babudungri	7.7	<0.5	<0.5	1.2
3	Phuljhari	2.2	<0.5	1.3	5	Chota Talsa	0.7	3	1.5	2.1
4	Manajhari	0.9	<0.5	3.8	2.3	Kudada	0.6	<0.5	<0.5	0.8
5	Balidungri	3.5	<0.5	0.7	1.3	Matku	0.9	<0.5	<0.5	0.6
6	Bakra	3.7	2.3	1.4	2.1	Dhatkidih	2.4	<0.5	1.6	1.9
7	Katsakra	4.8	3.4	4.1	4.9	Sundernagar	1	<0.5	1.3	0.7
8	Gohala	11.2	9	8.5	7.4	Talsa	3.1	<0.5	2.8	4.3
9	Latia	5.1	4.1	<0.5	3.5	Turamdih	0.7	<0.5	0.9	0.6
10	Mosabani	3.4	1.5	3.1	2.5	Nandup	21.4	<0.5	27.5	20.3
Geomean		**4.26**	**2.84**	**2.61**	**3.49**		**1.82**	**–**	**2.0**	**1.64**

detection limit. Comparison of the concentration range of U(nat) in groundwater with the US-EPA standards (US-EPA, 1994b, 2003) and WHO standards (WHO, 2011) shows that the radionuclide lies within the prescribed limits (30 µg.L^{-1}) for both the study areas. The statistical analysis of the data indicated geometric mean concentrations of U(nat) in the Bagjata and Banduhurang areas were found to be 3.31 µg.L^{-1}and 1.83 µg.L^{-1}, respectively, with geometric standard deviations of 2.0 µg.L^{-1}.

3.1. Excess lifetime cancer risk due to intake of uranium through drinking water

The data was used for calculating the excess lifetime cancer risk assessment by the US-EPA method (US-EPA, 1993). The excess lifetime cancer risk due to the consumption of water was calculated to be in the range of (8.81×10^{-6})-(4.34×10^{-5}) for the Bagjata area, with the highest risk in Gohala village (uranium concentration of 11.2 µg.L^{-1} in June 2006), and in the range of (3.36×10^{-6})-(9.55×10^{-5}) for the Banduhurang area, with the highest risk in Nandup village (uranium concentration of 27.5 µg.L^{-1} in January 2007) (Tab. II). The risk in the study area is within the acceptable excess individual lifetime cancer risk value of 1×10^{-4}. However, the risk in Nandup and Gohala villages is very close to the threshold value of 1×10^{-4} and will require remediation and management of drinking water after the mining

TABLE II

Range of excess lifetime cancer risk due to intake of uranium through drinking water.

Location	Intake[1]	Excess lifetime cancer risk
Bagjata	5.02-24.7	(8.81×10^{-6})-(4.34×10^{-5})
Banduhurang	1.92-62.1	(3.36×10^{-6})-(9.55×10^{-5})

SF_o in Risk/pCi (6.4×10^{-11}). [1] Daily Intake in pCi.

TABLE III

Range of intake and hazard quotient of uranium due to ingestion of groundwater.

Location	Intake[1]	*ADD*[2] (average daily dose)	HQ (hazard quotient)
Bagjata	0.007-0.037	0.00014-0.0007	0.05-0.23
Banduhurang	0.003-0.093	0.00005-0.0018	0.02-0.6

RfD_o in mg/ kg body weight/day (0.003 for uranium). [1] Intake in mg/day. [2] *ADD* in mg/ kg body weight/day.

starts in both the proposed uranium mining areas of Bagjata and Banduhurang. The elevated levels of uranium and the associated high risk may be attributed to the uranium mineralisation of the study area.

3.2. Daily intake estimate of uranium through drinking water and hazard quotient

However, to know the chemical risk associated with the ingestion of uranium through water, the hazard quotient (*HQ*) was estimated in both the study areas. The estimated exposure and hazard quotient due to intake of drinking water to the local population are given in Table III. The hazard quotients suggest that the uranium in the drinking water poses no threat to the local people. The *HQs* of all the locations in both the areas were below the threshold value of 1, as suggested by US-EPA. The *HQ* ranges from 0.05 to 0.23 for the Bagjata area, while for the Banduhurang area the *HQ* ranged from 0.02 to 0.6. The maximum *HQs* were found in Gohala and Nandup villages, respectively.

4. Conclusions

The concentration ranges of U(nat) in the groundwater of both the study areas are within the US-EPA and WHO standards of 30 $\mu g.L^{-1}$. The excess lifetime cancer risk due to the consumption of water in all the locations of both the study areas is within the acceptable excess individual lifetime cancer risk value of 1×10^{-4}. However, as the risk at some locations is very near to the threshold value of 1 chance in 10 000, it may stipulate remediation after mining starts. The elevated

levels of uranium and the associated high risk may be attributed to the uranium mineralisation of the study area. The hazard quotients of all the locations in both the areas were also below the threshold value of 1, as suggested by US-EPA. The study suggests that the concentration of U(nat) in the groundwater presently does not pose any serious threat to local people but needs close investigation in the near future and stringent remedial measures should be adopted in the high risk locations.

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