Environ Monit Assess (2009) 154:15–22
DOI 10.1007/s10661-008-0373-8

Estimation of uranium and radon concentration in some drinking water samples of Upper Siwaliks, India

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Received: 8 September 2007 / Accepted: 5 May 2008 / Published online: 20 June 2008

Abstract Uranium and radon concentration was assessed in water samples taken from hand pumps, natural sources and wells collected from some areas of Upper Siwaliks, Northern India. Fission track registration technique was used to estimate the uranium content of water samples. The uranium concentration in water samples was found to vary from 1.08 ± 0.03 to 19.68 ± 0.12 μg l^{-1}. These values were compared with safe limit values recommended for drinking water. Most of the water samples were found to have uranium concentration below the safe limit of 15 μg l^{-1} (WHO, World Health Organization, *Guidelines for drinking-water quality* (3rd ed.). Geneva, Switzerland: WHO, 2004). The radon estimation in these water samples was made using α-scintillometry to study its correlation with uranium. The radon concentration in these samples was found to vary from 0.87 ± 0.29 to 32.10 ± 1.79 Bq l^{-1}. The recorded values of radon concentration were within the recommended safe limit of 4 to 40 Bq l^{-1} (UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiations, *Sources and effects of ionizing radiation*. New York: United Nations, 1993). No direct correlation was found between uranium concentration and radon concentration in water samples belonging to Upper Siwaliks. The values of uranium and radon concentration in water were compared with that from the adjoining areas of Punjab state, India.

Keywords Uranium · Radon · Fission track · Etched track detector · Alpha Scintillometry

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Introduction

Uranium is present almost in all soils, rocks and waters. Water passing through and over rock and soil formations dissolves many compounds and minerals, including uranium, so varying amounts of it are present in some water sources. It occurs in dispersed state and has a heterogeneous distribution in the earth due to geo-chemical processes, which have slowly recycled the crustal material to and from earth's mantle. High values of uranium in drinking water and foodstuffs may lead to harmful effects in human beings. Their exposure to natural uranium is through food, water and to certain extent from air. According to an estimate (Cothern and Lappenbusch 1983) food contributes about 15% of ingested uranium while drinking water contributes about 85%. The estimation of uranium in water may be significant for

the hydrogeochemical prospection for uranium and for health risk assessments. Uranium has both chemical and radiological toxicity with the two important target organs being the kidneys and lungs (WHO 1998; ATSDR 1990, 1999). Retention of uranium in the kidney has been attributed to the creation of complexes with proteins and phospholipids in the proximal tubules, considered to be the main site of kidney damage. Abbasi (1998) has discussed the possible mechanism for the toxic action of the uranyl ion in the kidney. Uranyl ion circulates in the blood plasma as a relatively inert but acid labile bicarbonate–uranyl complex; this could be filtered into the kidney tubules and uranyl ion is set free by the action of hydrogen ions. The uranyl ion is thought to be librated and concentrated in the tubular lumen as a result of normal tubular action. The ions, however, cause damage to kidney structures which, if sever enough, results in kidney failure. Uranium estimation of water systems of India has been reported by some workers (Singh et al. 1984, 1995, 2001; Ramola et al. 1988; Sarin et al. 1992).

Radon is heaviest noble gas and only gaseous decay product in uranium decay series. It is a colourless, odourless inert gas, which is 7.5 times heavier than air. Hence its presence in drinking water is not felt during its consumption. Radon is a naturally occurring radioactive gas that may cause lung cancer if present in high concentrations. Natural water usually contains dissolved radon due to the presence of ^{226}Ra, a member of natural ^{238}U decay series, in soils and rocks through which the water has filtered. In many situations such as showering, washing clothes and flushing toilets, radon is released from the water and mixes with the indoor air. Thus radon from water contributes to the total inhalation risk associated with radon in indoor air. Although radon in drinking water does not pose a direct health risk (Cross et al. 1985), the main concern is that the levels of radon in indoor air of dwellings can be enhanced partially by radon derived from the water supply (Hess et al. 1985). During recent years, radon monitoring has become a global phenomenon due to its health hazard inside dwellings (ICRP 1987; Mishra and Ramachandran 1997; Nazaroff and Doyle 1985). Radon measurements have been carried out in the recent past (Virk et al. 1998; Virk and Sharma 2000; Virk 1997) in soil gas, groundwater and in air.

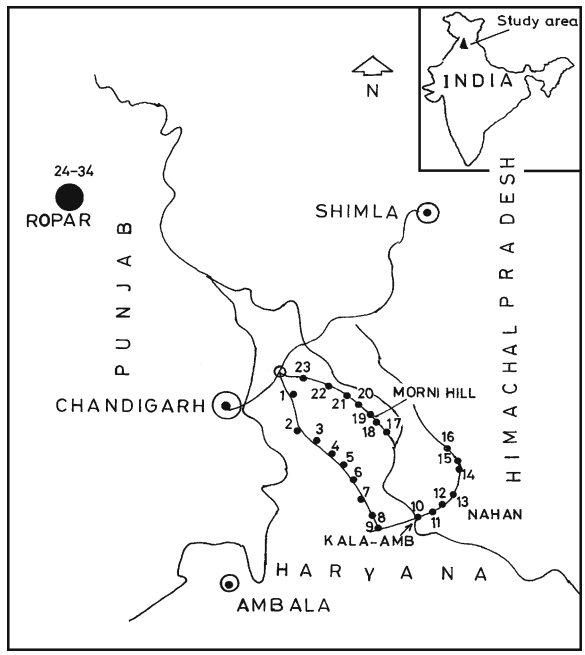

Fig. 1 Map showing the area surveyed with sampling locations

In the present study, concentration of uranium and radon has been assessed in drinking water samples collected from different areas belonging to the Upper Siwaliks of Kala Amb, Nahan and Morni Hills, Northern India. The latitude and longitude of Kala Amb, Nahan and Morni Hills are 30°30′0 N 77°12′0 E, 30.55° N 77.3° E and 30°41′60 N 77°4′60 E respectively. The area surveyed in the present investigations is shown in Fig. 1. The work in this study area has been carried out for the first time. The purpose of this study is to investigate the uranium and radon levels of groundwater being used for drinking as potable water and to determine the health hazards, if any, to the population groups belonging to the Upper Siwaliks of Kala Amb, Nahan and Morni Hills, Northern India and in the adjoining areas of Punjab state.

Geology of the area

The Upper Siwaliks consist lithologically either of very coarse conglomerates, the boulder- conglomerates or massive beds of sand, grit and brown

and red earthy clays. The former occur at the points of emergence of the large rivers – the Ravi, Tavi, Chenab, and Jhelum – and of their chief tributaries, whereas the later occupy the intervening ground. The clays in the upper parts of the series are indistinguishable from the alluvial clays of the Punjab plains into which they pass by an apparently conformable passage upwards. Fossils are numerous in the Upper Siwaliks at some localities. This area appears to have been a Favourite haunt of a highly diversified elephant population, as is evident from the profusion and wide distribution of their skeletal remains. Insicors of *Eliphas*, *Stegodon*, *Mastodon*, their molars, skull plates, madibles maxillae, limb bones etc., are commonly found in the sands and conglomerates. Other fossils are referable to *Bubalus, Bos, Hipopotamus, Rhinocerous, Sus, Equus, Cervus,* Apes, *Gavialis* and numerous *Chelonian* bones (Wadia 1989).

Experimental techniques

Uranium estimation

Fission track registration technique developed by Fleischer and Lovett (1968) and recently revised by Ilic and Durrani (2003) was used for the estimation of uranium in drinking water samples. In this technique, a drop of water of known volume was dried on a circular lexan disk in a dust free atmosphere. A non-volatile constituent of the water was left over the lexan disk in the form of a thin film. It was then covered with another lexan disk to make it a pair. All such pairs were then packed in an aluminium capsule and irradiated with thermal neutrons in the IC-1 position of the CIRUS Reactor at BARC, Trombay; Mumbai. A blank lexan disk was also irradiated along with the sample disks in order to calculate the background. After irradiation the lexan disks were etched in 6.25 N NaOH at 70 °C for 25 min. The whole area of the droplet was manually scanned with the optical microscope at a magnification of ×400. The uranium concentration C (in μg l^{-1}) was determined using the formula (Fleischer and Lovett 1968)

$$C = \mathrm{TM}/(\mathrm{VG}\sigma\mathrm{N}\Phi\mathrm{E})$$

Where N is the Avagadro's number (6.023×10^{23}); T, the total number of tracks formed; V, the volume of 1 drop (ml); M, the atomic weight of uranium isotope ^{238}U; G the geometry factor (assumed unity); Φ, the total thermal neutron dose 2×10^{15}(nvt); σ the fission reaction cross section (4.2×10^{-24}) and E, the etching efficiency factor, taken as unity for lexan plastic.

Radon estimation

Scintillometry technique (Alpha-Scintillometer GBH2002 with Lucas cell) was used for the estimation of radon concentration in ground water and mineral water, Here silver activated zinc sulphide phosphor, ZnS (Ag), was used as a scintillation material. Water samples were collected at ambient temperature and pressure from various sources in some areas belonging to the upper Siwaliks of Kala Amb, Nahan and Morni Hills of Haryana and Himachal Pradesh states in the month of June 2005 and simultaneously in the adjoining areas of Punjab state for comparative study. The volume of water taken in each case is 1 l. Radon sampling was complicated by the fact that the gas easily escapes from water and therefore has to be done without any aeration, which might lead to outgassing. So the water sample should be collected in such a way that there should be no bubbling. Radon gas emanated from radium present in the water or dissolved in it was sucked by pump connected to a radon bubbler with an extraction efficiency of more than 90%. The electronic digital counter records the alpha counts and radon concentration in water is measured by using the calibration constant (10 counts = 1 Bq l^{-1}). The counting was done for 10 minutes. Since the measurement was done on the spot so no time delay was taken in this case. The calibration constant was recommended by the International standards so there was no mathematical formula for it. The detection limit for the Lucas cell used in Alpha-Scintillometer is 0.02 Bq l^{-1}.

Results and discussion

Table 1 reports the results for uranium and radon concentration in drinking water samples collected

Table 1 Values of Uranium and radon concentration in water samples in different villages around the Upper Siwaliks and Punjab, Northern India

S. No.	Location	Source	Uranium concentration ($\mu g\ l^{-1}$)	Radon concentration ($Bq\ l^{-1}$)
Upper Siwaliks				
1	Kharagmangoli	Hand pump	3.85 ± 0.05[a]	5.35 ± 0.73[b]
2	Ramgarh	Tubewell	9.52 ± 0.08	3.06 ± 0.55
3	Billa	Tubewell	6.33 ± 0.07	4.29 ± 0.65
4	Alipur	Hand pump	19.68 ± 0.12	4.39 ± 0.66
5	Batour	Hand pump	11.09 ± 0.09	2.03 ± 0.46
6	Raipur Rani	Tubewell	2.58 ± 0.04	5.48 ± 0.74
7	Pyarewala	Hand pump	12.03 ± 0.10	3.25 ± 0.57
8	Laha	Hand pump	2.27 ± 0.04	3.89 ± 0.62
9	Raomajra	Hand pump	1.11 ± 0.03	6.19 ± 0.78
10	Kala Amb	Tubewell	3.24 ± 0.05	4.07 ± 0.63
11	Devi ka Bagh	Tubewell	3.83 ± 0.05	29.11 ± 1.70
12	Ambwala	Tubewell	6.38 ± 0.07	13.07 ± 1.14
13	Nahan	Tubewell	9.73 ± 0.09	25.34 ± 1.59
14	Do Sadka	Tubewell	3.49 ± 0.05	32.10 ± 1.79
15	Laadu	Natural source	5.13 ± 0.06	3.18 ± 0.56
16	Banethi	Natural source	5.40 ± 0.07	2.62 ± 0.51
17	Kanderan	Natural source	1.14 ± 0.03	21.35 ± 1.46
18	Badyal	Natural source	2.96 ± 0.05	5.44 ± 0.73
19	Morni	Natural source	2.07 ± 0.04	4.52 ± 0.67
20	Tikari	Natural source	1.08 ± 0.03	3.93 ± 0.62
21	Bhoori	Natural source	2.00 ± 0.04	15.30 ± 1.23
22	Mandana	Hand pump	1.95 ± 0.04	0.87 ± 0.29
23	Bearwala	Hand pump	5.33 ± 0.06	2.62 ± 0.51
Ropar (Punjab)				
24	Gharbhaga	Hand pump	9.44 ± 0.09[a]	4.57 ± 0.67[b]
25	Katali	Hand pump	11.40 ± 0.10	5.14 ± 0.71
26	Alampur	Hand pump	3.33 ± 0.05	5.19 ± 0.72
27	Lodhimajra	Hand pump	7.06 ± 0.08	6.06 ± 0.77
28	Lohgarh fide	Hand pump	19.25 ± 0.12	5.75 ± 0.75
29	Rattanpura	Hand pump	7.03 ± 0.08	5.18 ± 0.72
30	Noohan	Hand pump	2.24 ± 0.04	6.69 ± 0.81
31	Daburji	Hand pump	1.93 ± 0.04	5.91 ± 0.76
32	Gonumajra	Hand pump	2.55 ± 0.05	4.67 ± 0.68
33	Ghanoulia	Hand pump	3.59 ± 0.05	4.98 ± 0.70
34	Haripur	Hand pump	6.82 ± 0.07	8.01 ± 0.89

[a]Statistical counting error: $\sigma = \pm(1/\sqrt{N}) \times$ uranium content, where N is number of tracks

[b]Statistical counting error: $1\sigma = \pm(1/\sqrt{N}) \times$ radon concentration, where N is number of α-particles recorded by scintillometer in 1 min

from some areas of Upper Siwaliks, Northern India. The uranium concentration was found to vary from $1.08 \pm 0.03\ \mu g\ l^{-1}$ in Village Tikari (Haryana State) to $19.68 \pm 0.12\ \mu g\ l^{-1}$ in village Alipur (Haryana State) while the radon concentration was found to vary from $0.87 \pm 0.29\ Bq\ l^{-1}$ in village Mandana (Haryana State) to $32.10 \pm 1.79\ Bq\ l^{-1}$ in village DoSadka (Himachal Pradesh State). For Comparative study of the Upper Siwaliks region and adjoining area of Punjab, the values of uranium and radon concentrations of the Ropar district of Punjab State were also reported in Table 1. The uranium concentration in water samples was found to vary from $1.93 \pm 0.04\ \mu g\ l^{-1}$

Table 2 Range of uranium concentration in drinking water worldwide

Sr. No.	Country	Range of uranium concentration ($\mu g\ l^{-1}$)	Average value ($\mu g\ l^{-1}$)	References
1	Ontario, Canada	0.05–4.21	0.40	OMEE 1996 Moss et al. 1983 Moss 1985
2	New York, USA	0.03–0.08	–	Fisenne and Welford 1986
3	USA	–	2.55	USEPA 1990, 1991
4	Argentina	0.04–11.0	1.3	Bomben et al. 1996
5	Japan	–	0.0009	Nozaki 1970
6	Norweign	18% samples had uranium concentration in excess of 20 $\mu g\ l^{-1}$	–	Frengstad et al. 2000
7	New Mexico	> 20	–	Hakonson-Hayes et al. 2002
8	Central Australia	> 20	–	Hostetler et al. 1998
9	Jordan	0.04–1400	2.4	Gedeon et al. 1994, Smith et al. 2000
10	Kuwait	0.02–2.48	–	Bou-Rabee 1995
11	United States	0.01–652	–	Drury et al. 1981, Edgington 1965, Cothern and Lappenbusch 1983
12	South Greenland	0.5–1.0	–	Brown et al. 1983
13	Turkey	0.24–17.65	–	Kumru 1995
14	India	0.08–471.27	–	Talukdar et al. 1983, Bansal et al. 1985, 1988, Singh et al. 1993, 2003
15	India	1.08–19.68	–	Present investigations

in Village Daburji to 19.25 ± 0.12 $\mu g\ l^{-1}$ in village Lohgarh-fide while the radon concentration in water samples was found to vary from 4.57 ± 0.67 Bq l^{-1} in village Gharbhaga to 8.01 ± 0.89 Bq l^{-1} in village Haripur. It can be seen that the uranium and radon content in the drinking water samples varied from source to source and also with location. The health and environmental protection agencies had recommended safe limit of uranium and radon in drinking water for human beings. WHO (2004) had recommended 15 $\mu g\ l^{-1}$ of uranium in water as safe limit, whereas United States EPA (2003) had recently recommended 30 $\mu g\ l^{-1}$ of uranium in water as the safe limit for drinking purpose. However UNSCEAR (2000) recommended safe limit as 9 $\mu g\ l^{-1}$ and ICRP (1979) had recommended the safe limit as 1.9 $\mu g\ l^{-1}$. These levels were set to represent a concentration that does not result in any significant risk to health over the lifetime drinking of water. Results revealed that in about 78% of water samples from upper Siwaliks area and about 72% of water samples from Punjab area, the concentration of uranium lies below the recommended safe limit of 9 $\mu g\ l^{-1}$ (UNSCEAR 2000), whereas 22% drinking water samples from Upper Siwaliks area and about 28% of water samples from Punjab area had shown higher content of uranium and were not fit for the drinking purpose. The measured uranium content in water samples was below the recommended level of WHO (2004) except one sample from Alipur (Upper Siwaliks region) and one sample of Lohgarh-fide (Punjab area). However when compared with USEPA (2003) recommended level of 30 $\mu g\ l^{-1}$, all the analyzed samples from Upper Siwalik region and adjoining area of Punjab revealed uranium values below the safe limit. The uranium concentrations higher than 20 $\mu g\ l^{-1}$ were generally observed in water samples coming from uranium rich zones (Kumar et al. 2006). All the water samples have values less than 20 $\mu g\ l^{-1}$ so these are safe for drinking purposes. The uranium values in drinking waters were also reported by some workers. The value of uranium in drinking water from the United States reported by Drury et al. (1981), Edgington (1965) and Cothern and Lappenbusch (1983) were in the range of 0.01–652 $\mu g\ l^{-1}$, 0.11–640 $\mu g\ l^{-1}$ and 0.02–6.99 $\mu g\ l^{-1}$ respectively. Brown et al. (1983) reported uranium concentrations in water samples

in South Greenland in the range of 0.5–1.0 μg l^{-1}. Kumru (1995) reported uranium concentration in water samples from Turkey in the range of 0.24–17.65 μg l^{-1}. In India Guwahati drinking water samples (Talukdar et al. 1983) were reported to contain 0.08–5.32 μg l^{-1} of uranium. The Aligarh (in UP state of India) tubewell water (Bansal et al. 1985) contained 38.37–471.27 μg l^{-1} of uranium. The range of uranium in domestic Indian waters reported by Bansal et al. (1988) was 0.6–19.2 μg l^{-1}. The drinking water samples from Jhansi and Allahabad cities of UP state of India were reported (Singh et al. 1993) to have uranium content in the range of 0.87– 6.45 μg l^{-1}. Our uranium values in drinking water samples from the study area were more than those reported by Brown et al. (1983) in the water samples of South Greenland, less than the range reported by Bansal et al. (1985) but lie in the range reported by other workers as given in Table 2.

On the other hand the recorded radon concentrations in all the water samples from different areas belonging to the Upper Siwaliks of Kala Amb, Nahan and Morni Hills, Northern India and adjoining areas of Punjab were within the internationally recommended safe limit of 4 to 40 Bg l^{-1}(UNSCEAR 1993) and hence safe for drinking purposes. Variation in radon content in all water samples may be due to different degrees of agitation and variation in meteorological parameters. No regular trend of the variation of radon with source was observed. Our results revealed that there was no direct correlation of radon concentration with uranium content in water samples collected from different areas belonging to the Upper Siwaliks of Kala Amb, Nahan and Morni Hills of Haryana and Himachal Pradesh states and also from the adjoining areas of Punjab state as reported in Ganges waters in UP Himalayas earlier by Singh et al. (1987) also. Variation in fluid velocity may influence radon concentration (Andrews 1977). However detailed investigations will be carried out in the future studies.

Acknowledgements The authors are thankful to the residents of the study area for their cooperation during the fieldwork and lab staff of G.N.D. University, Amritsar especially Mr. Santokh Singh for providing necessary help.

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