Uranium and radon surveys in western Himalaya

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The water samples from mountain springs, streams and river systems in the western Himalaya were collected and analysed in the laboratory for uranium and radon contents. It is observed that the Himalayan river system is conspicuous by its high dissolved uranium and radium concentration. The water samples contain from 0.89 ppb to 63.4 ppb of uranium and from 34 Bq/l to 364 Bq/l of radon. The radon emanation in soil is measured by the track-etch method, emanometry and alpha-logger technique. The daily and long-term variation of radon was monitored in some mineralized zones of HP state with high uranium content in the soil. The maximum values of radon are recorded in Chhinjra, Rameda, Samurkala and Kasol areas of HP.

GEOCHEMICAL investigations for uranium deposits are based on the ability of uranium and its disintegration products, radium and radon, to dissolve in water and migrate together in Himalayan rivers and streams. One of the conspicuous characteristics of the Himalayan river system is its high dissolved uranium concentration, ~2 μg/l, compared to the global average of 0.3 μg/l in river waters. The Ganges and Brahmaputra, together, transport about 1000 tons of dissolved uranium to the estuaries of the Bay of Bengal annually[1,2].

Radon estimation in soil-gas, groundwater and atmosphere is an established technique in uranium exploration[3,4], environmental hazard assessment[5,6] and more recently in earthquake prediction[7–10]. ^{222}Rn is a long-lived isotope out of the three radon isotopes and is thus more mobile in the natural environment than other elements in the U-series. It is used as an effective tracer in understanding geo-physical processes that induce fluid motion in the ground. Prolonged exposure to radon and its daughter products may account for an increasing incidence of lung cancer among mine workers. Because of its importance in human life, it is of considerable interest to measure radon in air, soil and water for uranium exploration, environmental pollution, and earthquake prediction in western Himalaya.

There are various methods of U-estimation, viz. gamma ray spectrometry, mass spectrometry, laser flourimetry, autoradiography, neutron activation analysis and fission track-etch technique. The last one was used in our sample analysis due to its simplicity and lower detection limit.

The soil samples were collected from bore holes and dried in an oven at 150°C for 2 h. 50 mg of soil sample was mixed thoroughly with 100 mg of methyl cellulose powder used as a binder and the mixture was pressed into a pellet, about 1.3 cm dia and 0.1 cm thickness, using a hand press. Lexan polycarbonate discs of the same diameter were pressed against both sides of each pellet and then packed in an aluminium capsule, along with a standard glass dosimeter pellet. The capsule was irradiated in CIRUS Reactor at BARC, Trombay using a thermal neutron fluence of 10^{16} n/cm^2. After irradiation, Lexan discs were etched in 6.25 NaOH solution at 70°C for 40 min. The fission track density was measured using a Carl Zeiss binocular microscope with a calibrated eye-piece graticule. Comparison between the track densities on the Lexan discs surrounding the soil pellets and the dosimeter glass pellet gives the average value of U content by the relation[11,12]

$$C_{ppm}(\text{Sample}) = \frac{\rho(\text{Sample})}{\rho(\text{Standard})} \times C_{ppm}(\text{Standard}), \qquad (1)$$

where ρ represents the induced fission track density and C_{ppm} denotes the U content.

The experimental procedure for uranium estimation in water is based on the fission track technique[11,13]. A known volume of water (2 drops ≈ 0.04 cm^3) of each sample was allowed to evaporate on Lexan plastic discs (1.3 cm dia) in an air-tight enclosure. The non-volatile constituents of water were left over the discs in the form of a thin film/scale. The discs were packed in an aluminium capsule and sent for irradiation as in case of soil samples. After irradiation, Lexan discs were etched and the total number of fission fragment tracks counted. The detection limit of this method is 0.01 ppb, with a precision of 5–10%. U content in water was determined using the formula[13]

$$C_w = \frac{TM}{VGN_A E\sigma\phi}, \qquad (2)$$

where T is the total number of tracks counted over the disc, M the atomic weight of uranium (238), V the volume of water drop (0.04 cm^3), N_A the Avogadro number (6.023×10^{23}), G the geometry factor which is taken as unity, E the etching efficiency factor for Lexan plastic, σ the fission cross-section for ^{238}U (4.2×10^{-24} cm^2), and ϕ the thermal neutron fluence (5×10^{15} n/cm^2).

Both the track-etch technique and radon emanometry were used to estimate radon concentration in the soil-gas. In the track-etch method, a radon–thoron discriminator[14], with cellulose nitrate (LR-115 type II) film as track recorder, was used. The discriminator was kept in the auger hole 60 cm deep for a period of four weeks. After retrieval, the detector film was etched in 2.5 N NaOH solution at 60°C for two hours. The track density was measured by a binocular microscope and the

Table 1. Radon and U-concentrations in the soils of different geological areas

Place	Radon concentration (Bq/l)				U concentration (ppm)	
	Emanometry		Track-etch method		Track etch method	
	Min.	Max.	Min.	Max.	Min.	Max.
Himachal Pradesh						
Chhinjra	4.44 ± 0.37	567.68 ± 0.37	9.26 ± 7.77	92.87 ± 6.66	6.62 ± 0.42	86.93 ± 1.75
Samurkala	2.22 ± 0.37	53.65 ± 2.59	3.33 ± 0.37	151.33 ± 7.77	2.85 ± 0.24	116.14 ± 3.19
Kasol	7.77 ± 0.74	3468.01 ± 304.51	23.68 ± 1.85	4385.61 ± 377.77	5.50 ± 0.32	12.89 ± 0.55
Rameda	6.29 ± 0.74	803.64 ± 1.11	101.01 ± 7.77	3201.24 ± 91.76	1.85 ± 0.11	117.94 ± 2.76
Panjab						
Amritsar	2.22 ± 0.37	9.26 ± 0.74	5.92 ± 0.37	13.32 ± 0.74	2.56 (mean value)	

Table 2. Rn and U anomalies identified in water samples of western Himalaya

Sample location	Area	Rn content (Bq/m^3)	U content (ppb)	Remarks
Shat-Chhinjra	Kulu	323.01 ± 27.75	8.02 ± 0.07	Related to Shat-Chhinjra and Kasol mineralization[18]
Kasol (Hot spring)	Kulu	364.45 ± 30.34	37.40 ± 0.41	Related to Shat–Chhinjra and Kasol mineralization[18]
Maldeota	Dehradun	323.01 ± 22.94	63.40 ± 0.40	Related to mineralization of Mussoorie Syncline[19]
Paritibba	Dehradun	203.50 ± 23.31	26.47 ± 0.41	Related to mineralization of Mussoorie Syncline[19]
Jungle-Chitti	Garhwal	207.20 ± 22.2	12.64 ± 0.14	–
Nand Paryag	Garhwal	159.10 ± 22.2	33.40 ± 0.30	Related to Pokhri–Tunji mineralization[20]
Nangal (Choe)	Siwalik	223.11 ± 19.24	21.08 ± 0.24	–

radon was estimated by using calibration factor[15] of 1 track/mm^2/h = 82.5×10^3 Bq/m^3.

In radon emanometry, the auger holes, each 60 cm in depth and 6 cm dia, were left covered for 24 h. The soil-gas probe was fixed in the auger hole and connected to an alpha detector in a close-circuit[16]. The soil-gas is circulated through ZnS (Ag) coated chamber for 15 min till the radon forms a uniform mixture with the air. The detector was then isolated and radon alpha counts are recorded after four hours when equilibrium was established between radon and its daughters. The alpha counts were converted to radon activity in Bq/m^3 using the calibration factor[15].

The apparatus designed for estimation of radon in running tap or well water was discarded and the discrete sampling method[17] was adopted for convenience. 100 ml of each sample was collected in radon-tight reagent bottles of 1 litre capacity and connected to a conical flask through a hand-operated rubber pump and a glass bulb containing $CaCl_2$ to absorb moisture. LR-115 type II detector foils were kept suspended in the conical flask for 15 days. The radon gas was transferred from the reagent bottle to the flask by bubbling water and sucking the gas with the help of the rubber pump. This close-circuit technique is quite effective in radon estimation in dry or wet air. The detector foils were etched in 2.5 N NaOH solution at 60°C for 2 h and scanned under a Olympus microscope at a magnification of 600 ×. Track density was converted to radon concentration in Bq/m^3 with a precision of 5–10%.

Table 3. Uranium and radon content in the water channels of river Ganga

Sample location	No. of samples studied	U content (ppb)	Rn content (Bq/m^3)
Badrinath	2	3.95 ± 0.07	65.12 ± 11.84
Ram Dungi	2	2.39 ± 0.05	187.96 ± 15.91
Karan Prayag	2	3.56 ± 0.07	133.94 ± 25.16
Rudr Prayag	2	4.81 ± 0.05	166.87 ± 18.50
Dev Prayag	1	0.89 ± 0.02	224.22 ± 21.83
Rishi Kesh	2	8.00 ± 0.09	128.39 ± 16.65
Hardwar	2	7.79 ± 0.09	47.73 ± 11.10

Uranium and radon concentrations in the soil samples collected from different geological areas of Himachal Pradesh in the lower Siwaliks of western Himalaya are summarized in Table 1. There are extreme variations of radon and U contents even at the same site suggesting disequilibrium in the radioactive U-series. The maximum values of radon are recorded in Chhinjra, Samurkala and Rameda areas of Himachal Pradesh, which are identified for the uranium mineralization[18]. These results are corroborated by the gamma activity and *in-situ* ura-

Table 4. Rn and U contents in water samples of western Himalaya with regional threshold values

Sample location	No. of samples	Rn content (Bq/m^3) Min.	Rn content (Bq/m^3) Max.	Regional threshold (Bq/m^3)	U content (ppb) Min.	U content (ppb) Max.	Regional threshold (ppb)
Kulu	25	64.38	364.45	231.62	0.89	37.40	5.62
Kangra	30	34.04	202.76	172.05	0.20	2.31	2.70
Punjab Siwaliks	33	105.82	223.11	197.21	0.42	21.08	5.86
Garhwal	19	47.73	345.21	212.75	0.89	33.40	8.01
Dehradun	13	130.24	323.01	190.55	2.41	63.40	8.72

nium content in the soil of the area. This clearly indicates that radon can be used favourably for the exploration of uranium ore. Radon anomaly is also recorded in Kasol which is, however, not correlatable with the uranium content in the soil. Generally, the track-etch method yields higher radon values compared to emanometry because of its integrating nature of measurement.

Radon and uranium anomalies identified in water samples collected from rivers, streams and thermal springs of western Himalaya are reported in Table 2. The highest value of U content (63.40 ± 0.40 ppb) was observed in Maldeota area which is related to uranium mineralization of Mussoorie syncline[19], with U content as high as 612 ppb in the phosphorite samples[12]. The Kasol hot spring also records high U content of 37.40 ± 0.41 ppb and the highest radon concentration of 364.45 ± 30.34 Bq/m^3. The U contents in water samples of mountain springs falling into river Ganga show anomalous values which may be explained due to Pokhri–Tunji mineralization[20]. It is obvious that radon and uranium anomalies reported in Table 2 are related to uranium mineralization in the area through which the water channels flow.

Water samples were collected from streams and channels of river Ganga from Badrinath to Hardwar during September 1982 and analysed in the laboratory for uranium and radon activity. The results are summarized in Table 3. The highest U content in Ganga water was reported near Rishikesh and Hardwar where the river enters the plains. No correlation seems to exist between U content and radon activity reported in water samples. Similar results were reported by Sarin *et al.*[2] about the U content in the river Ganga and its tributaries. The minimum U content of 0.89 ± 0.02 ppb is determined at Dev Prayag, the junction of Bhagirathi and Alaknanda. However, the radon content is estimated to be 224.22 ± 21.83 Bq/m^3, which is highest for Ganga water. The high radon values may be due to radium separated from uranium and precipitating for a long time on the walls of fractured rocks.

Regional threshold values of radon and uranium concentration in water samples collected from springs, streams and rivers of western Himalaya are listed in Table 4, along with the minimum and maximum values. Regional threshold for uranium in the Ganga river basin (Garhwal, Dehradun) is found to be higher t and Satluj river basins (Kulu, Kangra and P waliks).

1. Sarin, M. M., Krishnaswami, S., Somayajulu, B. Moore, W. S., *Geochim. Cosmochim. Acta*, 1990, 1396.
2. Sarin, M. M., Krishnaswami, S., Sharma, K. K a J. R., *Curr. Sci.*, 1992, **62**, 801–805.
3. Ramola, R. C., Sandhu, A. S., Singh, M., Singh, S H. S., *Nucl. Geophys.*, 1989, **3**, 57–69.
4. Ghosh, P. C. and Bhalla, N. S., *Indian J. Pure A* 1975, **13**, 713–714.
5. Ramola, R. C., Singh, M., Singh, S. and Virk, H. S., *Radioactivity*, 1992, **15**, 95–102.
6. Fleischer, R. L., Giard, H. R., Mogro-Campero, L. G., Alter, H. W. and Gingrich, J. E., *Health Phys* 957–962.
7. Fleischer, R. L., *Geophys. Res. Lett.*, 1981, **8**, 477–4
8. Ramola, R. C., Singh, M., Sandhu, A. S., Singh, S H. S., *Nucl. Geophys.*, 1990, **4**, 275–287.
9. Virk, H. S. and Singh, B., *J. Geophys. Res. Lett.* 737–740.
10. Virk, H. S., *Nucl. Geophys.*, 1995, **9**, 141–146.
11. Fleischer, R. L. and Lovett, D. B., *Geochim. Cosmc* 1968, **32**, 1126–1128.
12. Singh, S. and Virk, H. S., *Indian J. Pure Appl. Phys* 125–126.
13. Virk, H. S. and Harinder Kaur, *Curr. Sci.*, 1979, **48**,
14. Singh, M., Ramola, R. C., Singh, N. P., Singh, S H. S., *Nucl. Tracks Radiat. Meas.*, 1984, **8**, 415–418
15. Singh, M., Ramola, R. C., Singh, N. P., Singh, S H. S., *Nucl. Tracks Radiat. Meas.*, 1986, **12**, 739–74
16. Ghosh, P. C. and Bhalla, N. S., Proceedings of tl Symposium on Radioactivity and Meterology of Ra AEET, Bombay, 1966, p.14.
17. Singh, N. P., Singh, S. and Virk, H. S., *Indian J. Phys.*, 1987, **25**, 87–89.
18. Narayan Das, G. R., Parthasarthy, T. N., Taneja, P. rumal, N. V. A. S., *J. Geol. Soc. India*, 1979, **20**, 95-
19. Saraswat, A. C., Sankaran, A. V., Varadraju, H. P. C. and Bargaje, V. B., Proceedings of the UNES session on Geochemical Prospecting, Colombo, 197C
20. Dar, K. K., *J. Geol. Soc. India*, 1964, **5**, 110–120.

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