ENVIRONMENTAL RADIOACTIVITY SURVEYS IN WESTERN HIMALAYAS

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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 was observed that Himalayan river system is conspicuous by its high dissolved uranium and radium concentrations. Water samples contained from 0.89 to 63.40 ppb of uranium and from 34 to 364 Bq/1 of radon. The radon emanation in soil was measured by track-etch method, emanometry, and alpha-logger techniques. Daily and long-term variation of radon was monitored in some U-mineralised zones of Himachal Pradesh and Uttranchal States with high uranium content in soil. There is a need to undertake epidemiological study correlating cancer risk with high uranium and radon values in the environment.

INTRODUCTION

The measurement of uranium and radon in environment, in general, and in the Himalayan ecosystem, in particular, is of special interest to mankind. It has long been known that radon is a causative agent of lung cancer, when present in high concentrations, as observed in uranium mines (Archer et al., 1976; Sevc et al., 1976). The health hazard of radon is principally due to its short-lived daughters: ^{218}Po, ^{214}Bi and ^{214}Po. During recent years, several reports have demonstrated the ever-increasing interest in monitoring radon in indoor environment of dwellings all over the world (Jonassen, 1975; Abu-Jarad and Fremlin, 1981; Alter and Fleischer, 1981; Nazaroff and Doyle, 1985; Ramola et al., 1992).

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/1, compared to the global average of 0.3 μg/1 in river waters. The Ganges and Brahmaputra, together, transport about 1,000 tonnes of dissolved uranium to the estuaries of the Bay of Bengal annually (Sarin et al., 1990,1992). Radon estimation in soil-gas, ground water and atmosphere is an established technique in uranium exploration (Ghosh and Bhalla, 1975; Ramola et al., 1989), environmental hazard assessment (Fleischer et al., 1980; Singh J. et al., 1989) and more recently in earthquake prediction studies (Fleischer, 1981; Ramola et al., 1990; Virk and Singh, 1994; Virk, 1995).

^{222}Rn is a long-lived isotope out of the three radon isotopes and is thus more mobile in natural environment than other elements in the uranium series. It is used as an effective tracer in understanding geophysical 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 the 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 the Western Himalaya.

URANIUM/RADON MEASUREMENT TECHNIQUES

Uranium estimation in soil

There are various methods of uranium estimation , viz., gamma-ray spectrometry, mass spectrometry, laser flourimetry, autoradiography, neutron activation analysis and fission track-etch technique. The latter one was used in our sample analysis due to its simplicity and lower detection limit. Soil samples were collected from bore holes and dried in oven at 150^0C for 2 hr. Fifty mg of soil sample was mixed thoroughly with 100mg of methyl cellulose powder used as a binder and the mixture was pressed into a pellet, about 1.3 cm diameter and o.1 cm thickness, using a hand press. Lexan polycarbonate discs of the same diameter were pressed against both sides of each pellet. The capsule was got irradiated in CIRUS reactor at BARC, Mumbai using a thermal neutron fluence of 10^6 n/cm^2. After irradiation, Lexan discs were etched in 6.25 N NaOH solution at 70^0C for 40 min. Fission track density was measured using a Carl Zeiss binocular microscope with a calibrated eye-piece graticule. The comparison between track densities on the Lexan discs surrounding the soil pellets and dosimeter glass pellet gives the average value of uranium content by the relation (Fleischer and Lovett, 1968; Singh and Virk, 1983):

$$C_{ppm}\,(\text{Sample}) = [p\,(\text{Sample}) / P\,(\text{Standard})]\; C_{ppm}\,(\text{Standard}) \quad \ldots\ldots..(1)$$

Uranium estimation in water

The experimental procedure for uranium estimation in water is based on fission track technique (Fleischer and Lovett, 1968; Singh et al., 1987a; Virk and Kaur, 1979). A known volume of water (two drops $\cong$ 0.04 cm^3) of each sample was allowed to evaporate on Lexan plastic discs (1.3 cm diameter) in an air-tight enclosure. 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 the 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%. The uranium content in water was determined using the following formula (Virk and Kaur, 1979):

$$C_w\,(TM) / (VGN_AE\,\sigma\phi) \quad \ldots\ldots..(1)$$

Where T= Total number of tracks counted over the disc,
M = Atomic weight of uranium (238),
V = Volume of water drop (0.04 cm^2),
N_A = Avogadro number (6.023×10^{23}),
G = Geometry factor which is taken as unity,
E = Etching efficiency factor for Lexan .plastic.
σ = Fission cross-section for ^{238}U (4.2×10^{-24} cm^2),
φ= Thermal neutron fluence (5×10^{15} n/cm^2),

Radon estimation in soil

Both track-etch technique and radon emanometry were used to estimate radon concentration in soil-gas. In track-etch method, radon-thoron discriminator (Fig. 1), with cellulose nitrate (LR-115 II) film as track recorder, was used (Singh et al., 1984). The discriminator was kept in the auger hole 60 cm deep for a period of 4 weeks. After retrieval, the detector film was etched in 2.5 N NaOH solution at 60^0C for 2 hr. Track density was measured by Carl Zeiss binocular microscope and radon was estimated by using calibration factor (Singh, M. et al., 1986) of 1 track//mm^2/hr=$82.5x10^3$ Bq/m^3.

In radon emanometry, the auger holes, each 60 cm in depth and 6 cm diameter, were left covered for 24 hr. The soil-gas probe was fixed in the auger hole and connected to an alpha-detector in a close-circuit (Fig. 2). The soil-gas was 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 were recorded after 4 hr 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 (Singh, M. et al., 1986).

Radon estimation in water

The apparatus designed for the estimation of radon in running tap or well water was discarded and the discrete sampling method (Fig.3) was adopted for convenience. Hundred ml of each sample was collected in radon-tight reagent bottles of one litre capacity and connected to a conical flask through a hand-operated rubber pump and a glass bulb containing Ca Cl_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 60^0C for 2 hr and scanned under Olympus microscope at a magnification of 600X. Track density was converted to radon concentration in Bq/m^3 with a precision of 5-10%.

Alpha Scintillometer (GBH 2002) with Lucas cell assembly (Fig. 4), supplied byInternational Environment Consulting, Germany, was used to record radon concentration in water. Radon gas emanating from radium dissolved in one litre of water was sucked by a pump connected to a radon bubbler with an extraction efficiency of more than 90%. Radon concentration is measured by converting alpha counts recorded in digital counter (10 counts=1 Bq/l). The detection limit for the Lucas cells used in 0.02Bq/l.

Radon estimation in indoor air

The sources of radon inside the dwellings are mainly soil beneath and the building materials used in the construction. For environmental survey, both the track-etch method and the electronic counters have been used. Plastic foils, LR-115 type II, 2 cm^2 each, were fixed on the glass slide with the help of scotch tape and suspended from the roofs of dwellings. After an exposure of one month, the detector foils were removed and

etched in the laboratory. The measured track density was converted to radon concentration in indoor air by using a calibraion factor (Singh, M. et al., 1986).

The electronic alpha-counter using pulse-ionisation chamber is found to be most suitable for radon estimation in environment. We have used Alpha-Guard PQ 2000 (Genitron Co., Germany) which is portable, direct reading and with a detection limit of 1 Bq/m^3. It has the advantage of measuring radon along with meteorological parameters in indoor environment, viz., temperature, pressure and humidity. Hence, it is possible to study radon correlation with meteorological variables during different seasons of the year. Alpha-Guard can be used to measure both instantaneous and integrated values of radon concentration inside the indoor air of dwellings. We have used to cross-check our radon results using this sensitive and rugged instrument.

RESULTS AND DISCUSSION

The uranium and radon concentrations in 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 uranium contents even at the same site suggesting disequilibrium in the radioactive uranium series. Maximum values of radon are recorded in Chhinjra, Samurakala and Rameda areas of Himachal Pradesh which were identified for uranium mineralisation (Narayan Das et al., 1979; Ramola, 1989). These results are corroborated by the gamma activity and in-situ uranium content in the soil of this 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 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 uranium content (63.40 ± 0.40 ppb) is observed in Maldeota area which is related to uranium mineralisation of Mussoorie syncline (Saraswat et al., 1970) with uranium content as high as 612 ppb in phosphorite samples. The Kasol thermal spring also records high uranium content of 37.40 ± 0.41 ppb and the highest radon concentration of 364.45 ± 30.34 Bq/m^3. The uranium contents in water samples of mountain springs falling into river Ganga show anomalous values which may be explained due to Pokhri-Tunji mineralisation (Dar, 1964). It is obvious that radon and uranium anomalies reported in Table 2 are related to uranium mineralisation in the area through which the water channels flow.

Water samples were collected from streams and channels of river Ganga from Badrinath to Hardwar and analysed in the laboratory for uranium and radon activity. The results are presented in Table 3. The highest uranium content in Ganga water is reported near Rishikesh and Hardwar, where the river enters the plains. No correlation seems to exist between uranium content and radon activity reported in water samples. Similar results are reported by Sarin et al. (1992) about the uranium content in the river Ganga and its tributaries. Minimum uranium 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. High radon

values may be due to radium separated from uranium and precipitating for a long time on the walls of fractured rocks (Bhimashankaran, 1974).

The radon concentration values in indoor environment are depicted in Table 4. A comparison can be made between radon values recorded in the houses of Amritsar (non-uraniferrous area) with those recorded in the houses of Rameda, Rawatgaon and Samurkala villages situated in the uraniferrous zones of H.P. state. These values are recorded under different environmental conditions and provide a wide range of variation due to ventilation, wind direction, weather conditions etc. Maximum radon concentration is found in indoor air of houses in Rameda area which is an order of magnitude higher than the value recorded in Amritsar (Singh, J. et al., 1989). High radon values are also recorded in the dwellings of Rawatgaon and Samurkala which may be due to presence of radioactive building materials (boulders etc.) used in the construction of houses or due to seepage of radon-rich soil-gas from the basement. The indoor radon values are found to be correlated with outdoor radon values only for the well-ventilated rooms. In general, anomalously high radon values in the houses of Rameda, Rawatgaon and Samurkala in H.P. state are beyond the intervention level (200-600 Bq/m^3) recommended by the International Commission on Radiological Protection (ICRP, 1994), and are a cause of serious concern for the general population living in these villages. The annual effective dose received by the village population in Rameda is 22.56 mSv (safety limit 10 mSv) (Singh, 1995) and the lifetime fatality risk calculated on the basis of ICRP (1991) is 11.28×10^{-4} on the average. Hence, there is a need for undertaking epidemiological health hazard to general population living in the uranium-mineralised zones of Western Himalaya.

Regional threshold values and uranium concentration in water samples collected from springs, streams and rivers of Western Himalaya are listed in Table 5 along with the minimum and maximum values. Regional threshold for uranium in the Ganga river basin (Garhwal, Dehradun) is found to be higher than Beas and Sutlej river basins (Kulu, Kangra and Punjab Siwaliks). Alpha Scintillometry technique has been used to collect radon data (Table 6) of some thermal/natural springs of Uttaranchal State. The values are found to be surprisingly low with variation from 0.8 ± 0.3 to 4.4 ± 0.7 Bq/l.

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Table 1. Radon and uranium concentration in soils of different geological areas in Himachal Pradesh.

Place	Radon concentration (Bq/l)				Uranium concentration (ppm)	
	Emanometry		Track-etch method		Track-etch method	
	Minimum	Maximum	Minimum	Maximum	Minimum	Maximum
Chinnjra	4.44±0.37	567.98±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

Table 2. Radon and uranium anomalies identified in water samples of Western Himalaya.

Sample location	Area	Radon content Bq/m^3	Uranium content (ppb)	Remarks
Shat-Chhinjra	Kulu	323.01±27.75	8.02±0.07	Related to Shat-Chhinjra and Kasol mineralisation (Narayan Das et al., 2979)
Kasol (Hot spring)	Kullu	364.45±30.34	37.40±0.41	Related to Shat –Chhinjra and Kasol mineralisation (Narayan Das et al., 1979)
Maldeota	Dehradun	323.01±22.94	63.40±0.40	Related to mineralisation of Mussoorie Sysncline (Saraswat et al., 1970)
Paritibba	Dehradun	203.50±23.31	26.47±0.41	Related to mineralisation of Mussoorie Sysncline (Saraswat et al., 1970)
Jungle Chitti	Garhwal	207.20±22.20	12.64±0.14	
Nand Paryag	Garhwal	159.10±22.20	33.40±0.30	Related to

				Polhri-Tunji Mineralisation (Dar, 1964)
Nangal (Choe)	Siwalik	223.11±19.24	21.08±0.24	

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

Sample Location	No. of samples studied	Uranium contents (ppb)	Radon 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

Table 4. Radon concentration in indoor enviroment.

Place	Radon concentration (Bq/m^3)	
	Minimum	Maximum
Amritsar	36.26±4.07	303.40±11.84
Rameda	1031.93±78.44	2413.51±217.19
Rawatgaon	348.17±33.67	1208.79±92.87
Samurkala*	442.00±0.00	1254.00±0.00

* Singh, J. (1995)

Table 5. Radon and uranium contents in water samples of Western Himalaya.

Sample location	No. of samples	Radon contents (Bq/m^3)		Regional threshold (Bq/m^3)	Uranium contents (ppb)		Regional threshold (ppb)
		Minimum	Maximum		Minimum	Maximum	
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

Table 6. Radon concentration in Thermal/Natural Springs of Uttaranchal State.

Name of Place	Source	Radon Conc. (Bq/l)
Suryakund, Yamunotri	Thermal Spring	0.8±0.3
Gangnani	Thermal Spring	2.6±0.5
Netala, Gangnani	Natural Spring	1.1±0.3
Gauri Kund, Kedar Nath	Thermal Spring	4.4±0.7
Kund (on way to Kedar Nath)	Natural Spring	2.6±0.5
Rudraprayag	Natural Spring	3.1±0.6