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Study of Naturally Occurring Radioactive Material Present in Deep Soil of the Malwa Region of Punjab State of India Using Low Level Background Gamma-Ray Spectrometry

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ABSTRACT

Background: Epidemiological observations such as mental retardation, physical deformities, etc., in children besides different types of cancer in the adult population of the Malwa region have been reported. The present study is designed to get insight into the role of naturally occurring radioactive material (NORM) in causing detrimental health effects observed in the general population of this region.

Materials and Methods: Deep soil samples were collected from different locations in the Malwa region. Their activity concentrations were determined using low-level background gamma-ray spectrometry. High efficiency and high purity germanium detector capped in a lead-shielded chamber having a resolution of 1.8 keV at 1,173 keV and 2.0 keV at the 1,332 keV line of ^{60}Co was used in the present work. Data were evaluated with Genie-2000 software.

Results and Discussion: Mean activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in deep soil were found to be 101.3 Bq/kg, 65.8 Bq/kg, and 688.6 Bq/kg, respectively. The mean activity concentration of ^{238}U was found to be three and half times higher than the global average prescribed by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). It was further observed that the activity concentration of ^{232}Th and ^{40}K has a magnitude that is nearly one and half times higher than the global average prescribed by UNSCEAR. In addition, the radioisotope ^{137}Cs which is likely to have its origin in radiation fallout was also observed. It is postulated that the NORM present in high quantity in deep soil somehow get mobilized into the water aquifers used by the general population and thereby causing harmful health problems.

Conclusion: It can be stated that the present work has been able to demonstrate the use of low background gamma-ray spectrometry to understand the role of NORM in causing health-related effects in a general population of the Malwa region of Punjab, India.

Keywords: Naturally Occurring Radioactive Material (NORM), Low-Level Background gamma-Ray Spectrometry, Deep Soil

Original Research

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Introduction

Naturally occurring radioactive material (NORM) namely ^{238}U, ^{232}Th, and ^{40}K have been present in our surroundings since time immemorial. The carcinogenic radon as well as polonium is produced from their decay. Anthropogenic interventions like agri-

culture, industrial and mining activities may lead to the increase in their concentration [1]. NORM is known to cause not only chemical toxicity and radiological toxicity but combined synergetic effect of both [2]. Their exposures may cause kidneys and lungs related diseases, DNA damage and sub-cellular organelles activity, etc. [3–5].

Lately, there has been an increasing concern about harmful health effects arising from NORM in the Malwa region of Punjab State located in the North Western part of India. This region has been reported to have a high incidence of cancer [6, 7]. The incidence of cancer in India is estimated to be 80 per 100,000 population whereas incidence of cancer in the Malwa region of Punjab State in India is observed to be 90 per 100,000 population [8]. A number of researchers [9–11] have reported that the concentration of uranium in the groundwater in the Malwa region is significantly higher not only with respect to the permissible limit of 15–30 ppb prescribed by international regulators like the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [12], the World Health Organization (WHO) [13] and the United States Environmental Protection Agency (EPA) [14] but also with respect to the concentration levels observed in other regions of India [15–17].

Schnug and Lottermoser [18] stressed the significance of fertilizer-derived uranium for groundwater contamination in regions with a low geogenic uranium background. Punjab State being the food bowl of India has seen excessive use of chemical fertilizers in the past few decades. Therefore contribution from phosphate fertilizers which are known to be enriched with NORM also cannot be ruled out.

In the present work, the low-level background high-resolution gamma-ray spectrometer has been employed to obtain the concentration of NORM in deep soil samples to understand its contribution in bringing about detrimental public health effects reported in its general population.

Materials and Methods

1. Samples Collection

Soil Samples observed to be free of roots from the depth ranging between 30 cm and 180 cm which are normally expected to remain undisturbed from anthropogenic activity, soil erosion, soil turnover, irrigation, phytoaccumulation, etc., were collected by using the standard Auger method from 15 different locations of the Malwa region of Panjab State in India. The region in Malwa from where the samples were collected is shown encircled in red in Fig. 1 [19].

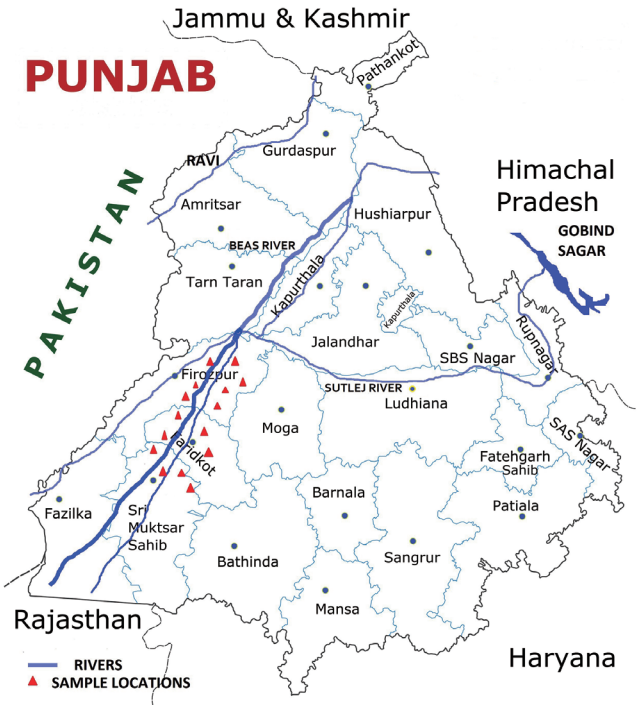

Fig. 1. Map showing the collection sites (encircled in red) in the Malwa region of Punjab State, India. Adapted from [19].

2. Sample Analysis

The samples after drying and pulverizing were placed in a petri dish after recording their mass. Each petri dish was sealed and kept for 5 weeks to ensure secular equilibrium between parent and daughter products. To avoid bias, the samples were encoded with neutral names. Their radionuclide content was determined using ultra-low-background gamma-ray spectrometric setup of the Radiation Counting Laboratory of Hochschule Mannheim, Mannheim, Germany, which is shown in Fig. 2.

The Petri dishes containing the samples were measured for 24 hours. Counting was carried out using 16k multichannel analyzers employing high-efficiency HPGe detectors kept in Pb shielded chambers. The detectors had a resolution of 1.8 keV at 1,173 keV line and 2.0 keV at the 1,332 keV line of ^{60}Co.

Data was evaluated with Genie-2000 (Canberra Industries Inc., Meriden, CT, USA) by using efficiency curves for each detector measured with an aliquot of a standardized commercial multi-nuclide calibration solution (Eckert & Ziegler, Berlin, Germany) which was carefully mixed with silica gel and placed into a Petri dish of the same dimension (i.e., 90

Fig. 2. Ultra-low-background gamma-ray spectrometric setup of Hochschule Mannheim, Mannheim, Germany.

mm or 70 mm diameter) as the samples and placed in identical positions as the samples.

The energy calibration was performed with the nuclides of the samples. A nuclide library based on the International Atomic Energy Agency (IAEA) isotope browser [20] was used for nuclide identification and activity calculation.

Background spectra were taken for 72 hours in order to keep the computational uncertainties small. Repeated background measurements revealed a stable situation during the counting campaign. In order to keep the background rates low, radiation shields built from 10 cm of old lead lined with 1 mm electrolytic copper surrounded the detectors. The internal volume of the shield was continuously rinsed by the evaporated nitrogen from the Dewar vessel.

3. Radioactivity Calculation

The activity of ^{238}U was estimated by measuring the peak area under the gamma line of ^{214}Pb occurring at 351.9 keV besides 609.3 keV gamma line of ^{214}Bi. The ^{232}Th activity was estimated by measuring the peak area under gamma lines of ^{228}Ac namely 911.2 keV and 968.9 keV. The ^{40}K activity was estimated using 1,461 keV gamma line peak area and activity of ^{137}Cs was estimated by measuring the peak area under the 661.7 keV gamma line. The activities were obtained together with their standard uncertainties by a routine fitting of Gaussian distribution to each line. The same is performed with the background spectra. The count rates of gamma lines of interest were automatically subtracted from the sample spectra by a routine in Genie-2000 yielding net count rates. In the next step of analysis, the count rate of each gamma line was calculated by dividing by the counting efficiency at its energy as obtained from a standard fitting routine in Genie-2000. With the aid of a radionuclide library with data from the IAEA isotope browser, the gamma lines were identified. By using the emission rates of each gamma line, the activity of the nuclide was calculated as the weighted mean of the activities obtained for the gamma lines of interest of each radionuclide.

Typically, the nuclides of the 4n and 4n+2 natural decay series were observed, in some cases, also ^{235}U and some of its descendants from the 4n+3 series. The thorium series reaches equilibrium quickly and, hence, in most cases, is found to be in secular equilibrium. This is different from the uranium series where hydrogeological separation of uranium and thorium from the radium is found frequently. Soluble uranyl and thorium cations can be dissolved in carbonaceous and sulfatic groundwater whereas radium is sparely soluble under these conditions. As a consequence, the direct progeny of ^{238}U, ^{234}Pa is not visible in the gamma-ray spectra, as well as ^{235}U. This is relevant as the only gamma-ray emission of ^{226}Ra at 186.2 keV (15.7%) coincides with the strongest gamma line of ^{235}U. The contribution of this can be estimated from other strong gamma lines and be subtracted from the sum peak if present. If gamma lines of ^{234m}Pa and ^{235}U were identified, the activity at 186 keV was attributed to ^{238}U in equilibrium with ^{226}Ra. In addition, the activity ratio of ^{235}U and ^{238}U was checked to be reasonable. In all other cases, activity was attributed to ^{226}Ra separated geochemically from its longer-lived parents.

Results and Discussion

Table 1 gives the mean activity concentration of ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs based on three measurements for each of the 15 deep soil samples namely M1 to M15 collected from different locations in Malwa region. Table 1 also gives the overall mean activity concentration of the radioisotopes of interest with its associated 1σ standard deviation. In the case of ^{137}Cs, only two samples yielded mean activity concentration above the limit of detection. Hence, a range is given. Table 2 gives the mean global NORM activity level prescribed by

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UNSCEAR [12]. The mean activity level of ^{238}U, ^{232}Th, ^{40}K in soils from an area close to Malwa region which has been reported in literature [21–23] is also shown in Table 2 along with the data obtained in the present work.

Table 1 shows that the activity concentration of ^{238}U ranges between 65.5 Bq/kg and 154.4 Bq/kg. It is also observed that the activity concentration of ^{232}Th ranges between 52.0 Bq/kg and 77.5 Bq/kg, whereas the activity concentration of ^{40}K ranges between 585.3 Bq/kg and 895.2 Bq/kg. The mean and the associated standard deviation values of ^{238}U, ^{232}Th, and ^{40}K are found to be 101.3 ± 23.5 Bq/kg, 65.8 ± 7.3 Bq/kg, and 688.6 ± 93.9 Bq/kg, respectively.

Table 1. Mean Activity Concentration of ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs (unit: Bq/kg)

Sample number	^{238}U	^{232}Th	^{40}K	^{137}Cs
M1	113.6	74.7	801.3	≤0.5
M2	87.7	62.9	716.8	≤0.5
M3	90.2	72.7	895.2	≤0.5
M4	88.9	67.0	719.9	≤0.5
M5	111.2	77.5	651.0	≤0.5
M6	65.5	52.0	663.6	1.4
M7	85.2	65.0	635.4	≤0.5
M8	82.7	62.5	788.8	≤0.5
M9	102.5	60.1	698.0	≤0.5
M10	140.8	64.6	585.3	≤0.5
M11	96.3	60.9	694.9	≤0.5
M12	154.4	59.3	616.1	≤0.5
M13	85.2	59.7	585.3	1.6
M14	98.8	72.7	629.1	≤0.5
M15	118.6	74.3	732.4	≤0.5
Mean ± SD[a]	101.3 ± 23.5	65.8 ± 7.3	688.6 ± 93.9	≤0.50–1.6

The activity concentration of ^{238}U ranges between 65.5 Bq/kg and 154.4 Bq/kg, the activity concentration of ^{232}Th ranges between 52.0 Bq/kg and 77.5 Bq/kg, whereas the activity concentration of ^{40}K ranges between 585.3 Bq/kg and 895.2 Bq/kg.
In the case of ^{137}Cs only two samples yielded mean activity concentration above the limit of detection. Hence, a range is given.

[a]Associated standard deviation value.

In addition to the aforementioned radioisotopes, ^{137}Cs with an activity concentration close to the limit of detection of ≤0.5 Bq/kg and, in some cases, around 1.5 Bq/kg was also observed. The presence of abovementioned radionuclides could perhaps be the trace to the radiation fallout, which may have found its way into the deep soil presumably as a result of irrigation and agricultural activities carried out over the years. A comparison of the data obtained in the present work with the global average data reported by UNSCEAR [12] shows that the ^{238}U activity concentration determined in the present work has a magnitude which is nearly three and a half times higher than the global average whereas the activity concentration of ^{232}Th and ^{40}K has a magnitude which is nearly one and half times higher than the global average.

It is further observed that the deep soil NORM activity concentration which has been determined for the first time in the present work is significantly higher than the activity concentration reported in literature for top soils [12, 21, 22]. This could perhaps be due to the deep soil NORM concentration remaining unaffected by phytoaccumulation and other farm practices of the local area which lead to transportation of matter from the lower to the upper part of the soil. It can be further hypothesized that the NORM present in the deep soil of the Malwa region could lead to the enrichment of the groundwater of Malwa region by getting mobilized from the terrestrial to the aquatic environment when conducive geochemical conditions occur. The groundwater enriched in NORM is then somehow finding its way into the general population causing health problems.

Conclusion

It can be stated that the NORM activity concentration of the deep soil determined in the present work from area where the local population has been reported to be affected by det-

Table 2. Activity Concentration Data Obtained in the Present Work along with the Data from Literature

	NORM (Bq/kg)				
	UNSCEAR [12]	Srivastava et al. [21]	Kumar et al. [22]	Srivastava et al. [23]	Present work
^{238}U	30[a]	20.9 ± 3.7	30.2 ± 0.5	42.7 ± 5.3	101.0 ± 23.5
^{232}Th	45[a]	34.7 ± 10.3	29.9 ± 0.6	45.8 ± 4.6	65.8 ± 7.3
^{40}K	420[a]	407.0 ± 113.0	291.0 ± 1.0	587.0 ± 52.8	688.0 ± 94.0

Values are presented as mean ± standard deviation.
The mean global NORM activity level prescribed by UNSCEAR [12]; the mean activity levels of ^{238}U, ^{232}Th, ^{40}K in soils from area close to the Malwa region which have been reported in literature [21–23].
NORM, naturally occurring radioactive material; UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation.
[a]Recommended value.

rimental health effects is comparatively higher and may lead to health-related problems if mobilized to aquifers. A more detailed study is needed to be carried out to get a clearer picture of the existing problem.

Conflict of Interest

No potential conflict of interest relevant to this article was reported.

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Author Contribution

Conceptualization: Srivastava A. Data curation: Chahar V, Chauhan N. Formal analysis: Chahar V, Chauhan N, Krupp D, Scherer U. Funding acquisition: Srivastava A, Scherer U. Methodology: Krupp D. Writing - original draft: Srivastava A. Writing - review & editing: Srivastava A, Krupp D, Scherer U. Investigation: Krupp D, Scherer U. Supervision: Srivastava A, Scherer U.

References

1. Bleise A, Danesi PR, Burkart W. Properties, use and health effects of depleted uranium (DU): a general overview. J Environ Radioact. 2003;64(2–3):93–112.
2. Busby C, Schnug E. Advanced biochemical and biophysical aspects of uranium contamination. In: De Kok LJ, Schnug E, editors. Loads and fate of fertilizer derived uranium. Leiden, Netherlands: Backhuys Publishers; 2008. p. 11–22.
3. Kang JK, Seo S, Jin YW. Health effects of radon exposure. Yonsei Med J. 2019;60(7):597–603.
4. Bjorklund G, Christophersen OA, Chirumbolo S, Selinus O, Aaseth J. Recent aspects of uranium toxicology in medical geology. Environ Res. 2017;156:526–533.
5. Harrison J, Leggett R, Lloyd D, Phipps A, Scott B. Polonium-210 as a poison. J Radiol Prot. 2007;27(1):17–40.
6. Blaurock-Busch E, Busch YM, Friedle A, Buerner H, Parkash C, Kaur A. Comparing the metal concentration in the hair of cancer patients and healthy people living in the Malwa region of Punjab, India. Clin Med Insights Oncol. 2014;8:1–13.
7. Blaurock-Busch E, Friedle A, Godfrey M, Schulte-Uebbing CE, Smit C. Metal exposure in the children of Punjab, India. Clin Med Insights Ther. 2010;2:655.
8. Lavit K, Shalini S. Mitigating cancer mortality in Punjab: a challenge for development. Indian J Econ Dev. 2017;13(2a):36–41.
9. Bajwa BS, Kumar S, Singh S, Sahoo SK, Tripathi RM. Uranium and other heavy toxic elements distribution in the drinking water samples of SW-Punjab, India. J Radiat Res Appl Sci. 2017;10(1):13–19.
10. Kumar A, Usha N, Sawant PD, Tripathi RM, Raj SS, Mishra M, et al. Risk assessment for natural uranium in subsurface water of Punjab State, India. Hum Ecol Risk Assess. 2011;17(2):381–393.
11. Alrakabi M, Singh G, Bhalla A, Kumar S, Kumar S, Srivastava A, et al. Study of uranium contamination of ground water in Punjab state in India using X-ray fluorescence technique. J Radioanal Nucl Chem. 2012;294(2):221–227.
12. United Nations Scientific Committee on the Effects of Atomic Radiation. Sources and effects of ionizing radiation (Volume 1). New York, NY: United Nations Scientific Committee on the Effects of Atomic Radiation; 2000.
13. World Health Organization. Guidelines for drinking-water quality. 4th ed. Geneva, Switzerland; World Health Organization; 2011.
14. United States Environment Protection Agency. 2011 edition of the drinking water standards and health advisories. Washington, DC: United States Environment Protection Agency; 2011.
15. Bhardwaj S, Shukla DP, Halder A. Spatial distribution of uranium and chemo-radiological assessment in Hamirpur district, Himachal Pradesh, India. J Radioanal Nucl Chem. 2020;324(2):467–480.
16. Kumar D, Singh A, Jha RK. Spatial distribution of uranium and basic water quality parameter in the capital of Bihar and consequent ingestion dose. Environ Sci Pollut Res Int. 2018;25(18):17901–17914.
17. Bajaj M, Eiche E, Neumann T, Winter J, Gallert C. Hazardous concentrations of selenium in soil and groundwater in North-West India. J Hazard Mater. 2011;189(3):640–646.
18. Schnug E, Lottermoser BG. Fertilizer-derived uranium and its threat to human health. Environ Sci Technol. 2013;47(6):2433–2434.
19. Central Water Commission [Internet]. Chandigarh, India: Government of India; 2020 [cited 2022 Mar 1]. Available from: http://cwc.gov.in/ibo/about-basins.
20. International Atomic Energy Agency. IAEA isotope browser app now available in multiple languages [Internet]. Vienna, Austria:

 https://doi.org/10.14407/jrpr.2021.00031

International Atomic Energy Agency; 2017 [cited 2022 Mar 1]. Available from: https://www.iaea.org/newscenter/news/iaea-isotope-browser-app-now-available-in-multiple-languages.
21. Srivastava A, Lahiri S, Maiti M, Knolle F, Hoyler F, Scherer UW, et al. Study of naturally occurring radioactive material (NORM) in top soil of Punjab State from the North Western part of India. J Radioanal Nucl Chem. 2014;302(2):1049–1052.
22. Kumar A, Kumar S, Singh J, Singh P, Bajwa BS. Assessment of natural radioactivity levels and associated dose rates in soil samples from historical city Panipat, India. J Radiat Res Appl Sci. 2017; 10(3):283–288.
23. Srivastava A, Chahar V, Sharma V, Sun Y, Bol R, Knolle F, et al. Study of uranium toxicity using low-background gamma-ray spectrometry. J Radioanal Nucl Chem. 2017;314(2):1367–1373.