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Study of uranium mobilization from Himalayan Siwaliks to the Malwa region of Punjab state in India

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Abstract Fossils, palaeosols and associated sedimentary rock samples from well-dated Siwalik sediments have been measured for their uranium content by low background gamma ray spectrometry with a view to study the role of geo-genic mobilization in enhancing the levels of uranium in ground water bodies of Malwa region in Punjab state, North Western India. Uranium activity in pure palaeosol and palaeosol associated samples i.e. calcrete and nodules varied between 46 and 214 Bq/kg whereas the same in pure fossil samples varied between 208 and 4837 Bq/kg. The data indicates a geo-genic contribution in the enhancement of uranium concentration in groundwater of the region.

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Introduction

The concerns about toxicity related to uranium in Malwa region of Punjab state located in North Western part of India is increasing day by day. Kumar et al. [1] have shown that the mean concentration of uranium in the groundwater is 73.1 ppb i.e. five times higher than the permissible limit of 15 ppb prescribed by the World Health Organization [2]. Kumar [3] also reported that the concentration of uranium in ground water samples collected from Bathinda District in Malwa region varied between 139 and 295 ppb. Alrakabi et al. [4] too found elevated levels of uranium in ground water samples obtained from this region. Several hypotheses have been postulated to explain the higher level of uranium concentration in the ground water, which ranged from fly ash to gulf war; fertilizers to geo-genic contribution as possible sources.

The Siwalik sediments are fluvial deposits ranging in the age from Middle Miocene (~18 Ma) to Middle Pleistocene (~0.75 Ma) formed in a foreland basin along the southern foothills of the Himalayas by detritus originating from sedimentary, igneous and metamorphic rocks upthrusted in the north along the Main Boundary Thrust (MBT) and the Main Central Thrust (MCT), (Fig. 1). Presently, the Siwaliks are exposed along the Himalayan foothills, but the majority of it lies buried under the Indo-Gangetic plain. The primary source of uranium in the ground water all over the world is uranium bearing minerals found in acidic magmatites, metamorphites and sediments. Being highly soluble and mobile U^{6+} gets liberated from such minerals when in contact with water

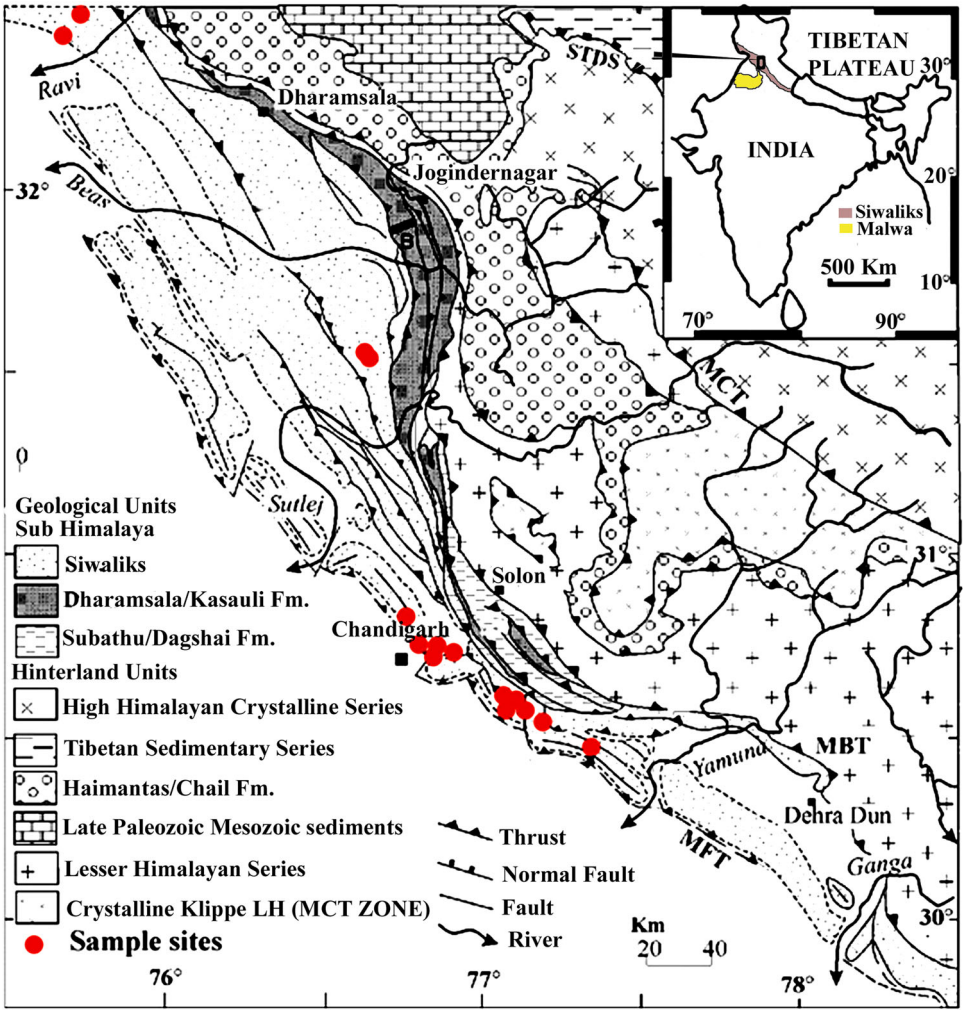

Fig. 1 Geological map of the area and the sample sites, modified after White et al. [23]

(ground/surface). U^{4+} being insoluble gets readily deposited where the conditions are reducing such as at the redox fronts of Siwalik palaeo-channels [5] or inside bones due to the decomposition of organic materials. This is usually followed by the replacement of Ca^{2+} of calcium carbonate cement in rocks or calcium phosphate of bones. An attempt was made to demonstrate the importance of Solid State Nuclear Track Detector Technique in the study of radioactivity in fossil bones from the Siwaliks [6]. Reasonable amount of uranium mineralization has been reported from the Siwalik sediments [7] as well as the Dharamsala Formation, which are precursors to the Siwaliks [8]. Uranium can get remobilized several times due to re-exposure following tectonic upheavals and/or reflow of ground/surface water through these deposits.

In the present work an attempt has been made to look into the geo-genic aspect of the problem by determining the activity of uranium in fossil and palaeosol and associated sedimentary rock samples of the Himalayan foothills, supposed to be the primary ground water recharge source of this region.

Materials and methods

Samples of fossils, palaeosol, sand and other associated sedimentary rocks were collected from the geologically well-dated (palaeomagnetically, biostratigraphically and tephrachronologically) sections of the Siwaliks exposed between Haripur Khol (H.P.) in the east and Jammu (J&K) in the west. The Siwalik fossils and sediments (palaeosols, mudstone/sandstone) have been dated using palaeomagnetic stratigraphy at Ghaggar River, Haripur Khol, Patiali Rao, Khetpurali, Parmandal and Haritalyangar [9–18]. Sections in which magnetostratigraphic studies have not been carried out have been biostratigraphically dated using fossils, for example Ramnagar (J&K) and Saketi (H.P.) sections [19].

Volcanic ash beds in India have been reported from the Ghaggar, Khetpurali near Chandigarh [20], Bali, Nagrota and Bada Khetar near Jammu. Range Rao et al. [10] dated these ash beds using zircon fission track method and came up with two dates 2.31 ± 0.54 and 2.8 ± 0.56 Ma. These acidic volcanic ash beds have been deposited as distal pyroclastic fallout in the north-western part of the Siwaliks at the end of the Neogene period. Its source has been assumed to be the Dacht-e-Nawar volcanic complex of East-Central Afghanistan, situated ~1000 km to the northwest of Jammu [21].

Table 1 gives the name of locations along with activity of uranium in Bq/kg. These sample sites have been marked using GARMIN GPS-12 and are situated well above the present-day water table of the areas. The samples were dried for about a day in an oven at 60 °C for the removal of moisture by offering a constant weight and then further pulverized and homogenized. Fifty grams of the dried, pulverized and homogenized palaeosol and fossil samples were hermetically sealed in Petri plates and kept aside for a time period of 40–45 days to establish secular equilibrium. After completion of this time period samples were analyzed at the Saha Institute of Nuclear Physics (SINP), Kolkata using CANBERRA's reverse electrode coaxial high-purity germanium detector, with resolution of 3.06 keV at 1332.5 keV and 50 % relative efficiency. The HPGe detector setup was placed inside CANBERRA MODEL 747 extra deep lead shield having 9.5 mm thick low carbon outer jacket, 10 cm thick low background lead as bulk shield with graded lining of 1 mm tin and 1.6 mm copper that prevented interference by lead X-rays. This protective assembly minimized interferences due to background radiation. Sample data was analyzed using Genie 2 k software. All electronic units and Genie 2 k software were procured from CANBERRA.

Samples were placed at 1 cm distance from the top of central HPGe detector. The energy calibration of the detector was done using ^{137}Cs, ^{60}Co and ^{133}Ba point sources. The efficiency calibration was done using the IAEA Uranium Ore Standard (Pitchblende), S-8. A weighed amount of IAEA standard (7.0 g corresponds to 102 dps) was taken in Petri dish, mixed thoroughly with silica gel so that the total weight of the standard became 50 g, equivalent to the sample size. The Petri dish containing the mixture of silica gel and IAEA standard was also hermetically sealed for 40–45 days for obtaining the secular equilibrium and was used for efficiency calibration. The density of fossil samples was about 1.7 kg m^{-3}, while that of silica gel along with the standard was around 2.1 kg m^{-3}.

For the present study, both background counting and sample counting were taken for a period of 75,000 s. ^{238}U activity was measured from 295.1 and 351.9 keV photo-peaks of ^{214}Pb, and 609.1 and 1764.2 keV photo-peaks of ^{214}Bi. As there is strong interference of ^{235}U on 186.11 keV peak of ^{226}Ra, the same has not been considered. Background correction was done for each of the samples. To validate the calibration method another 5 dps standard was prepared with IAEA Uranium Ore Standard (Pitchblende), S-8 taking 0.35 g pitchblende (corresponding to 5 dps) mixed thoroughly with silica gel so that the total weight became 50 g. This standard was analyzed in the same way for the four photo-peaks mentioned above and activity of ^{238}U has been calculated. The result has been appended in Table 1.

Results and discussion

Table 1 shows the activity of uranium in Bq/kg along with sample location information as well as the age of the samples. The ages of the samples were determined using palaeomagnetic stratigraphy as well as biostratigraphy besides zircon fission track dating method. It is observed that in general the activity of uranium in pure palaeosol and samples associated with palaeosol i.e., calcrete and nodules varied between 46 and 214 Bq/kg whereas the activity of uranium in pure fossil samples varied between 208 and 4837 Bq/kg. The lowest activity of uranium was observed in a 2.5 million years old palaeosol sample from Saketi where as the highest activity of uranium was observed in a 2 million years old mammalian fossil obtained from Moginand. The activity of uranium in top-soil from Malwa region of Punjab state has been reported [22] to be in the range of 15–27 Bq/kg which was comparable to the global average of 35 Bq/kg. The activity of uranium obtained for palaeosol in the present work was found to be two to ten times higher than the value obtained for top-soil. Further the activity of uranium obtained in the fossils in the present work was found to be nearly ten times to two hundred and thirty times higher than that of uranium in top soil.

Our results show that the fossils and the palaeosols presently situated at higher elevations, far above the present day water table, acquired anomalous quantity of uranium millions of years ago when they were buried at a deeper level where ground water containing uranium perhaps used to flow through them under anaerobic conditions.

Taking clue from the above mentioned hypothesis, it is proposed that the kind of high concentration of uranium the ground water in the Malwa region of Punjab has acquired, could only be perhaps due to uranium bearing water percolating through the recharge region (Himalayan foothills) and feeding the ground water of Punjab for millions of years. The other possibility could be the existence of uranium rich palaeo-channels of ancient rivers, that subsequently got dried up, buried below the water table of this

Table 1 Activity of U in fossils and palaeosols along with location and age

Sr. no.	Name and sample ID	Location	Siwalik formation and age	Uranium content (Bq/kg)
1	Fossil Mammalian Bone DKB-2	Devni Khadri (H.P.) 30° 31′ 08.23″ 77° 15′ 39.54″	Tatrot 2.5 Ma	208.4 ± 10.5
2	Palaeosol KHPS	Khetpurali 30° 40′ 41.50″ 76° 59′ 43.75″	Tatrot 2.5 Ma	76.1 ± 2.5
3	Fossil Elephant Jaw Bone MEJ	Masol 30° 49′ 41.78″ 76° 50′ 51.79″	Tatrot 2.5 Ma	2490.9 ± 60.3
4	Palaeosol HPS-D	Haritalyangar, H.P. 31° 31′ 27.91″ 76° 38’ 10.89″	DhokPathan 8.85 Ma	213.9 ± 2.0
5	Fossil Mammalian Bone DKB-1	Devni Khadri (H.P.) 30° 31′ 08.23″ 77° 15′ 39.54″	Tatrot 2.5 Ma	212.1 ± 8.3
6	Fossil Turtle Carapace MT	Masol 30° 49’ 41.78″ 76° 50’ 51.79″	Tatrot 2.5 Ma	683.1 ± 10.8
7	Palaeosol MPS-N	Moginand (H.P.) 30° 31′ 01.5″ 77° 13′ 14.4″	Tatrot 2.5 Ma	85.8 ± 2.3
8	Sandstone (between palaeosols) HSST	Haripur Khol (H.P.) 30° 28′ 02.52″ 77° 23′ 31.25″	Tatrot 2.5 Ma	1109.1 ± 30.2
9	Calcareous Palaeosol SPS	Saketi (H.P.) 30° 30′ 446″ 77° 15′ 180″	Tatrot2.5 Ma	45.7 ± 1.4
10	Palaeosol with nodules NPS	Nadah (Haryana) 30° 41′ 35.98″ 76° 53′ 44.18″	Pinjor 2 Ma	45.5 ± 3.5
11	Calcrete from Kanthro Village KPS	Kanthro (Saketi, H.P.) 30° 30′ 03.51″ 77° 14′ 57.69″	Tatrot 2.5 Ma	46.7 ± 3.5
12	Calcareous nodules RPS-B	Ramnagar (J&K) 32° 47′ 16.88″ 75° 18′ 54.58″	Chinji 13 Ma	60.0 ± 3.4
13	Maroon colour palaeosol HPSK	Kursai (Haritalyangar, H.P.) 31° 32′ 878″ 76° 37′ 089″	DhokPathan 9 Ma	48.4 ± 2.7
14	Large mammal bone fragment MB	Moginand village (Haryana) 30° 40′ 16.77″ 76° 56′ 23.32″	Pinjor 2 Ma	4837.4 ± 113.5
15	Large mammal bone fragment HB-K6	Kursai (Haritalyangar, H.P.) 31° 32′ 878″ 76° 37′ 089″	DhokPathan 9 Ma	1759.6 ± 31.6

Table 1 continued

Sr. no.	Name and sample ID	Location	Siwalik formation and age	Uranium content (Bq/kg)
16	Elephant Tusk	DevniKhadri (H.P.)	Tatrot	704.4 ± 24.5
	DKT	30° 31′ 08.23″	2.5 Ma	
		77° 15′ 39.54″		
17	Crocodilian Bone	Ramnagar, Kulwanta (J&K)	Chinji	1253.7 ± 26.9
	RBK	32° 46′ 00.89″	13 Ma	
		75° 24′ 44.90″		
18	Khetpurali Elephant Tooth	Khetpurali	Tatrot	1019.7 ± 23.0
	KET	30° 40′ 37.97″	2.5 Ma	
		76° 59′ 39.36″		
	Pitchblende standard 5 Bq			5.002 ± 0.003[a]

[a] Measured activity in Bq

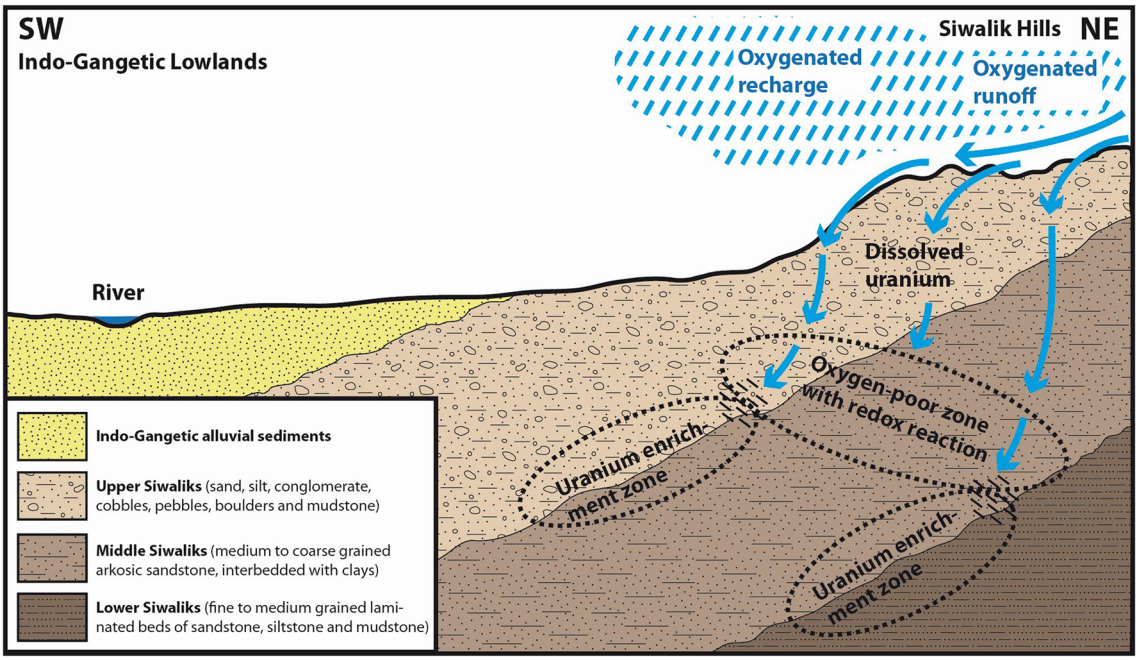

Fig. 2 Simplified geological cross-section of the suspected geochemical processes in the Siwalik sediments resulting in uranium enrichments and groundwater contamination, drawing by S. Mohr, modified after Lyle et al. [24] and Mugnier et al. [25]

region, which on and off kept feeding the ground water of this region. It is worth noting that the Malwa region being waterlogged and semiarid, experiences high surface evaporation, low mobility of the ground water, high soil salinity and high concentration of Total Dissolved Salts (TDS) in the ground water, creating anaerobic conditions, which in turn may facilitate uranium enrichment.

The Siwalik sediments, namely their sandstones, host considerable uranium mineralization [7]. The sediments underlie parts of the alluvial complex as shown in Fig. 2, which is showing a grossly simplified geochemical sketch of the relevant uranium transportation processes. It is probable that there are hydrogeological migration paths transporting geo-genic uranium from the deep-lying Siwalik sediments up to the shallow groundwater horizon of the region. This would be a sound explanation of the linear or scattered uranium hot spots in the groundwater reported.

All geological and geochemical data indicate that uranium toxicity in Punjab is probably related to the geo-genic factors of the region and perhaps the significant mobilization of uranium from the Himalayan Siwaliks to the Malwa region located in the north western part of Punjab state could be the cause of elevated level of uranium in its

ground water. Further work should be carried out in terms of source and direction of flow of ground water to arrive at definitive conclusion about the origin of the present uranium toxicity problem in the Malwa region of Punjab state.

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