

Radon, helium and uranium survey in some thermal springs located in NW Himalayas, India: mobilization by tectonic features or by geochemical barriers?

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Radon, helium and uranium measurements have been carried out in hot water springs in the Parbati and Beas valleys of Himachal Himalaya. Most of these hot springs are known as famous pilgrimage centers. The activity of dissolved radon in the liquid phase is found to vary widely, by an order of magnitude, between 10 and 750 Bq L⁻¹, whereas, the dissolved helium content in these thermal springs varies between 10 and 100 ppm. The uranium contents are low and vary from <0.01 to 5 µg L⁻¹. The measured values of radon, helium and uranium are possibly controlled by structural geology, namely the presence of pervious fault systems, and by the lithology of the leached host rocks. Redox-potential geochemical barriers cause the mobilization of uranile ions in solution (UO²⁺); the most plausible hypothesis is when the conditions are oxidising, confirming the importance of physico-chemical conditions up to the supergenic environment, to control the fluid geochemistry of the U–He–²²²Rn system. Some evidence is available from both geothermometric considerations and geochemical data which will be reported elsewhere, whereas the present study is focused on U decay series–noble gas geochemistry. The first analysis of collected ³He/⁴He data is consistent with a crustal signature at the studied thermal springs.

1. Introduction

Noble-gas geochemistry is currently recognized as a reliable tool for investigating the origin and processes involving fluids during their ascent to the surface.^{1–3} A better knowledge of the nature and evolution of deep fluids establishes a fundamental basis for geological studies,^{4–6} including fault tracing.^{7–12} In fact, fault zones are known to act as preferential pathways for migrating fluids, that can readily transmit a variety of fluids, including gases, to the surface.^{8,9,13} In some cases noble gases could act as indicators of old and recently activated fault segments.^{9–12} The He isotopic ratio can be used to discriminate component signatures that are different from crustal.^{7,14}

Geochemical studies of some thermal springs in India have been carried out in the recent past.^{15,16} The major geothermal springs in India are associated with tectonic belts, grabens and prominent fault zones. These springs are found along tectonic belts of Himalaya, the eastern and western fault zones of the Aravali range, along the Narmada-Son lineament and, moreover, a few hot springs are also found along the west coast fault zone.¹⁷

The study area of the Parbati and Beas valleys of Himachal Pradesh lies in the tectonic belt of the Himalaya. The area is drained by the main river Beas and its tributary Parbati. For the present investigation, thermal water samples were collected from Manikaran and Kasol areas in the Parbati valley and Kulu (Ramshila) and Manali (Kalath) in the Beas valley. The springs in the Parbati valley are located in the inner Himalayan range. Kasol is located at a height of 1560 m and Manikaran at 1680 m above mean sea level. The Parbati river flows almost in an E–W direction, forms the main drainage and is joined by a number of tributaries. The Parbati river originates from a

glacier in the Higher Himalaya and flows through gneisses, granite porphyroids, schists and quartzites of Varkrita, Jutogh and Chail formations and finally meets the Beas at Bhuntar (Fig. 1).

A major reverse fault system, NNW–SSE trending and eastward dipping, has been identified in Kasol.¹⁸ A dextral tear fault has been postulated to continue northward into the Beas valley, whereas another tear fault has been located near Manali. All these faults, as well as the main thrust contact, are part of the upstream of Manikaran. The fractures related to these faults appear to be fluid preferential pathways as subsidiary and tertiary channels for the upward migration of thermal fluid from deeper to shallower levels of the Parbati valley. Most of the rainfall leaves the area as direct surface runoff, a little amount infiltrates to augment the groundwater. The groundwater evolution, often very limited, occurs throughout the host-rocks, mostly of metamorphic-impervious nature, having the possibility to deepen only in the presence of faults, fractures, joints and permeable layers or zones which are favorably perched or are under slightly confined and unconfined conditions. Kulu discharge area (IND 7) also has gases escaping from depth, enriched mainly in CO₂ and able to carry other minor and trace components such as ⁴He and ²²²Rn.⁸ Direct infiltration of rainwater through joints, fractures and weathered zones is the main source of recharge to the springs. At least the following two types of water-bearing formations can be recognized.^{19,20}

1. *Fractured hard-primary rocks* of Varkrita and Jutogh Chails and window rocks of Kulu Rampur formations are characterized by springs and slight geo-gas leakage. The sectors affected by fractured systems, faults and thrusts are characterized by high secondary porosity and permeability.

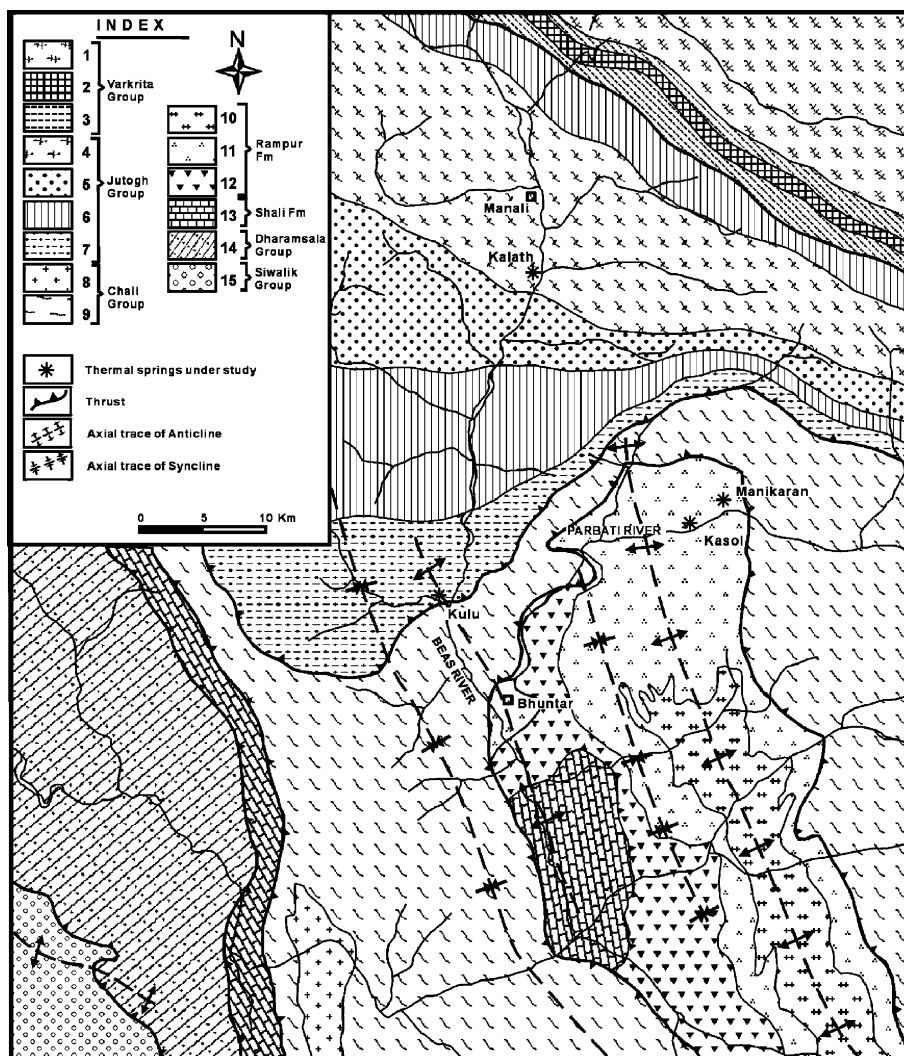


Fig. 1 Geological map of the study area. Legend: 1. Coarse grained quartzo-feldspathic biotite rich banded gneiss with tourmaline. 2. Quartz biotite garnetiferous schist interbedded with foliated micaceous quartzite. 3. Kyanite-sillimanite schist and gneiss. 4. Fine grained banded gneiss. 5. Foliated micaceous quartzite. 6. Biotiteschist with foliated micaceous quartzite. 7. Garnetiferous-biotite phyllonite and schist. 8. Intrusive Mandi granite. 9. Sericite-chlorite phyllite, quartzite, mylonitized gneiss, quartz porphyry, carbonaceous slate with thin bands of limestone. 10. Intrusive Bandal granite. 11. Rampur quartzite. 12. Volcanics. 13. Dolomite, limestone, quartzite, volcanics, shale and slate. 14. Fine grained micaceous sandstone, purple-green clay, claystone and micaceous siltstone. 15. Mainly conglomerate with medium to coarse grained sandstone, claystone and siltstone (ref. 40).

The groundwater flowing through this lithology strongly changes from one sector to another as a consequence of the segmentation of faults, as recognized elsewhere,⁷ involving isolated disconnected local bodies becoming pervious in favorable zones of jointing, fracturing and faulting. Both springs and geo-gas (enriched in Rn, He, CO₂, CH₄, etc.) leakage discharges from all sides of the individual aquifers at different elevations through fracture systems and joints and from weakened host rocks. The springs in rocks with secondary porosity show great seasonal variability in yield even within short distances.

2. *Fluvial and colluvial deposits*, lying along the lower and middle valley slopes near the confluence of two streams in the form of fans, terraces and old landslide deposits are highly porous and permeable. Therefore the springs show wide seasonal variability at the discharging sites. Fracture zones related to faults and other lineaments, characterized by sheared and crushed rocks are good locations for springs, even in rocks otherwise impermeable.^{19,20}

Several hot springs are located both in the Kulu and Manali areas in Himachal Pradesh. The famous thermal springs of Manikaran and Kasol along the Parbati valley have been known since historical times as famous pilgrimage centers. Both Manikaran and Kasol springs flow out from the Mani-

karan quartzite of the Rampur formation cropping out in the area. Here, hot water gushes out through the joint planes of the quartzite, which is traversed by several tension joints due to faulting and anticlinal folding.^{21,22} Moreover, the presence of the Jutogh thrust near Manikaran and Kasol plays an important role in the patterns of the groundwater supplying these hot springs. Possibly, groundwater goes deep down along the regional fault systems reaching reservoir temperatures which have so far never been calculated. Previous studies only refer to a generic steam-heat flow to explain the observed thermalism;²³ in this paper we tentatively fill this gap in the literature by running the geothermometric AQUACHEM²⁴ subroutines. Besides these hot springs in the Parbati valley, some others have also been investigated; which are located near Kulu (Ramshila) and Manali (Kalath), in the Beas valley.

New geochemical data (major, minor and trace elements, as well as dissolved gases and isotopes) have been collected during the March 2002 geochemical survey, allowing groundwater classification and reconstruction of water-rock interaction (WRI) processes and will be reported elsewhere, while this paper is specifically focused on the U-Th-Rn-He geochemical family and its geochemical mobility in groundwater, in the structural geology framework of this sector of the Parbati and Beas valleys of the Himachal Pradesh Himalayas.

Table 1 (a) Geochemical data for the studied thermal springs in the Parbati and Beas valleys, collected during the March 2002 geochemical survey. (b) Isotopic ratios for the 3 groups of springs sampled in March 2002

(a)

Name of spring	Group/formation	Rock types	Temp/ °C	Electr. cond./ μS cm ⁻¹	Radon/ Bq L ^{-1a}	Radon/ Bq L ^{-1b}	Helium conc. (ppm)	Uranium conc./ μg L ⁻¹	Eh/ mV	pH
IND2 Kasol 1	Rampur formation	Rampur quartzite	72.5	565	499.5 ± 2.2	579	70 ± 4.9	3.63	475	7.08
IND3 Kasol 2	Rampur formation	Rampur quartzite	69.0	545	653.5 ± 2.6	736	95 ± 5.3	4.36	515	7.08
IND4 Manikaran (Temple)	Rampur formation	Rampur quartzite	88.6	775	13.5 ± 0.4	16	40 ± 4.1	0.02	109	7.13
IND5 Manikaran (Shiv Temple)	Rampur formation	Rampur quartzite	87.4	605	21.4 ± 0.5	17	50 ± 4.4	<0.01	73	6.82
IND7 Kulu (Ramshila)	Jutogh group	Garnetiferous-biotite phyllonite and schist	35.0	6150	12.3 ± 0.3	16	9 ± 2.5	2.95	271	6.35
IND8 Manali (Kalath)	Varkrita group	Coarse grained quartzo- feldsp. biotite-rich banded gneiss with tourmaline	42.1	1881	21.2 ± 0.5	20	10 ± 2.6	0.11	293	6.44

(b)

Sample	Weight/g	³ He/ccstp g ⁻¹	⁴ He/ccstp g ⁻¹	³ He/ ⁴ He	R/Ra ^c	(R/Ra) corr. neon correction
IND2 Kasol 1	6.60	1.64E-12	1.06E-04	1.546E-08	0.011	0.011
IND5 Manikaran	6.20	4.97E-13	3.10E-05	1.602E-08	0.012	0.012
IND7 Kulu	6.39	2.27E-13	1.40E-05	1.620E-08	0.012	0.011

^a Using alpha scintillometer (by GNDU). ^b Using active charcoal collector (by INGV). ^c R/Ra is the ratio between ³He/⁴He of a generic sample and ³He/⁴He of the atmosphere.

2. Methodologies

The experimental techniques used for analysis of the collected samples are given below. For dissolved radon (²²²Rn), the collected water samples were analyzed by two different in-field methods allowing a comparison, whereas, for both dissolved helium (⁴He) and uranium measurements, the samples were collected and taken to the laboratory for analysis.

Dissolved ²²²Rn analysis

The first method (GNDU radon data in Table 1) uses an Alpha Scintillometer GBH 2002 (GBH Electronic™) with Lucas cell assembly (Fig. 2). It collects and records alpha counts after degassing/stripping the dissolved gases (with a certain efficiency being around 70%) from one litre of groundwater, during a time lapse of one minute, successively. The final alpha counts data is taken after 10 minutes, allowing the radon to reach a partial, reliable, equilibrium.^{2,25} Radon gas dissolved in the thermal waters was sucked by a pump connected to a radon

bubbler. The electronic digital counter records the alpha counts and the radon activity is measured by using an experimental calibration constant (10 counts = 1 Bq L⁻¹).

The second method (INGV radon data in Table 1) customized and calibrated² by a collaboration between the Department of Nuclear Engineering, University of Rome, Italy and INGV, Rome, Italy, was compared with other methods and direct radon-chamber measurements.² It makes use of Active Charcoal Collectors (ACC) to collect the radon stripped out from 1 litre of water by passing through 4 litres of air (99% stripping efficiency). The error is not reported in Table 1 but is within ±5%. The ACC are successively coupled with γ-spectrometry, by integrating the signal curves in the defined Region of Interest (ROI) of the decay chain of U and Th.²⁻⁵

Dissolved ⁴He analysis

Helium leak detector ASM 100 HDS (Alcatel®, France), coupled with the sniffing technique, was used for the analysis of dissolved helium in the thermal springs of the study area. The instrument hardware has been modified recently by INGV from a logarithmic voltage scale (with very low sensitivity towards low concentrations of ⁴He) to a linear voltage scale in the 1 ppm range. In this way, the ⁴He sniffer reaches a sensitivity of 0.01 ppm and it is precise for ⁴He soil gas surveys²⁶ where ⁴He content is always approximately 5.2 ppm.

Thermal spring water samples were collected in air tight bottles (100 mL). The sample bottle is connected to the sniffing probe through hypodermic syringes, which were also used to connect the sample bottle through a rubber tube to an air tight bottle containing silica gel (to absorb water vapours). ⁴He is sucked by a sniffer and signal output is displayed on a voltage scale which is later converted into ppm using calibrated curves. The concentration values of ⁴He in groundwater are later calculated (cc L⁻¹) using (i) this initial concentration of ⁴He in the free gas phase of head-space of the glass bottles, after the evacuation; (ii) the initial volume of groundwater from where the dissolved gases are extracted; (iii) Henry Law constant for ⁴He and grossly evaluated stripping efficiency, being almost 100%. The details of the method are described elsewhere.²⁶

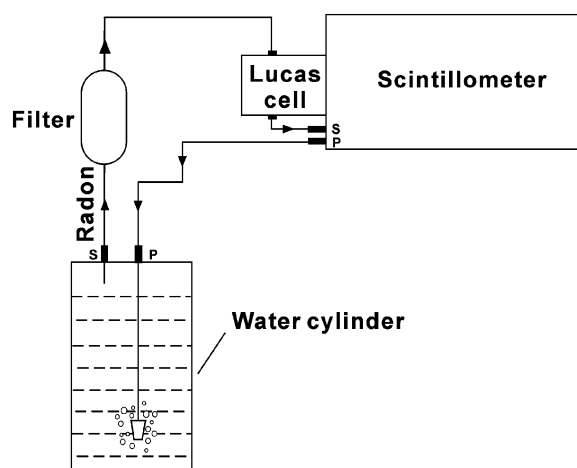


Fig. 2 Sketch diagram of the Alpha Scintillometer GBH 2002 (GBH Electronic™) with Lucas cell used in comparison with the γ-spectrometry coupled with the active charcoal collectors technique (ref. 2).

Uranium, pH, Eh and electrical conductivity measurements

The collected groundwater was also analyzed for uranium in the laboratory. The method for the determination of uranium at $\mu\text{g L}^{-1}$ level in waters is based on the direct introduction of liquid phase, without any chemical pre-treatment, into an inductively coupled plasma mass spectrometer (ICP-MS). The instrument used is a Perkin Elmer® ELAN 6000. The standard method provides a limit of detection of 2 ng L^{-1} and a good repeatability.

The pH and Eh (redox potential) of thermal spring water samples were measured *in situ* by Mettler Toledo® electrodes coupled with an ORION® 250A high impedance voltmeter. Electrical conductivity was measured together with temperature and automatic temperature compensation by a standard in-field instrument.

He isotopic ratio

Thermal waters were collected in copper tubes tightly closed at both ends by clamps; the tubes were directly connected to the high-vacuum inlet line of a MAP215 mass spectrometer ensuring negligible helium blanks compared to sample counts. $^3\text{He}/^4\text{He}$ ratios were calibrated²⁷ against an atmospheric standard ($R_a = 1.384 \times 10^{-6}$). In order to correct $^3\text{He}/^4\text{He}$ ratios for possible air contamination during sampling, both the pressure of incondensable gases and ^{20}Ne (of atmospheric origin) were measured for all samples presented in Table 1b.

3. Results and discussion

The results of the March 2002 geochemical survey are summarized in Table 1. The highest ^{222}Rn content of $653.5 \text{ (INGV} = 736) \text{ Bq L}^{-1}$ was found in Kasol 2, followed by $499.5 \text{ (INGV} = 579) \text{ Bq L}^{-1}$ for Kasol 1; the two springs are separated by a few metres only. Manikaran springs exhibited very low radon values, 13.5 and $21.4 \text{ (INGV} = 16 \text{ and } 17) \text{ Bq L}^{-1}$ as compared to the Kasol thermal springs, although both Kasol and Manikaran hot springs are associated with the same quartzite of the Rampur formation. The lowest radon value, $12.3 \text{ (INGV} = 16) \text{ Bq L}^{-1}$ was measured at Kulu (Ramshila) thermal spring, which also exhibits the lowest temperature (35.0°C).

Helium concentrations in the thermal springs are very well correlated with radon concentration, as found in other fault zones,^{2,3,7–10} with highest values of 95 ppm and 70 ppm in Kasol 2 and 1 respectively (Fig. 3). On the other hand, Kulu (Ramshila) exhibits the lowest helium value of 9 ppm in solution.

He isotopic composition of the 3 groups is found to be very similar ($R/R_a = 0.011\text{--}0.012$, Table 1b) with a strongly prevailing “crustal component”; no mantle signature is apparent, as expected for these geodynamic settings.^{7,14}

The high outlet temperatures of the Kasol and Manikaran thermal springs, ranging from 69.0 to 88.6°C , are hypothesized to be due to shallow circulation, to an anomalous geothermal gradient *i.e.*, greater than $200^\circ\text{C km}^{-1}$ in the region.²⁸ Table 2

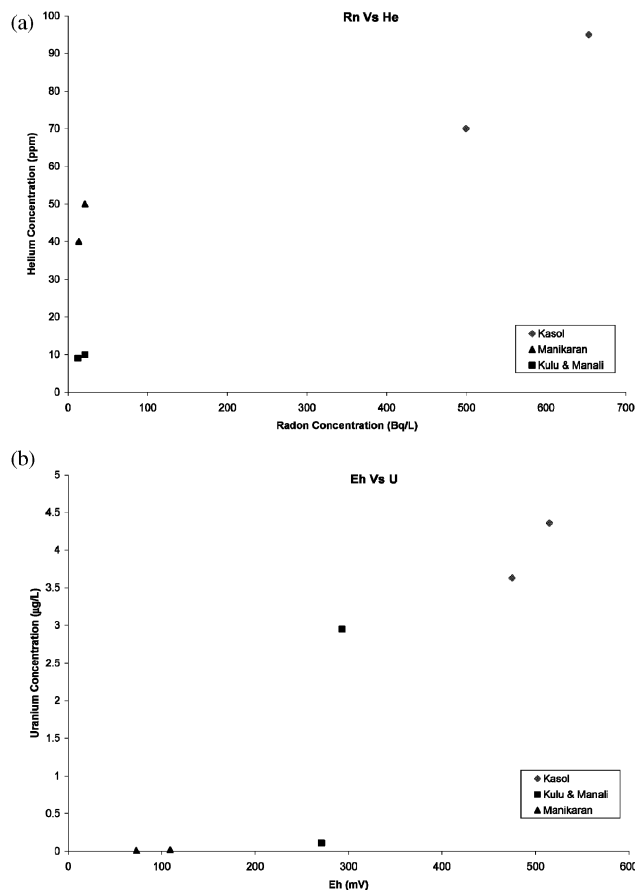


Fig. 3 Binary diagram of (a) Rn (Bq L^{-1}) versus He (ppm); (b) Eh (mV) versus uranium ($\mu\text{g L}^{-1}$) for the studied thermal springs.

displays geothermometric calculations performed using the AQUACHEM²⁴ code. For Kasol and Manikaran thermal springs, silica geothermometer calculations are based on quartz, as these waters are in thermodynamic equilibrium with this mineral, while for the same reason chalcedony has been considered for Kulu and Kalath waters. Saturation indices of silica phases have been calculated using the geochemical code PHREEQC.²⁹ Inferred deep temperatures are almost equal to or slightly higher than the discharging ones. These values could not reflect a water–rock equilibrium at depth (temperature of the reservoir), but, probably, they could be due to: (i) shallow water–rock re-equilibration (occurring at relatively low temperature) during the uprising of thermal waters through faults and fractures and (ii) mixing with cold shallow waters, causing the precipitation of silica. Na–K geothermometers give unreliable high temperatures for all waters, probably due to the absence of feldspar equilibria at depth. On the contrary, for the Manikaran springs, K–Mg and Li–Mg, while for Kulu and Kalath waters only, also Na–Li geothermometers, converge towards inferred temperatures ranging from 95 to 120°C . These values could represent a more reliable estimate of the

Table 2 Geothermometers calculated by the AQUACHEM code for the thermal reservoirs/springs sampled during the March 2002 geochemical survey

Sample	Geothermometer					
	SiO_2 (quartz)	SiO_2 (chalced.)	Na–K	K–Mg	Li–Mg	Na–Li
IND 2 Kasol 1	64	—	303	61	136	332
IND 3 Kasol 3	67	—	311	60	137	353
IND 4 Manikaran	78	—	282	92	115	376
IND 5 Manikaran	77	—	281	85	107	381
IND 7 Kulu	78	46	227	107	118	110
IND 8 Kalath	82	51	207	101	140	148

deep temperatures. The absence of any convergence of geothermometric estimation for the Kasol waters is noticeable (Table 2), which clearly indicates fast water circulation without any appreciable water–rock equilibrium. In the light of geothermometric considerations, a depth of 1–2 km can be estimated for the Manikaran, Kulu and Kalath hydrologic circuits (*i.e.* the depth of the geothermal reservoir), as found elsewhere for this kind of water geochemistry and lithology.⁷ The comparatively low outlet temperatures at Kulu (Ramshila) and Manali (Kalath) may be due to more advanced mixing with cold shallow waters.

The uranium concentration varies from <0.01 to $4.36 \mu\text{g L}^{-1}$. The two highest values of uranium *i.e.*, $4.36 \mu\text{g L}^{-1}$ and $3.63 \mu\text{g L}^{-1}$ were recorded in the hot waters of Kasol springs. Similarly, higher values of radon, up to 700 Bq L^{-1} and of helium, around 70 ppm (Table 1) were also observed in the same springs, which confirms the relationship between uranium mobilization and Rn-He presence in groundwater.^{2,3,7–10}

Generally,³ these higher values of radon and helium, as well as U, are attributed to a combination of both lithological factors (U-bearing minerals outcropping widely) and tectonic factors, also testified by diffuse fracture/shear filled veins and disseminated uranium mineralizations present in the Manikaran quartzite.^{30–32} The well known Chhinjra uranium mineralization^{33,34} lies within the investigated area. Hence, the highest values of uranium, radon and helium in spring waters in Kasol are possibly consequences of both the vicinity of radioactive source and its diffusion to the adjoining area through fractures, joints and faults *etc.*

On the other hand, despite the same lithological framework, we found lower values of uranium (<0.01 and $0.02 \mu\text{g L}^{-1}$), as well as of radon (13.5 and 21.4 Bq L^{-1}) and comparatively higher values of helium (40 and 50 ppm) in the Manikaran hot springs. This peculiar evidence could be linked to (i) higher residence time³⁵ at depth for the Manikaran groundwater, which allows both He enrichment and more prolonged and intense WRI processes allowing a partial equilibrium between rocks and circulating waters (see Table 2) and; (ii) physical-chemical conditions at the near-interface supergenic environment, *i.e.*, different acidic and reducing conditions or different geo-gas carriers for radon and helium.^{10,13}

In a completely different situation (as regards salinity and mineralization) the Ramshila hot-ferruginous spring of Kulu (IND 7 in Tables 1 and 2) is associated with the schistose rocks of the Jutogh group, often evolved in clayey secondary minerals and able to release into solution a huge quantity of salts and minor–trace elements/metals (Na–Cl chemistry, SO_4 , Al, B, As, Fe, Mn, Li, Sr, HCO_3 , *etc.*³⁶). These rocks, and most of the underlying primary rock, do not contain minerals that are relatively lower in uranium content, like apatite, zircon *etc.*, with respect to the Kasol framework. Therefore, it is not easily explainable that U, ^4He and ^{222}Rn are relatively lower than the Kasol springs.

In summary, apart from the lithological control on the U–He and Rn enrichments in solution for the three studied cases and the deepening along faults, which is different for each reservoir and maximum in both the Manikaran and Kulu areas, as emphasised by geothermometric and geochemical data, another factor must be recalled to differentiate the three case histories. This factor is the geochemical barriers generated at the supergenic environment by the peculiar fluid circulation conditions, as regards the oxidising/reducing or acidic/basic patterns, with consequent over-imposed geochemical barriers.³⁷ The thermodynamic properties of the U–Th nuclide series in solution, read in the pH–Eh binary diagrams, are important parameters to take into account when explaining mobility in the supergenic environment of the U–Th–Ra chain radionuclides.

While the pH values of the studied waters do not show a wide range of variation, *i.e.*, from 6.35 to 7.13, the redox

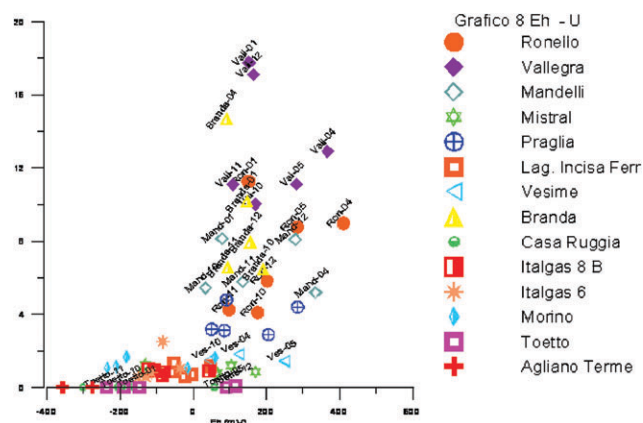


Fig. 4 Binary diagram of Uranium (ppb or $\mu\text{g L}^{-1}$) versus Eh (mV) of 15 groundwater sites monitored on a monthly basis in the Nizza Monferrato fault zone (after ref. 38), located within the Tertiary Piemonte Basin (NW Italy, Alpine range).

potential values (100–500 mV, Table 1, Fig. 3b) exhibit a notably wide range, as a consequence of both the presence of clayey-organic sediments (maximum in Kulu) and the presence of more or less reducing geo-gas rising up *i.e.*, reducing conditions at Manikaran, due to the presence of up to 0.5 ppm of H_2S and up to around $15 \mu\text{mol L}^{-1}$ of CH_4 as well as oxidising conditions at Kasol being lower than the uprising reducing geo-gas. The power of Eh conditions to mobilize U in solution, with consequent increase of radionuclides in solution, is well known in literature and by new experimental data analysis along other fault zones (Fig. 4 for the Nizza Monferrato, Tertiary Piemonte Basin, Italy fault zone³⁸). This occurs for radon also due to the lack of other geo-gas carriers in the area, as observed in the Tertiary Piemonte Basin as a whole. The geo-gas seems to be carried by CO_2 which in the studied area, as dissolved phase, reaches 25 mmol L^{-1} in Manikaran with respect to the content of 1–3 mmol L^{-1} in Kasol.

In general, the solubility of the uranile ion (UO^{2+}), coming from uranite or from monazite, increases with an increase in pH. However, in the present case, water pH does not show huge variations. Therefore, the more powerful parameter is Eh (redox potential) which controls the mobility of U and, consequently, results in the enrichment of U, helium and radon. Due to this, Kasol shows maximum values of U, radon and helium with respect to the other studied sites.

Moreover, we could suggest that, being minimum at Kasol, the “stripping/diluting effect”¹⁰ of the concurrent coming geo-gas (CO_2 mainly) toward the rest of rising trace gaseous components, as Rn and He, could have maximum dissolved He and Rn, where dissolved and free CO_2 is at a minimum, given parity of lithological background.

4. Conclusions

A group of thermal springs located in the Himachal Himalaya region (NW Himalaya, India), mostly inside the Parbati and Beas valleys, along seismically active fault systems and complex folded structures, were studied in the frame of an India–Italy joint geochemical survey in March 2002.

Apart from the geochemical settings as a whole (major, minor, trace elements as well as dissolved gases and isotopic composition), results have been only slightly outlined in this paper and will be discussed in future in wider framework. Here we focused on the relationships between uranium, radon and helium concentrations in these thermal waters and their geodynamical settings as well as their possible WRI processes, causing the observed U–He–Rn contents.

From the above study, two distinct hydrogeological-geochemical units can be discerned in the geothermal area of Manikaran and Kasol in the Parbati valley. In the Manikaran area, the unit comprising river terraces, valley fill material and alluvial cones contains relatively low salinity, immature waters, with not so fast circulation, characterizing a low enthalpy geothermal reservoir. Groundwaters here are circulating under unconfined conditions and are enriched along local faults by uprising geo-gas (CO_2 , H_2S and CH_4) able to drop the redox conditions as measured at the groundwater discharge sites. Here the U and Rn contents are relatively low. The He/Rn at Manikaran is instead higher than the other sites probably due to (i) higher residence time³⁵ at depth for the Manikaran groundwater, which allows both He enrichment and more prolonged and intense WRI processes, allowing a partial equilibrium between rocks and circulating waters (see Table 2) and; (ii) physical-chemical conditions at the near-interface supergenic environment, *i.e.*, different acidic and reducing conditions or different geo-gas carriers for radon and helium. WRI processes affecting the Manikaran waters are different to the Kasol ones, where a typical oxidising geochemical barrier³⁰ with UO^{2+} mobilization was envisaged, followed by enrichment of the U decay radionuclides (^{222}Rn and ^4He).

On the other hand, in fact, the Kasol area, where hard rock quartzites and associated phyllites are cropping out, is characterized by immature waters characterized by a fast circulation; waters do not reach any equilibrium with leached rocks and do not have time to become reducing and geo-gas enriched. The ascent rate of groundwater is allowed by the faulted host-rock: quartzite is highly jointed and fractured and forms a thick network of interconnected joints and fissures which allow radon and helium to migrate easily to the surface. In any case the relatively higher presence of uranium (up to 5 ppb) is linked to the oxidising conditions, which allow the mobilization of U as UO^{2+} , otherwise immobile in the Manikaran and Kulu pH-Eh settings.

The reservoir and geothermometric modeling kinetic patterns of the 3 cases, together with the rest of the geochemical data,³⁶ have yet to be fully explored. The high heat flow, strictly related to radioactive elements in the crust,³⁹ as ^4He and therefore ^{222}Rn , is closely related to the typical regional Alpine-Himalayan values of high heat generation. Further heat-flow investigations on the studied thermal waters area, together with the geochemical survey performed along the Parbati and Beas valleys, will have to be carried out to better understand the geodynamical settings of this sector of India.

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References

- G. M. Banwell and R. R. Parizek, *J. Geophys. Res.*, 1988, **93**, 355.
- C. Mancini, F. Quattrocchi, C. Guadoni, L. Pizzino and B. Porfidia, *Ann. Geofis.*, 2000, **43**(1), 31.
- F. Quattrocchi, M. Guerra, L. Pizzino and S. Lombardi, *Nuovo Cimento*, 1999, **22** C(3–4), 309.
- L. L. Chyi, T. J. Quick, T. F. Yang and C.-H. Chen, *Terr. Atmos. Oceanic Sci.*, 2005, (accepted).
- T. F. Yang, V. Walia, L. L. Chyi, C. C. Fu, C.-H. Chen, T. K. Liu, S. R. Song, C. Y. Lee and M. Lee, *Radiat. Meas.*, 2005, (accepted).
- V. Walia, H. S. Virk, T. F. Yang, S. Mahajan and B. S. Bajwa, *Terr. Atmos. Oceanic Sci.*, 2005, (accepted).
- M. Angelone, C. Gasparini, M. Guerra, S. Lombardi, L. Pizzino, F. Quattrocchi, E. Sacchi and G. M. Zuppi, *Appl. Geochem.*, 2005, **20**, 317.
- L. Pizzino, G. Galli, C. Mancini, F. Quattrocchi and P. Scarlato, *Nat. Hazards*, 2002, **27**(3), 257.
- L. Pizzino, P. Burrato, F. Quattrocchi and G. Valensise, *J. Seismol.*, 2004, **8**, 363.
- S. Salvi, F. Quattrocchi, M. Angelone, C. A. Brunori, A. Billi, F. Buongiorno, F. Doumaz, R. Funicello, M. Guerra, S. Lombardi, G. Mele, L. Pizzino and F. Salvini, *Nat. Hazards*, 1999, **20**, 255.
- C. C. Fu, T. F. Yang, V. Walia and C.-H. Chen, *Geochem. J.*, 2005, (accepted).
- V. Walia, T. C. Su, C. C. Fu and T. F. Yang, *Radiat. Meas.*, 2005, DOI: 10.1016/j.rad.meas.2005.04.011.
- T. F. Yang, C. Y. Chou, C.-H. Chen, L. L. Chyi and J. H. Jiang, *Radiat. Meas.*, 2003, **36**, 425.
- T. F. Yang, C.-H. Chen, R. L. Tien, S. R. Song and T. K. Liu, *Radiat. Meas.*, 2003, **36**, 343.
- J. R. Singh, *Indian Miner.*, 1989, **43**, 7.
- R. Singh and A. K. Bandyopadhyay, *Indian Miner.*, 1995, **49**, 55.
- S. C. Sharma, *Proc. Rare Gas Geochemistry*, ed. H. S. Virk, G.N.D. University, Amritsar, 1997, p. 193.
- R. Shanker, R. Pandi and C. L. Arora *et al.*, *Proceedings of the Second United Nations Symposium on the Development and Use of Geothermal Resources*, San Francisco, USERDA and USGS, Washington, D.C., 1975, p. 245.
- S. K. Bartarya, *Himalayan Geol.*, 1995, **6**, 17.
- V. M. Choubey, S. K. Bartarya and R. C. Ramola, *Environ. Geol.*, 2000, **39**, 523.
- B. L. Jangi, Gyan Prakash, K. J. S. Dua, J. L. Tussu, D. B. Dhimri and C. S. Pathak, *Proceedings of the Second United Nations Symposium on the Development and Use of Geothermal Resources*, San Francisco, USERDA and USGS, Washington, D.C., 1975, vol. 2, p. 1085.
- S. V. Srikantia and O. N. Bhargava, *Geol. Soc. India*, Geological Society of India Publication, Bangalore, India, 1988, 406.
- V. P. Sharma, *Geology of Kulu-Rampur belt*, H.P., *Mem.-Geol. Soc. India*, 1977, **106**, 235.
- Aquachem v.3.7, *Aqueous Geochemical Analysis, Plotting and Modeling*, Waterloo Hydrogeologic, Inc., 1998–1999.
- V. Walia, B. S. Bajwa and H. S. Virk, *J. Environ. Monit.*, 2003, **5**, 122.
- D. G. Jones, J. C. Baubron, S. E. Baubien, R. Bencini, C. Cardellini, D. Granieri, S. Lombardi, L. Penner, F. Quattrocchi, M. H. Strutt and N. Voltattorni, *Monitoring and characterisation of near-surface processes: soil gas monitoring*, in “*The IAEA Weyburn CO₂ monitoring and storage project*”, British Geological Survey Commissioned Report RR/05/033, Keyworth, Nottingham, 2005, 54 pp.
- Terrestrial Heat Flow and lithosphere structure*, ed. V. Cermak, L. Bodri and L. Rybach, Springer, Berlin, 1991, pp. 23–69.
- R. Shanker, *Indian Miner.*, 1988, **42**, 89.
- L. D. Parkhurst and C. A. J. Appelo, *User's guide to PHREEQC*, (version 2), USGS Water-Resources Investigations report, US Department of Interior, US Geological Survey, Denver, CO, 1999, 99–4259.
- V. M. Choubey, S. K. Bartarya, N. K. Saini and R. C. Ramola, *Environ. Geol.*, 2001, **40**, 257.
- N. P. Singh, S. Singh and H. S. Virk, *Nucl. Geophys.*, 1989, **3**, 119.
- H. S. Virk, *Curr. Sci.*, 1997, **73**, 536.
- G. R. N. Das, T. N. Parthasarathy and P. C. Taneja, *Proc. Indian Sci. Acad.*, 1972, **37**, 267.
- G. R. N. Das, T. N. Parthasarathy, P. C. Taneja and N. V. A. S. Perumal, *J. Geol. Soc. India*, 1979, **20**, 95.
- T. Torgersen and W. B. Clarke, *Earth Planet. Sci. Lett.*, 1997, **84**, 345–355.
- INGV, 2002, unpublished data.
- A. I. Perel'man, *Appl. Geochem.*, 1986, **1**, 669–680.
- F. Quattrocchi, R. Favara, G. Papasso, L. Pizzino, R. Bencini, D. Cinti, G. Galli, F. Grassa, S. Francophone and G. Volpicelli, *Nat. Hazard Earth Syst. Sci.*, 2003, **3**, 269–277.
- W. B. Clarke, W. J. Jenkins and Z. Top, *Int. J. Appl. Radiat. Isot.*, 1976, **27**, 515–522.
- D. K. Misra and V. C. Tewari, *Geosci. J.*, 1988, **9**, 153.