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Research Article

Deciphering Hydrochemistry and Fluid-Mineral Equilibria from Characteristic Low-Enthalpy Geothermal Waters of Himalaya and Eastern India

Archisman Dutta[a, c], Ahsan Absar[d], Parashar Mishra*[a, b], Sayandeep Banerjee[b], Pankaj Saini[e], Vishal V. Sakhare[f], Ajay Srivastava[a], Ayodhya Prasad Thapliyal[a] and Biswajit Ray[c]

[a]*Geological Survey of India, Northern Region, Lucknow-226024(UP), India.*
[b]*Department of Geology, Institute of Science, BHU, Varanasi-221005(UP), India.*
[c]*Department of Chemistry, Institute of Science, BHU, Varanasi-221005(UP), India.*
[d]*ONGC Energy Centre, SCOPE Minar, Lakshmi Nagar, Delhi-110092(DL), India.*
[e]*Geological Survey of India, Gangtok-737101(SK), India.*
[f]*Geological Survey of India, Central Region, Nagpur-440006(MS), India.*
*(*Corresponding author, E-mail: parashar.mishra@gsi.gov.in)*

Abstract

Geothermal energy, with associated low-carbon emissions compared to conventional fossil fuels, is presently one of the potential renewable energy sources. The decryption of the chemistry of thermal manifestations of geothermal systems, in terms of relative abundances of various chemical facies, is mandatory before launching systematic geothermal exploration in a given area as it provides valuable information on the basic characteristics of the geothermal reservoir. For the present study, three clusters of hot springs have been selected -1) Shyok-Nubra valleys geothermal prospect in NW Himalaya, 2) Subansiri valley geothermal areas in NE Himalaya and 3) the hot spring system occurring along Munger-Saharsa Ridge Fault Zone (MSRF) in the Eastern part of India. The study of these geothermal systems with low to moderately high temperatures of $35-75°C$, has been carried out with two main objectives. The first objective is to estimate reservoir temperature and other characteristics in the three above-mentioned hot spring groups using conventional chemical geothermometry techniques for the first-hand assessment of the quality of the geothermal resource and postulating its possible application. The second objective of the study is to understand the distribution of stable isotopes (δ^2H and $\delta^{18}O$) in the studied geothermal systems to reveal reservoir-related hydrological aspects of the geothermal system.

The three groups of hot springs show large variations in total dissolved solids (TDS) from <150 to about 1800 mg/L. Enigmatic and inexplicably low TDS values of 126 to 150 mg/L are reported from the three Bhimband hot springs along MSRF Zone. These springs discharge $Ca-Mg-HCO_3$ type water. The dearth of dissolved ionic species in these springs indicates lack of water-rock interaction in Bhimband area. Silica is the most abundant solute species and corresponds to a reservoir temperature of about 75°C. It may be inferred that relatively inert lithology in the reservoir and very high water-to-rock ratio might have rendered the observed chemical signatures to Bhimband hot springs. Shyok-Nubra springs at Changlung and Panamik are predominantly $Na-HCO_3$ type with TDS values of about 1700 and 550 mg/L, respectively. These hot springs give an indication of water-rock interaction in different types of lithology at temperatures of 120 to >150°C resulting in the observed chemical differences between the two. Thermal water at Taksing in NE Himalaya is also $Na-HCO_3$ type. Chetu hot spring with TDS of >1100 mg/L is anomalous in two ways. First, it has a very high SO_4 content of 358 mg/L and second, it has the lowest silica value. There is a possibility that the Chetu hot spring has its chemistry influenced by the dissolution of sulfates of Ca and Na. Preliminary stable isotope study indicates that the geothermal fluid is derived from meteoric sources. The lack of any indication for positive $\delta^{18}O$-shift suggests that reservoir temperatures of the springs are generally low.

Keywords: Thermal Waters, Hydrogeochemistry, Reservoir Temperature, Stable Isotopes, Himalayas, Bihar

Introduction

The geothermal resources of India are primarily low−enthalpic, non−volcanogenic and liquid−dominated systems (Das *et al.,* 2021). Geological Survey of India has classified seven principal geothermal provinces in India which are (1) Himalayan geothermal province, (2) Geothermal area along SONATA (SON−NARMADA−TAPI) lineament, (3) West Coast Geothermal Belt, (4) Cambay Geothermal Province, (5) Mahanadi Valley Geothermal Belt (6) Godavari Valley Geothermal Belt, and Sohna Geothermal Area in Aravalli ranges. About 350 hot springs have been identified in the country (Chatterjee *et al.,* 2022; Das *et al.,* 2022; Dutta *et al.,* 2023).

Hot springs are surface manifestations of various subsurface processes and their study allows developing an understanding of subsurface geochemical and hydrological processes (Sarolkar, 2020). The prevalent geothermal conditions in an area are
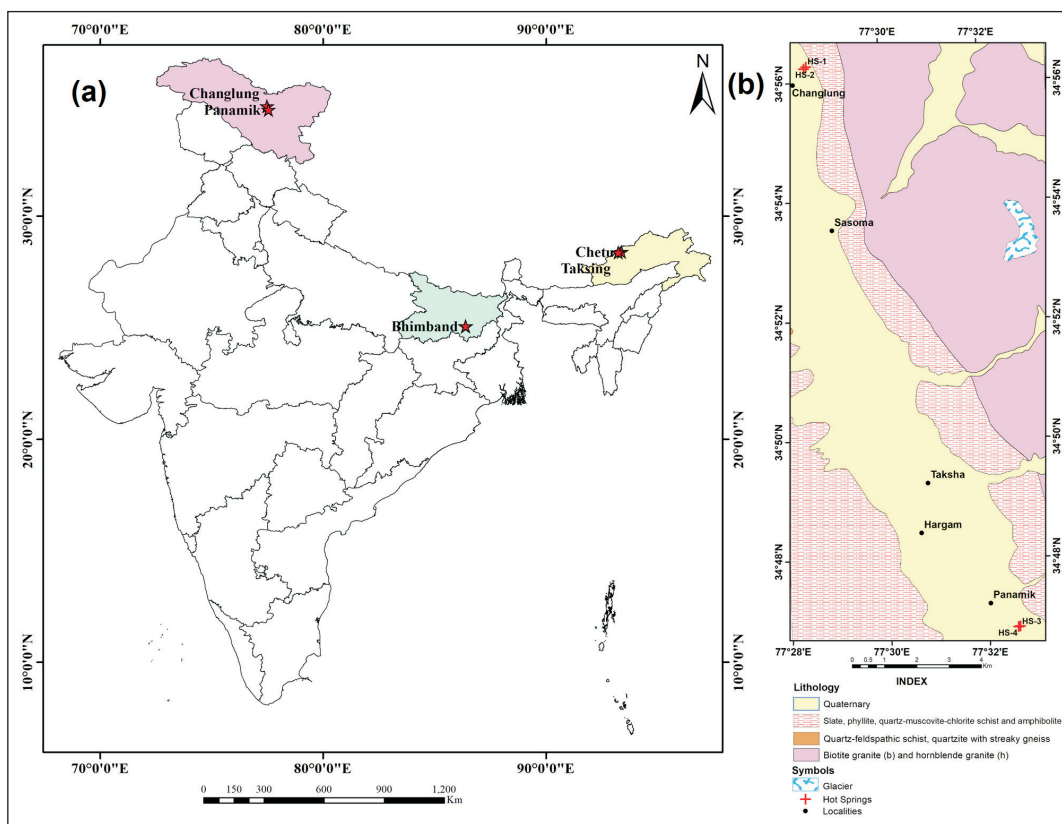
Fig.1. (a) Geothermal Provinces of the studied hot springs; (b) Geology of Nubra−Shyok Valley, Ladakh.

unravelled through the estimation of subsurface reservoir temperature, hydrological conditions and the extent of water-rock interaction. All these studies may be carried out by having a strong database on the chemistry of geothermal fluids, composition of geothermal deposits and stable isotope chemistry. Having this information on the given geothermal system is absolutely crucial for developing a preliminary conceptual model of the system preceding the deep drilling programme (D'Amore and Arnörsson, 2000; Nicholson, 1993; Sarolkar, 2020). For the present study, three hot springs regions have been selected, and are Shyok−Nubra valleys geothermal prospect in NW Himalaya, Subansiri valley geothermal areas in NE Himalaya and the hot spring system occurring along Munger−Saharsa Ridge Fault Zone (MSRF) in the Eastern part of India (Fig.1a). The present study reveals the origin of solutes in thermal waters and provides information on subsurface reservoir temperatures. Stable isotope studies have been used for getting information on the origin of thermal waters.

Study Area and Methodology

The geology of an area has a direct bearing on the chemistry of hot springs. It is the extent of water-rock interaction and the lithology of the area that determine the chemistry of hot springs. It is therefore imperative to have an idea of the rocks in the area from which hot springs acquire their solutes. The geology of the area around the thermal springs of Changlung and Panamik in Shyok−Nubra Valleys (Fig.1b) is dominated by Karakoram Granite of Palaeozoic age and Shyok Volcanics of Cretaceous - Eocene age. Karakoram Granite and Shyok Volcanics are separated by Shyok - Nubra Thrust trending NW-SE (Mishra and Debnath, 2018; Dutta *et al.,* 2023b; Mishra *et al.*, 2023). Chetu and Taksing hot springs are

located in the Crystalline Belt of the Se La Group of the Palaeoproterozoic age. These rocks consist of schist, gneiss, migmatites and other metamorphites (Dutta and Gupta, 2022; Chatterjee, *et al.,* 2022; Fig.2a). Another hot spring is located at Maza which could not be accessed and sampled because of ground realities. The springs of Bhimband area in Munger are located in quartzite and phyllite of Munger Group belonging to the Mesoproterozoic age (Dutta *et al.,* 2023a; Fig.2b).

Geothermal water sampling is done in 500 ml High-Density Polypropylene bottles for major cation and anion analysis. For stable isotopes (^{2}H, ^{18}O) analyses, 60 ml plastic bottles with double lids are used. Some of the physicochemical parameters like pH, EC, TDS, salinity, alkalinity, total hardness, Ca^{2+}, Mg^{2+}, and dissolved oxygen are measured on-site using calibrated Systronics water analyzer instrument and standard titrimetric techniques. Other chemical parameters like Na^+, K^+, and Li^+ are estimated using Systronics Flame Photometer. Chloride is determined by argentometric titration. Sulphate SO_4^{2-}, SiO_2, NO_3^-, PO_4^{3-} are measured by Thermo (type UV330) UV spectrophotometer. The inorganic charge balance error lies within ±5% which justifies the precision of the analysis. The stable isotopes δ^2H, and δ^{18}O are determined by an Isotope Ratio Mass Spectrometer (Isoprime 100) in the Isotope Hydrology Section, IRAD, BARC. Rare earth elements and trace elements are analysed using VARIAN 820 MS ICPMS instrument.

The chemical analyses of representative thermal water samples from the three geothermal provinces are given in Table 1. The temperature of the springs samples ranges from 35 to 75°C with pH values in the range of 5.37 - 8.44. Total Dissolved Solids (TDS) of these springs have been derived by summing up concentrations of major cations and anions. TDS shows a very wide range of
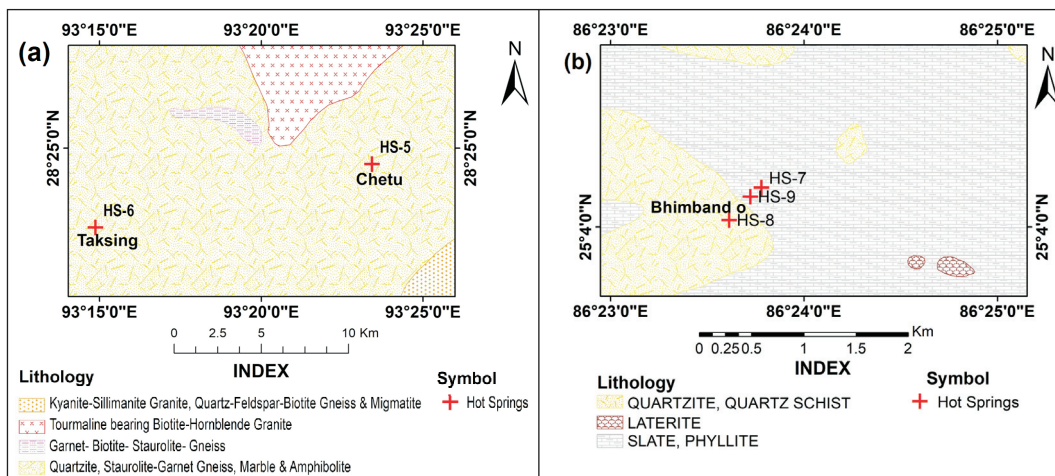
Fig.2. (a) Geology of the area around Chetu and Taksing in Subansiri Valley; (b) Geological map of Bhimband area, Bihar.

variations from 17 to 1695 mg/L. Himalayan thermal springs of Changlung and Chetu show the highest TDS value while Bhimband springs along MSRF zone show an extremely low TDS value.

Results and Discussion

Hydrogeochemical Facies

Anomalously large overall variations in TDS, particularly as observed in Changlung and Panamik geothermal areas of the Nubra Valley, manifest not only lithological differences but also the rate of ionic exchange, cumulative water to rock ratio and the residence time of the fluid underground. In Himalayan geothermal areas, Na is the most abundant cation but in MSRF zone it is enigmatically low at 1 mg/L. The latter, however, has a relative abundance of Ca and Mg. Among the anions, HCO_3 is most abundant. The dissolved silica concentration ranges between 28 and 118 mg/L (Table 1).

The Piper's Trilinear Diagram (Fig.3a) illustrates different chemical types of thermal waters. Waters discharging along MSRF zone are of Ca−Mg−HCO_3 type like most of meteoric waters (Dutta *et al.,* 2023a). Geothermal waters in north-west and north-east Himalayas are found to be Na−HCO_3 type, except Chetu (HS−5) which is Na−Cl type (Fig.3). Relatively high SO_4 content in springs HS−3, 4 and 5 (Table 1) suggests extraction of SO_4^{2-} from oxidation of pyrite minerals of rocks and/or through near surface oxidation of H_2S. Dissolution of some sulphate mineral as gypsum and thenardite may also account for high SO_4^{2-} concentration (Dutta *et al.,* 2023a).

The HCO_3 content in Changlung is about 5 times higher than that in Panamik. This is clearly a case of ion exchange in different chemical environments and lithological settings. The simplest explanation could be that Changlung has higher dissolved CO_2 which govern saturation of thermal waters with respect to calcite mineral, as per the following equilibrium reactions:

$$H_2O + CO_2 \rightleftharpoons H_2CO_3 \rightleftharpoons H^+ + HCO_3^-$$
$$Ca^{2+} + HCO_3^- \rightleftharpoons CaCO_3 + H^+$$
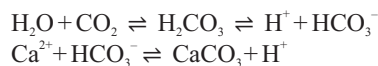
Deposition of calcite is observed under near surface conditions at Panamik. Calcite deposition, in case taking place under surface condition may however, indicate boiling. This would imply that while subsurface boiling is taking place at Changlung, it does not occur at Panamik. Changlung fluids represent less saturated condition with respect to calcite at the surface as the bulk of calcite has already been deposited subsurface. This assumption is supported by the fact that the SiO_2 content in Changlung thermal fluid is higher than that in the Panamik thermal fluid. This is because sub-surface boiling causes loss of CO_2 resulting in enhancing the fluid pH and concomitant increase in SiO_2 solubility. This, in turn, will reduce the solubility of calcite causing its supersaturation in thermal fluid. Deposition of calcite may result in scaling problems (Nicholson, 1993).

Origin of Solutes

Globally most geothermal systems give unequivocal evidence of having been derived from meteoric water. This simply implies that solutes in both thermal and non-thermal waters are acquired later during the course of the geochemical evolution of the hydrological system (Aher *et al.,* 2019). In determining the source

Table 1: Concentration of Major Cations and Anions in the three Geothermal Areas Studied

| Sample ID	Location Name	Temp. (°C)	Concentrations in mg/L										
			Calculated TDS	HCO_3^-	CO_3^{2-}	Cl^-	F^-	SO_4^{2-}	Ca^{2+}	Mg^{2+}	Na^+	K^+	SiO_2
HS-1	Changlung	69	1695	1050	0	80	12.5	53	8	6	425	13	116
HS-2	Changlung	75	1635	1060	0	85	12.5	55	8	5	430	13	118
HS-3	Panamik	75	485	210	0	15	12.5	101	12	<1	125	10	76
HS-4	Panamik	75	470	215	0	14	1	99	11	1	122	10	73
HS-5	Chetu	35	1132	326	0	100	2	358	131	7	182	9	28
HS-6	Taksing	57	582	380	30	36	7	26	15	6	177	7	48
HS-7	Bhimband-1	65	149	44	0	12	<0.3	15	22	4	1	<1	51
HS-8	Bhimband-2	63	122	29	0	10	<0.3	18	19	2	1	<1	45
HS-9	Bhimband-3	64	132	34	0	13	<0.3	16	8	11	1	<1	49
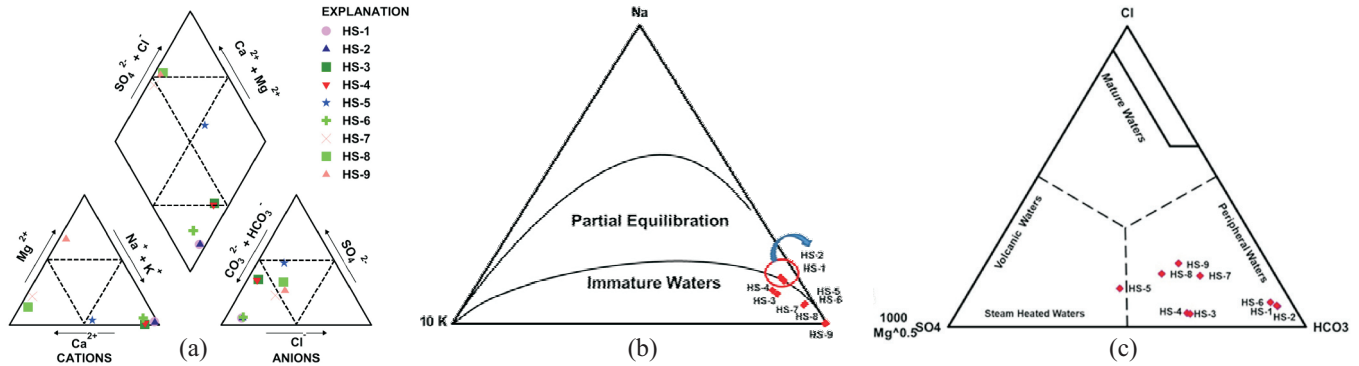
Fig.3. (a) Piper's Trilinear Diagram showing various chemical types of geothermal waters based on relative abundances of major cations and anions; (b) and (c) Giggenbach's Triangular Diagrams for major cations and anions, respectively.

of dissolved chemical species in solutions with varying TDS concentrations (from less than 100 to >10,000 mg/L), we naturally have to invoke processes like dissolution of water-soluble minerals and ion exchange as a result of water - rock interaction. Marked changes in the fluid chemistry may occur through the dissolution of sulfates, carbonates and chlorides. This may happen in both the cold and the hot water systems but may be restricted to only zones of the occurrence of these minerals. The bulk of chemical mass transport, however, takes place as a consequence of water - rock interaction. Water - rock interaction itself depends on parameters like the lithology in the reaction zone, temperature, water to rock ratio and duration of contact. The role of lithology is obvious (Pophare *et al.,* 2018; Singh *et al.,* 2022). Chemistry of thermal springs occurring in a carbonate terrain will obviously be different from those occurring in a granitic or basaltic terrain.

Temperature also plays an important role and most of the times solutions are more reactive in high temperature zones than in regions of normal thermal gradient. This explains the variations in the chemistry of cold and hot springs issuing from the similar terrain. Water to rock ratio is a parameter which itself is dependent on permeability in the reservoir rock. Higher permeability will result in larger amount of water to flow through the system resulting in high water to rock ratio. Restricted permeability zones, on the other hand, would result in relatively low water to rock ratio. Duration of rock-water contact, obviously, is another parameter controlling acquisition of solutes by geothermal solutions.

There is hardly any doubt that concentrations of dissolved solutes in the three geothermal regions being studied here are the result of solute acquisition under elevated temperature conditions under both silicate and carbonate dominated lithologies.While Himalayan hot springs tend to have formed in silicates-dominated reservoir rocks, those at Bhimband have evolved under a different geochemical environment.

In this context of the origin of solutes, it is imperative to mention about the anomalous chemical composition of the three Bhimband hot springs occurring in the vicinity of Munger Saharsa Ridge Fault (MSRF). In spite of moderately high surface discharge temperatures of around 65°C, similar to those of the Himalayan springs, Munger hot springs have an enigmatically low TDS of 120 to 150 mg/L. Silica concentration of 45 to 51 mg/L is followed by that of HCO_3 (29 to 44 mg/L). Among the cations Ca and Mg concentrations vary from <10 mg/L to 22 mg/L. Na records enigmatically low value of 1 mg/L. Geologically, the area consists of quartzite, slate, phyllite and laterite (Fig.3a). It seems that, hydrologically, the Munger hot springs have remained more or less

confined to chemically inert rock like quartzite. Low water-rock interaction in quartzite is also suggested to be a cause for lower silica content of about 50 mg/L. Moreover, the water-to-rock ratio must have been very high resulting in low relative concentration of cations. A Na value of 1 mg/L suggests that Munger fluids have not interacted with any sodium-bearing rock.

Mutual Relationships between Major Cations and Anions

The Na−K−Mg diagram (Fig.3b) is a vital tool to determine which waters are in geochemical equilibrium with rock-forming minerals and hence are ideal candidates to be used for geothermometry calculations for estimating reservoir temperatures. Most of the thermal waters of study areas fall under the Immature Water Zone close to the Mg vertex. This indicates that either these thermal waters have encountered vast lateral flow or during the course of their ascent got mixed with shallow groundwater table. These waters are also prone to lose their reservoir's identity during the course of lateral flow. Reservoir temperatures estimated by applying either cation ratio geothermometers or mineral composition geothermometers will be erroneous.

Thermal waters HS−1, 2, 5, and 6 (Table 1), being closer to phase boundary between the Partial Equilibrium Zone and the Immature Waters Zone may be used for the first order estimates of the reservoir temperature. In the Triangular Diagram Cl−SO_4−HCO_3 (Fig.3c) samples plot is close to the HCO_3^- apex. HCO_3/Cl ratios varying from 2.6 to 15.3 (from data given in Table - 1) suggest that excess HCO_3 may have been incorporated due to a combination of silicate rock weathering and carbonate dissolution. Large scale variation of Cl which is a conservative element (unaffected by dissolution-precipitation processes) from 10 to 100 mg/L indicates the diverse reservoir chemistry with limited scope of boiling except for the Changlung springs which are evidenced to be boiling sub-surface.

Chemical Geothermometry

Geothermometers are based on the solubility of specific minerals and temperature-controlled equilibrium during ion exchange reactions between certain minerals in thermal waters (Nicholson, 1993; Dutta and Gupta, 2022). Table 2 tabulates the results of the application of some commonly used geothermometers.

Thermometries that have been used (Table 2) are basically of two types: silica geothermometries and cation geothermometries.

Table 2: Reservoir Temperature estimates using various chemical geothermometries.

Sample ID	Surface Temp. (°C)	Reservoir Temperature in °C as estimated using various chemical geothermometries					
		Chalcedony conductive	Quartz conductive	Quartz adiabatic	Na-K-Ca Fournier and Truesdell	Na/K Giggenbach	K/Mg Giggenbach
HS-1	69	120	146	140	144	153	80
HS-2	75	121	147	141	144	152	82
HS-3	75	94	123	120	116	215	99
HS-4	75	92	121	118	118	217	96
HS-5	35	45	77	81	62	182	69
HS-6	57	70	100	101	101	168	65
HS-7	65	73	103	103	Not Applicable	Not Applicable	Not Applicable
HS-8	63	67	97	98	Not Applicable	Not Applicable	Not Applicable
HS-9	64	71	101	102	Not Applicable	Not Applicable	Not Applicable

Silica geothermometries relate silica concentrations in geothermal fluid directly to reservoir temperature. For groundwater and low-temperature hot springs relating silica concentration to chalcedony seems to work in most of the cases. For higher temperature systems, it seems appropriate to use quartz based thermometries. For systems where there is no boiling, the equation to be used is that for conductive cooling and in case of boiling springs or when there is evidence of subsurface boiling, the equation to be used is that of adiabatic cooling.

It is very significant to know that, in general, silica thermometries give reservoir temperature estimates at the lower side. This is because silica concentration measured at the surface, either in a hot spring or a well, is always lower than the silica concentration at the deeper or the reservoir level. This happens as the deeper silica concentration is prone to decrement on account of reservoir-related processes, such as, boiling (causing precipitation of silica) and mixing of ascending hot fluid with shallow groundwater. Therefore, generally speaking, reservoir temperature estimated using silica geothermometries are most likely to exist in the system.

Cation geothermometers are generally applicable to those geothermal systems in which rock forming minerals are in partial or full equilibrium with the geothermal fluids in accordance with the Na−K−Mg diagram (Fig.3a). Generally speaking, hot water systems with reservoir temperature around 150°C may be considered appropriate for the application of Na−K and Na−K−Ca cation geothermometries.

Therefore, considering all the chemical geothermometers, it can be concluded that the cation geothermometries are not applicable to the three samples from Bhimband. Based on silica geothermometries, reservoir temperatures of 67 to 100°C may be expected in these springs. This is rather anomalous in view of relatively high surface discharge temperatures of about 64°C of these springs. The quartz geothermometers give reservoir temperatures of 118 to 147°C for Changlung and Panamik geothermal systems of Nubra valley. These seem to be realistic temperatures. For Changlung, Na−K geothermometer gives temperature of about 150°C. Such a temperature may be possible at deeper levels as Na−K equilibrium is likely to be attained at 150°C. Na−K temperature is anomalously high at Panamik and gives indication of the lack of Na and K equilibrium. However, temperature of at least about 120°C, determined using silica values, is most likely to exist at deeper levels. Chetu and Taksing geothermal areas may have reservoir temperature of at least around 150°C as suggested by Na−K Geothermometry (Table 2). K−Mg temperatures are very low, generally below 100°C even for Changlung and Panamik. K−Mg equilibrium is considered to be a

low temperature phenomenon which may correspond to the latest equilibrium in the system related to the formation of clay.

Stable Isotope Systematics

All the spring waters fall very close to GMWL and bear similar characteristics of meteoric waters (Fig.4). The best fit line (BFL) obtained during linear fit of scattered dots furnish the following equation with definite slope and intercept:

$$\delta^2H = (8.0 \pm 4.49) \times \delta^{18}O + (7.78 \pm 0.34).$$

Thermal waters are devoid of any evaporative enrichment. Oxygen isotope exchange happens between silicate minerals in the reservoir or wall rocks with geothermal waters at temperature above 220°C and this gets reflected in positive $\delta^{18}O$ shift from GMWL (Craig, 1961). The reservoir temperatures estimated in the present work seldom reach to even 150°C and therefore there is absolutely no possibility of positive $\delta^{18}O$ shift. Thus, the waters retain their meteoric features.

Conclusions

The major cation and anion geochemistry shows the dominance of Na and K ions in thermal waters of the Himalayan terrain and Ca and Mg in thermal waters along the MSRF zone. Cation exchange, silicate rock weathering and carbonate dissolution are some of the principal geochemical processes involved in regulating total ion budget in thermal waters of study area. Changlung fluids are boiling under subsurface conditions
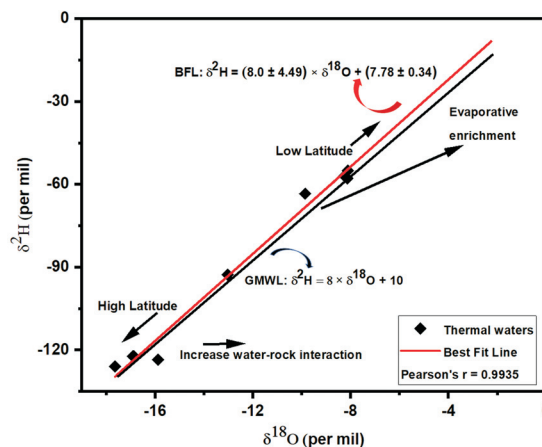

Fig.4. Stable Isotope characteristics of geothermal fluids in the three geothermal areas studied.

causing CO_2 degassing. This, in turn, causes precipitation of calcite resulting in the variations in Ca and HCO_3 contents in surface discharges compared to the deep single-phase geothermal fluid. SiO_2 content in Changlung thermal fluid is higher than that in the Panamik thermal fluid. This is probably because of sub-surface boiling, which causes loss of CO_2 resulting in enhancing the fluid pH with concomitant increase in SiO_2 solubility. Changlung fluids may be precipitating silica during their ascent to surface. Estimated reservoir temperature using silica thermometry is 77 to 147°C while Na/K thermometer may be applied to selected waters of Chetu, Taksing and Changlung. These geothermal areas give temperature estimates of 152 to 182°C. Thermal springs studied are meteoric in origin showing significant latitude/altitude effect without any evaporative enrichment. Because of relatively low temperatures of less than 150°C, there is no $\delta^{18}O$ shift. Munger hot springs have anomalous and rather enigmatic chemistry. Silica is the most abundant chemical species followed by bicarbonate. Low TDS of 120 to 150 mg/L and very low concentrations of major cations, particularly Na point to water-rock interaction in quartzite and a very high water to rock ratio.

Authors′ Contributions

AD: Investigation, Conceptualization, Methodology, Writing−Original Draft. **AA:** Conceptualization, Methodology, Writing−Review and Editing. **PM:** Software, Validation, Writing − Original Draft. **SB:** Visualization, Supervision, Editing. **PS:** Visualization, Supervision, Editing. **VVS:** Project Administration, Supervision, Editing. **AS:** Investigation, Validation, Formal Analysis. **APT:** Project Administration, Supervision, Editing. **BR:** Supervision, Visualization, Writing - Review and Editing.

Conflict of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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