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Hydrogeochemical and biological assessment of spring and stream water quality for its suitability for drinking in Kanglung locality, Trashigang, Bhutan

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ABSTRACT

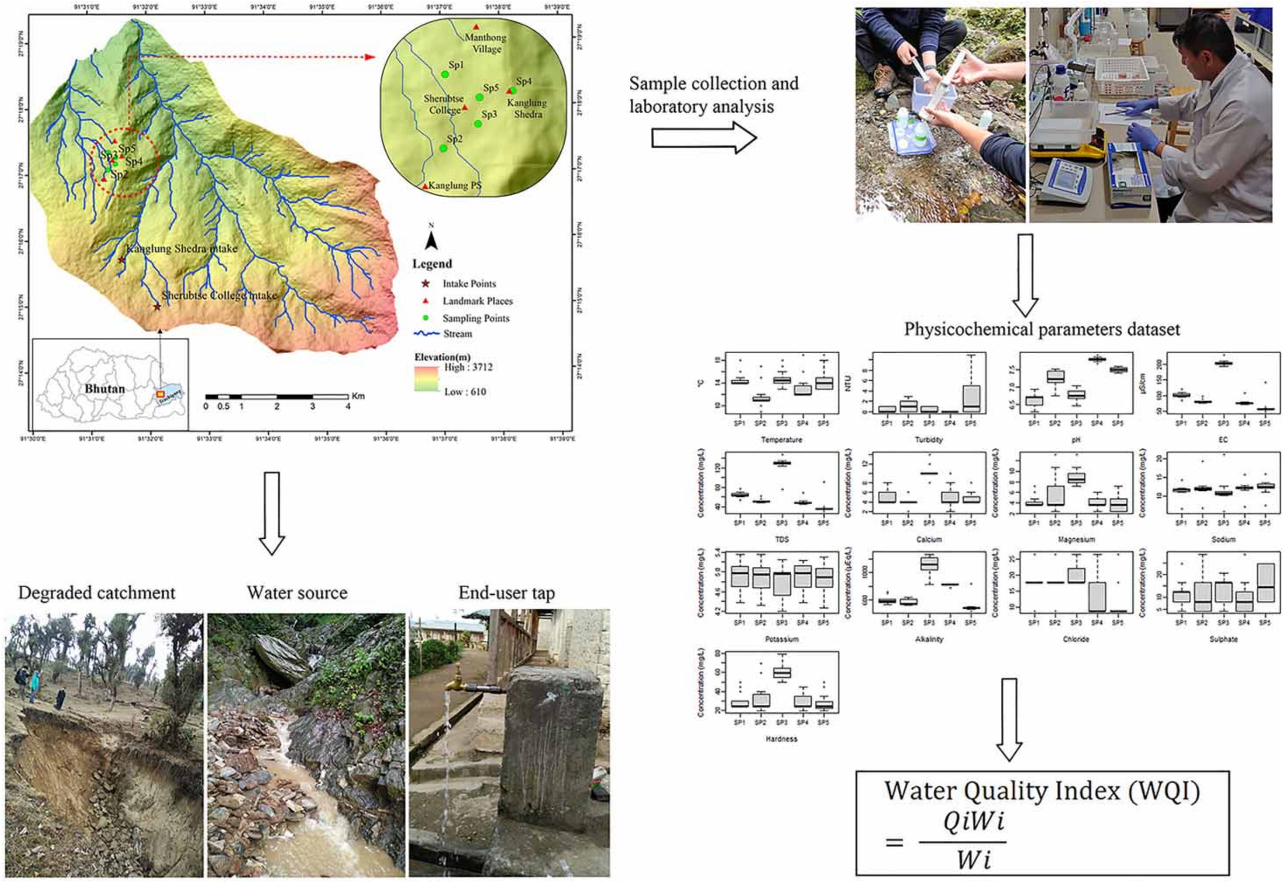
Springs are important water sources for domestic and agricultural uses in eastern Himalayas. This study describes the water quality scenario and major geochemical processes of the springs utilized for drinking in Kanglung locality, Bhutan. Water samples were collected at two end-user taps and three spring sources from October 2020 through April 2021 for laboratory analyses. Water samples analyzed met the WHO drinking water quality standard except for turbidity and thermotolerant coliform in some samples. The Water Quality Index (WQI) values of the spring water ranged from 5.75 to 41.64, majority falling under excellent class demonstrating potability of springs tapped. The order of anion and cation chemistry were $HCO_3^- > Cl^- > SO_4^{2-}$, and $Na^+ > Ca^{2+} > Mg^{2+} > K^+$, respectively. The scatter diagrams and molar ratios of major ions indicated silicate and carbonate weathering controlling the spring water chemistry. The piper diagram indicated that springs are predominantly Ca-Mg-Cl and Na-Cl types. Gibbs diagrams inferred that spring/stream chemistry is mainly controlled by rain-water followed by water-rock interaction. Statistical analyses indicated different sources of dissolved ions including carbonate and silicate weathering, atmospheric precipitation, dissolution of sulphate minerals, and oxidation of sulphides.

Key words: Bhutan, Himalaya, hydrogeochemistry, spring water quality, thermotolerant coliform

HIGHLIGHTS

- Precipitation and rock weathering are main processes in the acquisition of dissolved ions in springs in Kanglung.
- WQI values of springs ranged from 5.75 to 41.64 indicating 80% of the springs in Kanlung is suitable for drinking.
- Presence of thermotolerant coliform in some samples suggested that water must be boiled before drinking.

GRAPHICAL ABSTRACT



INTRODUCTION

In the Himalayas, spring water has traditionally been the primary source of water for drinking and other domestic purposes for centuries. Springwater is generally considered pristine; however, it can be easily contaminated when it emerges from the earth's surface. Globally, drinking water pollution has become a significant challenge attributable to rapid urbanization and industrialization. According to World Health Organization (WHO), about 435 million people take water from unprotected wells and springs, and around 144 million people depend on untreated surface water from lakes, ponds, rivers and streams (WHO 2019).

Although Bhutan has the highest per capita water use capacity globally (i.e., 94,500 m^3 per person per year), it is ironic that most rivers and streams are not accessible. The rivers and streams mostly flow at the bottom of the gorges and ravines, while settlements and farmland occupy the upper slopes and hilltops (ADB 2016). Consequently, springs or small mountain streams are the primary water sources for domestic and agricultural uses. However, climate change, population increase, and economic development are mounting pressures on these available water resources in terms of quality and quantity in the region (ADB 2016; Sharma *et al.* 2019). Recent studies (Tambe *et al.* 2012; Acharya 2021; Kulkarni *et al.* 2021) have demonstrated that springs and streams in the Himalayan areas, including Bhutan, are drying up or becoming seasonal, resulting in acute water shortages under changing climate conditions. The annual drinking water quality surveillance report of 2019 revealed that the availability of safe drinking water is still a challenge in Bhutan, which corroborates with past studies (Rahut *et al.* 2016; Wangdi & Clements 2017) carried out in some parts of Bhutan. From a total of 1951 drinking water samples collected nationwide for routine water quality surveillance, 47.4% tested for thermotolerant coliform, while some of the water samples had arsenic, lead, cadmium and iron concentrations exceeding the permissible limit. Yet, information on the water quality of the stream or spring water sources tapped for drinking is inadequate due to the limited coverage. Moreover, the routine analysis focuses mainly on the microbiological parameter. Physicochemical and biological characteristics of streams and springs are critical for the holistic understanding of water quality and determining its suitability for human consumption and other utility besides assessing its ecological health.

Physicochemical and biological characteristics of stream/spring waters mainly depend on natural factors such as underlying geological formations of the area, degree of chemical weathering of various rock types, atmospheric inputs and anthropogenic factors such as disposal of domestic and urban wastes, agricultural runoff, industrial and municipal effluents. All these factors make stream/spring waters vulnerable to microbial and mineral contaminations, including toxic trace elements, thus rendering them unfit for drinking and other purposes. Besides water quality information, spring/stream water chemistry are instrumental in inferring hydrological and hydrochemical processes in mountainous catchments. For instance, major ion chemistry is a powerful tool which is widely used for tracing source of solutes in groundwater aquifer system, the response of aquifers to hydrological events and describing the evolution of water as a result of water–rock interaction (Bhat *et al.* 2014). Despite its importance, this kind of data is virtually unavailable in Bhutan Himalayas.

Bhutan has a national drinking water quality standard for various physical, chemical and bacteriological parameters. While these standards are useful, they pose a particular problem in the case of reporting the water quality monitoring results to various stakeholders such as water service providers and the general public. This is due to an inherent complexity in comprehending the water quality results due to many individual variables. The water quality index (WQI), initially formulated by Horton (1965), later developed by Brown *et al.* (1970), and further improved by several researchers, provide an inclusive approach to interpreting water quality parameters of water sources for assessing potability (Aalipour *et al.* 2022). WQI aims to integrate complex water quality data and establish a simple criterion that describes overall water quality and is understandable to the general audience in deciphering water sources' status in a particular watershed (Seth *et al.* 2016). This approach has been widely employed in monitoring the water quality of surface waters, including spring and stream waters (Thakur *et al.* 2020; Tiwari *et al.* 2020; Ahsan *et al.* 2021) to help strategize appropriate management plans to improve water quality. Therefore, this study aimed to analyze the potability of major springs and streams in the Kanglung locality using drinking water quality indices and to infer major geochemical processes in the springs and streams. Our findings will provide insights on the quality of a spring and a small stream to support making an informed decision for the site-specific springshed management plans and interventions to improve drinking water quality in the area.

MATERIALS AND METHODS

Study area

Kanglung is under Trashigang District, Eastern Bhutan, at an elevation of about 1,930 m above sea level (a.s.l). The hydroclimate of the area is influenced mainly by the Indian summer monsoon typical of the eastern Himalayas. The monsoon season begins in June, peaks in July–August and ends in September, resulting in high streamflow. Annual rainfall measured at Kanglung meteorological station from 1996 to 2021 ranges from 885.6 mm to 1,487.3 mm. The maximum rainfall occurred in May and October, measuring 103.0 mm and 71.8 mm in 2020 and 2021, respectively. The average annual temperature maximum and minimum recorded at the station from 1996–2021 were 21.81 °C and 10.59 °C, respectively. Over 3000 people live in the Kanglung community comprising mainly of students, civil servants, corporate workers, entrepreneurs, and farmers (NSBO 2017). Recently, Kanglung has been identified as the regional educational hub because of its existing educational institutes, mainly Sherubtse College, Kunglung Thubten Choekhorling Shedra (a Buddhist monastery), and schools; and also due to its strategic location and favourable climatic conditions (Pokhrel 2015). Over the years, Kanglung has seen an increasing population partly owing to increased student intake at Sherubtse College and the establishment of new government, corporate and private facilities. The growing population has led to water shortages in the community mainly due to the increase in per capita water use. Moreover, the availability of clean drinking water is a recurrent issue in the community as the existing water supply is untreated (Tshering *et al.* 2021). Spring fed streams and springs are the primary source of drinking water and domestic application including irrigation for the Kanglung community. It is noteworthy to mention that the human encroachment at the water source areas has increased over the years due to the developmental activities like construction of roads and buildings. Moreover, some of the stream catchments in the study area have been witnessing frequent erosion due to intense surface runoff during storm events as an impact of land degradation at the upstream areas. Therefore, it is crucial to assess impact of various natural and manmade factors on the quality of these freshwater resources for the welfare of the community.

This study assessed one spring fed stream (Sp5) and four perennial spring waters (Sp1, Sp2, Sp3 and Sp4) used for drinking by the Sherubtse College (Sp2, Sp3 and Sp5), Kunglung Shedra (Sp4), and the nearby Manthong village (Sp1). The spring-water Sp1, Sp2 and Sp3 were sampled at the source while Sp4 and Sp5 were sampled at the end-user taps (Figure 1).

Field method

Eighty-nine grab water samples were collected weekly from October 2020 through April 2021. Both filtered and unfiltered water samples were collected for physico-chemical analyses. One-litre high-density polyethylene (HDPE) bottles were used to collect the unfiltered samples, while 125 mL HDPE bottles were used for collecting filtered samples. The sampling bottles were washed with deionized (DI) water and air-dried before sample collection. Water samples for major ions were filtered in the field using a pressure syringe filtering system through a 47-mm diameter Gelman A/E glass-fibre filter with a pore size of about 1.0 μm following the protocols discussed in Hill *et al.* (2018). The sampling bottles were rinsed at least five times with water samples during the sample collection. The HDPE bottle was filled with water sample and capped leaving no air space. The water samples collected were immediately transported to the Center for Science and Environmental Research (CSER) water chemistry laboratory, Sherubtse College, Royal University of Bhutan. The samples were stored at 4 °C until the analyses. Fifteen additional unfiltered samples for the thermotolerant coliforms analysis were collected in February 2021 (n=10) and December 2020 (n=5) in a sterile 125 mL HDPE bottle. The sample bottle was tightly capped, sealed with parafilm, and immediately sent to the Kanglung Basic Health Unit's Microbiological Laboratory, Trashigang, for analysis.

Laboratory method

The water samples collected were analyzed for physicochemical properties such as temperature, pH, electrical conductivity (EC), turbidity, calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), potassium (K^+), chloride (Cl^-), sulphate (SO_4^{2-}), acid neutralizing capacity (ANC), and total dissolved solute (TDS) following the standard methods developed for the analyses of water and wastewater by the American Public Health Association (Baird *et al.* 2017). The temperature was measured on-site using a glass laboratory thermometer (0–50 °C). The pH and EC were measured using the Fisher Accumet AB150 pH meter and the Mettler Toledo-SevenCompact Conductivity meter on the unfiltered samples. The Ca^{2+} and Mg^{2+} were estimated by the EDTA titrimetric method. The total hardness (TH) was computed from the determined concentrations of Mg^{2+} and Ca^{2+} and calculated as mg/L $CaCO_3$ using the equation described in Beyene *et al.* (2019): $TH=2.497\ (Ca^{2+}) + 4.118\ (Mg^{2+})$. The Na^+ and K^+ were determined with the flame photometer 128 (Systronics, India). The Cl^- was determined by the titrimetric (argentometry) method, while the gravimetric method was employed to determine the SO_4^{2-} concentration. The ANC (considered to equal bicarbonate alkalinity, HCO_3^-) was measured by the gran titration technique on a filtered water sample using Fisher Accumet AB150 pH meter and Gilmont GS-1200-A Micrometer Burette-Style Dispenser; 1/Ea, as per the standard operating procedure developed by Arikaree Environmental Laboratory, Institute of Arctic and Alpine Research University of Colorado, USA. A digital turbidity meter (Dufon, India) was used to measure the turbidity, and the gravimetric method was used to determine the TDS of the water samples. The analytical precision of the dissolved ions dataset was checked by charge balance error (CBE) given by the equation:

$$CBE\% = \frac{mEq\ (cations) - mEq\ (anions)}{mEq\ (cations) + mEq\ (anions)} \times 100 \qquad (1)$$

The CBE values of most of the samples were within the accepted range of $\pm 10\%$.

The analysis of thermotolerant coliforms was carried out following membrane filtration and colony count techniques to assess faecal contamination as per the protocol of RCDC (2019). The results were expressed as colony-forming units (CFU) per 100 mL of sample.

Data analysis

The major ions compositions were plotted on a piper trilinear plot using AquaChem 4.0 software to identify the main water types. The Gibbs diagram was plotted to assess the dominant hydrogeochemical processes on the chemical characteristics of spring water, viz., precipitation influence, water-rock interaction and evaporation. Correlation matrix and principal component analysis (PCA) were performed using the SPSS software package to understand the solute acquisition process in the system. The physicochemical parameters of the analytical results of spring water were also compared with the standard

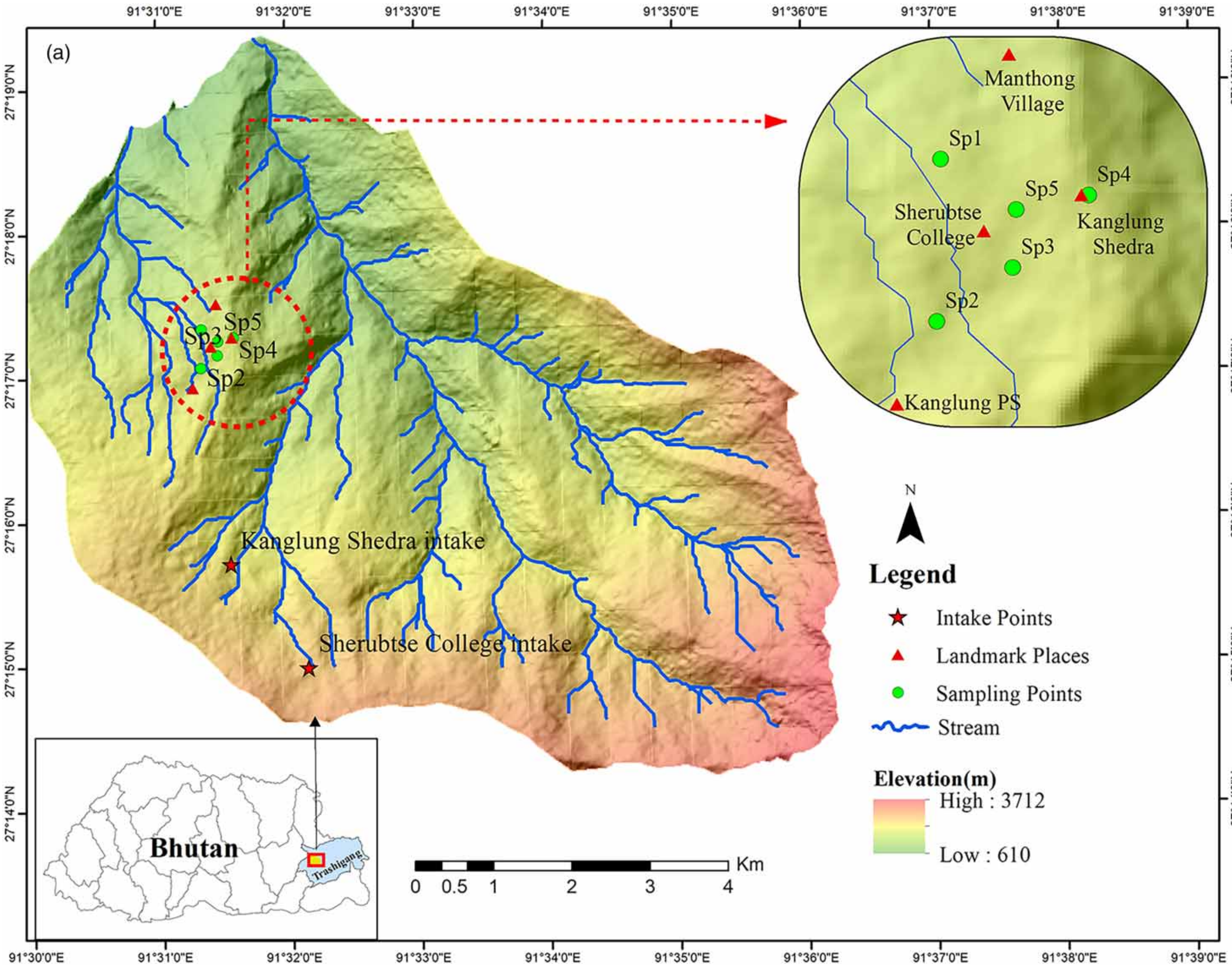


Figure 1 | (a) The study site in Kanglung community including landmark places, sampling locations, and intake points of drinking water sources for Kanglung Shedra and Sherubtse College (shown with red star). Top right inset: sampling points and location of Sherubtse College, Kanglung Shedra and Manthong village. Bottom left inset: regional locator. (b) The Land Use Land Cover (LULC) map of the study area. Note: 'Chhuzing' is an irrigated land while 'Kamzhing' is non-irrigated agricultural land in Bhutanese, respectively (*continued*).

guideline values for drinking water recommended by the World Health Organization (WHO) and the National Environmental Commission Secretariat (NECS), Bhutan.

The weighted arithmetic index method (Brown *et al.* 1972) was employed to determine the WQI of the spring water sample in the present study. Parameters used to calculate WQI were pH, EC, TDS, Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, Cl^{-}, SO_4^{2-}, TH and turbidity, and the limits were based on the WHO guideline values for drinking water quality (WHO 2011, 2017), and NECS. WQI is computed by the following equation (Thapa *et al.* 2020):

$$\mathrm{WQI} = \frac{\sum QiWi}{\sum Wi} \tag{2}$$

where, Wi = unit weight, Qi = quality rating scale. The quality rating scale for each water quality parameter was calculated as follows:

$$\mathrm{Qi} = \frac{(Vi - Vo)}{(Si - Vo)} \times 100 \tag{3}$$

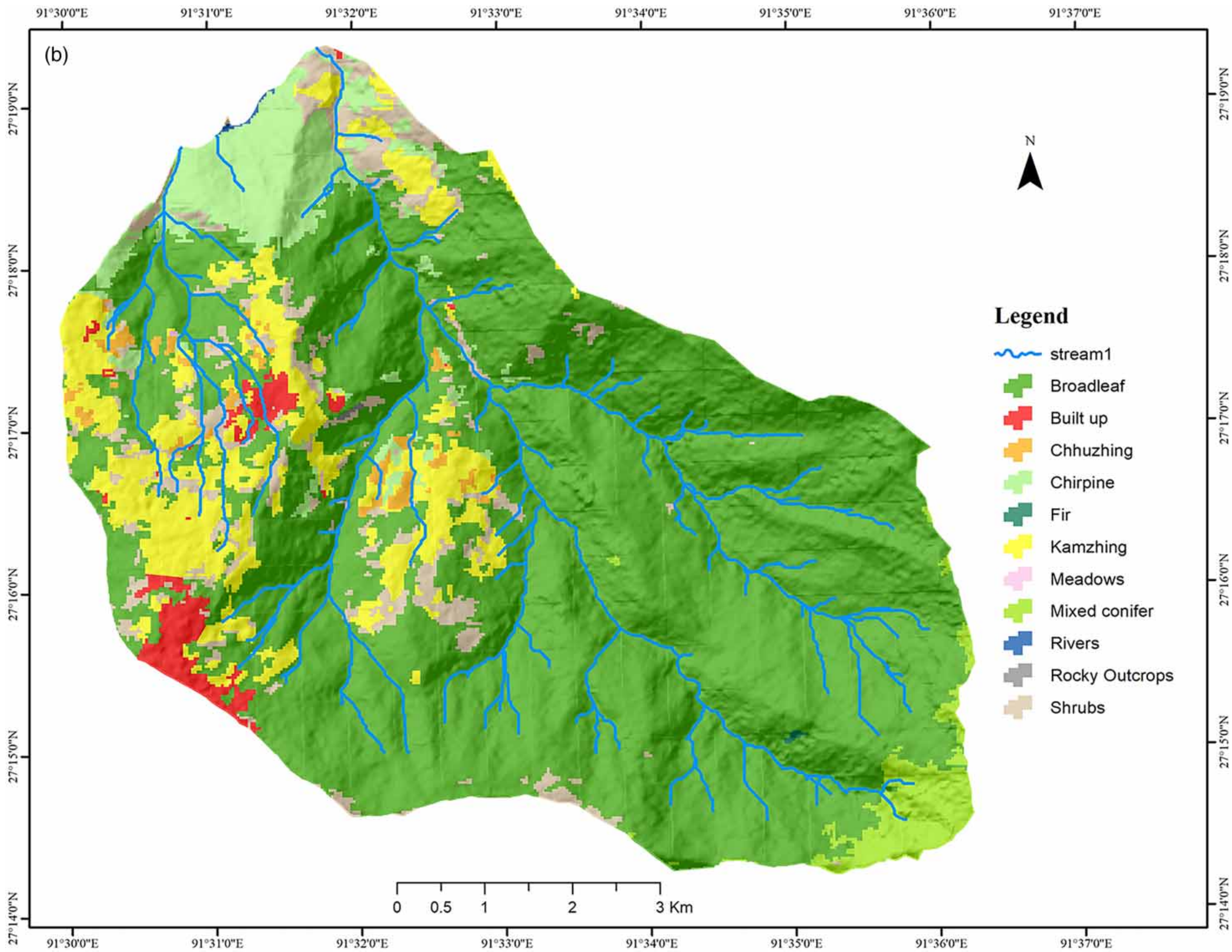


Figure 1 | Continued.

where, Vi = measured concentration value of water sample for each water quality parameter, Vo = ideal value of water quality parameters in pure state (Vo is 0 for all water quality parameters except pH, i.e., 7), Si = Standard value of water quality parameters (WHO). Unit weight Wi of each parameter was calculated by following the formula:

$$Wi = \frac{K}{Si} \tag{4}$$

where, K=proportionality constant calculated by the following formula:

$$K = \frac{1}{\sum \frac{1}{Si}} \tag{5}$$

The WQI obtained for each spring water was categorized as 0–25 excellent water quality, 26–50 good water quality, 51–75 poor water quality, 7–100 very poor water quality, and >100 unfit for drinking as per Ahsan *et al.* (2021).

RESULTS & DISCUSSION

Spring and stream water chemistry

The physicochemical characteristics of a stream and spring water samples including WHO drinking water standard are shown in Table 1. The temperature of the spring and stream water samples varied from 9 to 19 °C. The spring waters Sp1, Sp2 and Sp3 sampled at the source showed relatively stable water temperature than the Sp4 and Sp5 streams which were sampled at end-users tap across the study period (Figure 2). The relative constancy of water temperature across the springs Sp1–Sp3 demonstrates the thermal stability ascribed to minimum exposure of spring water to solar radiation and thermally buffered water emanating from the underlying rock (Lone *et al.* 2021). Turbidity measured across all the spring and stream water samples were generally below 3 NTU except for Sp5 (three samples exceeded 5 NTU) as shown in Figure 3, the majority falling under the permissible limit prescribed by WHO for drinking water. Low turbidity indicates the presence of less inorganic particulate matter and natural colloids such as slit and clay. Higher turbidity in Sp5 could be attributable to entry of soil or organic matter into the stream at the source area from the surface runoff during the storm events. The pH of the spring and stream water samples measured were slightly acidic to alkaline. The pH values in Sp1, Sp2 and Sp3 not only showed comparatively wide variation but also slighty more acidicity than Sp4 and Sp5 (Figure 2). The slightly acidic Sp1–Sp3 springs than streams (Sp4 and Sp5) indicates a closed system wherein growth of macrophytes occurs which on its decomposition produces organic acids (Bhat & Pandit 2020). However, in the present study, the pH of water samples lie within the permissible limit for drinking water. An average TDS and EC values of springs and streams were 67.0 ± 33.3 mg/L, and 105.7 ± 52.9 μS/cm, respectively. The lower EC values were measured in Sp4 and Sp5 streams while highest occurred in Sp3 spring indicating the different flow paths as well as an infleunce of anthropogenic activities. It is likely that Sp3 which is located at the lower elevation than Sp4 and Sp5 has a longer flow path and thereby extent of rock-water interaction increases accuring more dissolved ions. Moreover, mixing of wastewater from built-up areas (Figure 1(b)) at the vicinity of Sp3 source have likely resulted in higher EC or TDS values (Khadse *et al.* 2016). Similar altitudinal variation in the EC values of springs were reported in other Himalayan regions (Ansari *et al.* 2015; Bhat & Pandit 2020). The EC and TDS values in water samples were much lower than the maximum desirable limits for drinking water recommended by the WHO.

The HCO_3^- was the dominant anion contributing 54.3% of the total anions (TZ^-), followed by Cl^- and SO_4^{2-} accounting 23.4% and 22.2%, respectively. In cation chemistry, Na^+ was dominant accounting 42.9% of the total cations (TZ^+), followed by Ca^{2+} (20.6%), Mg^{2+} (18.9%), and K^+ (17.6%). The concentrations of weathering products were consistently higher in Sp3 except for Na^+ and K^+, corroborating with our assumption that this spring travels longer flow paths which is consistent with EC and TDS results (Figure 2). On the contrary, concentrations of dissolved ions, EC and TDS values were lower in Sp5 indicating minimum rock-water interaction likely owing to shorter flow path, and higher discharge resulting in dilution of dissolved ions. The possible sources for HCO_3^-, Ca^{2+} and Mg^{2+} are carbonate rocks dissolution while silicates of magmatic rocks are likley sources for Na^+ and K^+. Weathering of the Na^+ bearing minerals like plagioclase feldspar also release the soluble Na^+ products in water. The concentrations of all the major ions are much lower than the permissible limits prescribed for drinking water.

The TH in the study area ranged from 19.8 to 79.3 mg/L which falls in soft (0–60 mg/L) to medium (60–120 mg/L) category. The concentration of TH of spring/stream water samples are also within the desirable limit recommended for drinking water. No adverse effect due to water hardness on health was documented but extreme hard water is known to cause cardiac and kidney diseases (Narsimha & Sudarshan 2017).

Sources and processes controlling spring and stream water chemistry

To understand sources of dissolved ions in springs/streams in the study area, the scatter plots of equivalent concentration (mEq/L) of different parameters were used. The scatter diagrams of alkaline earth metals ($Ca^{2+} + Mg^{2+}$) and alkali metals ($Na^+ + K^+$) versus total cations (TZ^+) (Figure 3(b) and 3(c)) with an average molar ratios of 0.49 ± 0.11 and 0.51 ± 0.11, respectively, shows that most of the data points fall below 1:1 equiline revealing the higher contribution of total cations. The scatter plots of ($Ca^{2+} + Mg^{2+}$) and ($Na^+ + K^+$) with average molar ratios of 1.15 ± 0.57 had data points plotting both above (43%) and below (57%) the 1:1 equiline (Figure 3(a)). These scatter diagrams (Figure 3(a)–3(c)) and molar ratios all indicated the importance of both silicate and carbonate weathering in controlling the spring/stream water chemistry (Sharma *et al.* 2012). The molar ratios of Na^+/Cl^- of water samples range from 0.38 to 2.35. Out of 87 water samples, 56 had Na^+/Cl^- molar ratios greater than one and rest with value less than one or close to one. Samples having a Na^+/Cl^-

Table 1 | Overall physicochemical characteristics of stream and spring water

Parameter	Average (n=86)	Max	Min	STDV	WHO Standard 2017
Temperature (°C)	13.9	19.0	9.0	2.0	_
Turbidity (NTU)	1.0	11.0	0.0	2.0	5.0
pH	7.2	7.9	6.3	0.5	6.5–8.5
EC (μS/cm)	105.7	231.0	48.0	52.9	1,500
TDS (mg/L)	67.0	147.8	30.7	33.3	1,000
Ca^{2+} (mg/L)	5.7	14.0	2.0	2.7	75[a]
Mg^{2+} (mg/L)	5.2	13.2	2.4	2.5	50[a]
Na^{+} (mg/L)	11.9	21.1	5.9	2.0	200[a]
K^{+} (mg/L)	4.9	5.4	4.2	0.3	12[a]
Cl^{-} (mg/L)	15.7	26.6	8.9	6.0	250
HCO_3^- (mg/L)	36.4	63.8	19.9	12.0	500[a]
SO_4^{2-} (mg/L)	14.9	53.9	4.1	10.3	250
Total Hardness (mg/L)	35.8	79.3	19.8	15.8	500
Thermotolerant coliform (CFU/100 mL)	1.8	5.0	0.0	1.9	0

[a]WHO Standard 2011.

ratio greater than 1 indicate excess Na^+, which might be attributable to silicate weathering. However, with samples having Na^+/Cl^- ratio less than 1, it shows excess Cl^- due to carbonate weathering (Srivastava & Parimal 2020). In the Ca^{2+}+ Mg^{2+} versus HCO_3^- plot (Figure 3(d)), the samples spread mostly above and close to the 1:1 equiline with some samples poltting below the equiline. The points above and close shows that the HCO_3^- in the spring/stream water is controlled by the dissolution of alkaline earth metals as well as the calcite and dolomite minerals (Vishwakarma *et al.* 2018). The scatter plot of Ca^{2+}+Mg^{2+} versus HCO_3^-+SO_4^{2-} (Figure 3(e)) has sample points mostly plotted below or close to the 1:1 equiline line inferring the predominance of silicate weathering, and carbonate weathering as an additional process as the source of Mg^{2+} and Ca^{2+} in springs and streams (Kumar *et al.* 2008; Li *et al.* 2020). The excess of HCO_3^-+ SO_4^{2-} over Ca^{2+}+ Mg^{2+} concentrations can be attributable to the dissolution of silicate minerals (e.g., sodic plagioclases like albite) depicting silicate weathering and ion exchange as the dominant process in the study area (Vishwakarma *et al.* 2018; Egbueri 2019). Additionally, the scatter plot of Na^++ K^+ versus SO_4^{2-}+ Cl^- (Figure 3(f)) also had the locations of samples above, below and close to the 1:1 equiline. The location of samples below the 1:1 equiline indicate an impact of non-silicate minerals, whereas the locations above inferred contribution from the silicate minerals, confirming the influence of different sources of these dissolved ions in the springs and streams (Kumar *et al.* 2020).

The major cations and anions composition of the spring and stream water samples were plotted on a piper triangular diagram (Figure 4) to identify main chemical facies of the springs/streams (Piper 1944). The central diamond plot in Figure 4 indicated intermediate (mixed) chemical character of springs/streams are in the order NaCl>Ca-Mg-Cl>Ca-Mg-HCO_3->Na-HCO_3-Cl types. Most of the data points are scattered within zone 4, indicating that a strong acid exceeds weak acid, while some samples were also distributed in zone 3 suggesting that weak acid exceeds strong acid. In the lower left triangle, data points are clustered in zones B and D indicating that some are of no dominant type, some are sodium-potassium type in terms of cations. Similarly, in lower right triangle, most of the data points are distributed in zones B and E showing that some are of no dominant type, some are bicarbonate type in terms of anions. The occurrence of dominant Na-Cl and Ca-Mg-Cl types, and intermediate (mixed) Ca-Mg-HCO_3 and Na-HCO_3-Cl chemical facies indicated the influence of anthropogenic activity, atmospheric precipitation, weathering of carbonate and dissolution of silicate minerals, and higher mixing processes of springs and streams in the study area (Sajil Kumar & James 2016; Zhang *et al.* 2018). The dominance of NaCl type in the study area indicated rock water interaction with Na^+ leached from feldspar-rich reserves and Cl^- derived from anthropogenic activity (Senthilkumar *et al.* 2021).

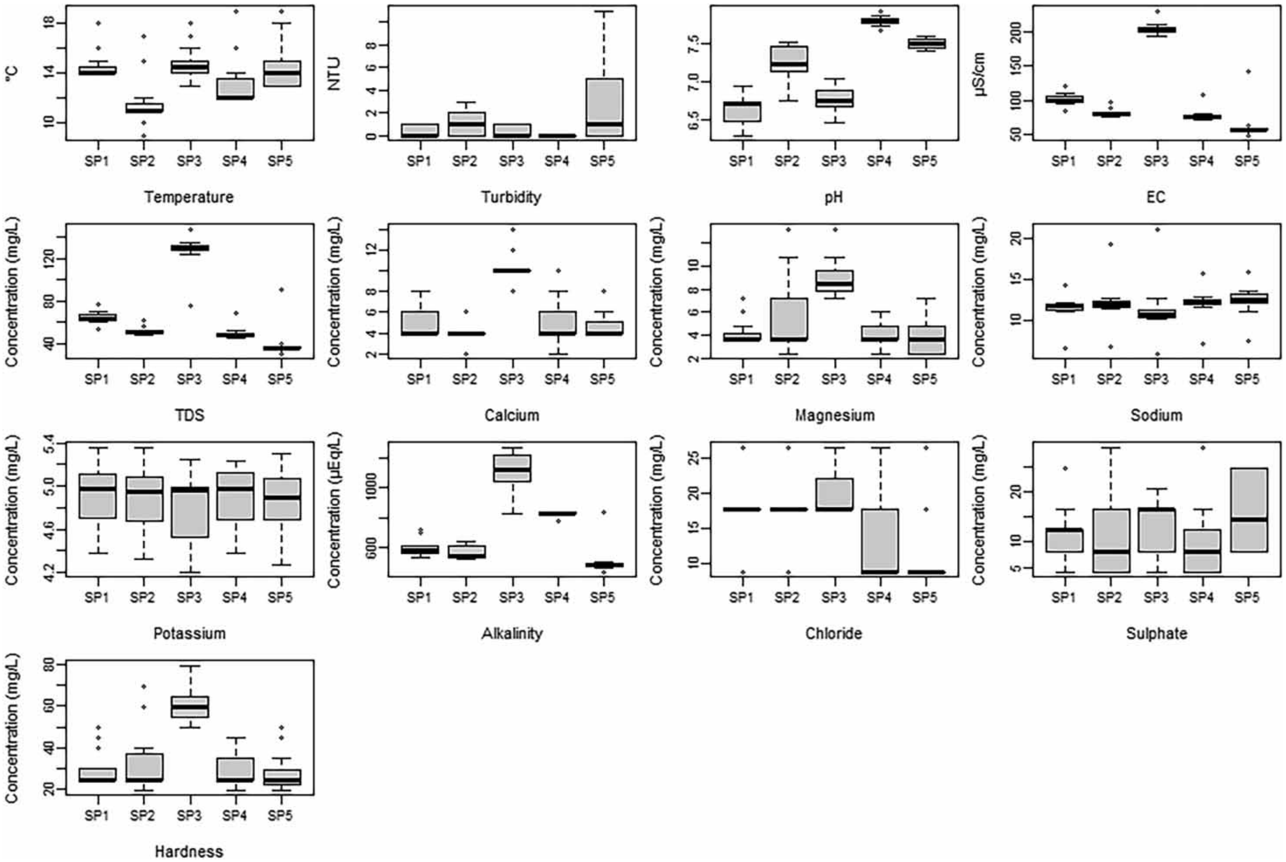


Figure 2 | Boxplot of the physico-chemical characteristics of a stream and spring waters.

In Gibbs diagrams, the variation of weight ratios of TDS versus $Cl^-/(Cl^-+HCO_3^-)$ and TDS versus $(Na^++K^+)/(Na^++K^++Ca^{2+})$ for anions and cations are plotted, respectively (Figure 5) (Gibbs 1970). These plots place water samples in the region between rock dominance zone and precipitation dominance zone. However, the majority of samples fall in the precipitation dominance suggesting that atmospheric precipitation is the primary dominant process in the acquisition of dissolved ions while weathering of the rock forming minerals is the secondary factor controlling the the hydrogeochemistry in the study area (Akoachere *et al.* 2018). The result also indicates interaction of precipitation water with local lithologies controlling the chemistry of spring and stream waters in the study area (Thakur *et al.* 2020). The precipitation dominance was reported in the groundwater at higher altitudes of the Himalayan region where ionic concentrations are low and more outflow and recharge from the water coming from the precipitation and melting of ice is anticipated (Kumar *et al.* 2020).

Correlation analysis

Correlation matrix between various physicochemical parameters of the springwater is shown in Table 2. The interrelationships among dissolved ions can indicate the solute acquisition processes (Singh & Ramanathan 2017) regulating springwater chemistry. EC and TDS have a strong positive correlation with Ca^{2+}, Mg^{2+}, HCO_3^- and TH indicating that springwater chemistry was mostly regulated by them, and presence of Ca^{2+}, Mg^{2+}, HCO_3^- and TH greatly infuences TDS, alkalinity and EC of the springwater (Mahato *et al.* 2018). A good correlation exists between Ca^{2+} and Mg^{2+}, Ca^{2+} and HCO_3^-, Mg^{2+} and HCO_3^- indicating origin of these ions are from carbonate rock weathering (Singh *et al.* 2015) resulting in Ca-Mg-HCO_3 water type in the study area. Low positive correlation of Cl^- on EC, TDS, Ca^{2+}, Mg^{2+}, and TH was also seen. The weak and negative correlations of Na^+ and K^+ with other major ions suggest that sodium and potassium mostly originated from Na/K-feldspars or Na/K-bearing minerals.

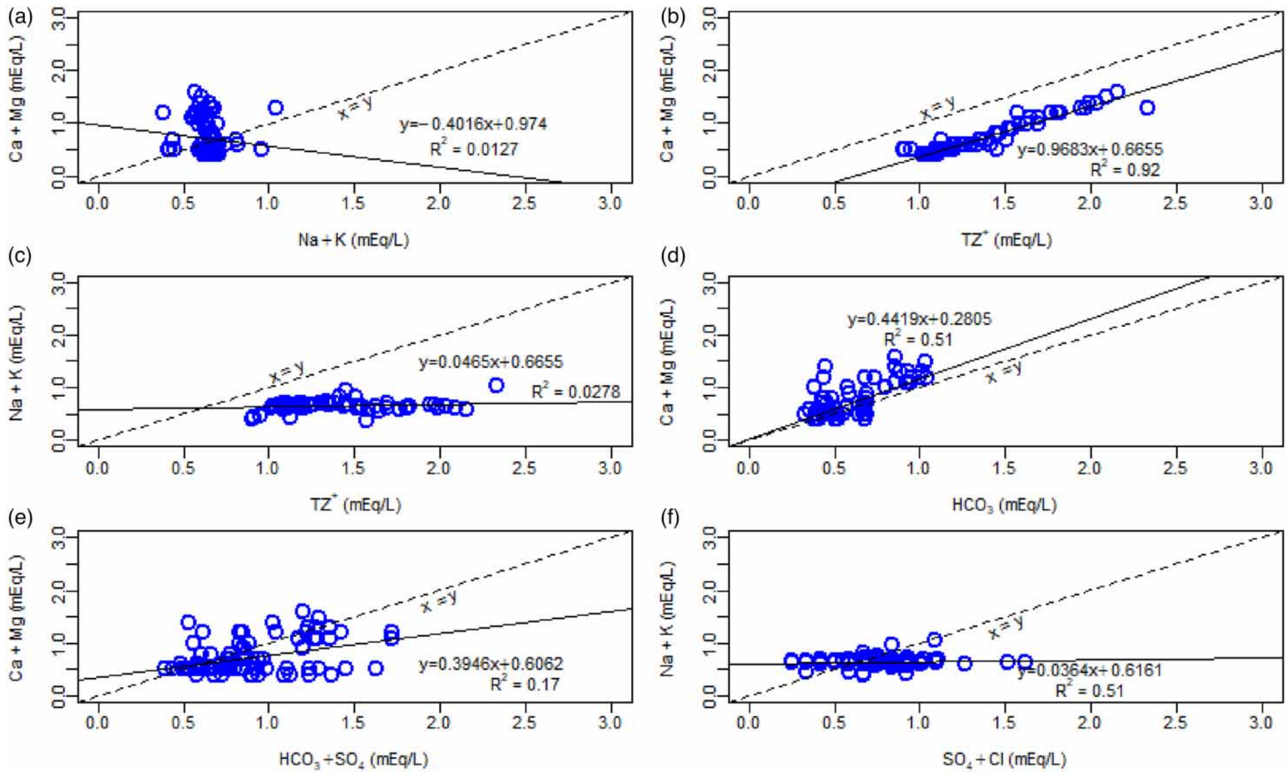


Figure 3 | (a) Scatter diagram between (Ca^{2+}+ Mg^{2+}) and (Na^{+}+ K^{+}); (b) (Ca^{2+}+ Mg^{2+}) and total cations (TZ^{+}); (c) Scatter diagram between (Na^{+}+ K^{+}) and total cations (TZ^{+}); (d) (Ca^{2+}+ Mg^{2+}) and (HCO_3^{-}); (e) Scatter diagram between (Ca^{2+}+ Mg^{2+}) and (HCO_3^{-}+SO_4^{2-}); (f) Scatter diagram between (Na^{+}+K^{+}) and (SO_4^{2-}+Cl^{-}).

Principal component analysis

R-mode factor analysis was used to identify major factors controlling the hydrochemistry of spring and stream waters in this study (Singh *et al.* 2012). Factor analysis of water samples generated five significant factors having eigenvalues greater then one (Table 3). These five factors explain about 80% of the total variance. Factor 1 accounts for 44.2% variance in the dataset and showed high loading of EC, TDS, Ca^{2+}, Mg^{2+}, HCO_3^{-} and TH, and negative loading of Na^{+} and K^{+} indicating the contribution from carbonate weathering. Factor 2 accounts for 11.1% variance in the dataset and had strong loading of Cl^{-} showing an input of atmospheric precipitation. Factors 3, 4 and 5 accounts for 8.6%, 7.9% and 7.8% variance in the dataset, respectively. Factors 3, 4 and 5 showed a high loading of SO_4^{2-}, K^{+} and Na^{+}, respectively, suggesting that these factors are associated with dissolution of sulphate minerals, oxidation of sulphides and contribution from weathering of silicate minerals.

Water quality index (WQI)

The relative weights of physicochemical parameters for expressing drinking WQI are provided in Table 4. The WQI values of the spring and stream waters computed for Sp1, Sp2, Sp3, Sp4 and Sp5 were 5.75, 23.05, 7.84, 22.64 and 41.64, respectively, falling in the excellent class except for Sp5 which falls in the good class. The result indicated that Sp1, Sp2, Sp3, and Sp4 can be used for drinking, and other allied purposes; however, Sp5 might require some pretreatment prior to drinking. The source of contamination in Sp5 is suspected from surface runoff at upstream water sources that washdown particulate and dissolved organic matter during the storm events. Similarly, poor water quality of Sp5 could be attributable to aging water suppy facilities.

Thermotolerant coliform

In this study, thermotolerant coliform ranged from 0 to 5 CFU/100 mL in spring and stream water samples collected in February and December coinciding with lean or dry winter season in the study area (Table 5). The result indicated that the spring waters Sp2, Sp3, and stream Sp4 were free of thermotolerant coliform contamination, while spring Sp1 and stream Sp5 had thermotolerant coliform exceeding the WHO drinking water standard (0 CFU/100 mL). The Sp5 had consistently higher

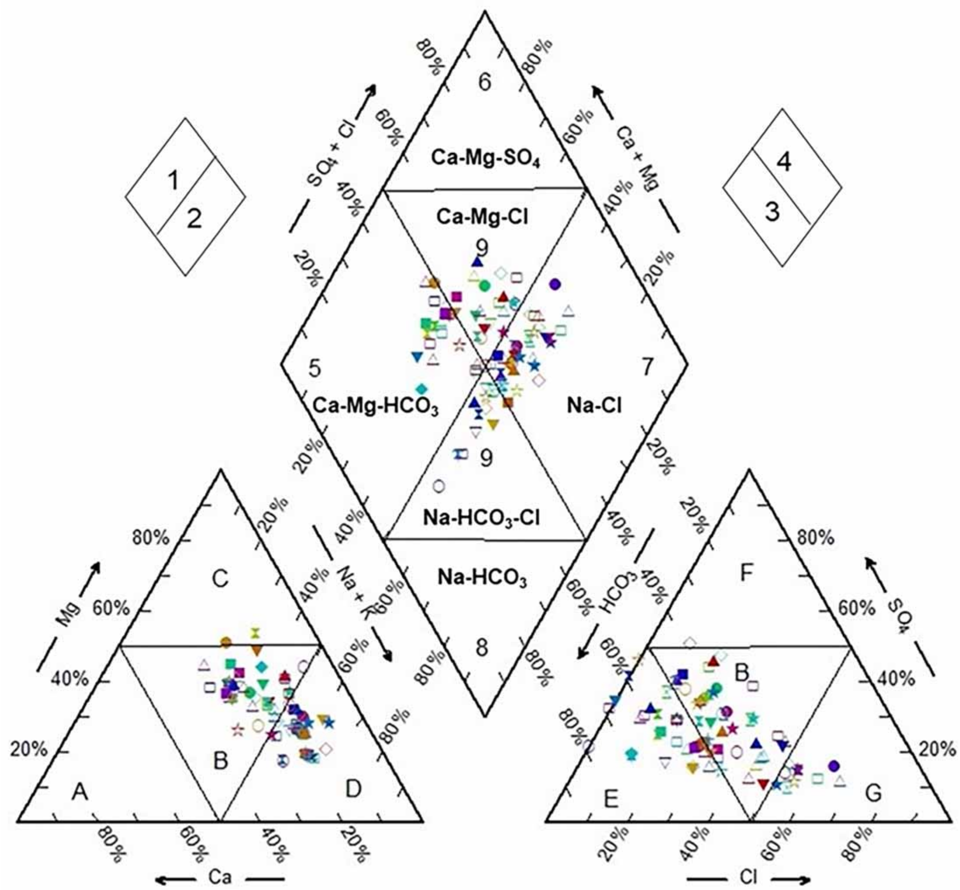


Figure 4 | Piper triangular diagram showing hydrogeochemical character and hydrochemical facies in the spring/stream water.

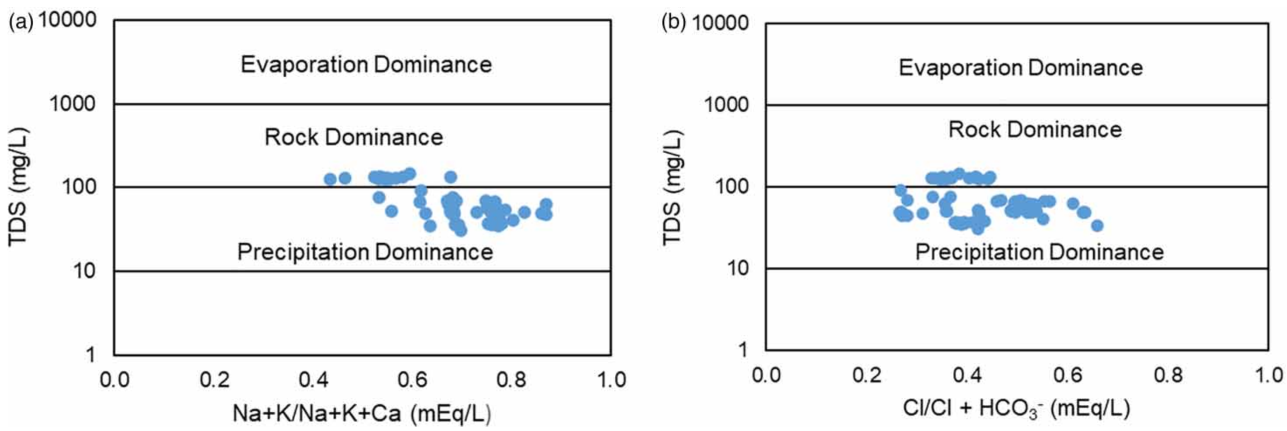


Figure 5 | Gibbs diagram I (a) & II (b) showing mechanism controlling spring/stream water chemistry.

thermotolerant coliform contamination. The occurrence of coliform in Sp5 could be attributable to entry of soil or organic matter (containing livestock and wild animal droppings) into the stream at the source area. The rangeland at the upstream catchment of the Sp5 stream is grazing area for the domestic animals which could be the source of organic matter. Similarly, the thermotolerant coliform contamination in the Sp1 could be due to faecal contamination from the leaking septic system, and domestic wastewater as its water source is located near the residential area (Figure 1(b)). The presence of thermotolerant

Table 2 | Correlation matrix of various physico-chemical parameters of spring/stream water samples

	Temp.	Turb.	pH	EC	TDS	Ca^{2+}	Mg^{2+}	Na^{+}	K^{+}	HCO_3^-	Cl^-	SO_4^{2-}	TH
Temp.	1												
Turb.	−0.05	1											
pH	−0.21	0.137	1										
EC	0.31*	−0.24	−0.61	1									
TDS	0.30*	−0.25	−0.60	0.99*	1								
Ca^{2+}	0.34*	−0.10	−0.41	0.83*	0.82*	1							
Mg^{2+}	0.20	−0.07	−0.44	0.69*	0.65*	0.72*	1						
Na^{+}	0.04	0.02	0.20	−0.20	−0.20	−0.11	−0.073	1					
K^{+}	0.05	−0.09	0.20	−0.11	−0.08	−0.20	−0.262	0.04	1				
HCO_3^-	0.12	−0.22	−0.18	0.82*	0.81*	0.78*	0.58*	−0.12	−0.12	1			
Cl^-	−0.02	−0.07	−0.51	0.45*	0.45*	0.45*	0.38*	0.05	−0.06	0.33	1		
SO_4^{2-}	0.16	−0.09	−0.02	0.054	0.051	−0.04	−0.122	0.06	0.195	0.08	−0.189	1	
TH	0.28*	−0.09	−0.46	0.80*	0.77*	0.89*	0.96*	−0.09	−0.25	0.71*	0.44*	−0.10	1

*Correlation is significant at the 0.01 level.

Table 3 | Principle and varimox rotated R-mode factor loading matrix of spring/stream water chemistry

Variables	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Communalities
Temp.	0.20	0.25	0.76	−0.18	0.12	0.72
Turb.	−0.18	−0.12	0.10	−0.73	0.09	0.60
pH	−0.31	−0.79	−0.19	0.01	0.30	0.86
EC	0.88	0.31	0.13	0.16	−0.18	0.94
TDS	0.86	0.32	0.13	0.19	−0.17	0.92
Ca^{2+}	0.90	0.21	0.13	−0.08	0.02	0.89
Mg^{2+}	0.82	0.25	−0.05	−0.23	0.02	0.78
Na^{+}	−0.07	−0.01	0.04	0.04	0.93	0.87
K^{+}	−0.22	−0.07	0.24	0.60	0.18	0.50
HCO_3^-	0.91	−0.09	0.00	0.19	−0.03	0.88
Cl^-	0.31	0.77	−0.15	0.09	0.22	0.78
SO_4^{2-}	0.01	−0.24	0.71	0.34	−0.08	0.69
TH	0.91	0.25	0.02	−0.19	0.02	0.93
Eigen value	5.74	1.44	1.12	1.03	1.02	
% of Variance	44.17	11.07	8.58	7.93	7.81	
Cumulative %	44.17	55.24	63.81	71.75	79.55	

coliform in water sources in the dry season suggested that the contamination will be more in wet seasons due to surface runoff during the storm events.

CONCLUSIONS

The springwater and streams are the primary water sources for various domestic applications in the Himalayas. However, these water resources are drying up and face increasing threats due to growing population and developmental activities, including climate change leading to reduced flow and poor drinking water quality. The present study assesses major springs and streams in the Kanglung locality to determine the water quality scenario and major geochemical processes controlling springwater chemistry. The order of anion and cation concentrations in water samples were $HCO_3^- > Cl^- > SO_4^{2-}$ and

Table 4 | Highest permitted values of WHO drinking water standards and relative weights for WQI analysis of springwater

Parameters	Highest permitted values in water (Si)	Relative weights (Wi)
PH	8.5	0.260
Turbidity	5	0.442
EC (μS/cm)	1,500	0.001
TDS	1,000	0.002
Ca^{2+} (mg/L)	75	0.029
Mg^{2+} (mg/L)	50	0.044
Na^{+} (mg/L)	200	0.011
K^{+} (mg/L)	12	0.184
Cl^{-} (mg/L)	250	0.009
SO_4^{2-} (mg/L)	250	0.009
HCO_3^{-} (mg/L)	500	0.004
TH (mg/L)	500	0.004
∑Wi		1

Table 5 | Thermotolerant coliform (CFU/100 mL) in spring water samples collected in the post-monsoon season

Sample	14-Dec-20	17-Feb-21	20-Feb-21
Sp-1	1	1	0
Sp-2	0	1	0
Sp-3	0	0	0
Sp-4	0	0	0
Sp-5	2	5	0

$Na^{+}>Ca^{2+}>Mg^{2+}>K^{+}$, respectively. The scatter diagrams and molar ratios of major ions indicated the importance of silicate and carbonate weathering in controlling the springwater chemistry and indicated the influence of different sources of dissolved ions. The Piper triangular diagram demonstrated hydrochemical facies of spring water were predominantly Ca-Mg-Cl and Na-Cl types. Gibbs diagrams suggested that atmospheric precipitation and weathering of the rock-forming minerals are the dominant hydrogeochemical processes controlling dissolved ions in the study area. The water quality parameters measured in most water samples were within the permissible limits recommended for drinking water; however, turbidity and thermotolerant coliform exceeded the permissible limits in a few samples (Sp1 and Sp5). The WQI values of all the springs and streams samples fall under excellent class except for stream Sp5. The WQI results demonstrated that springs Sp1, Sp2, Sp3, and stream Sp4 are potable and suitable for various domestic purposes; however, Sp5 might require some pretreatment before drinking. As stream Sp5 is the main drinking water source for Sherubtse College, providing water to a major population centre in the Kanglung community, appropriate mitigation measures are necessary to improve the stream water quality in the catchment area. Our results also suggest better strategies and institutional mechanisms for strengthening the management of these water sources to reduce pollution. Overall, our study demonstrates that the WQI is an efficient tool for providing usable and understandable water quality information for the general public, policymakers and water service providers. Additionally, the study provides baseline water quality information on major streams and springs in the Kanglung area.

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AUTHOR CONTRIBUTIONS

TL & PL: sampling and laboratory analyses, and data analysis. TD: conceptualization, writing original draft manuscript, data analysis, review and editing, supervision, and project administration. ST: preparing the study area maps.

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COMPLIANCE WITH ETHICS REQUIREMENTS

This study did not contain any studies with human participants or animal experiments performed by any of the authors.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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