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Vulnerability of karst aquifer to contamination: a case study of Liddar catchment, Kashmir Himalayas

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ABSTRACT

Water samples (n=100) were collected from karst springs to understand the hydrogeochemical processes controlling the chemical composition and quality of groundwater in karstified mountainous catchment of western Himalaya. The results suggest that congruent carbonate dissolution (calcite dissolution, dedolomitization), incongruent silicate weathering and ion exchange processes are the dominant processes controlling the spring water chemistry. Geochemical classification shows that springs water belongs to $Ca\text{-}HCO_3$ and $Mg\text{-}HCO_3$ type. Although, the concentrations of various ions in the spring waters are mostly within the prescribed specifications of World Health Organization (WHO, 2006) and Indian standards (BIS, 2012). Some samples (<1%) showed high levels of Fe (~5.8 mg L^{-1}) and F^-(~ 1.7 mg L^{-1}). The sharp increase of No_3^- (2 to 12 mg L^{-1}) and Cl^- (12 to 31 mg L^{-1}) with the onset of agricultural practices season and after the significant rain events suggest that the karst springs are vulnerable to contamination from point sources. The contamination of the karst aquifer, if remained unchecked, could pose a great threat to the health of the people using the water of the aquifer.

Keywords: Kashmir Himalayas, karst springs, hydrogeochemical processes, water quality, vulnerability.

INTRODUCTION

Water, the most essential resource that support existence of life on earth, plays a key role in the metabolic breakdown of essential molecules as protein and carbohydrates, as well as blood in animals and sap in plants. Besides, it regulates transportation as well as removal of waste material, enhances good health and nutritional status of any population (Meinzen-Dick, 1998) that is why through all ages, the human race has striven to locate and develop it (Kumar *et al.*, 2004). Kashmir, a region that bears complete stratigraphic record from Archean to recent (GSI, 1977) is bestowed with abundant fresh water resources both as surface (glaciers, lakes, streams) and subsurface (groundwater). In context to groundwater, the complex series of sedimentary formations, affected by intrusions of igneous rocks, fold, faults and wide spaced joints provide the basic framework for the hydrogeology and have resulted four independent or semi-independent hydrogeological units which constitute the major groundwater reservoirs in the Kashmir region(Jeelani, 2008). Among them, the Triassic Limestone aquifers are of strategic importance for the local community as they provide pristine water supply for drinking and irrigation in the form of large karst springs with high specific yield, i.e., 1000 m d^{-1} (Jeelani, 2008; Jeelani *et al.,* 2011; Jeelani and Shah, 2016). However, during a few years, the region has witnessed a tremendous increase in establishment of new housing colonies and/or human settlements and agricultural and horticultural practices within the immediate catchments of these fragile ecosystems. Groundwater quality within these karst aquifers can be put at high risk from the sensitive exokarstic features due to throwing/dumping into the recharge areas and make groundwater resources highly vulnerable to contamination. Attention on water contamination and its management has become a need of the hour because of far reaching impact on human health (Babu *et al.*, 2006; Jeelani *et al.,* 2014; Saleem *et al.,* 2015). Monitoring the hydrochemistry of karst springs, provide a useful information on hydrogeochemical characteristics of the karst aquifer, identification of subsurface flow routes of recharging water as well as the process that control the composition and quality of water (Rao, 2008; Jehangir *et al.,* 2011; Kumar *et al.,* 2014). In context to this, an effort has been made to understand and to assess the hydrogeochemical processes that control the composition and quality of water emerging from karst aquifers in the form of springs. The study will help to create suitable management plans to protect groundwater from these highly fragile ecosystems.

STUDY AREA

The study area includes Liddar mountainous catchment which lies between latitudes 33° 20′ N and 34° 15′ N and longitudes 74° 30′ E and 75° 35′ E (Fig. 1) with an elevation ranging from1600-5200 m above sea level (asl). The climate shows a great variability and marked seasonality in temperature and precipitation. Extreme daily maximum and minimum temperatures measured at Pahalgam weather station at 2100 m asl, range from 35°C in summer to -15°C in winter. March normally receives the maximum rainfall of the year (183 mm) and November the least (36 mm). The geology of the study region preserves a geological record of the Himalayan Orogenesis as well as the following sedimentary and volcanic processes. The main structural features observed in the stratigraphic sequences of the study region are faults, wide spaced joints and folds, which is turned into anticlines and synclines of several km wide. The study region is rich in surface (glaciers, streams, lakes) and subsurface water resources, among which large karst springs are the prominent.

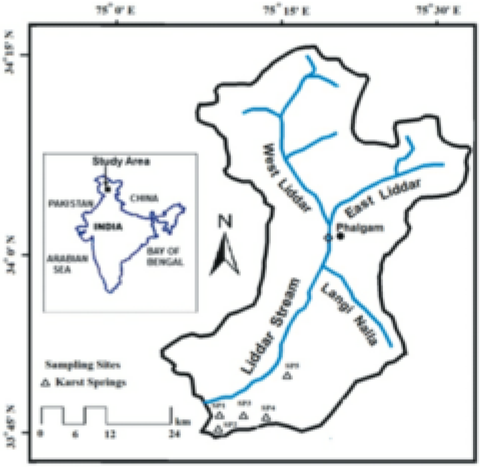

Fig.1: Location map showing the sampling sites (Note: SP1: Andernag, SP2: Malakhnag, SP3: Gujnag, SP4: Sayeednag: Sp5: Martandnag).

METHODOLOGY

Water samples (n=100) were collected monthly during 2013-2014 for chemical analysis from five major karst springs of Liddar catchment (Fig.1). To avoid the effect of floating debris, the samples were collected at depths >10cm below the water surface. The water samples were filtered using 0.45µm nucleomembrane to separate the suspended sediments and were later separated into two aliquots of 500 ml volume in HDPE bottles. One of the filtered aliquot was acidified with environmental grade (ultrapure) HNO_3 (2 *pH)* for cations (Ca^{2+}, Mg^{2+} Na^+ and K)$^+$ and trace elements (Fe and F), whereas other aliquot was preserved unacidified for anion measurements. In addition, one unfiltered sample (250 ml) was also collected for alkalinity measurements. Acidification of water samples preserves most trace metals and reduces precipitation, microbial activity and adsorption to walls of the sample container. Acidification is not required for anions determination, as it converts most of anions, thus doesn't permit to determine the individual anion from the sample. The standard methods were adopted to analyse the dissolved chemical constituents (APHA, 2001). The physical parameters such as water temperature, pH, Electric conductivity (EC) measured *in situ* by HACH (HQD Sension$^+$). Turbidity was measured by Portable Fluorometer (GGUN-FL30). Chemical analysis was carried out at Hydrogeology Laboratory, Department of Earth Sciences, University of Kashmir. Ca^{2+} and Mg^{2+} were determined by Ethylene diamine tetra acetate (EDTA) titration using murexide as indicator, whereas the Cl^- estimation was done by titrating the samples against $AgNO_3$ (0.02 N) using potassium chromate (5%) as an indicator. The HCO3- was determined by the titration of the water sample against HCl (0.01N) in which methyl orange was used as an indicator. The Na^+ and K^+ concentration in the samples were determined by using flame emission photometer (Systronics-130). SO_4^-, F^-, Fe^{2+} and NO_3^- were determined by UV-VIS spectrophotometer (EI-1371) at 420, 570, 510, 220nm, after following the standard procedures prior to their determination (APHA, 2001). During the analytical procedures, blanks and standards were run to check the reliability of the methods adopted. In majority of the water samples, the total cation charge ($TZ^+ = Ca^{2+} + Mg^{2+} + Na^+ + K^+ + Fe^{2+}$ in meq L^{-1}) balanced that of the total anions ($TZ^- = HCO_3^- + Cl^- + SO_4^- + NO_3^- + F^-$ in meq/L) within analytical uncertainties and the normalized inorganic charge balance (NICB = $(TZ^+ − TZ^- / TZ^+) \times 100\%$) was within ± 6%.

RESULTS AND DISCUSSION

Hydrochemistry

During the monitoring period, both physical and major chemical ions in karst springs waters varied considerably both spatially and temporally. The statistical summary of physico-chemical constituents in the karst springs are provided in table 1. Generally, spring water is fresh, transparent and ordour less. The pH which describes the intensity of acidity or basicity of water/fluid ranged from 7.5-9.5 with an average of 8 and therefore suggests alkaline nature of groundwater. However, the spatial variability of pH in karst springs were observed. The springs with lower discharge (Malakhnag and Gujnag) shows more pH (9-9.5) compared to springs with higher discharge (Andernag, Martandnag and Sayeednag). The water temperature which depend largely on the ambient temperature of the infiltrating water, thermal conductivity of rocks groundwater movement and its residence time, varied from (10-23 °C) with an average of 14°C. Water temperature was found lower (10-15°C) in karst springs with higher discharge like Martandnag and Andernag. Whereas the springs with lower specific yield like Malakhnag, Gujnag and Sayeednag, possessed higher water temperature (20-23°C). Electrical conductivity (EC) which is an index to represent the total concentration of soluble salts in water, varied from 283- 473 µS cm^{-1} with an average of 368 µS cm^{-1}. Generally, EC was mostly higher in Malakhnag and Gujnag springs and lower in Andernag, Martandnag and Sayeednag. In karst springs the turbidity varied from 0.72 - 4.4 nephelometric turbidity unit (NTU). Except, Andernagand Martandnag (0.72 -1.1NTU) all the karst springs exhibited higher values of turbidity (2.2–4.4 NTU).

Among the cations (TZ^+) the calcium (Ca^{2+}) dominated the cation budget by 53% followed by magnesium (Mg^+) 29%, sodium(Na^+) 14%, potassium(K^+) 4% and iron (Fe^{2+}) 1.5%. Ca^{2+}, Mg^+, Na^+, K^+ and Fe^{2+}, followed the same trend.The overall mean values of these ionic constituents varied from:Ca^{2+}: 54.5-105.3 mg L^{-1}, Mg^+: 30.8-63.7 mg L^{-1}, Na^+:10.1- 36 mg $L^{-1}$and K^+: 2-12.2 mg L^{-1}, Fe^{2+}: 1-5.8 mg L^{-1}. Ca^{2+} and Mg^+ was found higher in Andernag, Martandnag, Sayeednag and lower in Malakhnag and Gujnag. Higher proportion of Mg^{2+} (51-63 mg L^{-1}), Na^+, K^+ and Fe^{2+} was observed in Malakhnag and Gujnag springs. Among the anions (TZ^-), bicarbonate (HCO_3) made 90% of anion budget followed by sulphate (SO_4^-) 4.7%, chloride (Cl^-) 3.4%, nitrate (NO_3^-) 0.75% and fluoride (F^-) 0.18% of the total anion budget in the spring waters except in Malakhnag where SO_4^- mark 2nd place in abundance order. HCO_3^- SO_4^-, Cl NO_3^- and F varied from, 285-550 mg L^{-1}, 9.4-70mg L^{-1}, 7 to 31 mg L^{-1}, 1.8-12mg L^{-1} and 0.1-1.7 mg L^{-1}, respectively. Except, Malakhnag and Gujnag springs, the anions, showed higher concentration in all karst springs. Malakhnag showed higher values of SO_4^- (54-70 mg L^{-1}). It was observed that all major chemical constituents in spring waters exhibited more or less similar monthly and /or seasonal pattern with lesser ionic concentration during high flows (April-October) and higher concentration during low flow rates (November-March). Except, NO_3^-, Cl^- and F^-, all the ionic constituents (Ca^{2+}, Mg^+, Na^+, K^+ Fe^{2+}, HCO_3^- and SO_4^-) exhibited higher amounts in December/January and lower in July/August months. NO_3^-, Cl^- and F^- showed higher amounts during high flow period (May-September). The higher concentration of these ionic constituents in spring waters of the region is a strong evidence of speedy levels of anthropogenic activities within the source areas of the springs.

Sources of solutes

Variation in groundwater chemistry is usually used as an indicator to understand water-rock interactions and geochemical processes in the aquifers under different flow conditions. Ca^+ Mg vs HCO_3, Na vs Cl and Ca^++ Mg vs HCO_3 + SO_4, Na^++ K^+ vs TZ^+ (Fig.2) and Ca^++Mg-HCO_3-SO_4 vs Na-Cl plots, Ludwig-Langliar and piper trilinear diagrams (Fig. 3and 4) were used to identify and to understand the geochemical processes to evaluate the causes that control the spring water chemistry in the region. Scatter plot of Ca^++ Mg vs. HCO_3 (Fig. 2) shows that data falls above the equiline which suggest carbonate dissolution as a major sources of solutes besides some contribution from other sources. In Ca^+ Mg over HCO_3 +SO_4 (Fig. 2) dominancy of Ca and Mg over HCO_3 and SO_4 suggests the interaction of water-rock with silicate lithology also before emergence from the aquifer. Most of the data points lie above the aquiline in Na vs Cl^- plot (Fig. 2) which reflect the contribution from silicate lithology, because Cl is mostly associated with alkalies than alkaline earth elements. About 99% of samples showed Na / Cl^- ratio, > 1 which indicates that sodium is released from silicates as a result of silicate weathering. The presence of Fe and F in spring water is considered as lithogenic and may have released from the silicate rocks (e.g., Panjal Traps). The strong rock-water interaction followed by higher concentration of HCO_3 and pH value is one of the main reason for higher levels F in the spring waters.

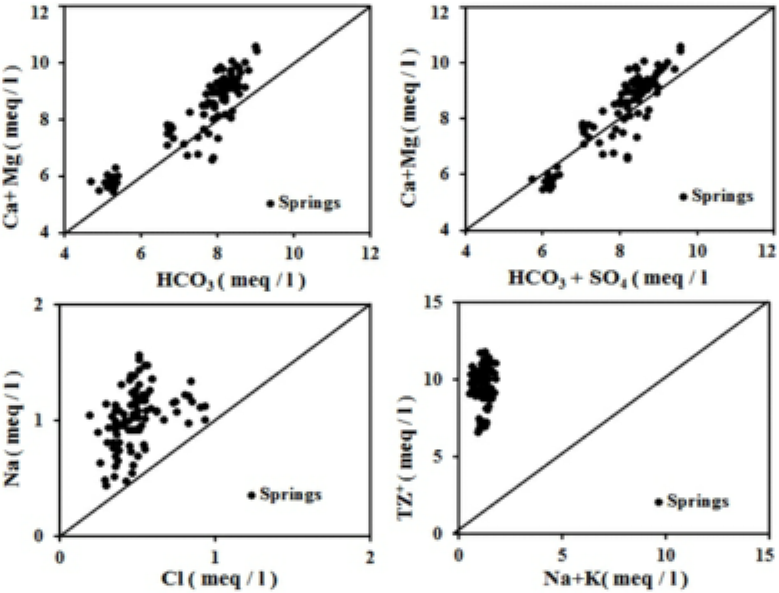

Fig.2: Scatter plots showing the sources of major ions in karst springs.

Table 1*:* Statistical summary of physico-chemical parameters in karst springs of the study area

Springs	Statistics	p H	EC	Turbidity	Water Temp	Ca	Mg	Na	K	Cl	HCO_3	SO_4	NO_3	Fe	F
			[illegible]	NTU	°C	Concentration: (mg L^{-1})									
Andernag	*Max*	8.7	469	1.1	14.0	105.3	63.7	30.9	5.3	16.2	550	33.4	4	3.4	1.3
	Min	6.9	392	0.93	10.0	87.2	49.1	20.0	3.1	12.2	487	24.0	1.8	2.1	0.5
	Average	7.9	436	-	12.2	98.1	55.1	24.7	4.0	14.1	514	27.6	3.9	2.7	0.9
Malakhnag	*Max*	8.7	448	4.4	22.3	65.8	37.0	28.6	6.6	14	327	70	8	5.8	1.5
	Min	7.5	299	3	17.3	54.5	30.8	20.2	2.4	12	285	42.0	2.9	2.2	0.7
	Average	7.9	394	-	19.5	59.3	33.7	23.9	4.5	12.0	314	54.3	4.1	3.7	1.0
Gujnag	*Max*	9.4	473	3.5	20.0	84.2	51.5	35.0	12	31	489	24.0	11	3.2	1.2
	Min	7.2	328	3	15.0	70.5	38.2	21.0	7.2	13.1	408	14.1	3.0	1.7	0.1
	Average	8.2	380	-	17.2	79.4	43.5	28.3	9.9	15.5	430	17.4	4.3	2.4	0.9
Sayeednag	*Max*	9.2	455	3	17.0	100.8	60.4	36.0	12	30	500	17.3	12	3.3	1.7
	Min	7.3	319	2.2	11.1	68.0	36.9	21.0	7.1	17.1	457	11.0	3.1	1.3	0.6
	Average	8.1	355	-	13.9	89.3	49.2	28.9	9.9	19.0	481	14.5	4.4	2.2	1.0
Martandnag	*Max*	8.6	448	0.91	15.9	101.0	54.8	30.1	6.2	14	498	16.6	8	4.6	1.3
	Min	7.2	298	0.72	12.0	73.6	36.3	10.1	2.4	8.6	470	11.0	2.9	1.3	0.6
	Average	8.0	341	-	14.3	94.1	48.6	20.2	4.6	12.3	486	14.0	4.5	2.5	0.9

Carbonate and silicate weathering

Quantification of carbonate weathering was estimated by Ca^{+} Mg/HCO_3 molar ratios. The Ca^{+} Mg/HCO_3 ratio close to 1, gives meaningful estimate of carbonate weathering. However, carbonate weathering involves dissolution of calcite and dolomite minerals. Therefore, to quantify calcite and dolomite weathering $Mg^{2+/}Ca^{2+}$ molar ratios are used (Szramek *et al.*, 2011). Mg^{2+}/Ca^{2+} of less than 0.1 indicates pure calcite dissolution whereas the ratio approaching to 1 refers pure dolomite dissolution and $Mg^{2+/}Ca^{2+}$ ratio of 0.33 represents equal contribution from both (Szramek *et al.*, 2011). The karst springs within the study region tend to have wider ranges (0.09-1.1) in $Mg^{2+/}Ca^{2+}$ ratios, which points toward a slight advantage of calcite dissolution over dolomite. It was observed that 77% of water samples showed calcite dissolution whereas 33% water samples showed dolomite dissolution. Although, quantification of silicate weathering is more difficult due to the incongruent degradation of silicates (Das and Kaur, 2001) however, the ratio between Na^{+} + K^{+} and total cations (TZ^{+}) gives useful estimation of silicate weathering (Stallard and Edmond, 1983). It was observed that majority of the samples lie near the Na^{+} + K^{+} = 0.2 TZ^{+} line in Na^{+} + K^{+} vs TZ^{+} plot (Fig. 2), thus indicate the role of silicate weathering in the geochemical processes, in contributing Na^{+} and K^{+} ions to the groundwater. The Na^{+} + K^{+}/TZ^{+} ratio (0.4-1.3) used as an index to evaluate the contribution of cations through silicate weathering also suggested release of Na^{+} and K^{+} to groundwater through same process.

Hydrogeochemical facies

The diagnostic chemical character of water solutions in hydrologic systems is generally determined with the application of hydrochemical facies (Jeelani *et al.*, 2014; Saleem *et al.*, 2015). Ludwig-Langliar (Ludwig-Langliar, 1942) and Piper Trilinear (Piper, 1944) plots has been used to characterize the water types in these karst springs of the region. Two hydrogeochemical facies were identified, Ca-HCO_3, Mg-HCO_3 Ca-Mg-HCO_3 type (Fig.3,4). The characterisation of groundwater with respect to Ca-HCO_3 and Mg-HCO_3 water types indicates the overall chemical character falling with the normal alkaline earth water group. The distributions of major chemical constituents in piper trilinear plot showed overlapping region, indicating similarity in the chemical constituents of spring water except Malakhnag and Gujnag springs. Malakhnag spring showed higher concentration of sulphate and Gujnag spring showed higher values of magnesium than other karst spring which suggested that these springs have a different drainage route with different lithology or environmental condition.

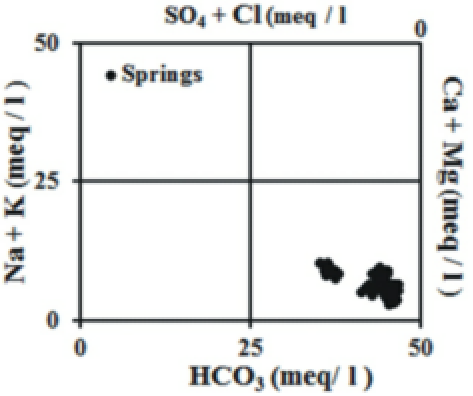

Fig. 3: Langliar Ludwig Plot showing the sources of major ions in karst springs.

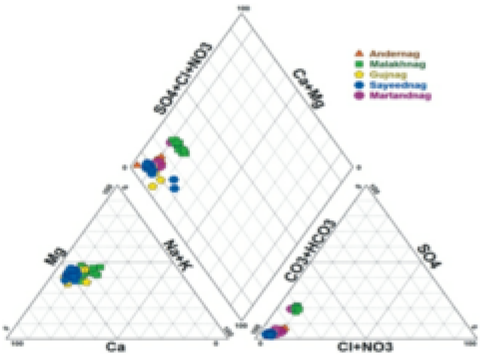

Fig. 4: Piper trilinear plot showing the hydrogeochemical facies in karst springs.

Vulnerability of karst aquifer to contamination

The water quality plays a vital role in promoting standards of human health and agricultural production. About 80% of diseases and over one third of the total deaths in the developing countries are caused by the use of contaminated water (UNESCO, 2000). Although spring water is excellent for domestic purposes when comparing the concentrations of measured ionic constituents in spring waters with WHO (2006) and BIS (2012) specifications for drinking water (Table 2), but the higher concentrations of NO$_3^-$ and Cl$^-$ as well as the concentration of some trace element (eg. F and Fe) in spring waters of the region is a matter of concern. The concentration of Cl and NO$_3$ has shown their increasing impact. The deterioration in spring water quality starts with the onset of the agriculture practice season and after significant rain events. We have noticed a significant increase in NO$_3$ and Cl concentration, from 2 mg L^{-1} to 12 mg L^{-1} and 12 mg L^{-1} to 31 mg L^{-1}, in summer and autumn months, respectively. Higher amounts of Fe (1-5.8 mg L^{-1}) and F(0.1-1.7 mg L^{-1}) was observed in all the karst springs. Occurrence of higher amounts fluoride in groundwater has a considerable impact on human physiology. The results of the analysis indicate that 1% of samples have higher F$^-$ concentration. Besides, we also examined sharp increase in turbidity in Gujnag (3.7-5 NTU: nephelometric turbidity unit), Malakhnag (4.4 NTU) and Sayeednag (0.96-2.4 NTU following significant rain events.

Dominant exokarstic and endokarstic features identified across the catchment, during hydrogeological field examination revealed that catchment is highly karstified. The exokarstic features seems to be the feeding channels of karst groundwater. The speedy levels of stone quarrying have exposed the solution features and internal drainage system of the carbonate aquifers (eg. Brakpora- Braridalow and Keherbal-

Wantrag transects). Any small thing (garbage etc.) would enter the groundwater through these exokarstic features and pollute the groundwater. Besides the use of polyethene is rampant in the catchment areas which may release the toxic chemical and pollute the pristine groundwater. It could be clearly seen that karst landforms (Karen fields, sinkholes, dolines) developed on karst ridges, particularly, along the course of stream banks are used as dumping sites. The dumping and/or throwing of solid/ municipal wastes into their karstified areas and/or recharge areas is a major concern in the study area. It was identified that during the high flow period, the dumping sites and other karst landforms remain under water and consequently adds toxicity to the stream flow which is ultimately supplied to the water table. The deterioration of water quality particularly the increase of faecal coliform, faecal streptococci is already reported during few years. Jeelani (2010) also have reported increasing concentrations of Fe and Cr and microbial bacteria (coliform and streptococci), in some karst springs of the region. The author observed that Total coliform, Faecal coliform and Faecal streptococci in the range of: 13-260, 2-92: 3-15 (MPN/100 mL), which is far above the permissible and desirable limits of WHO (2006) specification. Keeping in view the rapid flow and high permeability of karst aquifers, it is suggested that karst springs in the region are vulnerable to contamination. The study recommends that the identified recharge areas must be protected, restricted and conserved from the anthropogenic activities.

Table 2: Comparison of observed water quality parameters with the WHO (2006) and BIS (2012) drinking water specifications.

Parameters	WHO (2006)		BIS (2012)		Current study
	Desirable limit	Permissible limit	Desirable limit	Permissible limit	
p H	7-8.5	6.5-9.2	6.5-8.5	No relaxation	6.4 - 9.4
TDS (mg L^{-1})	500	1000	500	2000	442 - 739
Turbidity (NTU) (mg L^{-1})	1	< 5	1	5	0.72 - 4.4
HCO_3 (mg L^{-1})	-	-	200	600	285 -550
Ca (mg L^{-1})	150	500	75	200	54.5 - 105
Mg (mg L^{-1})	75	200	30	100	30 - 63.7
Na (mg L^{-1})	120	400	-	-	Oct-36
K (mg L^{-1})	50	150	-	-	2.4 - 12
SO_4 (mg L^{-1})	200	400	200	400	9.4 - 70
Cl (mg L^{-1})	250	500	45	No relaxation	8.6 - 31
NO_3 (mg L^{-1})	10	45	250	1000	1.8 - 12
Fe (mg L^{-1})	0.3	1	1	1.5	1 - 5.8
F (mg L^{-1})	-	1.5	0.3	No relaxation	0.1 - 1.7

CONCLUSION

Hydrogeochemical study was carried to understand the hydrogeochemical processes governing the evolution of the groundwater in karst settings. Ca^{2+} was the dominant cation and HCO_3^- was the dominant anion in groundwater. The order of abundance of cations followed Ca > Mg >Na>K>Fe and in anions the order follows HCO_3 > SO_4> Cl > NO_3> F. $CaHCO_3$and $MgHCO_3$, water types suggest preferential flow dominantly through limestone and dolomite rocks. However, higher concentration of SO_4 and Na, apart from Ca, Mg, and HCO_3 in Malakhnag and Gujnag springs reveals flow through both silicates and carbonate rocks. Congruent carbonate dissolution (calcite dissolution, dedolomitization) and incongruent silicate weathering were found to be the dominant processes controlling the spring water chemistry. The concentrations of various ions in most of the spring water samples are within the WHO (2006) and BIS (2012) standards indicating the waters are good for drinking purposes. However, growing concerns of wide limestone quarrying and spread of human settlements within the immediate catchments of the karst springs is a major concern due to the high permeability and least attenuation of the pollutants in the karst environments.

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