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Assessment of Temporal Variation in Hydrogeochemical Facies of River Water in the Central Himalayan Region

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Abstract: The water quality of a region is greatly influenced by the environment and its quality is a function of rock-water interaction and anthropogenic processes. 90% of the rural population of Uttarakhand depends on rivers and springs sources for their daily uses. Therefore, water characterisation and monitoring over time is important for human health and aquatic life. The basic understanding of hydrochemistry can be extracted by a trilinear piper diagram. The study also intends to develop hydro-chemical analysis for water quality at various locations in Uttarakhand to represent four major river catchments. The study attempted over 50 locations from 2010 to 2019 during pre and post-monsoon seasons. Four anions and four cations were used to understand hydrogeochemical facies. Result reveals that the most dominant hydrogeochemical facies were Ca-Mg-HCO_3 in the entire region followed by Ca-Mg-Cl or Ca-Mg-SO_4 facies due to limestone-dominant geology. Na-Cl or Na-SO_4 facies enhanced due to agriculture activities while $NaHCO_3$ facies due to the rapid weathering processes along the river course over the period. The study recommended for the area where the water type is either Na-Cl or Na-SO_4 or Na-HCO_3, intensive water resource planning should be implemented. These areas are highly vulnerable to human health costs.

Key words: Hydrogeochemical facies, water quality, spatio-temporal, Himalayas.

Introduction

Unplanned management of water resources at the individual and sectoral levels has reduced the quality and availability of safe drinking water and has led to human and animal infestation throughout the food cycle which increases the risk of various diseases. The main source of drinking water in hilly areas is fresh surface water, which is susceptible to contamination by various processes of human and natural activity. Both human and natural activities can alter the physical, chemical, and biological properties of water, with consequences for human and ecosystem health.

The majority of previous studies were conducted on a local scale in either the urban or rural domain. Several types of research have been conducted on both raw and supply water separately; however, limited studies have been conducted to determine the severity of water quality at large geographical coverage. As a result, the study proposes to look at the severity of water quality in river water in the spatio-temporal domain was chosen to examine intra-regional variations. A thorough assessment of the qualitative aspects of surface water is needed to meet the ever-increasing demand for water for various purposes. Changes in the underlying rock formations cause spatial variations in water quality.

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Surface water is an essential parameter to be studied for the sustainable development of urban and rural human life. However, the study also intends to develop hydrochemical analyses for water quality at various locations in Uttarakhand to represent the entire state under four major river catchments.

The water quality of a region is greatly influenced by its environment. Water quality variability is a function of the physical and chemical processes of water with underlying geology and anthropogenic interaction (Subramani et al., 2005; Tyagi et al., 2013). Surface and groundwater carry residues of surface materials from terrestrial and subsurface geology and can contain various contaminants (Tong et al., 2016). This depends on above-ground land cover and below-ground geology and soil properties. Minerals and rocks (Smedley & Kinniburgh, 2002), flora and fauna (Cerejeira et al., 2003), industrial waste (Carneiro et al., 2010), and urban runoff (Lapworth, et al., 2012) contribute to higher levels of pollution. About 90% of the rural population of the study area depends on rivers and springs for their daily water use (Tyagi et al., 2013) in Uttarakhand. The major drinking water source is gathered from small tributaries and springs similar to any other part of the Himalayas. The improper sewage system (Hassan et al., 2015), urbanisation process (Padmaja et al., 2017), development activities (Chandra et al., 2006) and agriculture practices (Hassan et al., 2015) which was noticed as the main responsible factors for water pollution of rivers in the recent time. Numerous pieces of research were conducted in past and ensured the issue related to surface water quality in this region by studying several water quality parameters (WQPs). The anthropogenic activities contaminated the Bhimtal Lake (Pant et al., 2017; Tiwari et al., 2015) and also increased the coliform and nitrate concentration in Nainital Lake (Das et al., 2007; Pande et al., 1983). Water quality parameters above the permissible limit of drinking water were reported in Udham Singh Nagar, Champawat, Pithoragarh (IMIS, 2009), Almora (Kumar et al., 1997) and Nainital district (CWP, 2005) in surface water sources. The quality of lake water deteriorated due to human activities in Dodi Tal (Kumar et al., 2019) and Badhani Tal (Kumar and Sharma, 2018).

Therefore, characterizing water, and identifying quality changes and trends in water quality over time is important for human health and aquatic life. The basic understanding of hydrochemistry can be extracted by a trilinear piper diagram by plotting cations and anions (Subramani et al., 2005), which are represented by the percentage of total milliequivalents per liter (meq/L) (Jasrotia et al., 2018).

Study Area

Uttarakhand is one of the ecologically rich Himalayan states but due to rapid industrialisation, tourism and urbanisation activities, the water bodies in the state and their qualities are deteriorating which is mostly caused by sewerage and wastewater discharge. However, in the recent past, there have been reports of the state encountering problems with water resources. Most of the districts in Uttarakhand are reported to have poor quality drinking water sources reported by DST researcher groups as well as other reports have also shown higher contamination as per BIS (10500), 2012. Hence, there is a need for systematic hydrochemical study, which can provide a solution to the water resource problems of the mountainous state and region. In this study, the four major river catchment; Yamuna, Upper Ganga, Ramganga and Sharda or Ghaghar of Uttarakhand has been taken into consideration to represent water quality threats in the Himalayan region. The study attempted over 50 source locations from 2010 to 2019 during pre and post-monsoon seasons which cover major rivers of the central Himalayas (Figure 1).

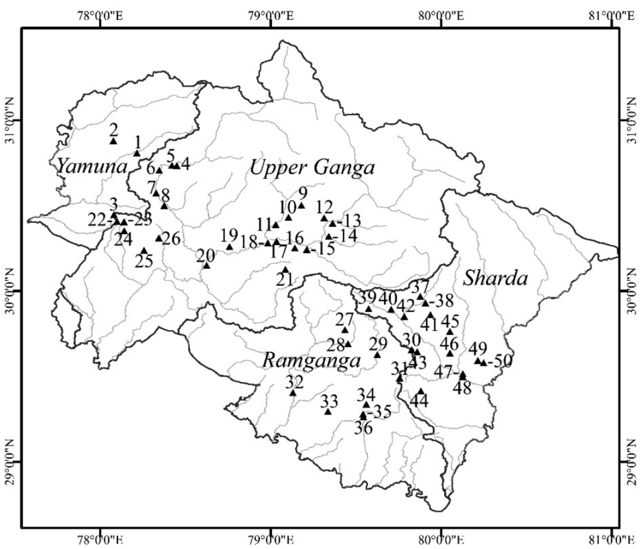

Figure 1: Location of surface water sample in Uttarakhand.

Data Used and Methodology

Sampling Method

The location of 50 drinking water supply sources was tested in this study during the pre and post-monsoon periods of 2018 and 2019 through primary investigation.

To see the decadal change at the same location during the pre and post-monsoon periods of 2010 and 2011, data was taken from the previous research as the secondary source. Previous research was carried out between 2010 and 2011 at 50 locations of river water sources in Uttarakhand which were taken as a reference period and primary data were collected in 2018 and 2019 at the same location. The number of sites was restricted due to the limitation of previous pre-monsoon and post-monsoon datasets for the years 2010 and 2011. These locations were revisited in 2018 and 2019 for data collection (Annexure-A). The Indian Standard IS:10500 (2012) prescribes the methods of tests for drinking water and formulated IS: 10500 drinking water standards for good human health.

All the water samples were collected in 1-L high-density polyethylene (HDPE) bottles pre-washed with 10% nitric acid (HNO_3) and rinsed with double-deionized water. All HDPE bottles were thoroughly rinsed three times with the sample water to be sampled to avoid unpredictable changes in characteristics as per standard protocols. A total of eight parameters of drinking water quality were analysed to understand the hydrogeochemical facies process (Table 1).

Analysis of Data

The basic concept of hydrochemistry is to diagnose the chemical properties of aqueous solutions in hydrological systems. Various chemical processes occur within the geological unit resulting in a change in the chemical properties of the water. Facies reflect the influence of chemical processes that occur between minerals and water (HatariLabs, 2018) within the petrological framework (Back, 1960). The water carried abundant ions in the form of positively charged (cations) and negatively charged (anions). Due to electroneutrality, cations and anions are in equilibrium and constitute most of the dissolved solids in water. Therefore, the availability of cations and anions plays a significant role in the understanding hydrochemistry of water (Sadashivaiah et al., 2008). The ionic composition of water helps to produce ionic types based on classification primarily on dissolved cations and anions, which are expressed in meq/L. A milliequivalent (meq) is a measure of the molar concentration of an ion normalised by the ion's ionic charge. Dominant dissolved ions should account for at least 50% of the total (USGS, 2020).

A piper diagram consists of two trilinear diagrams on the lower side and one diamond-shaped diagram on the upper middle in which left trilinear diagrams represent cations and right trilinear diagrams represent anions (Figure 2). Three axis of cations contain the value sodium plus potassium ($Na^+ + K^+$), calcium (Ca^{++})

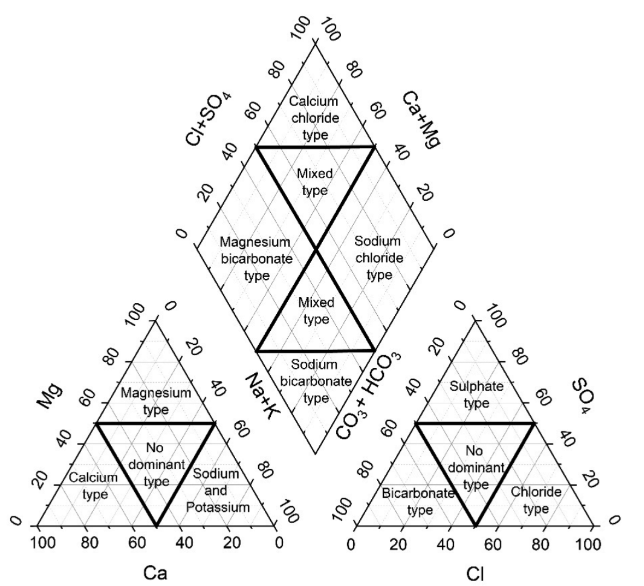

Figure 2: Piper diagram showing various hydrochemical facies.

Table 1: Quality parameters, technique and test methods

No.	*Name of parameter*	*Technique used*	*Test method*	*Standards (BIS-10500, 2012)*	
				Desirable	*Permissible*
1.	Alkalinity (CO_3 +HCO_3)	Titration	IS 3025 pt-23-2003	200	600
2.	Sodium (Na)	Atomic absorption spectrophotometer (AAS)	APHA 23rd-3111B	-	200 (WHO, 2009)
3.	Potassium (K)	AAS	APHA 23rd-3111B	-	-
4.	Calcium (Ca)	AAS	APHA 23rd-3111B	75	200
5.	Magnesium (Mg)	AAS	APHA 23rd-3111B	30	100
6.	Chloride (Cl)	Spectrophotometer	APHA 23rd-4500-Cl-B	250	1000
7.	Sulphate (SO_4)	Spectrophotometer	APHA 23rd 4500 SO4-E	200	400

and magnesium (Mg^{++}) in % of meq/L, while anions axis consists of bicarbonate plus carbonate (HCO_3^- + $CO3^{--}$), sulphate ($SO4^{--}$) and chloride (Cl^-) in % of meq/L. The diamond field covers the combination of cations and anions (RockWare, 2020). These trilinear diagrams bring out the chemical relationships among water samples with geology in terms of ore under definite terms rather than possible plotting methods (Sadashivaiah et al., 2008).

The modified piper diagram also known as the Chaddha diagram was developed (Chadha, 1999) for identifying the chemical process in natural water. The chemical processes are represented by four quadrants: recharging water (Ca-Mg-HCO_3 type), reverse ion-exchange water (Ca-Mg-Cl type), seawater/end-member waters (Na-Cl type) and base ion-exchange water (Na-HCO_3 type) (Ravikumar et al., 2017). Further, Chadha (1999) diagram was classified into eight types (Ravikumar et al., 2017), that is, alkaline earth (Ca+Mg) exceeds alkalies (Na+K); alkalies exceed alkaline earths; weak acids (CO_3 +HCO_3) exceed strong acids (SO_4+Cl); strong acids exceeds weak acids; magnesium/calcium bicarbonate type (temporary hardness); calcium-chloride type (permanent hardness); Sodium-chloride type (saline); sodium-bicarbonate type (alkali carbonate) and mixed type (no cation-anion exceed 50%).

Result and Discussion

In this study, Piper plots from 2010 to 2019 were created to understand the water chemistry of samples. This analysis indicates that the majority of the samples over the years were calcium bicarbonate type water. This means that over 50% of the total cation milliequivalents are present as calcium and over 50% of the total anion milliequivalents are bicarbonate. During 2010-11, most of the water samples clustered and compacted in the zone of alkaline earth and weakly acidic anions outnumbering both alkali metal and strongly acidic anions, indicated by the Ca-Mg-HCO_3 water type (Figure 3). In the later period (2018-19), the cluster was converted to a dispersed pattern and few samples moved toward Ca-Mg-Cl or Ca-Mg-SO_4 water types (Figure 4). The result reveals that water quality modified over the period. The ion exchange process is also altered due to seasonal changes. The water chemical processes described by the Chaddha diagram can be divided into four classes (Table 2).

The position of water sample data in scatter plot in piper chart Chaddha's plot lies in (type 1), which is a Ca-Mg-HCO_3 type of water which is also depicted as recharging water. Recharging water was formed when water enters the surface runoff from the surface, carrying dissolved carbonates in the form of HCO_3 and the geochemical origin of Ca and Mg. The Carbonate-salt weathering is expected to be more predominant due to calcium and magnesium ions in these locations.

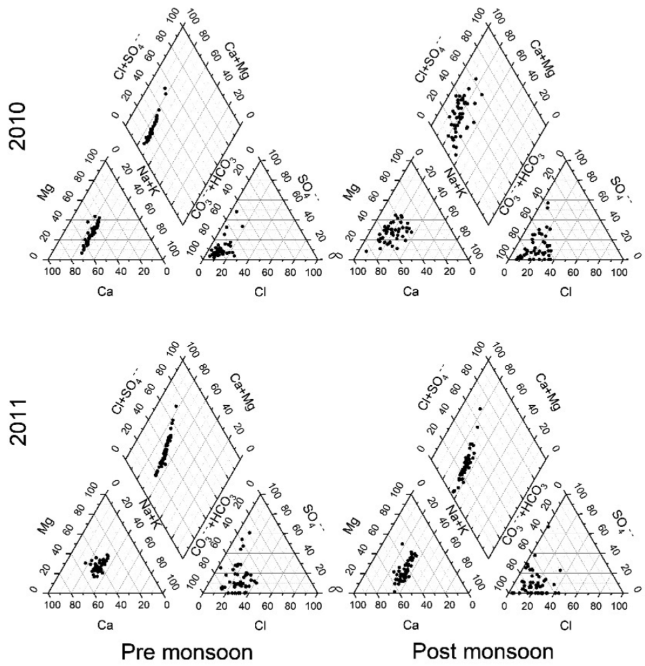

Figure 3: Piper diagram showing water types in Uttarakhand from 2010 to 2011.

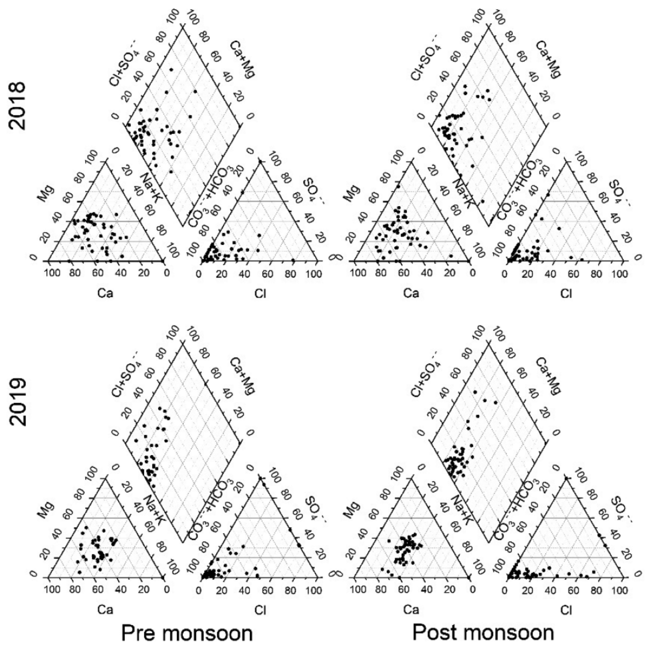

Figure 4: Piper diagram showing water types in Uttarakhand from 2018 to 2019.

The Ca-Mg-Cl or Ca-Mg-SO_4 type (type 2) is indicated to be predominant in limestone-dominant geology. A Na-Cl type (type 3) was also detected in some samples which could be due to the interaction between water and halite minerals and the source of chlorine in water systems through fertiliser uses. Water-type $NaHCO_3$ (type 4) results from silicate weathering, where sodium, potassium, calcium, and magnesium silicates interact with carbonic acid to form various cations in water systems.

The most dominant water types were type 1 (Ca-Mg-HCO_3) in the entire region and type 2 (Ca-Cl type) mostly observed in limestone-dominant geology suggesting that groundwater-surface water interactions decreased over this period (Table 3). Subterranean contributions are relatively high during the pre-monsoon season due to the absence of precipitation in the basin. In this case, pre-monsoon subsurface geology contributes to water-type dominance when there is a strong association of surface-subsurface interactions.

It was also observed that the type 3 (Na-Cl type) water samples increased over the period (Figure 5), this could be due to agriculture extension and the use of fertiliser in the basin. This sample value is high during the pre-monsoon period than the post-monsoon due to high surface runoff. Temporal data indicates there is limited scope to conclude halite minerals or salt in geology which is also supported by research on halite minerals in the Himalayas (Gaury et al., 2018). The water type $NaHCO_3$ (Type 4) is also identified during the analysis, indicating a rapid silicate weathering process in the river system and consistent with extreme precipitation events or rapid construction activity in the river basin.

Water type 4 mostly appears in Bhagirathi and Alaknanda Rivers during 2018-19 (Figure 6) due to rapid contractions work of hydro-power plant and *Char Dham* road construction along these river courses at locations no. 6, 7, 8, 11, 17 and 20 (Figure 1). It was also observed that most of the water type 2 and 3 is predominant due to limestone-dominant and halite minerals geology (Figure 6) near Mussoorie at locations 3, 22, 23 and 24 (Figure 1). Sample locations 27 and 28 are situated in the Ramgamga river basin which was

Table 2: Source of different water types

Types	*Water type*	*Goegenic source*	*Anthropogenic source*
Type 1	Recharging water (Ca-Mg-HCO_3 type)	Calcite, dolomite and limestone Gypsum (Carbonate minerals) (Singh et al., 2015)	
Type 2	Reverse ion exchange water (Ca-Mg-Cl or Ca-Mg-SO_4 type)	Cement material (Gypsum) (Chadha, 1999)	
Type 3	Saline /end-member waters (Na-Cl type or Na-SO_4)	Halite minerals (Joji, 2016; Gaury et al., 2018)	Fertilisers (Potash) (Molly Hunt, 2012)
Type 4	Base-exchanged water (Na-HCO_3 type)	Kaolinitic clay, sandstone, shale, limestone, dolomite, phyllite, schist, granite, gneiss, and intrusives. Silt, sand, and clay (Chadha, 1999)	

Table 3: Various water types in Uttarakhand

		2010		*2011*		*2018*		*2019*	
		Pre	*Post*	*Pre*	*Post*	*Pre*	*Post*	*Pre*	*Post*
Type 1	$CaHCO_3$	26	44	44	39	29	30	31	26
	$MgHCO_3$	12				9	11	10	12
Type 2	$CaSO_4$		1	3		1	1		
	$CaCl_2$	3				1	2		3
	$MgSO_4$		5	3	11				
	$MgCl_2$							1	1
Type 3	Na_2SO_4	2				1		5	2
	NaCl	3						2	2
Type 4	$NaHCO_3$	4				9	6	1	4
Total samples locations		50	50	50	50	50	50	50	50

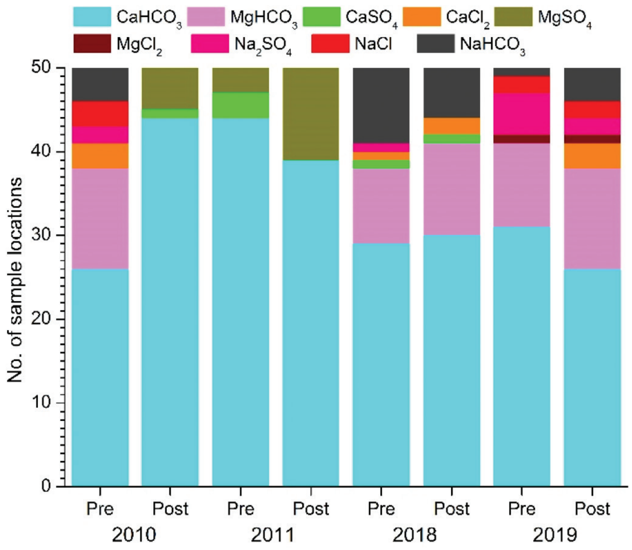

Figure 5: Changing profile of various water types over the period.

Figure 6: Spatio-temporal change in water types (Chadha, 1999).

converted from type 1 to type 3 and 4 due to agriculture extension activities. Sharda (Ghaghar) basin was not much influenced by human activities such as fertiliser practices and construction activities. Therefore, most of the water sample was observed under type 1 and type 2 based on local geological interactions. Overall, the Uttarakhand sample of water type is shifting towards type 3 and type 4 which indicated the impact of human and climate change on the basin and needs proper intervention.

Conclusion and Discussion

This study focused on the surface source of drinking water and its quality issues. The study was conducted from 2010 to 2019 in two seasons (pre and post-monsoon). Most of the sample depicts recharging water (Ca-Mg-HCO_3) and their sample value increased during the post-monsoon period whereas Mg-HCO_3 samples increased due to sedimentation of the surface runoff throughout the time. Regional geology has yield type 2 (Ca-Mg-Cl or Ca-Mg-SO_4) which is the dominant geology of limestone but has a seasonal influence on its occurrence. Due to agriculture extension and increased fertiliser uses over time, Na-Cl or Na-SO_4 type (type 3) water increased in recent times which is earlier not detected. This type of water increased during the pre-monsoon period and decrease during post-monsoon due to the impact of high surface runoff. The $NaHCO_3$ (type 4) of water was observed due to the rapid weathering process either extreme rainfall events or high construction activities along the river course.

Overall, water quality in Uttarakhand is moving towards type 3 and type 4, indicating strong impacts of human activities and climate change on water quality. Finally, in areas where the water type is either Na-Cl or Na-SO_4 or Na-HCO_3, intensive water resource planning should be implemented. These areas are highly vulnerable to human health costs. These selected locations can be treated individually.

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Consent to Participate

All authors are aware of the submission of the manuscript in this journal is associated with raw river water quality assessment and "Consent to Participate" is not applicable in this study. This paper does not include human subjects and/or animals trails.

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Competing Interests
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.

Availability of data and materials
The datasets generated and/or analysed during the current study are not publicly available due to State Agency Confidentiality request. The Analysis and testing were carried out at National Accreditation Board for Testing and Calibration Laboratories. The laboratory is consortia of multiple agencies which includes State Agency Uttarakhand Jal Sansthan (UJS) but are available from the corresponding author on reasonable request. One of the co-author is Co-ordinator of this laboratory.

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