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Groundwater quality assessment for drinking and irrigation purposes in arid to semiarid region of Indus Basin of South Punjab, Pakistan

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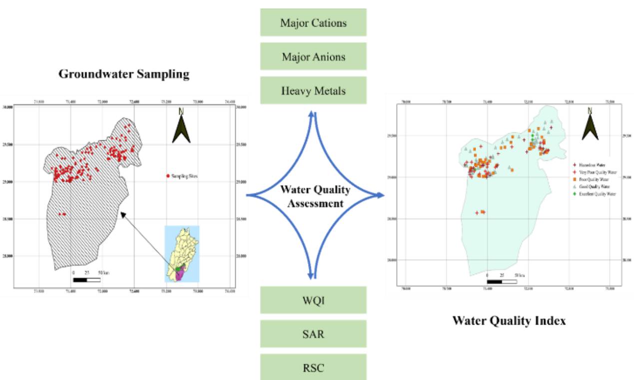
Graphical abstract



Abstract

As the groundwater pollution is increasing, it is crucial to assess groundwater quality and characterize hydrogeochemistry accurately for long term water supply. In this study groundwater samples were collected from 120 locations from Bahawalpur district and analyzed for electrical conductivity (EC), pH, total dissolved solids (TDS), sodium (Na^+), potassium (K^+), calcium (Ca^{2+}), magnesium (Mg^{2+}), carbonate (CO_3^-), bicarbonate (HCO_3^-), sulfate (SO_4^{2-}), chloride (Cl^-), nitrate (NO_3^-), fluoride (F^-) and heavy metals. The results obtained from the analysis of samples showed that the SAR of nearly 65% of samples collected from Bahawalpur district were in acceptable limit (<6 meq L^{-1}), while 12% found to be unfit for irrigation (>10 meq L^{-1}). Similarly, the RSC values depicted that 8% samples were unfit while 64% samples were falling in the category of fit irrigation water. Most of the water samples were predominantly Ca^{2+}-HCO_3^- and Na^+-HCO_3^- type, that were controlled by various processes of cation exchange, water-rock interaction, dissolution and evaporation. Some samples fall in the middle of diamond and lower triangles of Piper diagram are showing no dominant type of water (mixed water type) due to the complex influence of rock-water interactions as well as anthropogenic activities. The Water Quality Index (WQI) showed that out of total 120 sampling sites in Bahawalpur the number of excellent water samples were only 2% while 18% water samples were characterized as hazardous. Moreover, the number of samples falling in the categories of good, poor and very poor quality water were 36%, 29% and 13% respectively.

Keywords: Bahawalpur, groundwater quality, heavy metals, hydrogeochemistry, water quality index

1. Introduction

Water is the most valuable and crucial resource for all life on earth especially for human development (Loganathan and Ahamed 2017). Aquifers which are exploited on every part of earth and serve as the prime source of drinking water for >1.5 billion people globally contain over 97% of liquid fresh water and 30% of all fresh water (Oskin 2018). As one of the most fundamental humanitarian objectives dependable access to inexpensive, clean water is still a significant global concern in the 21st century. The current worldwide water problem is largely caused by industrial water use, as a result of a significance rise in both industry and population (Santos *et al*. 2014). Presently, above 2 billion people live in areas facing high water stress and their number would remain to rise (He *et al*. 2021). The problems related to water are likely to grow worse in the coming decades with water stress occurring worldwide even in the regions currently considered water rich (Malato *et al*. 2022).

The quality of water represents a vital role for its use in domestic, industrial as well as agricultural purposes. With respect to chemical contaminants the water quality is considered with respect to soluble ions, heavy metals, hardness and nitrate in rural as well as urban areas of the world (Amarasooriya and Kawakami 2019, Ahamad *et al*. 2020). Many countries in Asia are facing increased level of water pollution due to the addition of untreated waste water containing organic and inorganic pollutants into water resources (Uddin and Jeong 2021). Since the middle of the 20th century, groundwater resources have considerably decreased in both quantity and quality because of anthropogenic constraints, population growth, expanding urbanization, pollutant runoff, poor sanitation and industrial and agricultural development (Ray *et al*. 2017).

There is growing evidence that irrigation water may need to be filtered sooner rather than later due to the occurrence of heavy metals in food. The world's freshwater supply is under pressure from growing irrigation but there are additional factors contributing to the freshwater access (Mishra 2023). Freshwater bodies are being redistributed to a once-balanced hydrosphere as a result of changes in the water cycle brought about by climate change in different parts of the world (Chen 2019). Due to water's ability to regulate energy on a planetary scale this redistribution has the effect of intensifying weather patterns making dry seasons drier, summers hotter and the natural recharge rates of reservoirs and aquifers decreasing (McMahon *et al*. 2011). As the recharge of subsurface freshwater supplies not being able to keep up with the pace of withdrawal, agriculture is severely impacted and irrigation becomes less dependable (Janakarajan and Moench 2018). The continued flow of saline wastewater into the groundwater without treatment has posed a hazard to aquatic, terrestrial and wetland ecosystems. Currently salinization affects around 33% of the world's irrigated farmland (Munns 2005). Irrigating with saline water can cause salt to build up in the soil, reducing yield and deteriorating soil resources (Feizi *et al*. 2010). Many more areas with good-quality groundwater are threatened with contamination as a consequence of unnecessary withdrawals of groundwater (Roy and Shah 2002). Similarly, contamination of water bodies with heavy metals especially in developing countries has become a matter of great concern (Gao and Chen 2012). Unlike organic pollutants in the environment, heavy metals are more persistent and tend to accumulate in different parts of the environment.

The problem of groundwater pollution in numerous parts of Pakistan has developed so severe that without crucial steps are taken, extensive groundwater resources may be spoiled (Daud *et al*. 2017). According to a report by International Monetary Fund (IMF), Pakistan is listed in the top countries facing severe water shortages. The Pakistan Council of Research in Water Resources (PCRWR) declared that by 2025 most of the country will experience very little or no clean water availability (Ahmed *et al*. 2020). At present, only 20% of population has admittance to clean drinking water while remaining 80% of the country's population relies on polluted water mainly by sewerage, pesticides, fertilizer and industrial effluents (Sahoutara 2017). Such level of water pollution is responsible for nearly 30% of deaths and 80% of all diseases in Pakistan (Daud *et al*. 2017). Moreover about 17% area of Punjab and 75% in Sindh is underlain by saline groundwater (TDS>3000 ppm) which cannot be used for both drinking and irrigations purposes.

The Bahawalpur district at the edge of Cholistan desert is a fast developing and urbanizing district in the Punjab province, where groundwater is the main source for domestic and irrigation purposes. A few studies have been conducted on groundwater quality in response to deterioration due to overexploitation, excessive use of chemical fertilizers and other anthropogenic activities (Aamer *et al*. 2014, Mohsin *et al*. 2019). However, a detailed geochemical analysis and classification of groundwater quality has not been evaluated in the said district. Therefore, the current study was planned to evaluate groundwater quality of Bahawalpur district where main objectives were (1) to analyze various physicochemical parameters to pinpoint the suitable and unsuitable groundwater quality areas for drinking and irrigation purposes, (2) to evaluate the groundwater suitability by comparing the measured parameters with the guideline values, and (3) to prepare the spatial distribution map of the Water Quality Index (WQI).

2. Material and methods

2.1. Study area

Bahawalpur district is situated in southern part in Punjab province of Pakistan and lies between 27°-80' to 29°-70' N latitudes and 70°-54' to 72°-50' E longitude along the south eastern bank of river Sutlej near the Cholistan desert. With the population size of 3.6 million people and population density of 150 persons per km^2, it is 12th largest city of Pakistan (Pakistan Bureau of Statistics 2017). It covers an area of almost 24,830 km^2 with small industries in urban areas and temperature normally ranges between 13 to 48°C with annual rainfall around 150 mm (Govt. of Punjab and World Bank 2006).

As the Bahawalpur is an agricultural district nearly 73% population in villages is involved in agricultural production. Most of the suburban areas are irrigated with effluent released from the industries as well as municipal waste water for the production of crops, vegetables and forages. Moreover, groundwater extracted by either hand or electric pumps is the main source of drinking water in the selected area. In this study 120 sampling sites with four replications for each were selected from the study area on the basis of population density and groundwater consumption (drinking or irrigation purpose) for the assessment of groundwater quality. The map of the Bahawalpur district indicating the sampling points is shown in Figure 1.

2.2. Physiochemical and heavy metal analysis

Physiochemical analysis of water samples was carried out for the determination of cationic, anionic and trace elements according to their standard procedures. All chemicals and reagents used during the analysis were of the analytical grade. Moreover, all the instruments were standardized before actual determination with accurate standards of each element/parameter.

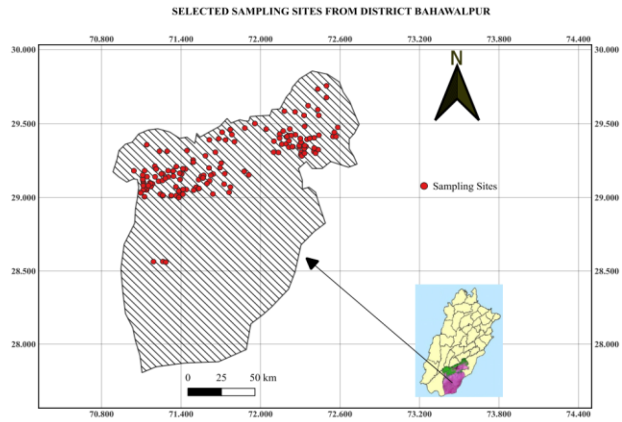


Figure 1. Map of Bahawalpur district indicating sampling points.

In the field, water pH, electrical conductivity (EC), temperature and total dissolved solids (TDS) were measured using pH meter (Milwakee H0030229), EC meter (OHAUS ST3200C), thermometer and TDS meter (Hanna TDS meter HI98311) respectively (Clesceri *et al*. 1998). The concentrations of major cations (K^+, Na^+ and Ca^{2+}) were measured through a specific procedure using Flame Photometer (BWB XP made in UK) after calibrating the instrument with standards of known concentrations. Four replications for each of the selected area were run for cations determination and the instrument was calibrated after every 20 samples (Hameed *et al*. 2022). The combined concentration of Ca^{2+} and Mg^{2+} and hardness of water were measured by titrating sample solution using ethylene diamine tetra acetic acid (EDTA, 0.01 N) in the presence of eriochrome black T (EBT) and a buffer solution at pH 10 and 12 respectively (Nasrin *et al*. 2014). A titration method of USSL, (1954) was followed for the measurement of CO_3^- and HCO_3^- using 0.1N H_2SO_4 as titrating agent and indicator solutions of phenolphthalein and methyl orange respectively. Cl^- was also analyzed by titration method using 0.2N silver nitrate ($AgNO_3$) in the presence of potassium chromate (K_2CrO_4) indicator. Anions including NO_3^- and SO_4^- were measured through UV/VIS Spectrophotometer (CECIl CE7400S) at a wavelength of 229 and 420 nm respectively (APHA 1998).

The analysis of heavy metal (Fe, As, Cr, Pb, Ni) concentration were carried out through Atomic Absorption Spectrophotometer (Agilent Technologies, AAS 200, USA). Quality control of heavy metal determination was measured with blank, reference standard solutions and duplicate samples and the analytical error was estimated to ≤10% (Estefan 2013). Fluoride concentration in collected water samples was determined through standard SPADNS colorimetric method using UV/VIS Spectrophotometer (CECIl CE7400S) at a wavelength of 570 nm (Arancibia *et al*. 2004). Piper (1944) trilinear diagram and a Durov (1948) plot were used to understand the geochemical progression of groundwater in particular. The diagrams in this study were plotted using the AquaChem software (AquaChem v4.0). Gibbs (1970) projected two diagrams to know the natural mechanisms of surface water chemistry. Gibbs diagrams basically depend on two ratios which are calculated by the following equations;

$$Gibbs\ ratio I = \frac{Cl}{\left(Cl + HCO_3\right)} \quad (1)$$

$$Gibbs\ ratio II = \frac{Na^+ + K^+}{Na^+ + K^+ + Ca^{2+}} \quad (2)$$

Where all the ionic concentrations are stated in meq L^{-1}.

2.3. Sodium Adsorption Ratio (SAR) and Residual Sodium Carbonate (RSC)

The Sodium Adsorption Ratio (SAR) (Lesch and Suarez 2009) and Residual Sodium Carbonate (RSC) were calculated by using the equations given below (USSL 1954);

$$SAR\left(meq\ L^1\right) = \frac{Na^+}{[\frac{Ca^{2+} + Mg^{2+}}{2}]^{1/2}} \quad (3)$$

$$RSC\left(meq\ L^1\right) = \left(CO_3 + HCO_3\right) - \left(Ca^{2+} + Mg^{2+}\right) \quad (4)$$

Where Na^+, Ca^{2+} Mg^{2+} CO_3^- and HCO_3^- are all measured in meq L^{-1}.

2.4. Water Quality Index (WQI)

Water Quality Index (WQI) was used to assess the groundwater quality for drinking purposes in the selected region (Adimalla *et al*. 2018). It is calculated using the following equations;

$$WQI = \frac{\sum Wn \times Qn}{\sum Wn} \quad (5)$$

Where;

$$Quality\ rating\ (Qn) = \frac{Vn - Vo}{Sn - Vo} \times 100 \quad (6)$$

$$Unit\ weight(Wn) = \frac{K}{Sn} \quad (7)$$

Here, Vn is the assessed value of parameters, Vo is the ideal or reference value, Sn is the suggested allowable limit of the parameter and K is constant of proportionality. WQI was classified giving to the criteria defined by the WHO (2004), (Table 1).

Table. 1. Criteria for the classification of groundwater quality on the basis of WQI.

Classification of water quality	WQI
Excellent quality water	0-25
Good quality water	26-50
Poor quality water	51-75
Very poor quality water	75-100
Unfit for drinking	>100

2.5. Heavy Metal Pollution Index (HPI)

The HPI signifies the total quality of groundwater with relation to heavy metals. The HPI model proposed is given by Mohan *et al*. (1996);

$$\text{HPI} = \frac{\sum_{i=1}^{n} W_i Q_i}{\sum_{i=1}^{n} W_i} \quad (8)$$

Where Qi is the ith parameter's subindex. n is the number of constraints taken into consideration and wi is the unit weightage of the ith parameter. The sub index (Qi) of the parameter is derived by;

$$Q_i = \sum_{i=1}^{n} \frac{\{M_i(-)I_i\}}{S_i - I_i} \times 100 \quad (9)$$

Wherein Si is the typical value of the ith parameter, Ii is the ideal value and Mi is the observed value of the heavy metal. Ignoring the algebraic sign, the sign (-) denotes the numerical variance between the two values. The critical heavy metal index value is 100 for drinking water.

3. Results

3.1. Physicochemical properties of water samples

3.1.1. pH, EC and TDS

The physicochemical parameters of the water samples collected from Bahawalpur district are shown in Table 2. pH, EC and TDS are the most basic parameters to identify the quality and usage of water. The guideline values for pH, EC and TDS for drinking water are 6.5 to 8.5, <400 µS cm^{-1} and <1000 mg L^{-1} respectively while recommended EC for irrigation water is <1150 µS cm^{-1}. The analyzed data showed that in the total 120 selected areas of Bahawalpur pH ranged from 6.81 to 8.42 while EC and TDS values were in the range of 144.8 to 5292.2 µS cm^{-1} and 226 to 8269 mg L^{-1} respectively. The frequency distribution of samples revealed that the pH of more than 80% samples were between 7 to 7.5, 14% between 7.5 to 8, 5% between 6.5 to 7 and nearly 1% between 8 to 8.5. On the other hand, EC of 40% samples were in the range of 100 to 500 µS cm^{-1}, 37% in the range of 500 to 1000 µS cm^{-1}, 9% in the range of 1000 to 1500 µS cm^{-1} and 11% above 1500 µS cm^{-1}. Similar trend was also observed in the values of TDS where 53% samples ranged between 200 to 1000 mg L^{-1}, 30% ranged between 1000 to 2000 mg L^{-1}, 8% ranged between 2000 to 3000 mg L^{-1} and nearly 8% samples were above 3000 mg L^{-1}. It can be seen from the Table 2 that a few sampling sites showed much more EC and TDS values as compared to average values of most of the samples. All such sites were observed to be far away from Sutlej River which indicates that groundwater in the vicinity of Sutlej River can be considered good quality but with the increase in the distance from the river water gradually becomes contaminated. Additionally, the availability of most of the water in this river in only restricted to monsoon season.

Table 2. Physicochemical parameters of groundwater samples collected from Bahawalpur district. All values in mg L^{-1} except EC in µS cm^{-1} and pH.

Parameters	pH	TDS	EC	Na^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	SO_4^{2-}	CO_3^-	HCO_3^-	NO_3^-
Mean	7.3	867	1356	210.6	6.9	50.1	30.9	188.0	135.7	0	359.2	3.59
Max	8.4	5292	8269	451.5	14.3	110.1	73.3	291.1	201.3	0	701.2	56.52
Min	6.8	144	226	66.5	0.9	16.3	8.1	83.7	46.1	0	151.1	0.01
SD	0.2	899	1406	84.1	3.0	21.3	12.6	50.07	32.81	0	102.9	6.6
Ideal v	7	0	0	0	0	0	0	0	0	0	0	0
CV	3.4	103	104	39.9	43.0	42.5	40.8	26.6	24.1	0	28.6	183.96
MOD	7.3	456	714	N/A	8.0	43.2	35.5	N/A	136.4	0	204.2	0.1
WHO	6.5-8.5	1000	400	250	12	75	50	250	250	-	500	10

Table 3. Heavy metal concentrations and drinking as well as irrigation water quality indices determined in the groundwaters of Bahawalpur. F and Fe are given in mg L^{-1}, As, Pb, Cr and Ni in µg L^{-1} while SAR and RSC in me L^{-1}. Remaining are index values.

Parameters	F	Fe	As	Pb	Cr	Ni	WQI	HPI	SAR	RSC
Mean	1.49	0.08	17	37	33	29	81	56	5.89	0.90
Max	9.80	2.06	100	120	56	90	442	162	16.00	3.30
Mini	0.15	0.00	0	2	5	4	20	6	2.00	0.01
SD	1.65	0.19	17	24	12	18	68	33	2.85	0.91
Ideal v	0.00	0.00	0	0	0	0				
CV	110.00	239.00	100	65	36	60	84	58	48.40	101.00
MOD	0.75	0.04	10	15	33	30	36	21	5.00	0.08
WHO	1.5	0.3	10	10	50	70				

3.1.2. Major cations

The groundwater Na^+ concentrations in Bahawalpur district were found within the range of 66.5 to 451.6 mg L^{-1} and according to the overall Na^+ distribution 53% samples showed less than 200 mg L^{-1}, 36% within the range of 200 to 300 mg L^{-1} and a few locations (<11%) were exceeding 300 mg L^{-1}. Comparatively K^+ concentrations in the groundwater samples were less than guideline value for drinking purpose except a few samples collected from selected sites of Bahawalpur. Almost 93% samples from Bahawalpur district were below 12 mg L^{-1} and 7% exceeding the line in terms of K^+ concentrations. The analysis of data showed that the Ca^{2+} concentration was uniformly distributed in all over the sampling sites ranging from 16.3 to 110.2 mg L^{-1} (13% samples observed to be high in Ca^{2+} content than 75 mgL^{-1} and 87% were containing less than 75 mg L^{-1}). The Ca^{2+} concentrations were not restricted to a specific area in all the selected locations but mixed concentrations (low and high) were observed in equal proportions. This might be because of equal distribution of calcareous soils all over the arid to semiarid region of southern Punjab. The data presented in Table 2 depicts that the distribution of Mg^{2+} ions was also coinciding with Ca^{2+} concentrations i.e., uniformly dispersed throughout the areas and the number of samples exceeding WHO guideline value were comparatively less (8%) in Bahawalpur.

3.1.3. Major anions

The Cl^- concentrations observed in the groundwater of Bahawalpur were in the range of 83.7 to 291.1 mg L^{-1} where total of 74 (61%) samples contained Cl^- concentrations below 200 mg L^{-1}, 28 (23%) were in the range of 200 to 250 mg L^{-1} and 18 (15%) were above 250 mg L^{-1}. Similar ranges (46.1 to 201.3 mg L^{-1}) of SO_4^{2-} concentrations have also been observed in the groundwater of Bahawalpur. The analyzed data reveals that > 90% of samples from Bahawalpur district were below 250 mg L^{-1} and in fact most of the values were even below 200 mg L^{-1} in terms of SO_4^{2-} content which indicates only a little contamination to such extent prescribed by WHO for SO_4^{2-} ion in drinking water (250 mg L^{-1}). In contrast HCO_3^- ions were present in higher concentrations in most of the areas selected for sampling. The concentrations were found to be varied among different locations but as a whole it ranged from 151.1 to 701.2 mg L^{-1}. Despite higher threshold level for drinking purpose more that 10% sampling sites were exceeding the 500 mg L^{-1} line recommended by WHO. The data arranged in the Table 2 depicts that most of the sampling locations were in the safe line in terms of NO_3^- contamination as the average concentration from maximum number of samples showed less than 10 mg L^{-1} concentrations except a few one from Bahawalpur district. As stated earlier almost 114 (95%) samples were below 10 mg L^{-1} and only 6 (5%) samples contained more that the said limit of NO_3^-. The fluoride concentrations in the case study were found in varying degrees ranging from 0.15 to 9.8 mg L^{-1} in the groundwater samples (Table 2). The determined concentrations revealed that there is a serious contamination of F^- in the groundwater of Bahawalpur as 29% of selected locations were exceeding 1.5 mg L^{-1} concentrations. Out of total 120 sampling sites 85 were in safe limit and 35 were in toxic range.

3.2. Sodium Adsorption Ratio (SAR) and Residual Sodium Carbonate (RSC)

Sodium adsorption ratio and residual sodium carbonate are two important parameters for quantifying the suitability of irrigation water for growing crops. Both the parameters are calculated by the mathematical interactions (equations 3, 4) of major cations (Na^+, Ca^{2+}, Mg^{2+}) and anions (CO_3^-, HCO_3^-) occurring in the water. Data regarding SAR and RCS of groundwater is represented in Figure 2 (a and b). On the basis of these ions SAR and RSC of most of the sampling sites were found fit for irrigation but still a significant number of sampling areas were showing greater SAR and RSC. On an average the SAR of nearly 65% samples from Bahawalpur district were in acceptable limit (<6 meq L^{-1}), while 12% sampling sites were found to be unfit for irrigation (>10 meq L^{-1}). Likewise, results were also found for RSC values which depicts that 64% water samples were falling in the category of fit irrigation water from Bahawalpur and the number of unfit water samples were 8%.

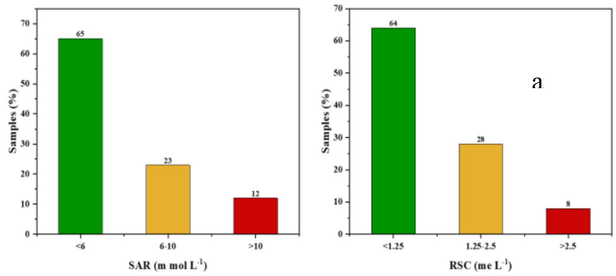


Figure 2. Distribution of SAR (a) and RSC (b) in selected areas of Bahawalpur. The graph bars in figure (a) are denoting percentage of samples (on the basis of SAR) falling in the category of fit (<6), marginal fit (6-10) and unfit (>10) for irrigation while graph bars in figure (b) are showing the percentage of samples (based on RSC) in the category of fit (<1.25), marginal fit (1.25-2.5) and unfit (>2.5) irrigation water.

3.3. Heavy Metal Pollution Index (HPI)

Heavy metal pollution index defines the overall contamination of water with a number of heavy metals including As, Pb, Cr, Ni and Fe etc. The classification of HPI of water includes five categories: <25 excellent, 25-50 good, 51-70 poor, 71-100 very poor and >100 hazardous water. In the case study total of 18% samples were found excellent and 13% hazardous in terms of HPI. Moreover 36% samples were in the category of good and 11% in the very poor water (Figure 3).

According to overall heavy metal occurrence in groundwater samples in selected sites iron ranged from 0 to 2.06 mg L^{-1}. Out of 120 sampling sites of Bahawalpur only 2 sites showed Fe concentration more than 0.3 mg L^{-1} while all other sites were below this limit. The data shown in the Table 3 indicates the As contamination in more than half of the sampling areas especially those areas which are comes under the effect of industrial units. High As contamination was also found to be linked with soil minerals such as Fe, Al and Mn oxyhydroxides which release As in groundwater due to changing soil pH and microbial actions. The overall range of As in groundwater was 0 to 100 µg L^{-1} where 47.5% samples was found to be none contaminated (<10 µg L^{-1}) while 52.5% samples were having As more than 10 µg L^{-1}. Data shown in the Table 3 shows that Pb concentration in most of the sampling areas was more than 10 µg L^{-1} and even greater than 50 and 100 µg L^{-1} in some of the samples. In the groundwater samples of Bahawalpur district it ranged from 2.5 to 120 µg L^{-1} and more than 100 (87%) samples were exceeding 10 µg L^{-1}, 5 out of them had Pb beyond 100 µg L^{-1}. The analyzed data for Cr concentrations reveals that there was only a certain number of samples showing more than 50 µg L^{-1} Cr in water samples collected from selected district (ranged from 5 to 56 µg L^{-1}). Like other heavy metals the groundwater contamination with Ni was also reported from many of the sampling areas in the selected district. On an average it was found between 4 to 90 µg L^{-1} in district Bahawalpur and out of 120 sampling sites nearly 5% were showing Ni concentration <70 µg L^{-1}. It is important to mention that similar to Cr, Ni concentrations in maximum number of samples (54%) were in the range of 20 to 50 µg L^{-1}.

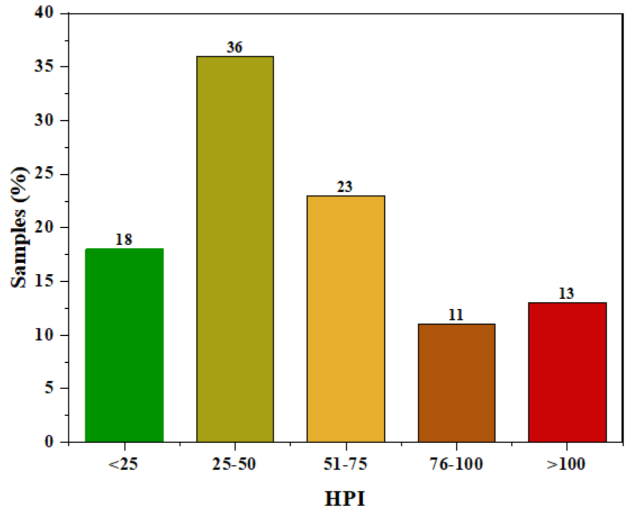


Figure 3. Range of HPI in the groundwater of Bahawalpur. Bars in the graphs are showing percentage of samples in the categories of excellent quality water (<25), good quality water (25-50), poor quality water (51-75), very poor quality water (76-100) and hazardous water (>100) on the basis of HPI.

3.4. *Major water type in aquifers of Bahawalpur*

All the major water types determined in selected area of Bahawalpur district can be classified as sodium-potassium/sodium-bicarbonate type (Figure 4a) except a few samples having sodium-chloride type. As shown in the figure dominant major cation is Na and the remaining cations are in the sequence of $K^+ > Ca^{2+} > Mg^{2+}$. While HCO_3^- is found to be major anion and rest of the anions are in the sequence of $Cl^- > SO_4^{2-} > CO_3^-$. Some of the samples (<20%) appearing in the middle of diamond and lower triangles of Piper diagram are indicating no dominant type of water (mixed water type) in the Bahawalpur district. Moreover, none of the sample was observed to be SO_4^{2-} or Mg^{2+} type because of lower distribution of these ions. Above data also indicates a single aquifer source in most of the sampling area. The results plotted on the Durov diagram shows that more than 75% of the samples appear along the dissolving or mixing line of the Durov plot (Figure 4b), supporting the idea that a mixed variety of water types predominate in the research area. Fresh recharge water with simple dissolving or mixing and no dominating large anion or cation is responsible for this tendency. Furthermore, a small percentage of samples (~13%) with Cl^- and Na^+ as the predominant anion/cation suggested a connection between the ground waters and the reverse ion exchange of Na-Cl waters.

The functional dissolution sources of ions were also determined by plotting Gibbs diagram (Figure 4c) of variability ratios of Na/(Na + Ca) and Cl/(Cl + HCO_3) as a function of TDS. A clear interaction can be observed from the plot between rock and groundwater chemistry and more specifically the major ions concentrations in the groundwater mainly resulted from rock-water interaction (weathering of rocks, formation of minerals and precipitation of carbonates) and evaporative sedimentation. The moderate TDS values and ions concentration is indicating dominancy of rock-water interaction as well as evaporative sedimentation. On the other hand, precipitation is not influencing the ion chemistry i.e., low precipitation causes high ion concentrations and moderate to low TDS, opposite is true for high precipitation.

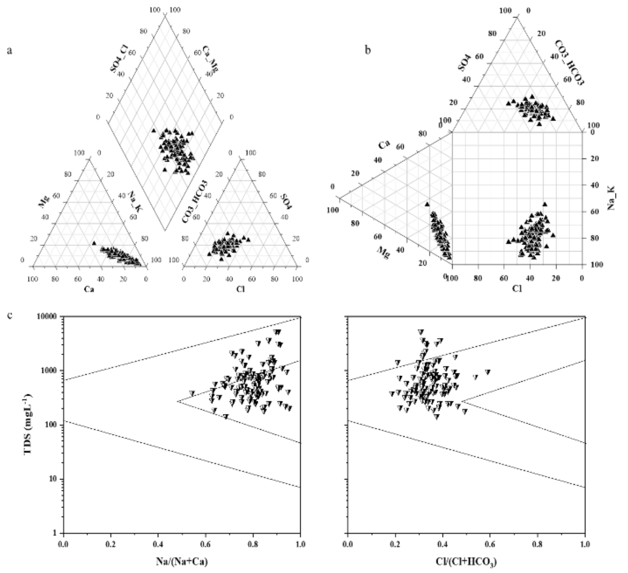


Figure 4. Piper plot in figure (a) is representing major water types while Durov diagram (b) is indicating major processes involved in the dissolution of major ions in the water systems in Bahawalpur. Occurring of clusters towards centers and Na-k to HCO_3 lines in Piper plots indicating the dominancy of sodium bicarbonate type water and mixed water type. Similarly points appearing near the middle line in Durov diagram reveals simple dissolution or mixing is the main process in groundwater chemistry. Gibbs diagram constructed between TDS and major cations and anions found in the groundwater of Bahawalpur is shown in figure (c). The points in the diagrams are showing that rock-water interactions and evaporative sedimentation are influencing the groundwater composition.

3.5. *Water Quality Index (WQI)*

Water quality index can be considered as a complete set of water quality which includes all the physicochemical and heavy metal constituents. Similar to HPI the classification of WQI also include five categories: <25 excellent, 25-50 good, 51-70 poor, 71-100 very poor and >100 hazardous water. WQI of selected site of Bahawalpur is shown in the Figure 5 and Table 3. Out of total 120 sampling sites in Bahawalpur the number of excellent and hazardous water samples were 3 (~2%) and 22 (18%) respectively. Moreover, the number of samples in categories of good, poor and very poor quality water were 44 (36%), 35 (29%) and 16 (13%) respectively.

3.6. *Correlation and cluster analysis*

The correlation matrix of physicochemical characteristics and heavy metals of water samples collected from Bahawalpur is given in Table 4. A number of parameters determined in the water samples can be seen having a positive correlation between each other like EC and TDS (r = 1), Ca^{2+} and HCO_3^- (r = 0.85), Mg^{2+} and HCO_3^- (r = 0.84), Ca^{2+} and Mg^{2+} (r = 0.80), Cl^- and SO_4^{2-} (r = 0.65), Cl^- and EC (r = 0.64), Na^+ and TDS (r = 0.61), Ca^{2+} and SO_4^{2-} (r = 0.52).

In addition to correlation matrix a dendrogram of cluster analysis was also constructed to further verify the grouping and correlation of different physicochemical and heavy metal constituents (Figure 6). Which generated four groups of sites having similar characteristics and same source of contamination in Bahawalpur area, group (G1) includes pH, F^-, Na^+ and K^+, group (G2) includes NO_3^-, Fe, Cr and Ni, group (G3) includes As and Pb and the group (G4) includes TDS, EC, Cl^-, SO_4^{2-}, Ca^{2+}, Mg^{2+} and HCO_3^-.

Table 4. Correlation matrix of all the physicochemical and heavy metal constituents in water samples (n=120) of Bahawalpur area. Highlighted values are indicating significance level at $P \leq 0.05$.

	pH	EC	TDS	Na^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	SO_4^{2-}	HCO_3^-	NO_3^-	F^-	Fe	As	Pb	Cr	Ni
pH	1.00																
EC	-0.05	1.00															
TDS	-0.05	0.99	1.00														
Na^+	-0.08	0.61	0.61	1.00													
K^+	-0.28	0.28	0.28	0.22	1.00												
Ca^{2+}	0.01	0.58	0.58	0.16	0.20	1.00											
Mg^{2+}	-0.11	0.58	0.58	0.17	0.26	0.81	1.00										
Cl^-	-0.24	0.63	0.63	0.28	0.39	0.51	0.56	1.00									
SO_4^{2-}	-0.26	0.61	0.61	0.15	0.38	0.52	0.50	0.65	1.00								
HCO_3^-	-0.08	0.56	0.56	0.16	0.18	0.85	0.85	0.50	0.50	1.00							
NO_3^-	-0.15	0.24	0.24	0.18	0.17	0.25	0.37	0.25	0.24	0.29	1.00						
F^-	0.39	0.10	0.10	-0.01	0.05	0.14	0.20	0.10	0.13	0.16	0.15	1.00					
Fe	-0.01	0.17	0.17	-0.11	0.01	0.10	0.11	0.13	0.17	0.08	-0.03	0.11	1.00				
As	0.05	-0.02	-0.02	-0.02	0.05	-0.09	-0.03	0.01	0.02	0.01	-0.10	0.03	-0.01	1.00			
Pb	-0.04	0.11	0.11	0.11	0.04	0.12	0.18	0.06	0.10	0.24	0.03	-0.08	0.04	0.22	1.00		
Cr	-0.04	-0.11	-0.11	0.15	-0.04	-0.08	-0.09	0.01	-0.17	-0.10	0.06	-0.09	0.03	-0.16	-0.06	1.00	
Ni	0.12	0.02	0.02	-0.04	-0.08	0.07	0.09	0.03	-0.11	0.09	0.05	-0.07	-0.03	-0.01	0.02	0.10	1.00

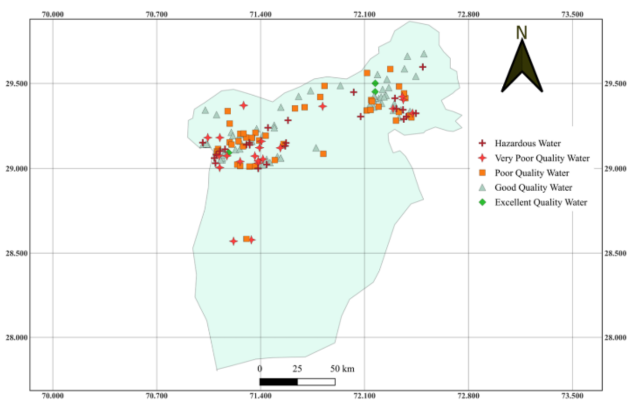


Figure 5. Spatial geographical distribution of WQI in the selected areas of Bahawalpur.

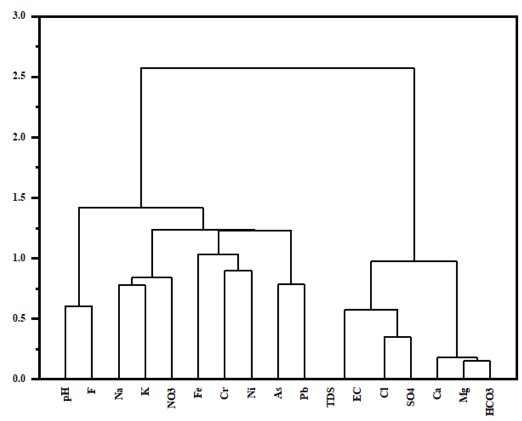


Figure 6. Dendrogram of cluster analysis (CA) of water samples collected from Bahawalpur constructed using Ward's method. The figure is indicating three distinct groups (parameters correlate within the group but differ from other group) of alike water samples from each site.

3.7. Principle Component Analysis (PCA)

The scores plots using principle component analysis were also generated between factor 1 (PC1) and 2 (PC2) to evaluate the enrichment of physicochemical parameters and heavy metals of water samples collected from the selected district. The scores plots obtained from water samples of Bahawalpur (Figure 7) demonstrated little variation in the relative dominance of main ions and other water quality criteria, such as trace metals. Groundwater samples were found to be substantially concentrated in main ions and metal contamination, as indicated by the score plots in the right side quadrants, which can be either lower or higher. However, pH and Cr were more prevalent in the distribution of sample in the left quadrants, either upper or lower.

4. Discussion

Groundwater quality assessment is a crucial process to evaluate the physical, chemical and biological characteristics of groundwater to ensure its fitness for numerous uses, such as drinking water, agriculture, industrial processes and environmental conservation. The analysis of water samples revealed that the ground water pH of water samples were slightly alkaline but values of most samples were within permissible limit for drinking water rendering to WHO defined range (6.5-8.5). The presence of HCO_3^- in the study area is responsible for alkaline pH of water which released from the weathering of carbonaceous rocks (López-Pazos *et al*. 2010). Parallel to pH, EC is also a significant parameter for the characterization of groundwater (Ouarekh *et al*. 2021). The EC of >90% water samples showed elevated values owing to the presence of dissolved salts in the water (Aamer and Sabir, 2014). The increasing EC also resulted in an increase of TDS because of ion exchange between groundwater and soil minerals (Baig *et al*. 2010).

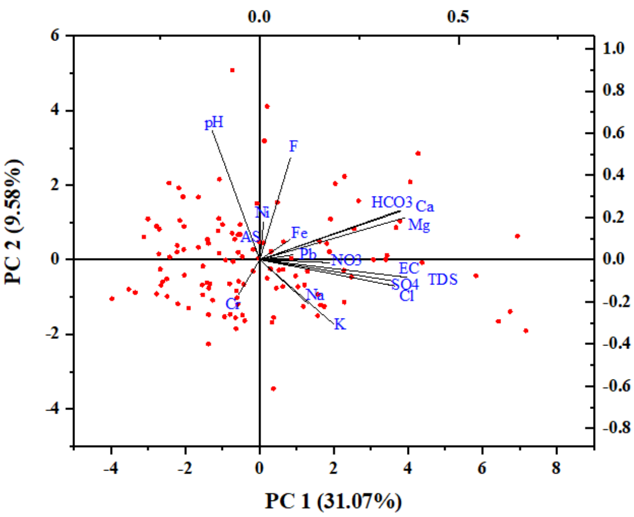


Figure 7. Score plot indicating the enrichment of different parameters found in the ground water of Bahawalpur district.

The majority of the contaminated water samples were unfit for human consumption because their TDS levels were beyond the WHO's recommended threshold of 1000 mg L^{-1} (WHO, 1993). According to Abbas *et al*. (2014), there is mounting evidence that water EC rises with residency duration and increased rock-water contact. Results of pH, EC and TDS found in line with those described by Alam *et al*. (2021).

Hydrochemical parameters showed that Na^+, K^+, Ca^{2+} and Mg^{2+} were major cations that were determined in the collected water samples. The main source of the higher Ca^{2+} and Na^+ levels in groundwater was the infiltration of soluble soil salts from irrigation or precipitation that contaminated surface water. High levels of Ca^{2+} and Na^+ can also result from sewage effluent contamination of drinking water (Abbas *et al*. 2014). On the other hand, Ca^{2+} and Na^+ are released on the weathering of silicate minerals, which may accumulate these types of ions in groundwater by water-rock interactions, leading to greater concentrations of Ca^{2+} and Na^+ in groundwater samples (Ramkumar *et al*. 2010). The overall groundwater K^+ concentrations were comparatively low in the water samples because K^+ have a tendency to be locked on the surface of clay minerals and disintegrates at a slower pace than Na^+ minerals (Mishra, 2020). The concentrations of Mg^{2+} were slightly above the permissible limits in a few samples collected from selected district. High concentrations of Mg^{2+} were caused due to diffusion of leachate from industrial, household and landfills sites (Abbas *et al*. 2014). Mehmood *et al*. (2012) conducted a selective study on the groundwater in parts of Bahawalpur city where they found similar trends as found in the current study for the heavy metal contamination as well as physicochemical parameters especially Cl^-, SO_4^{2-}, Ca^{2+} and Mg^{2+}.

The occurrence of anions was also observed in the similar pattern that of cations throughout the sampling sites. Out of all the anions HCO_3^- concentrations were in higher levels followed by Cl^-, SO_4^{2-}, NO_3^- and F^- because of the growing population and the significant increase in agricultural and residential waste in the examined area (Rasool *et al*. 2016). It is possible that the higher HCO_3^- concentrations were caused by local inputs such as fertilizers and household waste materials. The weathering of carbonaceous rocks is another possible reason behind elevated levels of HCO_3^- in the studied areas (Li *et al*. 2019). Nitrate is another anion dominating in the groundwater of selected sites especially in countryside areas of district Bahawalpur which primarily may occurred due to agricultural activities, decay of dead plants, sewage discharges and feces (Akhtar *et al*. 2021). The use of synthetic fertilizers such as DAP and urea for intensive agriculture is another source of contribution in soil and groundwater contamination with NO_3^-. FAO (2004) reports that during the past 30 years, Pakistan has grown its fertilizer usage, with Punjab Province accounting for the majority of this growth due to its enormous agricultural area.

The analysis of data also represented relatively higher to moderate levels of Cl^-, SO_4^{2-} and F^- contamination in samples. The possible reasons behind increased levels of Cl^- and SO_4^{2-} in groundwater are thought to be anthropogenic activities such as municipal seepage and sewage and other biowaste materials contributed in groundwater pollution. Additionally, the sampling sites come under the low lands of arid to semi-arid region where water naturally flows from North to South West, thus adding more concentrations of Cl^- and SO_4^{2-} in groundwater. When paired with Na^+ or Mg^{2+}, SO_4^{2-} overload can be corrosive and laxative, imparting an unpalatable taste (Ashraf and Foolad 2007). Groundwater contamination with SO_4^{2-} has been linked to anthropogenic causes, such as detergent use and seepage from city areas (Arshad and Umar 2022). Groundwater fluoride content rises as a result of prolonged water-rock interaction brought on by the low groundwater recharge sources such as rainfall in arid climate. Conversely, elevated evaporation leads to the precipitation of less soluble minerals ($CaCO_3$), hence diminishing the calcium ion availability in groundwater and facilitating the dissolving of fluoride minerals (Vithanage and Bhattacharya 2015). The presence of moderate to high concentrations of these cations and anions are responsible for high SAR and RSC values in few sampling locations of the selected areas. While most of the sampling locations showed SAR and RSC values in acceptable range which are in line with Mohsin *et al*. (2019).

By using the data attained from above mentioned parameters a Piper plot, Durov and Gibbs diagrams were designated to study major water types, sources of contamination and its interaction with external factors like precipitation and evaporation respectively. The Gibbs plot of data from the study area (Figure 4c) indicated the interaction between rock-percolation waters chemistry under subsurface. These plots revealed that weathering has a greater impact on the local groundwater quality just like evaporation. This implies that water-rock interactions cause concentrations of the main ions Na^+, Ca^{2+} and Mg^{2+} in water to rise. The main processes involved in water-rock interactions are ion exchange between clay minerals and water, chemical weathering of rock-forming minerals and dissolution-precipitation of secondary carbonates (Moghaddam and Fijani, 2008). Low rainfall (225-400 mm annually), considerable evaporation (>2000 mm annually) and low hydraulic conductivity of groundwater are characteristics of dry and semiarid climatic zones (Su *et al*. 2020, Khan *et al*. 2013a,b).

The spatial distribution of trace metals can be used to distinguish between zones with varying metal concentrations and to assess potential enrichment sources (Wang *et al*. 2020). Spatial distribution of elevated concentrations of As in the current study was mainly confined in the sampling locations chosen from urban areas especially near the industrial sites. High As contamination was also found to be linked with soil minerals such as Fe, Al and Mn oxyhydroxides which release As in groundwater due to changing soil pH and microbial actions. Transportation, intensive use of agricultural insecticides and leaching of weathered mafic and ultramafic rocks are the reasons behind increasing level of Pb in groundwater samples. The occurrence and distribution of moderate to lower concentrations of Fe and Ni were also attributed to the river inflows (Zhang *et al*. 2022). High concentrations of heavy metals are commonly summarized as heavy metal pollution index (HPI) which depends on the overall concentrations of the metal ions. Additionally, the increased use of pesticides and fertilizers in Punjab province is a key source of As, Pb, Ni, Cd, Cr and Fe in drinking water of the study area (NFDC 2016). The body of research makes clear that excessive use of agrochemicals, such as fertilizers and pesticides can result in high quantities of metals which can contaminate groundwater and have negative health effects on people (Singh 2011).

5. Conclusion

In conclusion majority of the analyzed groundwater samples are acceptable and suitable for drinking as well as irrigation purposes. The distribution of the major cations and anions showed that they are below the guideline/permissible limits and the predominant ions are Na^+, Ca^{2+}, HCO_3^-, Cl^- and SO_4^{2-}. It was observed that more than 50% of the groundwater samples are $Na\text{-}HCO_3$ water, less than 10% are $Ca\text{-}SO_4$ and $Mg\text{-}SO_4$ water while 30% samples showed mixed water type. On the basis of these ions SAR and RSC of most of the sampling sites are found fit for irrigation but still a significant number of sampling areas are showing greater SAR and RSC. The SAR and RSC of more than half of the samples (65 and 64% respectively) collected from Bahawalpur are in acceptable limit (<6 meq L^{-1}), while 12% and 8% samples are found to be unfit for irrigation (>10 meq L^{-1}) respectively. The heavy metal distribution in the groundwater samples collected from the study area is in the order Fe > Pb > *Cr*> Ni > *As*. The WQI of all the water samples showed that out of total 120 sampling sites in Bahawalpur the number of excellent water samples are only 2% while 18% samples are in the category of hazardous water samples. The remaining water samples shows WQI in the range of good (36%), poor (29%) and very poor quality (13%) water. Therefore, this research suggests the treatment of contaminated groundwater before human consumption. Also, the use of groundwater for irrigation purposes should be considered according to the level of groundwater contamination and soil quality.

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