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Evaluation of toxic heavy metal concentration in aquifer system for groundwater system development using multivariate statistical techniques

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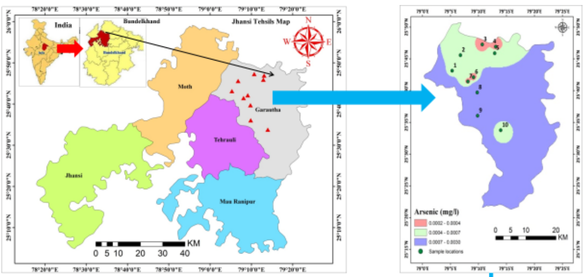
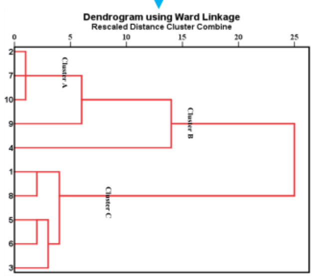
Graphical abstract



Heavy metals	Principal component analysis		
	1	2	3
Al	-0.241	**0.765**	0.384
As	0.078	0.325	**0.843**
Cd	***-0.611***	-0.344	-0.239
Co	***-0.590***	-0.262	0.343
Cr	-0.226	***-0.906***	0.136
Cu	**0.908**	-0.100	-0.283
Fe	**0.725**	0.030	-0.061
Mn	0.202	**0.960**	-0.052
Ni	-0.171	-0.216	**0.838**
Pb	**0.880**	0.166	-0.178
Zn	**0.876**	-0.018	0.376
Initial Eigenvalue	4.175	2.566	1.774
Variance %	37.954	23.331	16.132
Cumulative %	37.954	61.285	77.416



Abstract

Groundwater is a vital resource for human consumption. The study aimed to evaluate toxic metal concentrations in groundwater systems and determine pollutant sources using multivariate methods including cluster analysis (CA), principal factor analysis (PCA), and Pearson correlation coefficient (r). The results were compared with World Health Organization (WHO 2017) and Bureau of Indian Standard (BIS 2012) standards, indicating that Al concentration observed within prescribed values and other Cd, As, Zn, Pb, and Cu were less than the acceptable values, as well as the rest of Fe, Mn, and Ni levels in groundwater were mostly within acceptable values. The PCA results showed three factors (F1, F2, and F3) were responsible for the data structure, which was specified as 37.954%, 23.331%, and 16.132%, as well as total variance of dataset associated with 77.416%, respectively. Factor 1 showed strong positive loading (Cu, Pb, Zn), 2 (Al, Mn), and 3 (As, Ni), which demonstrated the contaminants source from natural and agricultural activities. Moreover, CA results revealed three clusters indicating low to high water pollution due to rock weathering and anthropogenic activities. Overall, results showed that 50% of groundwater samples were acceptable for potable and agricultural uses. Therefore, groundwater treatment is necessary before any use.

Keywords: Heavy metal concentrations, groundwater pollution, cluster analysis, principal factor analysis, and Pearson correlation coefficient

1. Introduction

Groundwater is an essential resource for all humanity as it is crucial for the existence of individuals; however, for over one billion citizens globally there is no secure exposure to drinking water, irrigation and industrial products (Carrard *et al.* 2019; Qadir *et al.* 2019a; Mukhtar *et al.* 2022). Approximately one-third of the global water usage is dependent on groundwater that allows 20% industrial water, 40% agriculture water, and 50% drinking water accessible (Hao *et al.* 2018; Akhtar *et al.* 2021). Groundwater extracts quickly and worldwide from the shallow aquifer system, and the extraction rates are 982 km^3/year. Late, the world's largest focus on the resolving of water quality problems, particularly in urban and developing countries, was gradually increased because the lack of access to clean waterways undermines the most significant right to existence (Regan *et al.* 2017). The progresses of urbanization, population growth, increase industrial applications, and agricultural fertilizers have also contributed to a quantitative decrease in groundwater quality over the last four decades (Krishna *et al.* 2015). A lot of the usages of the hazardous metals are used in the agriculture fertilizers and industrial production that change the biological, chemical, and physical characteristics of groundwater as well as affect the human health (Hari 2019). Furthermore, there is a natural cause of water contamination such as the interaction between the polluted surface water and groundwater, the interface of rocks and water, oxidation-reduction reactions, and weathering of minerals or rocks which affect the quality, quantity, and availability of groundwater (Zhou *et al.* 2019). Recent estimates show that approximately 80 percent of the world's population from around all over the world is at risk of harmful conditions caused by groundwater pollution (Akhtar *et al.* 2018; Ntanganedzeni *et al.* 2018).

A multiple of hazardous toxic pollutants and other dangerous chemicals emitted from distinct anthropogenic activities into the surface water resource, in particular in rivers and streams, thus polluted groundwater can lead to a serious health issue for humans (Alaya *et al.* 2014). For instances, high lead in water may lead to different diseases, including bone fragility, failure of the development of enamel, fluorosis, stiffness, and lameness of gait and molting of teeth, etc (Narsimha and Rajitha 2018). In addition, chromium often decreases cholesterol and fatty acids and regulates the amount of blood sugar and glucose, however, high levels in water may affect lung cancer, damage to the liver, blood pressure, tissue destruction, and high blood pressure (Dkhar *et al.* 2014; Akhtar and Rai 2019a). Furthermore, high zinc amounts may lead to diagnostic difficulties and have an effect on blood and the nervous system (Hafiza *et al.* 2015) and large amounts of arsenic may lead to serious confusion, lung cancer, diarrhea, headaches, sleepiness, and kidney cancer (Davenport 2014). Moreover, the previous study reported that cadmium has been highly toxic to the health of humans that it had been first reported to have chronically poisoned diseases in Japan such as itai-itai in the 20th century, and also causing a large number of infections including osteomalacia, osteoporosis, and renal tubular dysfunction, etc (Aoshima 2016; Kubier *et al.* 2019).

The previous analysis has shown that in some parts of the Jhansi district, groundwater pollution was recorded with Fe (329 - 944), Ni (0.3 – 85.7), Pb (39 - 531) and Mn (10 - 1342), µg/L which was due to the natural sources and anthropogenic activities (Akhtar *et al.* 2018, 2020). The pollution was increasing in the groundwater due to caused by human activity and natural sources in this district; therefore, it was important to investigate the groundwater quality of a study in this area. Furthermore, almost 80 percent of people of the Garautha Tehsil use the seasonal stream water and groundwater for domestic and agriculture purposes (Akhtar *et al.* 2018). Because the irrigation canals are accessible seasonally, the people of Garautha are dependent mainly on groundwater resources (Singh *et al.* 2013). Drought is also usually reported due to very high temperatures during the summer season. Most of the people of the study area, the municipals wastewater, and hazardous waste dumped into canals or drains then it percolates into the groundwater. In addition, people use pesticides and fertilizers for agriculture purposes in this Tehsil (Akhtar and Rai 2019b). Therefore, due to these human activities, it can change the physical and chemical properties of groundwater and it can cause several problems related to human diseases in this area.

The aim of this study is to evaluate the heavy metal level and the quality of the groundwater according to the permissible levels. Therefore, (1) to identify and investigate the heavy metals concentrations in groundwater by using (ICP-MS) (2) to evaluate the source of groundwater by using multivariate statistical techniques. This research paper will help the developers and the government agencies to supply households and farmers with the quality of water for their portable and agriculture purposes.

2. Materials and methods

2.1. Study area

Garautha Tehsil is located in the Jhansi district, India (Figure 1). It lies from 78.152° to 79.305° in the eastern part and 28.505° to 25.975° in the north part of district.

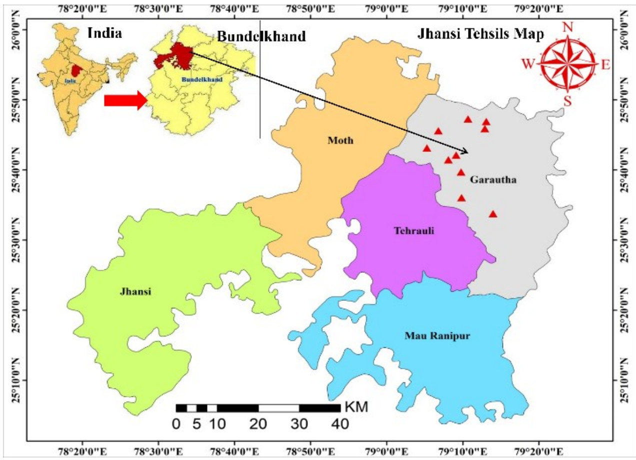


Figure 1. This is showed the Garautha Tehsil map and sampling of groundwater

According to the 2011 census, there are 59 villages in Tehsil. The drainage system lies in the Yamuna Sub-basin which is the part of Ganga River In this district. The main river of this Tehsil is Dhasan which is ephemeral in nature and there are few tributaries such as Lakheri, Kureri, and Sukhai and these tributaries flow from north to the northeast (Akhtar and Rai 2019b). The elevation ranges from about 150m over the ocean altitude to the north to about 165m to the south (CGWB 2017). The annual precipitation is approximately 840mm; with around 90% of the overall monsoon precipitation occurs in the month of September, July, and August. The temperature is sub-humid with the cool months from Nov-Jan in the winter seasons and Mar-Jul is the hot months in the warmer seasons while January is the coldest month about (24.10 °C) and May is the hot month about (42.6 °C). The irrigation system of this Tehsil contains 10% of lakes, 55% of canals and only 35% groundwater (CGWB 2017). The drawing map was prepared using the ASTER DEM (Qadir *et al.* 2019b). The topographical drainage maps were usually matching with those prepared by means of the ASTEM DEM data from 1965 to 1975 (Figure 2).

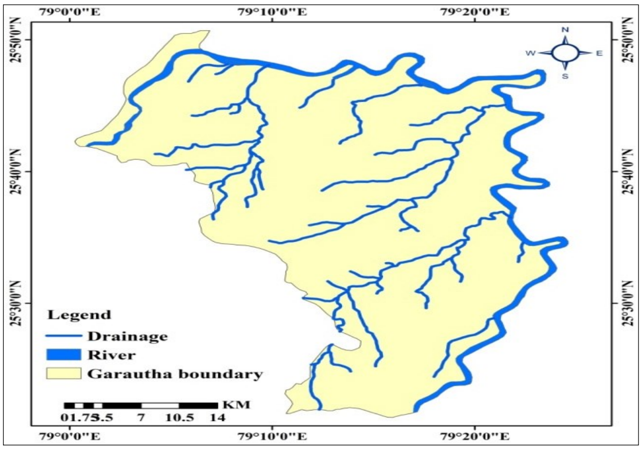


Figure 2. The diagram illustrated the drainage map of Garautha Tehsil (Akhtar *et al.* 2020)

2.2. Hydrology and geology

Garautha Tehsil is a granitic rock area with limited subsurface water system. According to report of (CGWB 2017), there are two divisions of the Jhansi district on the basis of geological formations: (1) the southern part of this area comprises granitic terrain (Bundelkhand granite) and Bundelkhand Gneissic Complex (BGC) and (2) the northern part is highly eroding composite alluvium or plain of Quaternary age (Figure 3). Therefore, the geological formations throughout this Tehsil are gneisses rocks from Precambrian to recently (BGC) in origin as well as granitic rocks. Moreover, the southern province comprises the weathered zone (average 20-40m thickness) of the BGC and the overlying residual soils largely form a various aquifer system (Akhtar *et al.* 2020).

In addition, the northern part consists of alluvium consists of pebbles, gravels, coarse sands, silt, clay and kankar with a maximum thickness of 60 m. In the certain part of this belong lies with consolidated and unconsolidated formations. The alluvium together with the Granite-Gneissic basement's underlying weathered zone forms a more or less homogeneous aquifer system. The alluvium soils in this area have been divided between black (lowland) soil and red soil (upland) and four subgroups based on color and topography. Black soils are further classified into mar and kabar and red soils are categorized as parwa and rakar (Akhtar and Rai 2019b). Such soils are shallow, gravely and highly porous, with a low content of organic material and low water capacity. The deep aquifers, typically recharged from natural runoff, are home to most groundwater. During several monsoon periods, variations in water levels are reported. The lowest level of groundwater in the northern part is around 20 mbgl. The table for groundwater ranges in general between 5 and 20 mbgl (CGWB 2017). The maximum well discharge was of approximately 46 litre/second for the alluvial aquifer. The dense, possible alluvial aquifers have been found to be confined to the eastern part of the Garautha Tehsil (Akhtar and Rai 2019b).

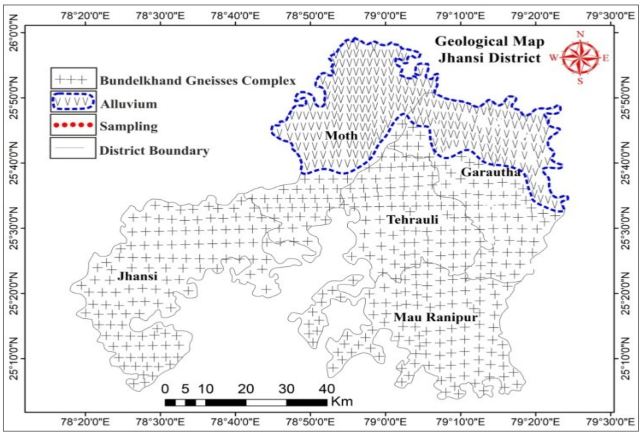


Figure 3. The diagram illustrated the geological map of study area (Akhtar *et al.* 2020)

2.3. Water sampling

A total of 10 groundwater samples were obtained from the hand pumps to analyze the concentration of heavy metals in groundwater. The groundwater samples were performed in acid-leached polyethylene bottles and collected from each location, which was thoroughly washed with water for analysis by adding ultrapure nitric acid (5ml / L). Perkin-Elmer Inductively coupled plasma-mass spectrometer (ICP-MS) was used for the study of groundwater samples. In order to classify the concentration of heavy metals to groundwater samples, a 10 ppb, 50 ppb, and 100 ppb combined heavy metal solution has been used to evaluate the calibration curve. The groundwater samples were absorbed in peroxide hydrogen and nitric acid with the help of the Anton Paar Multi-Wave PRO Microwave Response System for the removal/oxidation of organic materials and passed through the 0.45-micron filtering paper before (ICP-MS) analysis. Heavy metals as Al, Cd, Cr, Fe, As, Co, Pb, Cu, Mn, Zn, and Ni were examined and findings were compared to the limits of Bureau Indian Standards (BIS 2012) and the World Health Organization (WHO 2017) standards.

2.4. Multivariate statistical methods

Quantitative and independent assessment and interpretation of time and space variation in the quality of groundwater measurements, source of groundwater contamination by human activities (irrigation and wastewater), and natural processes, as well as geological and hydrologically active processes, is an important technique known as a multivariate statistical method (Bhuiyan *et al.* 2011; Brahman *et al.* 2013; Edokpayi *et al.* 2018). This approach is used to determine groundwater quality and source of groundwater pollution without losing useful information and tremendous economic development and the analysis was conducted using SPSS version 21 statistical software (Machiwal and Jha 2015; Bodrud-Doza *et al.* 2016). For this research, three multivariate approaches were used because of their importance for studies linked to groundwater such as the Cluster analysis (CA), the Pearson correlation coefficient (r), and the cluster analysis (CA).

3. Results and discussion

3.1. Heavy metal distribution in groundwater

In descriptive statistics which include the maximum, minimum, and average values of each metal across through standard deviation are shown the concentration of heavy metals in the analyzed groundwater samples, compared to drinking water requirements by (BIS 2012; WHO 2017), is listed in Table 1. The measurement of heavy metal in the groundwater samples is in the order of mean value such as Fe (0.832 mg/L) > Al (0.124 mg/L) > Zn (0.080 mg/L) > Mn (0.051 mg/L) > Cr (0.039 mg/L) >Ni (0.024 mg/L) > Cu (0.008 mg/L) > Pb (0.004 mg/L) > As (0.001 mg/L) > Cd (0.001 mg/L) > Co (0.001 mg/L). The concentration of Al (0.062 – 0.244 mg/L) in all groundwater was within the acceptable limit. The concentration of As (0.003 – 0.0002 mg/L), Cd (0.003 – 0.0002 mg/L), Cu (0.057 – 0.011 mg/L), Pb (0.007 – 0.001 mg/L) and Zn (0.154 – 0.019 mg/L) in all groundwater samples were lower than the acceptable limit by (BIS 2012) and (WHO 2017) and also the values of Co (0.014 – 0.003 mg/L) were than the permissible limit according to the (ATSDR 2004) (ATSDR 2004).

Table 1. The Table illustrated the drainage map of Garautha Tehsil

Heavy Metal	Min	Max	Mean	Std. Dev	WHO 2017	BIS 2012
Al	0.062	0.245	0.124	0.062	0.1	0.03 - 0.2
As	0.000	0.003	0.001	0.001	0.01	0.01 - 0.05
Cd	0.000	0.003	0.001	0.001	0.003	0.003 - NR
Co	0.000	0.001	0.001	0.000	-	-
Cr	0.012	0.057	0.039	0.017	0.05	0.05 - NR
Cu	0.003	0.015	0.008	0.004	2	0.02 - 1.5
Fe	0.459	1.559	0.832	0.332	0.3	0.3 – 1.0
Mn	0.011	0.122	0.051	0.039	0.1	0.1 - 0.3
Ni	0.015	0.028	0.024	0.004	0.07	0.02 - NR
Pb	0.002	0.008	0.004	0.002	0.01	0.01 - NR
Zn	0.019	0.154	0.080	0.050	3	5 - 15

Note: NR means not required

The concentration of Cr (0.057 - 0.011 mg/L) was observed that 5 samples within the permissible limit and 5 water samples were recorded less than the permissible value. Fe concentration (0.459 – 1.558 mg/L) was found to be 8 samples above the acceptable level and 2 samples within the permissible limit. The concentration of Mn (0.122 – 0.011 mg / L) was found to be below the acceptable limit of one sample and 9 samples were below the acceptable limit. Furthermore, Ni (0.007 – 0.001 mg / L) concentration was found to be 9 samples within the allowable limit and 1 sample below the acceptable range. The concentration of all heavy metal in the groundwater of the study area has been shown (Figures 4 and 5).

3.2. Source identification in groundwater system

The heavy metal concentration in PCA has been performed in this analysis for 11 heavy metals concentration from 10 sampling sites. The PCA findings demonstrated that the data for groundwater quality sets based more than 1 eigenvalue (which defines 77,416% of the total variance) and three main factors were extracted which is much stronger and contingent on identifying the major heavy metal distribution sources. The percentage of variance of 37.954 %, 23.331 % and 16.132 % respectively illustrate the total variance from one to three factors (Table 2).

Factor 1 demonstrated that the total variance of 37.954 % dataset and it suggests Cu, Pb, and Zn have a strong positive loading and Fe denotes the moderate loading while Cd and Co indicate the negative loading. This factor was associated with anthropogenic sources because the local inhabitants is used the fertilizers and pesticides for irrigation purposes. Exposure to agricultural practices, such as fertilizer usage and pesticides reported based on the previous study which is influences the Cu, Pb, Mn, and Zn (Deng *et al.* 2020).

Factor 2 indicates that 23.331 % of the total variance and denotes Al and Mn showed the strong loading and Cr stated the inverse loading. The main source of Al and Mn is generally leaching from the bedrock or minerals; therefore, it indicated that these metals originated from the natural source or geogenic source.

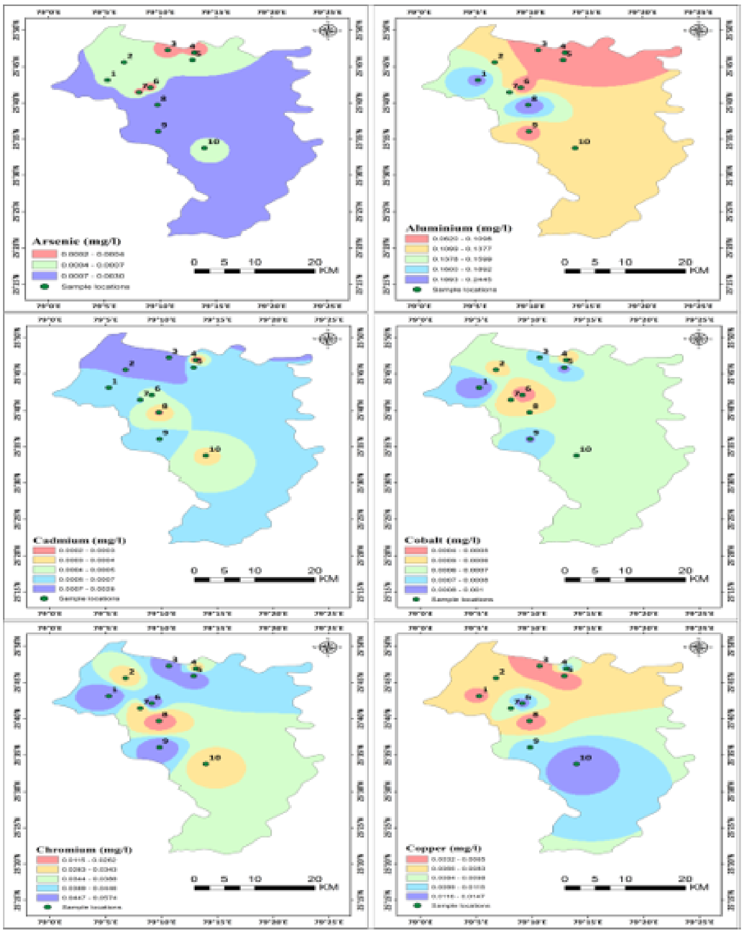


Figure 4. The map showed the concentration of Al, As, Cd, Co, Cr and Cu in the groundwater of the study area.

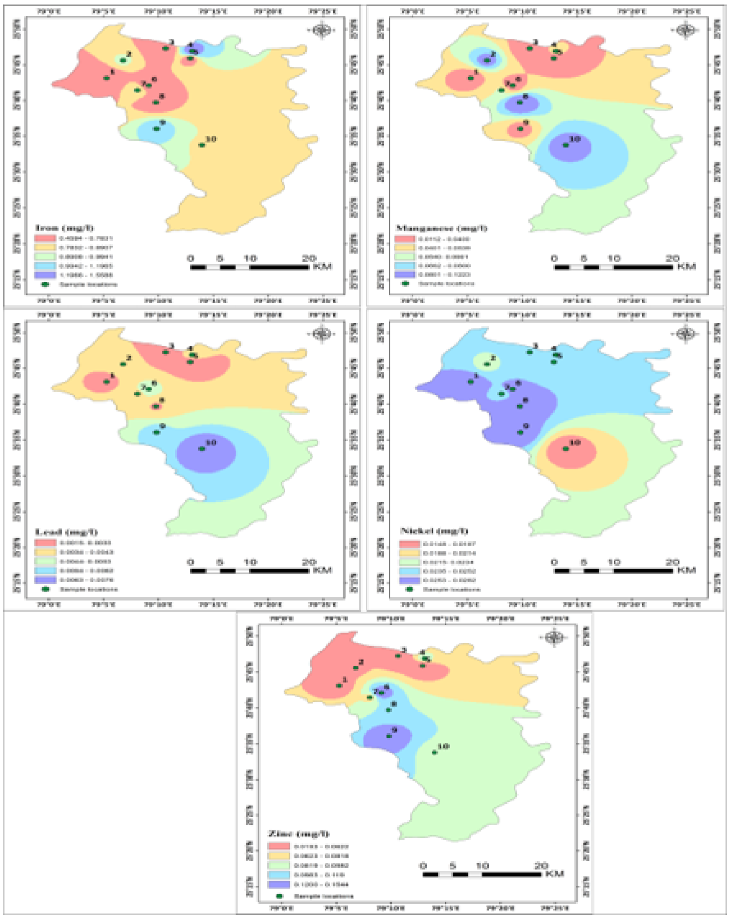


Figure 5. The map showed the concentration of Fe, Mn, Ni, Pb and Zn in the groundwater

Table 2: The obtained results of the principal component analysis

Heavy metals	Component		
	1	2	3
Al	-0.241	0.765	0.384
As	0.078	0.325	0.843
Cd	*-0.611*	-0.344	-0.239
Co	*-0.590*	-0.262	0.343
Cr	-0.226	*-0.906*	0.136
Cu	0.908	-0.100	-0.283
Fe	0.725	0.030	-0.061
Mn	0.202	0.960	-0.052
Ni	-0.171	-0.216	0.838
Pb	0.880	0.166	-0.178
Zn	0.876	-0.018	0.376
Initial Eigenvalues	4.175	2.566	1.774
Variance of percentage	37.954	23.331	16.132
Cumulative of percentage	37.954	61.285	77.416

The solubility of Mn-bearing minerals in groundwater depends directly on the main anions and the oxidation-reduction potential, especially with a relatively close-neutral pH (Rezaei *et al.* 2019). Factor 3 contributes 16,132% of the total variance and has demonstrated a strong positive loading of arsenic and nickel, suggesting naturally and anthropogenic sources. In addition, nickel and arsenic sources are possibly caused by the weathering of minerals or rocks in this region.

The results of Pearson correlation coefficients (r) (Table 3) indicates that Mn has a positive correlation with Al (r = 0.619), Pb with Zn (r = 0.709) and also As show a positive significant correlation with Ni (r = 0.496), therefore, this interaction between particular heavy metals demonstrates common sources, related connection and similar dependency in the movement processes in the study region. Furthermore, Cu has a significant positive correlation with Fe (r = 0.502), Pb (r = 0.921), Zn (r = 0.700) and negative correlation with Ni (r = -0.541) and this correlation between heavy metals is suggestive of the common and different sources. Moreover, the negative correlation of Cr with Mn (r = -0.921), Fe with Pb (r = -0541), and Ni with Pb (r = -0.488) and this relationship between heavy metals suggest an increase in one variable while the other decreases and inversely. Therefore, the negative relationship was showed the anthropogenic sources and positive correlation indicated the geogenic sources.

Table 3. Results of Pearson correlation coefficient

	Al	As	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Al	1									
As	0.473	1								
Cd	-0.275	0.203	1							
Cr	-0.483	0.173	0.37	1						
Cu	-0.279	0.212	0.462	0.075	1					
Fe	-0.298	0.008	*0.459*	0.331	0.560	1				
Mn	0.619	0.342	0.368	*0.923*	0.083	0.193	1			
Ni	0.160	0.496	0.016	0.229	*0.429*	0.088	0.311	1		
Pb	-0.052	0.059	*0.494*	0.257	0.921	0.541	0.365	*0.488*	1	
Zn	-0.092	0.412	*0.502*	0.126	0.700	0.458	0.155	0.186	0.709	1

The CA results have been divided into three clusters such as cluster A, cluster B, and cluster C (Figure 6). The first cluster A indicated that the insignificant contamination in (2, 7, and 10). Cluster B comprises (9, 4) that suggests the low to moderate polluted. Cluster C has 5 groundwater samples (5, 3, 1, 6, and 8) which are highly polluted. Cluster A comprises 30 % of groundwater is secure for domestic and drinking purposes (that is to be less reliant on anthropogenic factors), cluster B concludes 20 % groundwater samples are suitable for irrigation purposes, however, not suitable for drinking usages (that shows natural sources) where cluster C has 50 % of groundwater is not suitable for people usage (which indicate the more anthropogenic source).

As a consequence of geogenic and anthropological activities, heavy metals contamination has increased in groundwater across the Jhansi district, Bundelkhand.

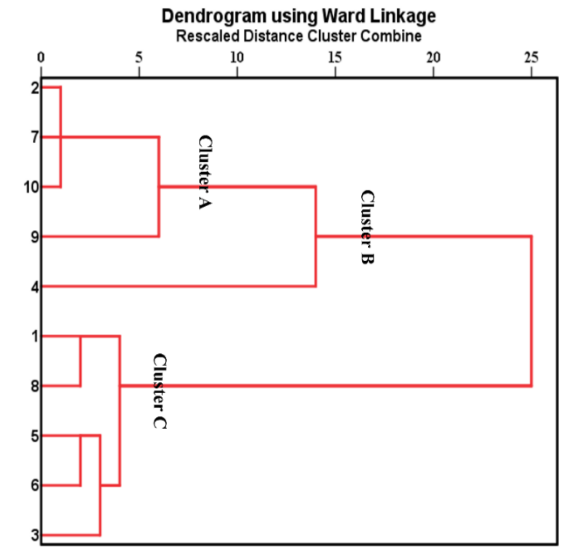


Figure 6. Dendrogram showed the Ward's linkage and Euclidean distance

4. Conclusion

The order was followed in this analysis by the mean heavy metal concentration in the groundwater samples as Fe (0.832 mg/L) > Al (0.124 mg/L) > Zn (0.080 mg/L) > Mn (0.051 mg/L) > Cr (0.039 mg/L) >Ni (0.024 mg/L) > Cu (0.008 mg/L) > Pb (0.004 mg/L) > As (0.001 mg/L) > Cd (0.001 mg/L) > Co (0.001 mg/L). The finding of results indicated that heavy metal concentrations of As (0.003 – 0.0002 mg/L), Cd (0.003 – 0.0002 mg/L), Cu (0.057 – 0.011 mg/L), Pb (0.007 – 0.001 mg/L) and Zn (0.154 – 0.019 mg/L) in total subsurface water samples were lower than the acceptable limits, except iron that 2 samples exceeded the value due to geogenic activities and rest of samples were within the permissible value set by the BIS (2012) and WHO (2017). Furthermore, findings from multivariable approaches such as PCA have been shown three factors that demonstrate data structures and total variance of 77.416 %of the dataset, specified by three variables such as 37.954 %, 23.331 %, and 16.132 % respectively and its outcome revealed that the groundwater pollution was due to agricultural fertilizers and pesticides and naturally in the study area. Moreover, the results of Pearson correlation coefficient (r) and HCA were demonstrated that anthropogenic and geogenic causes pollution the groundwater aquifers. Overall, the outcome of the results revealed that groundwater pollution is increasing in the study area due to geogenic factors (weathering of rocks/minerals) and human activities. The study thus suggests that 50% of groundwater is suitable for drinking and agricultural usage and 50% of samples are unsuitable for human's consumption, therefore, this research recommends any useful treatment of groundwater.

Data availability statement

The data has been preapared at the Nuclear Science Program, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, Selangor, Malaysia.

Acknowledgments

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Conflicts of Interest

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

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