

SOURCE OF GROUNDWATER IRON AND MANGANESE IN CHANDRAPUR DISTRICT, CENTRAL INDIA

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ABSTRACT

Groundwater sampling was carried out by grab sampling method from 36 sampling locations from the Chandrapur district in three seasons i.e. winter, summer, and post-monsoon. The samples were analysed for physicochemical parameters and heavy metals i.e. iron and manganese. Data obtained from the study area was interpreted by using multivariate statistical analysis i.e. principal component analysis, cluster analysis, correlation matrix and one way ANOVA to ascertain source apportionment of these two heavy metals. The results of the multivariate analysis revealed that iron and manganese both were associated with the lithogenic source. Groundwater iron concentration was higher when compared with manganese and at a number of sampling locations it was above the stipulated standard of BIS (Bureau of Indian Standards) (0.3 mg/L).

Keywords: *central India, Chandrapur, heavy metal, iron, manganese, source apportionment*

INTRODUCTION

A large percentage of the world population depends on groundwater as their main source of drinking water [1 - 3]. More than 50 % of the world's population depends on groundwater for drinking [4]. For many rural and small communities, groundwater is the only source of drinking water [5]. Over 50 % of the world's population is estimated to be residing in urban areas and almost 50 % of the mega-cities having populations over 10 million are heavily dependent upon groundwater and all are in developing world [6]. Over one billion people lack access to

clean safe water worldwide [7, 8]. In sub-Saharan Africa alone, up to 300 million rural people have no access to safe water supplies. Without safe drinking water near dwellings, the health and livelihood of families can be severely affected [9, 10].

Groundwater exploitation is generally considered as the only realistic option for meeting dispersed rural water demand [10]. This is because it is accessible anywhere; it is less capital intensive to develop and maintain; it is less susceptible to pollution and seasonal fluctuations and of natural good quality [7, 11]. However, the quality is under intense

stress from increasing demand and withdrawal, significant changes in land use pattern, climate change and pollution arising from geology and geochemistry of the environment [12, 13].

In India, 200 million people do not have access to clean drinking water. At present, only 85 % of the urban and 79 % of the rural population has access to safe drinking water. India is facing a water quality crisis. Toxic organic and inorganic pollutants already contaminate a growing number of groundwater reserves. With a number of avenues for its contamination, being a universal solvent, water tends to dissolve anything and everything that comes its way, thus changing its quality every time [6].

The United Nations considers universal access to clean safe water as a basic human right and an essential step towards improving living standard worldwide. The stress on water resources comes from multiple sources and the impact can take diverse forms [14]. Drinking water contamination with different chemicals and heavy metals, released from different anthropogenic sources has become a global concern [15]. The contamination of water resources has important repercussions for the environment and human health [16, 17]. About 2.3 billion individuals in the world suffer from diseases that are linked to water [18, 19].

Generally, drinking water containing different anions and heavy metals including Cd, Cr, Co, Hg, Ni, Pb, Zn, etc., has significant adverse effects on human health either through deficiency or toxicity due to excessive intake. The excessive ingestion of all these heavy metals including Cd, Cr, Co, Hg, Ni, Pb, and Zn has carcinogenic effects on human health [17].

Iron is one of the most abundant metals in the Earth's crust. It is found in natural freshwaters at levels ranging from 0.5 to 50 mg/L. Iron may also be present in drinking water as a result of the use of iron coagulants or the corrosion of steel and cast iron pipes during water distribution. Iron is an essential element in human nutrition. Estimates of the minimum

daily requirement for iron depend on age, sex, physiological status, and iron bioavailability range from about 10 to 50 mg/day [20].

Manganese is one of the most abundant metals in the Earth's crust, usually occurring with iron. Manganese is an essential element for humans and other animals and occurs naturally in many food sources. Manganese naturally occurs in many surface water and groundwater sources, particularly in anaerobic or low oxidation conditions and this is the most important source for drinking water. There have been epidemiological studies that report adverse neurological effects following extended exposure to very high levels in drinking water [20].

Drinking water quality in mafic and ultramafic rocks in northern Pakistan reported trace metals such as iron, manganese, nickel, chromium, and cobalt [21]. Iron in groundwater ranged from 134 to 5200 µg/L (mean ~ 1422 µg/L) [22]. Oyem et al. [23] reported higher iron content in groundwater of Boji-Boji Agbor area (27 %) and highest manganese content (31 %) in Boji-Boji Owa area of Nigeria. Melegy et al. [24] reported that about 50 % of the studied groundwater and surface water samples (n = 42) contained a high concentration of iron above drinking water guidelines of World Health Organization (WHO) (2011). As reported by Khan et al. [25] concentration of iron exceeded its permissible limit set by different organizations from some locations of Charsadda district, Pakistan. Ingestion of high level of iron can cause hemochromatosis with symptoms such as chronic fatigue, arthritis, heart diseases, cirrhosis, diabetes, thyroid diseases, impotence, and sterility. Iron, which facilitates persistent hepatitis B or C infection, also induced malignant tumours, colorectal, liver, lung, stomach and kidney cancers [26]. Utom et al. [27] reported up to 42 % of analysed groundwater samples iron concentration was beyond the Nigerian Industrial Standard; whereas, manganese concentration was in the range of 0.03 - 2.6 mg/L and 25 % sampling locations reported the concentration above the permissible limit.

According to Ocheri [28], variation in iron concentrations may be attributed to the geology of the environment, precipitation and runoff/infiltration, dissolution of iron minerals from rocks and soils, use of galvanized hand pump fittings and land use activities. Ibe et al. [29] reported high groundwater iron concentration that may be due to leaching of iron from iron scraps at the landfill site and from galvanized iron pipes in hand pumps equipped wells. The plausible source for the high iron concentration may be attributed to leaching from ferruginized sandstone and lateritic overburden. Iron concentration in groundwater may increase or decrease with the increasing depth of aquifers [30]. Hatva [31] reported iron and manganese contents in groundwater of Finland varied widely depending on aquifer structure, flow pattern and oxygen balance. Multivariate analysis showed that iron was associated with the lithogenic source [32]. Alam and Umar [33] reported relatively high concentrations for iron and manganese in a few samples. The groundwater iron source was associated with weathering followed by the dissolution of iron-bearing aluminosilicates, supported by sub-surface geology too; comprising of quartzite's underlined by granites. The origin of groundwater iron was attributed to the geogenic source [34]. Weathering processes along with corrosion products release iron in water [35]. Elevated manganese concentrations were associated with iron ores as well as lateritic mining [36]. According to Giri et al. [37], iron and manganese exceeded the IS 10500 standards in many locations. The elevated levels of iron and manganese were due to the natural occurrence of mineralization and background rock geochemistry. Summer season reported the elevated concentration of contaminants due to the decrease in the groundwater table. Chakrabarty and Sarma [38] attributed the possible source of origin of manganese as geogenic in nature. Bhuyan [39] reported groundwater was contaminated with iron which was attributed to its being geogenic in origin. Srinivasa Rao [40] iron was found to correlate considerably better with manganese in fluvial and coastal alluvium zones.

From the review of the related literature and researches, it was observed that selected studies have been carried out pertaining to groundwater heavy metals from the Chandrapur district. However, no significant emphasis was stressed upon groundwater iron and manganese and their source apportionment in particular. This is the identified gap in the research and new knowledge in this regard needs to be added to this subject domain. Hence, the purpose of this study is to analyse groundwater iron and manganese from the Chandrapur district and source of the same.

Study area

Chandrapur district (19°25' N to 20°45' N and 78°50' E to 80°10' E) is situated in the Vidarbha region of Maharashtra state of central India (Figure 1). The district is the easternmost district of the state. The district covers an area of 11,364 km² with elevation ranging from 106 m to 589 m asl (above sea level), the south-west part having a high level and south-east part with low level. The district comprises 15 administrative blocks and is surrounded by other districts such as Nagpur (north of northwest), Wardha (northwest), Yeotmal (west), Adilabad (south), Gadchiroli (east) and Bhandara (north). The district is bestowed with natural bounty in the form of dense forest and wildlife on one hand, and minerals such as coal, limestone, iron, copper, etc. on the other. Due to the abundant presence of natural resources and minerals, the district has witnessed sprawling coal mines, cement industries, pulp, and paper industry and a number of thermal power plants and at the same time Tadoba Andhari Tiger Reserve (TATR) which has one of the largest numbers of tigers in central India.

Climate and rainfall

The climate of the district is characterized by wide climatic conditions ranging from hot summer (in May temperatures rise up to 47 °C) to cold winter (December, temperature up to 7 °C) and general dryness throughout the year.

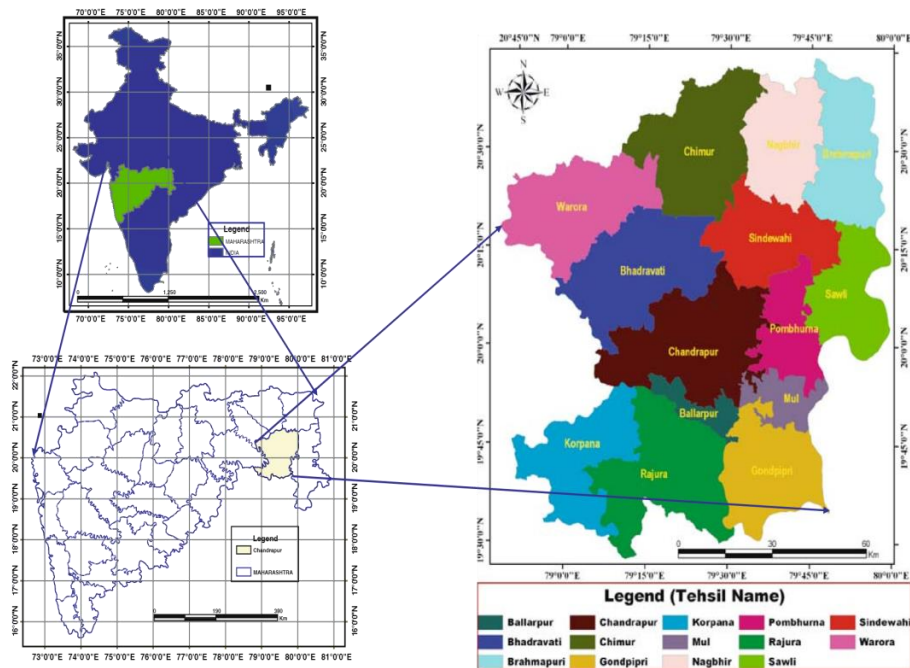


Figure 1. Chandrapur district with administrative blocks [41]

The district can be classified as tropical hot climate. The humidity was observed as 70 % during monsoon and 20 % in summer. The rainy season had reported rainfall from southwest monsoon (June - September) with annual rainfall ranging from 1200 - 1450 mm with an annual number of rainy days as 60 to 65. The rainfall is asymmetrically distributed in the district. The Warora administrative block receives comparatively minimum rainfall which gradually increases and reaches to a maximum around Brahmपुरi administrative block [42].

Geomorphology

Chandrapur district can be divided into two physiographic regions i.e., a plain region in valleys of Wardha, Penganga and Wainganga Rivers, and upland hilly region. The plain region is made up of widely spread and flat terrain occurring mostly along Wardha River. In Wainganga valley flat terrain exhibits rolling topography with residual hills in the southern part, while in the northern part (Brahmapuri administrative block) wide alluvial floodplains are observed. In Penganga valley, flat terrain covers the very little area in

the southwestern part of the district. The upland hilly region lies between Wardha and Wainganga Rivers comprising parts of Warora, Chandrapur, Mul, and Brahmपुरi administrative blocks. The southwestern part of the district in Penganga basin and covering parts of Rajura and Gadchander administrative blocks exhibit hilly topography. The entire area of the district falls in the Godavari basin. Wardha, Wainganga, and Penganga are the main rivers flowing through the district. These three rivers along with their tributaries rise in the upland within the district and drain the entire district [42].

Hydrogeology

The groundwater in Chandrapur district exists under confined/semi-confined and unconfined conditions. The depth of unconfined aquifer generally extends up to 20 m bgl (below ground level) and can be tapped by dug well. Pre-monsoon season reported the depth of water table in this aquifer in the range of 1.0 - 19.0 m bgl. The elevation of the water table varies from 230 m (NW) to 160 m (SE) asl. The groundwater flow is towards the Wardha River and its tributaries thereby confirming the affluent nature of the river. The coal belt's

peripheral area is the recharge zone and discharge belt is the area of hydraulic lows and natural drains. The potentiality of the unconfined aquifer is poor to moderate with a hydraulic conductivity of 2.0 to 6.0 m per day. Kamthis in the eastern limb of the Chandrapur region coal bed around Lohara, Durgapur and Bhatadi villages had the highest potential with a hydraulic conductivity of 18 - 32 m per day [41].

Geology

Geologically, Chandrapur district forms a part of Gondwana sedimentary basin. Lithologically Chandrapur district presents a variety of stratigraphic units right from Archean to recent alluvium and laterites. The brief description of these stratigraphic units includes:

Archean formations: Granites are holocrystalline rocks typically composed of quartz, feldspar and mica or hornblende and are of very varying grain; in this district, they are also associated with diorites and other holocrystalline basic rocks. Gneisses consist of gneiss proper - a foliated crystalline basic rock having much the same constituents as granite - with schists of hornblende, mica, and quartz and with much vein quartz. Dharwars, as they occur in the district, are highly altered shales (argillites) with some quartzites, sometimes ferruginous and with some micaceous schists. The Archeans comprises hard and fissured gneisses, quartzite. The Vindhyan metasediments are represented by flaggy and massive shale, limestone, sandstone, and ferruginous quartzite, covering an area of 1670 km². Groundwater in Archean crystallites and Vindhyan rocks occur under the table to semi-confined conditions in weathered and fractured zones. Aquifers in Archeans are characterized by a degree of weathering, secondary porosity, and effective inter-granular space; whereas, in Vindhyan, joint planes and fracture porosity developed during cooling and compression of sediments and in limestone the solution cavities play a major role in aquifer nature [43].

Purana formations: Resting unconformably on the gneisses occur the Vindhyan consisting mostly of sandstones, quartzitic sandstones, and quartzites with some shales and limestones. The Vindhyan of the district belongs to the Lower Vindhyan series [43].

Aryan formations: The rocks of the Talchir group, the lowermost member of the Gondwana series, are generally fine buff sandstones, greenish-gray silty shales and sandstones, underlaid by a bed containing boulders polished and striated; this striation or scratching supposed to be due to glacial action. The Barakar group is notable as containing all the workable beds of coal. Beginning from the top the arrangement of layers is: 1) coal, 2) sandstone and shales, 3) carbonaceous beds and 4) sandstones and shales. The Kamthi group is found resting unconformably on the Barakars. The rocks composing it include: 1) grit, more or less compact, 2) sandstones, coarse or fine-grained, with red blotchy streaks, with some conglomerate and 3) sandstones, argillaceous and ferruginous. Clays, usually red and green and shales of various colours occur intercalated among the sandstones. The rocks constituting the Kota-Maleri group are mainly red and green clays and argillaceous sandstones, the basal sandstones containing green clay-galls; limestone beds are found in association with the clays. The Deccan Trap series is composed of volcanic lavas and has been classified into upper, middle and lower traps; beneath it lie basal sedimentary beds, known as Lameta or Infratrappean, consisting of sandstones, sometimes calcareous, with limestone's, which are generally cherty and impure and some clays. Intercalated among the lava-flows occur volcanic ash beds and also some sedimentary beds; these latter are known as inter-trappean beds. The only traps found in the district belong to Lower Trap group. Laterites are next in succession to the trappean rocks and later still are the various deposits which include all the soils of the present area. In the river valleys, ossiferous gravels often cemented into a conglomerate of tolerable hardness are of frequent occurrence [43].

EXPERIMENTAL

Groundwater sampling and analysis

Thirty-six groundwater sampling locations comprising of hand pumps and dug wells from the Chandrapur district were identified (Figure 2 and Table 1). Stratified sampling was carried out for groundwater sampling during winter 2012, summer and post-monsoon 2013. Out of these sampling locations, 34 (94.44 %) were from hand pumps and two (5.55 %) from dug wells. The sampling locations were selected in such a way to ensure that the maximum study area be covered. Furthermore, these sampling locations were selected from rural areas where inhabitants were mostly dependent upon groundwater as a source of potable water and to carry out other domestic activities. Groundwater sampling was carried out by grab sampling method.

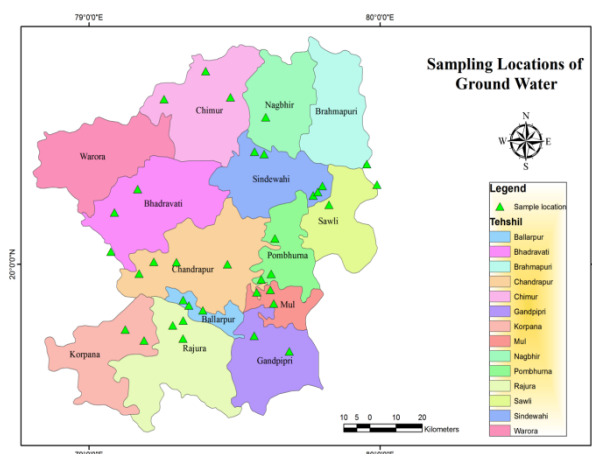


Figure 2. Groundwater sampling locations from the study area

In order to collect groundwater samples for analysis, two different capacities of polyethylene containers were selected. For analysis of general parameters (physicochemical), a narrow mouth polyethylene container of 1000 mL capacity (Poly lab, India) was selected, whereas, for heavy metals analysis a narrow mouth 100 mL capacity polyethylene container (Poly lab, India) was used. These both containers were thoroughly washed first with detergent then with distilled water followed by conc. HNO_3 (16 N, Merck) further by repeated washing

with distilled water in the laboratory. These containers were rinsed with a hand pump or dug well water before groundwater sampling and then the sample was collected into it. Heavy metals samples were preserved by adding conc. HNO_3 , 2 mL per 100 mL at the time of sampling. All reagents used while carrying out physicochemical analysis were of AR grade (Merck) and glassware was of borosilicate make. Double distilled water was used for the preparation of reagents. All reagents were prepared as stated in American Public Health Association (APHA) [44].

The concentrations of total heavy metals were determined after acid digestion with conc. HNO_3 [45]. Groundwater samples especially collected for determination of iron and manganese were acid digested in a pre-leached glass beaker on a hot plate at 95 °C and evaporated to 5 mL without boiling. While carrying this out, glass beakers were covered with a clean watch glass. This process resulted in the total extraction of metals from groundwater. After cooling, a small quantity of 1:1 conc. HNO_3 (16 N, Merck) was added into the digested sample and further refluxed for 15 min so as to dissolve any precipitate and residue resulting from evaporation. This digested sample after cooling was transferred into 25 mL volumetric flask and diluted up to 25 mL with double distilled water. This acid digested sample was used for the determination of iron and manganese concentrations. Heavy metals analysis was carried out by using ICP-OES (ICP-OES, Perkin Elmer, Germany, Dv 7000).

Statistical analysis

A multivariate statistical analytical approach, i.e., principal component analysis, cluster analysis, and correlation analysis was adopted for the interpretation of the data obtained. Principal component analysis (PCA) is used to infer the source of heavy metals (natural or anthropogenic). The components of PCA are rotated by a Varimax rotation. Cluster analysis is applied to identify different geochemical groups. It is formulated according to the Ward

Table 1. Groundwater sampling locations and characteristics (Water source: HP - Hand Pump, DW - Dug Well; Age - age of the hand pump or dug well)

Sampling location	Geographical details			Age (Years)	Depth (m bgl)	Average pH	Average TDS	Average Cl ⁻	Average Fe conc.	Average Mn conc.
	Latitude	Longitude	Altitude (m asl)							
Sonegaon (HP)	19°58'20.18"N	79°10'30.59"E	215	3	30.5	7.05	596.67	11.94	0.110	0.008
Telwasa (HP)	20°02'46.53"N	79°04'54.93"E	207	3	30.5	6.89	840.00	53.73	0.251	0.004
Belora (HP)	20°10'06.65"N	79°05'21.87"E	210	10	30.5	7.27	646.67	38.68	0.109	0.047
Sagra (DW)	20°14'57.66"N	79°10'03.50"E	240	57	15.2	7.25	1116.67	120.55	0.081	0.007
Pethbhansouli (HP)	20°32'05.79"N	79°15'46.12"E	209	3	30.5	7.05	833.33	91.86	5.090	0.412
Bhisi (HP)	20°37'49.43"N	79°24'03.33"E	287	1	45.7	6.8	1200.00	162.77	0.647	0.376
Pimpalgaon (HP)	20°32'42.30"N	79°29'16.55"E	246	25	76.2	7.02	1913.33	315.41	0.873	0.027
Mowada (HP)	20°15'47.70"N	78°59'38.90"E	198	10	54.8	7.11	783.33	65.80	0.173	0.003
Dongargaon (HP)	20°19'43.68"N	78°57'28.72"E	222	30	60.9	6.8	1440.00	223.44	0.871	0.372
Lohara (HP)	19°59'08.62"N	79°21'32.90"E	202	12	18.3	5.81	190.00	15.25	1.457	0.011
Chichpalli (HP)	20°00'00.99"N	79°28'54.27"E	226	12	21.3	6.93	3496.67	886.99	0.124	0.144
Dabgaon (T.) (HP)	19°57'06.42"N	79°35'52.71"E	215	3	91.4	6.87	1606.67	255.54	2.236	0.222
Naleshwar (HP)	19°58'15.72"N	79°37'57.79"E	215	12	42.7	6.57	1296.67	329.30	0.693	0.779
Karwan (HP)	20°05'03.19"N	79°38'33.42"E	205	8	45.7	7.33	673.33	58.68	0.128	0.053
Chikmara (HP)	20°13'36.52"N	79°46'23.80"E	214	25	30.5	6.98	1166.67	154.97	0.410	0.022
Pathri (HP)	20°11'54.59"N	79°49'49.52"E	240	20	30.5	6.73	586.67	79.44	0.190	0.057
Gunjewahi (DW)	20°15'21.25"N	79°48'11.22"E	230	60	10.7	7.44	400.00	17.41	0.081	0.003
Mangali Chak (HP)	20°14'03.28"N	79°47'16.26"E	224	25	60.9	7.04	466.67	19.66	0.176	0.003
Govindpur (HP)	20°28'54.27"N	79°36'45.26"E	271	25	45.7	6.93	1640.00	357.68	0.195	0.031
Ratnapur (HP)	20°21'8.87"N	79°34'14.42"E	250	10	30.5	6.87	996.67	158.05	1.441	0.113
Antargaon (HP)	20°21'39.96"N	79°36'11.34"E	246	15	60.9	7.49	616.67	8.17	0.164	0.003
Visapur (HP)	19°53'07.65"N	79°19'40.07"E	152	9	30.5	6.31	580.00	75.74	5.766	0.131
Ballarpur (HP)	19°52'01.24"N	79°20'56.78"E	243	5	18.3	6.12	560.00	63.70	18.213	0.045
Sasti (HP)	19°49'07.56"N	79°19'41.11"E	198	10	54.8	6.83	1980.00	269.49	2.270	0.088
Gowari (HP)	19°48'15.48"N	79°17'24.77"E	198	6	36.6	7.08	1006.67	102.43	0.308	0.003
Arvi (HP)	19°45'59.79"N	79°19'37.18"E	202	23	30.5	6.8	1003.33	97.69	0.524	0.005
Awarpur (HP)	19°47'32.39"N	79°07'45.38"E	216	2	60.9	7.13	1586.67	171.57	0.230	0.034
Lakhmapur (HP)	19°45'21.58"N	79°11'35.24"E	243	8	60.9	6.88	593.33	11.45	1.280	0.006
Kem (T.) (HP)	19°51'05.12"N	79°23'45.20"E	178	8	45.7	7.11	400.00	8.53	1.779	0.057
Ganpur (HP)	19°46'13.70"N	79°34'04.50"E	199	25	48.8	6.82	2720.00	435.26	0.601	0.004
Gondpipari (HP)	19°43'10.93"N	79°41'29.06"E	195	20	30.5	6.8	1446.67	230.97	1.562	0.287
Pombhurna (HP)	19°52'39.51"N	79°38'06.97"E	189	20	30.5	6.96	1246.67	177.45	0.310	0.008
Jam Tukum (HP)	19°55'06.70"N	79°37'40.47"E	174	20	76.2	6.9	1910.00	365.94	0.257	0.060
Dongar Haldi (HP)	19°54'56.71"N	79°34'57.48"E	187	6	36.6	7.01	1980.00	349.78	0.709	0.091
Durgapur (HP)	20°00'42.04"N	79°18'00.70"E	201	4	6.1	6.95	1866.00	219.72	0.256	0.286
Morwa (HP)	20°00'48.09"N	79°13'36.34"E	218	15	30.5	7.04	1180.00	116.27	0.251	0.003

algorithmic method. Outcomes are represented in a dendrogram, which illustrated the hierarchical arrangement of resulting clusters and values of distances between clusters (squared Euclidean distance). A correlation matrix is used to identify the relationship between the sampled elements [46]. The correlation coefficient is calculated in the form of a matrix [47]. One way analysis of variance (ANOVA) is used to estimate the measurement uncertainty across the whole site and for different sampling locations [48].

RESULTS AND DISCUSSION

Groundwater sampling locations are situated at different altitudes in the range of 152 - 287 m asl, year of installation from 1 to 60 years

(age), depth of 6 - 91 m bgl and iron and manganese concentrations variation in different seasons (Table 1). Groundwater samples are analysed for different physicochemical parameters: pH, total dissolved solids (TDS) (mg/L), chlorides (mg/L), iron (total) (mg/L) and manganese (total) (mg/L). These parameters average values are calculated from three seasons (winter, summer, and post-monsoon). The range is 5.8 - 7.4 (pH), 190.0 - 3496.66 (TDS), 8.170 - 886.98 (chlorides), 0.081 - 18.213 (iron) and 0.003 - 0.779 (manganese). The average value of total heavy metal content in the groundwater sample is in the order of iron > manganese. Maximum iron and manganese concentration from the samples is above the BIS permissible limit for respective metal (IS 10500:2012) (Table 2).

Table 2. Basic parameters for collected water samples

Parameter	Min.	Max.	Average	Std. Dev.	BIS
pH	5.8	7.4	6.9	0.3	6.5 - 8.5
TDS	190.00	3496.66	1182.38	699.24	500
Cl-	8.17	886.98	170.03	172.33	250
Fe	0.081	18.213	1.384	3.153	0.3
Mn	0.003	0.779	0.106	0.165	0.1

Std. Dev. - Standard Deviation; BIS - Bureau of Indian Standards (IS 10500:2012) permissible limit

Figure 3 (3a - 3d) depicts thematic maps for groundwater iron concentrations and Figure 4 (4a - 4d) depicts thematic maps for groundwater manganese concentrations for winter, summer, post-monsoon, and average concentration respectively. Minimum iron concentration in winter, summer and post-monsoon is below the detection limit (BDL), 0.164 mg/L (Sagra, DW) and 0.055 mg/L (Gunjewahi, DW) respectively; whereas, maximum 47.100 mg/L (Ballarpur, HP), 3.825 mg/L (Ballarpur, HP) and 4.022 mg/L (Visapur, HP) respectively. Maximum average iron concentration is in Ballarpur (HP) 18.213 mg/L and minimum in Gunjewahi (DW) 0.081 mg/L. The iron concentration in Ballarpur is 47.100 mg/L in winter, 3.825 mg/L in summer and 3.714 mg/L in post-monsoon. Seasonal variation in groundwater iron concentration is recorded. Maximum iron concentration is found to be elevated and above the permissible limit of 0.3 mg/L of the Indian Standard (2012) and aesthetic limit of WHO (2006) for iron. Groundwater manganese concentration in winter is in the range of BDL to 1.853 mg/L (Naleshwar, HP) in summer 0.003 mg/L (Morwa, HP) to 0.474 mg/L (Ganpur, HP); whereas, in case of post-monsoon it is in the range of 0.002 mg/L (Ganpur, HP) to 0.761 mg/L (Bhisi, HP). Average manganese concentration is in the range of 0.003 mg/L (Morwa, HP) to 0.779 mg/L (Naleshwar, HP). Seasonal variation in groundwater manganese concentration is recorded. Maximum manganese concentration is found to be elevated and above the permissible limit of 0.1 mg/L of the Indian Standard (2012) [49].

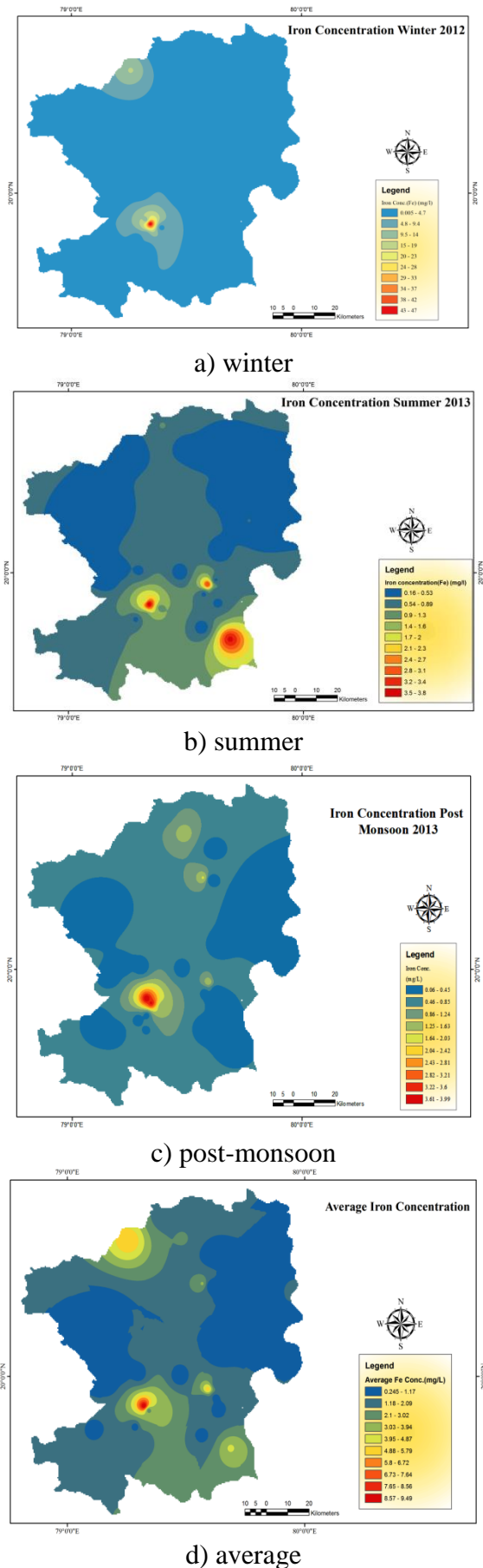


Figure 3. Thematic map of iron concentration

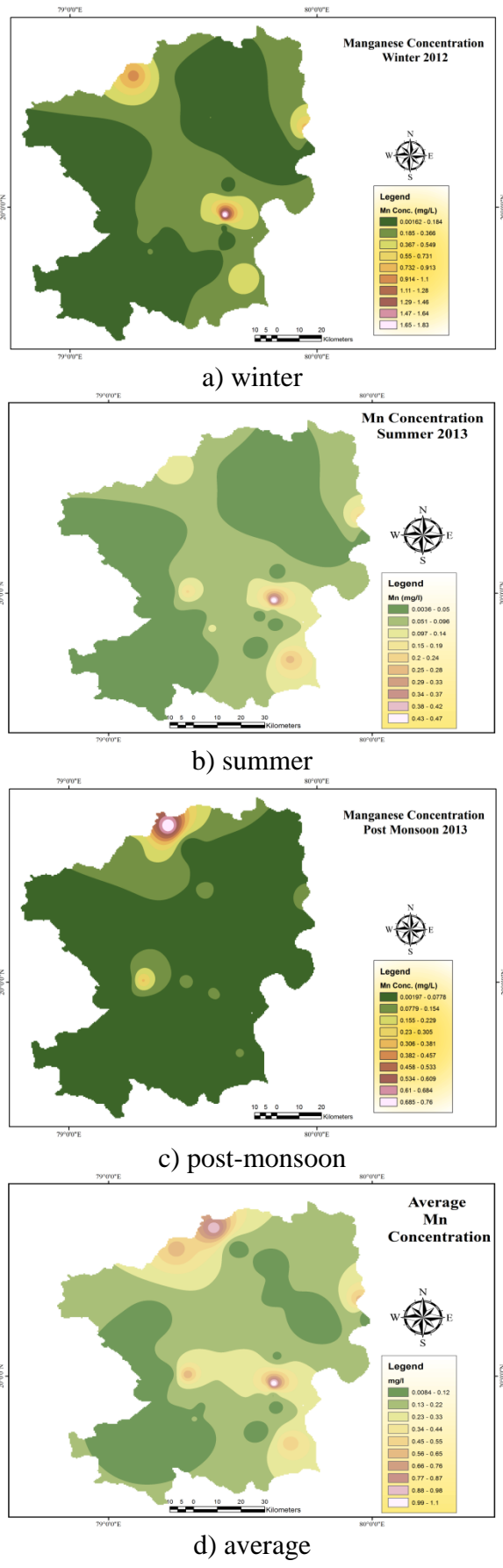


Figure 4. Thematic map of manganese concentration

Higher iron concentrations from hand pump are in agreement with results reported by (Satapathy et al., Rossister et al.) [41, 50]. Hand pumps being in close proximity to ores and minerals present in the Earth crust and water being a universal solvent that tends to dissolve these ores and minerals resulted together in a more elevated iron concentration than dug wells. Utom et al. [27] reported a minimum manganese concentration of 0.03 mg/L and maximum as 2.6 mg/L. Groundwater manganese concentration reported by Purushotham et al. [51] is in the range of 2.3 to 4340 $\mu\text{g/L}$ with an average of 2171 $\mu\text{g/L}$. Alam and Umar [33] reported manganese concentration range from 0.024 to 0.56 mg/L. Maximum manganese concentration (0.56 mg/L) in comparison with results of the existing study indicates that it is comparable with summer (0.474 mg/L). Maximum groundwater manganese concentration as reported by Agca et al. [52] is 1.026 mg/L; Cobbina et al. [53] is 1.05 mg/L; Hasan and Ali [54] is 9.98 mg/L; Homoncik et al. [55] is 1.9 mg/L; Melegy et al. [24] is 3.0 mg/L and Nawankwoala et al. [56] is 2.34 mg/L which again highlights that elevated manganese concentrations can be found in natural aquatic environment.

Principal component analysis

Principal Component Analysis (PCA) results for winter (Table 3) show that iron and manganese are grouped into a two-component model, which accounted for about 73 % of all the data variation. In the rotated component matrix, the first principal component (PC1, variance of 48 %) included iron while second principal component (PC2, variance of 25 %) is made up of manganese. Principal component (PC2) could be considered a natural component because the variability of heavy metals concentration appeared to be products of the study areas lithology. Of the component matrix, iron and manganese are observed to have rotated component matrix of 0.771 and 0.481 respectively, which is more than pH, TDS, and Cl^- . This suggests that the distribution of iron and manganese have a

lithogenic origin and therefore these two heavy metals are included in the second principal component.

As for the summer (Table 4), which accounted for ~ 72 %, of all the data variation, heavy metals are grouped into the two-component model. In the rotated component matrix, the first principal component (PC1, variance of ~ 41 %) includes iron and the second principal component (PC2, variance of ~ 30 %) is made up of manganese. As for the component matrix, it is observed that iron and manganese have rotated component matrix of 0.841 and 0.502 respectively which is more than pH, TDS, and Cl⁻. Factor loading plot for the summer is depicted in Figure 5b, which shows that iron and manganese at the positive axis of the plot and in comparatively close proximity with each other (as compared with winter) indicate that they are originated from a single

source –natural lithogenic origin. Compared to the winter (Figure 5a), iron and manganese are in close proximity to each other in the summer. The plausible reason which can be assigned to this observation is the prevailing environmental conditions in the aquatic environment. Due to a decrease in groundwater level in the summer, reduced (or no) dissolved oxygen and reduction in weathering and dissolution of minerals and ores present in the Earth crust can be assigned as contributing factors for such observations. In winter, dilution of heavy metals concentration, which got accumulated in the summer, resulted in observation as depicted in Figure 5a. Although iron and manganese are in similar axis and on the positive side of the plot, they are away from each other as compared with the winter season.

Table 3. Principal component analysis (total variance) (winter)

Component	Initial Eigen value			Extraction sums of squared loading			Rotation sums of squared loadings			Groundwater characteristics	Component matrix ^a		Rotated component matrix	
	Total	% Variance	Cumulative %	Total	% Variance	Cumulative %	Total	% Variance	Cumulative %		PC1	PC2	PC1	PC2
1	2.432	48.637	48.637	2.432	48.637	48.637	1.935	38.706	38.706	Fe	-.248	.776	-.263	.771
2	1.264	25.271	73.908	1.264	25.271	73.908	1.760	35.202	73.908	Mn	.437	.473	.428	.481
3	.683	13.662	87.570							pH	-.108	-.831	-.091	-.833
4	.578	11.553	99.123							TDS	.947	-.117	.949	-.098
5	.044	.877	100.000							Cl ⁻	.974	.007	.974	.026

Extraction method: Principal component analysis; Rotation method: Varimax with Kaiser normalisation;
^a Rotation converged in 3 iterations

Table 4. Principal component analysis (total variance) (summer)

Component	Initial Eigen value			Extraction sums of squared loadings			Rotation sums of squared loadings			Groundwater characteristics	Component matrix ^a		Rotated component matrix	
	Total	% Variance	Cumulative %	Total	% Variance	Cumulative %	Total	% Variance	Cumulative %		PC1	PC2	PC1	PC2
1	2.050	40.997	40.997	2.050	40.997	40.997	2.000	40.007	40.007	Fe	.172	-.829	-.095	.841
2	1.537	30.749	71.746	1.537	30.749	71.746	1.587	31.740	71.746	Mn	.411	-.393	.268	.502
3	.851	17.011	88.757							pH	-.256	.746	-.011	-.788
4	.522	10.440	99.196							TDS	.934	.297	.980	.009
5	.040	.804	100.000							Cl ⁻	.956	.227	.979	.082

Extraction method: Principal component analysis; Rotation method: Varimax with Kaiser normalisation;
^a Rotation converged in 3 iterations

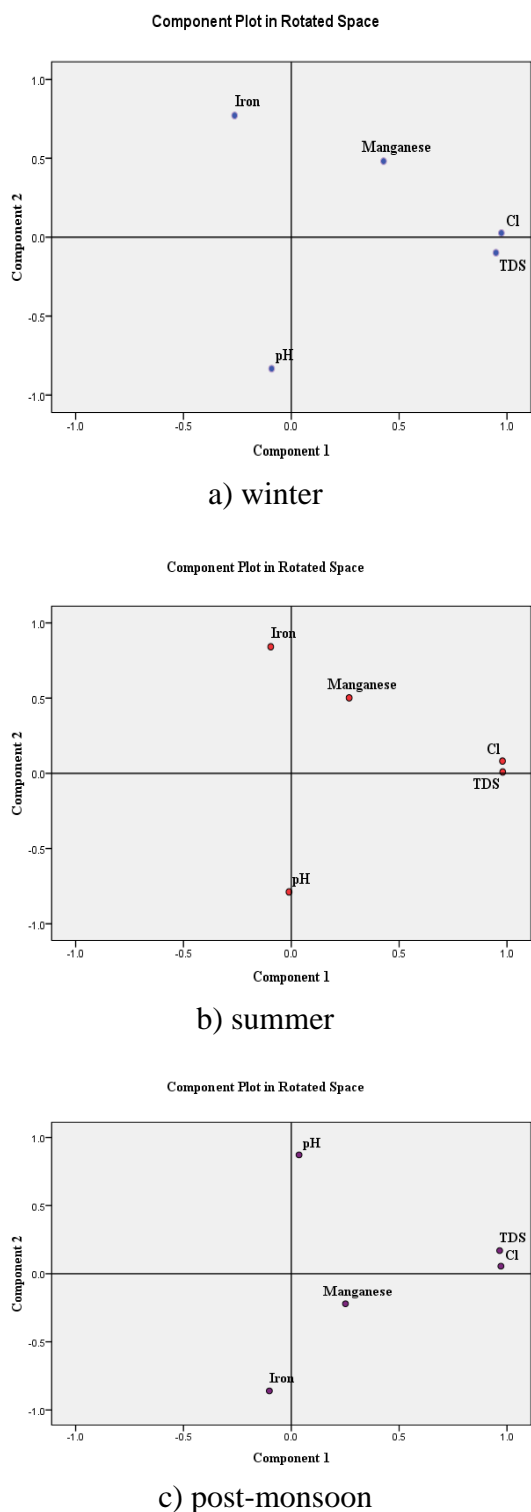


Figure 5. Plot of factor loading

As for the post-monsoon period (Table 5), the heavy metals distributed into the two-component model accounted for ~ 70 % of all the data variation. In the rotated component matrix, the first principal component (PC1, variance 42 %) includes iron and the second

principal component (PC2, variance ~ 28 %) is made up of manganese. The component matrix and rotated component matrix, which is divided into PC1 and PC2 as an anthropogenic and natural source of origin, shows that iron and manganese are found in quantity greater than 0.7 and 0.3 in the component matrix and in case of rotated component matrix they are -0.860 and -0.221 (Figure 5c). It may be stated that the post-monsoon period may have a negative impact on the concentration of these two heavy metals taken into consideration.

Rotated component matrix for winter and summer shows higher observations in PC2 than that of PC1 and they are further strongly correlated with iron at > 0.7 and manganese at > 0.4. This indicates that the source of heavy metals into groundwater during these two seasons is geogenic in origin. As for the post-monsoon season, in rotated component matrix, iron and manganese concentration reported negative observations; although negative, iron has a strong correlation (-0.860).

The PCA results suggest that two factors contribute to groundwater contamination and iron contribution is higher than that of manganese; these findings are in accordance with the observations obtained by Purushotham et al. [51] which states that iron contribution is higher than manganese. PCA carried out by Dwivedi and Vankar [32] showed iron and manganese have lithogenic sources, and similar conclusions are also drawn from the observations. These findings are also in accordance with results obtained by Mico et al. [57] which stated that iron and manganese appeared to be associated with parent rocks.

Cluster analysis

Iron

Cluster analysis results shown in Figure 6a for groundwater iron concentrations in winter revealed three major clusters: 1) Ballarpur, 2) Pethbhansouli and Visapur and 3) other 26 sampling locations. Those sampling locations

Table 5. Principal component analysis (total variance) (post-monsoon)

Component	Initial Eigen value			Extraction sums of squared loadings			Rotation sums of squared loadings			Groundwater characteristics	Component matrix ^a		Rotated component matrix	
	Total	% Variance	Cumulative %	Total	% Variance	Cumulative %	Total	% Variance	Cumulative %		PC1	PC2	PC1	PC2
1	2.103	42.057	42.057	2.103	42.057	42.057	1.952	39.036	39.036	Fe	-.497	.709	-.102	-.860
2	1.429	28.588	70.645	1.429	28.588	70.645	1.580	31.610	70.645	Mn	.117	.313	.251	-.221
3	.962	19.233	89.878							pH	.444	-.751	.036	.872
4	.454	9.075	98.953							TDS	.931	.308	.966	.169
5	.052	1.047	100.000							Cl ⁻	.882	.411	.972	.055

Extraction method: Principal component analysis; Rotation method: Varimax with Kaiser normalisation;

^a Rotation converged in 3 iterations

(n = 7, 19.44 %) where groundwater iron concentration is below the detection limit (BDL) are not included in cluster analysis. From these three clusters, it can be suggested that 26 sampling locations which formed a major cluster have comparable groundwater iron concentrations (0.006 - 5.714 mg/L); whereas, Pethbhansouli (HP) and Visapur (HP) have formed another cluster with iron concentrations of 14.313 mg/L and 11.536 mg/L respectively. Ballarpur (HP) with 47.100 mg/L has elevated groundwater iron concentration forming another cluster. The cluster analysis suggested that groundwater iron is geogenic in origin.

In the summer (Figure 6b), groundwater iron is clustered into three major clusters: 1) Ballarpur, Gondpipari, and Dabgaon (Tukum), 2) Visapur and 3) other 32 sampling locations. Out of these three clusters, Ballarpur, Gondpipari, and Dabgaon (Tukum) and Visapur are closest in terms of groundwater iron concentrations. The remaining major group have comparable iron concentrations. All other sampling locations are grouped into this cluster. From this dendrogram, the major cluster of 32 sampling locations suggested that iron has originated from one source which can be assigned as geogenic in origin.

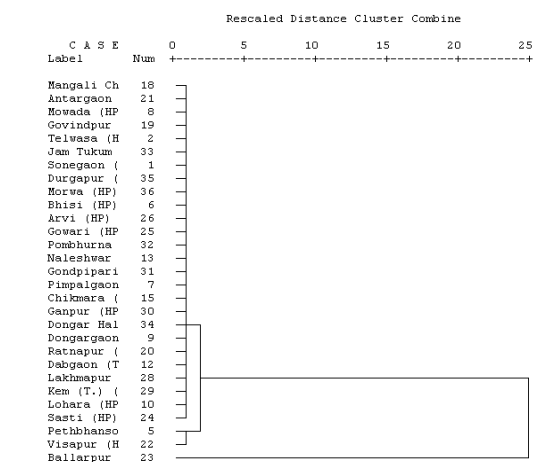
Similarly, cluster analysis for the post-monsoon period (Figure 6c) resulted in a dendrogram identifying close groundwater iron concentration groups. These three groups are: 1) Ballarpur and Visapur, 2) Dabgaon (Tukum), Ratnapur, Pimpalgaon and Kem (Tukum) and 3) remaining 30 sampling

locations. From this third major cluster it was evident that iron concentrations in groundwater are comparable. On the other hand, the second major cluster has comparable groundwater iron concentrations in the range of 1.2 to 1.7 mg/L. Visapur and Ballarpur cluster have near similar groundwater iron concentrations (4.022 mg/L and 3.714 mg/L, respectively). The close association of cluster 1 and cluster 2 is due to comparable groundwater iron concentrations. From the dendrogram which revealed a major cluster of 30 sampling locations indicated that groundwater iron concentration is geogenic in origin.

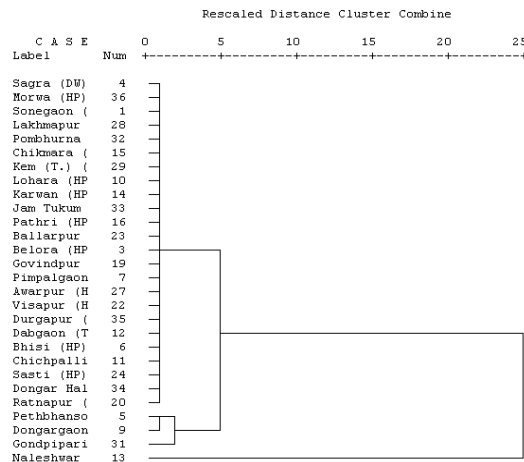
Manganese

Cluster analysis for groundwater manganese concentrations in winter (Figure 7a) indicates four major clusters: 1) Naleshwar, 2) Gondpipari, 3) Pethbhansouli and Dongargaon and 4) other 24 sampling locations. In this dendrogram 28 sampling locations are presented, other eight sampling locations due to 'BDL' are not presented. Sub-cluster included Pethbhansouli, Dongargaon, and Gondpipari. Clusters 1 and 2 have comparable results for groundwater manganese concentrations due to their closeness in the dendrogram. The major cluster of 24 sampling locations indicated groundwater manganese is geogenic in origin.

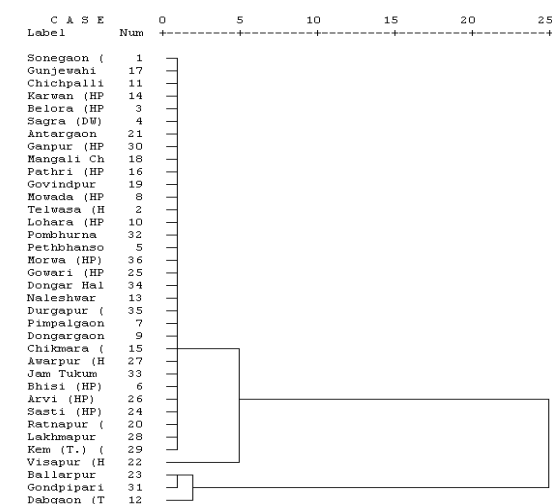
The summer (Figure 7b) revealed four major clusters: 1) Naleshwar, 2) Dongargaon, Durgapur, and Gondpipari, 3) Pethbhansouli, Dabgaon (Tukum), Chichpalli, Jam Tukum,



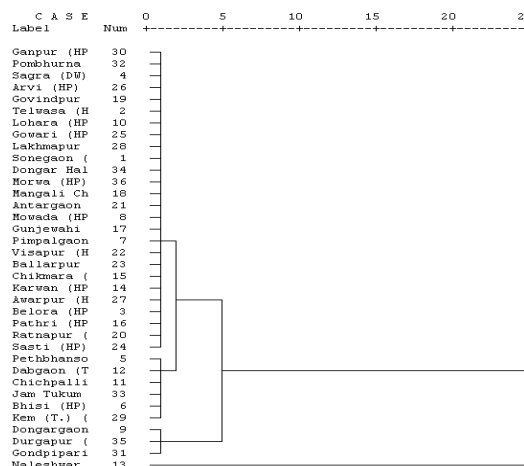
a) winter



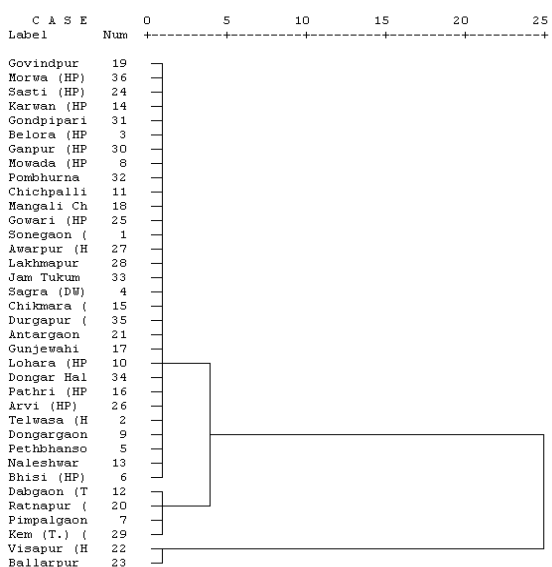
a) winter



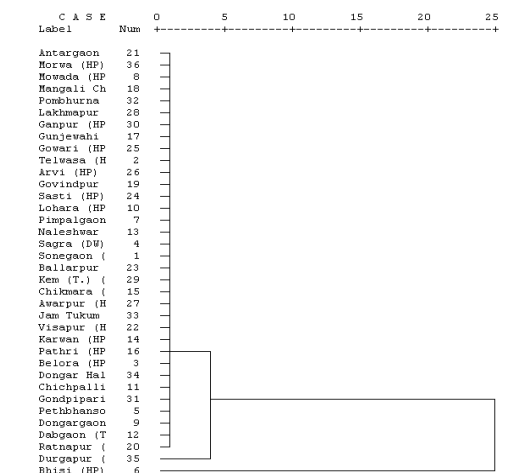
b) summer



b) summer



c) post-monsoon



c) post-monsoon

Figure 6. Cluster analysis of iron

Figure 7. Cluster analysis of manganese

Bhis, Kem (Tukum) and 4) other 26 sampling locations. Clusters 3 and 4, due to the similarity in groundwater manganese concentrations, have close proximity to each other. Cluster 1 which includes Naleshwar has highest groundwater manganese concentration (0.474

mg/L). From the dendrogram maximum sampling locations (n = 26, 72.22 %) it is evident that groundwater manganese concentrations are comparable with each other and it was revealed that the manganese is geogenic in origin.

Figure 7c indicates groundwater manganese for the post-monsoon period. The dendrogram indicates three clusters: 1) Bhisli, 2) Durgapur and 3) other 34 sampling locations. Bhisli (HP) has maximum (0.761 mg/L) groundwater manganese concentration, Durgapur (HP) with 0.312 mg/L and other 34 sampling locations have comparable concentrations. Clusters 1 and 2 are determined to be most similar in terms of groundwater manganese concentrations. From the third cluster, which forms the major cluster, it shows that groundwater manganese is geogenic in origin.

Correlation matrix

Correlation matrices for iron, manganese, pH, TDS, and Cl⁻ with significance level (1-tailed) in winter, summer, and post-monsoon are presented in Tables 6, 7 and 8 respectively.

Table 6. Correlation matrix (winter)

Particular	Fe	Mn	pH	TDS	Cl ⁻	
Correlation	Fe	1.000	.084	-.418	-.211	-.161
	Mn	.084	1.000	-.246	.181	.316
	pH	-.418	-.246	1.000	-.011	-.108
	TDS	-.211	.181	-.011	1.000	.942
	Cl ⁻	-.161	.316	-.108	.942	1.000
Sig. (1-tailed)	Fe		.312	.006	.108	.174
	Mn	.312		.074	.145	.030
	pH	.006	.074		.474	.265
	TDS	.108	.145	.474		.000
	Cl ⁻	.174	.030	.265	.000	

Table 7. Correlation matrix (summer)

Particular	Fe	Mn	pH	TDS	Cl ⁻	
Correlation	Fe	1.000	.243	-.455	-.029	-.010
	Mn	.243	1.000	-.142	.169	.205
	pH	-.455	-.142	1.000	-.020	-.118
	TDS	-.029	.169	-.020	1.000	.954
	Cl ⁻	-.010	.205	-.118	.954	1.000
Sig. (1-tailed)	Fe		.077	.003	.434	.477
	Mn	.077		.204	.163	.116
	pH	.003	.204		.455	.246
	TDS	.434	.163	.455		.000
	Cl ⁻	.477	.116	.246	.000	

Table 8. Correlation matrix (post-monsoon)

Particular	Fe	Mn	pH	TDS	Cl ⁻	
Correlation	Fe	1.000	.040	-.546	-.209	-.131
	Mn	.040	1.000	-.053	.112	.077
	pH	-.546	-.053	1.000	.187	.058
	TDS	-.209	.112	.187	1.000	.938
	Cl ⁻	-.131	.077	.058	.938	1.000
Sig. (1-tailed)	Fe		.408	.000	.111	.223
	Mn	.408		.380	.259	.327
	pH	.000	.380		.138	.369
	TDS	.111	.259	.138		.000
	Cl ⁻	.223	.327	.369	.000	

In winter (Table 6), correlation matrix for these selected five groundwater characteristics shows that iron and manganese have a correlation at a significance level of 0.312 (1-tailed), manganese and TDS with 0.145, pH and TDS with 0.474 and pH and Cl⁻ 0.265 at 1-tailed. The observations for significant (1-tailed) for the summer (Table 7) among different variables showed that iron has a significant relation (1-tailed) with TDS and chloride at 0.434 and 0.477 respectively. In the case of manganese with TDS and chloride, it is found to be 0.163 and 0.116 respectively significant. Observations for post-monsoon (Table 8) revealed that manganese has a correlation with iron (0.408), pH (0.380), TDS (0.259), and Cl⁻ (0.327).

One way ANOVA

The test statistics for groundwater iron (Tables 9 and 10) was $F(2,105) = 2.501$; $p < 0.087$ and manganese (Tables 11 and 12) is $F(2,105) = 4.595$; $p < 0.012$. The p statistic is used to test the null hypothesis. The p statistics computed for groundwater iron is found to be 0.087; whereas, for groundwater manganese, it is 0.012. These two calculated p values in comparison with alpha ($\alpha < 0.05$) reported that groundwater iron results are not statistically significant at this level (0.05); whereas, groundwater manganese is significant at this level and null hypothesis must be rejected.

Furthermore, the mean square between groups and within groups provides information pertaining to sampling variance and analytical measurement variance respectively. From these observations, it can be concluded that

sampling variation is higher (iron 57.319 and manganese 0.243) as compared with analytical measurement variance (iron 22.915 and manganese 0.053). Thus, it can be concluded that variation in groundwater iron and manganese concentration originate from a sample rather than analytical measurement

variation. Thus, it can be further concluded that errors from analytical measurements are minimum, and sampling variation has contributed to analytical measurement. It can also be concluded that spatial variation in groundwater iron and manganese concentration is there from the study area.

Table 9. Descriptive details for one way ANOVA for groundwater iron

Season	n	Mean	SD	Std. Error	95% confidence interval for mean		Minimum	Maximum
					Lower bound	Upper bound		
Winter	36	3.522	9.01	1.364	0.0673	5.609	BDL	47.100
Summer	36	0.730	0.909	0.151	0.423	1.038	0.164	3.825
Post-monsoon	36	0.582	0.920	0.153	0.271	0.894	0.055	4.022
Total	108	4.834	10.839	0.467	0.458	2.309	BDL	47.100

Mean, SD, Std. Error, Minimum and Maximum are reported in mg/L. BDL - below detection limit, SD - standard deviation

Table 10. One way ANOVA for groundwater iron

Heavy metal	Source of variations	Sum of squares	df	Mean square	F	Sig.
Fe	Between groups	114.638	2	57.319	2.501	0.087
	Within groups	2406.085	105	22.915		
	Total	2520.72	107			

df - Degree of freedom, F - F test, Sig. - Significant

Table 11. Descriptive details for one way ANOVA for groundwater manganese

Season	n	Mean	SD	Std. Error	95% confidence interval for mean		Minimum	Maximum
					Lower bound	Upper bound		
Winter	36	0.257	0.390	0.060	0.078	0.323	BDL	1.853
Summer	36	0.058	0.095	0.015	0.026	0.091	0.003	0.474
Post-monsoon	36	0.058	0.135	0.022	0.012	0.103	0.002	0.761
Total	108	0.373	0.620	0.022	0.060	0.151	BDL	1.853

Mean, SD, Std. Error, Minimum and Maximum are reported in mg/L. BDL - below detection limit, SD - standard deviation

Table 12. One way ANOVA for groundwater manganese

Heavy metal	Source of variation	Sum of squares	df	Mean square	F	Sig.
Mn	Between groups	0.485	2	0.243	4.595	0.012
	Within groups	5.547	105	0.053		
	Total	6.032	107			

df - Degree of freedom, F - F test, Sig. - Significant

One way analysis of variance for iron and manganese between groups and within the group as reported by Oyem et al. [23] is in accordance with the findings of this study. The levels observed between groups are higher those observed within a group, which indicates that variation in iron and manganese concentration is due to sampling variance and not due to analytical measurement variance.

Water source age, depth, Fe and Mn concentration

Correlation between water source age (years), depth of water source (m bgl), and iron and manganese concentration during winter (Table 13) found that the correlation of iron concentration with age and depth of water source is significant at 0.05 level. No correlations are observed between these five variables. In the summer (Table 14), groundwater iron concentration correlates with age of water source significantly at 0.05 level; whereas, correlation of manganese with iron is also significant at the same level. Post-monsoon observations pointed out (Table 15), iron concentration correlates with age of water source significantly at 0.05 level, iron and manganese correlations with altitude and age of water source are significant at 0.01 level. Furthermore, from the observations in these tables, it is evident that the age of water source and iron and manganese concentration are not correlated. The hand pump corrosion which may be an issue of concern as the age of hand pump progresses; however, the observations cannot provide proof for its contribution to groundwater iron concentration. Thus, it can be stated that the age of water source and groundwater iron concentrations are not correlated with each other. This finding indicates that the source of groundwater iron is of geogenic origin and may not originate from hand pump corrosion. The findings are in accordance with Hasan and Ali [54] that there is no clear trend between the age of tube-well and manganese concentration.

Table 13. Pearson's correlation coefficient between water source characteristics (winter)

	Altitude	Age	Depth	Fe	Mn
Altitude	1				
Age	0.17196	1			
Depth	0.07183	- 0.1707	1		
Fe	- 0.0496	- 0.2125**	- 0.2009**	1	
Mn	- 0.0712	- 0.1438	0.03149	0.08414	1

* Significant at 0.01 level; ** 0.05 level

Table 14. Pearson's correlation coefficient between water source characteristics (summer)

	Altitude	Age	Depth	Fe	Mn
Altitude	1				
Age	0.17196	1			
Depth	0.07183	- 0.1707	1		
Fe	- 0.1388	- 0.2129**	0.08912	1	
Mn	- 0.0092	- 0.118	0.05821	0.24266**	1

* Significant at 0.01 level; ** 0.05 level

Table 15. Pearson's correlation coefficient between water source characteristics (post-monsoon)

	Altitude	Age	Depth	Fe	Mn
Altitude	1				
Age	0.17196	1			
Depth	0.07183	- 0.1707	1		
Fe	- 0.373*	- 0.2392**	- 0.0129	1	
Mn	0.3173*	- 0.2686*	- 0.033	0.04001	1

* Significant at 0.01 level; ** 0.05 level

The water source contributing to groundwater iron concentration from wells where casing pipes are very old and corroded is ruled out by Alam and Umar [33]. This observation is in agreement with the findings of the study. Pearson's correlation coefficient for the age of water source and iron and manganese concentration reported negative weak to moderate correlation in all the seasons studied. Pearson's correlation coefficient between water extraction depth and iron and manganese concentration could not be established, which is broadly consistent with Daughney [58]. If groundwater is extracted from greater depths

where typically and significantly more reducing conditions prevail than in the shallow groundwater, then a correlation between water extraction depth and metal concentration would be probable. However, the lack of correlation between water extraction depth and concentrations of iron and manganese in groundwater indicates that such indirect relationships are not significant.

CONCLUSION

The PCA carried out on groundwater iron and manganese identified two principal components controlling their variability. Iron and manganese have been included in PC2, which is controlled by lithogenic sources. Cluster analysis of groundwater iron and manganese concentration from winter, summer and post-monsoon season showed that maximum sampling locations are forming a major cluster and in some cases, sub-cluster is also observed. The results of this cluster analysis show that a major cluster group originates from one source and it can be defined as geogenic in origin.

In the correlation analysis, it is found that iron and manganese are not significantly correlated with each other. One-way analysis of variance (ANOVA) for iron and manganese revealed between-group observations are higher than within-group observations, thus variation in iron and manganese concentration is due to sampling variance and not to analytical measurement variance. Age, altitude, and depth (in general) of water source have no significant correlation with groundwater iron and manganese concentration.

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