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Assessing Groundwater Contamination due to Heavy Metals and Their Spatial Distribution in an Industrial Area of Western India

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Abstract

The study was undertaken in and around the industrial establishments located in Ankleshwar region of Gujarat, India for understanding the variation in Cd, Cr, Cu, Mn and Ni level of groundwater in pre- and post-monsoon periods. The concentration of Cd exceeded the acceptable limits of BIS 2012 for drinking water in both the seasons whereas Cr, Cu, Mn and Ni had concentration levels above acceptable limits in the pre-monsoon season and below acceptable limits in the post-monsoon season. The average HPI value in the pre-monsoon season was 537.2 and it came down to 359.5 in the post-monsoon season, but these values exceeded the critical value of 100 in both the seasons and they were attributable to the discharge of effluents from the industries. Spatial distribution map was prepared using GIS for a better understanding of the levels of heavy metals contamination in the sampling sites.

Key words: Contamination, Groundwater, Effluents, Heavy-metal pollution index (HPI), Seasonal variation

Introduction

Chemical estates at Ankleshwar and Panoli were set up to boost industrial development and achieve the overall economic growth of Gujarat in the 1970s. These estates spread over an area of around 2600 hectares and consist of over 1700 industries manufacturing varieties of chemicals including pesticides, drugs, pharmaceuticals, petroleum products, textiles, plastics and dyes. These estates are dominated by small-scale industries which do not have effluent treatment plants of their own. There are several methods for removal of heavy metals from industrial effluents and these methods are costly making them unaffordable to small-scale industries (Carrondo *et al.,* 1979). This being so, two Common Effluent Treatment Plants (CETPs) are installed at these estates. Even these CETPs do not serve the intended purpose because proper treatability study of influents was not held prior to designing these plants. As a result, waste water being collected for treatment is discharged into the rivulet carrying toxic, dark brown or black effluents round the year. The toxic water percolates down to the aquifers and contaminates the groundwater used as drinking water. In addition, the interaction between groundwater and the polluted surface water too results in the transfer of heavy metals in the effluents to the groundwater system. The farmers in the adjoining areas use water from the rivulet for irrigating crops. The uptake of trace elements found in the effluents by plants leads to its entry into the food chain with the potential to cause health problems. Hapke (1996) and Sathawara *et al.* (2004) have also stated that persistent pollutants from water get transferred into the food chain and eventually reach the humans through plants and animals. The World Health Organization (WHO**)** in 1973 classified trace elements as essential (Zn, Sn, Cu, Mo and Cr), probably essential (Mn, Si, Ni and Vd) and potentially toxic with some possibility of essential functions (Pb, Cd, Hg, As and Li). The classified trace elements are toxic to the body depending on the concentration levels from high to low in the above stated order (Batley, 1983; Merian, 1991). Heavy metals are persistent and chemically stable (Hapke, 1996); hence the

removal of these trace elements is not possible once they contaminate the groundwater. However, attempts can be made for their dilution.

The study area falls in a region through which the Narmada River flows. The discharges from industrial complexes and municipality located in the catchment area are the major cause of degradation of the Narmada River (Jain *et al.,* 2008). Trace elements like Mn, Cu, Cd, Ni, Cr, Pb and Co were higher in tube well water of Nawagam-Vatava region of Gujarat because of effluents flowing in Khari canal (Parmar and Patel, 2009). The concentration of heavy metals such as Pb, Ni, Cr, Zn and Cu was found to be higher in the pre-monsoon season along the course of Sabarmati River and its tributary Kharicut canal (Rita *et al.,* 2013). The Central Pollution Control Board (CPCB, 2009), India has designated the Ankleshwar and Panoli industrial estates as a red zone. This has stressed the need for undertaking a study for a deeper understanding of the extent of the problem of pollution because of heavy metals.

This study was undertaken to find out the extent to which the areas are affected by industrial pollution. In specific terms, the study aimed to identify heavy metals concentrations, their seasonal variations and the contributing factors, and find out water quality based on heavy metal pollution index (HPI) and spatial mapping.

Materials and Methods

Groundwater samples were collected from 50 sites along Amla Khadi and Wandi Khadi, tributaries of Narmada as well as from locations close to the industrial estates of Ankleshwar and Panoli in Gujarat, India (Fig. 1). Effluents from the two industrial estates are discharged into the said tributaries. The sampling sites were specifically chosen in order to assess the impact of effluents from Ankleshwar Gujarat Industrial Development Corporation (GIDC) and Panoli GIDC on groundwater quality. Samples were collected from tube wells ranging in depth between 10 to 45 m below ground level during two different seasons, *viz.* pre-monsoon (PRM) and post-monsoon (POM) in the months of May, 2016 and December, 2016, respectively. GPS readings were noted at the time of collecting the samples in the PRM season so that subsequent samples were collected from the same locations in the POM season. The tube wells were allowed to run for 30 minutes as a precautionary measure and thereafter samples were collected in prewashed poly-propylene bottles following the standard method (APHA, 2005). Water samples so collected were placed in a cooler with ice and transported to the

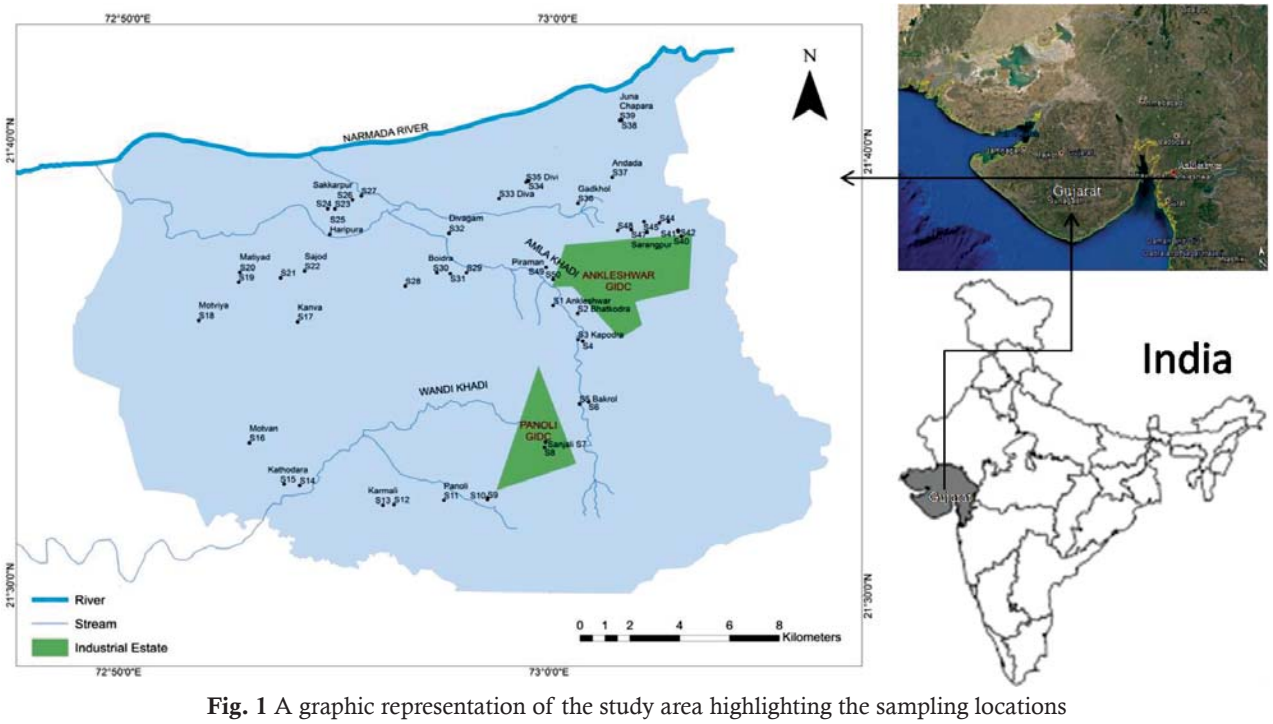

Fig. 1 A graphic representation of the study area highlighting the sampling locations

laboratory immediately for further analysis. The collected samples were filtered using Whatmann no.42 filter paper and acidified with concentrated nitric acid to a pH below 2.0 to prevent the occurrence of any undesirable change in the properties as required by standard procedure (Radojevic and Bashkin, 1999).

The concentration of heavy metals viz. cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni) and manganese (Mn) was analyzed by using atomic absorption spectrometry (ZEEnit 700 P, Analytic Jena) with specific lamp for each particular metal. The following wavelengths were employed: Cd (228.8 nm), Cr (357.9 nm), Cu (324.8 nm), Ni (232 nm) and Mn (279.5 nm)

The heavy metal pollution index (HPI) is a commonly used technique to measure the overall quality of water based on the cumulative presence of individual heavy metal. The HPI model (Mohan *et al.*, 1996) based on weighted arithmetic mean of heavy metal concentrations was used in this study. In this model the unit weightage is inversely proportional to the recommended standards of the corresponding parameter and the model is represented as:

$$HPI = \frac{\sum_{i=1}^{n} W_i Q_i}{\sum_{i=1}^{n} W_i}$$

where, Q_i is the sub index of the i^{th} parameter, W_i is the unit weightage of the i^{th} parameter and n is the number of parameters considered.

For calculation of the sub-index (Q_i) of the parameter, the following formula is given:

$$Q_i = \sum_{i=1}^{n} \frac{|M_i - I_i|}{S_i - I_i} \times 100$$

where, M_i is the monitored value of heavy metal of i^{th} parameter, I_i is the ideal value of i^{th} parameter and S_i is the standard value of i^{th} parameter. The numerical difference of the two values (M_i and I_i) has been taken as absolute value irrespective of the algebraic sign. The concentration limits given by Bureau of Indian Standards (BIS, 2012) were taken for this study and the critical pollution index of HPI value for drinking water is 100 as proposed by Prasad and Bose (2001). Principal Component Analysis (PCA) has been used to identify the factors which contribute to heavy metal pollution in the study area.

The data were statistically analyzed using STAR (Version: 2.0.1) statistical software package for calculation of average, standard deviation, variance, paired differences and level of significance. The level of significance was calculated at 95% confidence level. The HPI and Geographical Information System (GIS) were used to put the water quality data into a clear perspective and created an easy-to-comprehend format that highlighted the overall water quality condition with a view to facilitating remedial measures. The spatial distribution maps were prepared using Inverse distance weighted (IDW) interpolation technique of ArcGIS 10.1 software (Environmental Systems Research Institute, Redlands 380 New York Street 92373, California, United States).

Results and Discussion

In the pre-monsoon (PRM) season, the mean concentrations of Cd, Cr, Cu, Ni and Mn for the entire sampling areas were 0.015, 0.079, 0.106, 0.105 and 0.182 mg l^{-1}, respectively (Table 1). In the pos-monsoon (POM) season, the mean concentrations of Cd, Cu, Mn and Ni were 0.013, 0.016, 0.007 and 0.006 mg l^{-1}, respectively. The concentration of Cr during POM season was below detectable levels.

In 34 out of the 50 sites, Cd ranged from 0.015 to 0.032 mg l^{-1} in the PRM season and from 0.015 to 0.020 mg l^{-1} in the POM season, far exceeding the acceptable limits for drinking water. In the remaining 16 sites, no trace of Cd was found in both the seasons. The high levels of Cd are probably the result of discharge of industrial waste and leaching from sewage-laden landfills (Singh, 2003). Higher concentrations of Cd in the study area are possibly due to the presence of Cd in different industrial products like Ni-Cd batteries, TV tubes, alloys, pigments, plastics, solar cells and fungicides (Lawrence, 1981). For Cr, the values ranged from 0.000 to 0.225 mg l^{-1} in the PRM season and 34 sites showed concentrations above the acceptable limits in the PRM season and Cr was not found in any of the sites in the POM season. The discharges of industrial waste water from various industries such as metallurgical, refractories and chemical, mainly comprising of pigments, electroplating and tanning are the

Table 1. Summary statistics of heavy metals in PRM and POM seasons and its comparison with drinking water limits of BIS (2012) (unit: mg l^{-1})

Locations	Cd	Cr	Cu	Mn	Ni
PRM (n=50)					
Minimum	0.000	0.000	0.057	0.000	0.096
Maximum	0.032	0.225	0.235	0.446	0.298
Mean	0.015	0.079	0.106	0.105	0.182
SD	0.011	0.060	0.037	0.134	0.053
POM (n=50)					
Minimum	0.000	0.000	0.004	0.000	0.000
Maximum	0.020	0.000	0.030	0.076	0.082
Mean	0.013	0.000	0.016	0.007	0.006
SD	0.009	0.000	0.006	0.014	0.016
Acceptable limit	0.003	0.050	0.050	0.100	0.020
Permissible limit	No relaxation	No relaxation	1.500	0.300	No relaxation

PRM and POM depict pre-monsoon and post-monsoon, respectively

principal anthropogenic sources of Cr contamination (Kotasa and Stasicka,2000). Beyond detectable limits of Cr in POM season can be plausibly explained by less discharge of effluents containing Cr due to less industrial activity in the monsoon providing sufficient time for pre-monsoon Cr to settle down in the sediment. This is consistent with the observation that the released Cr in the natural/groundwater is particle associated and therefore gets deposited into sediments (Smith *et al.,* 1995).

Cu ranged from 0.057 to 0.235 mg l^{-1} in the PRM season and from 0.004 to 0.030 mg l^{-1} in the POM season. The presence of Cu may be due to the discharge of industrial effluents from electroplating, paints and dyes, petroleum refining, fertilizers, metallurgy, pesticide, iron and steel industries (Shrivastava, 2009). The concentration of Cu was above acceptable levels in all the sites in the PRM season, whereas it was within the acceptable limits in all the sites in the POM season. Seasonal differences are greatest for Cu because of greater sorption to solids and particulate loads due to erosion and movement of solids caused by greater flow of water (Brian and Michael,2009). The concentrations of Mn ranged from non-detectable to 0.446 mg l^{-1} in the PRM season and non-detectable to 0.076 mg l^{-1} in the POM season. In the PRM season the levels in 34 sites were found to be higher than the acceptable levels. In the POM season all sites were within the acceptable limits. Mn is found in most parts of the earth's crust as a natural mineral. This possibly explains the presence of Mn in the groundwater. However, the possibility of its presence due to the industrial waste cannot be entirely discounted. For Ni, the values ranged from 0.096 to 0.298 mg l^{-1} in the PRM season and from 0.000 to 0.082 mg l^{-1} in the POM season. The levels were dangerously high in all the sites in the PRM season. However, in the POM season the levels became much less in all the sites and only in 3 sites the levels stood higher than the acceptable levels. The source of Ni in groundwater may be due to its use in the food processing, metallurgical, chemical and pigments industries (Cempel and Nikel, 2006).

Concentration of Cd in both PRM and POM seasons was higher than the level prescribed for drinking water (BIS, 2012) in most of the sampling locations. In the case of Cr, Cu, Mn and Ni, the levels were above the acceptable limits in the PRM season in most of the sites and they dipped to acceptable levels in the POM season.

A paired t test of PRM and POM groundwater samples was done to compare the seasonal variations caused in respect of all the five heavy metals. Concentrations for all the heavy metals in POM season were significantly lower than the mean concentrations for these metals in the PRM season (Table 2).

Principal component analysis justified the contribution of two factors (PC-I and PC-II) to 95.8 % of the total variance (Table 3). Factor 1 comprising of Cd, Cr, Cu and Ni accounted for 84.9 % of the variance. This factor is associated

Table 2. Seasonal variations in heavy metal contamination in the groundwater of study area

Difference	Mean	Lower CL*	Upper CL*	SD	SE(M)	t value	*df*	Sig.(2-tail)
Cd PRM-POM	0.002	0.001	0.003	0.003	0.001	4.87	49	0.000
Cr PRM-POM	0.079	0.062	0.096	0.060	0.009	9.27	49	0.000
Cu PRM-POM	0.089	0.079	0.099	0.034	0.005	18.29	49	0.000
Mn PRM-POM	0.098	0.061	0.135	0.131	0.019	5.29	49	0.000
Ni PRM-POM	0.175	0.160	0.190	0.052	0.007	23.74	49	0.000

*At 95% Confidence Level (CL).
SD = Standard Deviation, SE(M) = Standard Error Mean, *df* = degrees of freedom

Table 3. Principal component loadings for heavy metals in groundwater of sampling locations

Elements	Mean	
	PC-I	PC-II
Cd	-0.462	-0.327
Cr	-0.460	-0.362
Cu	-0.474	-0.073
Mn	-0.367	0.867
Ni	-0.465	0.071
Eigenvalues	4.247	0.542
% of Variance	84.9	10.8
Cumulative %	84.9	95.8

PC-I and PC-II depict principal components I+II

with effluents from industries consisting of chemicals, pesticides, pharmaceuticals, petroleum products, engineering, textiles and plastics located in the study sites. This factor association converges with the strong positive loads of Cd, Cr, Cu and Ni due to anthropogenic origin (Ramesh and Purvaja, 1995). Factor 2 comprising of Mn contributed to 10.8 % of the variance. This happens as a result of the natural presence of manganese in the study areas.

The HPI values were in the range of 59.3 to 1035.6 (mean 537.2) and 4.1 to 574.7 (mean 359.5) in the PRM and POM seasons, respectively (Table 4). There was marked reduction in HPI values

Table 4. Heavy metal pollution index (HPI) values for each sampling sites in PRM and POM seasons.

Sampling ID	PRM	POM	Mean	Sampling ID	PRM	POM	Mean
S1	573.0	483.9	528.5	S27	665.5	534.6	600.0
S2	87.9	6.8	47.4	S28	62.4	12.0	37.2
S3	698.7	534.6	616.7	S29	104.8	7.1	56.0
S4	686.6	534.7	610.6	S30	68.9	10.1	39.5
S5	623.2	529.0	576.1	S31	79.5	13.1	46.3
S6	593.6	493.6	543.6	S32	146.6	8.5	77.5
S7	751.3	526.4	638.9	S33	868.5	540.8	704.7
S8	540.3	425.5	482.9	S34	927.7	532.5	730.1
S9	970.4	545.5	757.9	S35	963.9	539.8	751.9
S10	1035.6	547.8	791.7	S36	658.8	534.6	596.7
S11	759.8	538.4	649.1	S37	490.1	411.1	450.6
S12	842.9	553.3	698.1	S38	143.9	4.1	74.0
S13	782.5	532.4	657.5	S39	140.1	4.1	72.1
S14	894.0	574.7	734.4	S40	142.5	4.4	73.5
S15	819.7	541.8	680.8	S41	590.4	495.0	542.7
S16	61.2	15.0	38.1	S42	722.0	534.9	628.5
S17	85.1	12.3	48.7	S43	724.1	531.8	628.0
S18	82.6	12.0	47.3	S44	700.7	536.3	618.5
S19	111.0	12.0	61.5	S45	760.0	535.9	648.0
S20	59.3	12.0	35.7	S46	810.7	574.2	692.4
S21	114.0	12.0	63.0	S47	643.2	528.1	585.7
S22	640.2	490.9	565.6	S48	843.0	541.6	692.3
S23	747.3	534.6	640.9	S49	645.2	533.5	589.4
S24	829.5	531.8	680.7	S50	81.9	7.9	44.9
S25	578.8	450.0	514.4	Mean	537.2	359.5	448.4
S26	907.4	548.3	727.9	SD	324.1	244.3	281.5

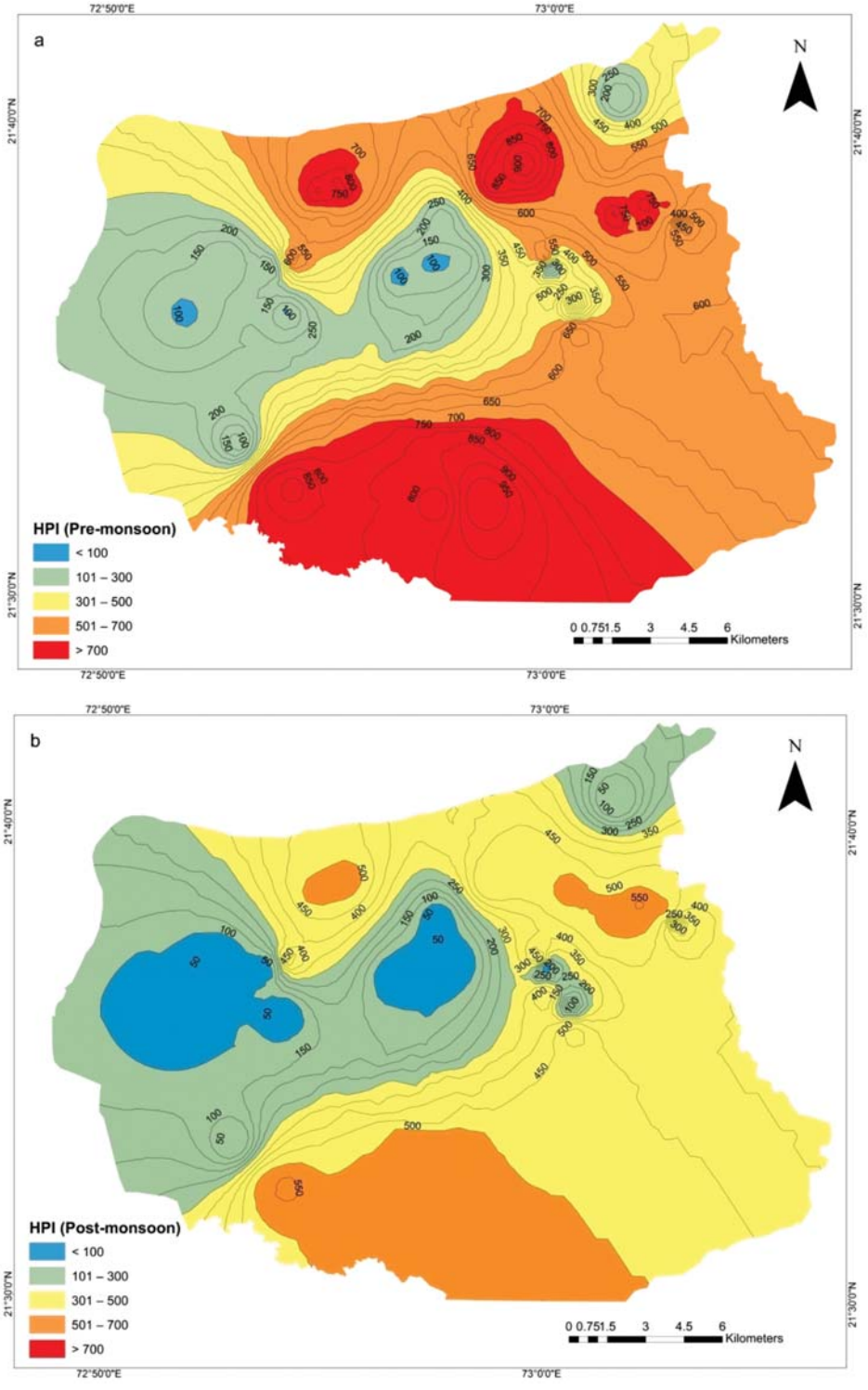

Fig. 2 Concentration contours showing spatial intensity of HPI in the study area during the PRM (a) and POM (b) seasons

from PRM season to POM season at all the sampling sites (Fig. 2a, b). High HPI values in the PRM season resulted from large number of industries coupled with indiscriminate disposal of not-so-well-treated effluents into the water flows. Low HPI values in the POM season are attributable to the dilution of heavy metals contained in the groundwater caused by rainfall. This is supported by the finding of Ramesh and Purvaja (1995) that heavy local precipitation dilutes the concentration of the trace elements. The critical value for HPI is 100 and obviously

any level of contamination above this value is unacceptable and makes water unfit for drinking. It is a significant finding that 82 % of sites in PRM season and 68 % of sites in POM season had HPI values above the critical value.

Conclusion

Findings emerged from the present study showed that. Cd content was above the acceptable levels for drinking water in both the PRM and POM seasons and showed less seasonal variability in groundwater in and around Ankleshwar and Panoli GIDC. Cu, Cr, Mn and Ni were found to be above the acceptable levels in the PRM season whereas these heavy metals were within the acceptable level in the POM season and showed considerable seasonal variability. HPI values indicated that 82 % of the samples in the PRM season and 68 % in the POM season were of poor quality water and unfit for drinking. These findings indicate that there is heavy contamination of groundwater due to the discharge of effluents from industries located in the region which calls for urgent measures to drastically reduce the contamination levels for the sake of restoration of public health.

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