Groundwater contamination with heavy metals in Chennai city, India – A threat to the human population

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Abstract. The goal of this research was to learn more about heavy metal pollution in groundwater in North Chennai during the pre-monsoon and post-monsoon seasons. The total no. of 108 groundwater samples were utilized for this study retrieved from both dug and bore wells. The analytical results indicate that all the determined trace metals show wide spatial and temporal variations. Spatial variation is mainly due to the different sources of contamination, and temporal variation is mainly due to the influence of rainfall and the associated changes in the hydrogeochemical conditions. The abundance order of trace elements based on average ionic concentration in pre-monsoon is Iron (Fe)> Manganese (Mn)> Chromium (Cr)> Copper (Cu)> Lead (Pb)> Nickel (Ni)> Zinc (Zn) and during post-monsoon Fe>Mn>Pb>Zn >Cr>Cu>Ni. Nemerow’s pollution index (NPI) gives an idea about the range of pollution for individual water quality parameters concerning a standard value. Our result showed that North Chennai city is polluted by Lead in both seasons.

1. Introduction

Due to the rock-water interaction, groundwater develops in the pores and fissures within geological formations, which are enriched with various ions, including major and trace elements [1] [2]. Drainage pattern, topography, geology, hydro-geomorphology, climatic conditions are the important factors that control the presence and flow of groundwater [3]. Although the behavior of trace elements in groundwater is not fully understood, the non-conservative characteristics of trace elements have drawn the attention of many researchers [4][5]. Groundwater is mainly used for irrigation, domestic and industrial needs based on its quality and quantity. Over the last two decades, the demand for groundwater increased drastically due to a decline in surface water resources, irrigation, climate change, industrialization, and urbanization. As a result, there is a significant decline in groundwater all over the world [6] [7][8].

The presence of heavy metals in groundwater poses a major hazard to the ecosystem. [9][10]. Although humans and animal’s bodies need certain heavy metals like Zn, Cu, and Fe in time of drinking and eating food that helps in the smooth running of the human body[11] [9]. When heavy metal concentrations exceed their threshold value, they become harmful to human health[12] [13] [14]. The high consumption of trace elements created toxicity whereas less consumption leads to nutritional deficits[15].
The heavy metals risk level affects human health that includes the occurrence of cancer, skin infections, birth imperfections, organ failure, and nervous system damage [16][17]. It enters the human body through direct absorption through food, drinking water, the air we breathe, and the skin[18].

The standard of water determines the health of the ecosystem [19][20]. Since water is a liquid it holds property to flow which permits the heavy metals in the aquatic environment to migrate easily and also for degradation, as a result, it spreads to other areas[21] [22]. The industries expel the effluents directly into the water bodies and this is the major threat to the organisms that reside there [23]. As a result, it's critical to identify the sources of heavy metal pollutants in water and to assess the pollutants so that a reference point for environmental healing and effective water management may be provided. Water contamination is primarily caused by humans[24].

Using principal correlation and component analysis, Nemerow’s pollution index [25] analyzed the heavy metal contamination and distinguished its source in different aquatic environments. Numerous attempts have been conducted to delineate the occurrence of trace metals in groundwater [26] [27]. However, there are limited studies have been carried out for assessing trace element contamination in Chennai City [28][29]. Since it adversely affects human health continuous monitoring of this contamination is required.

The rapid increase in the need for urban water in Chennai City developed because of its economic growth. This led to the pollution of water bodies due to wastewater discharge by various industries. The goal of this research is to investigate trace metal pollution in groundwater as well as the spatial distribution of heavy metals in the water system. In this study, we calculated the distribution of heavy metals such as Cobalt (Co), Chromium (Cr), Copper (Cu), Iron (Fe), Manganese (Mn), Nickel (Ni), Lead (Pb), and Zinc (Zn) in groundwater using a Nemerow’s pollution index (NPI).

2. Study Area
The investigation is taking place in the northern suburbs of Chennai. The region lies between the longitudes of 80°03ʹ and 80°17ʹ East and latitudes of 13°03ʹ and 13°16ʹ North, with a total area range of ~358.08 km² Figure 1. The climate in the research region is of the subtropical maritime monsoon type, which is consistent throughout the year. The highest temperature ranges from 37 to 44 degrees Celsius, while the minimum temperature ranges from 18 to 27 degrees Celsius. On average, the southwest monsoon (June–September) receives 35 percent rainfall, whereas the northeast monsoon (October–December) receives 60 percent rainfall of average approximately 1,200 mm/year [6]. The maximum elevation is about 10 meters and it is located in the western part of the study area and has a gradual slope towards the east above mean sea level (MSL). Geomorphology of the study area includes shallow to deep pediment and pediplain originated by denudation and the older deltaic plain originated by fluvial and coastal action and lakes also present. The geological range of the present study area is from Archean to Recent. Laterite and lateritic gravel, conglomerate, sandstone and claystone, medium to coarse sand with clay, sandy silt, black clay, and mud were the major litho units of the study area. Inceptisols are the widely distributed soils in the northern portion of Chennai City [30]. The area under examination has several small to large scale industries are present such as information technology parks, automobile companies, refineries, treatment plants, and dumping yards.

3. Material and Methods
Groundwater samples were obtained from 54 wells (38 bore wells + 16 dug wells) based on stratified random sampling. The exact coordinates of the samples were obtained by a GARMIN GPS (Global Positioning System). The samples were taken in June 2015 (premonsoon) and January 2016 (monsoon) because the NE monsoon is active from mid-October to mid-December (postmonsoon). A pre-calibrated digital pH meter (PCS tester 35) was used for immediate and accurate measurement of pH. With the help of flame Atomic Absorption Spectrometer (AAAnalyst 700 Perkin Elmer) the trace metal concentrations were calculated. For a better result, the instrument was calibrated using standard solutions (MERCK).
Before the measurement, the instrument was tested with standard materials for reference (SLRS Riverine Water, National Research Council, Canada).

The research area is the Puzhal watershed, drawn using DEM map that was downloaded from the Bhuvan website, and processed in ArcGIS 10.3 software using the Geo-Hydro tool. The SPSS software (SPSS, 2001) was utilized for statistical analysis. The results were compared to the Bureau of Indian Standards’ suggested levels (BIS, 2012) [31]. To calculate the extent of pollution Nemerow and Sumitomo (1970) introduced the Nemerow pollution index (NPI). Later several researchers have followed this index [32]. The NPI is a simplified pollution index and it is given as

\[ NPI = \frac{C_i}{L_i} \]

Where,
- \( C_i \) – Concentration of \( i^{th} \) parameter
- \( L_i \) – Permissible limit of \( i^{th} \) parameter

The value \( \leq 1 \) is considered as ideal Nemerow pollution index and the water quality standard determines the pollution level and is divided into four classes, that are 0< NPI< 1.0 - meeting standard quality (good condition), 1.0< NPI<5.0 - slight polluted, 5.0 < NPI< 10 - medium pollution and NPI> 10 - heavily polluted, see figure 1.

![Map of the study area with sampling locations](image)

**Figure 1** Map of the study area with sampling locations

### 4. Results and Discussion
4.1. Trace Elements

During pre-monsoon, pH value differs from 5.8 to 7.7 with a mean value of 6.96 ± 0.45 which indicates slightly acidic in nature. In post-monsoon time, pH value contrasts from 5.8 to 8.0 with an average of 7.19 ± 0.49 which indicates slight basic nature. According to BIS specified sample no (7, 9, 10, 14, 15, 17, 21, 46 and 49) total 17% of samples in pre-monsoon and sample no (9, 10, 14 and 15) total 10% of samples show below the permissible limit. A spatial distribution diagram of the variation of pH is shown in Figure 2. It is found that during both seasons, the groundwater pH was low near the north-western portion of the study area where more drinking water purifier companies are there. From the field observation, we found that area was exposed mostly laterite rock and low population density. Furthermore, the basin behaves acidic nature due to the presence of lateritic soil [33]. The use of fertilizers that produce acids like ammonium sulfate and superphosphate of lime as manure for agriculture could be a reason for the acidic nature of the soil [34], see figure 2.

![Figure 2 Spatial distribution map of pH](image)

The basic statistical values of the analytical results and suitability of water for drinking purpose is given in Table 1. The analytical outcomes indicate that all the determined trace metals show wide spatial and temporal variations. Spatial variation is mainly due to the different sources of contamination, and temporal variation is mainly due to the influence of rainfall and the associated changes in the hydrogeochemical conditions. The order of abundance of trace elements based on average ionic concentration in pre-monsoon is Fe>Mn>Cr>Cu>Pb>Ni> Zn and during post-monsoon Fe>Mn>Pb>Zn>Cr>Cu>Ni.

| Parameters | PRM AVG | MIN | MAX | POM AVG | MIN | MAX | BIS 2012 | WHO 2004 |
|------------|---------|-----|-----|---------|-----|-----|----------|----------|
| pH         | 7.0     | 5.8 | 7.7 | 7.2     | 5.8 | 8   | 6.5-8.5  | 7-9.2    |
| Fe         | 0.630   | 0.102 | 6.268 | 0.546 | 0.019 | 8.855 | 0.3       | 0.3      |
| Mn         | 0.478   | 0.006 | 7.13 | 0.518 | 0.001 | 5.063 | 0.1-0.3  | 0.05     |
| Cu         | 0.142   | 0.028 | 0.395 | 0.042 | 0.010 | 0.419 | 0.05     | 0.005    |
| Ni         | 0.082   | 0.014 | 0.22 | 0.025 | 0.000 | 0.113 | 0.02     | 0.02     |
| Pb         | 0.163   | 0.065 | 0.423 | 0.168 | 0.005 | 0.337 | 0.01     | 0.01     |
Zn 0.048 0.001 1.014 0.062 0.002 0.458 5.0-15.0 3
Cr 0.262 0.019 0.539 0.052 0.009 0.309 0.05

Pre-monsoon iron concentrations range from 0.102 to 6.268 mg/l with a mean of 0.72 0.898 mg/l, whereas post-monsoon iron concentrations range from 0.019 to 8.855 mg/l with a mean of 0.685 1.317 mg/l. The iron concentration is higher in the northwestern part due to agricultural activity and domestic sewage; the southern part is polluted by leaching from industrial and domestic sewage Figure 3. The dominancy of iron was noticed in both the seasons and the source may be laterite. Iron above 0.3 mg/l is understood to cause staining of garments and utensils. The better attention of iron is also no longer appropriate for the processing of food, liquids, ice, bleaching, dyeing, and plenty of other activities. The most common form of iron in groundwater is ferric hydroxide, which has a typical concentration of 0.5 mg/L. The lower iron concentration leads to diseases called "anemia" and excessive concentration of iron might cause a liver disease known as "haemosiderosis" [35].

Manganese levels range between 0.006 and 7.13 mg/l during the pre-monsoon season, with an average of 0.59±1.13mg/l. Mn concentrations range from 0.0014 to 5.063 mg/l in post-monsoon seasons, with a mean value of 0.59±1.0289 mg/l. The contour diagrams Figure 3 of manganese of both monsoons illustrate higher values in the north-eastern region due to agricultural and domestic effluents; the southern part is affected by domestic and industrial effluents. The variation diagrams indicate that there is not much seasonal impact on the study area, which indicates it is mostly of geogenic origin, see figure 3.
Figure 3 Spatial distribution map of iron and Manganese

Lead concentrations range from 0.065 to 0.423 mg/l during the pre-monsoon period, with a mean value of 0.166±0.059 mg/l. Pre-monsoon indicates the increase in the concentration of lead in the north-eastern part due to the influence of agricultural practice; central and southern parts are nearby waste disposal sites, close to the industrial area, and close to roadsides and Pb could be leached from waste disposal sites and come from atmospheric fallout Figure 4. Post-monsoon values of Pb range between 0.005 to 0.337 mg/l in the groundwater samples and the mean is 0.168 ± 0.08 mg/l. The post-monsoon data is suggesting irregular scattering of lead suggesting different sources Figure 4. The heavy rain may wash the paint from buildings, bridges, roads, petroleum products from garages, bus stops, agricultural land, urban runoff close to roadsides indicating leaching from waste disposal sites and atmospheric deposition. It also reported that the presence of lead in natural waters was unusual because its sulfide and carbonate (available forms of lead) forms were highly insoluble in water [36]. In pre-monsoon lead was the fifth most abundant metal whereas during post-monsoon it is the third most abundant metal which indicates heavy precipitation and the urban flood is the main cause of increased lead concentration in the northern part of Chennai city. The urban flood which occurred during December 2015 in Chennai city would have increased lead concentration in groundwater by various miscellaneous sources [37][38]. Pb is particularly carcinogenic as well as poisonous metal which creates continued health hazards such as irritability, headache, anemia, damaging the brain, kidney, and liver, blood pressure, vomiting, stomach, and lung cancer [38], see figure 4.
In pre-monsoon data the copper ranging from 0.028 to 0.395 mg/l with a mean of 0.14±0.07 mg/l whereas the post-monsoon values vary between 0.0103 to 0.419 mg/l with a mean of 0.0478±0.0568 mg/l. The variation diagram Figure 5 illustrates during post-monsoon almost all samples show below the desirable limit and in pre-monsoon samples show below the permissible limit which indicates the impact of heavy rain reduced the copper concentration in post-monsoon. The amount of copper is maximum occurs in the southwestern portion of the present study which is mostly covered by industrial and residential areas.

During pre-monsoon, the estimated zinc concentration is 0.001 to 1.014 mg/l with a mean of 0.065±0.143 mg/l. Zinc values in post-monsoon fluctuates from 0.0023 to 0.4582 mg/l with a mean of 0.0677±0.0939 mg/l. Although the variation diagram Figure 5 explains all the samples of both monsoons are within desirable limits of (WHO and BIS) standards but during the post-monsoon central part of the area under study slightly increases compared to pre-monsoon [39] [31]. The distribution diagram indicates middle and southern parts of the study area show significantly higher value, wells located near the plating and fabricating industries, see figure 5.
During pre-monsoon, the concentrations of nickel alter 0.014 to 0.22 with a mean value of 0.08±0.047 mg/l. In post-monsoon, the nickel value fluctuates from 0.003 to 0.113 with a mean of 0.025±0.027 mg/l. The higher value of nickel occurs in the south and central portion of the area under study which may be contaminated by domestic and industrial effluents Figure 6. The variation diagram Figure 6 indicates almost all samples from both seasons fall above permissible level, but during post-monsoon, its concentration drastically decreases due to dilution. The higher concentration observed in the study area may be an atmospheric accumulation of nickel from phosphate fertilizers, combustion of diesel oil and fuel oil, industrial emissions, automobile emissions, emissions from electric power utilities, the incineration of waste and disposal of sewage sludge as well as, from miscellaneous sources [40] [41][42].

During pre-monsoon the Cr concentration is 0.019 to 0.539 mg/l with a mean 0.26±0.149 mg/l. Spatial distribution diagram Figure 6 during pre-monsoon signifying higher concentration of chromium occur Ambattur and Avadi industrial area which located southwestern side of the investigated area. The value of Cr during post-monsoon alters from 0.009 to 0.309 mg/l with a mean value of 0.052±0.043 mg/l. The distributional diagram Figure 6 indicates that the Cr values peaks in post-monsoon occur in the NE portion area under study. The economic use of chromium (Cr) and its compounds are manufacturing metallic and other alloys, and pigment manufacturing[43]. According to the drinking water standard of WHO and BIS, the tolerable limit of chromium is 0.05 mg/l.
4.2. pH vs Total Metal Load (TML)

To classify types of groundwater the method based on pH and the total metal load of Ficklin [44] and revised by Caboi [45] was applied. The water pH and metal load (mg/l) are used to classify the metal load in the water. The metal load in the present study is calculated as Fe+Mn+Cu+Pb+Ni+Zn+Cr+Co. From the chart Figure 7, during pre-monsoon surface water samples from 18 locations representing 33% of all the samples are categorized as near neutral-low metal and 36 locations 67% of total samples as near neutral-high metal. During post-monsoon surface water samples from 37 locations representing 69% of all the samples are categorized as near neutral-low metal and 17 locations (31% of total samples) as near neutral-high metal load.

4.3. Correlation Matrix

The physicochemical characteristic analysis was done on all samples. It showed the changes in the water quality and also the water quality standards. In addition, the variations in the concentration of each parameter are more significant. The relation among the two variables is the correlation coefficient and is normally represented by the English alphabet ‘r’ and it shows the strength of the relationship among the two variables [46]. Liu et al. classified factor loadings as 0.3 represented as weak, 0.5 to 0.75 marked as moderate and >0.75 noticed as strong [47]. During pre-monsoon, there is also a strong correlation between Cu-Pb, Cu-Cr, Ni-Pb, Ni-Cr, Pb-Cr, moderate correlation between Mn-Ni, Cu-Ni. In the case of post-monsoon there is a moderate correlation between Fe-Mn, Mn-Ni, Mn-Cr, Cu-Zn illustrated in Table 2 and Table 3. Metals having a positive correlation indicates the same source of origin. During post-monsoon Pb having a negative correlation with other metals indicate the source is different. A decrease in the value of the correlation coefficient of heavy metal pairs was observed during the post-monsoon. This is due to the dissolution of metals and minerals in the groundwater not from external sources and similar observations reported by [12]. The weak negative correlation is an indication that pH plays a role in the release of metals [48], see figure 7.
4.4. Factor Analysis (FA)

It uses the principal component extraction technique with varimax rotation. FA was applied separately to the hydrochemical data set about the pre and post-monsoons duration. The results of FA are given in table 4. In the study area during pre-monsoon Factor-1 is signified by strongly load by Cu, Pb, Cr, and Ni with a total variance of 43.39%. These metals are widely used usually in industries paint industries, industrial smelting operations, and electroplating. These industries expel wastewater which holds a certain amount of metals apart from the combustion of fossil fuels, and the important outside source fertilizers from the agricultural areas [49][50] [10][51]. This indicates that industrial, agricultural, and urban pollution are significantly responsible for F-1. Factor-2 explains 19.04% of the total variance (Tv) with strongly loaded by Fe and Mn that indicates weathering activity. Factor-3 accounts for 12.97% of the Tv has moderately loaded by Zn and weakly loaded Fe. In different types of rock-forming minerals Iron generally occurs [52]. Zinc coexists with iron as these are having comparable geochemical affinities [53]. During post-monsoon Factor-1 is characterized by strong loadings on Mn and moderate loads on Ni, Zn & Cr and weak loads on Fe & Cu with a total variance of 27.30%. Positive loading of Mn and Fe indicates weathering activity but the presence of trace elements influences urban sewage pollution. Iron and manganese are mostly formed in reducing the environment. Precipitation of Iron and Manganese oxyhydroxide are capable to adsorb metals that lead to metal accumulation in groundwater [54]. High concentrations of Zn, Cu, and Cr are positive correlation indicates source may be roadside dust reported by Ng et al. [55]. Factor-2 explains 19.96% of the total variance was moderately loaded by pH and weakly loaded Mn, Ni, and Cr. Factor-3 explains 15.02% of the total variance was moderately loaded by Fe and weakly loaded Cu, see table 4.

Table 2 Correlation matrix among trace metals in groundwater during PRM (2015)

| Parameters | pH | Fe | Mn | Cu | Ni | Pb | Zn | Cr |
|------------|----|----|----|----|----|----|----|----|
| pH         | 1.00 |    |    |    |    |    |    |    |
| Fe         | -0.12 | 1.00 |    |    |    |    |    |    |
### Table 3: Correlation matrix among trace metals in groundwater during POM (2016)

|       | pH  | Fe   | Mn  | Cu  | Ni  | Pb  | Zn  | Cr  |
|-------|-----|------|-----|-----|-----|-----|-----|-----|
| pH    | 1.00|      |     |     |     |     |     |     |
| Fe    | 0.04| 1.00 |     |     |     |     |     |     |
| Mn    | -0.08| 0.32 | 1.00|     |     |     |     |     |
| Cu    | -0.18| 0.24 | 0.06| 1.00|     |     |     |     |
| Ni    | 0.14 | 0.18 | 0.42| -0.05| 1.00|     |     |     |
| Pb    | -0.21| -0.19| -0.05| 0.15| 0.04| 1.00|     |     |
| Zn    | -0.23| 0.15 | 0.23| 0.45 | 0.29| 0.18| 1.00|     |
| Cr    | -0.04| 0.06 | 0.56| -0.04| 0.26| 0.03| 0.17| 1.00|

### Table 4: Varimax rotated principal component analysis for groundwater samples during PRM and POM

| Parameters | PRM | F1  | F2  | F3  | POM | F1  | F2  | F3  |
|------------|-----|-----|-----|-----|-----|-----|-----|-----|
| pH         | .176| -.678| -.027| -0.18| 0.60| 0.23|     |     |
| Fe         | .227| .609 | .312 | 0.47 | 0.14| 0.68|     |     |
| Mn         | .203| .828 | -.143| 0.79 | 0.31| -.11|     |     |
| Cu         | .908| -.032| .149 | 0.37 | -0.64| 0.44|     |     |
| Ni         | .834| .304 | -.094| 0.61 | 0.33| -.13|     |     |
| Pb         | .891| .095 | .002 | 0.10 | -0.54| -0.52|     |     |
| Zn         | .074| .040 | .952 | 0.64 | -0.47| 0.07|     |     |
| Cr         | .939| -.073| .163 | 0.62 | 0.30| -.43|     |     |
| Total      | 3.47| 1.52 | 1.04 | 2.18 | 1.60| 1.20|     |     |
| % of Variance | 43.39| 19.04| 12.97 | 27.30 | 19.96| 15.02|     |     |
| Cumulative % | 43.39| 62.43| 75.40 | 27.30 | 47.26| 62.29|     |     |

4.5. *Nemerow’s pollution index*

NPI gives figures about the range of pollution for individual water quality parameters concerning its standard value. NPI values also help to identify pollutants region; which is a piece of vital evidence concerning worsening water quality of the area. According to NPI values in Chennai city, Figure 8 and Figure 9 illustrate lead is heavily polluted both sampling periods.
5. Conclusion:
It is found that during both seasons, low pH is occurring near the north-western part of the area under investigation. The use of fertilizers that produces acids like ammonium sulfate and superphosphate of lime as manure for agriculture could be a reason for the acidic nature of the soil. The trace elements abundance is in the order based on average ionic concentration in pre-monsoon is Fe>Mn>Cr>Cu>Pb>Ni>Zn and
during post-monsoon Fe>Mn>Pb>Zn>Cr>Cu>Ni. It is found that the Copper, Lead, Chromium, and Nickel are observed to be high after Fe in the groundwater samples which may be due to industrialization of the area and improper disposal of wastewater. The pH plays a minimal role in the release of metals was indicated by weak negative correlation. The present study concludes that groundwater samples are heavily polluted by Lead in both the season hence proper monitoring of the groundwater, measures to reduce contamination of Pb in groundwater of North Chennai.

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