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Assessment of sources and distribution metals in groundwater of Pondicherry region, India

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Abstract: The proposed study investigates the seasonal variation in the concentration and the source of the heavy metals like Mn, Cu, Pb, Zn and Fe in the groundwater samples of Pondicherry region. The study results reveal that, the heavy metal concentration is high during South West Monsoon season (SWM) compared to that of North East Monsoon season (NEM). The pH was near neutral and metal load representing most of the samples were low during NEM. Statistical analysis shows that the 63.7 % of the total variance is observed during NEM and 68.9% during SWM. Geographic information system (GIS) tool was considered for the study to understand the environmental pollution status of the groundwater systems of the study area and to identify the groundwater quality parameters. The multivariate statistical analysis explains that the source of trace metal in the groundwater is derived from natural origin except copper and lead as these contaminants were derived from anthropogenic activities. Based on the output of WATEQ4F, several species of heavy metals exist, in which the dominant species are Mn, $CuCl_2$, $PbCO_3$, Fe and Zn.

Keywords: Heavy metals, Groundwater, Statistical analysis, GIS

1. Introduction

Groundwater is a significant water resource for the drinking, agricultural and industrial sectors [1-3]. Human activities have increased the concentration of heavy metals in the groundwater like industry, agriculture return flow, and solid waste disposal [4,5]. Dissolved trace elements play a vital role in monitoring the groundwater quality content to evaluate the hazard of metal pollution towards community health [6]. Usually the trace metal concentration in groundwater is based on the natural factors like type of the aquifer, infiltrating rate, mineral weathering in the aquifers and residence time. The anthropogenic sources like landuse infiltration and contamination of soils, industrial effluents and manure dumping and polluted surface water-groundwater interaction. Such relations often result in groundwater pollution and ingestion of contaminated water [7,8]. Pondicherry region is depending only on the groundwater for drinking, irrigation, and agriculture practices and also the Pondicherry is known for the existence of industrial estates. This region is also affected by seawater intrusion. A number of previous studies in groundwater had been carried out in the proposed region, on groundwater potentiality, its quality determination and recharge [9-12], Subsurface investigation [13], Geomorphological examination [14], Saturation index analysis [15], Isotopic research and microbial contamination by [16,17], Groundwater budgeting [18], Dissolved organic carbon studies in groundwater accompanied by water level and rainfall to categorize the seasonal variation [19-21]. Geological formation and excessive mining activities leads high concentration of heavy metal in the groundwater of this region. The main motto of this study is to compute the heavy metal contamination, in the groundwater of the proposed region and determine its suitability for drinking water purpose. The study further aims to explore the spatial unpredictability of trace elements and other physico-chemical parameters in the groundwater of the study area.

1.1 Study area

The proposed area is in the east coast of India forming enclaves within the Cuddalore district of Tamilnadu.

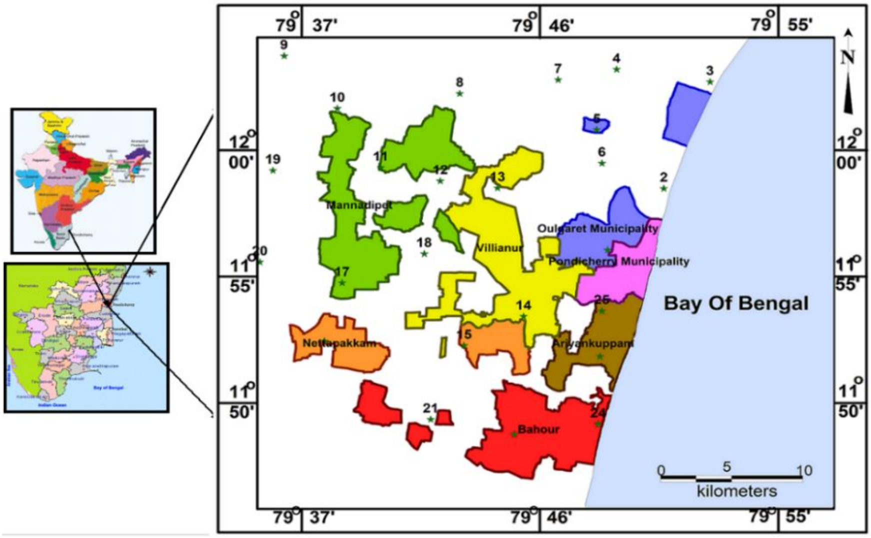

Figure 1. Sampling Location and Communes map of the study area

It is bounded by north latitudes 11° 45' and 12° 03' and east longitudes 79° 37' and 79° 53' and forms parts of survey of India topographical maps Nos. 58 M/9, M/13 and 57 P/12 and P/16. Pondicherry region encompasses 293 km^2 areas with seven communes (Figure 1) consisting of a total 179 villages. The study area is intersected by the deltaic channel of River Gingee and Pennaiyar. The Pondicherry region is covered by sedimentary formations, ranging in age from cretaceous to recent. In general, this region is a flat peneplain with an average elevation of about 15m above msl (mean sea level). Three major physiographic units are generally observed viz., i) Coastal plain, ii) Alluvial plain and iii) Uplands. The climate of Pondicherry region is humid and tropical. The mean monthly temperature ranges between 22 and 33 degrees Celsius. It receives an annual rainfall of 1281 mm. The position of groundwater table varies from 1.5 to 27 meters BGL [22,23]. The Pondicherry region is basically an agriculture region and the land used for crop production is around 56%. The Pondicherry is also known for the presence of industrial estates at Periyakalapet, Sedarapet, Karasur, Mudaliarpet and Mettupalayam. The general strike of the Cretaceous and Tertiary formations trends NE-SW with gentle dip. The Cuddalore formations though maintain the same strike shows a dip up to 10^0.

2. Methods and materials

A total of 50 groundwater samples were collected during Northeast Monsoon (NEM) and Southwest Monsoon (SWM) (Fig 1). One liter of water sample was collected in a polyethylene bottle and the samples were filtered using 0.45 µm membrane filter in field. Samples were sealed and brought to the laboratory and stored properly at 4°C until analysis. The field observation parameters like pH, EC TDS, Temperature and Alkalinity were measured in-situ by using the standard procedures [24]. The collected samples were analyzed for major cations, anions using standard procedures [24]. Samples were collected in 500ml polythene bottles for metal analysis. Prior to sampling, the bottles were rinsed with the water to be sampled and the samples were sealed by acidifying to pH ~ 2 with HNO_3 and kept back at a temperature of 4 °C until analysis. The samples were then spiked with the normal solution of the metals chosen for analysis. The collected water samples were filtered using a pre-conditioned plastic Millipore filter unit equipped with a 0.45-µm filter membrane for further elemental analysis. The metal ion concentrations were then measured using inductively coupled plasma mass spectrometry (ICPMS) (ICP-MS: Agilent Technologies 7700 series). Multivariate analysis was carried out by using SPSS Ver.10 software, [25] which describes the dispersion of the multiple measured parameters to obtain Eigen values and Eigen vectors.

3. Results and Discussion:

3.1 Geo chemical parameters study

The groundwater of the study area is illustrating by a broad variety of physicochemical parameters values Table 1 demonstrates some fundamental physicochemical parameters of groundwater during NEM and SWM. The order of dominance for major ion concentrations during NEM was Na>Ca>K>Mg and Cl> HCO_3>NO_3>PO_4>SO_4 and SWM was Na>K>Ca>Mg and HCO_3>Cl>NO_3>PO_4>SO_4. The predominant ions in the groundwater during NEM was Na and Cl with the average values of 184 and 359 mg/l and during SWM, Na and HCO_3 with the average values of 209 and 349 mg/l respectively.

Table 1. Minimum, Maximum, and Average concentration of physico chemical parameters in milligram per litre. (Except Temperature in °c, EC in µS/cm and pH)

		Ca	Mg	Na	K	Cl	HCO_3	SO_4	NO_3	PO_4	SIO_2	TDS	EC	pH	Temp
NEM (n=25)	Max	228.000	48.000	561.000	80.000	914.300	390.400	0.700	65.090	8.750	229.000	2309.000	3630.000	7.920	31.800
	Min	20.000	4.800	13.100	0.600	53.175	61.000	BDL	BDL	BDL	10.600	310.225	469.102	6.730	26.500
	Avg	101.407	15.733	184.270	24.896	358.740	272.104	0.369	16.500	0.928	125.193	1151.542	1805.992	7.367	29.541
SWM (n=25)	Max	104.000	62.400	360.100	247.800	514.025	597.800	0.990	184.160	4.450	229.000	1773.335	2343.000	9.500	33.400
	Min	40.000	2.400	58.500	4.700	106.350	73.200	0.130	BDL	BDL	47.000	494.205	751.000	5.430	29.100
	Avg	59.333	26.119	208.789	45.519	257.155	348.652	0.541	22.376	0.574	129.556	1091.747	1503.714	7.014	30.883

Electrical Conductivity is one of the important indices of water quality, which gives the measure of the dissolved salts and salinity. Higher EC in the water samples reflects leaching or dissolution of the aquifer material or mixing of saline water0. The groundwater samples have EC values during NEM ranges between 469 and 3630µS/cm (Table 1), during POM it ranges between 751 µS/cm and 2343 µS/cm. Comparison of the EC values with standard shows that a most of the samples were above the permissible limit.

The groundwater quality equivalent to freshwater, with lesser EC values during NEM were noted in the South eastern part of the study area (Figure 2). Higher EC concentration during NEM is observed in North Eastern part of the study region.

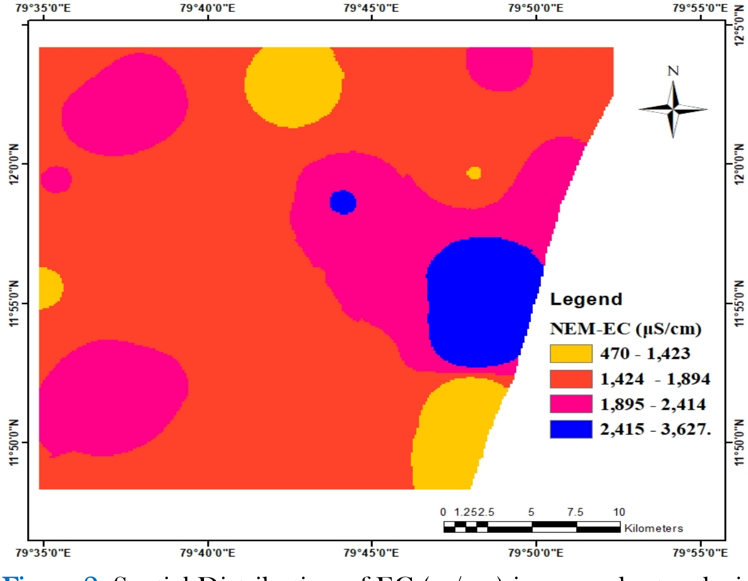

Figure 2. Spatial Distribution of EC (µs/cm) in groundwater during NEM

The monitoring wells with more than 3000 µS/cm indicates that the groundwater is highly mineralized /contaminated with saline intrusion. Higher EC values observed during SWM (Figure 3) in many hot spots in the study region.

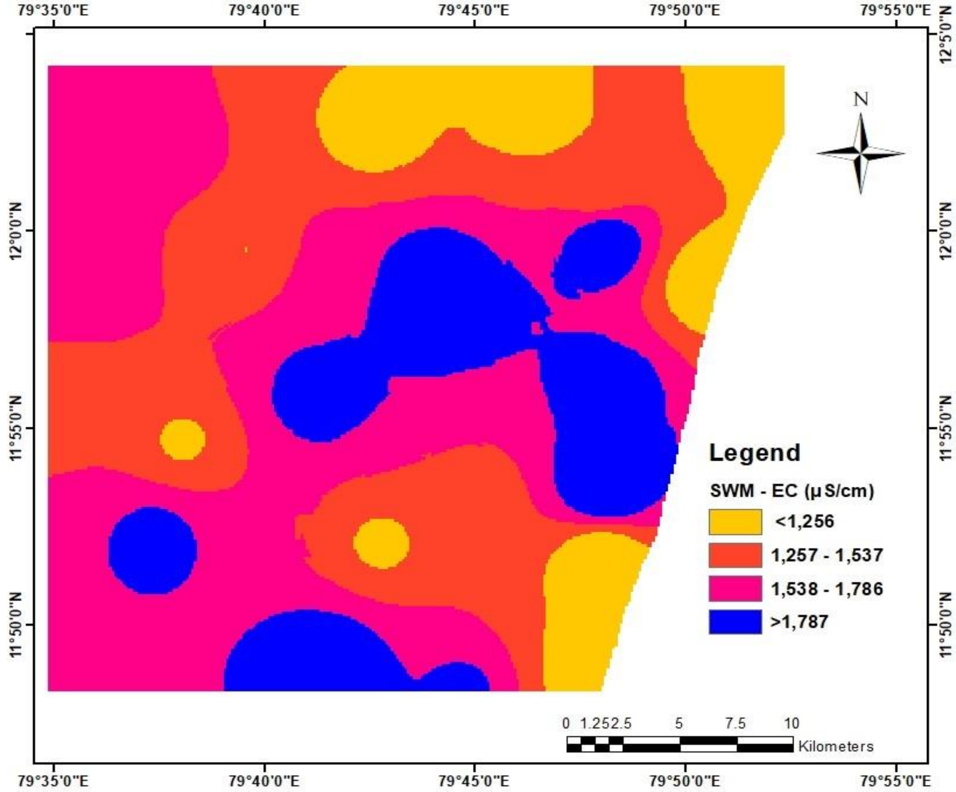

Figure 3. Spatial Distribution of EC (µs/cm) in groundwater during SWM

The groundwater (Figure 4) has dominant representation of Na-Cl and Ca-Mg-Cl facies. Most of the samples are of Na-Cl type, has high sodium and chloride may be due to the process of removal of other ions from the system either by adsorption or by precipitation due to the saturation or, they can also be due to seawater intrusion [10]. Migration of the samples from transition zone of mixed Ca- Mg-Cl and Na-Cl facies towards the sea water composition as few samples represent Ca-Mg-Cl facies, the groundwater in this region may also have influenced by seawater intrusion. This depicts the impact of seawater intrusion due to the dominance of the facies of both Na and Cl. The seawater composition is due to the diminishment in precipitation, depletion of water level with additional exploitation of groundwater during NEM.

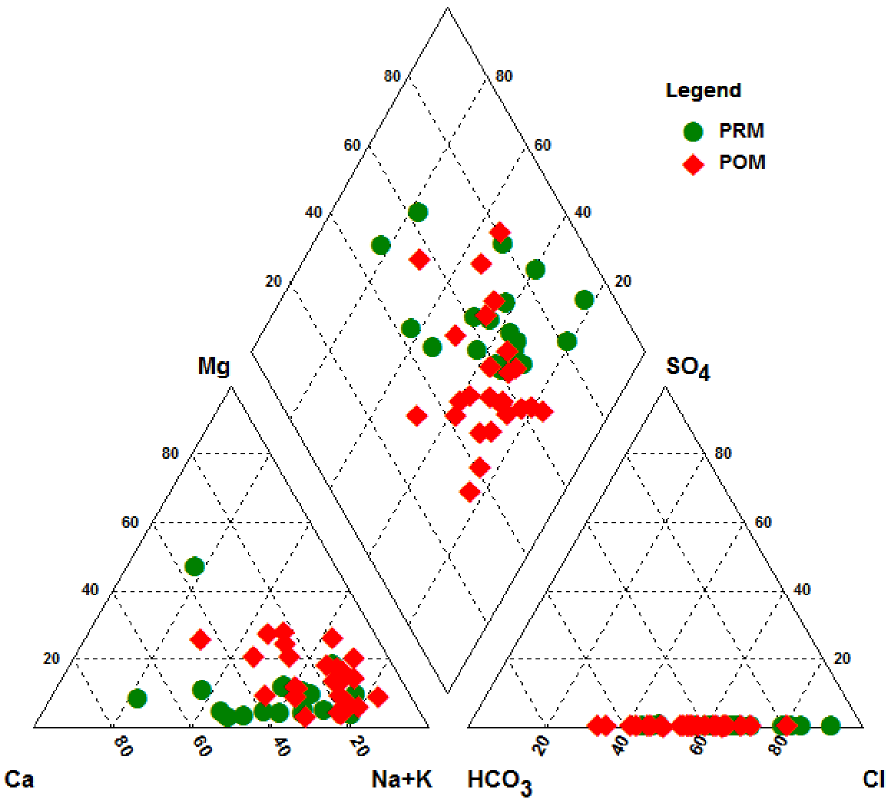

Figure 4. The distribution of major ions using a piper plot and the determination of water type in groundwater

3.2 Heavy metal concentration

The statistical information of measure heavy metal in provided in Table 2. The mean concentration of Mn , Cu, Pb, Zn and Fe of all the samples in NEM season was found as 0.37, 0.003, 0.08, 0.53, 0.38 mg/l and SWM season was establish as 0.06, 0.013, 0.014, 0.13, 0.48 mg/l respectively. The concentrations of the metals Cu, Pb, Fe during NEM were lower as compared to SWM. that the Mn, Pb, Zn and Fe were higher in concentration with respect to [28-30] limit during SWM.

The concentrations of Pb ranged from 0.01 to 0.18 mg/l in the NEM season and BDL to 0.29mg/l in the SWM season, 96 and 8% of samples from both the seasons exceeding the desirable limit of 0.01 mg/l (Table 2). The Pb concentration in the study area is confined to the southern part of the study area during NEM and southeastern part during SWM (Fig 5a,b). Higher concentration is due to the effect of agriculture return flow and other man-made activities. The concentration of the Fe of 36% and 24% of NEM and SWM exceeding the desirable limit of 0.3 mg/l. Higher Fe in water is noted most of the part of the study area (Fig 6a,b) due to the abundance of iron bearing minerals [31]. Fe in the region is due to the Weathering and rock-water interaction to the iron bearing litho-units. Earlier studies by [32,33] signify that mining activities release Fe in the environment. During NEM Higher concentration of the Fe is observed in North East and western and South Eastern part of the study area (Fig 6a,b). Higher concentration during SWM is observed in North Eastern part of the study area. The concentration of Zn in the study area ranged from 0.029 to 6.6 mg/l during NEM and BDL to 2.23 mg/l in SWM. it illustrates the broad difference in the contents. In few locations higher concentration of Zn was observed. Generally, both the seasons higher concentration is noted in the central part of the study area (Fig 7a,b). It is due to the industrial effluents and agricultural influence which is predominant in this region.

Table 2. Seasonal variation in metal concentration in the groundwater (all values in mg/l)

Mean	0.370	0.003	0.081	0.536	0.382
Std. Deviation	0.849	0.002	0.045	1.379	0.517
SWM	Mn	Cu	Pb	Zn	Fe
Minimum	BDL	BDL	BDL	BDL	BDL
Maximum	0.37	0.046	0.29	2.29	5.02
Mean	0.064	0.013	0.014	0.133	0.48
Std. Deviation	0.091	0.013	0.059	0.45	1.09
USEPA (2009)	0.05	1.3	0.015	5.0	0.30
WHO (2006)	0.1	2.0	0.01	4.0	0.30
BIS (2003)	0.1	0.05	0.01	5.0	0.30

The concentration of Mn of samples in the NEM and SWM is BDL to 3.7 and BDL to 0.37 respectively. The Manganese associate with complex sulphide minerals present in sedimentary rocks [31]. Higher level of Mn in the groundwater are suitable to the geological formation of the region [34]. During NEM higher concentration of Mn is noted in the South eastern part of the study region and during SWM higher level of Mn is noted in the southern part of the region (Figure 8a,b). The Copper level in the groundwater is ranged from BDL to 0.008 mg/l in NEM and BDL to 0.046 mg/l in SWM. All the samples are within the permissible limit of standards. Maximum concentration during NEM is noted in the northern region of the study area and central region shows the maximum level of Cu in the study area in SWM (Fig 9a, b).

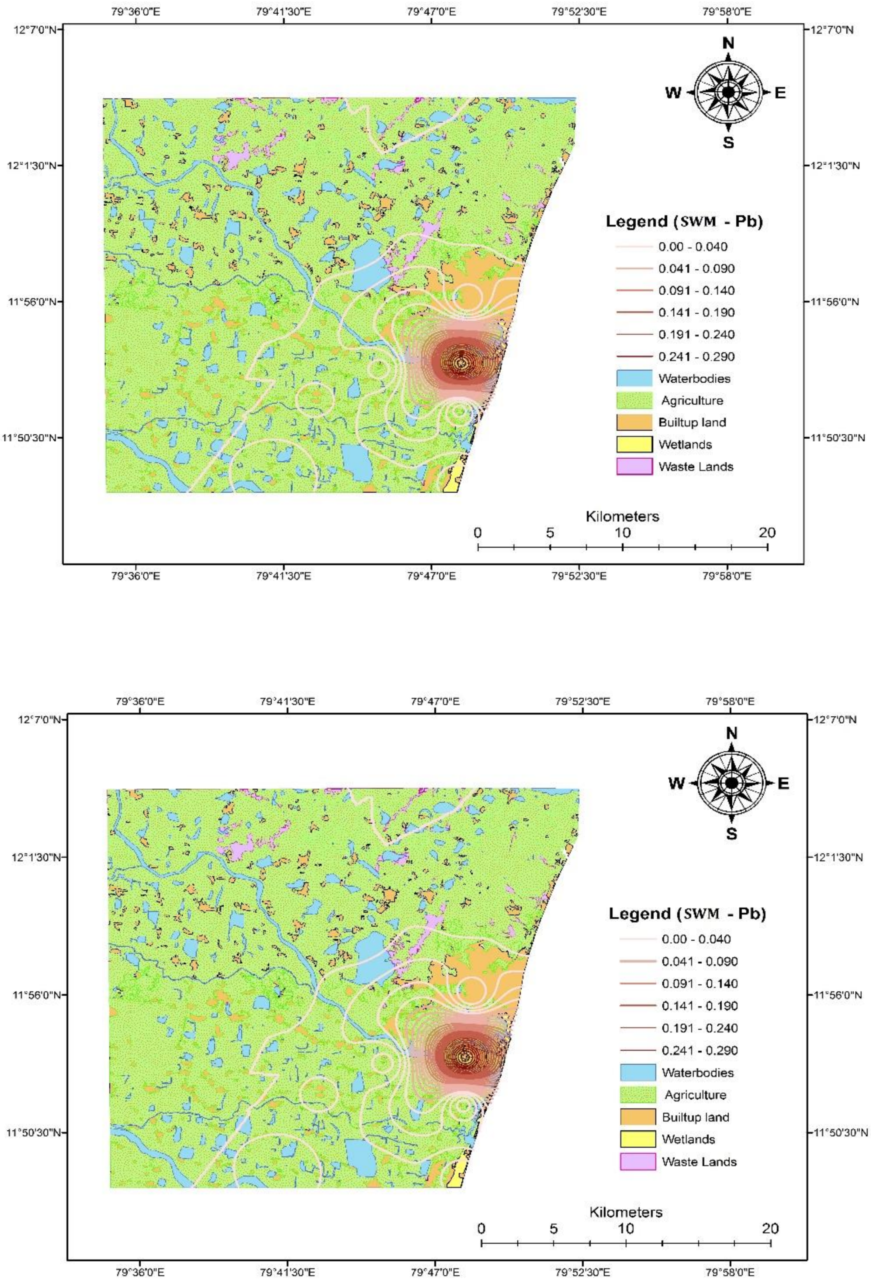

Figure 5. Concentration contour showing spatial distribution for Pb in groundwater throughout the study area in the a NEM, b SWM season overlaid on the landuse (all values in mg/l).

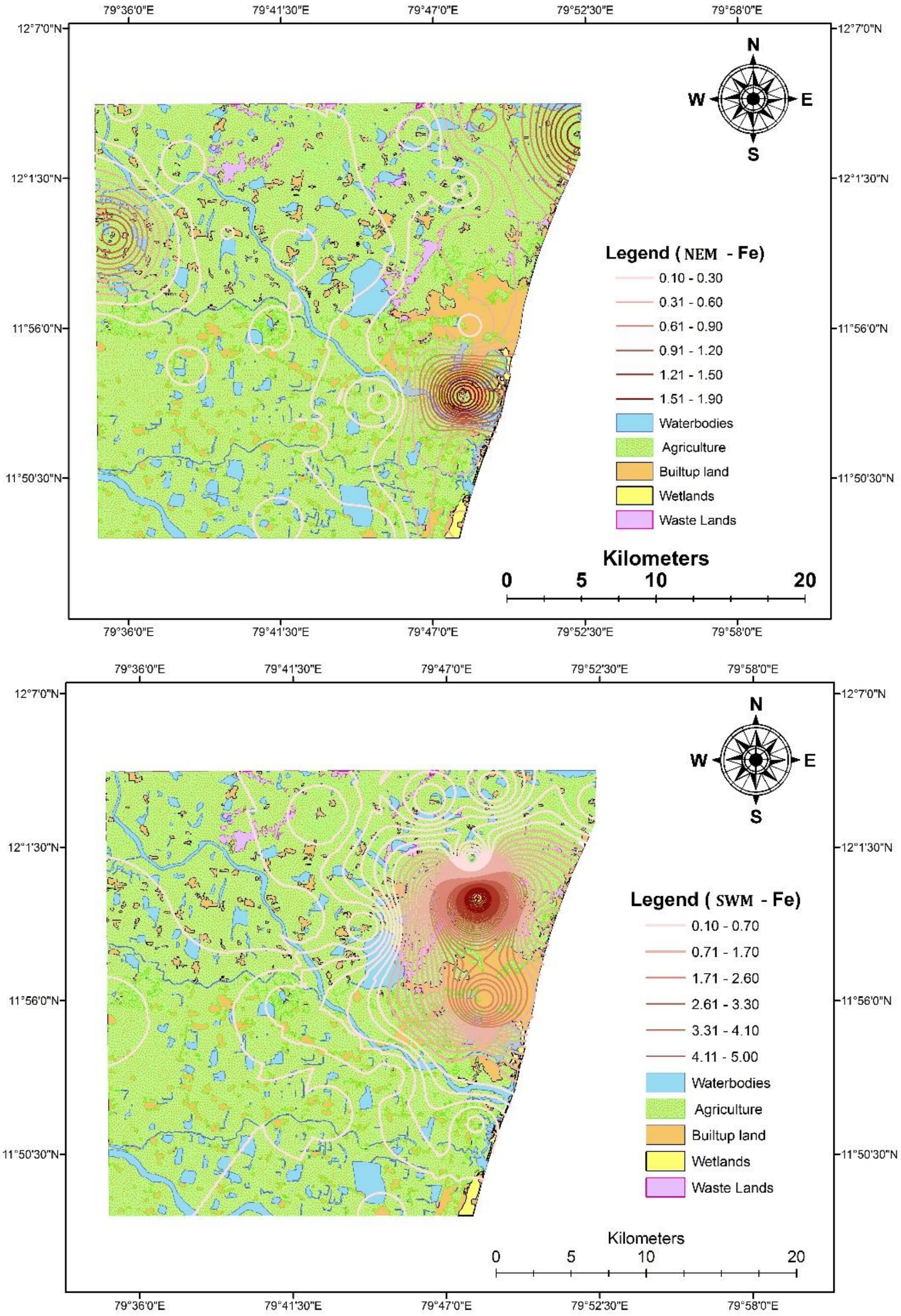

Figure 6. Concentration contour showing spatial distribution for Fe in groundwater throughout the study area in the a NEM, b SWM season overlaid on the landuse (all values in mg/l).

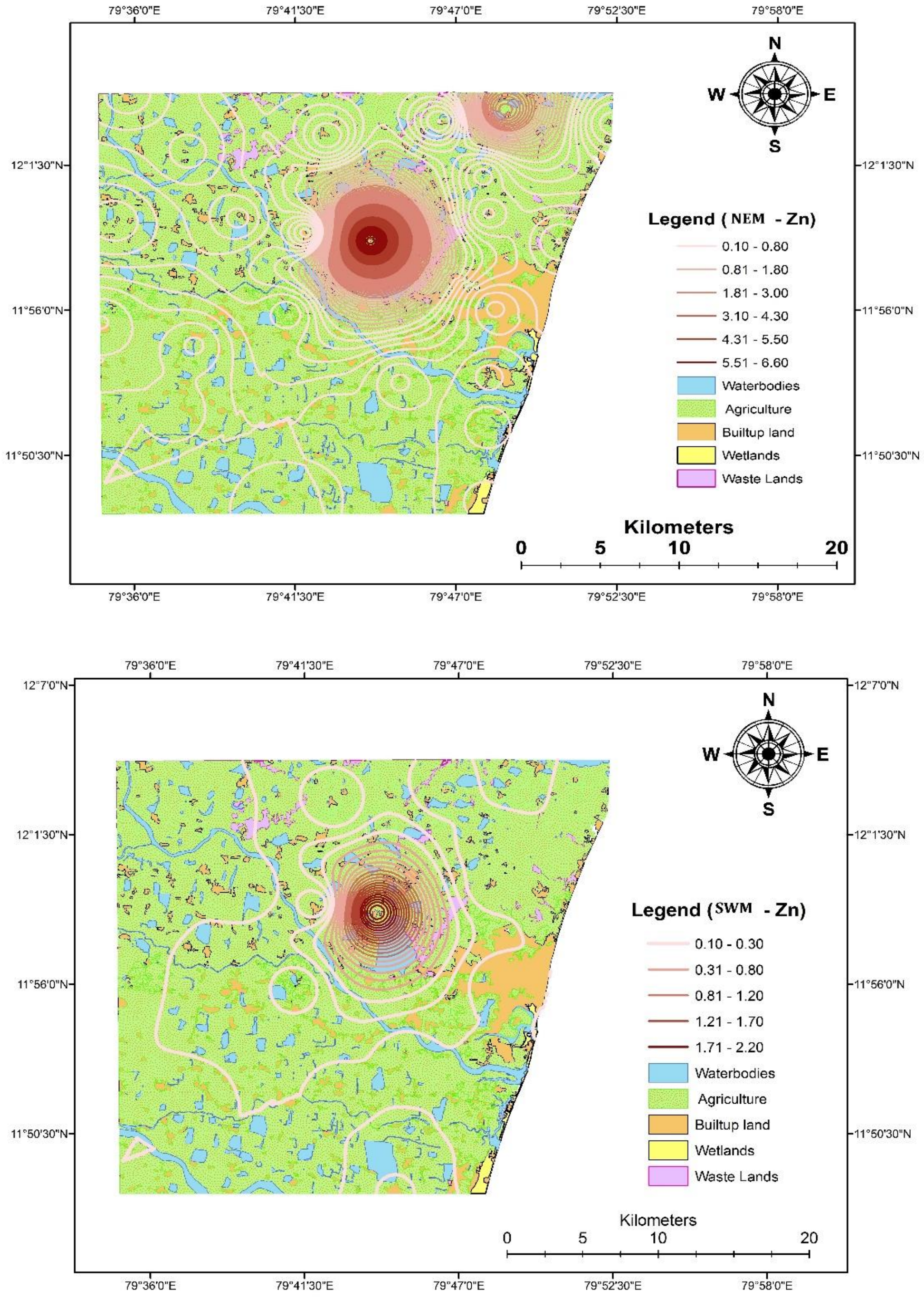

Figure 7. Concentration contour showing spatial distribution for Zn in groundwater throughout the study area in the a NEM, b SWM season overlaid on the landuse (all values in mg/l).

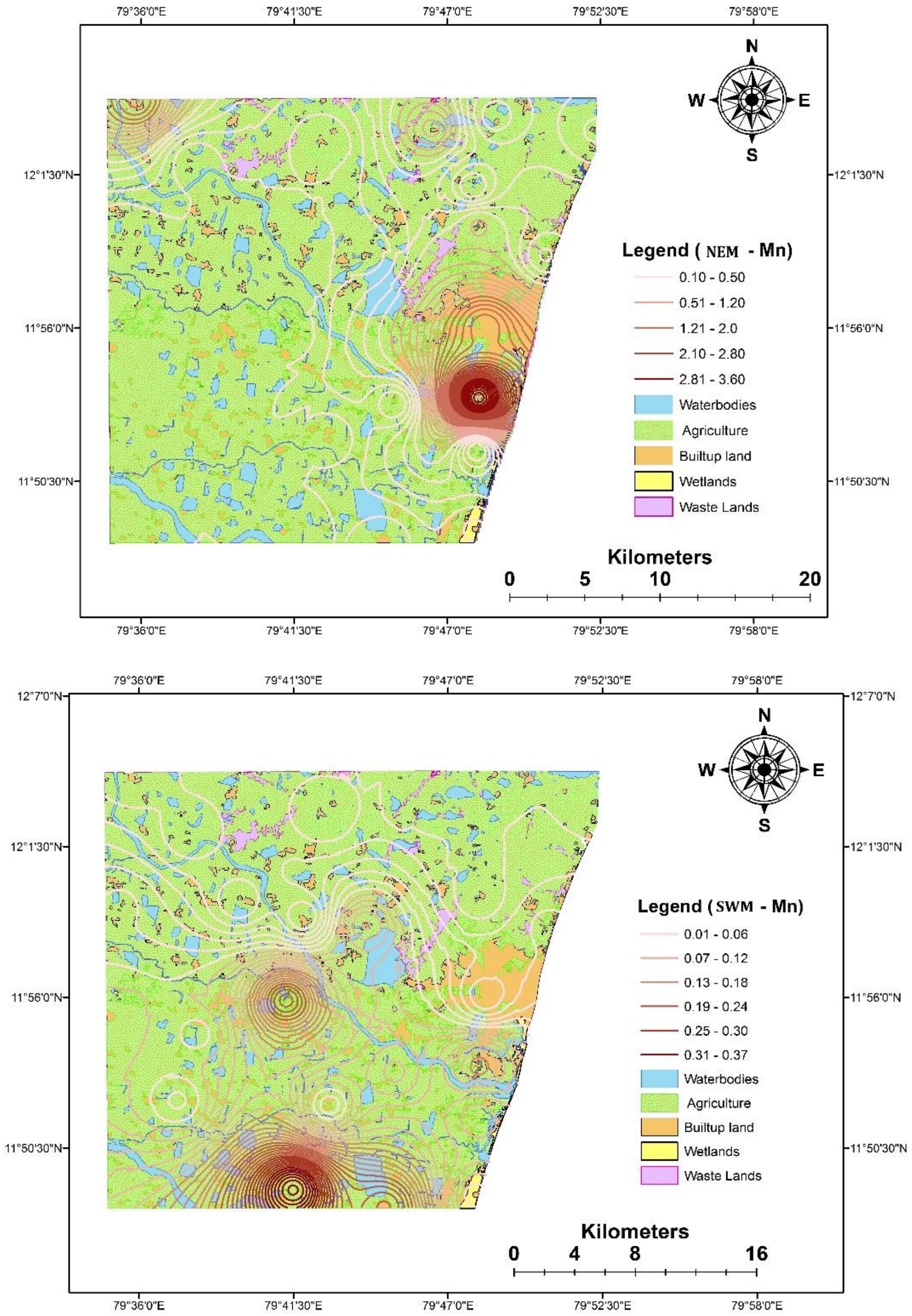

Figure 8. Concentration contour showing spatial distribution for Mn in groundwater throughout the study area in the a NEM, b SWM season overlaid on the landuse (all values in mg/l).

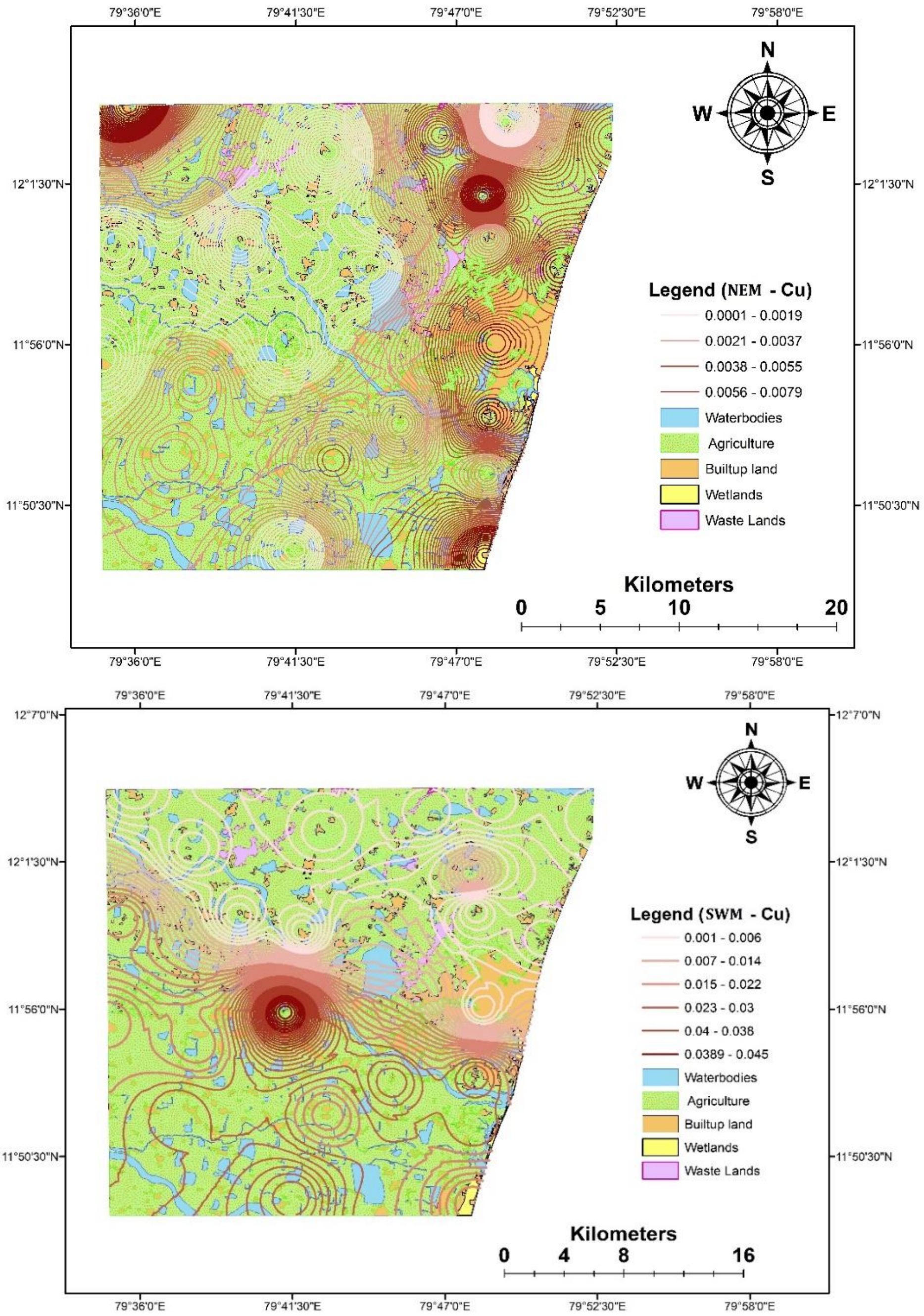

Figure 9. Concentration contour showing spatial distribution for Cu in groundwater throughout the study area in the a NEM, b SWM season overlaid on the landuse(all values in mg/l).

The potential sources of those metals are associated with natural processes or artificial influences. Under natural conditions, the dissolved ions within the water are attributed to the mineral assemblages within the rocks close to the land surface [35]. Composition of rock, minerals, texture, porosity, and regional structure also influence the concentration of heavy metals in the groundwater [35]. Meanwhile, manmade inputs, such as agriculture influence, industrial influence are different probable sources accredited to these variations. This, directly or indirectly, permit the seepage during the moving of waste along the pathways. Consequently, these contaminants can basically go through the soil and pollute the groundwater. The rainfall is also acting as a necessary part in controlling the movement of trace elements into the vadose zone [36].

Rainfall drains the huge quantity of pesticides which applied to control the grass growth in the cultivated land of the study area. Pesticides containing metal compounds are brought via surface runoff then penetrate into the groundwater, which leads to groundwater pollution [37]. The distribution of these contaminants is restricted to a small area once they penetrate into the aquifer. Therefore, it will be limited within the dumping area and is not diffused or discrete to the other places. The shallow groundwater level might also play an important role in the concentration of heavy metal in the study area by rainfall recharge in NEM. Rainfall is the major source of groundwater recharge in the study area. The study area acquires 63 % rainfall during NEM and 29% of rainfall during SWM. However considering the past ten years rainfall data , the proposed area receives the lowest rainfall during 2016, NEM season. The difference in precipitation and decline of water level influence the climate as well as groundwater quality of the study area.

3.3 Water classification with respect to pH and Heavy metal concentration

The water classification was done by considering the pH value and heavy metal concentration in groundwater of the study area by adopting the techniques of [38,39]. The method uses the water pH and metal load (mg/l). In this study, the metal load is calculated by taking the sum of heavy metals like (Mn+Cu+Pb+Zn+Fe). 8% of total samples falls in the acid-low metal and 92% , in the near neutral-low metal window, during NEM. However, 48 % of total Samples falls in the acid-low metal and 48 % in the near neutral-low metal and 4% in the acid-high metal window during SWM.

3.4 Factor Analysis

Principal component analysis (PCA) was considered in this study to delineate the source of heavy metal in the groundwater. The PCA of groundwater demonstrate to facilitate the variables were interrelated to two main components in which 62.7 % of the total variance was justified. During NEM, three factors with eigen values (>1) was extracted. The first factor with 27.46 % of variance comprises EC, Mn, Cu, and Fe with high loadings (Table 3) which seemed to be associated to the earth's crust and the geological formation of the area. The higher loading of Cu and Fe is due to salinity, cation-exchange progression at soil water interface and industrial wastes [40], high loading of Fe may also be due to dissolution of lithogenic materials. The second factor contributed to temperature and Cu and account for 19% of the total variance. Cu show association with temperature since its source is external and may be linked with agricultural return flow. The third factor contribute to pH and Zn and account for 17.3%. which indicates the acidic environment is more appropriate for the release of Zn in the groundwater,

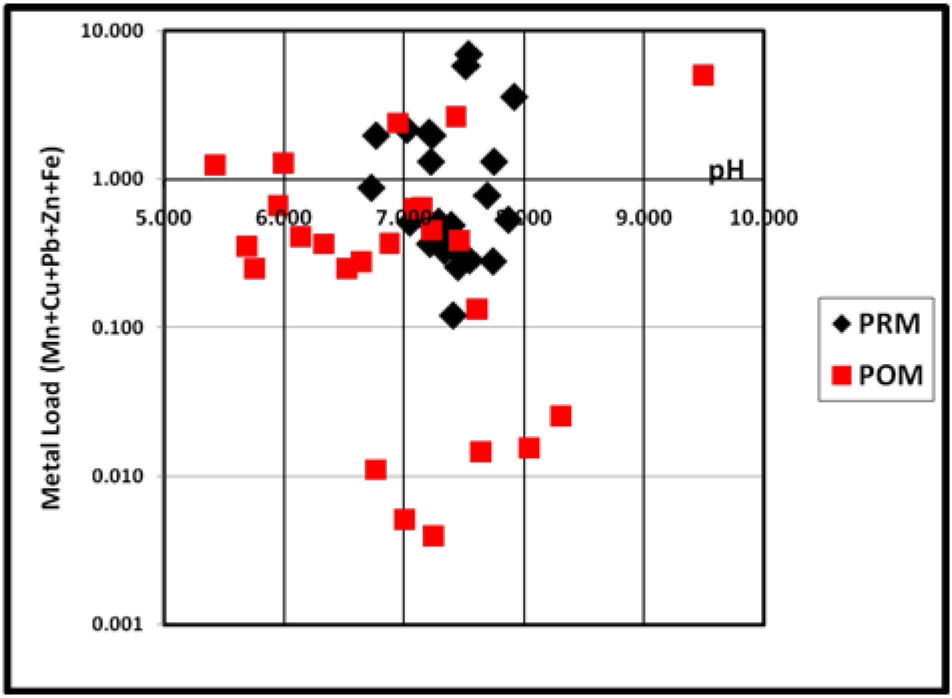

Figure 10. Classification of water samples based on the plot of metal load and pH

Table 3. Loading for the varimax 3-factors model

NEM	1	2	3	SWM	1	2	3
EC	.782	-.252	.198	EC	.730	.485	.318
pH	-.099	.116	.766	pH	-.295	.711	.239
Temp	-.168	.804	.229	Temp	-.478	.489	-.020
Mn	.906	.080	-.004	Mn	.799	-.207	.215
Cu	.548	.536	-.262	Cu	.760	-.373	-.052
Pb	-.002	-.674	.237	Pb	.598	.037	-.400
Zn	.028	-.246	.734	Zn	.101	.000	.870
Fe	.653	-.032	-.207	Fe	.060	.871	-.196

3.5 Chemical species of heavy metals

Chemical species of heavy metals were directly qualified to toxicity [42,43]. Florence (1982) has denied the term speciation analysis as the determination of the individual physico-chemical forms of the element, which together make up its total concentration in a sample. According to speciation analysis involves the use of analytical methods that can provide information about the physico-chemical forms of the elements. [44] distinguishes physical speciation, which involves differentiation of the physical size or the physical properties of the metal, and chemical speciation, which entails the differentiation among the various chemical forms.

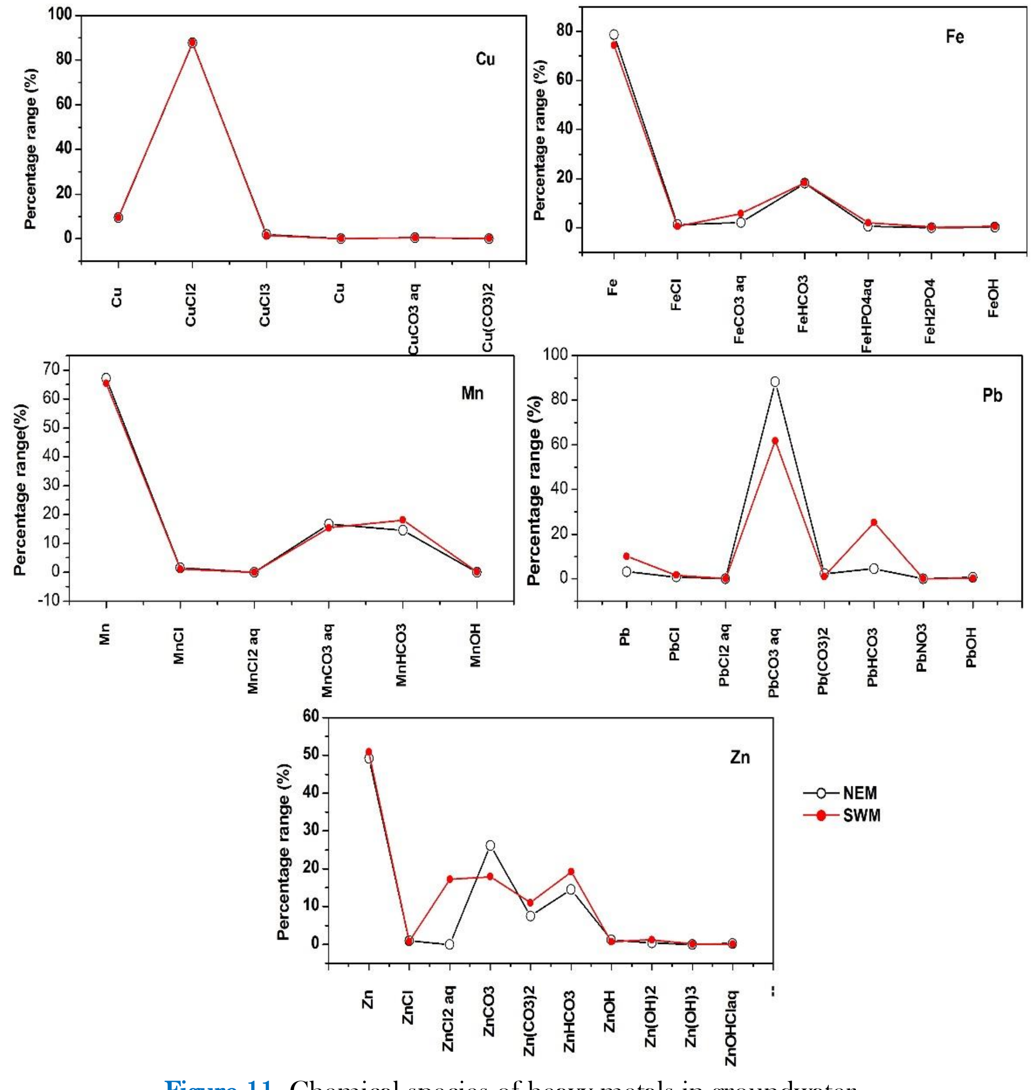

Figure 11. Chemical species of heavy metals in groundwater

The species distribution of the heavy metals ions, Cu, Fe, Mn, Pb and Zn in the groundwater samples of the study area were calculated by WATEQ4F program in percentage and in mol/L. All metals were found to exist as free and complex species. According to these computations, Cu was mainly present as chloride complexes. Pb was mainly present as carbonates complexes. Fe, Mn and Zn is mainly existing as free metal ions during NEM and SWM (Figure 11).

Copper: Chloride forms of copper constitute the largest part in both seasons. With the pollution of groundwater increasing, copper makes a significant percentage of its complexes with carbonates and hydroxides. Such complexes in both seasons comprise less than 1%. With increased pollution, there are significantly fewer ionic forms of copper (~10,3%). There are more chloride compounds of copper in the groundwater in both the seasons, but the differences are small, and the total amounts of migration forms reaches high.Zinc, Iron and Manganese. The ionic form of zinc, iron and Mn prevails free metal compounds and is more than 90%. (Fig 11) With increased pollution in shallow water, the formation of carbonate and bicarbonate compounds also increases. Sulphate and chloride compounds of zinc are detected more often in fewer amounts. Zn can be enhancing in the groundwater as zinc sulfide responded to Mg in the groundwater.

Lead. Carbonate migration forms prevail in both seasons, and their percentages remain stable. With increased pollution in groundwater, the amounts of ionic lead forms decrease to 19%. The free metal, chloride and sulphate complexes of lead are formed in both the seasons, but their percentage is not significant.

4. Conclusion

The heavy metal concentration in groundwater of Pondicherry region reveals that the metal concentrations were higher in the South West Monsoon season as compared to the North East Monsoon season. The Mn, Pb, Zn and Fe concentration in the groundwater samples of SWM found to be higher and above the permissible limit. Few samples fall in the high metal window which should be of great concern as the water is used for both domestic and irrigation purpose in the study area. It is inferred from PC analysis that the source of Fe and Mn are from weathering, rock water interaction and industrial sewage. The source of Pb is identified to be from anthropogenic and mainly related to agricultural return flow and industrial effluents. Thus, anthropogenic sources and weathering processes are the two main factors which influence the heavy metal concentration in the groundwater of the study area. The chemical species of Pb in groundwater was mainly in the form of $PbCO_3$ and Pb^{2+}. The $PbCO_3$, $MnCO_3$ and $PbHCO_3^+$ were dominant species in groundwater in some sub urban areas probably because of the concentration of HCO_3 and pH values influenced by lithology. The chemical species of Mn in shallow groundwater was mainly Mn^{2+}. Hence the study provides an insight on the levels, sources and distribution of some important metals in groundwater of the Pondicherry region.

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