Geochemical Characteristics and Evaluation of Minor and Trace Elements Pollution in Groundwater of Tuticorin City, Tamil Nadu, India using Geospatial Techniques

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ABSTRACT

In this work the major and trace elements pollution levels in groundwater of Tuticorin city of Tamil Nadu was investigated. This coastal city with numerous large and small scale industries has increasingly affected the water quality by dumping effluents. The mean major and trace elements levels in groundwater in descending order: Si > Sr > Zn > Fe > Rb > Se > Al > Mn > Cu > As > Cr > Ba > V > Ni > Pb > Mo > Co > Sb > Cd > Ag. The concentrations of As, Se, Pb, Ba, Al, Fe and V (0.084, 0.150, 0.057, 0.837, 0.357 and 0.052) in some of the groundwater samples exceed the standard value of world health organization (WHO, 2004). Interrelationship between the elements was done by correlation matrix and factor analysis which indicates the contribution from industrial and irrigation fields. Spatial interpolation of inverse distance method (IDW) maps of all the elements suggested that discharge of effluents from chemical factory (STERLITE, TAC, SPIC, HEAVY WATER PLANT, NILA SEA FOODS), municipal wastewater, fertilizers were added to the natural sources. Results of this research suggests that proper measures should be taken to protect the vital groundwater resources in the Tuticorin city.

INTRODUCTION

Major and trace elements above permissible limit in groundwater have direct consequences on human health and environment. Their occurrence in groundwater is often guided by two main pathways such as natural and anthropogenic. Natural pathways are associated with rock weathering, leaching, rock water interaction and resident time. On the other hand, anthropogenic activities such as burning of fossil fuels and discharge of industrial effluents and sewage into the water bodies can contribute significant concentration of trace elements in the groundwater (Handa 1981; Leung and Jiao, 2006). Metals like zinc (Zn) and copper (Cu) have a great role in balancing the metabolic activity of an organism and have a narrow concentration ranges. Other elements like aluminium (Al), cadmium (Cd) and lead (Pb) show extreme toxicity even at trace levels (Vanloon and Duffy, 2005; Puthiyasekar et al. 2010; Selvam et al. 2015a,b,c).

Increase in the trace metal contents of an aquatic system may begin from the daily human activities, a mixture of degrees of geochemical reactions occurring in the sub-surface of the earth, runoff addition, aerial authentication and percolation-related phenomena (Ibe and Duruike, 2005; Selvam et al. 2013a,b). Among the possible contaminants in water, heavy metals have attracted an unprecedented attention owing to their high toxicities even at low concentrations. Heavy metals are known to have a wide range of oxidation states which establish their physicochemical forms like simple hydrated, complex and colloidal particles (Matthew, 1996; Tokalioðlu et al. 2000; Selvam et al. 2014a,b,c; Venkatramanan et al. 2016). The different oxidation states of trace metals possess varying degrees of toxicity hence, the need for speciation. Most transition metals are very toxic while some micro-elements are very essential to the human biochemical systems in trace amounts. Speciation, which is the analytical study of heavy metal to determine their form of occurrence in any given sample, has wide applications in biological, geochemical, environmental, and water treatment processes. Speciation, in water analysis, is accomplished through the use of ion-exchange resin to selectively separate the exchangeable ions from the solid metals.

The present study area and surroundings have numerous large and small scale industries. The large scale industries in the study area (other than infrastructure providers i.e. the port and the power plant) are Sterlite (copper), SPIC (fertilizers and chemicals) and Kilburn chemicals (titanium dioxide). Moreover, cottage industries like the textile mills are also present. The presence of large industries can lead to the establishment of small scale industries that have product synergies with these units (Selvam 2014). In the past three years rapid industrial development on the coast due to the establishment of large scale industries with thermal power plant, fertilizer, chemical and mineral processing plants, textile mills, match factories and nearly 500 small scale industries. Since the area has increased population there is concern regarding the quality of the groundwater aquifer system (Ramesh et al. 1995; Mandal and Sengupta 2006; Selvam 2012a,b; Singaraja et al. 2015).

There are few studies published in recent years on heavy metal pollution indices and bacterial contamination in groundwater sources in Tuticorin city, India (Balasubramanian et al. 1993; Selvam 2015; Singaraja et al. 2016a). Hence, current research work will deliver an efficient scientific basis of major and trace elements concentrations and may provide useful vision for future study.

MATERIALS AND METHODS

Description of research site

Tuticorin city extends over approximately 154 sq km and lies between 8°43' - 8°51' N latitude and 78°5' - 78°10' E longitude in the southern part of Tamil Nadu, India (Fig. 1). Topographic elevation ranged from 0 (near the coastline) to 27 m (msl) in western part. The slope is gentle in the western and the central part further it is nearly flat in the eastern part. The salt pans, chemical industries and irrigation fields are situated in this surrounding area. The port was constructed in the year of 1986 after urbanization. During the summer season, vegetables/leguminous crops are full-fledged, whereas crops like rice,
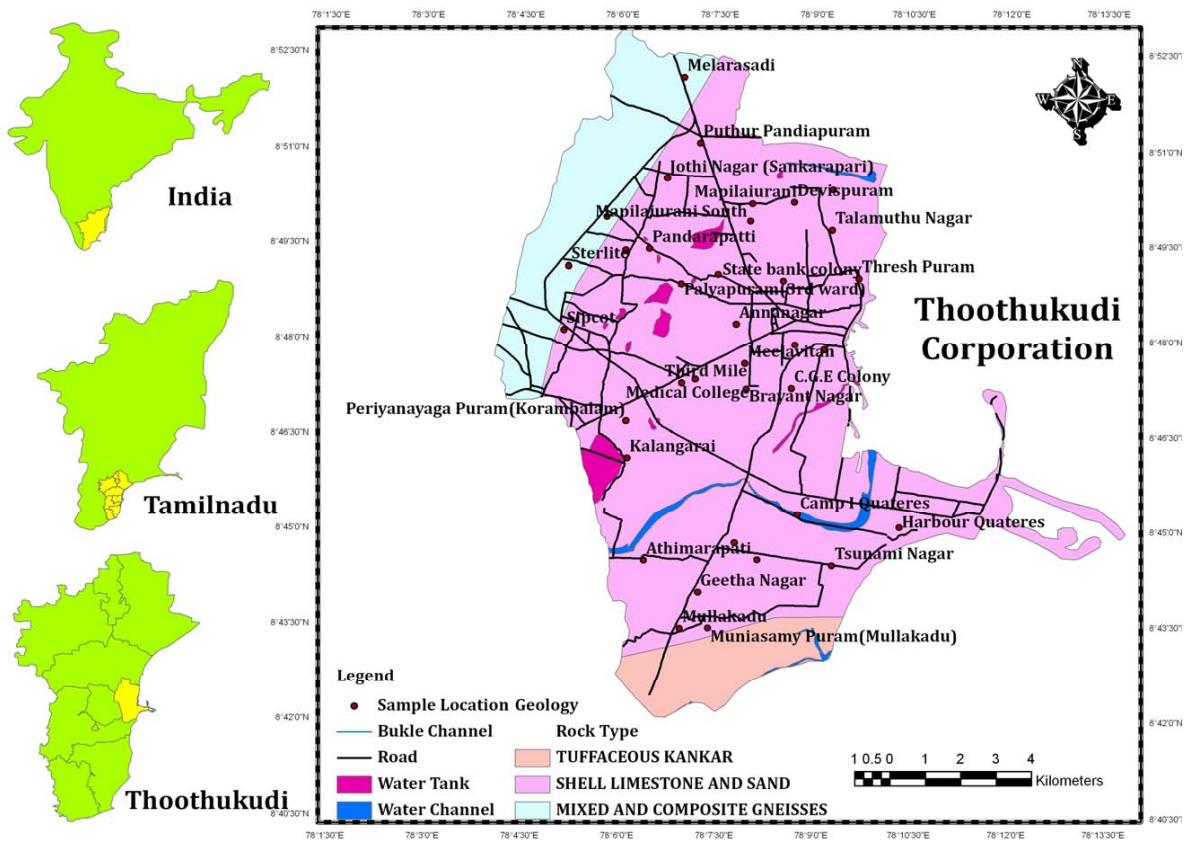
Fig.1. Location of the study area and the sampling sites

sugar-cane, maize, and guinea-corn are grown in monsoon season. The other part of the city has industries manufacturing chemicals, salt, flower dying, copper wire, copper alloy, alkali chemicals and fertilizers, petro-chemicals and plastics, heavy water, chemical dying (Selvam 2016; Singaraja et al. 2016b).

GEOGENIC SETTING

Tuticorin city has 90% sedimentary rocks of Tertiary to Recent age consisting of shell limestone and sand, tuffaceous kankar, sand (Aeolian deposits) while other parts are covered by mixed and composite gneiss of Proterozoic age (Fig. 2). Sedimentary rocks are fully covered with fossiliferous shells of gastropods and pelecypods. The average thickness of the bed ranged from 5m to more than 20m which extends in the NE-SW direction and dipping 10° to 15° towards SE. Archaean group of rocks also occur and are represented by crystalline, metamorphic, and finely foliated rocks with a NW to SE trend (Balasubramanian et al. 1993; Selvam et al. 2016). The soils are divided into 3 types viz., black soils in the western part (Sankarapari area), red soil (sandy loam to sandy soil) in the central part and alluvial sandy soils (Coastal area) in the eastern parts. Mainly unconsolidated and semi consolidated rock formations with fractured crystalline rocks are the water bearing aquifer zone. The maximum and average thickness of alluvium is 45 m and 25m bgl, respectively. The recorded average annual rainfall in this region is 877mm. NE monsoon contributes 65.4% of rainfall. Two major groundwater bearing rock formations are shell limestone and aeolian deposits with confined and semi-confined condition. Hydraulic properties of the aquifers exhibited are both vertical and horizontal dimensions. Water table depth ranged from 1 to 10m bgl in post-monsoon and 2 to 15m bgl in pre-monsoon. The mean maximum and minimum temperature was noted is 37.6°C (June) and 19.9°C (January), respectively. The temperature is relatively high and increases the rate of evaporation of the surface water and also the actual evapotranspiration, which is nearly equal to that of the annual precipitation (Balasubramanaian et al. 1993; Selvam et al. 2015). Based on the scanty contribution of rainfall the recharge of groundwater system is very less.

SAMPLING AND ANALYSIS

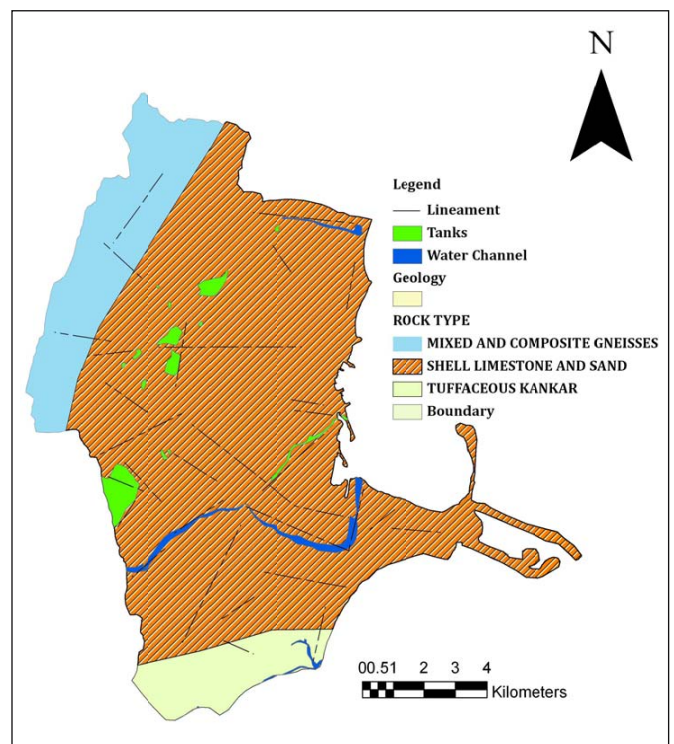A total of 36 groundwater samples were collected from 20 open

Fig.2. Geology map of the study area

wells and 16 bore wells with depth varying between 6 to 86 mbgl in pre-monsoon (PRM) season 2013. The coordinates of the sampling stations are marked with the help of a hand held GPS (Garmin, etrex). The water samples were collected in pre-cleaned, acid washed (1N HCl) HDPE bottles of 500 ml capacity. The samples were filtered, acidified with nitric acid (1N) and analyzed for trace elements at geochemistry laboratory of National Geophysical Research Institute (NGRI), Hyderabad using Inductively Coupled Plasma Mass Spectrometer (ICP-MS) Model ELAN DRC II, Perkin–Elmer Sciex® Instrument, USA. The Acidified groundwater samples were directly fed into the ICP-MS by conventional pneumatic nebulization, using a peristaltic pump with a solution uptake rate of about 1 ml/min. The nebulizer gas flow, sample uptake rate, detector voltage and lens voltage were optimized for a sensitivity of about 50,000 counts for a 1 ng/ml solution. Calibration was performed using the certified reference material NIST 1640a (National Institute of Standards and Technology, USA) to minimize matrix and other associated interference effects. Relative standard deviation (RSD) was found to be better than 6% in the majority of the cases, which indicates that the precision of the analysis is reasonably good. The results were evaluated in accordance with the drinking water quality standards given by the World Health Organization (WHO 2004).

GEOSTATISTICAL TECHNIQUES

Statistical analyses were carried out by SPSS version 19.0. Factor analysis was used for data interpretation and source identification. The analytical results of 20 selected trace element concentrations are reported in mg/l. Pearson's correlation analysis was used to know the relationship between the elements. Factor analysis is used to identify the most important components contributing to the data structure and the interrelationships among variables (Lall and Sharma, 1996; Selvam and Sivasubramanian 2012). It was used to enumerate the natural and anthropogenic contributions. This is represented by 1st step array of correlation coefficients for all pairs of variables were calculated. Then, the matrix was diagonalized and its principal components (eigenvectors) were obtained. Two types of factor loadings obtained, factor I is associated to the largest eigenvalue and is able to explain the greatest amount of variance in the data set. The second factor

Table 1. Statistical measures such as minimum, maximum, average and standard deviation

Elements	Concentration (mg/l)		Average	SD	Percentage of samples exceeding allowable limits in WHO 2004
	Min.	Max.			
Al	0.011	0.837	0.084594	0.136	9
Si	0.026	23.45	2.235606	5.907	*
V	0.0021	0.052	0.01008	0.010	*
Cr	0.0015	0.08	0.013086	0.013	3
Mn	0.00095	0.424	0.040315	0.078	0
Fe	0.0018	0.357	0.123033	0.080	9
Ni	0.000082	0.011	0.005192	0.003	0
Co	0.000077	0.0273	0.002004	0.005	*
Cu	0.0027	0.2365	0.031661	0.060	0
Zn	0.00094	0.87	0.203493	0.197	0
As	0.00087	0.0837	0.015796	0.025	30
Se	0.00027	0.1497	0.086987	0.050	32
Rb	0.00028	0.8501	0.107435	0.249	*
Sr	0.00043	2.00036	0.504263	0.447	*
Mo	0.000025	0.0086	0.002297	0.002	0
Ag	0.000022	0.0024	0.000272	0.000	*
Cd	0.000012	0.0021	0.000296	0.000	0
Sb	0.000015	0.007	0.000889	0.002	3
Ba	0.00053	0.057	0.012131	0.012	0
Pb	0.00045	0.018	0.004375	0.003	6

(orthogonal and uncorrelated with the first one) explains most of the remaining variance, and so forth (Antony Ravindran and Selvam 2014b; Sonkamble et al. 2012).

Inverse Distance Weighted

Tuticorin city map was digitized from survey of India toposheet using ArcGIS 9.3 software and location points are imported by GPS (GARMIN 12). Further, IDW interpolation technique was used for spatial analysis. This method regulated the cell values by linearly-weighted mixture and set of location points. This weightage is a function of inverse distance and then input point is from the output cell location, the less importance it has in the calculation of the output value. The output value for a cell using IDW is partial to the sort of the input values used to interpolate. For the IDW is a weighted distance average, the average need not be greater than the highest or less than the lowest input value. The sampling is sufficiently dense to represent the local variation that needs to be simulated. Thus, IDW technique is ideal for analysis in respect of water quality data from various sampling points densely spread out. If the sampling of input points is scant or very irregular, the results may not adequately characterize the preferred surface.

RESULTS AND DISCUSSION

Interrelationship between the Elements

Pearson's correlation coefficient values are given in Table 3 and the significant level is $p < 0.01$. pH correlated with Cd and Pb with a correlation coefficient of 0.66 (p<0.01) and 0.65 (p<0.01), respectively. It indicates that contamination sources from industrial and irrigation fields. Meanwhile, EC and Mn have a correlation coefficient of 0.44 (p<0.01) and displays significant correlation between TDS and Cd, Al and Cd, Cd and Cu, Cr and Cu, Cu and Mn, Mn and Pb, Ni and Pb which indicate that good correlations with respective correlation coefficient (r) values of 0.78, 0.62, 0.33, 0.44, 0.31, 0.45 and 0.58. But, Fe show by very low correlation with other elements due to the local geogenic origin. As does not show any correlation because its concentration is controlled by other sources. This inter-relationship of elements clearly denoted that contamination of groundwater from the processes of chemical industries and landfill leachate/municipal sewage systems (Tariq et al. 2010).

Factor Loadings

It terms the processes falling in the investigated area and identification of the factors allows genetic understanding of the environment. Thus, it is a valuable tool to state the factors that impact on groundwater hydrochemical processes. The obtained results shows that operation are high factor loadings (close to 1 or >1) and low factor loadings (close to 0) attained for the correlated variables. The best numbers of factors are known as from eigen value > 1 represented by good interpreted data set. (Mohamed El Alfy, 2010). There are five factors loading with total variance of 75.40% and this given in Table 2. Factor 1 exhibited 40.62% of the total variance with significant loading on Al, V, Fe, Ni, Zn, Se, Sr, Mo, Ag and Cd. This is due to the influence of industries. Factor 2 accounted the 11.67% of the total variance with high loading of Mn, Cd, Pb. This indicates that the influence of natural activity is one of the most important factors controlling groundwater chemistry of this city. Another factor which controls the chemistry is use of manganese and phosphate fertilizers in irrigation. Factor 3 represented by 9.21% of the total variance with good loadings of Cr, V, Sb elements while Factor 4 revealed that 7.68% of the total variance with significant loading of V, Mo, Cd. This factor loading may be attributed to anthropogenic activities like domestic, agricultural runoff and industrial wastes discharge and also natural phenomena. Based on the factor loadings it is suggested that major

Table 2 Factor analysis results of trace elements in groundwater

Parameters	Factors				
	1	2	3	4	5
Al	.245	.629	-.105	-.089	.216
Si	-.693	-.149	-.320	-.049	.137
V	.136	-.184	.479	.663	.147
Cr	-.044	.473	.764	-.069	-.258
Mn	-.582	.596	.429	-.140	-.066
Fe	.831	.392	-.055	-.134	.112
Ni	.859	.326	-.115	.189	.008
Co	-.698	.173	-.066	.122	.245
Cu	-.894	.226	.068	.028	.155
Zn	.608	.069	.158	-.098	.442
As	-.951	.109	-.046	.005	.140
Se	.908	.121	.090	-.025	.140
Rb	-.850	-.016	-.241	.012	.252
Sr	.581	-.222	-.003	-.281	-.297
Mo	.556	-.119	.161	.637	.128
Ag	.326	.092	.007	-.015	.670
Cd	.428	.501	-.472	.339	-.290
Sb	-.680	.242	.395	.045	-.038
Ba	.495	.121	.085	-.575	.182
Pb	-.209	.731	-.448	.224	-.122
Total	**8.124**	**2.335**	**1.844**	**1.537**	**1.241**
% of Variance	40.622	11.676	9.218	7.684	6.204
Cumulative %	40.622	52.298	61.516	69.200	75.404

and trace elements are mainly contributed from the industrial sectors and are also of local geogenic origin.

Geospatial Interpolation (IDW)

Pb concentration in groundwater ranged from 0.00045 to 0.018 mg/l with an average of 0.0045 mg/l. The maximum allowable limit of lead ion concentration in groundwater is 0.01 mg/l as per WHO 2004 standards (Fig. 3). Pb is the most significant of all the heavy metals because it is toxic, very common and harmful even in small amounts (Gregoriadou et al. 2001). It is found in trace amounts in various foods, notably in fish. Some old houses having lead water pipes can cause contamination of drinking water. The desirable limit of Pb for drinking water is specified as 3 ppm (WHO 2004). The analysis of water in our study has lesser concentrations of lead than the recommended guideline value. The not permissible (NP) limit of Pb in PRM was observed towards southeast portion of coastal area.

Arsenic concentration in the groundwater varies from 0.00087 to 0.084 mg/l with an average of 0.0158 mg/l. The maximum permissible limit of arsenic in groundwater is 0.01 mg/l (WHO 2004). As contamination patches were observed in NW, NE and central portion due to industrial (STERLITE, SPIC, TAC, HEAVY WATER PLANT) waste leaching and percolation into the aquifer system (Fig. 4). Arsenic trioxide is obtained as a byproduct from dusts and residues produced during the treatment of metal ores such as gold and copper (Puthiyasekar et al. 2010; Venkatramanan et al. 2015). Arsenic is highly toxic and affects human health drastically.

Chromium concentration in the groundwater varies from 0.0015 to 0.08 mg/l with an average of 0.013 mg/l. only one sample exceeds the permissible limit in NE portion, which may be due to industrial activity (STERLITE) (Fig. 5). The most common man-made sources of chromium in groundwater are burning of fossil fuels, mining effluent, and effluent from metallurgical, chemical, and other industrial operations (Leung and Jiao, 2006). Cr levels greater than 0.05 mg/l. However, the effect of chromium in drinking water has not been thoroughly investigated but chromium is known to produce lung tumors when inhaled.

The concentration of Fe in the groundwater varies from 0.0018 to 0.357 mg/l, with an average of 0.123 mg/l (Fig. 6). The maximum permissible limit of iron in groundwater is 0.3 mg/l (WHO 2004). NP limit of iron in PRM was observed in patches in central portion of the study area. Occurrence of iron in drinking water leads to unpleasant taste and may increase the hazard of pathogenic organisms. Since most of these organisms need iron to grow. More than about 0.3 mg/l of iron, stains laundry and utensils reddish brown. The bioavailability of iron in drinking water has not been investigated properly. It is known that iron influences the uptake of copper and lead.

Al varied from 0.011 to 0.837 mg/l with an average of 0.084 mg/l (Fig. 7). The not permissible (NP) limit of aluminium (0.2 mg/l) in PRM was observed in north and central portion of the study area. Aluminum occurs naturally in some rocks and mine tailings. Aluminum forms by weathering of feldspars. High concentration of Al in groundwater causes premature senile dementia (Alzheimer's disease).

The concentration of selenium in the groundwater varies from 0.00027 to 0.1497 mg/l and average of 0.0869 mg/l (Fig. 8). The maximum permissible limit of selenium in groundwater is 0.01 mg/l (WHO 2004). As per WHO standards seven samples exceeded the limit in the study area having sedimentary rocks. Se is derived from sedimentary rocks and in groundwater as a mixture of selenite and selenate. Selenium from hazardous waste sites and from farmland will end up in groundwater or surface water through irrigation and runoff. Selenium poisoning can affect the growth, skin discolouration, bad teeth, and some psychological and gastro-intestinal problems. Diminutive amount of selenium was beneficial, but excess amount was toxic.

Sb in groundwater between 0.000015 and 0.007 mg/l with an average of 0.00088 mg/l (Fig. 9). The maximum permissible limit of Sb in groundwater is 0.005 mg/l (WHO 2004). Only one sample had more than desirable level, that is, 0.005 mg/l.

Cadmium concentration ranged from 0.000012 to 0.0021 mg/l with an average of 0.000296 mg/l. Acute cadmium poisoning symptoms are similar to those of food poisoning. It is associated with kidney disease and linked to hypertension. Up to 325 mg of cadmium is not

Table 3 Correlation coefficients for parameters of groundwater samples

	pH	EC	TDS	Al	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn
pH	1											
EC	-0.02	1										
TDS	0.12	0.09	1									
Al	-0.22	0.02	0.09	1								
Cd	**0.66**	0.17	**0.78**	**0.62**	1							
Cr	0.03	0.18	0.12	0.23	0.11	1						
Cu	0.12	0.21	0.33	-0.11	**0.33**	**0.44**	1					
Fe	-0.45	0.12	0.25	-0.23	0.15	-0.11	0.22	1				
Mn	-0.22	**0.44**	-0.22	-0.33	-0.22	0.21	**0.31**	0.12	1			
Ni	0.16	-0.12	-0.15	0.12	-0.33	0.14	-.23	-0.11	0.23	1		
Pb	**0.65**	-0.56	0.39	0.16	0.15	0.09	-0.11	0.11	**0.45**	**0.58**	1	
Zn	0.22	-0.12	0.33	0.29	0.22	-0.22	-0.03	0.14	0.05	0.06	0.02	1
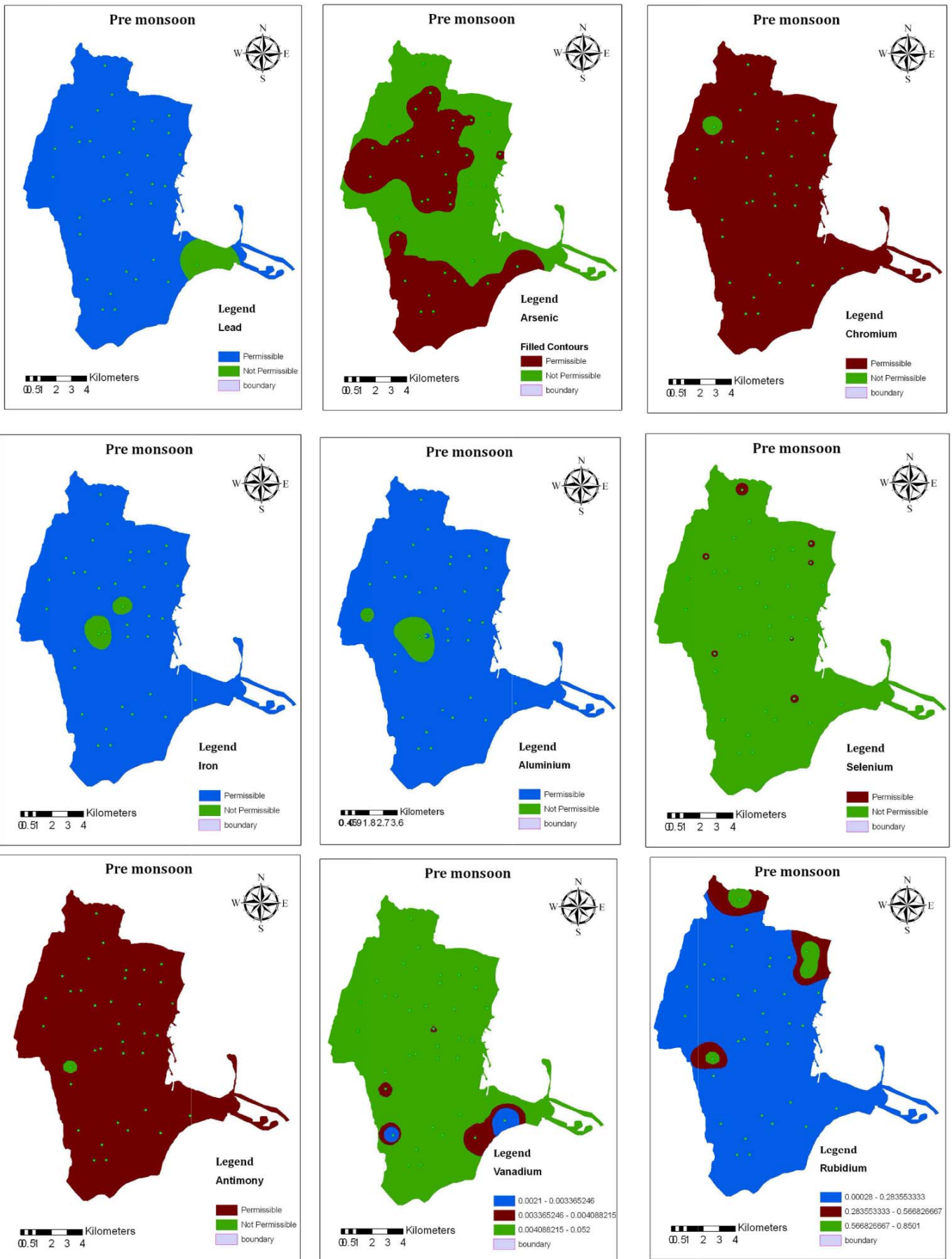
Figs.3-11. Spatial variation maps showing **(3)** Lead **(4)** Arsenic, **(5)** Chromium, **(6)** Iron, **(7)** Aluminium, **(8)** Selenium, **(9)** Antimony, **(10)** Vanadium and **(11)** Rubidium concentration in the study area.

fatal but toxic symptoms occur at 10 mg. In the study area, cadmium concentration is too low and below the permissible limit of WHO 2004.

Nickel level ranged from 0.000082 to 0.011 mg/l, with an average of 0.005192 mg/l. The permissible limit for Ni is 0.02 mg/l for drinking water (WHO 2004). It is noted that none of the samples are above the permissible limit. Ni compounds induce nasal, laryngeal and lung cancer (Lessard et al. 1978; Singaraja et al. 2015; Venkatramanan et al. 2014). Molybdenum concentration in groundwater varied from 0.000025 to 0.0086 mg/l, with an average of 0.00229 mg/l. The permissible limit for Mo is 0.07 mg/l for drinking water (WHO 2004). Mo concentration in the study area is within the permissible limit. Ba in groundwater ranged between 0.00053 and 0.057 mg/l, with an average of 0.012131 mg/l. The permissible limit for Ba is 0.7 mg/l for drinking water (WHO 2004).

Cu concentration varied from 0.0027 to 0.2365 mg/l with an average of 0.0316 mg/l. The permissible limit for Cu is 2 mg/l for drinking water (WHO 2004) and it is found to be within the permissible limit. Zn level in groundwater ranged from 0.00094 to 0.87 mg/l with an average of 0.20 mg/l. The permissible limit for Zn is 3 mg/l for drinking water (WHO 2004). Zn concentration is found to be within the permissible limit in all groundwater samples. Manganese concentration ranged from 0.00095 to 0.424 mg/l with an average value of 0.040 mg/l. The permissible limit for Mn is 0.5 mg/l for drinking water (WHO 2004). Cobalt concentration in the groundwater is between 0.000077 and 0.0273 mg/l with an average value of 0.0020 mg/l. Vanadium concentration in the groundwater varied from 0.0021 to 0.052 mg/l with an average of 0.01008 mg/l (Fig. 10). Silver concentration in groundwater varied from 0.000022 to 0.0024 mg/l, with an average of 0.00027 mg/l. The concentration of strontium ranged from 0.00043 to 2.00036 mg/l, with an average of 0.504263 mg/l. Rubidium varied from 0.00028 to 0.8501 mg/l, with an average of 0.1074 mg/l (Fig. 11). WHO drinking standard have not given any guidance value for cobalt, vanadium, silver, strontium and rubidium concentration in the groundwater. However, existence of these elements in ultra trace quantity highlights the presence of natural as well as anthropogenic influences in the groundwater of the study area.

CONCLUSION AND RECOMMENDATION

According to this research, As, Se, Pb, Ba, Al, Fe and V elements suggested that above the permissible limit for WHO (2004) drinking standard. The mean concentration of other elements was lower than the guideline value. These elements are polluted and effluents mainly originated from copper industries, fertilizer factories, co-operative distilleries and municipal waste water. Inter-relationship of elements represented by correlation and factor loading indicated that Se, Al, Pb and Sb were linked and controlled by mixed origin from anthropogenic and geogenic sources while As, Fe and Cr were derived from man-made activities. Spatial interpolation of inverse distance method (IDW) maps suggested that discharge of effluents from chemical factory (SPIC, TAC, HEAVY WATER PALNT, STERLITE, and NILA SEA FOODS), municipal wastewater, fertilizers and addition to natural sources. Concentrations of elements were mostly distributed to south and NW direction in this study area. In view of these findings, there is need to observe more closely the environment under review and put in place appropriate checks and balances to preserve the health of communities within the industrial areas, particularly as the effects of major and trace elements are pose great dangers to humans health, animals and plants. Moreover, government should approve some treatment technologies (Central effluent treatment plant and Iron oxyhydroxides and the adsorption/co-precipitation removal mechanism) in this city to reduce these elements contamination in natural water resources.

The unprocessed wastes emerging from the industries must be supervised for preserving the standards recommended by the pollution control board for various industries in this city. The chemical analysis of the treated effluent from the CETP (Common effluent treatment plant) was established to hold some toxic metals of As, SE and Fe. As the CETP receives wastes from various industries in transporters and it is advised to check for the elements and TDS concentrations before permitting the raw effluents into the CETP. The present study delivered the baseline data for assessment of elements contamination in Tuticorin city. Periodical observing the water quality has to be continued to check the increase in TDS concentrations of groundwater.

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