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Distribution of heavy metals in surface water of Ranipet industrial area in Tamil Nadu, India

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Abstract Ranipet industrial area is about 120 km from Chennai on Chennai-Bangalore highway and is a chronic polluted area identified by Central Pollution Control Board of India. It is one of the biggest exporting centers of tanned leather in India. The total number of industries located in and around Ranipet town are 240 tanneries along with ceramic, refractory, boiler auxiliaries plant, and chromium chemicals. Studies were carried out to find out the contamination of surface water bodies due to industrial effluents. The results reveal that the surface water in the area is highly contaminated showing very high concentrations of some of the heavy/toxic metals like Cadmium ranging from 0.2 to 401.4 μg/l (average of 51.1 μg/l), Chromium 2.4–1,308.6 (average of 247.2 μg/l), Copper 2.1–535.5 μg/l (average of 95.5 μg/l), Nickel 1.6–147.0 μg/l (average of 36.7 μg/l), Lead 6.4–2,034.4 μg/l (average of 467.8 μg/l) and Zinc 20.8–12,718.0 μg/l (average of 3,760.4 μg/l). The concentration levels of these metals are much above the permissible limits in surface water and are health hazards especially for the people working in the tannery industries. It was observed that the people in the area are seriously affected and suffering from occupational diseases such as asthma, chromium ulcers and skin diseases. Distribution of metals, their contents at different locations, and their effects on human health are discussed in this paper.

Keywords Tannery industry · Heavy metals · Inductively coupled plasma-mass spectrometry · Ranipet-India

Introduction

Water pollution is a major problem related to the economic/industrial growth of any country. The number of industries in India, during the last decade, has grown more than ten times and accordingly the problems related to environmental degradation have increased many folds. There is a need for sustainable development of economic growth and industries. Some of the industries release their effluents either on the open land or in surrounding surface water bodies contaminating the soil, surface water and ultimately groundwater. Government of India is aware of these problems and have started looking into the remedial measures to clean some of the highly contaminated surface water bodies. Involvement of very high costs of remediation will make this process slow and therefore, it is essential that the contamination of water bodies is controlled rather than remediation.

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Leather production is a major industry in India, which makes significant contribution to the country's foreign exchange earning and provides employment opportunities to about three million people. Dissolved chromium reaches concentration levels of some g/kg^{-1} due to anthropogenic pollution from tanneries, which is generally linked to the use of Cr (VI) compounds in several industries applications such as plating, metallurgy, pigments, and leather tanning (Nriagu 1988; Appelo and Postma 1996; Tariq et al. 2006). During the process of leather manufacturing, several chemicals like Cr $(SO_4)_3$, NaCl, Ca $(OH)_2$, H_2SO_4 etc. are extensively used. Therefore, the resultant effluent is enriched with chromium and sodium salts (NaCl and SO_4). The tannery waste is characterized by its strong color (reddish dull-brown), high BOD, high pH, and high total dissolved solids. Tannery effluents, being voluminous and highly puerile, when discharged untreated, damage the normal life of the receiving water bodies; and if allowed to percolate into the ground for a prolonged period, will pollute the groundwater permanently and make it unfit for drinking, irrigation, and domestic consumption. Groundwater is the main source of water supply for domestic use and drinking purpose in rural areas. It is established that a single tannery can cause the pollution of groundwater around the radius of 7–8 km (Bhaskaran 1977; CLRI 1990; Ansari et al. 1999). Chromium present in effluent is primarily in the less toxic trivalent form Cr (III) but when this effluent is discharged into the soil, due to varying environmental conditions, Cr (III) is oxidized to toxic hexavalent form, which seldom remains as Cr (VI) (Anderson 1999; Govil et al. 2004; Srinivasa Gowd et al. 2005).

The relation between pollution and health problems is now well documented and reasonably well known by the general public (Barzilay et al. 1999). In Tamilnadu state, where more than 60% of India's economically important tanning industry is located, tannery waste containing chromium and sodium compounds has, over many years, contaminated 55,000 ha of agricultural land. Thousands of farmers lost their farms, or part of their earning capacity because of this contamination. Incidence of respiratory diseases among workers exposed to occupational and environmental risks of tannery industry at Ranipet industrial area is reported (Mahimairaja et al. 2000). In many areas of Tamilnadu, groundwater is not suitable for domestic use, forcing villagers to travel 4–5 km for water. Much of the groundwater is unsuitable for irrigation, and hundreds of wells in the region can no longer be used (Thangarajan 1999; Selvakumar and Manoharan 2002; Mondal et al. 2005). This is also one of the contaminated sites identified by Central pollution control board, India.

The primary objective of the present study was to assess the heavy metal contamination and their distribution in surface water.

Materials and methods

Study area

Ranipet industrial area is a part of Vellore District of Tamilnadu, located in 79°19′–79°22′ E longitude and 12°53′–12°57′ N latitude and is a chronic polluted area and one of the biggest exporting centers of tanned leather (Fig. 1). Many small-scale tanneries are processing leather in the study area and discharging their effluents on the open land and surrounding water bodies (Govil et al. 2004). The total numbers of tannery industrial units located in and around this town are 240 besides other industries like ceramic, refractory, boiler auxiliaries plant, and chromium chemicals.

Industries located in Ranipet are discharging effluents into Puliathengal, Vanapadi, and Thandalam lakes and it is a matter of increasing concern, as these industries are located in Palar river basin. Studies of groundwater also indicated the high concentrations of chromium in Palar river basin, which is much more than the permissible limit in drinking water. These tanneries are polluting the Palar River, causing ecological degradation and health hazards (Thangarajan 1999).

Geology

Geologically the study area is covered by crystalline rocks of Archaean age consisting of Granites and some basic intrusive bodies (Fig. 2). The alluvium consisting of fine to coarse sand and clay occurring in the area is of a fluviatile origin and restricted to the course of Palar river and major streams (Subramanian and Selvan 2001).

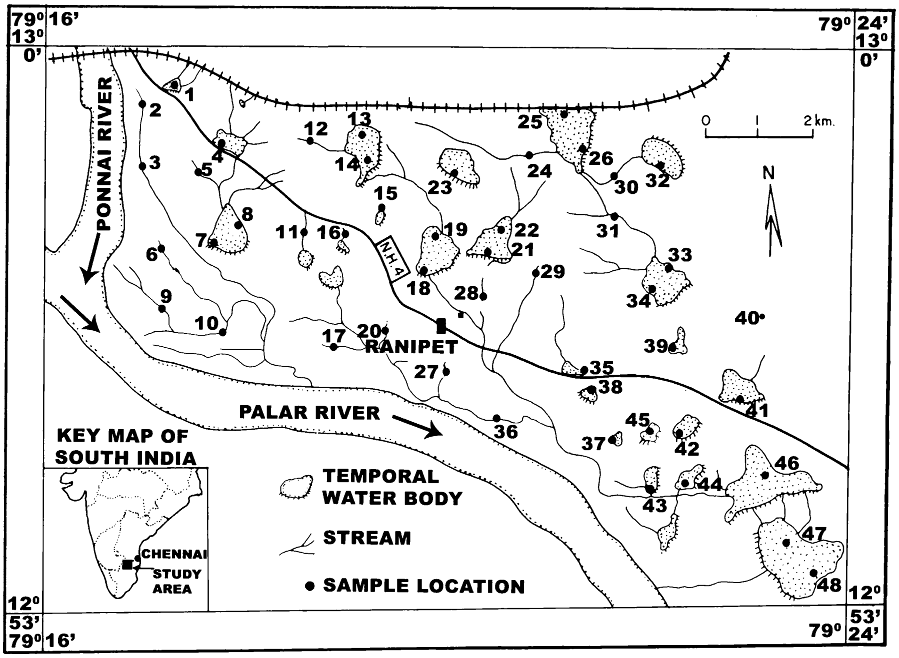


Fig. 1 Location map of the study area

Sample collection

For the assessment of surface water quality, 48 water samples were collected during May 2006 from the water bodies and streams distributed throughout the area. These samples were collected in 1-l capacity, double cap polythene bottles. pH and total dissolved solids were measured in the field by using water proof pocket testers, manufactured by Eutech Instruments, Singapore. Elico make specific conductance bridge and pH meter were used to determine the specific conductance at 25°C and pH, respectively. The samples were filtered and acidified with nitric acid to maintain the pH of the samples less than 2 for

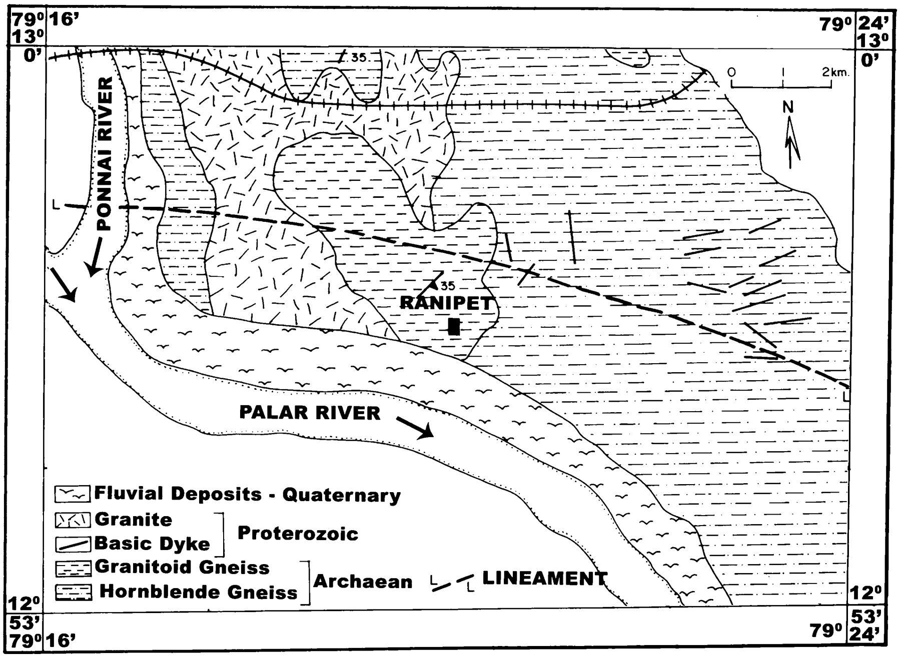


Fig. 2 Geology map of the study area

Table 1 Trace element concentration in (μg/l) in NIST 1640 analyzed by ICP-MS

Element	Certified value	Observed value
^{114}Cd	22.79	22.958±0.483
^{52}Cr	38.6	38.663±0.257
^{63}Cu	85.2	85.405±0.958
^{58}Ni	27.4	27.471±0.197
^{208}Pb	27.89	27.858±0.098
^{64}Zn	53.2	53.004±0.425

trace metal analysis by ICP-Mass Spectrometer. The water sample bottles were thoroughly washed with acid, and then with distilled water in the laboratory before filling the bottle with the sample. The bottle was rinsed to avoid any possible contamination in bottling and every other precautionary measure was taken. The precise locations of the sampling points were determined in the field through the development of the GARMIN 12 Channel Instrument, based on the principles of Global Positioning System (GPS), the exact longitude and latitude of the sampling. The location of the sampling points is shown in Fig. 1.

Sample analysis and instrumentation

Trace metals in the water samples were determined using ICP-MS (Model-ELAN DRC II, Perkin-Elmer Sciex Instrument, USA). The water sample was introduced, after filtration and acidification, into ICP-MS instrument 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 voltages and lens voltage were optimized for a sensitivity of about 50,000 counts/s for 1 ng/ml solution of Indium. The oxide and doubly charged ion formation levels measured on CeO^{+} and Ba^{++} were found to be <2.5%. The instrumental and data acquisition parameters are given elsewhere (Balaram and Gnaneshwar Rao 2003). Calibration was performed using NIST 1640 (National Institute of Standards and Technology, USA), which is a standard reference material for trace elements in natural water, to minimize matrix and other associated interference effects and to check the precision and accuracy of the analysis. NIST 1640 was also analyzed as unknown sample and the results were found to be within permissible errors (<5% RSD) (Table 1). Lower limits of detection for all the elements were better than 1 μg/ml (Buurman et al. 1996; Govil et al. 1997).

Results and discussion

Total heavy metal concentrations

Table 2 gives the maximum, minimum and mean values of heavy metal concentrations. The heavy metals with enrichment levels exceeding the normally expected distribution in water give rise to concern over the suitability for drinking and irrigation (Ferguson 1990; Alloway 1995; Zhang and Selinus 1998; Barzilay et al. 1999; Govil et al. 1999; Romic and Romic 2003; Krishna and Govil 2004). High levels of these elements are observed in some pockets, very close to tannery industries, which indicate that the source of these elements could be the industrial effluents.

Cadmium

Cadmium has a very low maximum contaminant level (MCL) in drinking water, which is 10 μg/l due to its toxicity (WHO 1996). It exists in the aqueous solution in the +2 valence state. Cadmium occurrence in the environment is from both natural and anthropogenic

Table 2 Statistical summary of metal concentrations in water (μg/l)

	Cd	Cr	Cu	Ni	Pb	Zn
Mean	51.1	247.2	95.5	36.7	467.8	3,760.4
Median	17.9	81.9	50.6	30.8	317.8	2,276.6
SD	87.1	338.1	120.7	31.7	456.6	3,796.4
Threshold value[a]	10.0	50.0	30.0	20.0	10.0	300.0
Minimum	0.2	2.4	2.1	1.6	6.4	20.8
Maximum	401.4	1,308.6	535.5	147.0	2,034.4	12,718.0

[a] Maximum permissible concentration as defined by USEPA (2003).

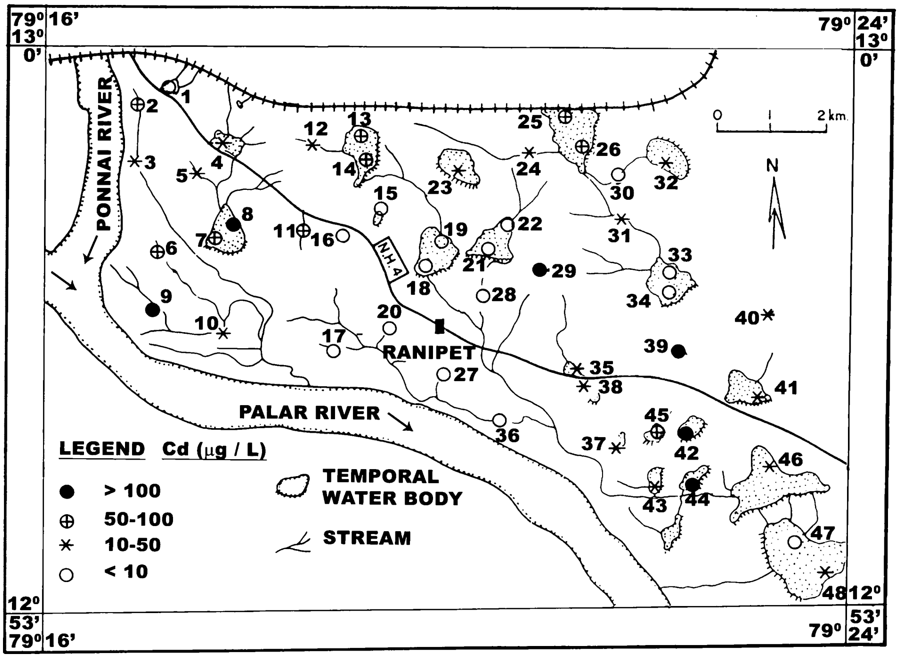


Fig. 3 Distribution of Cadmium (μg/l) in water

sources. Cadmium is a relatively mobile element. Environmental levels are greatly enhanced by the existing industrial operations as Cd is commonly used as a pigment in paint, plastics, ceramics and glass manufacture. The concentration of Cd in the water samples ranges from 0.2 to 401 μg/l, with an average of 51.1 μg/l (Fig. 3). Cadmium is highly toxic, producing symptoms such as nausea, vomiting, respiratory difficulties, cramps and loss of consciousness at high doses. Chronic exposure to metal can lead to anemia, anosmia (loss of sense of smell), cardiovascular diseases, renal problems and hypertension (Mielke et al. 1991; Robards and Worsfold 1991).

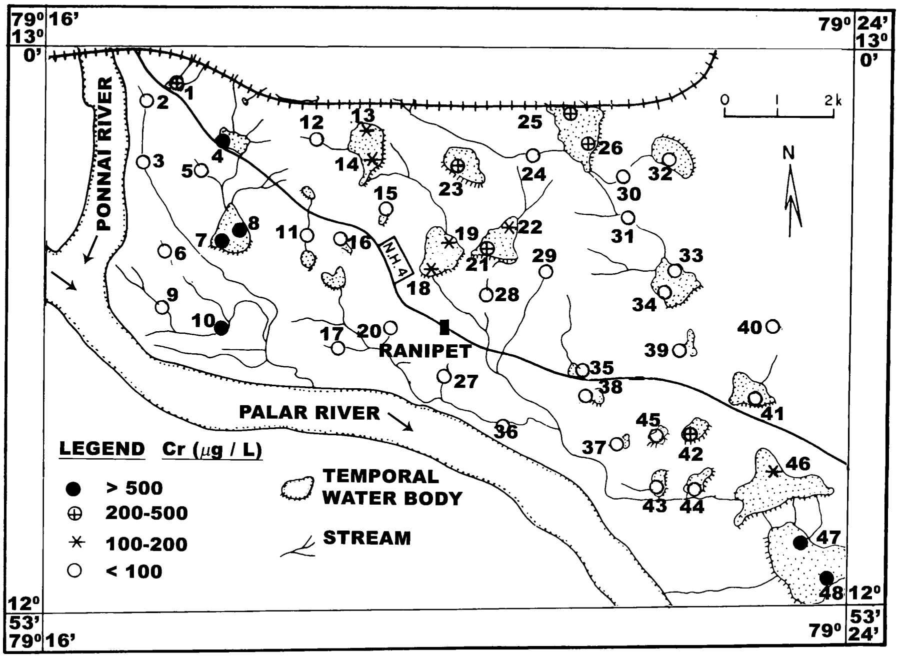
Chromium

Cr (III) is considered as an essential trace element for the maintenance of an effective glucose, lipid and protein metabolism in mammals. On the other hand, Cr (VI) is toxic for biological systems. Chromium is water soluble and extremely irritating and toxic to human body tissue owing to its oxidizing potential and permeability of biological membranes (Anderson 1999). Chromium levels in the study area ranged from 2.4 to 1,308.6 μg/l, with an average of 247.2 μg/l (Fig. 4). The normal range of chromium in water is 100 μg/l (USEPA 2003). As the surrounding rocks are predominantly granite, where chromium concentration is always below 50 mg/kg, it is not possible to derive such high levels of Cr from rocks (Edmunds and Savage 1991; Fendorf 1995; Edmunds and Smedley 1996). Therefore, the source of Cr appears to be anthropogenic from the existing tannery industries, where they are using chromium and its compounds. Chromium is an essential trace element, required for the metabolism of lipids and proteins and to maintain a normal glucose tolerance factor. High doses of chromium cause liver and kidney damage and chromate dust is carcinogenic (SEGH 2001; Mugica et al. 2002).

Copper

Copper values were found to be abnormally high in the study area. The water samples in the area contain in the range of 2.1–535.5 μg/l with an average value of 95.5 μg/l (Fig. 5). The normal threshold value prescribed in water is 30 μg/l. Copper accumulation in the water may be due to the plastic industry, blast furnace, steel and application of agrochemicals in the agro-based industry. Copper is however, characterized by the so called point sources of contamination, namely uncontrolled, active or untended waste dumps. Such places imply great contamination risks

Fig. 4 Distribution of Chromium (μg/l) in water



for water (Romic et al. 2001; Govil et al. 1999). From the aspects of groundwater hazards in this area, only the quantities of metals leached to the solemn/gravel substratum contact are dispensed in the aquifer, thus potentially contaminating it. The transfer of contamination and its dispersion in the aquifer is a very slow process and changes in metal concentration in the upper aquifer layers are not manifested in a short time either. Overdoses of copper may also lead to neurological complications, hypertension, liver, and kidney dysfunctions (Larocque and Rasmussen 1998; Rao et al. 2001; Krishna and Govil 2004).

Fig. 5 Distribution of Copper (μg/l) in water

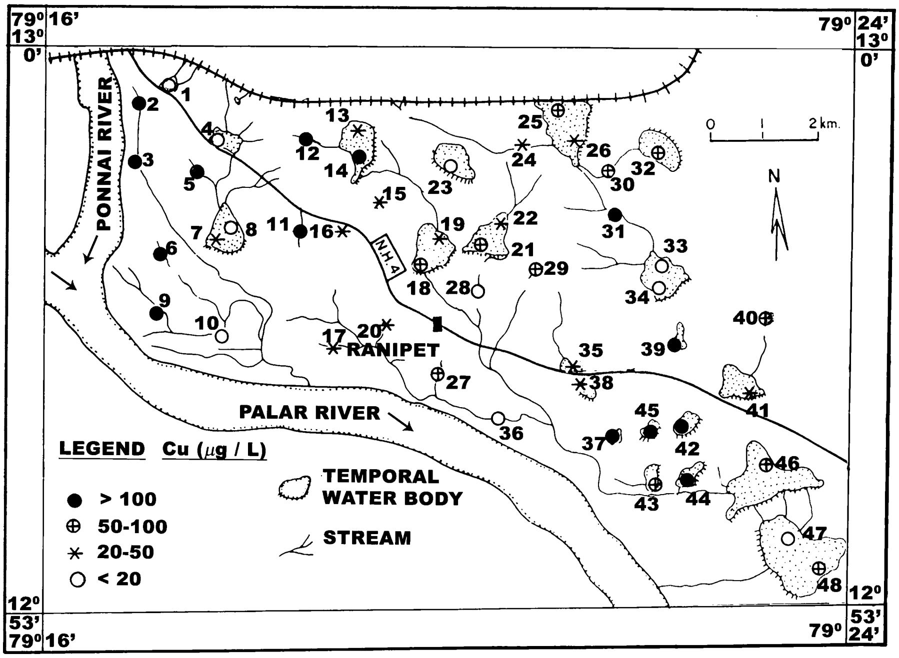


Table 3 Correlation coefficient matrix of trace metals in water

	Cd	Cr	Cu	Ni	Pb	Zn
Cd	1.00					
Cr	0.12	1.00				
Cu	0.18	−0.29	1.00			
Ni	0.18	−0.29	0.73	1.00		
Pb	0.43	−0.08	0.53	0.68	1.00	
Zn	0.28	−0.25	0.20	0.33	0.48	1.00

Nickel

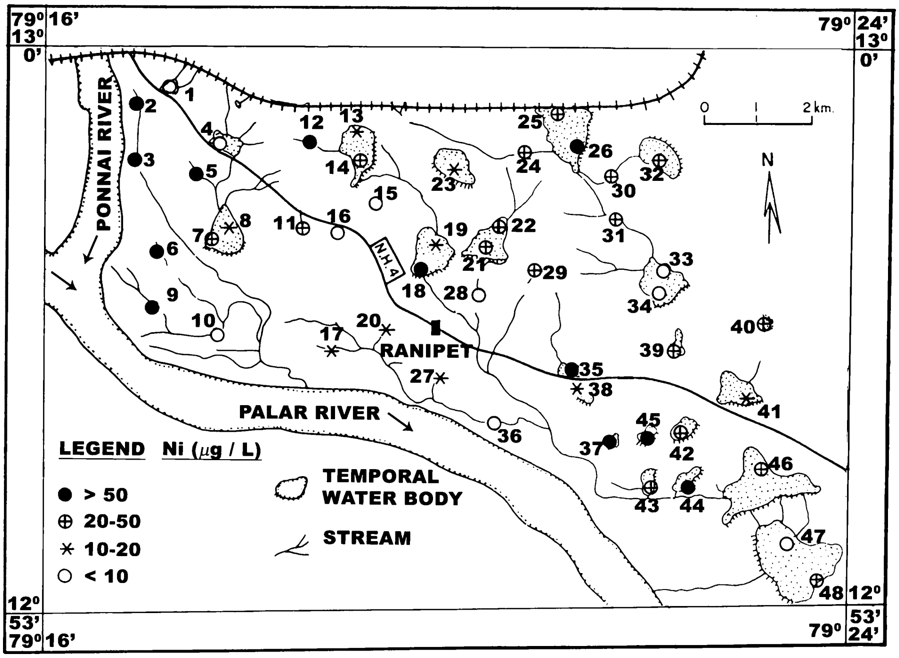
The concentration of nickel ranges from 1.6–147.0 μg/l in the study area with an average of 36.7 μg/l. The normal threshold value prescribed in water is 20 μg/l (WHO 1996). It was found to be associated with Cu and Pb (Table 3) indicating that an increase in Ni concentration in water leads to corresponding increase in the levels of Cu and Pb. Sources of nickel in water include contamination from municipal sewage sludge, wastewater from sewage treatment plants, and groundwater near landfill sites. Industries like ceramics, steel and alloys, electroplating, and refractory are contributors of nickel to water. Anthropogenic sources, such as combustion of fuel or waste released from industries, where nickel is extracted from ore, may have some effect on increase in Ni concentrations, as confirmed in this study.

The geochemical map in Fig. 6 show the spatial distribution of Ni in the study area. The most common adverse effects of nickel exposure are skin allergies, dermatitis. Rhinitis, nasal sinusitis, and nasal mucosal injury, which are reported in workers chronically, exposed to nickel compounds in the study area. The lung is the target organ for nickel toxicity in humans.

Lead

The average concentration of lead in water samples of the study area is 467.8 μg/l and ranges from 6.4 to 2,034.4 μg/l (Fig. 7). Pb was found to be associated with Cu, Ni and Zn (Table 3). Pb–Zn and Cu–Ni are elemental pairs inherently associated with each other because of their intrinsic chemical characteristics. It is obvious that similar relationship is found in the water samples of the study area. Lead is released from smelting, motor-vehicle exhaust fumes and from corrosion of lead pipe work. Lead salts used in ceramic industry, which is located in the study area, have on occasion caused poisoning. Lead solubility is controlled principally by $PbCO_3$ and low-alkalinity

Fig. 6 Distribution of Nickel (μg/l) in water



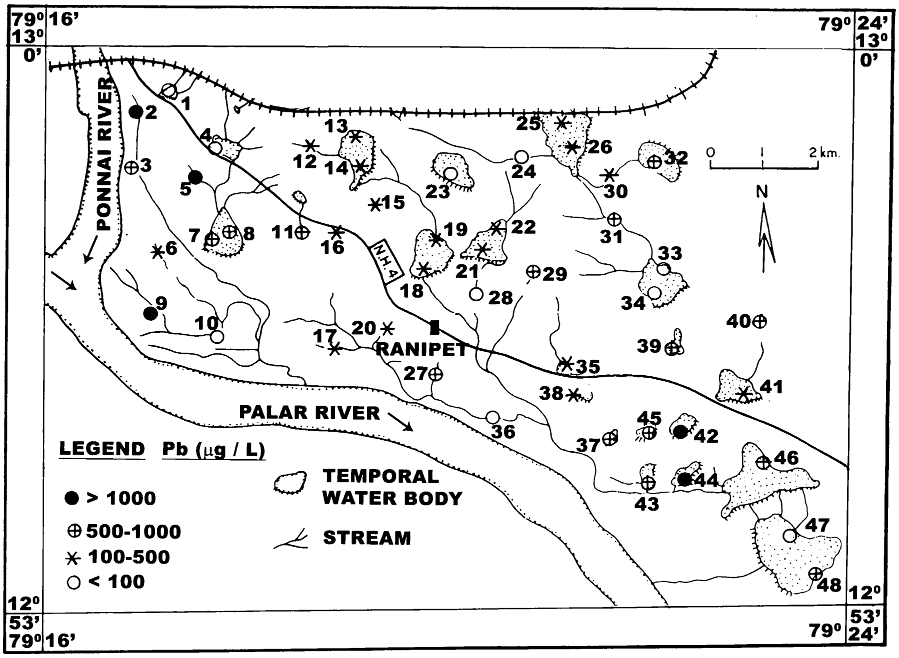


Fig. 7 Distribution of Lead (μg/l) in water

and low-pH waters can have higher Pb concentrations (Hem 1989). The WHO maximum recommended concentration for Pb has recently been reduced from 50 to 10 μg/l because of concerns about chronic toxicity, although its concentration rarely exceeds in natural waters. Lead is a cumulative poison, initiating hypertension, tiredness, irritability, anemia, behavioral changes and impairment of intellectual functions in affected patients (Tebbutt 1983). Long term exposure to lead or its salts (especially soluble salts or the strong oxidant PbO_2) can affect adversely to nervous system and kidneys (Mugica et al. 2002).

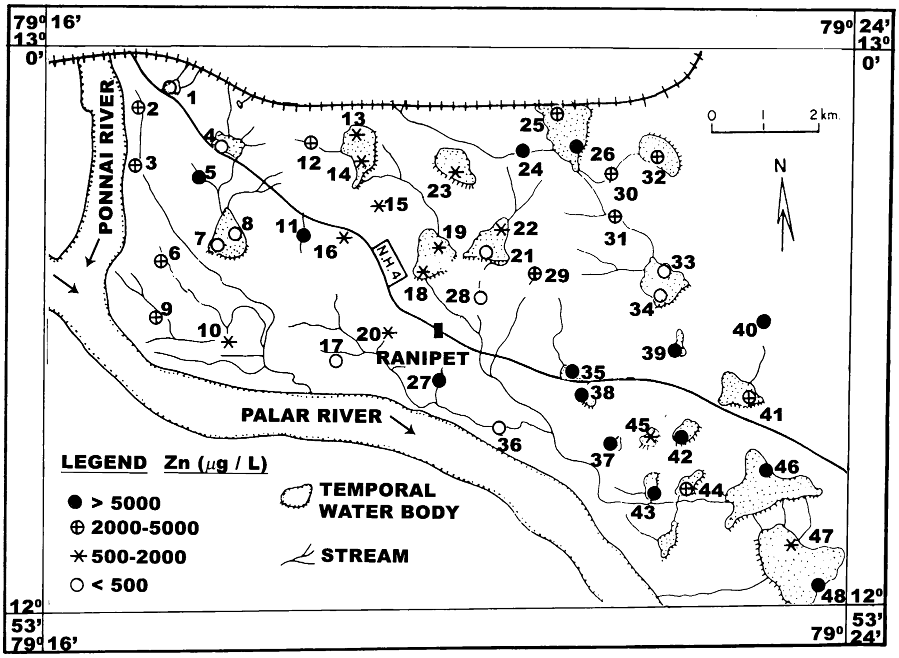


Fig. 8 Distribution of Zinc (μg/l) in water

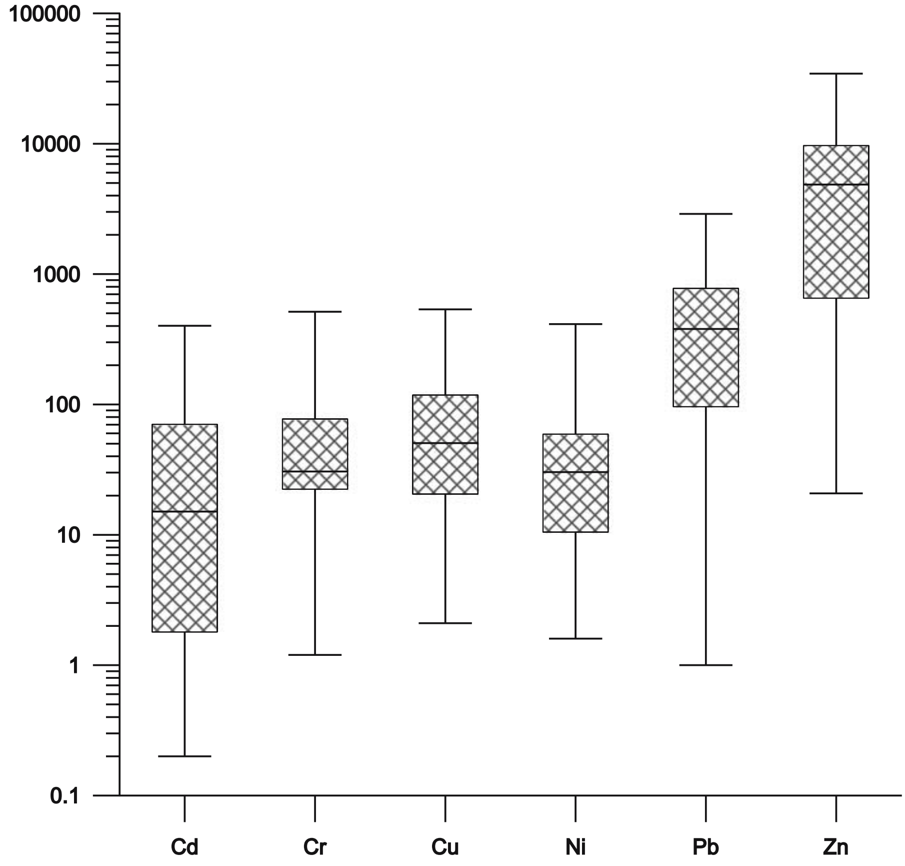


Fig. 9 Heavy metal concentrations (μg/l) and its variations in Box–Whisker plots

Zinc

Zinc belongs to a group of trace metals, which are essential for the growth of humans, animals and plants and are potentially dangerous for the biosphere when present in high concentrations. The main sources of pollution are industries and the use of liquid manure, composted materials and agrochemicals such as fertilizers and pesticides in agriculture (Romic and Romic 2003). The average Zinc concentration in the study area is 3,760.4 μg/l, ranges from 20.8–12,718.0 μg/l (Fig. 8), which is very high. The normal threshold value prescribed in water is 300 μg/l (BIS 1991) and only six samples showed Zinc concentration lower than the normal threshold value (Fig. 9). In India 47% of soils are depleted in Zinc, however the high concentration of Zn are found in the nearby industrial areas, which clearly represents the source to be anthropogenic and not natural (Aswathanarayana 1995). Moreover, it is very unlikely that 3947.9 μg/l of Zn could be derived from acidic rocks present in the study area. Higher contamination of Zn causes hematological disorders and deteriorates human metabolism.

Conclusions

The results of the study show that the water in the Ranipet industrial area is significantly contaminated by Cd, Cr, Cu, Ni, Pb, and Zn, which may give rise to various health problems or diseases. The detected levels of total metal contamination in many of the samples were found to exceed international threshold values. High Cr values confirm the contamination originating from the tannery waste. Ni and Zn showed a strong anthropogenic influence. However, it has been assumed that the anomalous concentrations of Nickel in the vicinity of the highway and industry are of anthropogenic input. Copper is characterized by strongly scattered anthropogenic influence. This particularly relates to the uncontrolled waste dumps and liquid waste from industries. High concentration of Cd and Zn obtained in the water of the study area indicated that the pollution of these heavy metals in the area originated mainly from the industries in the area, either from dumping of industrial waste or from release of uncontrolled effluents on to the ground which contaminate the water bodies and streams present in the study area. There is an urgent need to measure toxic metals in industrial effluents before dumping them in Common Effluent treatment Plant (CETP). It is also desirable to check the outlet water of CETP after treatment to ensure that these toxic metals are not released into the adjoining water bodies. Alternatively, a pipeline can be laid in the area, which would carry the effluents to a safer place for treatment. The water in the area requires remediation as per environmental quality criteria and regular monitoring of toxic metals enrichment in the area.

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