

Research Paper

Assessment of groundwater quality and human health risk associated with chromium exposure in the industrial area of Ranipet, Tamil Nadu, India

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ABSTRACT

Rapid urbanization and industrialization leading to an increase in groundwater contamination is a serious environmental concern in India in recent years. The risk of groundwater contamination is highly pronounced in and around the Ranipet industrial area causing a threat to human health and a balanced ecosystem. In this study, 40 groundwater samples were collected in and around the industrial area of Ranipet which is largely producing chromium (Cr) and chromium-based chemicals. The heavy metal contamination and water quality index (WQI) were evaluated to determine groundwater quality and related human health risk assessment using the model proposed by the USEPA for adults. Based on the WQI range, it is found that more than 50% of groundwater samples are poor and non-potable. Also, the concentration of heavy metal chromium in the groundwater exceeded the acceptable limit, i.e., 0.05 mg/L. The human health risk assessment indicates that the chronic daily intake of groundwater for adults in the study area is in the order of $Cr > Fe > Pb > Cd$ indicating chronic toxicity. It was also observed that the carcinogenic risk is higher than the acceptable limit (1×10^{-6}) as a result of higher chromium intake via ingestion. The outcome of the present study will support the stakeholders in decision-making toward regional sustainable groundwater management.

Key words: carcinogenic risk, chromium exposure, chronic toxicity, groundwater, heavy metal, water quality index

HIGHLIGHTS

- The study conducted in the industrial area of Ranipet depicts the groundwater quality and heavy metal contamination.
- Chromium, the carcinogenic metal, was found to exceed the permissible limit of the World Health Organization standard.
- Human health risk assessment and incremental lifetime cancer risk for the study area show that the community is likely to suffer from carcinogenic health effects.

1. INTRODUCTION

Groundwater is the most precious, essential, and natural resource for life on Earth. It has become a vital source for several activities such as household needs, drinking, industrial, agriculture, and other purposes. For the past few decades, the human water demand is met from the utilization of groundwater. It is a valuable asset for the economic growth of a country, especially in arid and semi-arid regions. Groundwater quality is one of the major considerations of humankind since it is directly connected with human wellness. The assessment of groundwater's physical, chemical, and biological characteristics is important before consumption. In developing countries like India, groundwater contamination has become a serious concern in the post-industrialization era. Over the years, high levels of groundwater contamination are observed in most parts of the country due to the percolation of toxic elements from industrial effluents, landfills, and diffused pollutants from pesticides and fertilizers. Deterioration in groundwater quality can be due to natural processes such as the geological formation of rocks and numerous anthropogenic activities such as improper disposal and release of effluent from industries into surface water bodies which migrates under the action of leaching from unsaturated zone to groundwater effortlessly (Shankar *et al.* 2008; Li *et al.* 2021). Especially in the vastly industrialized and densely populated regions with shallow aquifers, groundwater contamination by anthropogenic wastes is a serious issue (Arumugam & Elangovan 2009; Krishna Kumar *et al.* 2014).

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Numerous studies have been conducted in various parts of India to determine the groundwater contamination and their sources of pollution (Lakshmanan *et al.* 2003; Shankar *et al.* 2008; Rao *et al.* 2011; Brindha & Elango 2012; Selvakumar *et al.* 2017; Acharya *et al.* 2018; Chaurasia *et al.* 2018; Dhawde *et al.* 2018; Adimalla & Qian 2019). Various models have been implemented for assessing groundwater vulnerability and to identify the risk zones across the country (Khan *et al.* 2010; Alam *et al.* 2014; Bhuvaneshwaran & Ganesh 2019; Bera *et al.* 2021) to highlight the requisite groundwater management plans and protection measures. Hence, effective regulation and reclamation of the ecological environment polluted by toxic elements and heavy metals have become a serious scientific issue affecting the health and development of the economy.

A few heavy metals such as copper (Cu), zinc (Zn), and iron (Fe) are essential for human body metabolism, while the excess concentration of other heavy metals like chromium (Cr), cobalt (Co), arsenic (As), and cadmium (Cd) is highly toxic even at low concentration. Higher concentrations of such heavy metals in potable water can cause various health issues such as kidney, liver damage, and gastric cancer (He *et al.* 2018). A higher amount of Cr via food or contaminated water can cause kidney damage, intestinal bleeding, and gastrointestinal stromal tumors (Muhammad *et al.* 2011). The characterization of the heavy metal content in groundwater is necessary to fathom the source, fate, transport, and potential health risk (Lu *et al.* 2018). Health risk assessment is an effective tool used in the qualitative assessment of human health to the existing environment in terms of hazard quotient. As per the study conducted using Sobol sensitivity model-based human health risk evaluation, children face more severe health risks than women and men in the leather tanning industrial region of South India (Karunanidhi *et al.* 2021). Due to the severity of the problem, existing conditions of groundwater quality have to be assessed and an action plan for remediation measures has to be implemented with immediate effect.

The specific study aimed to (i) assess the groundwater quality in the industrial area of Ranipet, (ii) determine the heavy metals concentration in groundwater, and (iii) assess the carcinogenic human health hazards of chromium in adults through oral intake. This study is important as the groundwater in the study region is majorly used for residential and irrigation purposes, and the impact of groundwater contamination on human health has not been investigated in the past. Thus, the present study provides insight into the human health risk assessment due to the presence of high heavy metal concentration in the groundwater of the study area.

2. STUDY AREA

The study region (Figure 1) in the Ranipet industrial area is an industrial hub located near Chennai city in southern India. The Palar river which runs through the district is one of the commonly used sources of drinking water. The major rock formation is Charnockite, Gneiss, Granite, Syentis, Pyroxineite, Sandstone, Shale, and Alluvium with basic sedimentary rock type. Due to the establishment of the industrial complex by the State Industries Promotion Corporation of Tamilnadu Ltd (SIPCOT) at Ranipet, the region consists of various sectors of sodium and chromates manufacturing units; tanning industries, pharmaceutical companies, and common effluent treatment plants. Residents and industries mostly rely on the groundwater for domestic, farming, industrial activities, and other purposes. The growing industrial activities in the study region with negligible environmental concern leading to improper disposal of effluent causes the contamination of groundwater by heavy metals. Also, the heaps of chromium sludge which are left behind the premises of closed down factories in the old SIPCOT sector for more than two decades without proper disposal plan and detoxification of water gathers dust and contributes to the increased chromium content in groundwater which is higher than the permissible limit. The carcinogenic elements present in these effluents will have an adverse health effect on the population of the region when it is exposed for a long duration.

3. MATERIALS AND METHODS

3.1. Sample collection and analysis

Forty groundwater samples were collected that include 30 existing borewells, 6 open wells, and 4 hand pumps covering the entire area of interest shown in Figure 1 as per standard EPA procedures. The location details of the sample collection were collected using handheld GPS. Water samples were collected after pumping stagnated water for 10 min from the bore wells, open wells, and hand pumps (see Figure 2(a)–2(c)). Each sample was collected in prewashed 1,000 ml polyethylene bottles. It was then secured, labeled (as shown in Figure 2(d)), and carefully transported to the laboratory.

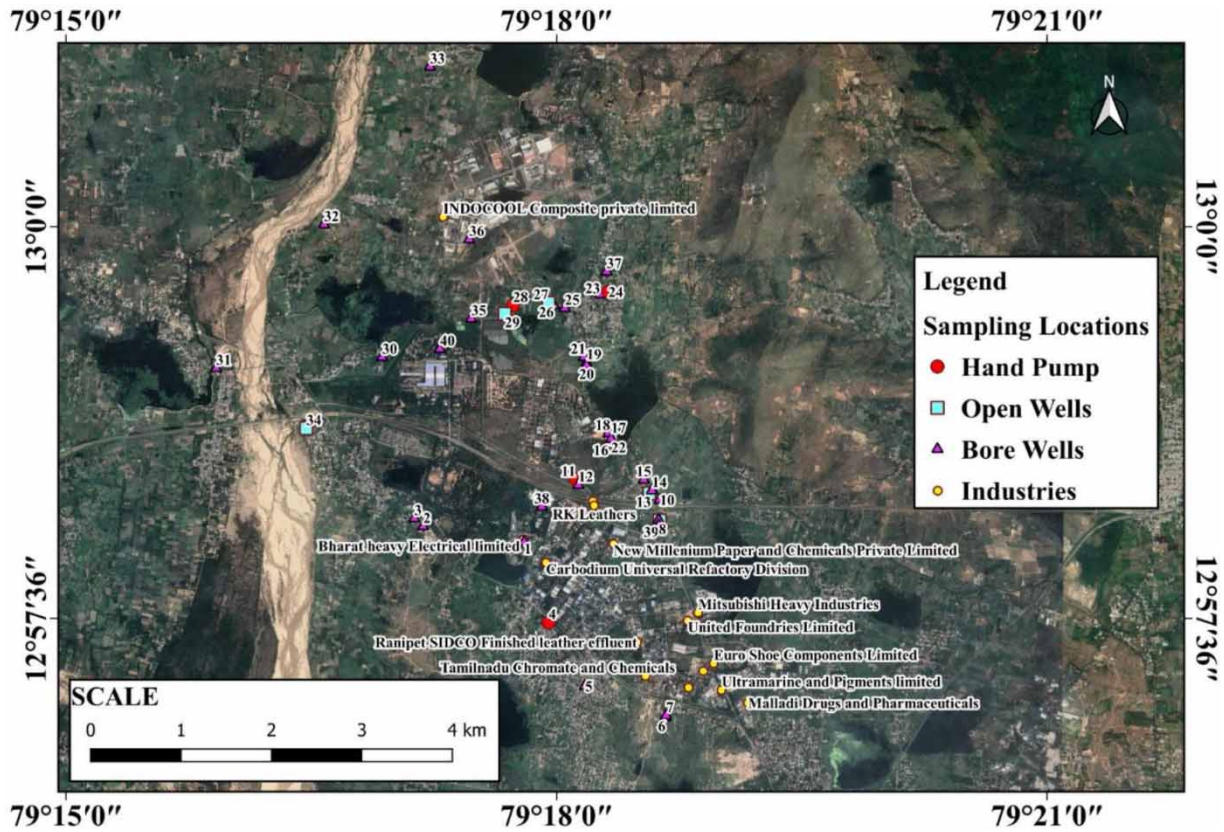


Figure 1 | Study area.

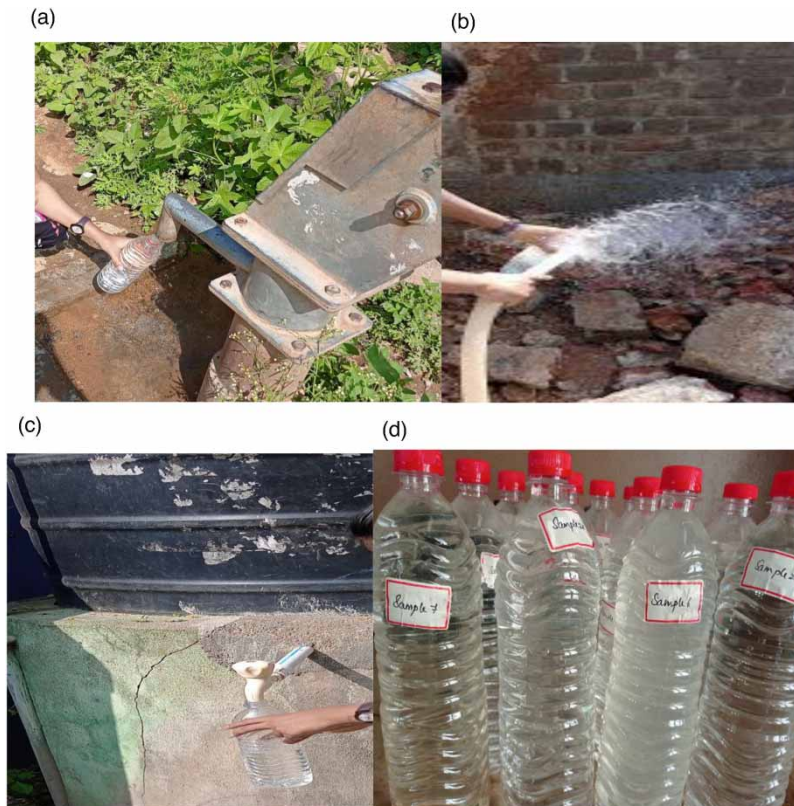


Figure 2 | Groundwater samples collected from various sources in the study region: (a) hand pump, (b) open well, (c) borewell, and (d) collected and labeled samples.

Physiochemical parameters for the samples were determined using standard methods. The samples collected were analyzed within 5 days. At the laboratory, the titrimetric method was performed to determine total hardness (TH), bicarbonate (HCO_3^-), carbonate (CO_3^{2-}), chloride (Cl^-), calcium (Ca^{2+}), and magnesium (Mg^{2+}) as shown in Figure 3. The samples collected were investigated for possible heavy metals. For the analysis of heavy metals, a few drops of concentrated HNO_3 acid were added to the water samples to maintain the pH ~ 2 and also to avoid the precipitation of heavy metals. Fifty milliliters of each sample were taken in a conical flask and 10 ml of concentrated HNO_3 solution was added and heated for digestion. It was then allowed to cool and was filtered using Whatman filter paper no. 42. The concentration of chromium (VI) was then measured for the filtrate spectrophotometrically at 540 nm using the diphenylcarbazide colorimetric technique.

The accuracy of charge balance error (%CBE) for each sample is calculated using the following formula:

$$\% \text{CBE} = \frac{\sum \text{cations} - \sum \text{anions}}{\sum \text{cations} + \sum \text{anions}} \times 100 \quad (1)$$

where all the ions are expressed in meq/L. The calculated %CBE was found to be below $\pm 10\%$ which indicates the accuracy of the data.

Then, the quality of the collected groundwater samples for drinking purposes is estimated based on parameters like water quality index (WQI; Tyagi *et al.* 2020) and its deleterious effect on human health by human health risk assessment (Adimalla & Qian 2019). The WQI is determined as

$$\text{WQI} = \frac{\sum Q_i W_i}{\sum W_i} \quad (2)$$

$$Q_i = \left[\frac{V_i - V_0}{S_i - V_0} \right] \times 100 \quad (3)$$

$$W_i = \frac{K}{S_i} \quad (4)$$

$$K = \frac{1}{\sum \left(\frac{1}{S_i} \right)} \quad (5)$$

where Q_i is a rating based on the concentration, W_i is the unit weight of each water quality parameter, S_i is the standard i th parameter, V_i is the calculated concentration of the parameter in the sample, V_0 is the standard value of parameters in pure water, and K is the proportionality constant. The range of WQI values as depicted in Table 1 determines the water quality status for potability.

Heavy metal enters the human body through various routes such as oral, dental, and inhalation. Oral intake has a high influence when compared to other means. So, this study considers the human health effects via oral intake only, and



Figure 3 | Testing of samples in the laboratory.

Table 1 | Water quality index and water quality status (Bengal *et al.* 2007)

Water quality index value	Water quality status
0–25	Very good
26–50	Good
51–75	Average
76–100	Poor
>100	Non-potable

others are neglected. The Chronic Daily Intake (CDI) of heavy metal through oral consumption considering various parameters (as given in Table 2) and hazard quotient are estimated by the following formulas.

$$CDI = \frac{C \times CR \times EF \times ED}{BW \times AT} \quad (6)$$

$$HQ = \frac{CDI}{RfD} \quad (7)$$

$$ILCR = CDI \times CSF \quad (8)$$

RfD is the reference dose of heavy metals, and *CSF* is the cancer slope factor which is defined as the health risk caused by the exposure to an average amount of 1 mg/kg/day of carcinogenic element (USEPA 2014; Teng *et al.* 2015).

Heavy metal in water is a serious threat to organisms which leads to both gastric cancer and non-carcinogenic effects through consumption. The United States Environmental Protection Agency (USEPA) defined incremental lifetime cancer risk to be a cumulative probability of an individual to develop any type of cancer over a lifespan as the result of exposure to heavy metals (Mohammadi *et al.* 2019). Non-carcinogenic and carcinogenic risks are calculated and it is defined that their values should not exceed 10^{-6} (USEPA 2004).

4. RESULTS AND DISCUSSION

4.1. General hydrochemical parameters

The physiochemical parameters (as given in Table 3) of the collected groundwater samples in the study area were analyzed. The basic parameter pH of the groundwater samples collected is slightly acidic and alkaline ranging from 6.35 to 8.51 with an average of 7.22. All the samples collected were found to be within the World Health Organization (WHO) standard (BIS 2012). However, pH does not usually have a direct impact on human health. The concentration of electrical conductivity which is the measure of free ions concentration in groundwater varies from 230 to 2,315 $\mu\text{S}/\text{cm}$. This may be due to the higher concentration of free-moving ionic concentration. Hardness in water is generally caused mainly by calcium and magnesium ions. The total hardness in the groundwater samples ranged from 85 to 314 mg/L with a mean value of 164.75 mg/L. 25% of the samples exceeded the standard of 200 mg/L, while the TDS for the study area varies from 212 to 1,905 mg/L with an average value of 751.3 mg/L. According to the WHO, drinking water with a TDS level less than 500 mg/L is always

Table 2 | Parameters for determining chronic daily intake (CDI) for the drinking water

Parameters	Symbols	Units	Values
Consumption rate	CR	L/day	2.2
Exposure frequency	EF	day/year	365
Exposure duration	ED	years	70
Body weight	BW	kg	70
Average time	AT	days	25,550
Concentration of heavy metal	C	mg/L	Study values

Table 3 | Physiochemical parameters of groundwater

Parameter	Units	Minimum	Maximum	Mean	WHO (2008)
pH	–	6.35	8.51	7.22	6.5–8.5
EC	µS/cm	230	2,315	1,067.65	–
TDS	mg/L	212	1,905	751.30	500
TH	mg/L	85	314	164.75	200
TA	mg/L	115	426	265.77	120
Ca ²⁺	mg/L	37	253	107.70	75
Mg ²⁺	mg/L	24	99	56.80	50
Cl ⁻	mg/L	92	907.3	298.98	250

considered to be good for human health, whereas water with a TDS level greater than 500 and 1,000 mg/L is classified as hard and brackish water, respectively. So, 12.5% of total samples in the study region are found to be unpalatable for human health. Even though the major ions Ca²⁺, Mg²⁺, and Cl⁻ are essential for human health, the increase in the concentration of these ions will lead to serious disorders in metabolic activities. The concentration of Ca²⁺, Mg²⁺, and Cl⁻ in the samples varied from 37 to 253 mg/L, 24 to 99 mg/L, and 92 to 907.3 mg/L, respectively. In this study, 67% of samples exceeded the permissible limit for Ca²⁺, 42% exceeds the limit of Mg²⁺ and 52% of samples exceed the limit of Cl⁻. Sampling results presented in Table 6 should show more statistical parameters such as standard deviation, skewness, and confidence intervals.

4.2. Assessment of groundwater quality for drinking

In the present research, the WQI varies from 42.71 to 117.97 with a mean value of 78.54. Based on the WQI about 55% of water samples collected were poor and non-potable (Table 4). This result strongly implies that the groundwater in the study area is highly contaminated by heavy metals due to the effluent discharge from industries.

4.3. Heavy metals in groundwater

Chromium occurs in higher concentrations of wastes from electroplating, paints, dyes, chrome plating, tanning, and leather industries. A trace amount of these metals are important to human health. However, in higher concentrations, they can be toxic and cause serious health effects. The recommended level of chromium in drinking water is 0.05 mg/L. The study shows a variation from 0.012 to 40.5 mg/L while the other heavy metals such as Fe, Cd, and Pb were found to be within limits given by the WHO (Table 5), and the statistical data such as standard deviation, skewness, and confidence intervals for the heavy metals are presented in Table 6. The 95% confidence interval for all the parameters states that the values are true for all the samples collected. As the encompassing rocks are crystalline rocks of granites and basic dykes and do not influence the groundwater contamination (Srinivasa Gowd & Govil 2008), the intrusion of a high concentration of chromium into the groundwater appears to be mainly due to effluents from the surrounding chromium-based industries (Figure 1). And also, the groundwater samples collected at the proximity of the industrial locations showed the presence of Cr⁶⁺ exceeding the permissible limit. This confirms the source of higher Cr⁶⁺ concentration is anthropogenic from existing tanneries,

Table 4 | Water quality index of collected groundwater samples

Water quality index	Water quality status	Number of samples	% of samples
0–25	Excellent	–	–
26–50	Good	4	10
51–75	Moderate	14	35
76–100	Poor	20	50
>100	Non-potable	2	5

Table 5 | Heavy metal concentration in groundwater (all units are in mg/L)

Heavy metals	Minimum	Maximum	Mean	WHO (2008)
Cr ⁶⁺	0.01	40.52	4.173	0.05
Fe	0.013	0.57	0.19	0.3
Cd	0.0001	0.004	0.001	0.01
Pb	0.0001	0.053	0.006	0.1

Table 6 | Statistical data for various physiochemical parameters and heavy metals

Parameters	Standard deviation	Skewness	Confidence interval	
			Lower	Upper
pH	0.618	0.366	7.05	7.413
EC	527.840	0.402	913.48	1,239.33
TDS	396.954	1.415	639.68	869.733
Cl ⁻	196.528	1.481	239.80	362.580
Ca ²⁺	54.212	1.063	93.153	124.949
Mg ²⁺	20.502	0.372	50.051	62.999
TH	60.722	0.966	147.477	183.6744
TA	89.719	0.018	239.235	292.271
Cr	11.291	2.555	0.997	8.007
Fe	0.126	0.870	0.1540	0.233
Cd	0.001	1.125	0.00083	0.0014
Pb	0.011	3.145	0.0032	0.010

chromium-based industries, and their improper waste disposal. Discharge of tannery effluents into open land leads to leaching and seepage of contaminants into the groundwater over a long period accounting for an elevated level of chromium.

4.4. Evaluation of health risk assessment

The result analysis of the collected groundwater samples considering various parameters indicates that the groundwater is severely contaminated by Chromium metal. Based on the WQI, it is found that only 10% of the water samples are suitable for drinking purposes, while others are of poor quality and non-potable. Hence, this will have an adverse impact on the health of families in and around the region through regular consumption and it is essential to conduct the human health risk analysis for the study area.

Considering the concentration of various heavy metals and the drinking water intake, the carcinogenic health risk assessment is performed as per the model suggested by the USEPA. The hazard quotient and incremental lifetime cancer risk are calculated and presented in Table 7. The carcinogenic risk heavy metals were found to be within limits except for chromium. The incremental lifetime cancer risk via oral intake for chromium varies from 0.01288 to 0.05218 with an average value of 0.00537. This is significantly higher than the permissible limit (1×10^{-6}) recommended by the Ministry of Environmental Protection (USEPA 2004).

Elevated concentration of chromium in groundwater can be hazardous to human health if consumed over a long period of time and the main potential human health threats are cancer, respiratory problems, anemia, gastrointestinal hemorrhage, ulcers, liver, and kidney failure.

Table 7 | Hazard quotient and incremental lifetime cancer risk from exposure to heavy metals

Samples	Hazard Quotient				Incremental lifetime cancer risk		
	Cr	Fe $\times 10^{-5}$	Cd $\times 10^{-9}$	Pb $\times 10^{-8}$	Cr $\times 10^{-5}$	Cd $\times 10^{-8}$	Pb $\times 10^{-7}$
1	0.00012	0.104	6.285	2.244	1.546	1.917	2.671
2	0.38238	5.971	0.6285	3.367	Below the desired limit (BDL)	0.1917	4.007
3	0.22523	BDL	0.0125	4.489	5.154	0.3834	5.342
4	0.42428	BDL	0.0251	0.2244	BDL	0.7668	26.71
5	0.00523	4.399	0.0125	0.5612	BDL	0.3834	66.78
6	0.30275	BDL	0.6285	2.469	BDL	0.1917	2.938
7	0.39180	BDL	0.6285	5.387	BDL	0.1917	6.411
8	0.00209	BDL	0.6285	2.244	BDL	0.1917	2.671
9	0.00628	3.771	0.01257	5.612	BDL	3.834	6.678
10	0.00041	4.399	6.285	2.244	5.154	0.1917	2.671
11	0.00052	1.361	0.0125	1.167	6.442	0.3834	13.89
12	0.00020	1.676	0.6285	3.142	2.577	0.1917	3.739
13	0.00020	BDL	0.0188	8.305	2.577	0.5751	9.884
14	0.00010	BDL	0.1257	0.1346	1.288	3.834	16.02
15	0.00052	BDL	0.6285	0.1212	6.442	0.1917	14.42
16	0.00010	2.514	0.3142	8.979	1.288	9.585	10.68
17	0.00031	1.676	0.6285	5.387	3.865	0.1917	6.411
18	0.00041	BDL	0.01257	6.734	5.154	0.3834	8.014
19	0.00010	0.1047	6.285	2.244	1.288	1.917	2.671
20	0.00041	0.4190	0.01885	4.714	5.154	0.5751	5.60
21	0.00020	BDL	0.6285	0.1795	2.577	0.1917	21.3
22	0.00010	0.1047	0.2514	0.9203	1.288	7.668	0.0109
23	0.00031	0.2095	6.285	0.0118	3.865	1.917	0.01415
24	0.00026	0.4190	0.1885	0.6285	3.221	5.751	0.7479
25	0.00031	BDL	6.285	2.244	3.865	1.917	2.671
26	0.00104	BDL	0.1257	3.367	BDL	3.834	4.007
27	0.00020	BDL	0.6285	9.42	2.577	0.1917	0.01121
28	0.00010	BDL	0.6285	9.652	1.288	0.1917	0.01148
29	0.00010	1.885	0.1257	6.285	1.288	3.834	7.479
30	0.00041	BDL	6.285	2.693	5.154	1.917	3.205
31	0.00020	BDL	0.1885	2.244	2.577	5.751	2.671
32	0.00010	1.676	0.6285	8.979	1.288	0.1917	0.01068
33	0.00020	BDL	0.0125	6.734	2.577	0.3834	8.014
34	0.00041	BDL	0.6285	7.183	5.154	0.1917	8.548
35	0.00031	BDL	0.0125	2.244	3.865	0.3834	2.671
36	0.00020	2.199	0.6285	0.022	2.577	0.1917	2.671
37	0.00031	1.466	6.285	2.244	3.865	1.917	2.671
38	0.00020	BDL	0.0125	5.387	2.577	0.3834	6.411
39	0.00020	BDL	0.6285	4.489	2.577	0.1917	5.342
40	0.00020	BDL	0.6285	6.734	2.577	0.1917	8.014
Minimum	0.00010	0.1047	6.285	0.02244	1.288	1.917	0.2671

(Continued.)

Table 7 | Continued

Samples	Hazard Quotient				Incremental lifetime cancer risk		
	Cr	Fe $\times 10^{-5}$	Cd $\times 10^{-9}$	Pb $\times 10^{-8}$	Cr $\times 10^{-5}$	Cd $\times 10^{-8}$	Pb $\times 10^{-7}$
Maximum	0.42428	5.971	0.0251	1.189	0.05218	7.668	1.415
Mean	0.04372	1.998	0.6882	1.388	0.00537	2.099	1.652

5. CONCLUSION

In this research, 40 groundwater samples were taken from the industrial area of Ranipet to assess the groundwater quality using the WQI and the human health risk through oral intake. The following conclusion can be drawn from this work.

1. The groundwater collected was both acidic and alkaline. This was mainly due to the dominant carbonate (HCO_3^-). The main carcinogenic contaminant in the study area was found to be chromium (Cr).
2. Based on the WQI, only 10% of groundwater samples were found to be suitable for drinking purposes. In addition, based on the heavy metal analysis for the collected samples, the chromium concentration was found to exceed the permissible limit. This is due to the proximity and improper disposal from such chromium-based industries in and around the study area.
3. The human health risk assessment shows that adults are likely to suffer from the cancer-causing effects due to the higher content of Chromium in groundwater samples. Thus, exposure to such heavy metal for a longer duration of time cause the chance of higher carcinogenic risk in an adult population.

Accumulation of heavy metals in drinking water and subsequent intake might pose a potential risk to human health. Also, chronic long-term exposure to high doses of chromium, especially Cr^{6+} promotes carcinogenicity and mutagenicity among humans and animals through their oral intake. With growing awareness and scientific knowledge concerning environmental and health consequences due to contaminants in drinking water, the need for an integrated framework for groundwater quality management has gained serious attention. Government should invest in valuation and formulate an action plan to understand the current scenario of damages made to natural resources and also to reverse the effects of severe groundwater contamination due to anthropogenic activities. A fundamental shift in federal environmental policies inclusive of regulations on risk reduction and resource valuation will prevent environmental degradation and its adverse impacts in the near future. Thus, the findings of this study will help regulatory authorities and stakeholders in decision-making toward regional sustainable groundwater management and remediation of contaminated sites promoting a healthier environment.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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