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Nitrate Contamination of Groundwater in Selected Industrial and Non Industrial Sites, Coimbatore District, Tamil Nadu: Exposure and Health Risk Assessment

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ABSTRACT

Groundwater resources are essential for sustaining human health, agricultural productivity, and industrial growth. In recent years, anthropogenic influences have caused a sharp decline in ground water quality, which is also having an impact on human health in many parts of India. It is crucial to analyze the risk of contamination to human health and the quality of the water supply. Coimbatore is one of the rapidly growing industrial areas in South India. The industrial areas of ground water have a significant impact on ground water quality due to anthropogenic activities such as discharge of Industrial waste, chemicals and pollutants into the ground which make water unsuitable for human and industrial use. Nitrate is the emerging contaminant which can cause a deadly disease methehemoglobinemia. Hence the present study is focused to analyze the ground water of selected industrial and non industrial sites of Coimbatore city. Nitrate concentration ranged from 32.90 to 88.13 mg/l in the samples industrial and non-industrial sites, and over 37% of both the sites had nitrate levels that above the Bureau of Indian Standards' and WHO's acceptable limits (45 mg/l and 50 mg/l, respectively). The USEPA human health risk assessment tool was used to examine the potential health hazards of increased nitrate intake for both adults and children.

Key words : *Ground water quality, Industrial and Non industrial sites, Nitrate contamination, Methemoglobinemia, Health hazard index, South India*

Introduction

Ground water is the ultimate source for consumption and agricultural purposes. Nearly 1/3 of India's population depends upon ground water for consumption, industrial needs and agricultural activities. Variation of groundwater quality in an area is a function of physical and chemical parameters that are greatly influenced by geological formation and anthropogenic activities (Banerjee and Prasad, 2020). The industrial activities and the influence of humans over the past decades have increased the need for groundwater more than ever before (Kaviarasan *et al.*, 2016). Nitrate contamination is an emerging issue in India. One third of states in India have excess nitrate in their drinking water, and Tamil Nadu is one of the state. Coimbatore is also called as the 'Manchester of South India', as it contains large number of textile and manufacturing industry. Therefore the ground water is influenced by the anthropogenic activities. Nitrate is naturally present in the atmosphere as plants and lightning storms (Kumar *et al.*, 2019). The nature of water assets are affected due to the anthropogenic activities like agricultural fertilizers, sewages, septic tanks, animal dung, residential wastewater and industrial waste water. As per WHO and BIS, the maximum permissible limit of nitrate is 50 mg/l in drinking water (WHO, 2017). Therefore nitrate become one of the widespread contaminants in ground water and the ingestion of nitrate will lead to methemoglobinemia, which is commonly called as blue baby syndrome. Children's are more prone to this disease; this will affect the ability of blood to carry oxygen throughout the body. It also cause other diseases such as thyroid dysfunction, breathing problems, nuisances, tiredness and multiple sclerosis that can result from high nitrate intake (Ahada and Suthan, 2018).

The USEPA Human health risk assessment tool is used to assess health hazards of various pollutants. Number of studies have been done on the risk to human health associated with nitrate contamination of ground water in India. And the reports also said that the health of the women and children are profoundly affected for health risks from elevated levels of nitrate (Karunanidhi *et al.*, 2019 and Sajil Kumar, 2020).

Hence, the present study is focused to understand the nitrate contamination in selected Industrial and Non Industrial sites of Coimbatore and to determine the Human health risk of nitrate exposure for male, female and children's with the help of USEPA Health Risk assessment tool.

Materials and Methods

Study area

The investigated region lies in the district of *Coimbatore* in Tamil Nadu province, South India. The city is situated at 11° 00′ N latitude and 77° 00′ E longitude and occupies an area of about 3104 km^2. Groundwater is commonly found in fractures and fault regions in the hard rocks and in porous media in the alluviums associated with river systems. Dug wells and bore wells are the essential methods of water extraction.

Methodology

A total of 10 ground water samples were collected from different industrial and non industrial sites from various places of Coimbatore. The selected in-

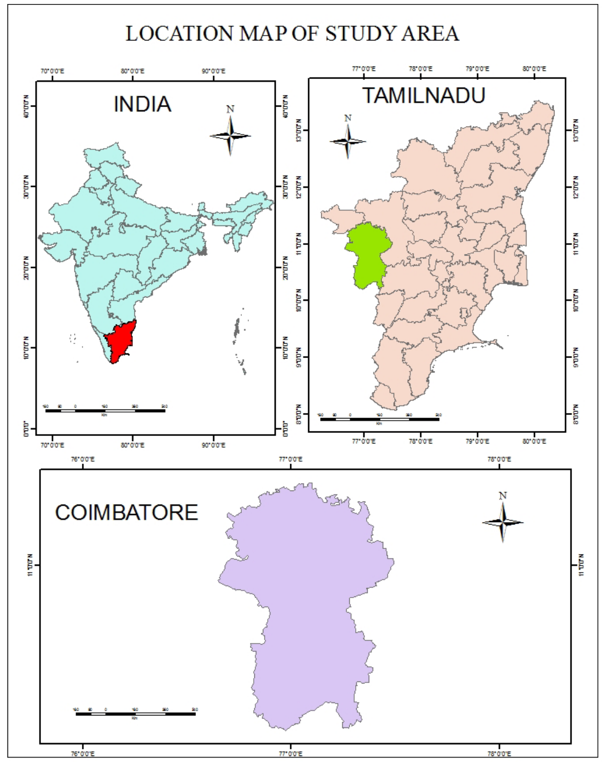

Fig. 1. Location map of study area

dustrial sites are Athipalayam, Arasur, Ganapathy, Peelamedu and Saravanampatti. The non industrial sites are Nava India, Avarampalayam, Ondipudhur, Chinniyampalayam and Neelambur. Water has pumped out before sampling to stabilize the physicochemical parameters. Samples were collected in pre-cleaned polyethylene bottles, sealed, and brought to the laboratory for analysis. Samples were kept at 4 °C. The pH and EC of the sample was determined using pH meter (Systronics pH system 361) and EC meter (Systronics conductivity TDS meter 307). The samples were analyzed for bicarbonate (HCO^{3-}), chloride (Cl^-), sulfate (SO_4^{2-}), nitrate (NO^{3-}), calcium (Ca^{2+}), sodium (Na^+), and potassium (K^+) based on the American Public Health Association (APHA 2012) procedures in the laboratory. The Ca^{2+} and Mg^{2+} were determined using the standard Ethylene di-amine tetra acetic acid (EDTA) titration method and Na^+ and K^+ by flame photometer (Systronics Flame Photometer 128). A standard solution of sulfuric acid (H_2SO_4) and methyl orange indicator were used to determine the HCO^{3-}, while the concentration of Cl^- was determined by using silver nitrate ($AgNO_3$) and potassium chromate (K_2CrO_4) indicator. A UV–visible spectrophotometer was used to estimate the Nitrate and Sulphate (Systronics Visiscan 167).

Human Health Risk Assessment

Human beings were exposed to the toxic elements, mainly by inhalation, ingestion, and dermal pathways. In this study, the health impact of nitrate on males, females, and children has been investigated. The standard procedures and reference values suggested by the United State Environmental Protection Agency (USEPA, 2014) are used in the health risk assessment. Exposure risks by ingestion and dermal pathways were calculated using equation 1 and 2

$CDD_{in} = (C_{water*}IR_*EF_*ED)/(BW_*AT)$ .. (1)

$CDD_{de} = (Cwater_*SA_*KP_*EF_*\ ED_*\ ET_*CF)/(BW_*AT)$ (2)

In the equation CDD_{in} and CDD_{de} represents the chronic daily dose by ingestion and dermal effects respectively, C_{water} is the concentration of nitrate in ground water (mg/l).

IR - ingestion rate of water

EF - exposure frequency

ED - exposure duration in years

BW - body weight in kg

AT - average residence time in days / years

SA - exposed skin area in cm^2 (adults = 16,600 cm^2 ; children = 12,000 cm^2)

KP - dermal permeability coefficient for water (0.001, no unit)

ET - water exposure time in hours/day (0.4 h/day for adults and children)

CF - conversion factor (0.001 for adults and children).

The hazard quotient (HQ) of Nitrate exposure via ingestion can be calculated by

$HQ_{in}=CDD_{in}/RfD$ .. (3)

$HQ_{de}=CDD_{de}/RfD$ .. (4)

HQ_{in} is the ingestion-based hazard quotient

HQ_{de} is the dermal-based hazard quotient

RfD is the reference dose of NO^{3-}, i.e., 1.6 mg/kg/day (USEPA 2014).

HQ <1 has negligible non-carcinogenic effects, while on the other hand HQ >1 shows significant non-carcinogenic health hazards.

Hazard index (HI) is the overall risk of exposure via both digestion and dermal pathways.

$HI=HQ_{ingestion}+HQ_{dermal}$

The other hand Hazard Quotient HQ >1 shows significant non-carcinogenic health hazards (Adimalla *et al*., 2018).

Results and Discussion

The physico- chemical parameters of the ground water samples are presented in the Table 1.

pH, Electrical Conductivity, TDS

The pH requires the optimum range of 6.5-8.5 (WHO 2017). The samples from this study showed the pH value of 6.89-7.86, with the average ranges 7.4. This indicates that the study region showed slightly alkaline condition. The carbonates and bicarbonates in the aquifers are the major controlling factors for pH value (Saha and Paul, 2019).

The electrical conductivity is a measure to the capacity of water to conduct electrical current, which is directly related to the concentration of salts dissolved in water. In this study the electrical con-

Table 1. Physio chemical parameters of Industrial and Non Industrial Sites

Sample site	1	2	3	4	5	6	7	8	9	10
pH	7.08	6.89	7.42	7.48	7.35	7.57	7.67	7.68	7.83	7.86
EC	4300	5700	2200	2000	2100	1800	3300	2000	4100	4200
TDS	2500	3400	1300	1200	1600	1100	1900	1400	2400	1500
HCO_3	80	76	86	96	84	76	82	44	70	84
TH	4440	5883	1480	2035	2590	1184	1110	925	5106	1295
Ca	1620	2100	375	795	630	570	615	330	525	585
Na	44.8	86.2	31.8	67.1	54.2	29.2	25.1	13	24.3	27.3
K	32.68	61.79	27.6	18.36	15.59	15.91	25.51	16.59	53.4	19.24
Cl	146.7	888.7	277.2	187.5	171.2	220.1	195.6	464.7	953.9	399.5
SO_4	90	118	20.5	27.3	58.3	18	62	52	43	29
P	0.02	0.01	0.01	0.02	0.01	0.01	0.03	0.03	0.04	0.03
NO_3	72.35	88.13	77.61	46.05	40.79	46.05	59.20	48.68	32.90	56.57

ductivity varied from 1800-5700 µS/cm, which is higher than the permissible limit according to WHO 2017. The higher concentration of salts in rocks gets dissolved and it may be the cause of higher electrical conductivity (Karthik *et al*., 2019).

The total concentration of dissolved substances in water is Total dissolved solids. It consists of both inorganic salts and traces of organic matter. The value of TDS ranges from 1100- 4370 ppm. The high TDS may be due to the percolation of channel water containing solids, agricultural wastes, and industrial seepages (Udhaya Kumar, 2016).

Chloride, Alkalinity, Sulphate

The inorganic fertilizer usage, road salts, animal feeds, effluents from Industrial activities and septic tanks are the major causes of chloride concern in groundwater. In the present study the chloride value was observed in the range of 146 to 953 mg/l, with a average value of 350.7 mg/l which is above the acceptable limit of 250 mg/l (WHO 2017).

The alkalinity of water samples measured to give an idea about the salts present in it. The water flows through the soils and sedimentary rocks from which the hydroxide, carbonate and bicarbonates dissolve in it and causes alkalinity (Kumar *et al*., 2021). The maximum permissible level is 600 mg/l (WHO). The highest amount of alkalinity was found in an Industrial sample, 96 mg/l. The lowest amount of alkalinity was found in a Non Industrial sample, 44 mg/l.

Sulphate concentration ranges from 18 to 118 mg/l, with an average of 49 mg/l. The samples from the location are within the permissible limit 250 mg/l (WHO, 2007). The non industrial sampling site shows higher concentration than the industrial sampling site which is due to natural decomposition of rocks, minerals, agricultural runoff and atmospheric deposition.

Total hardness, Sodium, Potassium

The sum of calcium and magnesium ions in water is known as total hardness. The lowest value of 925 mg/l found in the non industrial location (8) and the 5883mg/l highest value of found in the industrial location (2). The average value of hardness was found to be 2475 mg/l, which is higher than the allowable limit of 300 mg/l. This higher hardness might be attributed to the effluents from the nearby industry (Jeyaraj, 2016).

Among the cations sodium is the dominant ion, and its concentration varies between 13.01 mg/l to 86.26 mg/l. The level of potassium is 15.59 mg/l to 61.79 mg/l, which is remarkably higher than the permissible limit of 12 mg/l (WHO, 2017).

Nitrate Concentration of Ground Water

The level of nitrate in groundwater ranges from 32.90 to 88.13 mg/l, with the average concentration of 56.83 mg/l. The acceptable limit of nitrate in drinking water, as per WHO guidelines, is 50 mg/l (WHO 2017). The Industrial site Arasur (88.13 mg/l) and Ganapathy (56.17 mg/l) shows higher concentration of nitrate. In the non industrial site, Ondipudhur (59.2 mg/l) and Neelambur (56.5 mg/l) shows higher concentration. The nitrate (NO^{3-}) is naturally found in the environment and is an impor-

Table 2. Health risks of Nitrate contamination in Children and Adults via Dermal and Ingestion exposure

Sample Industrial	Chronic daily intake			Dermal Absorbed Dose			HQ(oral)			HQ (dermal)			Hazard Index(HI)		
	M	Fe	C	M	Fe	C	M	Fe	C	M	Fe	C	M	Fe	C
1	3.37	5.93	7.34	0.01	0.00	0.00	2.11	3.71	4.59	0.00	0.00	0.00	2.11	3.71	4.59
2	4.11	7.23	8.94	0.01	0.00	0.01	2.57	4.51	5.59	0.00	0.00	0.00	2.57	4.51	5.56
3	3.62	6.36	7.87	0.01	0.00	0.01	2.26	3.98	4.92	0.00	0.00	0.00	2.27	3.98	4.93
4	2.14	3.77	4.6	0.00	0.00	0.00	1.34	2.36	2.92	0.00	0.00	0.00	1.34	2.36	2.92
5	1.90	3.34	4.14	0.00	0.00	0.00	1.18	2.09	2.58	0.00	0.00	0.00	1.19	2.09	2.59
Sample Non-Industrial	Chronic daily intake			Dermal Absorbed Dose			HQ (oral)			HQ(dermal)			Hazard Index (HI)		
	M	Fe	C	M	Fe	C	M	Fe	C	M	Fe	C	M	Fe	C
1	2.14	3.77	4.67	0.00	0.00	0.00	1.34	2.36	2.92	0.00	0.00	0.00	1.34	3.71	4.59
2	2.76	4.85	6.00	0.00	0.00	0.00	1.72	3.03	3.75	0.00	0.00	0.00	1.73	4.51	5.59
3	2.27	3.99	4.94	0.00	0.00	0.00	1.41	2.49	3.08	0.00	0.00	0.00	1.42	3.98	4.93
4	1.53	2.69	3.33	0.00	0.00	0.00	0.95	1.68	2.08	0.00	0.00	0.00	0.96	2.36	2.92
5	2.63	4.64	5.74	0.00	0.00	0.00	1.64	2.90	3.58	0.00	0.00	0.00	1.65	2.09	2.59

tant plant nutrient. It is present at varying concentrations in all plants and is a part of the nitrogen cycle. Nitrate can reach both surface water and groundwater as a consequence of agricultural activity (including excess application of inorganic nitrogenous fertilizers and manures), from wastewater disposal and from oxidation of nitrogenous waste products in human and animal excreta, including septic tanks (Zhang *et al*., 2019). Surface water nitrate concentrations can change rapidly owing to surface runoff of fertilizer, uptake by phytoplankton and denitrification by bacteria, but groundwater concentrations generally show relatively slow changes. Some ground waters may also have nitrate contamination as a consequence of leaching from natural vegetation (WHO 2017) (Tanega *et al*., 2019).

According to results, the percentage of low risk (<45 mg/l) and high risk (45-100 mg/l) were Neelambur, Saravanampatti, Peelamedu, Nava India, Chinniyampalayam, Avarampalayam, Ondipudhur, Athipalayam, Ganapathy, Arasur respectively. The higher concentration of nitrate in the study area poses a risk to human drinking water source. Some of the studies by Narsimha 2018 and Sajilkumar, 2020 have also indicated that the long term exposure of nitrate in drinking water can causes methemoglobinemia in humans.

Human Health risk assessment of nitrate

The higher concentration of nitrate (>45 mg/l) can cause human health risks (WHO, 2017). The USEPA developed system is used to assess the human health risks (Sener *et al*., 2017). The results of Hazard quotient obtained for oral and dermal pathways of nitrate are illustrated in Table 2.

The chronic daily intake (CDI) values of nitrate ranging from for 1.53 and 4.11 mg/kg/day for males, 2.69 and 7.23 mg/kg/ day for females and 3.33 and 8.94 mg/kg/day for children. CDI values were further used in determining the hazard quotient via ingestion.

The dermally absorbed dose (DAD) was calculated as a step of determining the HQ_{Dermal}. A DAD value of nitrate for males ranges from 0.005 and 0.014 mg/kg/day, with an average of 0.007 mg/kg/day. DAD for females ranges from 0.003 and 0.009 mg/kg/day, with an average of 0.005 mg/kg/day. For children, DAD ranged 0.004 and 0.011 mg/kg/day, with an average of 0.006 mg/kg/day.

In this study, HQ through the oral pathway for men ranges from 0.96 and 2.57. The average value is 1.38, which is higher than the recommended value of 1(USEPA 2014); it shows that non-carcinogenic risk is visible for men in the study area.Most of the sample showed the highest value. From a total of 10 samples, 38% exceeded the permissible value. HQ through the oral pathway for female varied between 1.68 and 4.51. The average value is 2.42, which is higher than the recommended value of according to USEPA 2014; thus, a non-carcinogenic risk is visible for female in the study area (Marghade *et al.,* 2020). HQ through the oral pathway for children varied between 2.08 and 5.59. The average value is 3.00, which is higher than the recommended value of 1 according to USEPA 2014 (Jayarajan, 2021).

The Hazard Quotient(HQ) for Nitrate ranging from 0.003 to 0.008 for men, 0.002 to 0.006 for women and 0.002 and 0.007 for children. Similar studies conducted in various places of India such as Sulur taluk (Karunanidhi *et al.,* 2019), Texa valley, Tirupur region (Karunadhi *et al*., 2020), multiple areas of Coimbatore and Tirupur district (Sajil Kumar 2020), Punjab (Ahada and Suthar, 2017), and Northwest China (Zhang *et al.,* 2019) indicates that children and women are more exposed to risk because of their lower body weight compared with men. From the results, its showed that children are more suspectible to non-carcinogenic risks due to nitrate toxicity.

Conclusion

The present study investigated the physical and chemical properties, the existence of nitrate concentration in ground water, and the health risks associated with nitrate in selected industrial and non-industrial sires of the Coimbatore District. Nitrate concentration ranges from 32.90 to 88.13 mg/l with an average value of 56.83 mg/l. A total of 37% of both groundwater samples had NO_3- levels that were higher than permissible limit, demonstrating both natural and man-made causes affect the level of nitrate in ground water.

The study area has not shown any risk from nitrate exposure (HQ Dermal 1 in all samples), however there may be a health risk through ingestion. The HQ via ingestion exceeded one in 38% of males, 40% of females, and 44% of children. The order of human health risk is children> females> males. So Nitrate enrichment should be controlled by agricultural and anthropogenic activities with the help of corrective actions like frequent inspection of the water supply, health examinations, and inspection of subterranean pipelines can lessen the harmful effects of nitrate-contaminated drinking water.

Conflict of Interests

The authors declare that they have no conflict of interests.

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