Desalination and Water Treatment
www.deswater.com
doi: 10.5004/dwt.2023.30096

316 (2023) 1–21
December

Spatial distribution of nitrate and fluoride pollution and risk assessment in drinking water resources of Chennai, Kancheepuram, and Tiruvallur districts of Tamil Nadu, India

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Received 31 March 2023; Accepted 15 October 2023

ABSTRACT

Nitrate and fluoride are among the key chemicals that cause large scale health effects through drinking water exposure. In this study, the nitrate and fluoride content from drinking water sources of the Chennai River basin districts of Chennai, Kancheepuram and Tiruvallur in Tamil Nadu, India were analyzed to map their spatial distribution. Out of the 103 groundwater samples analyzed, 84 (81.6%) and 79 (76.7%) samples were found to possess below permissible limits of nitrate (45.0 mg/L) and fluoride (1.0 mg/L), respectively. Chennai city, with high population density was not found to be a nitrate and fluoride endemic region. The maximum nitrate concentrations of 143.9 and 132.0 mg/L were found to occur in water sourced from two relatively isolated hotspots, a borewell in Sottupakkam and an openwell in Sirunallur, respectively of Kancheepuram district. Tiruvallur (maximum 2.7 mg/L at Tiruvallur town) and Kancheepuram (maximum 2.6 mg/L at Chengalpattu town) districts had more than 10 fluoride hotspots each with content higher than the permissible limit, which is most probably due to fluoride leaching from mineral-rich rocks in these regions. The fluoride concentration levels appeared to correlate better against water quality parameters (total dissolved solids, electrical conductivity, alkalinity, total hardness, chloride and sulphate) studied when compared with the nitrate concentration levels. The fluoride concentrations also appeared to have a relatively higher correlation with alkalinity and hardness which can be attributed to concomitant release of bicarbonate, calcium and magnesium ions during the fluoride leaching process from mineral-rich rocks in these regions. Furthermore, the non-carcinogenic health risk associated with nitrate and fluoride in water ingestion in infants, children and adults was evaluated with the United States Environmental Protection Agency (U.S. EPA) method. The percentage exceedance of total hazard index limit for infants, children and adults in Chennai district was 95.8%, 58.3% and 8.3%, respectively, while it was 100.0%, 100.0% and 61.0%, respectively in Kancheepuram district and 97.4%, 97.4% and 57.9%, respectively in Tiruvallur district. These results predict that infants and children from these three districts appear to be highly susceptible to non-carcinogenic nitrate and fluoride induced health hazards, while adults are predicted to be more or less immune to the associated health risks.

Keywords: Chennai; Kancheepuram; Tiruvallur; Fluoride; Nitrate; Non-carcinogenic health risk; Hazard quotient; Total hazard index

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1. Introduction

According to the World Water Assessment Programme (WWAP), groundwater supplies almost half of all the drinking water in the world [1]. Groundwater is one of the principal components in hydrological cycle and is interconnected with other biological and chemical cycles. Hence, groundwater pollution is known to induce imbalances in ecosystem leading to severe socio-economic effects. India is also reported to be the topmost groundwater abstracting county in the world [2]. Tamil Nadu, in particular, is a state with inadequate water resources and seasonal rainfall. Hence, it is one of the Indian states over-exploiting its groundwater resources. Tamil Nadu is mostly in the shield area with 73.0% of its area covered under hard crystalline formations, while the rest comprises of unconsolidated sedimentary formations. As far as groundwater reserves are concerned, insufficiency is the major crisis in hard rock environment while salinity is the issue in sedimentary areas [3]. Chennai, the capital city of Tamil Nadu, is the fourth-most populous urban agglomeration in India and is situated adjacent to the Bay of Bengal. Chennai metropolitan city abuts the neighboring districts of Kancheepuram and Tiruvallur. These districts constitute the Chennai River basin region in Tamil Nadu, India. Apart from industries, agriculture and allied activities form the major occupation in these districts.

Nitrogen and fluorine are essential elements for all living organisms, but become toxic if ingested in excess. When nitrate is present in excess, it may be microbially transformed to nitrite in the digestive system. The nitrite can oxidize the ferrous to ferric ion in haemoglobin and form methaemoglobin. The methaemoglobin with its poor ability to bind with oxygen is hindered in its ability to carry oxygen from the lung to the various tissues leading to methemoglobinemia or blue baby syndrome [4]. Excess fluoride ion ingested through drinking water is of concern because of its possible accumulation and damage to the human tissues. Many epidemiological studies have established that high fluoride intake affects the bones and teeth [5]. According to Indian Standard Specifications, the permissible limits for nitrate and fluoride in drinking water are 45.0 and 1.5 mg/L, respectively [6].

Nitrate ion is the stable form of oxygenated nitrogen involved in the nitrogen cycle. It can easily leach into groundwater due to its relatively poor affinity towards the soil. The sources of nitrate are fertilizers from agricultural runoff, animal manure, untreated sanitary sewage, decomposition of food/diary/meat/other organic products, leakage from septic systems, industrial waste and detergents/soaps from urban runoff etc. [7]. Fluorine exists in nature in the form of fluorides in a number of minerals such as fluorspar, cryolite and fluorapatite. These fluoride-rich minerals can leach and accumulate in groundwater sources, especially when present in high concentrations [8]. Hence, the information about concentration of pollutants and associated human health hazard indices, especially for pernicious nitrate and fluoride in drinking water sources is essential for urban planning as well as designing and installing industrial and household point-of-use water treatment systems. The presence of common anions in water such as chloride, sulphate and bicarbonate may also affect the efficiency of conventional water treatment technologies such as ion-exchange to remove nitrate and fluoride. In addition to this, other water quality parameters such as total dissolved solids (TDS), total hardness etc. have to be assessed to examine the suitability of groundwater for drinking and irrigation purposes [9].

The nitrate and fluoride released into the environment can affect the water quality as well as the vegetation and food chain. They can be either directly ingested or be absorbed through the skin adversely influencing human health [10,11]. The classical method utilized to assess the risk involved involves comparing the pollutant levels with the guideline limits of regulatory agencies. However, this is a rather simplistic method and does not take into consideration, extent of exposure, age of subjects etc. Hence, a more scientific evaluation is the probability-based risk-assessment methodology recommended by United States Environmental Protection Agency (U.S. EPA) [12,13]. The U.S. EPA method has been widely adopted by many researchers to estimate the adverse health effects associated with prolonged human exposure to contaminants in soil, water, and air [14]. U.S. EPA has classified nitrate and fluoride as non-carcinogenic contaminants, and the health risk assessment methodology has been used effectively by many researchers to assess health risk involved in exposure to nitrate and fluoride in drinking water.

In the first part of this work, the spatial mapping of nitrate and fluoride pollutants concentration levels in the groundwater resources of Chennai River basin districts, Chennai, Kancheepuram and Tiruvallur of Tamil Nadu, India consisting of urban and rural areas has been undertaken. In this investigation, drinking water from 103 sources in these districts were collected, their physico-chemical characteristics analyzed and the parameters mapped to provide the spatial information. The data from the survey was compared with the Indian Standards to map the spatial distribution, especially hotspots with high pollutant density. This spatial information is also essential for urban and rural planning, design water treatment plants, assess health risks of resident population and provide information about the probable cause for any diseases or ill effects in the future [15–17]. In the past, several studies have reported the non-carcinogenic health risk associated with nitrate and fluoride in India, with many of them reporting elevated levels suggesting high-risk for infants and children [18–22]. However, there are no reports for densely populated Chennai city and Chennai river basin region. In the second part of this work, health risk assessment for the nitrate and fluoride exposure in these districts was conducted by calculating the chronic daily intake (CDI), the hazard quotient (HQ) and total hazard index (THI) for three age groups of subjects, infants (<2 y), children (2–16 y) and adults (≥16 y). Overall, the spatial distribution and health risk assessment information about nitrate and fluoride contamination in the Chennai River basin districts is expected to help plan any pre-emptive or remediation measures in the future.

2. Study area

2.1. Geography and geology

The Chennai River basin is situated in the north-western region of Tamil Nadu state in India. It is located

between latitudes 12°15′00″N to 13°30′00″N and longitudes 79°15′00″E to 80°30′00″E. The rivers flow from west to east to meet the Bay of Bengal in this region. The major rivers in the region are Araniar, Kosasthalaiyar, Cooum and Adyar. Apart from this many minor streams fed mainly by rains exist in the southern part of the basin [23]. The Chennai River basin has three contiguous districts, *viz.* Chennai, Kancheepuram and Tiruvallur abutting the Bay of Bengal. These districts were chosen as the study area (Fig. 1). Fig. 1A and B are political maps of India and the state of Tamil Nadu, respectively, while Fig. 1C is the Geographic Information System based map of the study area. Chennai, Kancheepuram and Tiruvallur districts cover an area of around 175, 4,483 and 3,394 km^2, respectively. The population density in these districts are reported to be 26,553, 892 and 1,098 per km^2, respectively [24]. The districts are part of the eastern coastal region consisting of mainly plains with a few hilly areas and possessing elevation ranging from 0 to around 100 m. The soil in these districts are classified mainly as clay, shale and sandstone. The hydrogeology in these districts are mainly sand, sandstone, weathered and fractured granites, gneisses and charnockite.

The Palar is a major perennial river flowing through the Kancheepuram district. It originates in the Western Ghats and drains into the sea. Apart from this, there are smaller rivers such as Cheyyar, Kiliyar and Vegavathi which end up as small tributaries of Palar River. Seasonal rivers, Araniar, Kosasthalaiyar and Thondiar are fed by rains and drain into river basins which are used for drinking and irrigation purpose. In Tiruvallur district, the Araniar, Kosasthalaiyar, Adayar and Cooum are major rivers which eventually make their way to the ocean. The district has two important water bodies, Poondi reservoir and Puzhal (Red Hills) tanks.

The 16 taluks (administrative areas in Indian states) of Chennai district are: Alandur, Aminjikarai, Ambattur, Ayanavaram, Egmore, Guindy, Madhavaram, Maduravoyal,

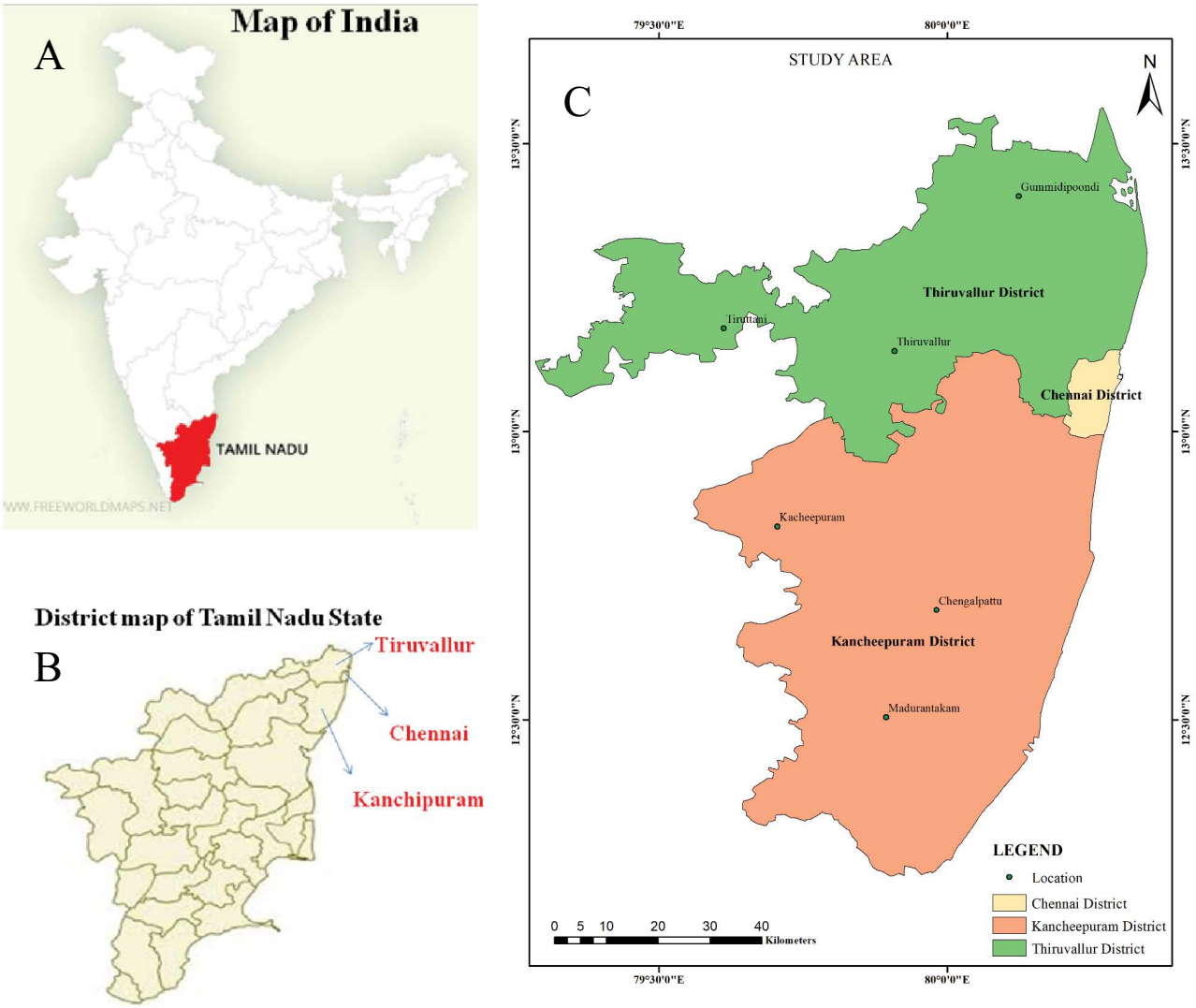

Fig. 1. Location of the study area in the map of Tamil Nadu state, India. Panel A: political map of India, Panel B: political map of the state of Tamil Nadu in India, Panel C: Geographic Information System map of the study area comprising of three districts in Tamil Nadu.

Mylapore, Mambalam, Purasaiwakkam, Perambur, Sholinganallur, Tiruvottiyur, Tondiarpet and Velachery. A total of 24 samples were collected from these Chennai taluks and the sampling locations are as marked in Figs. 2A and 3A.

Kancheepuram district is situated on the northern east coast of Tamil Nadu and is adjacent to Bay of Bengal, Chennai city and Tiruvallur district. A total of 41 water samples were collected from the 11 taluks of Kancheepuram district *viz.* Chengalpattu, Cheyyur, Kancheepuram, Maduranthakam, Pallavaram, Sriperumbudur, Tambaram, Thiruporur, Uthiramerur, Tirukalukundram and Walajabad. The exact locations used for the sampling are marked in Figs. 2B and 3B.

Tiruvallur district sharing borders with Chennai and Kancheepuram districts consists of 9 taluks. A total of 38 drinking water samples were collected from 7 taluks of Tiruvallur district *viz.* Avadi, Gummidipoondi, Ponneri, Poonamallee, Tiruvallur, Thiruttani and Uthukkottai. Figs. 2C and 3C mark the locations used for sampling the groundwater from Tiruvallur district.

3. Materials and methods

3.1. Sampling and analytical methods

A total of 103 water samples were collected during September 2018 by following standard methods (APHA) [25]. Sampling was carried out from the household borewell (83 samples) and openwell (15 samples) drinking water sources. In the study area, especially Chennai district, treated lake water routed through pipes is the major domestic drinking water source. Drinking water was sampled from (i) borewells, (ii) openwells, (iii) treated lake water (3 samples) and (iv) untreated lake water (1 sample) supplied by the authorities and (v) pond water (1 sample). The original source of the treated lake water were Chembarambakkam lake (spread over 15.38 km^2 in the outskirts and supplying water to Chennai), Maduranthakam lake (spread over 9.71 km^2 in Kancheepuram district) and Ambattur – Athipattu lake (spread over 1.78 km^2 in Chennai). The untreated water samples were collected directly from the two water bodies, Puzhal (Red Hills) lake (spread over 18.21 km^2 in Chennai) and Padianallur pond (a small pond near Chennai used for local water supply).

The water samples were collected in fresh 250 mL polyethylene bottles. In borewells, the water was initially pumped out for around 10 min to ensure that stagnant water did not make it to the sample. The water samples were labeled and stored at 4°C in the refrigerator before analysis. Nitrate in water samples were analyzed at 202 nm with UV-Visible spectrophotometry (Hitachi UV Spectrophotometer UH5300 Spectrophotometer, Japan, Model NO-3J1-0015). Fluoride ion concentrations in water samples were measured using fluoride ion selective electrode-based ion meter (Model No-LMION-40, Mark Labman, Serial No-1.7710, India). Water quality parameters were analyzed using water analyzer purchased from Siscon (India) Pvt. Ltd. The turbidity and sulphate concentration were analysed by Nephelo-Turbidity Meter. Electrical conductivity (EC), TDS, salinity, dissolved oxygen and pH were analyzed using standard electrodes. Alkalinity, total hardness, calcium, magnesium and chloride were measured by standard titrimetry. All chemicals purchased were AR grade from Siscon (India) Pvt. Ltd. All analysis were conducted in triplicates.

3.2. Human health risk assessment methodology

U.S. EPA defines human health risk as the probability of harmful effects to human health due to exposure to environmental pollutants. The human health risk assessment may be defined as the methodology used to evaluate the probability of occurrence of any harmful health impacts over a determined time period. The assessment methodology includes different phases such as: (i) measurements characterizing the nature and extent of the environmental pollutant in a given location, (ii) determining which pollutants have the potential to cause harm to human health, (iii) assessing the dose-dependent effects of the pollutant on human subjects, (iv) assessing the frequency and magnitude of exposure that has occurred to humans in the given location and (v) characterization of the risks for human health involved in the case-studied. In this case, the harmful effects of exposure to nitrate and fluoride, their permissible limits and non-carcinogenic health risks are well documented. Hence, in this study, at the end of the first survey part, the concentration of nitrate and fluoride at various locations were used to calculate the CDI, HQ and THI [26–28]. This information was used to characterization of the risk for infants (<2 y), children (2–16 y) and adults (≥16 y) based on the following calculations.

$$\mathrm{CDI} = \frac{C \times \mathrm{IR} \times F \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \tag{1}$$

where CDI of nitrate and fluoride *via* oral ingestion is in mg/(kg·d), while C, IR, F, ED, BW and AT represent the concentrations of nitrate and fluoride in groundwater (mg/L), ingestion rate (L/d) [29], exposure frequency (d/y) [30], exposure duration (y) [31], average body weight (kg) [32] and average exposure time (d) [30], respectively. The parameters utilized to characterize the CDI for infants, children and adults are presented in Table 1.

$$\mathrm{HQ} = \frac{\mathrm{CDI}}{\mathrm{RfD}} \tag{2}$$

where RfD is the reference dose for nitrate (1.6 mg/(kg·d)) and fluoride (0.04 mg/(kg·d)) [13].

$$\mathrm{THI} = \mathrm{HQ}_{\mathrm{nitrate}} + \mathrm{HQ}_{\mathrm{fluoride}} \tag{3}$$

with THI ≤ 1 and >1 suggesting no health risk and high hazard levels to human subjects, respectively [33,34].

4. Results and discussion

4.1. Physico-chemical parameters and quality of groundwater

As the first part of the study, the physico-chemical analysis of the water samples collected from the study area, Chennai, Kancheepuram and Tiruvallur districts was

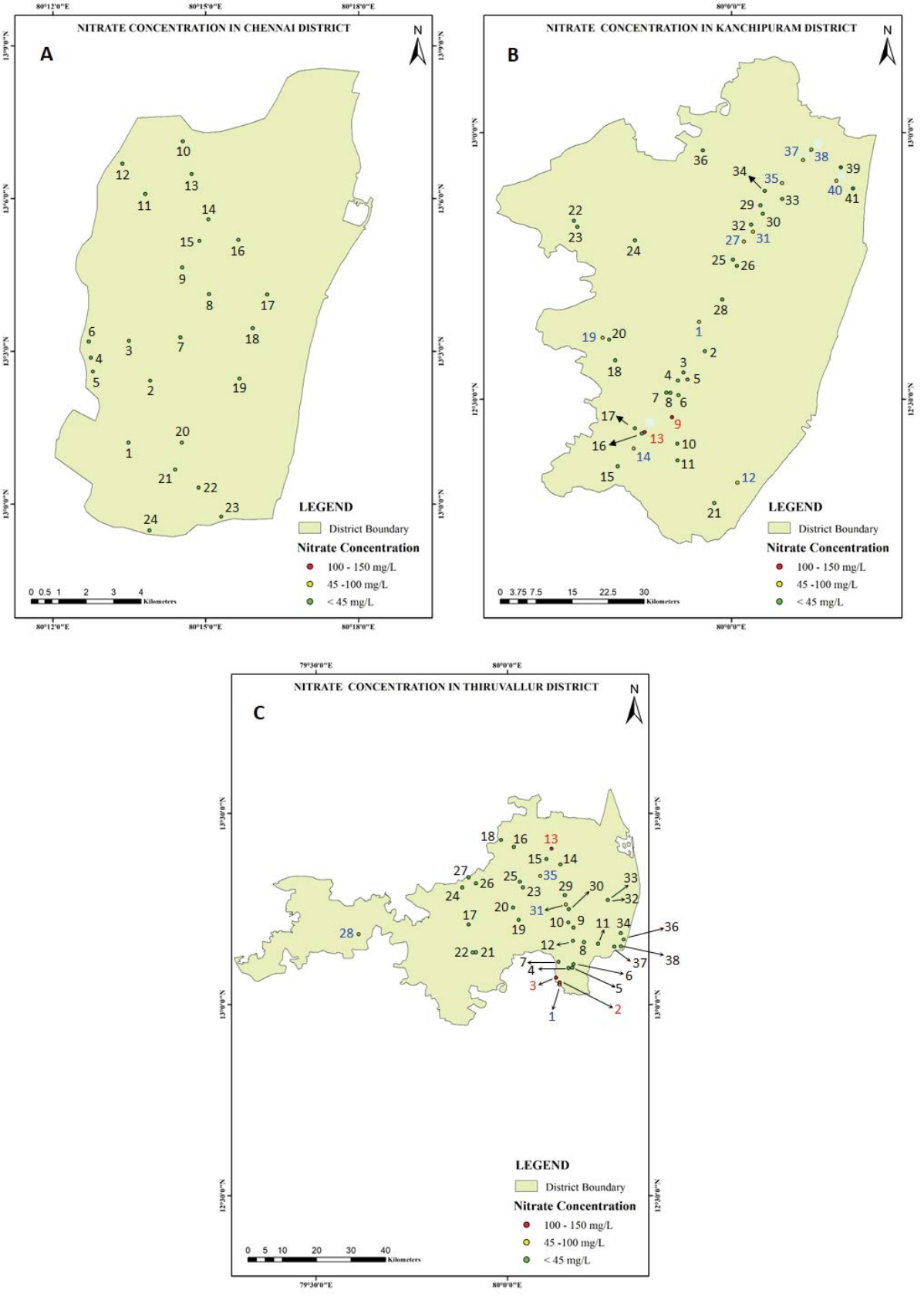

Fig. 2. Nitrate distribution in (A) Chennai, (B) Kancheepuram and (C) Tiruvallur district of Tamil Nadu state, India.

conducted. This data was used to establish the spatial distribution of nitrate and fluoride in the study area. The permissible value/range of parameters, specified as per Indian Standards, were used to establish the suitability of the water for drinking purpose [6]. A summary of these physico-chemical parameters and their values/ranges are shown in Table 2. The collected drinking water samples were found to be colorless and odorless. Only 11.7% of the samples were observed to have turbidity above the permissible limit. Around 79 samples exhibited zero turbidity, with 13 samples having turbidity between 1–3 NTU. In all the districts, 86 samples exhibited the pH values within the permissible limit. The drinking water samples from 9 spots were acidic in nature (range 4.1–6.0) and 9 samples were above the permissible alkaline range (8.6–9.5). EC, an indication of ionic strength, varied between 0.3–9.6 mS/cm. Salinity was found to range between 190.0–5,870.0 mg/L with a mean of 1,005.0 mg/L. The average dissolved oxygen content in the samples was 4.9 mg/L. The average TDS of samples sourced from the area studied was 896.0 mg/L, much higher than the standard drinking water limit of 500.0 mg/L. Around 76.7% of the samples were found to exceed this standard TDS limit. The average total hardness was observed to be 323.5 mg/L. The water hardness is mainly due to the presence of cations Ca^{2+} and Mg^{2+}. Around 26.2% and 51.5% of the samples were found to have calcium and magnesium levels above their permissible limits, respectively. In 74.0% of the samples, alkalinity (usually contributed by bicarbonate) exceeded the drinking water permissible limit. Chloride concentration was in the range of 20.0–2,172.0 mg/L (mean value of 325.5 mg/L). Average sulphate concentration in these districts was found to be under the permissible limit. The maximum values of TDS, EC, salinity, total hardness, calcium, magnesium, chloride and sulphate were observed for the borewell sample collected from Ambattur industrial estate of Tiruvallur district. This was most probably due to the industrial activity in this region. Overall, the results of the physico-chemical analysis show that groundwater samples from the study area can be used for drinking after treatment, especially for TDS, hardness and alkalinity.

Table 1
Parameters utilized for calculating the chronic daily intake used in the non-carcinogenic human health risk assessment of nitrate and fluoride in water pollution

Parameters	Infants	Children	Adults
IR (ingestion rate *via* water, L/d)	0.65	1.50	3.00
F (exposure frequency, d/y)	365.00	365.00	365.00
ED (exposure duration, y)	0.50	6.00	30.00
BW (average body weight, kg)	6.94	25.90	64.70
AT (averaging time, d)	182.50	2,190.00	10,950.00

4.2. Nitrate pollution in groundwater

4.2.1. Chennai district

The general physico-chemical analysis presents an overall picture of the groundwater quality in the districts. However, this study focused on the hypothesis that ingestion of above permissible limit nitrate and fluoride groundwater may be a potential health hazards to the resident in the in the study area. To analyze this, as shown in Fig. 2A–C, the spatial distribution of nitrate concentrations in drinking water were mapped for Chennai, Kancheepuram and Tiruvallur districts, respectively. The color-coded map has the concentrations classified into three discrete ranges *viz.* <45.0 mg/L, 45.0–100.0 mg/L and 100.0–150.0 mg/L. The sampling locations latitude, longitude and the nitrate concentrations are documented in Tables 3–5. Overall, around 84 (81.5%) water samples were found to contain nitrate below the tolerance limit (45.0 mg/L). In Chennai districts, none of the 24 locations had nitrate levels above the permissible limit. Hence, Chennai city and district, in spite of hosting a high population density, does not appear to be a nitrate endemic region.

Table 2
Statistical summary of physico-chemical parameters and its comparison with Indian Standards for drinking water

Parameter	Range	Mean	Indian Standards	Percentage of sample above the limit
Turbidity (NTU)	0.0–131.0	5.2	5	11.7
pH	4.1–9.5	7.7	6.5–8.5	17.5
Total dissolved solids (mg/L)	137–5,310	896	500	76.7
Electrical conductivity (mS/cm)	0.3–9.6	1.6	–	–
Salinity (mg/L)	190–5,870	1,005	–	–
Dissolved oxygen (mg/L)	3.1–7.1	4.9	–	–
Alkalinity (mg/L)	100–750	315	200	74.0
Total hardness (mg/L)	90.0–2,600.0	323.5	200	67.0
Ca^{2+} (mg/L)	20.0–529.1	68.5	75	26.2
Mg^{2+} (mg/L)	7.3–311.9	38.8	30	51.5
Cl^{-} (mg/L)	20.0–2,172.0	325.5	250	45.6
SO_4^{2-} (mg/L)	12.0–1,442.0	193.7	200	24.3
NO_3^{-} (mg/L)	0.0–143.9	25.8	45	18.4
F^{-} (mg/L)	0.3–2.7	0.9	1	23.3

Table 3
Nitrate and fluoride concentrations in different locations of Chennai district, Tamil Nadu, India

No.	Location	Latitude/Longitude	Nitrate (mg/L)	Fluoride (mg/L)	Remarks
1	Saidapet	13°01′12.7″N 80°13′28.9″E	12.1	0.5	Densely populated urban residential area with few industries
2	T. Nagar	13°02′25.5″N 80°13′54.5″E	1.6	0.6	Densely populated commercial and residential area
3	Kodambakkam	13°03′12.7″N 80°13′29.6″E	7.1	0.5	Densely populated urban residential area
4	Vadapalani	13°02′52.7″N 80°12′44.7″E	9.0	0.6	Densely populated urban residential area
5	Saligramam	13°02′47.5″N 80°11′53.8″E	4.3	0.7	Densely populated urban residential area
6	Virugambakkam	13°02′54.1″N 80°11′49.7″E	2.5	0.7	Densely populated urban residential area
7	Nungambakkam	13°03′16.7″N 80°14′30.2″E	2.4	0.9	Densely populated lake reclamation urban area
8	Egmore	13°04′07.7″N 80°15′03.8″E	1.3	0.5	Densely populated urban residential area
9	Kilpauk	13°04′39.0″N 80°14′32.2″E	1.0	0.8	Densely populated urban residential area
10	Aminjikarai	13°07′07.6″N 80°14′33.0″E	2.7	0.4	Densely populated urban lake reclamation area
11	Ayanavaram	13°06′05.7″N 80°13′48.7″E	1.6	0.6	Densely populated urban residential area
12	Villivakkam*	13°06′45.7″N 80°12′08.2″E	33.9	0.5	Urban area with railway locomotive and allied factories
13	Perambur	13°06′29.1″N 80°14′43.5″E	41.0	0.4	Urban area with railway locomotive and allied factories
14	Otteri	13°05′35.8″N 80°15′03.2″E	1.2	0.6	Densely populated lake reclamation urban area
15	Purasaiwakkam	13°05′10.1″N 80°14′52.6″E	1.1	0.7	Densely populated commercial and residential area
16	Vepery	13°05′11.5″N 80°15′38.5″E	6.8	0.5	Densely populated commercial and residential area
17	Chintadripet	13°04′07.3″N 80°16′12.5″E	3.4	0.6	Densely populated commercial and residential area
18	Triplicane	13°03′27.4″N 80°15′55.4″E	2.2	0.9	Densely populated urban residential area beside the seashore
19	Mylapore	13°02′28.0″N 80°15′39.9″E	9.2	0.5	Densely populated urban residential area near the seashore
20	Kotturpuram	13°01′12.7″N 80°14′31.8″E	0.3	0.5	Densely populated urban residential area
21	Guindy	13°00′40.8″N 80°14′24.0″E	15.7	0.6	Urban area with student population and small-scale industries
22	Adayar	13°00′19.5″N 80°14′51.5″E	0.2	0.3	Densely populated urban residential area near the seashore
23	Thiruvanmiyur	12°59′22.0″N 80°15′21.1″E	5.0	0.9	Densely populated urban residential area beside the seashore
24	Velachery*	12°58′57.9″N 80°13′05.2″E	8.0	0.6	Densely populated lake reclamation urban area

*Openwell source

Table 4
Nitrate and fluoride concentrations in different locations of Kancheepuram district, Tamil Nadu, India

No.	Location	Latitude/Longitude	Nitrate (mg/L)	Fluoride (mg/L)	Remarks
1	Mamandur	12°38′42.6″N 79°56′23.1″E	92.5	1.3	Highway town with busy food-outlet and agricultural activity
2	Padhalam	12°35′24.9″N 79°57′01.6″E	7.6	1.3	Village with rocky terrain near Palar River and agricultural activity
3	Melavalaipettai*	12°32′26.3″N 79°54′16.8″E	2.7	0.9	Openwell water from highway village near lake with agricultural activity
4	Karunguzhi*	12°32′06.8″N 79°54′06.2″E	1.0	1.2	Highway town on rocky terrain close to lakes and agricultural fields
5	Karunguzhi	12°32′06.8″N 79°54′06.2″E	2.4	1.0	Highway town on rocky terrain close to lakes and agricultural fields
6	Maduranthakam**	12°30′43.1″N 79°53′08.9″E	2.2	1.2	Treated lake water from densely populated town near Maduranthakam lake
7	Maduranthakam	12°30′44.4″N 79°53′06.6″E	36.8	0.8	Densely populated town near Maduranthakam lake
8	Maduranthakam	12°30′44.4″N 79°53′06.6″E	38.2	0.6	Densely populated town near Maduranthakam lake
9	Sirunallur*	12°27′58.9″N 79°53′19.0″E	132.0	1.2	Openwell water from village with paint factory and agricultural fields
10	Chittamur	12°24′58.8″N 79°53′56.3″E	39.1	0.9	Village with agricultural fields and a manufacturing industry
11	Pazhuvur	12°23′06.1″N 79°53′56.7″E	13.4	0.8	Village 3 km from Chittamur with agricultural activity

Table 4 (Continued)

Table 4

No.	Location	Latitude/Longitude	Nitrate (mg/L)	Fluoride (mg/L)	Remarks
12	Cheyyur*	12°20′36.5″N 80°00′41.0″E	49.8	0.7	Openwell water from village close to fresh water Udaiyur lake
13	Sottupakkam	12°26′18.3″N 79°50′14.5″E	143.9	0.9	Busy highway rural town with colleges, agricultural fields and food-outlets
14	Acharapakkam*	12°24′28.0″N 79°49′01.3″E	60.0	0.9	Openwell water from densely populated highway town with food-outlets
15	Tozhupedu	12°39′46.2″N 79°27′14.0″E	15.3	0.9	Highway town with food-outlets and agricultural fields
16	Melmaruvathur*	12°26′08.0″N 79°49′55.5″E	39.5	1.2	Openwell water of highway town with nearby lakes and rocky terrain
17	Melmaruvathur	12°26′12.9″N 79°49′19.0″E	4.3	0.7	Highway town with high visitor density, nearby lakes and rocky terrain
18	Kamalapoondi*	12°34′24.2″N 79°46′55.9″E	7.1	1.2	Rural village near Uthiramerur on very rocky and hilly terrain
19	Uthiramerur	12°36′55.1″N 79°45′31.5″E	65.0	0.7	Temple town with tourists, high population density and nearby lakes
20	Uthiramerur*	12°36′53.7″N 79°45′34.1″E	44.4	1.0	Openwell water of highway town near lakes and rocky terrain
21	Vedal	12°18′18.3″N 79°58′04.4″E	7.9	1.2	Rural village near Bay of Bengal shore, but very rocky and hilly terrain
22	Ennaikaran	12°50′06.6″N 79°42′18.6″E	14.0	1.7	Suburb of Kancheepuram town with dense population and rocky terrain
23	Kancheepuram	12°49′24.1″N 79°42′41.0″E	2.1	0.9	Densely populated temple town with many water tanks
24	Walajabad	12°47′53.2″N 79°49′09.4″E	30.0	0.9	Densely populated town with a railway station on Palar Riverbank
25	SP Koil	12°45′42.9″N 80°00′12.0″E	8.5	0.9	Highway town close to MM Nagar and associated industries
26	SP Koil*	12°45′13.3″N 80°00′03.0″E	6.1	0.7	Openwell water from highway town near MM Nagar and industries
27	MM Nagar	12°47′45.0″N 80°01′24.9″E	54.1	0.6	Highway town abutting Tamil Nadu Government's Industrial zone
28	Chengalpattu	12°41′15.0″N 79°58′59.3″E	34.2	2.6	Densely populated urban area next to Palar River and rocky terrain
29	Urapakkam*	12°51′46.6″N 80°04′06.5″E	19.1	1.4	Openwell water from highway town with residential population
30	Guduvancheri*	12°50′52.1″N 80°03′32.9″E	7.9	0.5	Openwell water from highway town with residential population
31	Potheri	12°48′51.7″N 80°02′25.7″E	66.0	0.5	Highway college campus town with very high student population density
32	SRM Nagar	12°49′21.6″N 80°02′38.1″E	28.5	0.5	Highway college campus town with very high student population density
33	Vandalur	12°52′40.8″N 80°04′57.7″E	2.4	0.9	Highway college campus town with very high student population density
34	Vandalur	12°53′33.6″N 80°05′06.3″E	38.7	1.2	Highway town close to railway station on rocky terrain
35	Perungalathur	12°54′20.5″N 80°05′42.8″E	49.2	0.6	Highway town with high population density
36	Sriperumbudur	12°58′01.7″N 79°56′47.5″E	22.8	0.6	Highway town with temple tanks, residential and industrial areas
37	Chromepet	12°56′55.6″N 80°08′04.5″E	65.5	0.9	Chennai city suburb with tanning industry cluster
38	Pallavaram	12°58′06.1″N 80°08′59.2″E	86.6	0.7	Chennai city suburb with tanning industry cluster
39	Pallikaranai	12°56′07.1″N 80°12′19.5″E	19.1	0.4	Chennai city suburb with marshlands and residential areas
40	Medavakkam	12°54′36.7″N 80°11′48.8″E	65.0	0.3	Chennai city suburb with high population density and small industries
41	Sholinganallur	12°53′44.8″N 80°13′40.4″E	25.0	0.3	Chennai city suburb near the sea with residential and commercial areas

*Openwell water;
**Treated lake water.

Table 5
Nitrate and fluoride concentrations in different locations of Tiruvallur district, Tamil Nadu, India

No.	Location	Latitude/Longitude	Nitrate (mg/L)	Fluoride (mg/L)	Remarks
1	Poonamallee	13°02′30.5″N 80°07′33.2″E	57.4	2.3	Groundwater from densely populated commercial and residential area
2	Poonamallee**	13°02′30.5″N 80°07′33.2″E	106.5	1.2	Treated lake water from densely populated commercial and residential area
3	Thiruverkadu	13°04′13.5″N 80°07′22.7″E	120.5	1.1	Temple town commercial area next to the bus station
4	Ambattur Ind. E.	13°05′45.3″N 80°09′34.8″E	7.8	1.6	Government of Tamil Nadu designated industrial area
5	Ambattur Ind. E.**	13°05′45.3″N 80°09′34.8″E	2.7	1.2	Treated lake water from Government of Government's Industrial area
6	Ambattur Ind. E.	13°06′21.2″N 80°10′21.3″E	2.5	2.1	Government of Tamil Nadu designated industrial area
7	Ambattur	13°06′43.5″N 80°08′00.2″E	21.8	0.7	Densely populated residential area near Ambattur lake
8	Puzhal	13°09′49.2″N 80°12′00.1″E	25.5	0.5	Sparsely populated area beside Puzhal (Red Hills) lake shore
9	Padianallur	13°12′25.9″N 80°10′31.4″E	0.3	2.0	Densely populated North Chennai suburban area near Puzhal lake
10	Padianallur****	13°12′25.9″N 80°10′31.4″E	20.1	1.0	Pond water from pond in Chennai city suburb
11	Madhavaram	13°09′33.7″N 80°14′12.8″E	19.3	0.6	Chennai suburb adjacent to residential college and a semi-industrial area
12	Red Hills Lake***	13°10′00.8″N 80°10′17.2″E	1.0	0.8	Lake water from banks of Puzhal (Red Hills) lake with no habitation
13	Gummidipoondi	13°24′28.9″N 80°06′54.5″E	105.9	0.8	Town in highly industrialized belt
14	Kavaraipettai	13°21′59.6″N 80°08′19.3″E	8.4	0.7	Highway small town close to Panapakkam lake and Kilikodi lake areas
15	Thandalachery	13°22′50.8″N 80°06′06.3″E	23.7	1.1	Rural area near Kavaraipettai north of Chennai with rocky terrain
16	Thaanipoondi	13°24′45.3″N 80°01′00.6″E	0.8	1.6	Rural area near Tamil Nadu state border with hilly terrain
17	Neyveli village	13°12′34.8″N 79°53′53.2″E	8.9	0.9	Groundwater from banks of Poondi reservoir with poor habitation
18	Sathyavedu	13°26′10.4″N 79°57′27.2″E	36.0	0.4	Border town with residential population density and proximity to industries
19	Thamaraipakkam	13°13′15.3″N 80°01′43.8″E	3.9	0.8	Highway junction residential town close to Kususthalai river
20	Vengal	13°15′13.5″N 80°00′54.0″E	0.0	0.7	Highway village with sparse population
21	Kakkalur byepass	13°08′13.3″N 79°55′03.7″E	19.2	0.6	Densely populated near Tiruvallur town and near Vaagai Lake and highway
22	Tiruvallur	13°08′12.8″N 79°54′33.0″E	1.2	2.7	Densely populated urban town North of Chennai near lakes
23	Vadamadurai	13°18′24.6″N 80°02′25.8″E	14.3	1.2	Highway town North of Chennai on the banks of Arani River
24	Uthukkottai	13°20′02.4″N 79°53′34.5″E	29.1	0.7	Sparsely populated highway junction area on the Arani Riverbank
25	Periyapalayam	13°18′44.1″N 80°02′52.1″E	28.4	0.7	Sparsely populated village on the Arani Riverbank
26	Tharatchi village	13°19′01.1″N 79°55′03.9″E	23.3	1.2	Rural village on Arani Riverbank and upstream of Vadamadurai town
27	Uthukkottai	13°20′08.7″N 79°53′51.1″E	18.5	0.7	Small town on Arani Riverbank and close to a lake
28	Thiruttani	13°11′04.2″N 79°36′38.8″E	50.1	0.9	Temple town with commercial and residential areas and high visitor traffic
29	Panchetti	13°17′10.5″N 80°09′00.1″E	16.1	1.0	Highway industrial town on riverbanks and downstream of Vadamadurai
30	Karanodai	13°14′59.2″N 80°09′37.6″E	5.9	0.8	Small highway town on Kosasthalaiyar riverbanks and north of Puzhal lake

Table 5 (Continued)

Table 5

No.	Location	Latitude/Longitude	Nitrate (mg/L)	Fluoride (mg/L)	Remarks
31	Janappanchatram	13°15′44.4″N 80°09′09.8″E	59.9	0.6	Highway town with food-outlets and residences next to Kosasthalaiyar river
32	Minjur	13°16′25.5″N 80°15′44.0″E	16.1	0.8	Densely populated suburban town north of Chennai
33	Minjur*	13°16′25.5″N 80°15′44.0″E	16.6	0.8	Openwell water from densely populated suburban town north of Chennai
34	Ennore	13°11′11.6″N 80°18′22.2″E	0.4	0.3	Industrial town near seashore and Kosasthalaiyar riverbank
35	Arani	13°20′11.4″N 80°05′08.7″E	52.9	0.8	High population density area in Arani town
36	Tiruvottiyur	13°09′51.9″N 80°18′01.3″E	42.1	0.5	Densely populated city area close to the seashore
37	Tiruvottiyur	13°09′07.9″N 80°17′46.9″E	21.0	0.6	High population density area near industries
38	Tiruvottiyur*	13°09′07.9″N 80°17′46.9″E	41.8	0.8	Open well water from high population density area near industries

*Openwell water;
**Treated lake water;
***Lake water;
****Pond water.

4.2.2. Kancheepuram district

In Kancheepuram district, 29 of the 41 spots sampled had lower than permissible limit of nitrate. Seven highway towns, Mamandur (92.5 mg/L nitrate, highway town with high tourist traffic, motels and food outlets), Pallavaram (86.6 mg/L nitrate, Chennai city suburb with tanning industry cluster), Chromepet (65.5 mg/L, Chennai city suburb with tanning industry cluster), Acharapakkam (60.0 mg/L nitrate, highway town with motels and food outlets), MM Nagar (54.1 mg/L nitrate, highway town with food outlets), Potheri (66.0 mg/L nitrate, highway college-town with hostels and food outlets) and Perungalathur (49.2 mg/L nitrate, high population density highway town) adjoining the Grand Southern Trunk (GST) road with high population density were found to have considerably high nitrate levels. Apart from this, three other locations were found to have higher than permissible nitrate concentrations, temple town Uthiramerur (65.0 mg/L nitrate, high tourists and population density with nearby lakes), Medavakkam (65.0 mg/L nitrate, newly developed Chennai city suburb with high population density and small industries) and Cheyyur (49.8 mg/L nitrate, rural village proximity to fresh water Udaiyur lake). The maximum nitrate level of 143.9 mg/L was observed in a borewell sample collected from Sottupakkam of Kancheepuram district. Openwell water sample from Sirunallur in the same district was also found to contain nitrate levels as high as 132.0 mg/L. Sottupakkam is a rural town with agricultural activity, but located at the junction of two major highways with many restaurants, motels and a residential high student-density college. The urban sewage and fertilizer from agricultural field run-offs are the probable reasons for high nitrate levels in this hotspot. Sirunallur, a rural area abounded by agricultural fields is located in the proximity of a paint factory and the discharge from this unit is probable reason for high nitrate levels. Chromepet and Pallavaram locations are in the vicinity of leather processing industries and is the likely reason for high nitrate concentrations in these Chennai city suburbs. It is also important to point out that the treated Maduranthakam lake water distributed in Kancheepuram district, as expected was found to contain low nitrate concentration of 2.2 mg/L, showing the efficacy of the treatment process in the town.

4.2.3. Tiruvallur district

In Tiruvallur district, a majority (31 out of 38 spots) of the drinking water samples were found to be safe with respect to nitrate levels. Of these, the treated lake water at Poonamallee highway location (106.5 mg/L nitrate, on road location with commercial and residential areas), Thiruverkadu town (120.5 mg/L nitrate, temple-town commercial district next to bus-stand), Gummidipoondi industrial town (105.9 mg/L nitrate, town amidst highly industrialized belt) were found to be hotspots with nitrate concentrations >100.0 mg/L. Apart from this, four more locations, groundwater at Poonamallee highway location (57.4 mg/L nitrate, on road location with commercial and residential areas), Thiruttani town (50.1 mg/L nitrate, commercial district in high visitor-traffic temple town), Janappanchatram village (59.9 mg/L nitrate, highway junction with food-outlets, motels and residences adjacent to the Kosasthalaiyar river) and Arani town (52.9 mg/L nitrate, high residential population density town) were found to have above permissible limits of nitrate. It is interesting to note that the treated lake water and groundwater collected at the same Poonamallee highway location had very high and relatively lower nitrate levels, respectively, an indication of the poor efficiency of the water treatment plant. Temple-towns, Thiruverkadu and Thiruttani suffer from high-tourist traffic and the sampling spots chosen for with their proximity to the bus station and temples, as expected show high nitrate levels. Gummidipoondi is a heavily industrialized town, while Arani and Janappanchatram are high-population density highway towns with the sampling spot located in the midst of residential areas. The treated Chembarambakkam lake water unexpectedly exhibited a very high nitrate level

of 106.5 mg/L, while the treated Ambattur – Athipattu lake water produced much lower nitrate level of 2.7 mg/L. It is interesting to note that, Red Hills lake and Padianallur pond (untreated water samples) have merely 1.0 and 20.0 mg/L nitrate, respectively. The locations in these three districts were chosen anticipating high nitrate pollution levels due to either anthropogenic activity associated urban sewage run-off, agricultural activity induced fertilizer run-off and industrial activity related pollution and results of the investigation have proved that the assumptions to be true.

4.3. Fluoride pollution in groundwater

4.3.1. Chennai district

Fig. 3A–C marks the drinking water locations sampled for surveying fluoride concentrations in the three districts studied. The locations are color-coded based on three discrete fluoride concentration ranges, <1.0 mg/L, 1.0–1.5 mg/L and 1.5–3.0 mg/L. The water sampling locations latitude, longitude and the fluoride concentrations are documented in Tables 3–5. Overall, taking all the 103 samples in consideration, the minimum, maximum and average concentration of fluoride ions in groundwater samples investigated were found to be 0.3, 2.7 and 0.9 mg/L, respectively. Interestingly, the average value of 0.9 mg/L is below current standard prescribed limit in India (1 mg/L, Table 3) for fluoride. In Chennai district, all the 24 surveyed locations exhibited meager levels of fluoride content, below the permissible limit suggesting that high-population density induced urban sewage run-off, leaching from fluoride ores from rocks and/or industrial activity may not contribute significantly to fluoride pollution in the city (Fig. 3A). Overall, the concentration based analysis results for Chennai district show that residents are not likely to suffer from any ill effects associated with high fluoride in water content.

4.3.2. Kancheepuram district

In Kancheepuram district, out of the 41 samples, 12 were found to possess fluoride concentrations above the permissible 1.0 mg/L limit. The highest fluoride levels of 2.6 mg/L was detected in densely populated Chengalpattu town which is present near Palar River and rocky terrain. A second hotspots was located at Ennaikaran (1.7 mg/L fluoride, densely populated Kancheepuram town suburb in the vicinity of rocky terrain) with both locations exhibiting fluoride concentrations >1.5 mg/L and marked with red colored tag in the Fig. 3B. The other locations with excess fluoride levels were: Mamandur (1.3 mg/L fluoride, highway town with high tourist traffic, motels and food outlets), Padhalam village (1.3 mg/L fluoride, rocky terrain on the shores of the Palar River), openwell in Karunguzhi (1.2 mg/L fluoride, highway town on rocky terrain near two lakes), Treated water in GST road near Madurantakam town (1.2 mg/L fluoride, high population density town near a lake), openwell in Sirunallur (1.2 mg/L fluoride, openwell location in village with paint factory), openwell in Melmaruvathur town (1.2 mg/L fluoride, highway temple-town with high-visitor traffic), openwell in Kamalapoondi (1.2 mg/L fluoride, very rocky terrain with hills near Uthiramerur), Vedal (1.2 mg/L fluoride, rural area with rocky terrain and hills near the sea), openwell in Urapakkam (1.4 mg/L fluoride, openwell location on urbanized highway town with high population density) and Vandalur (1.2 mg/L fluoride, urbanized highway town). Many of these locations are present amidst rocky terrain of Kancheepuram district, next to water bodies.

4.3.3. Tiruvallur district

In contrast to this, the survey of Tiruvallur district for fluoride levels shows that among the 38 selected spots surveyed, 26 locations had less than the permissible limit of 1 mg/L. However, 6 sampling spots each were found to present fluoride levels in the 1.0 to 1.5 mg/L and 1.5 to 3.0 mg/L ranges, respectively. The hotspots for fluoride contamination were Tiruvallur town (2.7 mg/L fluoride, urban area north of Chennai with high population density and near a lake), Poonamallee (2.3 mg/L fluoride, on road location with commercial and residential areas), Ambattur Industrial estate (1.6 mg/L fluoride, industrial area surrounded by high residential population density), Ambattur industrial estate (2.1 mg/L fluoride, industrial area surrounded by high residential population density and 2 km away from the previous spot), Padianallur (2.0 mg/L fluoride, north of Chennai city and close to Puzhal (Red Hills) lake) and Thaanipoondi (1.6 mg/L fluoride, rural area near the state border with hilly terrain). The other locations with high fluoride content were openwell Poonamallee (1.2 mg/L fluoride, on road location with commercial and residential areas), Thiruverkadu town (1.1 mg/L fluoride, temple-town commercial district next to bus-stand), openwell Ambattur industrial estate (1.2 mg/L, industrial area surrounded by high residential population density), Padianallur pond (1 mg/L fluoride, north of Chennai city and close to Puzhal (Red Hills) lake), Thandalachery (1.1 mg/L fluoride, rocky terrain rural area near Kavaraipettai, north of Chennai), Vadamadurai (1.2 mg/L fluoride, highway spot on the Araniar river shore and north of Chennai), Tharatchi village (1.2 mg/L fluoride, village on Araniar river shore and upstream of Vadamadurai) and Panchetti (1 mg/L fluoride, on Araniar river shore and downstream of Vadamadurai). Many of the locations in the three districts, especially those of Tiruvallur districts were strategically chosen based on their proximity to water bodies. The results show that as expected, locations close to lakes and riverbeds appeared to have enhanced fluoride levels. This is also observed explicitly in the Araniar river shore locations of Vadamadurai, Tharatchi village and Panchetti. The presence of fluoride minerals-rich hilly and rocky areas capable of leaching the fluorides and transporting them downstream through rivers, streams and lakes in the likely cause for presence of high fluoride levels in the ponds, lakes and their shores of these regions.

Tamil Nadu possess major minerals, limestone, magnesite, graphite, bauxite, lignite and iron ore. It also has deposits of minor minerals such as quartz, feldspar, gypsum etc. However, these minerals do not occur in the three districts studied. [35]. In fact, fluorosis in patients are mainly reported from Salem, Ariyalur, Srivilliputhur, Tiruchengode, Namakkal and Dindigul districts of Tamil Nadu [36]. The three districts have so far not been reported as fluoride rich groundwater regions of Tamil Nadu and our results reiterate these earlier findings, but throw light on hotspots

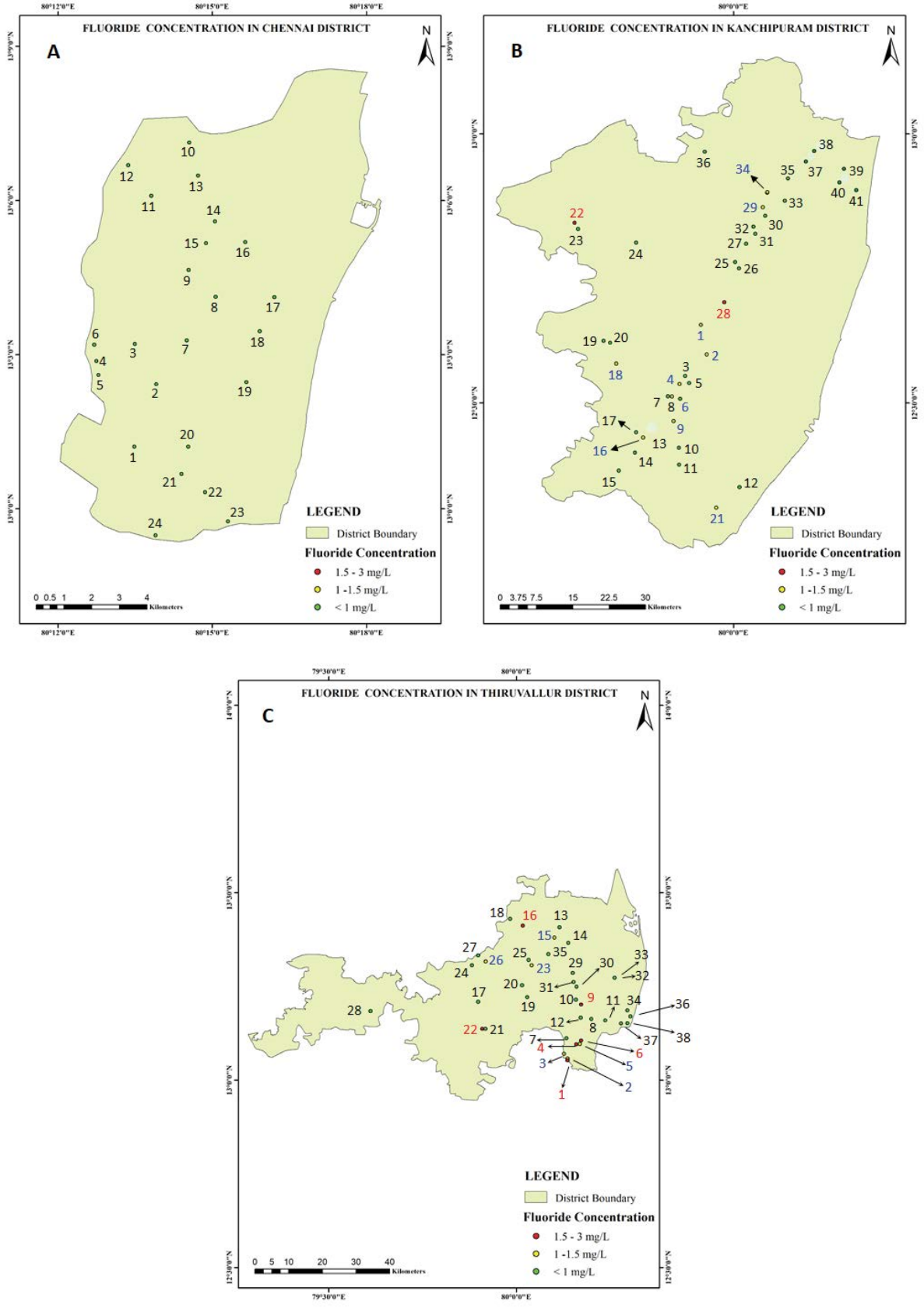

Fig. 3. Fluoride distribution in (A) Chennai, (B) Kancheepuram and (C) Tiruvallur district of Tamil Nadu state, India.

where the fluoride concentrations are likely to be high enough to cause fluorosis.

4.4. Influence of water quality parameters on nitrate and fluoride content in groundwater

The mapping of the spatial distribution of nitrate and fluoride content in the study area has to be complemented with additional analysis to understand their relationship with other water quality parameters. A strong positive correlation, say between fluoride concentrations and total hardness data in a water sample would suggest that these parameters are related and that fluoride leaching and accumulation at the water source is directly or indirectly responsible for the enhanced total hardness of the water sample. Hence, a linear regression analysis was employed to assess the dependence of nitrate and fluoride levels on water quality parameters (TDS, EC, alkalinity, total hardness, chloride and sulphate) as shown in Fig. 4A–F. The R^2 values from these plots can be used to assess the degree of correlation between the variables. A glance at the R^2 values shows that fluoride ion concentration appears to correlate better against the water quality parameters than the nitrate ion concentration. For example, in Fig. 4A, nitrate and fluoride concentrations plotted against TDS show that the R^2 value to be 0.088 and 0.105, respectively. The comparison of these R^2 values can be used to infer that fluoride appears to have better correlation with TDS than nitrate. The study of the other correlations in Fig. 4B–F, shows that a similar trend with fluoride presenting better correlation than nitrate against the water quality parameters.

It is also interesting to note that while most plots showed marginal differences in correlation between fluoride and nitrate, with alkalinity, fluoride exhibited a relatively marked difference in correlation (R^2 = 0.317) than nitrate (R^2 = 0.078) (Fig. 4C). The relatively higher correlation between alkalinity and fluoride levels has been attributed to the concomitant discharge of bicarbonate ions during the dissolution of fluoride bearing minerals into the groundwater [37–41]. This theory is also supported by the relatively higher regression coefficient for fluoride levels against total hardness levels (Fig. 4D). The fluoride minerals dissolution process is also expected to release calcium and magnesium ions which go onto increase the hardness levels in groundwater. It is also important to note that in this study, the regions with rocky and hilly terrain exhibited higher fluoride content in groundwater resources supporting the above inferences.

The nitrate concentration levels with their R^2 values appear to show positive but relatively poor correlation with the water quality parameters. As reported in earlier literature, the major sources of nitrate in drinking water is sewage run-off from domestic activity and fertilizer run-off from agricultural activity [42–45]. This run-off might not affect the water quality parameters and this is most probably the rationale for nitrate levels in this study to correlate poorly against the water quality parameters.

4.5. Nitrate and fluoride based non-carcinogenic human health risk assessment

Chennai and its adjoining districts with high population density, industrial activity, rivers flowing to the Bay of Bengal, stagnant water bodies and lake reclamation areas may be expected to more susceptible to nitrate and fluoride pollution. The rivers turn more polluted as their flow eastward towards the Chennai basin and are therefore more likely to transport and accumulate nitrate and leached fluoride in downstream water bodies. Chennai has three polluted rivers, Kosasthalaiyar, Cooum and Adyar meandering through it from west to east to reach the sea. Tiruvallur district also has the polluted Araniar, Cooum and Kosasthalaiyar rivers flowing through it with two major water bodies, Poondi reservoir and Puzhal (Red Hills) lake. Kancheepuram has the Adyar, Cheyyar and Palar Rivers flowing through it to the sea along with Odiyur, Maduranthakam and Chembarambakkam lakes. The drinking water is sourced to the city and its suburbs from a combination of groundwater, ponds and lakes. Hence, water from these districts were surveyed for their physico-chemical parameters and analyzed specifically for the most widespread non-carcinogenic nitrate and fluoride water contaminants. A preliminary analysis of the results summarized for the three districts (Table 2) show that groundwater nitrate and fluoride contamination as per BIS acceptable limits is exceeded by only 18.4% and 23.3% of the samples, respectively [6]. However, these preliminary results are only an indication and a more probability-based prediction methodology based on CDI and HQ calculations is needed to assess the extent of the health risk.

Although, water can be absorbed dermally, it is considered negligible and oral ingestion is considered the most prominent route of introduction. Hence, this study focused on the hypothesis that ingestion of nitrate and fluoride contaminated groundwater can a potential non-carcinogenic health hazards to infants, children and adults in the study area. The extent of this hazard was assessed with the CDI and human HQ calculations outlined in the methods section. The results of this analysis are summarized in Tables 6–8 for Chennai, Kancheepuram and Tiruvallur districts.

4.5.1. Chennai district

The results for $HQ_{nitrate}$ analysis of Chennai district predicts negligible minimum values in the dataset, but the maximum values for infants, children and adults are 2.40, 1.48 and 0.79, respectively (Table 6). At the outset, Chennai district appears to be relatively safe for adults with many locations showing $HQ_{nitrate} < 1.00$. While, most locations in the district show very poor nitrate accumulation and $HQ_{nitrate}$ values in groundwater, two locations, openwell water at Villivakkam (33.9 mg/L nitrate, $HQ_{nitrate}$ = 1.98, 1.23 and 0.65 for infants, children and adults, respectively) and Perambur (41.0 mg/L nitrate, $HQ_{nitrate}$ = 2.40, 1.48 and 0.79 for infants, children and adults, respectively) exhibit above limit $HQ_{nitrate}$ values for infants and children. In these locations, adults appear to be predicted to be relatively safe from the risk associated with ingestion of nitrate contaminated drinking. These results mirror the concentration-based analysis results of section 4.2 for Chennai district which show that Villivakkam and Perambur to be two locations with relatively high, but below permissible limit nitrate concentrations. Villivakkam and Perambur neighborhoods of Chennai city have an Indian railway locomotive

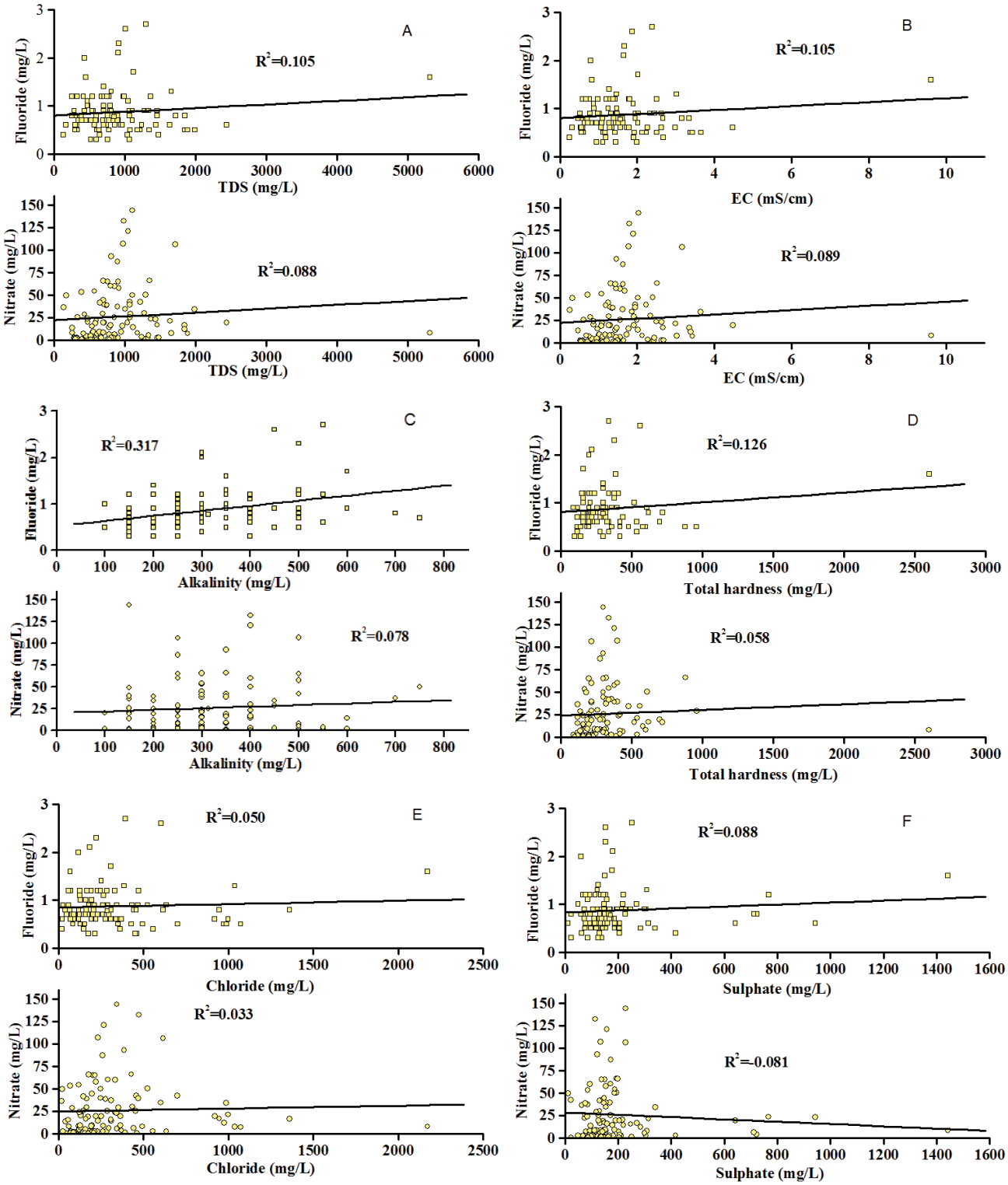

Fig. 4. Linear regression plots analyzing the dependence of nitrate and fluoride on (A) total dissolved solids, (B) electrical conductivity, (C) alkalinity, (D) total hardness, (E) chloride and (F) sulphate.

manufacturing facility along with the presence of many allied industries. The effluents emanating from these industries are likely to contribute to the high nitrate levels in these localities. The data analysis also shows that only 8.33% of the samples exceed the HQ_{nitate} limits for infants and children, indicating that apart from Villivakkam and Perambur, other locations in the district are not likely to pose any nitrate induced health hazard for residents. This is also reflected

Table 6
Nitrate and fluoride hazard quotient and total hazard index in different locations of Chennai district, Tamil Nadu, India

	Location	HQ - nitrate			HQ - fluoride			THI		
		Infants	Children	Adults	Infants	Children	Adults	Infants	Children	Adults
1	Saidapet	0.71	0.44	0.23	1.17	0.72	0.39	1.88	1.16	0.62
2	T. Nagar	0.09	0.06	0.03	1.40	0.87	0.46	1.50	0.93	0.49
3	Kodambakkam	0.42	0.26	0.14	1.17	0.72	0.39	1.59	0.98	0.52
4	Vadapalani	0.53	0.33	0.17	1.40	0.87	0.46	1.93	1.19	0.64
5	Saligramam	0.25	0.16	0.08	1.64	1.01	0.54	1.89	1.17	0.62
6	Virugambakkam	0.15	0.09	0.05	1.64	1.01	0.54	1.79	1.10	0.59
7	Nungambakkam	0.14	0.09	0.05	2.11	1.30	0.70	2.25	1.39	0.74
8	Egmore	0.08	0.05	0.03	1.17	0.72	0.39	1.25	0.77	0.41
9	Kilpauk	0.06	0.04	0.02	1.87	1.16	0.62	1.93	1.19	0.64
10	Aminjikarai	0.16	0.10	0.05	0.94	0.58	0.31	1.09	0.68	0.36
11	Ayanavaram	0.09	0.06	0.03	1.40	0.87	0.46	1.50	0.93	0.49
12	Villivakkam*	1.98	1.23	0.65	1.17	0.72	0.39	3.16	1.95	1.04
13	Perambur	2.40	1.48	0.79	0.94	0.58	0.31	3.34	2.06	1.10
14	Otteri	0.07	0.04	0.02	1.40	0.87	0.46	1.48	0.91	0.49
15	Purasaiwakkam	0.06	0.04	0.02	1.64	1.01	0.54	1.70	1.05	0.56
16	Vepery	0.40	0.25	0.13	1.17	0.72	0.39	1.57	0.97	0.52
17	Chintadripet	0.20	0.12	0.07	1.40	0.87	0.46	1.60	0.99	0.53
18	Triplicane	0.13	0.08	0.04	2.11	1.30	0.70	2.24	1.38	0.74
19	Mylapore	0.54	0.33	0.18	1.17	0.72	0.39	1.71	1.06	0.56
20	Kotturpuram	0.02	0.01	0.01	1.17	0.72	0.39	1.19	0.73	0.39
21	Guindy	0.92	0.57	0.30	1.40	0.87	0.46	2.32	1.44	0.77
22	Adayar	0.01	0.01	0.00	0.70	0.43	0.23	0.71	0.44	0.24
23	Thiruvanmiyur	0.29	0.18	0.10	2.11	1.30	0.70	2.40	1.48	0.79
24	Velachery*	0.47	0.29	0.15	1.40	0.87	0.46	1.87	1.16	0.62
	Minimum	0.01	0.01	0.00	0.70	0.43	0.23	071	0.44	0.24
	Maximum	2.40	1.48	0.79	2.11	1.30	0.70	3.34	2.06	1.10
	Mean ± Std. Dev.	0.42 ± 0.59	0.26 ± 0.36	0.14 ± 0.19	1.41 ± 0.39	0.87 ± 0.24	0.46 ± 0.13	1.83 ± 0.62	1.13 ± 0.38	0.61 ± 0.19
	% Samples > 1.00	8.33	8.33	0.00	87.50	29.17	0.00	95.83	58.33	8.33

*Openwell water

in the skewed standard deviation values that is comparable in magnitude to the mean values ($HQ_{nitrate}$, mean ± standard deviation of 0.42 ± 0.59, 0.26 ± 0.36 and 0.14 ± 0.19 for infants, children and adults, respectively) in the data set. These relatively high $HQ_{nitrate}$ levels in these locations are most probably due to a combination of untreated human sanitary sewage, decomposition of food and leakage from septic systems.

Table 6 also lists the $HQ_{fluoride}$ predicted by the U.S. EPA based methodology. The minimum values in the dataset were all <1.00, lower than the acceptable limit, but the $HQ_{fluoride}$ maximum values were 2.11, 1.30 and 0.70 for infants, children and adults, respectively. The $HQ_{fluoride}$ mean ± standard deviation for the dataset is 1.41 ± 0.39, 0.87 ± 0.24, 0.46 ± 0.13 for infants, children and adults, respectively. The mean values data suggest that only infants are likely to be susceptible to fluoride in drinking water associated health hazards. The dataset also shows that densely populated lake reclamation area, Nungambakkam (0.9 mg/L fluoride, $HQ_{fluoride}$ for infants, children and adults = 2.11, 1.30 and 0.70, respectively), densely populated residential area on the sea shore, Triplicane (0.9 mg/L fluoride, $HQ_{fluoride}$ for infants, children and adults = 2.11, 1.30 and 0.70, respectively) and densely populated residential area on the sea shore, Thiruvanmiyur (0.9 mg/L fluoride, $HQ_{fluoride}$ for infants, children and adults = 2.11, 1.30 and 0.70, respectively) with their near-acceptable threshold fluoride concentrations to be responsible for the high mean $HQ_{fluoride}$ values predicted for infants and children in Chennai district. Nungambakkam is a lake reclamation area which his likely to accumulate nitrate from rain-water run-offs from surrounding areas. Triplicane and Thiruvanmiyur are beside the seashore and likely to accumulate nitrate due to rain-water run-offs from interior areas. Apart from these locations, there are other locations in the district which appear to have $HQ_{fluoride}$ > 1.50 for infants namely, very densely populated Saligramam, Virugambakkam, Kilpauk and Purasaiwakkam. The fluoride accumulation in these urban areas are most probably due to combination of human activity as well as leached fluoride ores being deposited by rain-water run-offs.

The composite HQ values for the nitrate and fluoride concentrations, THI values for Chennai district with

Table 7
Nitrate and fluoride hazard quotient and total hazard index in different locations of Kancheepuram district, Tamil Nadu, India

	Location	HQ - nitrate			HQ - fluoride			THI		
		Infants	Children	Adults	Infants	Children	Adults	Infants	Children	Adults
1	Mamandur	5.41	3.35	1.79	3.04	1.88	1.00	8.46	5.23	2.79
2	Padhalam	0.44	0.28	0.15	3.04	1.88	1.00	3.49	2.16	1.15
3	Melavalaipettai*	0.16	0.10	0.05	2.11	1.30	0.70	2.27	1.40	0.75
4	Karunguzhi*	0.06	0.04	0.02	2.81	1.74	0.93	2.87	1.77	0.95
5	Karunguzhi	0.14	0.09	0.05	2.34	1.45	0.77	2.48	1.53	0.82
6	Maduranthakam**	0.13	0.08	0.04	2.81	1.74	0.93	2.94	1.82	0.97
7	Maduranthakam	2.15	1.33	0.71	1.87	1.16	0.62	4.03	2.49	1.33
8	Maduranthakam	2.24	1.38	0.74	1.40	0.87	0.46	3.64	2.25	1.20
9	Sirunallur*	7.73	4.78	2.55	2.81	1.74	0.93	10.54	6.52	3.48
10	Chittamur	2.29	1.42	0.76	2.11	1.30	0.70	4.40	2.72	1.45
11	Pazhuvur	0.78	0.49	0.26	1.87	1.16	0.62	2.66	1.64	0.88
12	Cheyyur*	2.92	1.80	0.96	1.64	1.01	0.54	4.55	2.82	1.50
13	Sottupakkam	8.42	5.21	2.78	2.11	1.30	0.70	10.53	6.51	3.48
14	Acharapakkam*	3.51	2.17	1.16	2.11	1.30	0.70	5.62	3.47	1.85
15	Tozhupedu	0.90	0.55	0.30	2.11	1.30	0.70	3.00	1.86	0.99
16	Melmaruvathur*	2.31	1.43	0.76	2.81	1.74	0.93	5.12	3.17	1.69
17	Melmaruvathur	0.25	0.16	0.08	1.64	1.01	0.54	1.89	1.17	0.62
18	Kamalapoondi*	0.42	0.26	0.14	2.81	1.74	0.93	3.23	1.99	1.06
19	Uthiramerur	3.80	2.35	1.26	1.64	1.01	0.54	5.44	3.37	1.80
20	Uthiramerur*	2.60	1.61	0.86	2.34	1.45	0.77	4.94	3.06	1.63
21	Vedal	0.46	0.29	0.15	2.81	1.74	0.93	3.27	2.02	1.08
22	Ennaikaran	0.82	0.51	0.27	3.98	2.46	1.31	4.80	2.97	1.58
23	Kancheepuram	0.12	0.08	0.04	2.11	1.30	0.70	2.23	1.38	0.74
24	Walajabad	1.76	1.09	0.58	2.11	1.30	0.70	3.86	2.39	1.28
25	SP Koil	0.50	0.31	0.16	2.11	1.30	0.70	2.60	1.61	0.86
26	SP Koil*	0.36	0.22	0.12	1.64	1.01	0.54	2.00	1.23	0.66
27	MM Nagar	3.17	1.96	1.05	1.40	0.87	0.46	4.57	2.83	1.51
28	Chengalpattu	2.00	1.24	0.66	6.09	3.76	2.01	8.09	5.00	2.67
29	Urapakkam*	1.12	0.69	0.37	3.28	2.03	1.08	4.40	2.72	1.45
30	Guduvancheri*	0.46	0.29	0.15	1.17	0.72	0.39	1.63	1.01	0.54
31	Potheri	3.86	2.39	1.28	1.17	0.72	0.39	5.03	3.11	1.66
32	SRM Nagar	1.67	1.03	0.55	1.17	0.72	0.39	2.84	1.76	0.94
33	Peerkankaranai	0.14	0.09	0.05	2.11	1.30	0.70	2.25	1.39	0.74
34	Vandalur	2.27	1.40	0.75	2.81	1.74	0.93	5.08	3.14	1.68
35	Perungalathur	2.88	1.78	0.95	1.40	0.87	0.46	4.28	2.65	1.41
36	Sriperumbudur	1.33	0.83	0.44	1.40	0.87	0.46	2.74	1.69	0.90
37	Chromepet	3.83	2.37	1.27	2.11	1.30	0.70	5.94	3.67	1.96
38	Pallavaram	5.07	3.13	1.67	1.64	1.01	0.54	6.71	4.15	2.21
39	Pallikaranai	1.12	0.69	0.37	0.94	0.58	0.31	2.05	1.27	0.68
40	Medavakkam	3.80	2.35	1.26	0.70	0.43	0.23	4.51	2.79	1.49
41	Sholinganallur	1.46	0.90	0.48	0.70	0.43	0.23	2.17	1.34	0.71
	Minimum	0.06	0.04	0.02	0.70	0.43	0.23	1.63	1.01	0.54
	Maximum	8.42	5.21	2.78	6.09	3.76	2.01	10.54	6.52	3.48
	Mean ± Std. Dev.	0.42 ± 0.59	0.26 ± 0.36	0.14 ± 0.19	1.41 ± 0.39	0.87 ± 0.24	0.46 ± 0.13	1.83 ± 0.62	1.13 ± 0.38	0.61 ± 0.19
	% Samples > 1.00	60.98	51.22	24.39	92.68	75.61	12.20	100.00	100.00	60.98

*Openwell water;
**Treated lake water.

Table 8
Nitrate and fluoride hazard quotient and total hazard index in different locations of Tiruvallur district, Tamil Nadu, India

	Location	HQ - nitrate			HQ - fluoride			THI		
		Infants	Children	Adults	Infants	Children	Adults	Infants	Children	Adults
1	Poonamallee	3.36	2.08	1.11	5.39	3.33	1.78	8.75	5.41	2.89
2	Poonamallee**	6.23	3.85	2.06	2.81	1.74	0.93	9.04	5.59	2.98
3	Thiruverkadu	7.05	4.36	2.33	2.58	1.59	0.85	9.63	5.95	3.18
4	Ambattur Ind. E.	0.46	0.28	0.15	3.75	2.32	1.24	4.20	2.60	1.39
5	Ambattur Ind. E.**	0.16	0.10	0.05	2.81	1.74	0.93	2.97	1.84	0.98
6	Ambattur Ind. E.	0.15	0.09	0.05	4.92	3.04	1.62	5.06	3.13	1.67
7	Ambattur	1.28	0.79	0.42	1.64	1.01	0.54	2.92	1.80	0.96
8	Puzhal	1.49	0.92	0.49	1.17	0.72	0.39	2.66	1.65	0.88
9	Padianallur	0.02	0.01	0.01	4.68	2.90	1.55	4.70	2.91	1.55
10	Padianallur****	1.18	0.73	0.39	2.34	1.45	0.77	3.52	2.18	1.16
11	Madhavaram	1.13	0.70	0.37	1.40	0.87	0.46	2.53	1.57	0.84
12	Red Hills Lake***	0.06	0.04	0.02	1.87	1.16	0.62	1.93	1.19	0.64
13	Gummidipoondi	6.20	3.83	2.05	1.87	1.16	0.62	8.07	4.99	2.66
14	Kavaraipettai	0.49	0.30	0.16	1.64	1.01	0.54	2.13	1.32	0.70
15	Thandalachery	1.39	0.86	0.46	2.58	1.59	0.85	3.96	2.45	1.31
16	Thaanipoondi	0.05	0.03	0.02	3.75	2.32	1.24	3.79	2.35	1.25
17	Neyveli village	0.52	0.32	0.17	2.11	1.30	0.70	2.63	1.63	0.87
18	Sathyavedu	2.11	1.30	0.70	0.94	0.58	0.31	3.04	1.88	1.00
19	Thamaraipakkam	0.23	0.14	0.08	1.87	1.16	0.62	2.10	1.30	0.69
20	Vengal	0.00	0.00	0.00	1.64	1.01	0.54	1.64	1.01	0.54
21	Kakkalur byepass	1.12	0.69	0.37	1.40	0.87	0.46	2.53	1.56	0.83
22	Tiruvallur	0.07	0.04	0.02	6.32	3.91	2.09	6.39	3.95	2.11
23	Vadamadurai	0.84	0.52	0.28	2.81	1.74	0.93	3.65	2.26	1.20
24	Uthukkottai	1.70	1.05	0.56	1.64	1.01	0.54	3.34	2.07	1.10
25	Periyapalayam	1.66	1.03	0.55	1.64	1.01	0.54	3.30	2.04	1.09
26	Tharatchi village	1.36	0.84	0.45	2.81	1.74	0.93	4.17	2.58	1.38
27	Uthukkottai	1.08	0.67	0.36	1.64	1.01	0.54	2.72	1.68	0.90
28	Thiruttani	2.93	1.81	0.97	2.11	1.30	0.70	5.04	3.12	1.66
29	Panchetti	0.94	0.58	0.31	2.34	1.45	0.77	3.28	2.03	1.08
30	Karanodai	0.35	0.21	0.11	1.87	1.16	0.62	2.22	1.37	0.73
31	Janappanchatram	3.51	2.17	1.16	1.40	0.87	0.46	4.91	3.04	1.62
32	Minjur	0.94	0.58	0.31	1.87	1.16	0.62	2.82	1.74	0.93
33	Minjur*	0.97	0.60	0.32	1.87	1.16	0.62	2.84	1.76	0.94
34	Ennore	0.02	0.01	0.01	0.70	0.43	0.23	0.73	0.45	0.24
35	Arani	3.10	1.91	1.02	1.87	1.16	0.62	4.97	3.07	1.64
36	Tiruvottiyur	2.46	1.52	0.81	1.17	0.72	0.39	3.64	2.25	1.20
37	Tiruvottiyur	1.23	0.76	0.41	1.40	0.87	0.46	2.63	1.63	0.87
38	Tiruvottiyur*	2.45	1.51	0.81	1.87	1.16	0.62	4.32	2.67	1.43
	Minimum	0.00	0.00	0.00	0.70	0.43	0.23	0.73	0.45	0.24
	Maximum	7.05	4.36	2.33	6.32	3.91	2.09	9.63	5.95	3.18
	Mean ± Std. Dev.	1.59 ± 1.76	0.98 ± 1.09	0.52 ± 0.58	2.33 ± 1.25	1.44 ± 0.77	0.77 ± 0.41	3.92 ± 2.06	2.42 ± 1.27	1.29 ± 0.68
	% Samples > 1.00	55.26	31.58	15.79	94.74	78.95	15.79	97.37	97.37	57.89

*Openwell water;
**Treated lake water;
***Lake water;
****Pond water.

respect to minimum, maximum, mean ± standard deviation values were 0.71, 6.71, 2.25 ± 1.35 for infants, 0.44, 4.15, 1.39 ± 0.83 for children and 0.24, 3.18, 0.74 ± 0.45 for adults. The minimum values in the dataset are all much lower than the prescribed THI < 1.00 limit, however the relatively high maximum and the standard deviation values suggest that there a few areas in Chennai district which suffer from very high THI. Hence, it is not surprising to note that in contrast to the results of the concentration-based spatial distribution data of Table 3, Table 6 shows that many areas of Chennai possess THI > 1 and would therefore pose a health hazard from the nitrate and fluoride in water pollution for infants and children. Two areas that need specific mention are: openwell water of Villivakkam (THI = 3.16, 1.95 and 1.04 for infants, children and adults, respectively) and Perambur (THI = 3.34, 2.06 and 1.10 infants, children and adults, respectively), which mirror the results of $HQ_{nitrate}$ and $HQ_{fluoride}$ analysis. As stated earlier, these are located near Indian railway locomotive manufacturing factory and other allied units. The high nitrate and fluoride concentrations as well as the THI > 1.0 levels can be ascribed to the effluents discharged from these units. A few other locations that indicated high THI are densely populated lake reclamation urban area, Nungambakkam (THI = 2.25, 1.39 and 0.74 for infants, children and adults, respectively), densely populated urban residential area beside the sea shore, Triplicane (THI = 2.24, 1.38 and 0.74 for infants, children and adults, respectively), urban area with student population and small-scale industries, Guindy (THI = 2.32, 1.44 and 0.77 for infants, children and adults, respectively) and densely populated urban residential area beside the sea shore, Thiruvanmiyur (THI = 2.40, 1.48 and 0.79 for infants, children and adults, respectively). The THI percentage exceedance rate for Chennai district is 95.83%, 58.33% and 8.33% for infants, children and adults, respectively, which is marked by a significant difference between adults and non-adults. Overall, the results show that in spite of concentration analysis data providing lower than permissible levels of nitrate and fluoride in all locations of Chennai district (Table 3), the non-carcinogenic nitrate and fluoride based health hazard assessment data (Table 6) suggests that infants and children in certain hotspots to be highly vulnerable than adults.

4.5.2. Kancheepuram district

The $HQ_{nitrate}$, $HQ_{fluoride}$ and THI results from 41 locations in Kancheepuram district are shown in Table 7. The data statistics for $HQ_{nitrate}$ shows that the minimum values are near negligible, while the maximum values of infants, children and adults are 8.42, 5.21 and 2.78 mg/L, respectively from the Sottupakkam sample (143.9 mg/L nitrate), with the values far exceeding the maximum permissible limit. Apart from this, the mean ± standard deviation values for infants, children and adults are 1.93 ± 2.02, 1.2 ± 1.25 and 0.64 ± 0.67 mg/L, respectively, all of which are marked by standard deviations comparable in magnitude to the mean values. This can be attributed to the markedly high nitrate levels of four locations including Mamandur, a highway town with high tourist traffic, motels and food outlets (92.5 mg/L nitrate, $HQ_{nitrate}$ for infants, children and adults = 5.41, 3.35 and 1.79, respectively), openwell water from Sirunallur located in village with paint factory (132 mg/L nitrate, $HQ_{nitrate}$ for infants, children and adults = 7.73, 4.78 and 2.55, respectively), Sottupakkam, a rural town with agricultural activity, but located at the junction of two major highways with many food-outlets, motels and a residential high student-density college (143.9 mg/L nitrate, $HQ_{nitrate}$ for infants, children and adults = 8.42, 5.21 and 2.78, respectively) and Pallavaram, a Chennai city suburb with tanning industry cluster (86.6 mg/L nitrate, THI for infants, children and adults = 5.07, 3.13 and 1.67, respectively). These four locations are predicted to have significantly high $HQ_{nitrate}$ for infants > 5.00.

Apart from this, other Kancheepuram district location with 4.00 < $HQ_{nitrate}$ for infants < 5.00 are, Potheri, a highway college campus town with very high student population density (66 mg/L nitrate, $HQ_{nitrate}$ for infants, children and adults = 3.86, 2.39 and 1.28, respectively), Chromepet, a Chennai city suburb with tanning industry cluster (65.5 mg/L nitrate, $HQ_{nitrate}$ for infants, children and adults = 3.83, 2.37 and 1.27, respectively), Uthiramerur, a temple town with tourists activity, high population density and nearby lakes (65.0 mg/L nitrate, $HQ_{nitrate}$ = 3.80, 2.35 and 1.26, respectively), Medavakkam, a Chennai city suburb with high population density and small industries (65.0 mg/L nitrate, $HQ_{nitrate}$ = 3.80, 2.35 and 1.26, respectively), open well water of Acharapakkam, a densely populated highway town with food-outlets and railway station (60.0 mg/L nitrate, $HQ_{nitrate}$ for infants, children and adults = 3.51, 2.17 and 1.16, respectively) and MM Nagar, a highway town abutting Tamil Nadu Government's Industrial zone (54.1 mg/L nitrate, $HQ_{nitrate}$ for infants, children and adults = 3.17, 1.96 and 1.05, respectively). Mamandur, Pallavaram, Potheri, Chromepet, Acharapakkam, MM Nagar and Sottupakkam are all highway towns, while Uthiramerur is a temple town with high visitor traffic and Medavakkam is a newly developed Chennai city suburb with high population density. The high nitrate concentration and associated $HQ_{nitrate}$ values in these as well as other locations listed in Table 7 are most probably due to a combination of untreated sanitary sewage, decomposition of food and agricultural run-offs.

The maximum value of the $HQ_{fluoride}$ values for infants, children and adults in Kancheepuram district are 6.09, 3.76 and 2.01, respectively in Chengalpattu (2.6 mg/L fluoride), which is a densely populated town next to Palar River situated amidst rocky terrain (Table 7). The $HQ_{fluoride}$ mean ± standard deviation values are 2.15 ± 0.96, 1.33 ± 0.6 and 0.71 ± 0.32 mg/L, respectively. The high mean values for infants and children suggest considerable health hazard from fluoride contamination in Kancheepuram district. Apart from Chengalpattu, the high mean $HQ_{fluroide}$ values also emanates from following locations: Ennaikaran (1.7 mg/L fluoride, highway town near Kancheepuram town with rocky terrain, $HQ_{fluoride}$ for infants, children and adults = 1.64, 1.01 and 0.54, respectively), openwell water of Urapakkam (1.4 mg/L fluoride, urbanized highway town with high population density, $HQ_{fluoride}$ for infants, children and adults = 3.28, 2.03 and 1.08, respectively), Mamandur (1.3 mg/L fluoride, highway town with high tourist traffic, motels, food outlets and agricultural activity $HQ_{fluoride}$ for infants, children and adults = 3.04, 1.88 and 1.00, respectively) and Padhalam, (1.3 mg/L fluoride, village on Palar

River shore with agricultural activity and rocky terrain, $HQ_{fluoride}$ for infants, children and adults = 3.04, 1.88 and 1.00, respectively). The highway towns listed above are likely to accumulate fluoride from leaching of ores from the rocky terrain of Kancheepuram district.

The percentage $HQ_{nitrate}$ exceedance values for infants, children and adults are 55.56%, 50.00% and 19.44%, respectively, while the values for $HQ_{fluoride}$ are 100.00, 80.56 and 13.89, respectively, which indicates that majority of the infants and children in Kancheepuram district are susceptible to nitrate and fluoride in water contamination health hazards. The relatively high $HQ_{nitrate}$ and $HQ_{fluoride}$ values also results in very high THI values for many locations in the district. For example, the three highest THI were observed for Sirunallur (THI = 10.54 for infants, 6.52 for children and 3.48 for adults), Sottupakkam (THI = 10.53 for infants, 6.51 for children and 3.48 for adults), Mamandur (THI = 8.46 for infants, 5.23 for children and 2.79 for adults) Chengalpattu (THI = 8.09 for infants, 5.00 for children and 2.67 for adults) and Pallavaram (THI = 6.71 for infants, 4.15 for children and 2.21 for adults). The mean ± standard deviation values were 4.22 ± 2.16, 2.61 ± 1.33 and 1.39 ± 0.71 for infants, children and adults, respectively. The percentage exceedance for the THI limit were 100.0, 100.0 and 61.0, respectively. These data put-together can be used to show that infants and children in all locations sampled in Kancheepuram district are highly susceptible to non-carcinogenic nitrate and fluoride health hazards.

4.5.3. Tiruvallur district

The $HQ_{nitrate}$, $HQ_{fluoride}$ and THI values for the 38 samplings spots of Tiruvallur district are produced in Table 8. The maximum and mean ± standard deviation values of $HQ_{nitrate}$ are 7.05 and 1.59 ± 1.76 for infants, 4.36 and 0.98 ± 1.09 for children and 2.32 and 0.52 ± 0.58 for adults. The stark difference between the maximum and mean values show that a few spots with very high $HQ_{nitrate}$ are contributing to the relatively high standard deviation. Around 55.26%, 31.58% and 15.79% of the locations in Tiruvallur district were found to exceed the acceptable $HQ_{nitrate}$ levels for infants, children and adults, respectively. The locations with $HQ_{nitrate} > 2.00$ for infants are temple town Thiruverkadu with the location next to a commercial area and the bus station (120.5 mg/L nitrate, $HQ_{nitrate}$ = 7.05, 4.36 and 2.33 for infants, children and adults, respectively), treated lake water from densely populated mixed commercial and residential area of Poonamallee (106.5 mg/L nitrate, $HQ_{nitrate}$ = 6.23, 3.85 and 2.06 for infants, children and adults, respectively), highly industrialized town, Gummidipoondi (105.9 mg/L nitrate, $HQ_{nitrate}$ = 6.20, 3.83 and 2.05 for infants, children and adults, respectively), highway town Janappanchatram with food-outlets and residences next to Kosasthalaiyar river (59.9 mg/L, $HQ_{nitrate}$ = 3.51, 2.17 and 1.16 for infants, children and adults, respectively), groundwater sample of densely populated and mixed commercial and residential are in Poonamallee (57.4 mg/L nitrate, $HQ_{nitrate}$ = 3.36, 2.08 and 1.11 for infants, children and adults, respectively), high population density Arani town (52.9 mg/L nitrate, $HQ_{nitrate}$ = 3.10, 1.91 and 1.02 for infants, children and adults, respectively) temple town, Thiruttani with commercial and residential areas and high visitor traffic (50.1 mg/L nitrate, $HQ_{nitrate}$ = 2.46, 1.52 and 0.81for infants, children and adults, respectively), densely populated Chennai city suburb of Tiruvottiyur which is also close to the sea shore (42.1 mg/L nitrate, $HQ_{nitrate}$ = 2.45, 1.51 and 0.81 for infants, children and adults, respectively), open well water of Tiruvottiyur at VOC Nagar near industries (41.8 mg/L nitrate, $HQ_{nitrate}$ = 2.11, 1.30 and 0.70 for infants, children and adults, respectively) and Tamil Nadu border town of Sathyavedu with proximity to industries (36.0 mg/L nitrate, $HQ_{nitrate}$ = 2.11, 1.30 and 0.70 for infants, children and adults, respectively). The reasons for the high nitrate levels in these locations are most probably evident from the densely population and/or tourist traffic in these areas.

The $HQ_{fluoride}$, maximum, mean ± standard deviation and exceedance values for percentage of locations above acceptable limits are 6.32%, 2.33% ± 1.25% and 94.74% for infants, 3.91%, 1.44% ± 0.77% and 78.95% for children, 2.09%, 0.77% ± 0.41% and 15.79% for adults. The locations with high fluoride content are Tiruvallur (2.7 mg/L fluoride, urban area with lake, $HQ_{fluoride}$ for infants, children and adults = 6.32, 3.91 and 2.09, respectively), Poonamallee (2.3 mg/L fluoride, on road location with commercial and residential areas, $HQ_{fluoride}$ for infants, children and adults = 5.39, 3.33 and 1.78, respectively), Ambattur Ind. E. (2.1 mg/L fluoride, industrial area surrounded by high residential population density, $HQ_{fluoride}$ for infants, children and adults = 4.92, 3.04 and 1.62, respectively), Padianallur (2 mg/L fluoride, north of Chennai city and close to Puzhal (Red Hills) lake, $HQ_{fluoride}$ for infants, children and adults = 4.68, 2.90 and 1.55, respectively), Ambattur Ind. E. (1.6 mg/L fluoride at two locations in industrial area surrounded by high residential population density, $HQ_{fluoride}$ for infants, children and adults = 3.75, 2.32 and 1.24, respectively) and Thaanipoondi (1.6 mg/L fluoride, near the state border with hilly terrain, $HQ_{fluoride}$ for infants, children and adults = 3.75, 2.32 and 1.24, respectively).

The percentage exceedance values for $HQ_{nitrate}$ are 55.26%, 31.58% and 15.79% and $HQ_{fluoride}$ are 94.74%, 78.95% and 15.79% for infants, children and adults, respectively. This data suggests that infants and children in many locations in Tiruvallur district are highly susceptible to nitrate and fluoride in water-based health hazards. This is also reflected in the analysis of the THI data. The THI maximum and mean ± standard deviation values for Tiruvallur district are 9.63 and 3.92 ± 2.06 for infants, 5.95 and 2.42 ± 1.27 for children and 3.18 and 1.29 ± 0.68 for adults. The Tiruvallur district locations with high $HQ_{nitrate}$ and $HQ_{fluoride}$ values also lead to the following top 5 highest THI values: Thiruverkadu (THI = 9.63 for infants, 5.95 for children and 3.18 for adults), treated lake water at Poonamallee (THI = 9.04 for infants, 5.59 for children and 2.98 for adults), groundwater of Poonamallee ((THI = 8.75 for infants, 5.41 for children and 2.89 for adults, Gummidipoondi (THI = 8.07 for infants, 4.99 for children and 2.66 for adults), and Tiruvallur ((THI = 6.39 for infants, 3.95 for children and 2.11 for adults). It is also important to point out that an industrial area such as Ennore presents a relatively low nitrate and fluoride content in water and corresponding THI and this is most probably due to the efficient central effluent treatment plant. The THI percentage exceedance rates are 97.37%, 97.37% and 57.89% for infants, children and adults, respectively which suggest that

almost all the sampling spots in Tiruvallur district (except Ennore) to have a very high non-carcinogenic health risk index for nitrate and fluoride contamination in drinking water, in particular for infants and children.

5. Conclusions

The objective of the investigation was to assess the risk associated with nitrate and fluoride contamination in drinking water resources of three contiguous river basin districts, Chennai, Kancheepuram and Tiruvallur in Tamil Nadu, India. Drinking water from 103 locations in these districts were collected and initially analyzed for their physico-chemical properties including nitrate and fluoride content. The results was used to establish that highly urbanized Chennai district is free from nitrate and fluoride in water pollution. Also, a majority of the areas in Kancheepuram and Tiruvallur districts appear to have nitrate and fluoride concentrations within the standard limits, with a few locations having >100.0 mg/L of nitrate and >2.5 mg/L of fluoride concentrations. The majority of the samples collected exhibited TDS, alkalinity and total hardness levels above the permissible limit. The nitrate and fluoride concentration levels data have been tabulated and a map providing this spatial information has been produced as a result of this work. When compared to nitrate content, fluoride content in drinking water samples expressed higher correlation against the studied water quality parameters. The results also reiterates published literatures that attribute this correlation to the probable leaching of bicarbonate, calcium, magnesium etc. along with the leaching of fluoride from mineral-rich rocks to the drinking water sources. However, it is important to note that these inference were made based on the concentration of contaminants in the drinking water. Hence, this inference is at the most only an indication based on comparison with prescribed permissible limits and not necessarily a scientifically proven methodology to assess the probability of associated health risks. Hence, the U.S. EPA non-carcinogenic health risk assessment methodology for nitrate and fluoride was used to assess the associated health risks for infants, children and adults. The results of this analysis show that the mean THI values for infants, children and adults are 1.83, 1.13 and 0.61, respectively in Chennai, while it is 4.22, 2.61 and 1.39, respectively in Kancheepuram district and 3.92, 2.42 and 1.29, respectively in Tiruvallur districts with almost all of them exceeding THI limit of 1.00. This is also reflected in the high percentage THI limit exceedance values of 95.8%, 58.3% and 8.3% in Chennai district, 100.0% 100.0% and 61.1% in Kancheepuram district and 97.4%, 97.4% and 57.9% in Tiruvallur district, respectively. These results show that infants and children in these districts are overwhelmingly susceptible to non-carcinogenic nitrate and fluoride in drinking water induced health risks. The information can also be used for planning and development of urban and rural areas, agricultural activities etc. As far as the authors know, this is first spatial distribution and health risk assessment study of fluoride and nitrate levels in Chennai and neighboring districts of Tamil Nadu. The results of the study would form the basis for further studies on remediation strategies to mitigate fluoride and nitrate pollution in these districts.

Acknowledgements

This study is conducted with the financial support of Department of Science and Technology-Water Technology Initiative Programme (DST–WTI), New Delhi, India. The reference number of this project sanctioned to the corresponding author is DST/TM/WTI/2K16/238.

The authors also acknowledge the guidance of Dr. R. Sivakumar, Professor, Department of Civil Engineering, SRM IST with the GIS maps.

Conflict of interest

On behalf of all authors, the corresponding authors state that there is no conflict of interest.

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