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S. Selvam^{a,*}, K. Jesuraja^{a,b}, Priyadarsi D. Roy^c, S. Venkatramanan^d, Ramsha Khan^e, Saurabh Shukla ^e, D. Manimaran ^a, P. Muthukumar ^a

^a *Department of Geology, V.O. Chidambaram College, Thoothukudi, 628008. Tamilnadu, India*

^b *Regsitration No: 18212232061030, Affiliated to Manonmaniam Sundaranar University, Tirunelveli, 627 012, Tamil Nadu, India*

^c Instituto de Geología, Universidad Nacional Autónoma de México (UNAM), Ciudad Universitaria, Ciudad de México, CP 04510, Mexico

^d *Department of Disaster Management, Alagappa University, Karaikudi, Tamil Nadu, India*

^e *Faculty of Civil Engineering, Institute of Technology, Shri Ramswaroop Memorial University, Barabanki, UP, 225003, India*

HIGHLIGHTS GRAPHICAL ABSTRACT

- The study measured hazardous heavy metal in water during the COVID-19 lockdown.
- Order of heavy metal enrichments (Zn *>* Fe *>* Cu *>* As *>* Cr *>* Pb *>* Cd) in both phases.
- TCR values also demonstrated "higher risk of cancer" in children and adults.
- The study suggested that the Thamirabarani River system remained hostile to human health.

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ABSTRACT

Variation in levels of toxic heavy metals in river system during the COVID-19 pandemic lockdown might potentially assist in development of a public health risk mitigation system associated with the water consumption. The water quality of Punnakayal estuary in the Thamirabarani River system from the south India, a vital source of water for drinking and domestic purposes, industrial usage, and irrigation was assessed here. A comparitive assessment of physico-chemical variables (pH, EC, TDS, DO, BOD, turbidity and NO3), microbiological parameters (total coliform bacteria, fecal coliform bacteria, fecal streptococci and escherichia coli) and toxic metals (As, Cr, Fe, Cu, Zn, Cd, and Pb) suggested a decrease of 20% in the contaminant ratio during the lockdown period in comparison to the pre-lockdown period. The Health risk assessment models (HQ, HI, and TCR) highlighted carcinogenic and non-carcinogenic hazards for both children and adults through the ingestion and dermal adsorption exposures. The HI values for both As and Cr exceeded the acceptable limit (*>*1) during the lockdown period, but the potential risk for children and adults remained low in compaisio with the pre-lockdown period. Our results suggested that the Thamirabarani River system remained hostile to human health even during the lockdown period, and it requires regular monitoring through a volunteer water quality committee with private and government participations.

* Corresponding author. *E-mail address:* geoselvam10@gmail.com (S. Selvam).

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1. Introduction

The outbreak of corona virus or COVID-19 has led to a complete lockdown in 213 countries ([World Health Organization, 2020](#page-19-0); [Bherwani](#page-18-0) [et al., 2020; Gautam and Trivedi, 2020](#page-18-0); [Sivakumar, 2020\)](#page-19-0). The first case of COVID-19 was reported in China [\(Travaglio et al., 2021;](#page-19-0) [Kachroo,](#page-18-0) [2020;](#page-18-0) [Selvam et al., 2020a; Sinha, 2020\)](#page-19-0) and subsequently, it caused an epidemic of acute respiratory syndrome (SARS-CoV-2) all over the globe ([Gautam and Hens, 2020\)](#page-18-0). Till now, India has a total number of cases of 1,32,05,926 with a mortality rate of 1.28% i.e., 1,68,436 deaths. The Tamil Nadu state in south India has 33,659 active cases with a mortality rate of 1.40% i.e., 12,863 deaths (Source: [https://www.mygov.in/](https://www.mygov.in/covid-19/) [covid-19/](https://www.mygov.in/covid-19/)). This epidemic has also caused irreversible changes to the socio-economic and environmental conditions as the prolonged closure of various industrial edifices had detrimental effects on the economic prosperity. However, the progressive effects on different ecosystems and environment were applauded. For example, [Selvam et al. \(2020a](#page-19-0), India), [Lian et al. \(2020](#page-18-0), China), [Nakada and Urban \(2020,](#page-18-0) Brazil), [Ropkins and](#page-19-0) [Tate \(2021,](#page-19-0) UK) and [Stratoulias and Nuthammachot \(2020](#page-19-0), Thailand) documented better air quality. Similarly, the upgrades in surface water qualities was noticed by [Selvam et al. \(2020b](#page-19-0), India, [Qiu et al. \(2020](#page-19-0), China), and [Kassem and Jaafar \(2020,](#page-18-0) Lebanon). [Dutta et al. \(2020\)](#page-18-0) and [Patel et al. \(2020\)](#page-19-0) documented water quality improvements in the Ganges and Yamuna rivers as a result of the lockdown in India.

The water pollution has been evaluated through estimation of heavy metals ([Selvam et al., 2020a](#page-19-0)). It has two vital sources, i.e. (i) natural (ii) anthropogenic. The natural resources include metals released from rock weathering and their eventual discharge to the water bodies [\(Ravindran](#page-19-0) [and Selvam, 2014](#page-19-0)). The anthropological sources include the emission of heavy metals by emancipation of industrial effluents, fossil fuels/industrial burning, and discharge of sewage into the surface water bodies ([Selvam and Sivasubramanian, 2012;](#page-19-0) [Singaraja et al., 2015;](#page-19-0) [Pan](#page-19-0)[neerselvam et al., 2021](#page-19-0)).

Recent studies have reported that human health risk assessment using Heavy Metal Pollution Index (HMPI), and Heavy Metal Toxicity Load (HMTL) approaches, carcinogenic and non-carcinogenic health risk approach and human health risk index analysis. The chronic exposure to low concentrations of metals like Pb, Cd, Cr might cause brain and kidney damage, and other chronic kidney disease (CKD). Heavy metals like Cd, Pb, Cu, Ni, U, As, and Fe, are the main nephrotoxic heavy metals that can cause tubular damage and glomerulopathies ([Binesh](#page-18-0)[pour et al., 2020](#page-18-0); [Karaouzas et al., 2020; Mukherjee et al., 2020](#page-18-0); [Proshad](#page-19-0) et al., 2020; Tokatli and Ustaoğlu, 2020).

Thamirabarani River system is one of the vital water sources in the state of Tamil Nadu (south India) for drinking, irrigation and middle scale industrial usages (Source: [https://en.wikipedia.org/wiki/Thamira](https://en.wikipedia.org/wiki/Thamirabarani_River) [barani_River\)](https://en.wikipedia.org/wiki/Thamirabarani_River). Past researches in this region have reported heavy metal contamination in this river and related estuaries through the industrial (tannery) effluent, municipal sewage effluent, and urban development ([Selvam et al., 2015](#page-19-0); [Singaraja et al., 2015](#page-19-0); [Muthukumaravel et al.,](#page-18-0) [2021\)](#page-18-0). Therefore, the general assumption was that the reduced activities of above sources during the lockdown would have improved the surface water quality of the Thamirabarani channel. However, there has been no study to assess the effects of COVID-19 lockdown. Thus, the purpose of this study was to (i) estimate hydro-chemical variances between the lockdown and pre-lockdown periods, (ii) assess the pollution vulnerability of industrial and anthropological demeanor on surface water quality, (iii) measure possible health hazards to children and adults from domestic, irrigational and livestock usages of surface water during the lockdown and their differences with the pre-lockdown period, (iv) identify the pollution sources and estimate the reduction percentage of toxic heavy metals, and (v) provide the community with suggestions and solutions to protect the water system through eco-friendly environmental activities.

Fig. 1. Location map of the study area along with sampling points.

2. Study area

The Thamirabarani River basin is located in the coastal district of Thoothukudi, Southern Tamil Nadu, India. This district was detached from the adjacent Tirunelveli district in 1986 on the basis of primary augmentation of industrial efficiencies and relevant coastal economic evolutions ([Selvam et al., 2013](#page-19-0)). Geographically, it extends between N 8.5838889–8.6194444 latitude and E 77.9225–78.1297222 longitude and includes 223.32 sq.km of the total delta basin [\(Fig. 1\)](#page-2-0). The Thamirabarani River originates from the Pothigai hills of the Western Ghats and flows through the districts of Thoothukudi and Tirunelveli before joining the Bay of Bengal. It provides water for irrigation and electricity production [\(Arisekar et al., 2020](#page-18-0)). The primary crops include paddy, banana, ground nut, brinjal, ragi, sorghum, coconut, pulses, ginger, tea and rubber in upper reaches of the river course [\(Arisekar et al., 2018](#page-18-0)).

The delta region is underlined by the Archaean gneisses followed by granites and charnockites ([Jesuraja et al., 2021\)](#page-18-0). Alluvium (Quaternary), Dune Terri sands and Tertiary sediments outcrop in the delta region and the marine and fluvial-marine deposits are present along the coast ([Narayanaswamy and Lakshmi, 1967;](#page-19-0) [Magesh et al., 2016](#page-18-0); [Satheeskumar et al., 2020\)](#page-19-0). The primary land use encompasses agricultural lands, barren lands and salt pans other than emigrations and aquifers ([Satheeskumar et al., 2020\)](#page-19-0). Both the groundwater and surface water are vulnerable to anthropogenic activities related to fishing and tourism [\(Selvam et al., 2021\)](#page-19-0), both of which generated annual revenue of 8.9 crores in 2017–2018 (source: [http://www.townpanchayat.in/ti](http://www.townpanchayat.in/tiruchendur) [ruchendur](http://www.townpanchayat.in/tiruchendur)). All chemical plants are pertinent to salt industry, petrochemicals and plastics industry [\(Jesuraja et al., 2021](#page-18-0)). Small scale industries belong to paper, soft drink manufacturing, textile, PVC pipe manufacturing, and soap manufacturing.

3. Sampling and analysis

A total of 20 water samples were collected uniformly from the Punnakayal estuary of Thamirabarani River basin, and subsequently located in [Fig. 1](#page-2-0) using GPS (HANNA 2130). These samples were collected in the pre lockdown period (28–29 January 2020), and during the COVID-19 lockdown (6–7 May 2020). The water samples were collected from 10 cm below the water surface and stored in 2 L polyethylene containers. As per the guidelines of American Public Health Association (APHA), we used ultrapure $HNO₃$ for on-site acidification (pH *<* 2) to avoid microbial activity and adsorption/precipitation on the bottle walls [\(APHA, 1995\)](#page-18-0). Different physical parameters such as pH, total dissolved solids (TDS), electrical conductivity (EC), turbidity, and dissolved oxygen (DO) were measured using deluxe water and soil analysis kit (model no: 191).

In laboratory, the UV–visible spectrophotometer (Systronic), quantified $NO₃$ and the AAS (Atomic Absorption Spectroscopy Perkin Elmer, Elan Drce) measured the absorptions of metals (Cr, Cu, Fe, Cd, Pb and Zn) and metalloids (As) by using The NIST (National Institute of Standards and Technology) standard (1640a) for the QA/QC (Quality Assurance and Quality Control) resolution."Cetripur" was used for the multi-element (Merck) calibration. The MPN (Most Probable Number) method ([ISO, 2000](#page-18-0)) and estimation of total coliform, fecal coliform, Escherichia coli, and fecal streptococci determined the microbial quality. The MPN method also estimated the number of coliforms of lactose enzymes that produce gas per 100 ml of water sample.

4. Computations of metal pollution codes

4.1. Heavy metal pollution index (HMPI)

HMPI comprehensively assessed the influence of each dissolved heavy metal on overall surface water quality ([Mohan et al., 1996](#page-18-0); [Vetrimurugan et al., 2016;](#page-19-0) [Jorfi et al., 2017](#page-18-0); [Wagh et al., 2018;](#page-19-0) [Rezaei](#page-19-0) [et al., 2019;](#page-19-0) [Karaouzas et al., 2020](#page-18-0)). It was computed using the formula:

Table 1

$$
HMPI = \frac{\sum_{i=1}^{n} W_i Q_i}{\sum_{i=1}^{n} W_i}
$$
\n(1)

Where n and *Wi* refer to number of heavy metals and unit weight of the ith heavy metal, respectively.

In step 1, the sub-index (Q_i) of ith heavy metal was computed using equation (2):

$$
Q_i = \sum_{i=1}^{n} \frac{|M_{con}(-) I_i|}{S_i - I_i} \times 100
$$
 (2)

Where M_{con} (μ g/L) refers the computed value of ith heavy metal. S_i is the standard permissible of ith metal for drinking purpose [\(World Health](#page-19-0) [Organization, 2017](#page-19-0)) for the heavy metals (μg/L) and *Ii* refers to the ideal limits of ith heavy metal.

In step 2, the unit weight (W_i) of each metal was assessed using the equation (3):

$$
W_i = \frac{k}{S_i} \tag{3}
$$

Where k refers to proportionality constant and it is considered as 1 for all the metals ([Wanda et al., 2012; Qu et al., 2018](#page-19-0)).

Finally, HMPI was computed using the Eq. (1) and it classified the heavy metal pollution in surface water bodies into three categories such as low contamination (HMPI*<*15), medium contamination (HMPI: 15–30) and high contamination (HMPI*>*30, [Edet and Offiong, 2002](#page-18-0); [Wanda et al., 2012; Qu et al., 2018](#page-19-0); [Zakir et al., 2020](#page-19-0)).

4.2. Human health risk index analysis

Consumption of drinking water contaminated with toxic metals increases the risk of non-carcinogenic and carcinogenic diseases in humans [\(Bineshpour et al., 2020](#page-18-0): [Qu et al., 2018\)](#page-19-0). We utilized the methods specified by U.S. Environmental Production Agency (USEPA) to appraise the non-carcinogenic (As, Cr, Fe, Cu, Zn, Cd, and Pb) and carcinogenic risks from the dissolved metals (As, Cr, Cd, and Pb) by *S. Selvam et al.*

following [USEPA \(2013\)](#page-19-0).

4.2.1. Non-carcinogenic health risk approach

[USEPA \(2004\)](#page-19-0) initiated a health risk technique for measuring the non-cancerous human health risks from the heavy metal elements in groundwater and surface water through ingestion, inhalation and exposure to skin. Primarily, the risk was caused by direct water intake and absorption or skin contact [\(Saha et al., 2017;](#page-19-0) [Qu et al., 2018](#page-19-0); [Mukherjee et al., 2020](#page-18-0); [Saleem et al., 2019; USEPA, 2020](#page-19-0)). It computes the pollutant dose consumed in human using chronic daily intake (CDI), which reflects the dose of pollutants in kilogram per day captivated through the digestion pathway (CDI_{ingestion})and dermal absorption (CDI_{dermal}) using Eqs. (4) and (5), respectively [\(USEPA, 2011](#page-19-0); Zhang [et al., 2017;](#page-19-0) [Jehan et al., 2020;](#page-18-0) Tokatli and Ustaoğlu, 2020).

$$
CDI_{\text{integration}} = Con_{\text{water}} \times \frac{(IR \times EF \times ED)}{(BW \times AT)} \tag{4}
$$

$$
CDI_{dermal} = Con_{water} \times \frac{(SA \times Kp \times ET \times EF \times ED \times CF)}{(BW \times AT)} \tag{5}
$$

where, *Conwater* refers to trace metal concentration in surface water (μg/ L). [Table 1](#page-3-0) reveals the non-carcinogenic health impact parameters and their input assumptions used for estimating the exposure to heavy metals through intake and skin absorption.

In second step, we calculated the Hazard quotient (HQ) from CDI (CDI_{ingestion} and CDI_{dermal}) and RfD (RfD _{ingestion} and RfD _{dermal}) using Eq. (6) [\(Saha and Paul, 2019](#page-19-0); [Imran et al., 2019\)](#page-18-0);

$$
HQ_{integration} / HQ_{dermal} = \frac{CDI_{ingestion} / CDI_{dermal}}{RfD_{ingestion} / RfD_{dermal}}
$$
 (6)

At the final step, the total potential non-carcinogenic risks were appraised by estimating the hazard index (HI) by using Eq. (7) ([Rupa](#page-19-0)[kheti et al., 2017](#page-19-0); [Jehan et al., 2020](#page-18-0); [Karthikeyan et al., 2021](#page-18-0));

$$
HI = HQ_{ingestion} + HQ_{demal} = \frac{CDI}{Rfd}
$$
 (7)

The toxic metals with HI and HQ of *>*1 can have adverse effects and with *<*1 have no adverse effects on human health [\(USEPA, 1989](#page-19-0); [Vetrimurugan et al., 2016](#page-19-0), [2017](#page-19-0); [Yang et al., 2017](#page-19-0); [Mohammadi et al.,](#page-18-0) [2019\)](#page-18-0).

4.2.2. Carcinogenic health risk approach

We considered the heavy metals as carcinogenic to humans (As) and likely to be carcinogenic to humans (Cd) in order to assess the carcinogenic and non-carcinogenic risks according to the IARC report [\(IARC,](#page-18-0) [2013\)](#page-18-0). Both Pb and Cr were included in the non-carcinogenic risk assessment. Even though, Cu, Fe and Zn are not classified in the IARC report, they were involved in the non-carcinogenic risk assessment (e.g. [Chan et al., 1998](#page-18-0)).

The carcinogenic risks (CR) were evaluated using the following equations (Eqs. (8) and (9)) ([Benhaddya, 2020\)](#page-18-0):

$$
CR = CDI \times CSF \tag{8}
$$

$$
TCR = CR_{\text{integration}} + CR_{\text{dermal}} \tag{9}
$$

The standard assumption values of cancer slope factor (CSF) to measure the risks are 0.0005, 0.0015, 0.0061 and 0.0000085 ppb/day for Cr, As, Cd and Pb, respectively ([Gao et al., 2019;](#page-18-0) [Tokatli and](#page-19-0) Ustaoğlu, 2020). The acceptable or tolerable carcinogenic risk range is 0.000001–0.0001. If CR or TCR of an element exceeds 0.0001, the effect might be detrimental on human health [\(Qu et al., 2018;](#page-19-0) [Mohammadi](#page-18-0) [et al., 2019; Gao et al., 2019;](#page-18-0) [Tabassum et al., 2019](#page-19-0)).

Fig. 2. Spatial map for TDS distribution during Pre-lockdown and lockdown.

4.3. Heavy metal toxicity load approach

HMPI or HI (HMPI*>*100 or HI *>* 1) indicate the suitability of water as it reveals the accurate ratio of surplus metal ([Saha and Paul, 2019](#page-19-0); [Proshad et al., 2020\)](#page-19-0). It concedes many ideas for predicting and mitigating the pollution of water bodies. This estimation technique predicts the concentration of excess metal in water and the amount that must be removed to make it harmless for human use. HMTL (Heavy metal toxic load) estimates toxic heavy metal in a water source that seduces human health, and it was computed using the following equation [\(8\)](#page-4-0) ([Saha and](#page-19-0) [Paul, 2019\)](#page-19-0):

 $HMTL = \sum_{i=1}^{n} c.HIS$

where c, n and HIS denote the concentration of heavy metal, number of heavy metals and risk severity score, respectively. HIS scores for As (1676), Cr (1149), Cu (805), Zn (913), Cd (1318), and Pb (1531) were considered from [ATSDR \(2018\).](#page-18-0) They are based on the frequency of hazardous material occurrence on National Priority List (NPL), prepared by ATSDR. The permissible limit of HMTL was below 5888.527 mg/l ([Saha and Paul, 2019](#page-19-0); [Proshad et al., 2020](#page-19-0)). The HIS score was multiplied by acceptable limit of specific concentration, which is considered the permissible limit for toxicity load and the permissible toxicity load is given in [Table 9.](#page-17-0) HTML Result identifies that the surplus percentage of removal of heavy metals from surface aquifers beyond the permissible toxicity load is essential for human health.

Fig. 3. Interpretation of Nitrate risk during Pre-lockdown and lockdown.

5. Results and discussion

We assessed water quality of the Punnakayal estuary during the lockdown and pre-lockdown periods using the physic-chemical variables, dissolved metals and microbiological parameters.

5.1. Physicochemical parameters

Mean pH of the pre-lockdown (7.78), and lockdown (7.63) periods did not show significant difference ([Table 2](#page-4-0)). Higher pH of TSW - 11, 12, 20 during the lockdown period was due to warmer water temperatures. Both the mean EC and TDS of the lockdown period were 22.04% and 29.96% lower than the pre-lockdown. TDS of 75% of surface water samples in the pre-lockdown and 50% of samples of lockdown interval exceeded the permissible limit of WHO (2017; 1000 mg/l) [\(Fig. 2](#page-5-0)).

These changes are due to absence of agricultural activities during the first phase of lockdown as well as the dilution effect from rainfall. It also revealed the reduction in water consumption for the industrial purposes. Compared to the [Central Pollution Control Board \(CPCB\) \(1979\)](#page-18-0) and [BIS](#page-18-0) [\(1982\)](#page-18-0) water quality standards, about 45% of both the pre-lockdown (1.26–9.26 mg/l) and lockdown (1.11–6.89 mg/l) samples had DO in class C, revealing unsuitability for drinking. The mean BOD in both periods (5.4 mg/l in pre-lockdown and 4.8 mg/l in lockdown) showed no significance difference and it grouped only few samples in class A, representing the suitability for drinking without conventional treatment but after disinfection (Source: [Central Pollution Control Board \(CPCB\),](#page-18-0) [1979](#page-18-0) and [BIS, 1982](#page-18-0)). Turbidity limits before and during the lockdown are 1.56–9.65 (NTU) and 1.21–6.62 (NTU), respectively. Most samples from both periods exceeded the acceptable limit of [BIS \(2012\).](#page-18-0) Higher turbidity loads also revealed natural causes such as erosion of more silt

Fig. 4. Heavy metal occurrence difference on bar chart during Pre-lockdown and lockdown.

Fig. 5. Spatial variation map for chromium distribution during Pre-lockdown and lockdown period.

Fig. 6. Spatial variation map of iron distribution during Pre-lockdown and lockdown period.

and mud and anthropogenic causes such as agriculture, sand mining, construction, and algae from domestic wastes. NO_3 ⁻ is a prime contaminant in agro-terrains ([Adimalla et al., 2018;](#page-18-0) [Zhang et al., 2018](#page-19-0); [Chandrasekar er al., 2021\)](#page-18-0). It was quantified as 35–72 mg/l during the pre-lockdown and 12–56 mg/l during the lockdown. Comparison with [World Health Organization \(2017\)](#page-19-0) suggests 50% of pre-lockdown and only 15% of lockdown samples were under high risk. We detected 27% discrepancy between the two periods and plenty of samples of the pre-lockdown period showed high health risk (e.g.[Adimalla and Qian,](#page-18-0) [2019,](#page-18-0) [Fig. 3](#page-6-0)).

5.2. Metal concentrations

Concentrations of chromium (Cr), copper (Cu) and zinc (Zn) showed *<*20% difference between the lockdown and pre-lockdown periods ([Table 2](#page-4-0) and [Fig. 4\)](#page-7-0). However, the concentrations of arsenic (As), iron (Fe), lead (Pb) and cadmium (Cd) decreased *>*20% during the lockdown in comparison to the pre-lockdown period.

Arsenic: It is one of the most dangerous toxic components, and can lead to immune disorders, reproductive dysfunction and skin cancer ([Kabata-Pendias and Szteke, 2015](#page-18-0); [Kacmaz, 2020;](#page-18-0) Tokatli and Ustaoğlu, [2020\)](#page-19-0). Concentration of 0.012–0.099 mg/l in pre-lockdown and 0.001–0.098 mg/l during the lockdown suggested that all of them exceeded the permissible limit ([World Health Organization, 2017\)](#page-19-0) for drinking (0.01 mg/l). Despite the COVID-19 lockdown, the anthropological activities related to poultry waste, fertilizer plants, brick making plants, pot design making plants and agricultural practices continued in this region ([Selvam et al., 2014a,](#page-19-0) [2017](#page-19-0))

Chromium: The maximum Cr concentration was 0.08 mg/l in prelockdown samples and reduced to 0.07 mg/l in the lockdown samples.

Fig. 7. Spatial variation map of lead distribution during Pre-lockdown and lockdown period.

About 45% of the pre-lockdown samples exceeded the permissible limit (0.05 mg/l) and only 15% of the lockdown samples exceeded the permissible limit ([Fig. 5\)](#page-7-0). In both the phases, all samples near the coast exceeded the allowable limit of 0.05 mg/l ([World Health Organization,](#page-19-0) [2017\)](#page-19-0). Reduction in chromium concentration during the lockdown indicated decline in the activities related to the chemical industries (e.g. DCW Industry) [\(Selvam et al., 2017](#page-19-0)). Similarly, there was less utilization of petroleum product in heavy vehicle workshops of this region (e.g. [Hua](#page-18-0) [et al., 2016;](#page-18-0) [Sakthivel et al., 2016\)](#page-19-0).

Iron: For local aquifers the main contributors of iron (Fe) are industrial effluent, acid-mine drainage, and sewage. In this research the iron (Fe) concentration is varied from 0.123 to 0.458 mg/l pre lockdown and 0.102–0.321 mg/l during the lockdown [\(Fig. 6\)](#page-8-0). This result shows that 55% of pre-lockdown samples have an acceptable limit of 0.3 mg/l ([World Health Organization, 2017](#page-19-0)), while 5% of samples exceed the allowable limit during the lockdown period. The maximum occurred value of Fe on COVID-19 phases is very close to permissible limit (0.3 mg/l) and shows that this may be due to the shutting of metallurgical industries during the COVID-19 lockdown period, from which wastes are discharged into water bodies and from landfills [\(Milivojevi](#page-18-0)ć et al., [2016\)](#page-18-0).

Copper: This trace element is an important nutrient for the human body [\(Muhammad et al., 2014](#page-18-0); [Samantara et al., 2017\)](#page-19-0). We detected almost no change between the two COVID-19 stages with mean values of 0.218 mg/l during pre-lockdown and 0.201 mg/l during the lockdown. None of them exceeded the drinking limits (2 mg/l) of the [World Health](#page-19-0) [Organization \(2017\)](#page-19-0) standard.

Zinc: Absence of Zn affects the metabolism and the immune system, resulting in infections in humans, anemia and birth defects in pregnant women and delayed sexual maturity in men ([ATSDR, 2005;](#page-18-0) [Samantara](#page-19-0)

Fig. 8. Statistical significance of biological parameters (MPN ml/l) in groundwater samples before and during the lockdown period related to COVID-19.

[et al., 2017](#page-19-0); [Karunanidhi et al., 2021](#page-18-0)). Concentration of Zn in pre-lockdown samples (0.568–0.999 mg/l) and lockdown period (0.523–0.999 mg/l) remained similar. All of them remained suitable for drinking (*<*5 mg/l). It showed the absence of potential sources of Zn such as industries related to rubber, paint, bronze, die-casting metals, brass and other alloys in this region.

Cadmium: It ranged from 0 to 0.002 mg/l in pre-lockdown samples and 0–0.001 mg/l in the lockdown samples. Both remained within the permissible limits ([World Health Organization, 2017](#page-19-0); *<*0.003 mg/l). The two periods (pre-lockdown and lockdown) showed a significant difference of up to 42%. Cadmium can flow from phosphate fertilizers into the soil and surface water, and it is also sourced from cadmium-based batteries and cadmium coated materials [\(ATSDR, 2008](#page-18-0); Tokatli and Ustaoğlu, 2020). Since there is no homeostatic mechanism to control, the exposure to very low levels of Cd can cause adverse overall effects on humans ([Carter and Fernando, 1979](#page-18-0)).

Lead: It varied from 0.006 to 0.019 mg/l with an average of 0.012 mg/l in the pre-lockdown and between 0.004 and 0.009 mg/l with an average of 0.007 mg/l during the lockdown. About 65% of the surface

water samples of the pre-lockdown interval exceeded the recommended limit (0.01 mg/l), but all the lockdown samples were within the allowable range for drinking [\(Fig. 7](#page-9-0)). Industrial discharges from foundries, battery production amenities, contaminated land runoffs and sewage are the main bases of Pb in the Thoothukudi coastal region. ([Selvam et al., 2017](#page-19-0)). Deficiency or minimal disposal of effluents from these industries and manufacturing unit and reduction in petroleum related transportation during the lockdown interval might have led to lower Pb concentration.

5.3. Microbiological parameters

The expressive statistics of microbiological concentration are presented in [Table 2.](#page-4-0) Total coliform bacteria population varied between 6.2 and 189 MPN ml/l and 1.2–95 MPN ml/l in the lockdown and prelockdown samples, respectively. The maximum population of fecal coliform bacteria was 42.92 MPN ml/l in the lockdown and 97.09 MPN ml/l in the pre-lockdown period. Escherichia coli bacterial population ranged from 0 to 62 MPN ml/l in the lockdown samples and 0 to 77 MPN ml/l in the pre lockdown samples. The reduction in bacteria during the lockdown period was due to closure of fishing companies and tourism contamination (Fig. 8). However, all fecal streptococci population was low (Est *<*10) both in the pre-lockdown and lockdown samples. Escherichia coli Total coliforms, and faecal coliforms are contributed to the water system by humans and other warm-blooded animals. They survive the sewage treatment plants in large numbers and protect their pathogens for a longer period ([Selvam et al., 2017](#page-19-0)). [Selvam et al.](#page-19-0) [\(2020a\)](#page-19-0) have observed that the closure of industries and other commercial activities in the study area provide favorable condition for the growth of large bacterial population.

5.4. Pollution Indices

Based on the method used by [Edet and Offiong \(2002\)](#page-18-0), we evaluated the HMPI (heavy metal pollution index) for the pre-lockdown and lockdown phases for As, Cr, Fe, Cu, Zn, Cd, and Pb (Table 3). The computed HPI values varied between 15.18 to 81.25, and 10.67 to 46.92 with an average values of 51.81 and 31.61 in the pre-lockdown samples and lockdown samples, respectively. As per the classifications, about

Table 3

Heavy metal pollution index (HMPI) and decreased percentage of studied metal in Thamirabarani River for Pre-lockdown and Lockdown phase.

Sampling Point	Pre-lockdown phase		Lockdown phase		Decreased % (Pre lockdown -
	Pre lockdown	Degree of pollution as per HMPI scale	Lockdown	Degree of pollution as per HMPI scale	Lockdown)
TSW ₁	48.23	High Pollution	35.97	High Pollution	25.41
TSW ₂	76.41	High Pollution	39.72	High Pollution	48.02
TSW ₃	25.60	Medium Pollution	19.62	Medium Pollution	23.36
TSW ₄	34.97	High Pollution	30.03	Medium Pollution	14.14
TSW ₅	81.25	High Pollution	39.62	High Pollution	51.23
TSW ₆	38.65	High Pollution	30.08	Medium Pollution	22.17
TSW ₇	15.18	Medium Pollution	10.67	Low Pollution	29.68
TSW ₈	46.75	High Pollution	11.47	Low Pollution	75.46
TSW ₉	48.04	High Pollution	12.25	Low Pollution	74.49
TSW 10	46.36	High Pollution	14.73	Low Pollution	68.23
TSW 11	78.95	High Pollution	37.42	High Pollution	52.60
TSW 12	81.25	High Pollution	39.62	High Pollution	51.23
TSW 13	69.68	High Pollution	43.38	High Pollution	37.74
TSW 14	43.56	High Pollution	39.49	High Pollution	9.35
TSW 15	29.79	Medium Pollution	22.12	Medium Pollution	25.77
TSW 16	59.43	High Pollution	34.55	High Pollution	41.86
TSW 17	50.72	High Pollution	44.07	High Pollution	13.10
TSW 18	51.87	High Pollution	46.92	High Pollution	9.55
TSW 19	66.81	High Pollution	42.67	High Pollution	36.14
TSW 20	51.86	High Pollution	43.40	High Pollution	16.32
Min	15.18	Low Pollution (0%)	10.67	Low Pollution (20%)	9.35
Max	81.25	Medium Pollution (15%)	46.92	Medium Pollution (20%)	75.46
Mean	51.81	High Pollution (85%)	31.61	High Pollution (60%)	36.85

Fig. 9. Result of HMPI values display on spatial map for Pre-lockdown and lockdown period.

15% and 85% of the pre-lockdown samples were categorized as medium and high pollution, respectively. Similarly, about 20%, 20% and 60% of the lockdown samples were grouped in low pollution, medium pollution and high pollution groups, respectively. Among all the heavy metals, Cd and Pb played important roles in adjusting the HMPI. Results showed no significant depravity between the two COVID-19 phases except for the sampling stations at the central part of the study region (TSW 7 - (15.18–10.67), TSW 8 - (46.75–11.47), TSW 9 - (48.04–12.25), and TSW 10 - (46.36–14.73). The HMPI values of the central part were transferred from the high pollution to low pollution class and there was no change in the rest as they were continued to receive sewage and municipal effluents as well as pollutant from agricultural activities (i.e. Phosphate fertilizers) (Fig. 9).

5.5. Health risk assessment

The hazard indices (HI) of non-carcinogenic risk and carcinogenic risk were based on hazard quotients (HQ) of the ingestion and dermal adsorption pathways. They showed the total potential human health risks on children and adults from various heavy metals.

5.5.1. Non-carcinogenic health risk

The non-carcinogenic risk for children and adults was evaluated for the toxic As, Cr, Fe, Cu, Zn, Cd and Pb [\(Tables 4 and 5](#page-12-0)). In the prelockdown period for children, the HQ ingestion ranged from 1.280–10.560, 0.107–0.853, 0.013–0.049, 0.098–0.239, 0.061–0.107, 0–0.128 and 0.137–0.434 for As, Cr, Fe, Cu, Zn, Cd and Pb, respectively. Similarly, the dermal pathway HQ ranged from 0.032–0.266, 0.440–3.520, 0.001–0.003, 0.003–0.008, 0.003–0.005, 0.000–1.320

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Table 8

and 0.001–0.004 for As, Cr, Fe, Cu, Zn, Cd and Pb, respectively ([Table 4](#page-12-0)). For children, the HQ ingestion of As exceeded the limit (*>*1) in all samples and HQ dermal result showed values beyond the hazard quotient limit in 75% samples for Cr and 35% samples for Cd. During lockdown period and for children, the HQ ingestion values ranged from 0.107–10.453, 0.107–0.747, 0.011–0.034, 0.089–0.236, 0.056–0.107, 0.000–0.064 and 0.091–0.206 for As, Cr, Fe, Cu, Zn, Cd and Pb, respectively. The dermal pathway HQ ranged from 0.003–0.263, 0.440–3.080, 0.001–0.002, 0.003–0.008, 0.003–0.005, 0.000–0.660 and 0.001–0.002 for As, Cr, Fe, Cu, Zn, Cd and Pb, respectively. In the lockdown period, the HQ ingestion pathway of As also exceeded the limit (*>*1) in 100% of the samples and HQ dermal results of Cr remained above the hazard quotient limit in 60% of the surface water samples.

In the pre-lockdown period for adults, the HQ ingestion ranged between 1.143–9.429, 0.095–0.762, 0.012–0.044, 0.088–0.214, 0.054–0.095, 0.000–0.114 and 0.122–0.388 for As, Cr, Fe, Cu, Zn, Cd and Pb, respectively. The HQ through dermal pathway varied between 0.015–0.124, 0.206–1.646, 0.000–0.002, 0.002–0.004, 0.001–0.003, 0.000–0.617 and 0.002–0.007 for As, Cr, Fe, Cu, Zn, Cd and Pb, respectively [\(Table 5](#page-13-0)). For the adult, the HQ ingestion of As exceeded the limit (*>*1) in 100% samples and HQ dermal of chromium remained above the hazard quotient limit in 50% of the samples. During the lockdown period, for adults, the HQ values via intake pathway were 0.095–9.333, 0.095–0.667, 0.010–0.031, 0.079–0.211, 0.050–0.095, 0.000–0.057 and 0.082–0.184 for As, Cr, Fe, Cu, Zn, Cd and Pb, respectively. The HQ dermal pathway were 0.001–0.123, 0.206–1.440, 0.000–0.001, 0.001–0.004, 0.001–0.003, 0.000–0.309 and 0.001–0.003 for As, Cr, Fe, Cu, Zn, Cd and Pb, respectively. During the lockdown, As in about 95% samples and Cr in 40% samples exceeded the hazard quotient limits (*>*1) of ingestion and dermal contact pathways.

The pre-lockdown and lockdown HI indices (HI; sum of all HQ values by intake or skin absorption pathway) for children and adults were higher for the water intake pathway compared to the skin contact pathway ([Table 6](#page-14-0)). Two-term HI values for ingestion and skin absorption exposure to As and Cr were higher than the acceptable range of noncarcinogenic metals. However, the pollution impact or pollution rate of lockdown period was relatively lower than the pre-lockdown. For example, the average HI intake for children was 6.616 for As and 2.485 for Cr during the pre-lockdown. They decreased to 5.149 (As) and 1.988 (Cr) during the lockdown. The closure of several industries in this region, limited use of petrochemicals in agriculture and reduction of other

anthropological contributions such as discharge of domestic wastewater, municipal waste, and chemical waste from industries during the lockdown might have led to less heavy metal contribution and recued health risks.

5.5.2. Carcinogenic health risk

The carcinogenic risk (CR) was computed for As, Cr, Cd and Pb and [Table 7](#page-15-0) presents the total carcinogenic risk (TCR; sum of CR from ingestion and dermal contact exposure) for children and adults. In the pre-lockdown samples and for children, the TCR varied between 0.0005819–0.0048010, 0.0001633–0.0013064, 0–0.0003944 and 0.0000017–0.0000052 for As, Cr, Cd and Pb, respectively. Similarly for adults, the TCR values in the pre-lockdown samples varied between 0.0005171–0.0042658, 0.0001462–0.0011693, 0–0.0003526 and 0.00000155–0.0000047 for As, Cr, Cd and Pb, respectively. In carcinogenic elements the following elements are As, Cr and Cd (*>*95% samples) beyond the permissible carcinogenic limit. Only Pb (100%) remained within the acceptable or tolerable carcinogenic limit (0.000001–0.0001) for children and adults.

In the lockdown samples, the TCR for children varied between 0.0000485–0.0047525, 0.0016033–0.0112231, 0–0.0001972 and 0.0000011–0.0000025 for As, Cr, Cd and Pb, respectively. The TCR values for adults varied between 0.0000431–0.0042227, 0.0001444–0.0010108, 0–0.0001752 and 0.0000009–0.0000022 for As, Cr, Cd and Pb, respectively. Again all the toxic metals (As, Cr and Cd), except for Pb exceeded the carcinogenic range (0.000001–0.0001) causing risk to children and adults. In this study did not show any significant improvements in surface water pollution but compare to prelockdown period, during lockdown period the pollution level is much reduced.

5.5.3. Heavy metal toxicity load

We computed HMTL to evaluate the concentration of pollutants that might cause non-carcinogenic health risk and furnished the data about the percentage of metals that needs removal from the specific samples (Table 8). The ranges of As, Cr, Cu, Zn, Cd, and Pb that are the most threatening to human health were selected from the ATSDR material priority list to calculate HTML [\(ATSDR, 2017](#page-18-0)). It varied between 773.70 and 1382.30 mg/l with an average of 1101.78 mg/l for the pre-lockdown samples and between 665.63 and 1311.45 mg/l with a mean of 990.04 mg/l for the lockdown samples. DCW industrial waste

Table 9

^a Denotes that those samples are within permissible toxicity load.

leaks and increased usages of agro-based petrochemicals in the study area may lead to increased HTML results. It is necessary to remove 90%, 38%, 47% of As, Cr, and Pb in the pre-lockdown samples. Similarly, almost similar amount of As (90%) and slightly less Cr (29%) must be removed from the lockdown samples to make it suitable for human health. However, Pb remained below the permissible toxicity load in all the lockdown samples. In both the pre-lockdown and lockdown periods, the concentrations of Cu, Zn and Cd were suitable for human activities (Table 9).

6. Remediation for human welfare

Our results suggested that the surface water of Punnakayal estuary in the Thamirabarani River system poses severe non-carcinogenic and carcinogenic hazards to human health. The pollution sources such as industrial effluents, domestic wastewater, sand mining and agroventures can be minimized by proper management of the surrounding petrochemical, and beverages manufacture units as well as improving the wastewater drainage system. The lockdown period bestowed a good opportunity to understand the paramount importance of nature in our daily lives. Furthermore, it also provided an insight to realize that the conservation and sustainability of natural water systems can be inhibited by effectively managing the pollution sources. According to HTML results, As, Cr and Pb required greater attention in the pre-lockdown samples, whereas only As and Cr were peril to human in the lockdown samples. Therefore, the implementation of pertinent strategy technique of water quality management might help to minimize the pollution of water bodies. In environmental studies various researchers were proposed various innovative solutions for arsenic and chromium remediation, especially [Marinho et al. \(2019\)](#page-18-0) was discussed various aqueous solutions and the analytical methods used for their detection and quantification of arsenic and chromium elements. This study advocates to form a special panel to routinely monitor the surface water quality and mitigate the risk from exposure to potential heavy metals, especially from As and Cr in the Thamirabarani River ecosystem.

7. Conclusions

The present study evaluated the influence of COVID-19 pandemic lockdown on surface water quality of the Punnakayal estuary in the

Thamirabarani River system of south India by estimating reduced absorptions of As, Cr, Cu, Cd, Fe, Pb and Zn. Toxic heavy element contamination risk assessment codes, health risk assessment methods and some pollution load approach described the water quality prior to lockdown and the lockdown periods. We did not observe any changes in the order of heavy metal enrichments (Zn *>* Fe *>* Cu *>* As *>* Cr *>* Pb *>* Cd) in both phases as the industrial ejects, domestic sewage and agricultural applications continued during the lockdown period. However, the quantity or impurity ratio was reduced compared to the prelockdown period. In the pre-lockdown surface water, the concentrations of Cu, Zn and Cd remained within permissible limits of [World](#page-19-0) [Health Organization \(2017\)](#page-19-0) in all samples and hazardous As, Cr, Fe and Pb exceeded the permissible limits in 100%, 40%, 45% and 65% samples, respectively. During the lockdown period, As remained similar with 100% samples exceeding the permissible limit, but relatively less samples had Cr (15%) and Fe (5%) above the permissible limits. HQ non-carcinogenic risk on children and adults from the ingestion and skin absorption of hazardous Fe, Cu, Zn, Cd and Pb were within the reliable range for both the periods. HI results, however, indicated more delicate to non-cancer risks in children compared to adults from both As and Cr. TCR values also demonstrated "higher risk of cancer" in children and adults from As, Cr, and Cd, even during the lockdown and "no carcinogenic dilemma" from Pb.

Credit author statement

S. Selvam: Writing – original draft, Writing – review & editing, Visualization, Supervision, Project administration, Methodology, Funding acquisition. **K.Jesuraja**: Writing – original draft, Data curation, Resources. **Priyadarsi D. Roy**: Writing – review & editing, Investigation, Project administration. **S. Venkatramanan**: Data curation, Resources, Software, Visualization. **Ramsha Khan**- Software, Resources, Formal analysis. **Saurabh Shukla**: Software, Resources, Formal analysis. **D. Manimaran**: Software, Resources, Formal analysis. **P. Muthukumar**: Software, Resources, Formal analysis.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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