Environmental Pollution 294 (2022) 118553

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Potential health risk caused by heavy metal associated with seafood consumption around coastal area☆

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A R T I C L E I N F O

Keywords:
Heavy metals
Seafood
Target hazard quotient
Hazard index
And health risk

A B S T R A C T

The current study investigated seasonal fluctuations in diversity of fish and heavy metal concentrations in coastal areas, as well as the possible human health risks associated by the heavy metals (Mercury, Lead, Chromium, Cadmium, Copper and Zinc). From five different locations across the coastal area, 44 finfish species from 11 orders and 33 families were collected. Four finfish species such as *Mugil cephalus, Lates calcarifer, Etroplus suratensis*, and *Chanos chanos* were used to estimate and assess the heavy metal concentrations based on abundance and distribution across coastal area. Results revealed that the metal concentration in these fish species, water, and sediment were all found to be significantly comparable. During the southwest monsoon season, the highest concentrations of metals were found in *Chanos chanos, Mugil cephalus*, and *Lates calcarifer*. A hazard index and a target hazard quotient were calculated to determine the human-related health risk. Except for Hg and Cd in children, the anthropological health hazard assessment revealed that most element exposure doses are safe for both children and adults.

1. Introduction

Seafood is an important protein source for humans since it provides all the required amino acids (Fuentes et al., 2009). Furthermore, due to growing awareness about the nutritional and therapeutic benefits of fish meat, consumption has expanded significantly (Medeiros et al., 2012). Though they have a great dietary value, the health risks associated with seafood must be considered because they acquire toxins and contaminants from the environment. In recent years, aquatic environments are continuously exposed to heavy metals pollution because of the increasing natural and anthropogenic activities (Hong-Giang et al., 2021) which possess a severe environmental threat to invertebrates, fish and humans (Yujun Yi et al., 2011). These trace metals are classified into essential and non-essential metals since, negligible amount of trace metals are essential to organisms. However, beyond certain levels, they can be toxic or lethal to organisms (Elbeshti et al., 2018). Since heavy metals are non-biodegradable and cannot be destroyed or created by humans, they are inert in the environs and considered as conservative contaminants if left undisturbed (Wilcock, 1999).

Fishes place a topmost level in the aquatic food chain and exhibit different sensitivities towards changes in their habitat. Hence, fishes are used to study the aquatic ecosystem pollution, as the possibilities of bioaccumulation are significantly high (Naigaga et al., 2011). Naturally, trace metals are available in the environment and are easily dissolved and readily utilized by catch fish and other marine organisms. Hence, analyzing the heavy metal concentrations in fishes will not only help us

☆ This paper has been recommended for acceptance by Jörg Rinklebe.

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https://doi.org/10.1016/j.envpol.2021.118553
Received 18 September 2021; Received in revised form 27 October 2021; Accepted 17 November 2021
Available online 3 December 2021

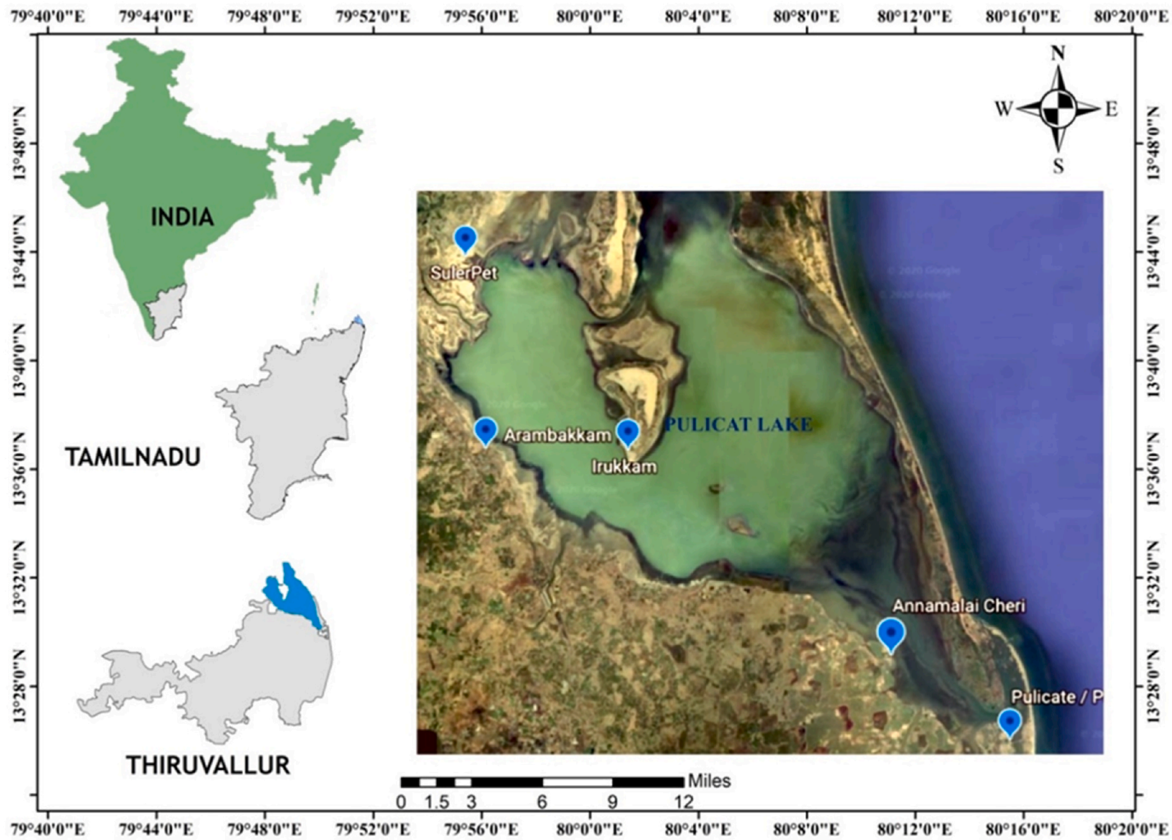

Fig. 1. The study area map.

to understand the pollutant contamination but also aid in detecting the anthropological health hazard associated with the consumption of finfish species.

Pulicate Lagoon is located along the coastal region of the Bay of Bengal. The brackish water environment supports unique biological diversity and acts as a perfect spot for breeding and nursing ground habitat for the marine organism (Kamala Kannan et al., 2008). The siltation process in this region is significantly high due to heavy rainfall received during the monsoon season. Along with the natural process, anthropogenic activities like the use of damaged fishing gear, limestone extract are other environmental concerns of this lake. Apart from these factors, the Pulicate Lake receives excessive domestic, industrial, agricultural and aquaculture discharges which act as primary factors responsible for the environmental contamination (Ruddiman, 2005). Hence, the probability of pollution contaminants impacting aquatic animals also will be high. Numerous studies report on the accumulation of heavy metals in various finfishes, water and sediments at Pulicate Lake (Barath Kumar et al., 2017). However, the anthropological health hazard associated with the ingesting of commercially important fish is to be monitored and reported regularly (Minwei et al., 2021; Li et al., 2015; Ahmed et al., 2015a). Considering this, the study aims to assess the seasonal variations of Hg, Pb, Cr, Cd, Cu, and Zn in water, sediments and four finfish species (viz., *Mugil cephalus, Lates calcarifer, Etroplus suratensis, Chanos chanos*), to estimate the daily intake and target hazard quotient of heavy metals due to the ingesting of seafood, and to assess potential health hazard for local customers using HI index.

2. Materials and methods

2.1. Materials

Analytical grade chemicals were used for the research.

2.2. Study area

Pulicate Lake is located between 13°20′- 13°40′ N latitude and 80° 14′-80° 15′ E longitude nearly aligned to the Bay of Bengal. The estuary receives freshwater from Kalangi, Swarnamukhi and Arani watercourse and finally flows in Bay of Bengal. The lake has a stretch of 60 Km from North to South with a width of 0.2 Km to 17.5 Km. Fig. 1 shows the location of stations (Pazhaverkadu, Annamalaicheri, Arambakkam, Sulurpet and Irukkam) chosen for the study.

2.3. Sample collection and preparation

The coastal water samples, sediments and finfish were collected at Pulicate from January to December 2019.

2.3.1. Water samples analysis

The water samples were composed in acid washed polyethylene container (1Litre). The parameters such as temperature, pH, salinity, alkalinity, turbidity, dissolved oxygen (DO), biological oxygen demand (BOD), and chemical oxygen demand (COD) were analyzed using multi parameters probe (PROFESSIONAL PLUS MULTIPARAMETER WATER

QUALITY METER). Then the water was filtered with 0.45 μm Whatman No.1 filter paper and the pH were reduced to 3.5 using 0.1 N HCl and heavy metal analysis was performed. To analyze the heavy metals, present in the samples, American Public Health Association standard (2005) methodology was adopted and de-ionized water was used as blank.

2.3.2. Sediment samples analysis

Sediments samples were obtained by grab sampler and the collected sediments were kept in clean polyethylene bags. Sediment was dried in hot air oven at 105 °C, homogenized to fine powder (250 μm) and stored in plastic bottles. The determination of sediments' particle size was done using sieve shaker (Associated Scientific Engineering Works, India) and then chloric acid digestion method was adopted for heavy metals analysis (Shanthi & Ramanibai., 2012).

2.3.3. Fish samples analysis

The collected fish specimens were identified by following standard identification manuals and Fish Base. Obtained samples were stored in a refrigerator at 4 °C after proper labeling and then transported to the research laboratory for further investigation. The body parts of the selected fishes (Muscle) were dissected to estimate the heavy metal concentrations present in the samples.

2.4. Heavy metal analysis

The water, sediment and fish samples were digested using microwave digestion (MARS 6 Microwave Digestion & Extraction System) and the concentrations of Mercury, Lead, Chromium, Cadmium, Copper and Zinc were investigated using SPECTRO ARCOS Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES) analyzer (SPECTRO Systematic Apparatuses, Kleve, Germany). Manufacturer instructions were followed for blank preparation and sample handling.

2.4.1. Quality assurance and quality control

Construction of standardization curves, quality assurance, and quality control were all confirmed by taking into account various aspects (blanks, standardization curve, spiked sample, and average standard checks). Heavy metal investigation was performed using NIST standard Reference material 1640 (Boumans, 1991). Hg 194.227, Pb 168.215, Cr 267.716, Cd 228.802, Cu 324.754, and Zn 213.856 were used to guarantee the calibration curve. The metals have mid-point tests ranging from 0.21 percent to 5.2 percent. The percentage of spikes recovered ranged from 96.54 to 98.85.

2.5. Statistical analyses

The analysis was done in triplicate. Mean, SD and graphical representation of data were done using R software (version 3.4.5) [PCA-FactoMine R, factoextra and ggplot 2 Package]. Pearson correlation was tested to understand the association of heavy metals observed in sediment, water and fish. ANOVA was conducted to identify the significance in metal activity among seawater, sediment and fish. To find the variation in the distribution of heavy metals in seawater, sediment and fish "principal component analysis" was conducted.

2.6. Risk assessment

2.6.1. EDI

The calculation of Estimated Daily Intake (EDI) for meal size of seafood was done from fish muscles. The risk factors and permissible consumption range were calculated by following USEPA (2000a,b); (Naji et al., 2016).

$$\text{"EDI} = \text{C} \times \text{MS/BW"} \tag{1}$$

Where.

MS - meal size; C - concentration of metal present in the body parts (Muscle) (mg/kg) and BW - total body weight.

2.6.2. CRlim

$$\text{"CRlim} = \text{RfD} \times \text{BW/C"} \tag{2}$$

Where.

CRlim - concentrated allowable ingesting rate (Kg Day^{-1}); RfD - reference dosage for all trace metal (mg/kg d^{-1}); BW – mean consumer body weight (Kg) and C - activity of metals in the finfish muscle (mg/kg) (USEPA, 2012).

2.6.3. THQ

The THQ is estimated as the percentage between exposure and reference dosages, normally THQ was used to study the noncancerous health hazard assessment. THQ was measured according to the standard assumption of integrated (USEPA, 1989) threat analysis.

$$\text{"THQ} = \text{EF} \times \text{ED} \times \text{MS} \times \text{C/BW} \times \text{RfD} \times \text{AT"} \tag{3}$$

Where.

EF - frequency of exposure and it variation based on the frequency of consumption rate (For persons who eat fish 7 times a week is considered 365 days per year while for those who eat once in a month it is considered as 12 days per year); ED - exposure duration (adults-70 years; children-6 years); C - concentration of the metals in muscles (fish muscles parts examined mg/kg); RfD - oral reference dosage (mg/g/day); BW - entire body weight (For grown person it is assumed as 70 Kg; for children, it is assumed as 16 Kg); AT - average exposure time which is equal to EF x ED (Naji et al., 2016).

2.6.4. HI

The generally potential health hazard posed by metals and the sum of THQ values for separate metals HI was used.

$$\text{"HI} = \text{THQ Hg} + \text{THQ Cr} + \text{THQ Cd} + \text{THQ Pb} + \text{THQ Cu} + \text{THQ Zn"} \tag{4}$$

3. Results and discussion

3.1. The seawater quality

The water quality parameters such as temperature, pH, salinity, alkalinity, turbidity, DO, BOD and COD were analyzed from the water samples collected at Pazhaverkadu, Annamalaicheri, Arambakkam, Sulurpet, and Irukkam to observe the seasonal variations (S. Tables 1–3). In the marine and estuarine waters, temperature plays a significant role as it affects the life of creatures and physicochemical parameters (Sukumaran et al., 2013). The water temperature showed a higher value

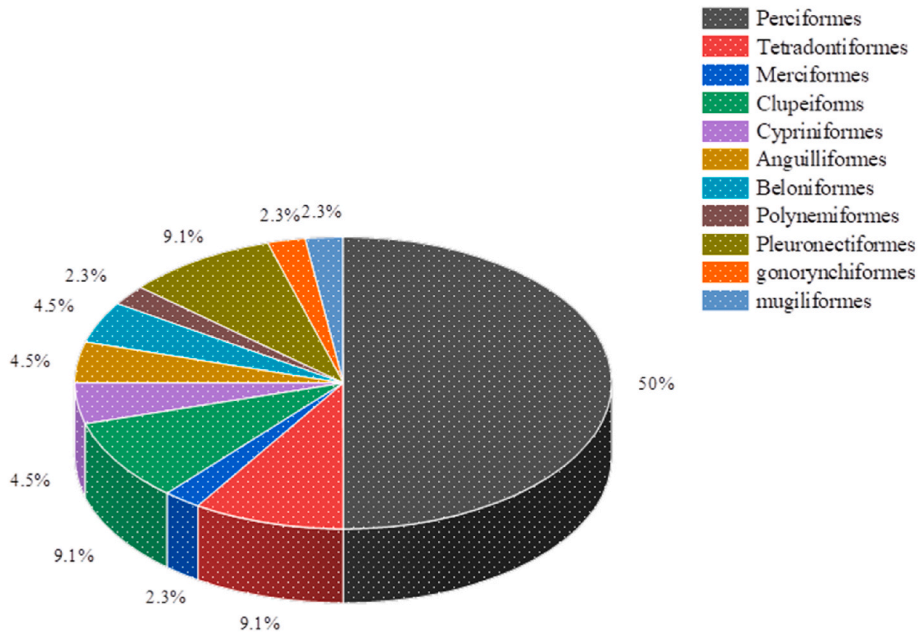

Fig. 2. Percentage composition of finfish species Orders in Taxonomic Classification.

in summer (32.4 °C) at Arambakkam and registered less in northeast monsoon (26.7 °C) at Irukkam. Salinity is one of the important water quality parameters as variations in salinity can influence the behavioral pattern and distribution of organisms in the estuarine and coastal environment (Karleskint et al., 2009). The maximum salinity of 33.9 PSU was recorded at three stations in summer (Pazhaverkadu, Annamalaicheri, Arambakkam), while the lowest value of 24.4PSU was observed at Irrukam. Generally, temperature and salinity will be high during summer due to the factors like increased solar radiation, and high evaporation whereas low during monsoon because of heavy rainfall and less air temperature (Saha et al., 2010).

The highest pH (7.8) value was registered at Annamalaicheri, Arambakkam, Sulurpet and Irukkam while the lowest pH (7.1) was documented at Irukkam in the southwest rainy season. The alkalinity was high (149.9 mg/L) in the northeast rainy season at Sulurpet whereas the less (121.5 mg/L) was noted at Pazhaverkadu during summer. Seasonal variations in pH values can associate with factors like reduction of carbon dioxide by photosynthesis through bicarbonate degradation, influences of freshwater, reduction in salinity and decay of organic matter (Paramasivam & Kannan., 2005). Turbidity is one of the important influencing parameters as it changes the distribution pattern of marine fishes (Laegdsgaard and Johnson, 2001). The highest turbidity of 6.1 NTU was observed at stations Pazhaverkadu, Annamalaicheri and Irukkam whereas the lowest turbidity (2.7 NTU) was measured during summer at Pazhaverkadu. Dissolved oxygen measurements showed the minimum concentration in summer (2.2 mg/L) while the highest concentration of DO was observed in northeast monsoon season (5.5 mg/L) at Sulurpet. The influence of fresh water and heavy mixing of atmospheric air into the water due to heavy wind which also attributes to the increase of DO during the northeast monsoon season (Morgan et al., 2006).

Less freshwater input, domestic and industrial discharges increase COD; further factors like microbial utilization of oxygen for the decomposition process and high primary productivity also attribute to high COD in the estuarine environment (Pillai, 1994). BOD and COD are used as a pointer of the seawater quality as the contents of both BOD and COD change water pollution (Maiti, 2001). The observed BOD was high in the northeast monsoon (90.2 mg/L) season and the less was recorded in summer (41.5 mg/L) at Pazhaverkadu and Arambakkam, respectively. High COD corresponds to microbial utilization of oxygen due to decay of organic matter and heavy freshwater inputs (Harrison, 1990). The high COD (352.3 mg/L) was observed in summer while less COD of (265.9 mg/L) was noted in the southwest monsoon season.

3.2. The sediment composition

The sediment contains particles of different sizes, shapes and chemical compositions and is transported by different waterways and deposited in the bottom of the seas and estuarine according to their textual properties (Ramesh and Anbu, 1996). The present study estimated the sediment quality like sand, silt, and clay. S. Tables 4–6 shows the different sediment composition percentages of samples collected at study locations for different seasons. The sand composition was found to be high (91.8%) during the northeast monsoon season while the less composition of sand was observed in (4.8%) in northeast monsoon at Arambakkam. The silt concentration was observed to be higher (78.3%) in the northeast monsoon season at Irukkam Island whereas the less silt concentration (2.4%) was recorded at the southwest season. The highest (22.3%) and lowest (3.8%) clay concentrations were recorded in the northeast monsoon season at Irukkam and Pazhaverkadu.

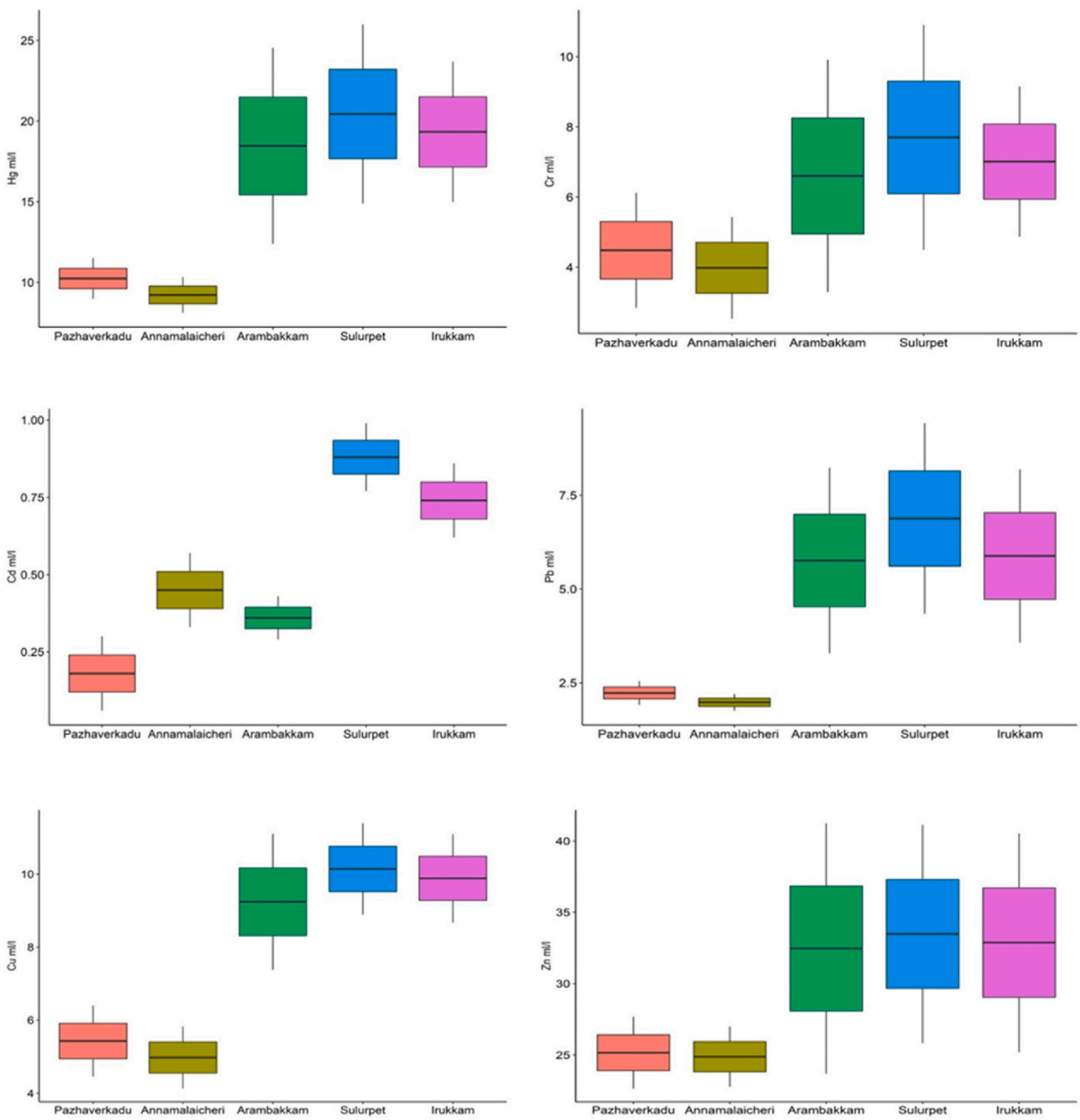

Fig. 3. The concentration of heavy metals present in water sample during southwest monsoon 2019.

Table 1
Seasonal variation in Heavy metals concentration analyzed in finfish species.

		SUM						
		Hg	Cr	Cd	Pb	Cu	Zn	Hg
Mugil cephalus	Mean	0.097	0.102	0.137	0.106	6.54	10.15	0.088
	SD	0.002	0	0.003	0.002	0.502	0.098	0.005
	Range	0.094–0.099	0.101–0.102	0.132–0.142	0.102–0.110	5.67–7.41	9.98–10.32	0.079–0.096
Lates calcarifer	Mean	0.095	0.107	0.131	0.109	6.56	10.49	0.086
	SD	0.006	0.005	0.002	0	0.058	0.035	0.005
	Range	0.0855–0.1053	0.0182–0.0982	0.1273–0.1341	0.109–0.1098	6.46–6.66	10.43–10.55	0.0772–0.095
Etropleus suratanesis	Mean	0.089	0.099	0.126	0.106	6.8	11.39	0.098
	SD	0.005	0.003	0.003	0.001	0.023	0.323	0.005
	Range	0.0802–0.0976	0.0929–0.1041	0.121–0.1318	0.1032–0.108	6.76–6.84	10.83–11.95	0.089–0.1068
Chanos chanos	Mean	0.097	0.108	0.131	0.113	6.75	10.88	0.075
	SD	0.005	0.001	0.003	0.002	0.335	0.156	0.006
	Range	0.0885–0.1059	0.1053–0.1097	0.1254–0.1366	0.109–0.117	6.17–7.33	10.61–11.15	0.0656–0.848

SUM - Summer season.
SW-M - Southwest monsoon season.
NE- M - Northeast monsoon season.

3.3. The finfish species diversity

Totally, 44 finfish species belonging to 11 orders and 33 families were identified from the fish species collected at five study locations. The families such as Siganidae and Cichilidae were found to be the most dominant fish group followed by Tetraonidae, Theraponidae, and Engrualidae (S. Fig. 1). The maximum number of fish species was collected at Pazhaverkadu followed by Annamalaichery during the collection period. Out of all finfish species collected and identified, the most predominant fish species were *Mugil cephalus* and *Chanos chanos*as their abundance and presence were noted during all the seasons. Among all the orders of finfish species observed, Perciformes consisted 50% of fish species (Fig. 2). Generally, the physical variability of an ecosystem plays an important part in the diversity of fish species at estuarine and coastal waters. Further, differences in habitat characteristics like pH, turbidity, total dissolved solids, and conductivity in different locations due to variations in land-use patterns also influence the distribution of species variations (De Silva et al., 2007). Around 8.9% of finfish species present in India were also commonly found in the world (Nelson, 2006). Many reports have investigated the diversity of finfish species in Indian coastal waters and around 569 finfish species have been reported on the southeast coast of India (CMFRI, 1969).

3.4. Heavy metals in water

Concern over contamination by heavy metals in the marine environment is continuously increasing and global attention is significant due to their abundance, persistence and ecological toxicity (Islam et al., 2015a; Ahmed et al., 2015a, b). However, it has been reported that the anthropogenic activities are significant contributors to the ongoing increase of heavy metals in various aquatic habitats (Sánchez-Chardi et al., 2009) The activity of six heavy metals (Copper, Mercury, Lead, Zinc, Chromium and Cadmium) analyzed in water samples collected at Pazhaverkadu, Annamalaicheri, Arambakkam, Sulurpet, and Irukkam are illustrated in Fig. 3 (southwest monsoon), S. Fig. 2 (summer) and S. Fig. 3 (northeast monsoon).

The range in average concentration of Mercury, Chromium Cadmium Lead, Copper and Zinc, were 08.28 ± 1.14–21.46 ± 5.54 ppm/mL; 2.91 ± 1.48–8.50 ± 3. ppm/mL; 0.14 ± 0.2–1.81 ± 0.15 ppm/mL; 0.94 ± 0.28–7.87 ± 2.51 ppm/mL; 3.97 ± 0.84–10.21 ± 1.46 ppm/mL and 23.87 ± 2.19–34.36 ± 7.74 ppm/mL respectively. The maximum values for all the heavy metals (Mercury, Chromium, Cadmium, Lead, Copper and Zinc) analyzed were observed in northeast monsoon season at Sulurpet station while the minimum was noted in summer at Annamalaicheri. The high concentration in the northeast monsoon season might attribute to the drainage of a surplus amount of rainwater discharge (Thomas & Mohaideen., 2014). In addition to this around Pulicate, there are several industries like thermal power plant located in North Chennai, Continuous port activities at Ennore, and Manali petrochemical industries are releasing untreated effluents indirectly into the lake, this could also be the reason why northeast monsoon season exhibits elevated concentration (Prabhu et al., 2008). Aside from the above factors, the anthropogenic activities like untreated sewage disposal, industrial discharge and natural phenomena such as sediment movement from tides, underwater currents bring naturally present metals to the water column (Najmeddin et al., 2017).

3.5. Heavy metals in coastal sediment

The concentration of heavy metals present in the samples is illustrated in S. Fig. 4 (southwest monsoon), S. Fig. 5 (summer) and S. Fig. 6 (northeast monsoon). The average concentration of Hg estimated for all the stations ranged from 60.41 ± 2.26 ppm/g to 86.48 ± 3.52 ppm/g registering the maximum and minimum during summer at Arambakkam and Pazhaverkadu, respectively. Among the five stations, Sulurpet exhibited high chromium content (99.84 ± 3.18 ppm/g) in the northeast monsoon season and Pazhaverkadu showed less chromium content (55.16 ± 3.44 ppm/g) during summer. The cadmium concentration was higher in the northeast monsoon season at Sulurpet while the lower concentration was at Pazhaverkadu during summer. Similarly, lead concentrations were also found to be higher in Sulurpet and lesser in Pazhaverkadu during northeast monsoon season and summer, respectively. The high (62.30 ± 4.59 ppm/g) and less (12.98 ± 6.5 ppm/g) Cu concentrations were observed at Sulurpet and Pazhaverkadu during northeast monsoon and summer season, respectively. Similar to Cu, Zn concentrations were also higher (79.14 ± 3.46 ppm/g) in the northeast monsoon season at Sulurpet and lesser (62.30 ± 4.59 ppm/g) in summer at Pazhaverkadu. The observed activity of heavy metals in the sediments

	SW-M						NE- M			
Cr	Cd	Pb	Cu	Zn	Hg	Cr	Cd	Pb	Cu	Zn
0.097	0.127	0.094	5.51	9.25	0.107	0.112	0.146	0.13	7.54	10.65
0.004	0.005	0.002	0.56	0.098	0.002	0	0.005	0.002	0.56	0.28
0.0895–0.1047	0.1186–0.1348	0.0908–0.0980	4.54–6.48	9.08–9.42	0.104–0.109	0.115–0.112	0.1377–0.1543	0.1266–0.1342	6.57–8.51	10.17–11.14
0.098	0.129	0.097	5.86	9.78	0.141	0.116	0.141	0.131	7.76	11.79
0.006	0.002	0.005	0.404	0.15	0.029	0.005	0.003	0.006	0.058	0.15
0.0886–0.108	0.125–0.1324	0.0888–0.1056	5.16–6.56	9.52–10.04	0.1074–0.1977	0.1069–0.1257	0.1354–0.1462	0.1210–0.1418	7.66–7.86	11.53–12.05
0.088	0.117	0.088	5.87	10.78	0.088	0.099	0.125	0.115	6.83	11.38
0.003	0.006	0.009	0.485	0.266	0.005	0.005	0.003	0.008	0.427	0.439
0.0828–0.0926	0.1054–0.1278	0.0722–0.103	5.03–6.71	10.32–11.24	0.0792–0.0966	0.0899–0.1071	0.1195–0.131	0.101–0.1296	6.09–7.57	10.62–12.14
0.078	0.091	0.097	3.75	8.88	0.108	0.116	0.148	0.135	7.75	11.88
0.001	0.005	0.002	0.323	0.502	0.005	0.004	0.003	0.009	0.277	0.445
0.0753–0.0797	0.0824–0.0996	0.093–0.0101	3.19–4.31	8.01–9.75	0.99–0.116	0.1083–0.1227	0.1427–0.1451	0.1196–0.1504	7.27–8.23	11.11–12.65

samples was higher during the northeast monsoon season which can be attribute to the deposition of heavy metals from various waterways conjugate at Pulicate (MacFarlane et al., 2007). Rapid developments in industrial sectors in coastal regions have undoubtedly released heavy metals around the world coastal zones, especially during the last decade the increase in industries was significantly high. Hence, coastal zones and aquatic environments are used as sinks for metals that are released by these industries and lead to the increase of heavy metals in the sediments (Santos et al., 2005).

3.6. Activity of heavy metals in seafood

Heavy metals pollution has long been considered as a serious threat, since it accumulates in the biota and non-biota of the marine environment, which leads to bioaccumulation. The possibility of bioaccumulation of heavy metals through the food chain via fish is significantly high as it is consumed widely around the globe. Hence, heavy metal analysis in fish is profoundly important for human betterment (Tepe et al., 2008). The concentration of six heavy metals analyzed in four fish species (viz., *E. suratensis, M. cephalus, C. chanos,* and *L. calcarifer*) collected in different seasons is given in Table 1. The heavy metal activity studied in the muscle of four finfish species exhibited wide variation. The outcomes revealed that all the analyzed heavy metals were found to be elevated during the northeast monsoon season with varying finfish species used for the study. The observed high values of heavy metals in different species are as: Hg (0.1977 ppm/g) – *Lates calcarifer*; Cr (0.1227 ppm/g)-*Chanos chanos*; Cd (0.1541 ppm/g) – *Chanos chanos*; Pb (0.1504 ppm/g) – *Chanos chanos*; Cu (8.51 ppm/g) - *Mugil cephalus*; Zn (12.65 ppm/g) - *Chanos chanos*. It is well reported earlier that numerous aspects (such as metal activity, pH, temperature, salinity, and DO in seawater) contribute to the variation in trace metal concentrations in a marine environment. However, the increase of heavy metals in organisms is purely based on the physiological factors like muscles, organs, and nursing conditions of the creatures (Sunlu, 2006).

Similarly, the higher accumulation of trace metals in organisms is mostly contributed by the water and food chain bio-magnifications process thereby results in higher activity in high-trophic creatures (Suseno et al., 2010). In this study, the accumulation of trace metals in fish species was high in the northeast monsoon season than in other seasons which corroborate with increased heavy metal activity observed in seawater and sediments during the same month. The least concentration of Hg (0.0656 ppm/g), Cr (0.0753 ppm/g), Cd (0.0824 ppm) and Cu (3.19) were found in the southwest monsoon season from *Chanos chanos* species. Pb and Zn alone exhibited less concentration in the southwest monsoon season from species *E. suratensis* and *C. chanos*. The analyzed heavy metals in fish muscles were compared with FSSAI, (2015), MFR. (1985), USEPA (2000) WHO (1989), FAO (1983) and EC (2014) guidelines for seafood intake by humans. Except for Cu all other metals were found to be within maximum permissible limits (MPL) Table 2. Further the heavy metal activity measured in fish muscles were also associated with other locations and literature and it exhibited varying concentrations in different locations in various seafood organisms (S. Table 7).

3.7. Relationships between metal concentrations in seawater, coastal sediment, and seafood

Analysis of variance (ANOVA) was done to study metal concentrations observed in seawater, coastal sediment and fish species. The ANOVA results showed that metals are significantly varying from water, sediment and fish species (*$P < 0.005$; $F = 632$)*. However, Pearson correlation coefficient investigation was conducted to understand the association between metals and water, sediment, and finfish species (Table 3). The results implied a significant positive correlation between Cd, Pb, Cu and Zn. However, there was no negative relationship observed among metals. Nevertheless, a less positive relationship of Hg with other metals was observed implying the direct relationship of Hg with other metals.

Principal component analysis (PCA) was used to investigate the relationships between metals in the environmental matrix, which included seawater, sediment, and seafood (Fig. 4). From the results eigenvalues more than 1 (S. Table 8) were taken for the interpretation. PCA produced five components of these dimensions 1 and 2 accounted for 79.22% and 18% respectively. In dimension-1 water (0.861), Mcep (0.978), Lcal (0.981), Esur (0.985) and Chan (0.985) were exhibiting significant positive while negative relation was observed for all the variables except water sediment in dimension-2. Heavy metals such as Mercury, Chromium, Copper and lead shows significant negative relation in dimension-1 with sediment, water, and fish species. However, Cu and Zn showed positive relation in dimension-1 with water, sediment and

Table 2
Maximum permissible limits (MPL) of metals in fish muscle (μg g^{-1} wet weight) according to national and international guideline values.

	WHO (1989)	MFR (1985)	FAO (1983)	USEPA (2000)	EC (2014)	FoodSafetyStandards Authority of India (2015)	Pulicate lake, India (finfish)											
							Summer season				Southwest monsoon season				Northeast monsoon season			
							MC	LC	ES	CC	MC	LC	ES	CC	MC	LC	ES	CC
Hg	0.3	–	0.5	0.5	0.5	0.5	0.097	0.0954	0.0889	0.0972	0.088	0.0861	0.0979	0.0752	0.107	0.1407	0.0879	0.1084
Cr	30	30	30	120	–	–	0.1021	0.1073	0.0985	0.1075	0.0971	0.0983	0.0877	0.0775	0.1121	0.1163	0.0985	0.1155
Cd	50	–	–	8	–		0.1369	0.1307	0.1264	0.131	0.1267	0.1287	0.1172	0.091	0.146	0.1408	0.1254	0.1484
Pb	2	2	0.5	4	0.3	0.3	0.1064	0.1094	0.1056	0.113	0.0944	0.0972	0.0876	0.097	0.1304	0.1314	0.115267	0.135
Cu	1	1	0.5	2	0.5	0.3	6.54	6.56	6.8	6.75	5.51	5.86	5.87	3.75	7.54	7.76	6.83	7.75
Zn	100	100	40	120	30	–	10.15	10.49	11.39	10.88	9.25	9.78	10.78	8.88	10.65	11.79	11.38	11.88

(WHO World Health Organization; MFR Malaysian Food Regulation; FAO Food and Agriculture Organization of the United Nations (FAO); USEPA United States Environmental Protection Agency; EC European Commission; FSSAI Food Safety and Standards Authority of India; MC - *Mugil cephalus*; LC- *Lates calcarifer*, ES- *Etropleus suratanesis*, CC- *Chanos chanos*)

exhibited a closer association with fish species. The PCA analysis results in the increase of heavy metals are less likely in coastal water and coastal sediment as most of the heavy metals were in a negative relationship except Cu and Zn indicating the accumulation. This significant positive relation of Cu and Zn with fish species might be due to the availability of high concentrations in water and sediment thereby must have led to accumulation. It has been reported that the accumulation of Cu increases with the presence of increased concentration, since the ions of Copper compete with the ions of other heavy metals during absorption through gills; thus, increases the bioaccumulation of Copper in fish (Malik and Maurya, 2016). Similarly, Zn is also an important constituent of various enzymes in all living organisms, positive association of Zn with coastal water, coastal sediment and fish species might attribute to the higher concentration of Zn observed.

3.8. Evaluation of human health risk due to heavy metals present in fish samples

Considering the importance of fish, the present study investigates the EDI of fish muscle (formula 1) was applied. The calculated EDI for the heavy metals presents in fish samples for Pulicate Lake is presented in Table 4 for southwest monsoon, S. Table 9 for summer and S. Table 10 for northeast monsoon. The present study evaluated the EDI using RfD (Reference Dose Factor), RfD represents the daily exposure limit of particular metal to a human population without a significant hazard of harmful effects. As per the New York State Department of Health (2007) if the obtained ratio of EDI is similar or lesser than RfD ratio, the harmful effects would be minimum on the human population. However, a varying ratio of EDI exhibits varying effects on humans are as follows; the EDI ratio of 1–5 times – low risk, 5–10 times – reasonable risk, and the threat becomes high if the ratio is more than ten times the RfD. Hence, EDI is used as a reference point to identify the hazardous effects on humans by the metals at different doses. The estimated EDI values were compared with permissible limits of various heavy metals recommended by different organizations like Joint FAO/WHO Expert Committee on Food Additive (1982, 1989, and 2000), World Health Organization (1996, 2006) and National Research Council (1989).

In the present study EDI results obtained from children and adults were lesser than the permissible limits, implying the adverse effects due to metal exposure would be more unlikely. However, the toxic effects due to trace elements to human beings depends on regular consumption (Singh et al., 2010). The daily concentrated allowable consumption rate (formula 2) for polluted fish created on non-carcinogenic effects due to heavy metals analyzed for all the individual species of fish are summarized in Table 4 for southwest monsoon, S. Table 9 for summer and S. Table 10 for northeast monsoon. The estimated CRlim data for all the seasons and fish species were higher than EDI (CRlim > EDI) values and indicates that the fish are safe to consume. Nevertheless, the THQ and HI values also should be considered for the cumulative effects of heavy metals on humans.

The THQ values for all the heavy metals assessed (formula 3) during all the seasons for different finfish species are presented in Table 4 for southwest monsoon, S. Table 9 for summer and S. Table 10 for northeast monsoon. THQ is used to study the non-carcinogenic health effects for human beings by each metal due to the consumption of seafood (U.S. Environmental Protection Agency, 2014). THQ values estimated for all the metals for adults in all the fish species were lesser than one (<1) throughout the study period indicating no potential carcinogenic effect. However, the THQ estimated for children was lesser than one (<1) for all the metals (Cr, Pb, Cu, Zn) except Hg and Cd, implying significant risk to children who are below 16 years. Generally, human beings exposed to more than one heavy metal are highly vulnerable to toxic effects (Li et al., 2013). Hence, it is essential to determine the cumulative effects of trace metal on humans HI index which was evaluated using formula 4. The HI index was evaluated based on the THQ values, if the HI values exceed more than one (>1) the effects on humans would be significantly

Table 3
Inter-elemental correlation matrix of heavy metals in the seawater, sediment and seafood of the Pulicate Lake.

	Hg	Cr	Cd	Pb	Cu	Zn
Hg	1					
Cr	0.627	1				
Cd	0.743	0.218	1			
Pb	0.753	0.225	0.999	1		
Cu	0.772	0.238	0.998*	0.999	1	
Zn	0.773	0.239	0.997*	0.998	0.999	1

high. In the present study, the HI values calculated were significantly high (>1) for both children and adults as represented in Table 4 for southwest monsoon, S. Table 9 for summer and S. Table 10 for northeast monsoon indicating a potential risk to humans. This further creates concern over the residents of the Pulicate and avoiding excessive consumption of studied fish food might prevent the adverse effects due to heavy metals.

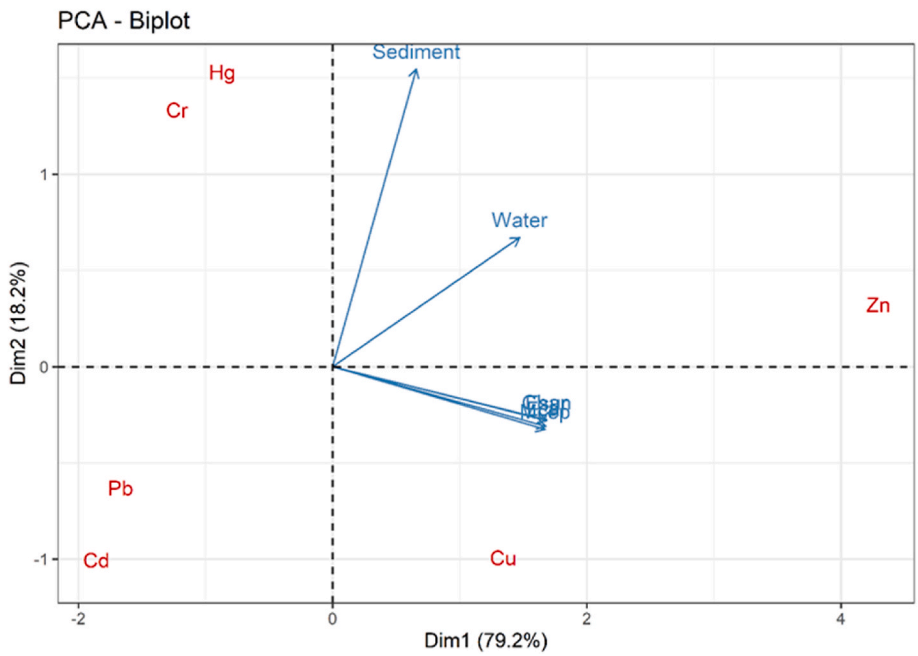

Fig. 4. PCA of heavy metals in environmental matrix, such as seawater, sediment and seafood (Mcep- *Mugil cephalus*; Lcal- *Lates calcarifer*; Esur- *Etroplus suratensis*; Chan- *Chanos chanos*).

Table 4
Target hazard quotient (THQ) values in finfish species during southwest monsoon season 2019.

				EDI		CRlim		THQ	
Species	Metal	M	Rfd	Adult	Child	Adult	Child	Adult	Child
Mugil cephalus	Hg	0.088	0.0005	0.000285	0.000627	0.397727	0.090909	0.570743	1.254
	Cr	0.0971	0.0015	0.000315	0.000692	1.081359	0.247168	0.209921	0.461225
	Cd	0.1267	0.001	0.000411	0.000903	0.552486	0.126283	0.41087	0.902738
	Pb	0.0944	0.004	0.000306	0.000673	2.966102	0.677966	0.076531	0.16815
	Cu	5.51	0.4	0.017868	0.039259	5.08167	1.161525	0.04467	0.098147
	Zn	9.25	0.3	0.029996	0.065906	2.27027	0.518919	0.099988	0.219688
Hazard Index (HI)								1.412724	3.103947
Lates calcarifer	Hg	0.0861	0.0005	0.000279	0.000613	0.406504	0.092915	0.55842	1.226925
	Cr	0.0983	0.0015	0.000319	0.0007	1.068159	0.244151	0.212515	0.466925
	Cd	0.1287	0.001	0.000417	0.000917	0.543901	0.12432	0.417356	0.916988
	Pb	0.0972	0.004	0.000315	0.000693	2.880658	0.658436	0.078801	0.173138
	Cu	5.86	0.4	0.019003	0.041753	4.778157	1.09215	0.047508	0.104381
	Zn	9.78	0.3	0.031715	0.069683	2.147239	0.490798	0.105717	0.232275
Hazard Index (HI)								1.420317	3.120631
Etroplus suratensis	Hg	0.0979	0.0005	0.000317	0.000698	0.357508	0.081716	0.634951	1.395075
	Cr	0.0877	0.0015	0.000284	0.000625	1.197263	0.27366	0.189599	0.416575
	Cd	0.1172	0.001	0.00038	0.000835	0.59727	0.136519	0.380063	0.83505
	Pb	0.0876	0.004	0.000284	0.000624	3.196347	0.730594	0.071019	0.156038
	Cu	5.87	0.4	0.019036	0.041824	4.770017	1.09029	0.047589	0.104559
	Zn	10.78	0.3	0.034958	0.076808	1.948052	0.445269	0.116527	0.256025
Hazard Index (HI)								1.439748	3.163322
Chanos chanos	Hg	**0.0752**	0.0005	0.000244	0.000536	0.465426	0.106383	0.487726	1.0716
	Cr	**0.0775**	0.0015	0.000251	0.000552	1.354839	0.309677	0.167548	0.368125
	Cd	**0.091**	0.001	0.000295	0.000648	0.769231	0.175824	0.2951	0.648375
	Pb	**0.097**	0.004	0.000315	0.000691	2.886598	0.659794	0.078639	0.172781
	Cu	**3.75**	0.4	0.012161	0.026719	7.466667	1.706667	0.030402	0.066797
	Zn	**8.88**	0.3	0.028797	0.06327	2.364865	0.540541	0.095989	0.2109
Hazard Index (HI)								1.155403	2.538578

4. Conclusion

The present study investigated the seasonal variations in species diversity and heavy metals concentrations in fishes in selected landing centers at Pulicate Lake. The heavy metals analyzed in water, sediment and fish muscle samples exhibited varying concentrations. High metal concentrations were observed during northeast monsoon in samples which clearly implies the influence of rainfall with heavy discharge along with pollutants from various industrial sources. Among four species studied, *Chanos Chanos* consists of a high concentration of Cadmium, Lead and Chromium. A high concentration of Mercury and Copper was observed in *Lates calcarifer* and *Mugil cephalus*. The potential health risk estimated (THQ and HI) due to the consumption of four finfish species indicated that there is a possible significant threat to children due to Hg and Cd exposure as the values exceeded the maximum permissible limits. Hence, research on toxic heavy metals present in various seafood organisms at the Pulicate region is essential to provide efficient data to ensure the safety of local residents as well as the consumers. Further, the study emphasizes regular monitoring of these hazardous trace elements has to be carried-out and recommends taking actions to control them.

Credit author statement

Kumar Pandion: Conceptualization, Investigation, Methodology, Writing – original draft; **Mohamed Khalith S. B**: Investigation, Methodology; Writing – original draft; **Balasubramani Ravindran**: Validation, Conceptualization, Writing – review & editing; **Murugesan Chandrasekaran**: Writing – review & editing; **Rajakrishnan Rajagopal**: Writing – review & editing; **Ahmed Alfarhan**: Validation, Data curation; **Soon Woong Chang**: Writing – review & editing; **Ramamoorthy Ayyamperumal**: Writing – review & editing; **Amitava Mukherjee**: Writing – review & editing; **Kantha Deivi Arunachalam**: Validation, Project administration, Funding acquisition, Resources, Supervision and Visualization.

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.

Acknowledgement

The authors are grateful to SRM Institute of Science and Technology, Kattankulathur-603203, for providing infrastructure and continuous support to carry out this work. The authors acknowledge King Saud University, Riyadh, Saudi Arabia for funding this research through Researchers Supporting Project No: RSP 2021/11. This work was supported by the National Research Foundation of Korea by the Korean Government (MSIT) (2020R1G1A1012069).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.118553.

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