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Heavy metal pollution in selected upland tributaries of Sri Lanka: comprehension towards the localization of sources of pollution

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ABSTRACT

This study aimed to assess the heavy metal (HM) profile of the main upland tributaries of three major rivers, the Mahaweli, the Deduru and the Gin Rivers, which are commonly used for urban water supply in Sri Lanka. The HM profiles of arsenic (As), cadmium (Cd), lead (Pb) and mercury (Hg) were investigated by ICP-MS. Land-use classification was performed to locate the main sources of pollution. Mean pH, TDS and conductivity showed significant inter-site mean differences ($p < 0.001$). The mean contents of the HMs, considering all rivers, were observed in the order [Hg] > [As] > [Pb] > [Cd] at the sources and [As] > [Hg] > [Pb] > [Cd] at the river mouths. Particularly, in the Mahaweli River, the mean As content was $0.08 \pm 0.05\ \mu g \cdot L^{-1}$ and showed an increasing trend from the source to the river mouth. In the Deduru River, the mean Hg content was $0.14 \pm 0.15\ \mu g \cdot L^{-1}$, and of all rivers studied, the highest content of $0.50 \pm 0.17\ \mu g \cdot L^{-1}$ was recorded. The Gin River showed significant inter-site mean differences ($p < 0.05$) in [Pb], [As] and [Hg]. In all rivers studied, [As] was significantly higher in water samples collected near agricultural lands and urban areas as compared with the other land-use/cover types, which was further proved by a significant positive correlation (coefficient = 0.479, $p = 0.0325$). We, therefore, emphasized that HM pollution is more likely due to anthropogenic activities in the upper catchment with less lithogenic contamination. However, national water quality management should be further strengthened and new policy enforcement is emphasized.

Key words: agrochemicals, arsenic, chronic kidney disease, human health, point pollution

HIGHLIGHTS

- Arsenic (As) content was higher at the river mouths.
- Mercury had the highest mean concentration at the sources.
- As concentration increased gradually from the source to the river mouth.
- Agricultural activities could be the source of As addition.
- Sri Lankan river waters are still safe to be used for drinking purposes.

INTRODUCTION

Water, as one of the most precious resources for every living entity, represents a basic daily need for the global population that reached 7.7 billion in 2019 (PRB 2019). As it becomes increasingly scarce, different conservation measures are applied worldwide. Sri Lanka is relatively well provided with water resources, and the annual per capita water supply in the country is above the threshold of 1700 mm (Falkenmark *et al.* 1989). The major drinking water sources in Sri Lanka are piped supply, protected or unprotected wells and sources such as reservoirs, streams and tributaries. According to the World Health Organization (WHO) & UNICEF (2006), 79% of the total population had access to improved drinking water, i.e. protected from external contamination and from faecal matter (free of *Escherichia coli* contamination) in 2004, and improved water sources comprise household water supply, public standpipes, protected dug wells, protected springs and rainwater collection. When domestic consumption is considered, one-third of the population has access to piped water. The country's population, however, mainly depends on groundwater sources since they are the preferred low-cost water source for most rural and peri-urban households, while 7% of the population uses water from rivers and reservoirs for drinking purposes (Bandara 2003). It has long been known that shallow open dug wells have provided the basic drinking domestic water supply to a major proportion of rural residents since medieval times.

Despite the fact that there is good supply of water to cater to the needs of the people, the rapid increase in population over recent decades has tended to exert pressure on both the quantity and the quality of drinking water supply (Bandara 2003). Moreover, Mahagamage & Manage (2014) reported that the majority of observed sites in the Kelani river basin, which hosts more than a quarter of the national population and provides about 80% of drinking water to the Colombo area, has a poor water quality index. Their results further showed that the water quality parameters are above the permissible levels recommended by the WHO and Sri Lankan Standards (SLS) for drinking water. In the 1980s, several studies attempted to alert governmental institutions about the above permissible limits of nitrate and coliform contents, heavy metal (HM) contamination and high fluoride content in drinking water (Dissanayake *et al.* 1987). Apart from this, in recent decades, a large amount of agricultural, industrial and domestic pollutants, especially HMs, have accumulated in coastal aquatic ecosystems like lagoons, estuaries, etc. (Kodikara 2021). Therefore, it is apparent that many water bodies are at risk of being polluted by HMs. In this context, it is more important to study the HM profiles of the water tributaries that are being used for providing drinking water.

HM pollution has been the subject of extensive discussion and is tied to the fact that drinking HM polluted water plays a significant role in the development of chronic kidney disease of unknown aetiology (CKDu) (Chandrajith *et al.* 2011) and some health issues in parts of Sri Lanka (Kodikara 2021). As CKDu is prevalent in human settlements where groundwater is used as the main drinking source, different risks factors, in particular, cadmium (Cd), lead (Pb), arsenic and their mixtures, which are considered to be of anthropogenic origin, have been found to be related to the disease (Bandara *et al.* 2011). As there is still no consensus about the aetiology of CKD and the rural population does not have better access to safe drinking water, the epidemic is likely to continue. As a result, the study of HM contents in drinking water will help in human health management. Therefore, this study aims at establishing the HM profiles of selected major upland tributaries of Sri Lanka which are being used as major sources of drinking water to the public. It is further believed that this study will create a strong baseline database that will be useful for water quality management and associated policy enforcement in the country. The following questions are addressed: (a) What are the HM profiles of the upland tributaries of the Mahaweli, Deduru and Gin Rivers? and (b) Which localities (source to the river mouth) contain the highest concentrations of HM pollution?

METHODOLOGY

Study site

Sri Lanka is located in the Indian Ocean between 05°55′ and 09°51′N latitude and 079°41′ and 081°53′ E longitude. It has a total land area of approximately 65,610 km^2 with a coastline of about 1738 km. The country is divided into four major climatic zones, namely, wet, dry, intermediate and arid zones. The wet zone is mainly confined to the southwestern region and the dry zone to the northern and eastern parts of the country. These two zones are separated by the intermediate zone. The arid zone, on the other hand, is found in the northwestern and southern parts of the country, and the climatic conditions are very different in the climatic zones. The country presents a network of 103 major river basins. The most important rivers in terms of size, length and usage are, in clockwise direction, Mahaweli (mean annual discharge 11,016 million m^3), Gal, Menik, Walawe (2,165 million m^3), Nilwala, Gin (1,903 million m^3), Bentota, Kalu (7,862 million m^3), Kelani (5,474 million m^3), Maha, Deduru (1,608 million m^3), Malvathu and Yan Rivers. Many of them are being used directly by the public or for supplying drinking water. Based on the level of use and size of the rivers, three major upland tributaries, namely, the Mahaweli (Central, North Central and Eastern Provinces), the Deduru (North Western Province) and the Gin (Southern Province) Rivers (Figure 1) were selected for the study.

The Mahaweli River is the longest river of Sri Lanka with a length of 335 km, crossing three different provinces: Central, North Central and Eastern. It starts from the upland territory within the wet zone and then flows towards the North East. Its

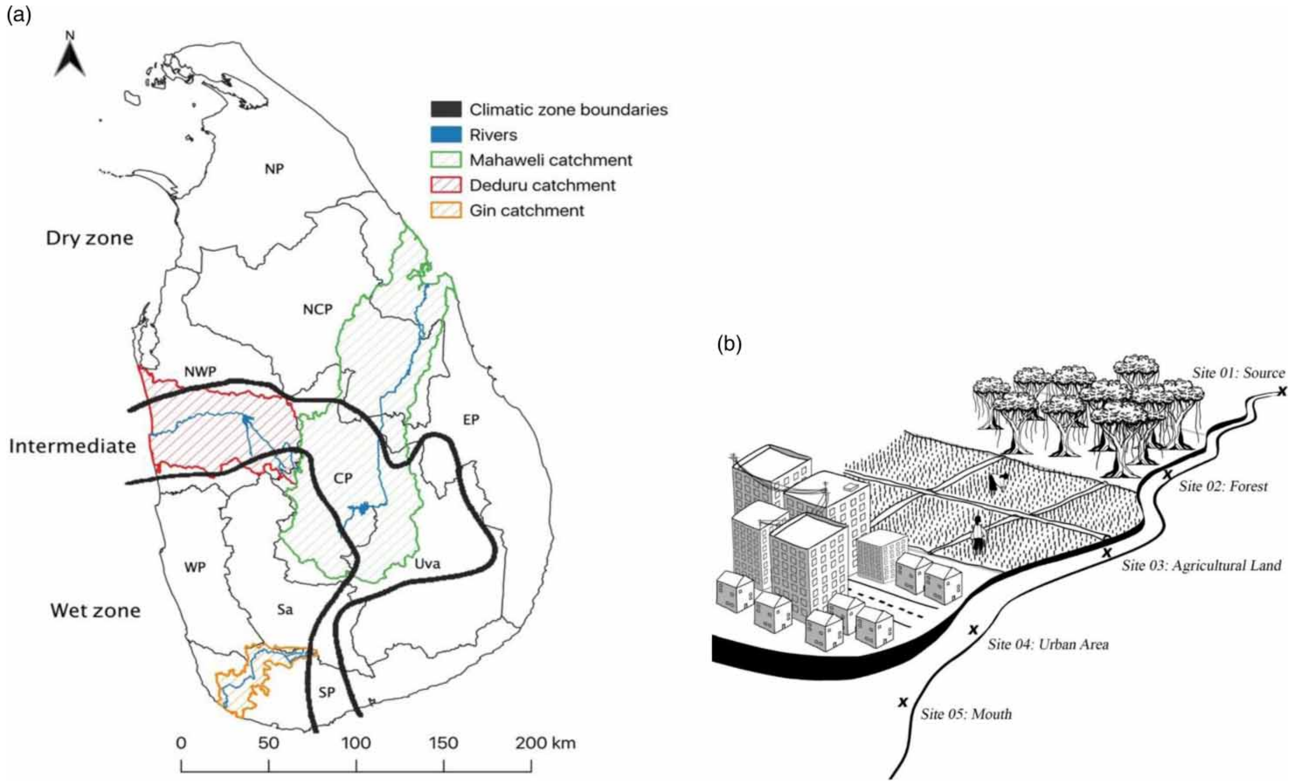

Figure 1 | (a) Provinces and climate zones of Sri Lanka. CP: Central Province; EP: Eastern Province; NCP: North Central Province; NP: Northern Province; NWP: North Western Province; Sa: Sabaragamuwa Province; SP: Southern Province; Uva: Uva Province; WP: Western Province. (b) Sampling design of the study which covers source, forest, ALs, UAs and river mouth in order.

catchment area, with a length of 10,227 km^2, is the largest in Sri Lanka covering around one-sixth of the territory. The stream feeds four major reservoirs, namely, Kothmale, Victoria, Randenigala, Rantambe and around 4000 small ones in order to irrigate agricultural land (AL). These small tank cascade systems are a part of the traditional water management system and the main source of drinking and irrigation water for farmers in the North Central Province (NCP) (Bandara *et al.* 2011). The Deduru River is 142 km long and runs through the North Western Province. Its basin of 2,616 km^2 is located entirely in the dry zone. The river feeds the Deduru reservoir and is also used for drinking purposes and irrigation of ALs. The Gin is also a large river, 113 km in length and essentially located in the Southern province, even if some tributaries have their source in the Sabaragamuwa Province. Its catchment of 932 km^2 is located in the wet zone. The stream is used for drinking purposes and irrigation systems (Amarathunga & Kazama 2016). Each tributary was selected on the basis of its capacity to mimic the baseline pattern (see Figure 1(b)).

Remote sensing and GIS work; land-use architecture of catchments

Archives of Google Earth Pro (version 7.3.3.7699_64-bit) satellite images were used in identifying the land-use/cover classes of the selected rivers. Accordingly, forest cover (FC), urban area (UA), AL, reservoir (R) and bare soil (BS) land-use classes were identified and were mapped using the on-screen digitization method (Madarasinghe *et al.* 2021). The catchment areas were extracted using QGIS (3.4.10-Madeira) and digital elevation model data source from the SRTM Downloader (3.1.5 plugin). Image attributes used in discriminating the aforementioned land-use classes are given in Table 1.

Water sampling

Surface water sampling was done at major sampling locations along the selected rivers, as shown in Figure 1(b). Accordingly, water samples were collected at **site 1**: close to the source; **site 2**: immediately after the FC and well before the first patch of AL; **site 3**: after the first patch of ALs; **site 4**: more downstream, immediate after the UAs; **site 5**: close to the river mouth but far enough to avoid brackish water contamination. Water samples were collected into a 250 ml glass sampler (DURAN®

Table 1 | Image attributes used in the study

Land-use class	Image attributes
Forest cover	Different shades of green; dense crowns; heterogeneous texture
Urban area	White, red and brown colours; polygonal shapes and straight lines
Agricultural land	Light green or brown-beige colours and the characteristic grid pattern (paddy); homogeneous stands (plantations); star-shaped crowns (palm or coconut); crowns turning reddish during the dry season (rubber)
Reservoir	Blue and turquoise colours; flat surface; BS in the dry season for seasonal reservoirs
Bare soil	Dark colour and smooth surface (rock hill); yellow or red colour and granular surface (open mines)

Laboratory bottle washed with 1% HNO_3 for 24 h before use) at a depth of 30 cm using a Ruttner sampler (HYDRO-BIOS, PMMA Water Sampler, China). At each location, five samples of 250 ml each were taken at different points along a transversal section of the stream in order to minimize local variations. Immediately after the water collection, the samples were labelled and stored in a refrigerator until the analyses were done. In total, 110 samples were collected during the year 2020 from the three selected rivers as follows: 35 samples (07 locations $\times$ 5 samples per location) from the Mahaweli River, 35 samples (07 locations $\times$ 5 samples per location) from the Deduru River and 40 samples (08 locations $\times$ 5 samples per location) from the Gin River.

Assessment of water quality and HM contents

As the water quality parameters, pH, conductivity and total dissolved sediments (TDS) were measured. A small fraction of 80 ml was taken from the original sample and was further subdivided into 20- ml portions, which were considered replicates. The pH of the water samples were measured by using a pH meter (WalkLAB HP9010, Singapore), while conductivity and TDS were measured by using a multimeter (Hanna HI9835, USA) at an ambient temperature of 31 °C after proper calibration. All measurements were done in triplicate. The contents of the selected HMs such as arsenic (As), Cd, mercury (Hg) and Pb in water samples were determined using inductively coupled plasma mass spectrometry (ICP-MS) (CCME 1991). Approximately 5 ml of the homogenized sample was measured into an EasyPrep high pressure microwave vessel and 10 ml of 65% nitric acid was added. The EasyPrep microwave digestion programme was followed to digest the samples using a microwave digester (CEM MARS 5, USA). The digested sample was quantitatively transferred and filtered using No. 542 Whatman filter paper followed by washing with deionized water and volume up to 25 ml. The prepared solution was used to analyse mineral content by ICP-MS (Agilent 7900, USA). *LOQs pertaining to HM analyses are given below.

Data analyses

The variables pH, conductivity, total dissolved solids (TDS), HM contents and land-use cover were treated as continuous variables, while sampling location (source to mouth) and sampling-site category were treated as categorical ordinal variables. All the parametric assumptions were met for the aforementioned variables, with an error margin of $\alpha = 0.05$, and parametric tests were performed taking the sampling location as a fixed factor. Each continuous variable was compared among the sampling locations using one-way ANOVA with null hypothesis: there was no significant difference in the means of the tested parameters among the sampling locations. Thereafter, the mean value of each tested parameter in each site was compared with the recommended values of the WHO (2017) and SLS (2013) using the Student's *t*-test. When the parametric conditions were not met, the non-parametric Kruskal–Wallis test was carried out. A non-parametric Kendall correlation test was performed to create the correlation matrix and to check the correlation between land-cover type and HM contents. Each significant correlation was tested with a linear regression model wherein all the assumptions of linearity, homoscedasticity, independence and normality were respected. All statistical analyses were performed using R (version 3.5.1) statistical software.

RESULTS

Land-use/cover architecture and cover estimation

The identified land-use/cover types, i.e. FC, UA, AL, R and BS, could be observed in all three rivers. According to land-use/cover estimation, the Mahaweli River margins are dominated by forest with 63.1% cover, while AL and UA constitute 23.0 and 13.9% in that order (Table 2 and Figure 2(i)). The catchment area of the Mahaweli River is mainly occupied by forest with 48.8% cover, followed by AL (31.1%). With regard to the Deduru River, the river margins are dominated by AL (69.9%), while

Table 2 | Distribution of land-use/cover classes (in hectare) among the selected tributaries

River catchment	Site	AL	FC	UA	BS	R
Gin	G1 [03]	3,798 (51.2)	2,445 (32.9)	1,178 (15.9)	0	0
	G2 [03]	1,967 (51.6)	1,518 (39.9)	323 (8.5)	0	0
	G3 [03]	9,012 (42.9)	10,703 (51.0)	1,262 (6.0)	6 (<0 .1)	1 (<0 .01)
	G4 [03]	9,960 (43.6)	9,851 (43.1)	3,004 (13.2)	0	18 (<0 .1)
	G5 [04]	13,426 (62.3)	3,127 (14.5)	4,965 (23.1)	18 (<0 .1)	3 (<0 .1)
	G6 [05]	7,556 (54.0)	895 (6.4)	5,492 (39.3)	11 (0.1)	23 (0.2)
	Total	45,719 (50.5)	28,739 (31.5)	16,224 (17.9)	35 (<0.1)	45 (0.1)
Mahaweli	M1 [01]	23 (7.1)	299 (92.9)	0	0	0
	M2 [02]	40 (7.4)	504 (92.6)	0	0	0
	M3 [03]	374 (41.0)	485 (53.2)	53 (5.8)	0	0
	M4 [04]	2,471 (49.4)	2,196 (43.9)	334 (6.7)	0	0
	M5 [04]	3,190 (48.3)	2,015 (30.5)	1,392 (21.1)	0	2 (<0 .1)
	M6 [04]	86,890 (34.7)	112,802 (45.0)	40,458 (16.1)	796 (0.3)	9,749 (3.9)
	M7 [05]	125,328 (28.6)	224,584 (51.3)	62,173 (14.2)	1,028 (0.2)	25,156 (5.7)
	Total	218,316 (31.1)	342,885 (48.8)	104,410 (14.9)	1,824 (0.2)	34,907 (5.0)
Deduru	D1 [01]	168 (49.0)	69 (20.1)	106 (30.9)	0	0
	D2 [02]	29 (13.6)	129 (60.6)	55 (25.8)	0	0
	D3 [03]	500 (35.7)	604 (43.1)	286 (20.4)	12 (0.8)	0
	D4 [04]	2,461 (60.7)	736 (18.2)	813 (20.0)	45 (1.1)	0
	D5 [04]	17,291 (56.0)	6,017 (19.5)	7,274 (23.6)	150 (0.5)	126 (0.4)
	D6 [04]	12,622 (61.8)	2,129 (10.4)	5,310 (26.0)	34 (0.2)	336 (1.6)
	D7 [04]	65,400 (62.9)	12,876 (12.4)	22,191 (21.4)	601 (0.6)	2,821 (2.7)
	D8 [05]	24,836 (60.7)	3,061 (7.5)	10,920 (26.7)	49 (0.1)	2,044 (5.0)
	Total	123,307 (61.0)	25,621 (12.7)	46,955 (23.2)	891 (0.5)	5,327 (2.6)

Percentages are given in brackets '()'. Sampling site category is given in brackets '[]' (see Figure 1(b)).

forest and UA constitute 14.9 and 15.2% cover, respectively. When the catchment area of the Deduru River is considered, the same trend remains with 61.0% of the surface covered by AL, followed by UA (23.2%) and FC (12.7%). In addition, FC decreases along the river in the direction from the source to the mouth. The banks of the Gin River are dominated by AL (45.4%) and UA (41.0%) with a low FC (13.6%). Moreover, the catchment area is dominated by AL (50.5%), while the rest is divided between FC (31.5%) and UA (17.9%). In the flow direction, FC decreases, while UA increases from the source to the mouth.

Assessment of water quality

The Mahaweli River (M for the Mahaweli River) had an average pH of 6.75 $\pm$ 0.34 within the range of 6.23–7.13. Sites M1 and M2 showed values significantly below the threshold ($p < 0.001$; see Table 3). According to the Kruskal–Wallis test results, the three parameters pH, TDS and conductivity showed significant inter-site mean differences ($p < 0.001$). The pH variation in the Mahaweli River was as follows: pH M1 = pH M2 < pH M3 < pH M4 = pH M5 = pH M6 = pH M7. The TDS showed a specific pattern, increasing from the source to the river mouth, as follows: TDS M1 = TDS M2 < TDS M3 = TDS M4 = TDS M5 = TDS M6 < TDS M7. Conductivity also followed a similar pattern of TDS. The Deduru River (D for the Deduru River) had a mean pH of 7.03 $\pm$ 0.37 within the range of 6.37–7.43. The Kruskal–Wallis test results showed that water pH, TDS and conductivity were significantly different ($p < 0.001$) among the sampling sites (source to the river mouth). The pH along the river showed the pattern of pH D1 = pH D2 = pH D3 = pH D4 = pH D5 < pH D6 = pH D7 = pH D8, while the TDS showed a slight increase along the sites as follows: TDS D1 < TDS D2 < TDS D3 < TDS D4 < TDS D5 < TDS D6 < TDS D7 < TDS D8. Conductivity followed a pattern similar to the TDS. The Gin River (G for the Gin River) pH values were within the range of 6.33–6.72, and the average pH was 6.47 $\pm$ 0.17. The sites G1, G2 and G3 showed values that were significantly lower than the threshold. According to the Kruskal–Wallis test results, water pH, TDS and conductivity were significantly different ($p < 0.001$) among the sampling sites (source to the river mouth). The parameter, pH, showed a significant difference between sites G2 and G5. The parameters TDS and conductivity showed a common pattern, which was a decrease from the source to the river mouth: TDS G1 > TDS G2 = TDS G3 > TDS G4 = TDS G5 < TDS G6.

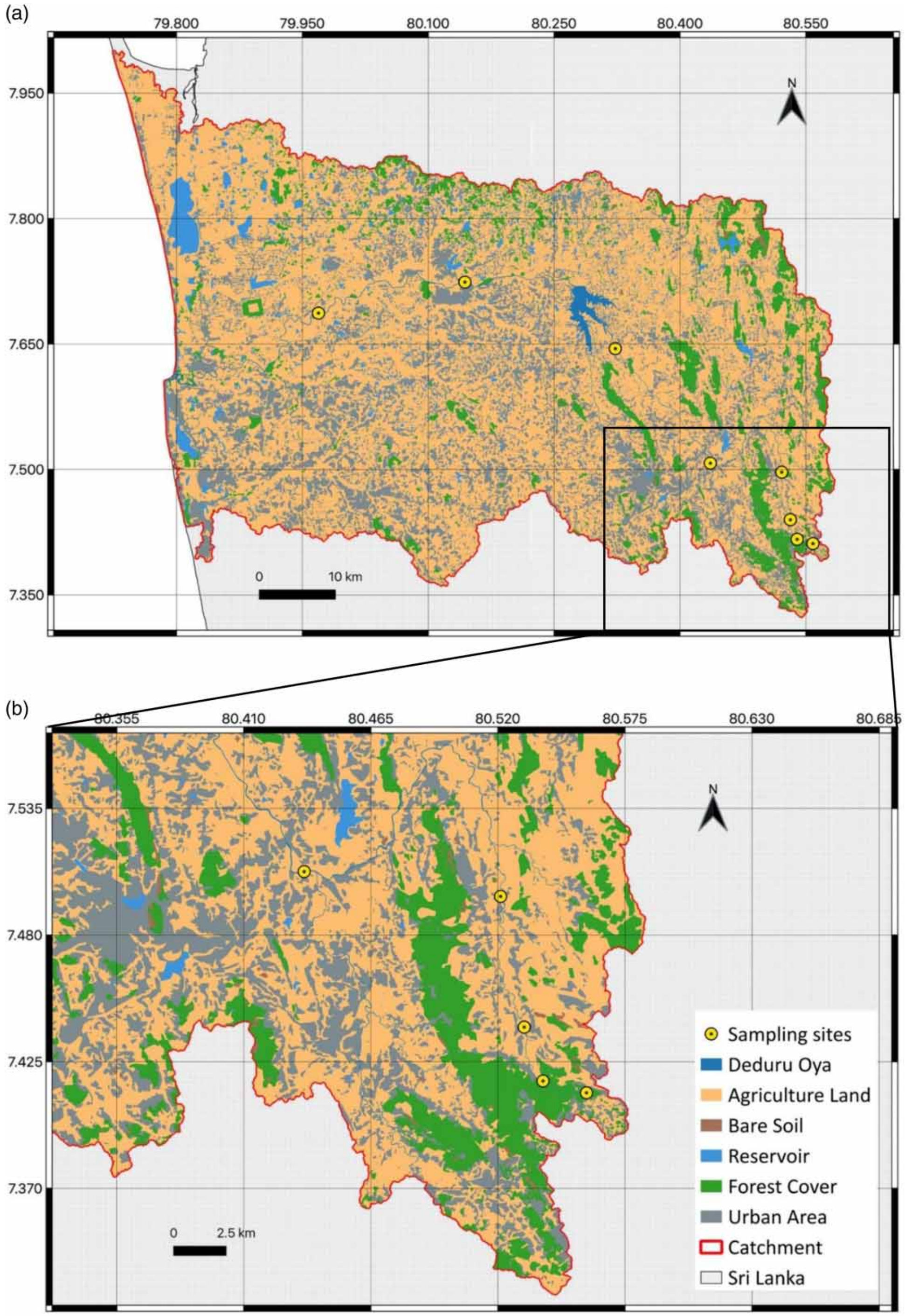

Figure 2 | (a) Land-use/cover map of the Mahaweli River; (b) detailed map of the Mahaweli River's catchment; (c) land-use/cover map of the Deduru River and (d) detailed map of the Deduru River's catchment; (e) land-use/cover map of the Gin River. Legends are given with respect to each map. The terms 'Ganga and Oya' are referred to as rivers. (*continued.*).

HM contents in tributary water

The measured HM contents in the selected main tributaries did not exceed the safety limits stipulated by the World Health Organization (WHO) and SLS guidelines (Table 4). Among the HMs studied, Cd was present in insignificant amounts,

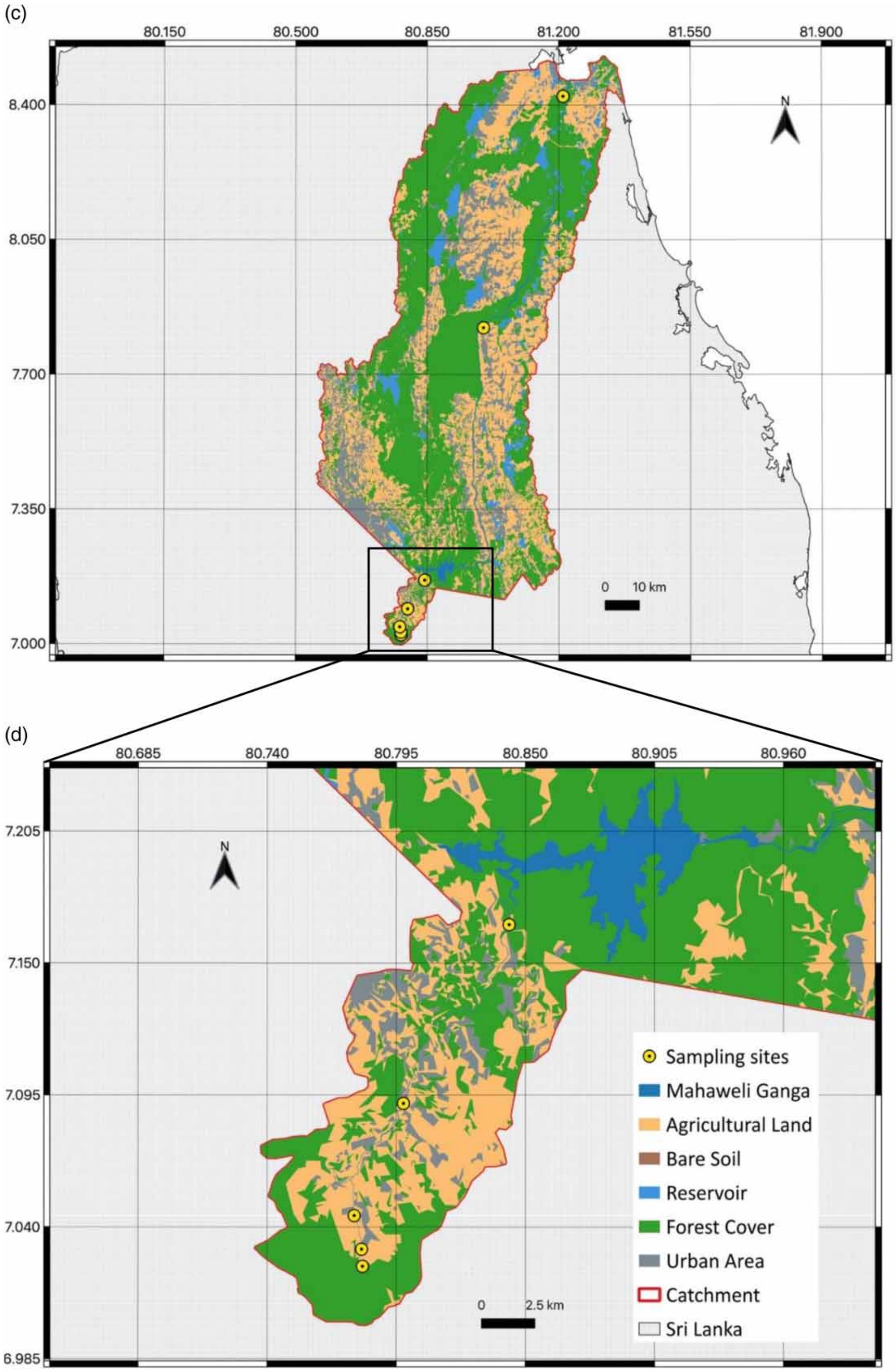

Figure 2 | (Continued).

frequently below the detection limit of 0.01 μg·L^{-1}. In the Mahaweli River, the measured Pb content was very low, the mean Hg content was 0.12 ± 0.07 μg·L^{-1} and the inter-site variation was not significant ($p > 0.05$). The mean As content was 0.08 ± 0.05 μg·L^{-1} and showed an increasing trend along the river (from the source to the river mouth): [As]M1 = [As]M2 < [As]M3 = [As]M4 = [As]M5 = [As]M6 < [As]M7 with [As]M6 > [As]M3.

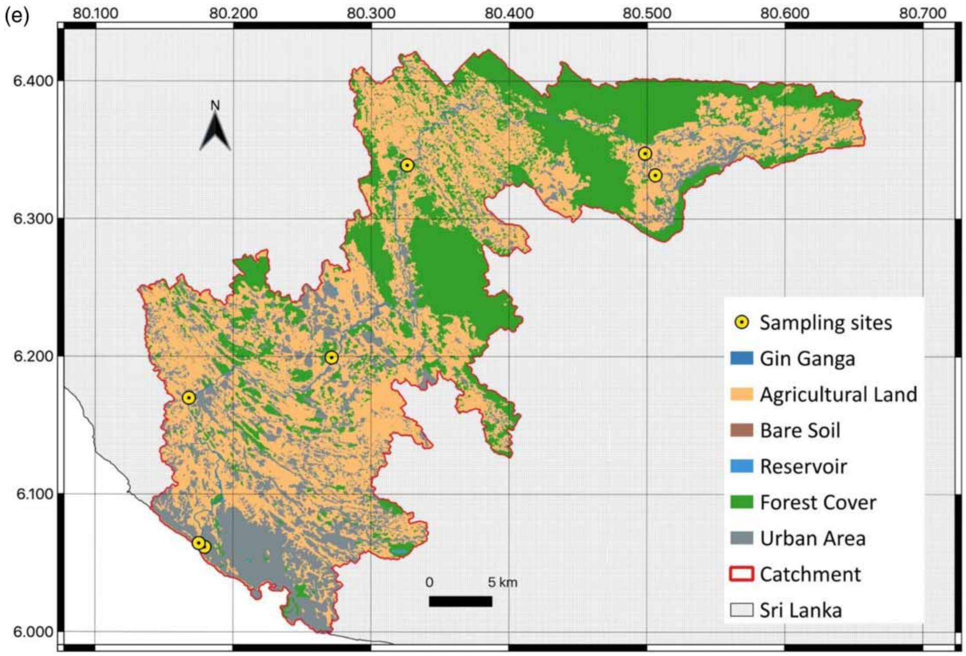

Figure 2 | (Continued).

Table 3 | Physicochemical properties of water samples taken from three tributaries

River	Site	pH	Conductivity (μS·m^{-1})	TDS (ppm)
Gin	G1 [03]	6.35 ± 0.10*	88.8 ± 1.7*	44.7 ± 0.6*
	G2 [03]	6.33 ± 0.04*	54.5 ± 0.4*	27.2 ± 0.3*
	G3 [03]	6.37 ± 0.10*	55.3 ± 0.8*	27.6 ± 0.3*
	G4 [03]	6.65 ± 0.42*	48.0 ± 2.1*	23.8 ± 1.0*
	G5 [04]	6.72 ± 0.34*	48.4 ± 0.8*	24.3 ± 0.5*
	G6 [05]	6.41 ± 0.15*	7,389.2 ± 4,301.2*	3,690.8 ± 2,121.7*
	Range	6.33–6.72*	48.0–7,389.2*	23.8–3,690.8*
	Average	6.47 ± 0.17*	1,280.7 ± 2,992.6*	639.7 ± 1,494.7*
Mahaweli	M1 [01]	6.23 ± 0.06*	66.6 ± 2.0*	33.7 ± 0.5*
	M2 [02]	6.27 ± 0.06*	69.4 ± 1.5*	34.6 ± 0.5*
	M3 [03]	6.72 ± 0.26*	168.5 ± 2.5*	84.4 ± 1.0*
	M4 [04]	7.13 ± 0.16*	128.3 ± 2.3*	64.0 ± 1.2*
	M5 [04]	7.03 ± 0.13*	151.1 ± 6.6*	75.7 ± 3.3*
	M6 [04]	6.83 ± 0.14*	123.8 ± 5.2*	61.9 ± 2.4*
	M7 [05]	7.04 ± 0.09*	250.2 ± 29.0*	132.2 ± 19.8*
	Range	6.23–7.13*	66.6–250.2*	33.7–132.2*
	Average	6.75 ± 0.34*	136.8 ± 58.3*	69.5 ± 31.1*
Deduru	D1 [01]	6.37 ± 0.19*	81.0 + 21.3*	40.5 ± 10.5*
	D2 [02]	6.66 ± 0.06*	104.7 ± 2.2*	52.8 ± 1.3*
	D3 [03]	7.01 ± 0.22*	113.3 ± 37.1*	56.7 ± 18.8*
	D4 [04]	7.01 ± 0.07*	149.0 ± 4.0*	74.3 ± 2.0*
	D5 [04]	7.03 ± 0.08*	180.6 ± 3.2*	90.0 ± 1.7*
	D6 [04]	7.37 ± 0.23*	245.1 ± 10.4*	123.0 ± 4.6*
	D7 [04]	7.43 ± 0.17*	239.4 ± 19.5*	122.2 ± 6.7*
	D8 [05]	7.35 ± 0.06*	299.4 ± 5.8*	150.5 ± 5.4*
	Range	6.37–7.43*	81.0–299.4*	40.5–150.5*
	Average	7.03 ± 0.37*	176.6 ± 78.2*	88.8 ± 39.6*

Asterisk '*' indicates a value significantly different from the recommended values. A sampling site category is given in brackets '[]' (see Figure 1(b)).

Table 4 | Concentration of Pb, Cd, As and Hg of water samples collected in the selected rivers

River	Site	Pb	Cd	As	Hg
Gin	G1 [03]	0.20 ± 0.04	0.01 ± 0.01	0.16 ± 0.02	0.31 ± 0.10
	G2 [03]	0.13 ± 0.06	0	0.07 ± 0.01	0.15 ± 0.06
	G3 [03]	0.14 ± 0.01	0	0.10 ± 0.01	0.13 ± 0.02
	G4 [03]	0.11 ± 0.05	0.01 ± 0.01	0.13 ± 0.01	0.07 ± 0.03
	G5 [04]	0.16 ± 0.04	0.01 ± 0.01	0.12 ± 0.01	0.04 ± 0.01
	G6 [05]	0.01 ± 0.01	0.01 ± 0.01	0.18 ± 0.02	0.02 ± 0.01
	Range	0.01–0.20	0–0.01	0.07–0.18	0.07–0.31
	Average	0.12 ± 0.07	< 0.01	0.12 ± 0.04	0.12 ± 0.11
Mahaweli	M1 [01]	0.02 ± 0.01	0	0.03 ± 0.01	0.15 ± 0.02
	M2 [02]	0.02 ± 0.01	0	0.03 ± 0.01	0.05 ± 0.02
	M3 [03]	0.02 ± 0.01	0.01 ± 0.01	0.08 ± 0.01	0.18 ± 0.14
	M4 [04]	0.01 ± 0.01	0	0.09 ± 0.01	0.19 ± 0.15
	M5 [04]	0.02 ± 0.01	0	0.09 ± 0.01	0.05 ± 0.01
	M6 [04]	0.02 ± 0.01	0	0.10 ± 0.01	0.06 ± 0.03
	M7 [05]	0.02 ± 0.01	0	0.17 ± 0.02	0.19 ± 0.15
	Range	0.01–0.02	0–0.01	0–0.17	0–0.19
	Average	0.02 ± 0.01	< 0.01	0.08 ± 0.05	0.12 ± 0.07
Deduru	D1 [01]	0.04 ± 0.02	0	0.07 ± 0.01	0.12 ± 0.01
	D2 [02]	0.03 ± 0.01	0.01 ± 0.01	0.06 ± 0.01	0.07 ± 0.01
	D3 [03]	0.01 ± 0.01	0	0.07 ± 0.02	0.06 ± 0.02
	D4 [04]	0.03 ± 0.02	0	0.09 ± 0.01	0.06 ± 0.01
	D5 [04]	0.02 ± 0.01	0	0.16 ± 0.02	0.09 ± 0.03
	D6 [04]	0.04 ± 0.02	0	0.26 ± 0.01	0.11 ± 0.03
	D7 [04]	0.04 ± 0.01	0	0.29 ± 0.01	0.50 ± 0.17
	D8 [05]	0.03 ± 0.01	0	0.29 ± 0.03	0.13 ± 0.02
	Range	0.01–0.04	0–0.01	0.06–0.29	0.06–0.50
	Average	0.03 ± 0.01	< 0.01	0.16 ± 0.10	0.14 ± 0.15

Values are given in μg·L^{-1}. A sampling site category is given in brackets '[ ]' (see Figure 1(b)).

Interestingly, [As] was significantly higher in water samples collected near AL and UA as compared with the other land-use/cover types. With regard to the Deduru River, Pb content showed a pattern similar to that of the Mahaweli River, i.e. the measured Pb content was low. However, the mean Hg content was 0.14 ± 0.15 μg·L^{-1} and showed significant inter-site mean differences ($p < 0.05$). The Hg content in site D7 was the highest (0.50 ± 0.17 μg·L^{-1}) to be recorded in all rivers studied. The As contents were also significantly different among the sampling sites ($p < 0.05$). The mean value of As content was 0.16 ± 0.10 μg·L^{-1} and the variation among the sites followed an increasing trend as follows: [As]D1 = [As]D2 = [As]D3 = [As]D4 < [As]D5 < [As]D6 = [As]D7 = [As]D8. Similarly, in the Mahaweli River, [As] was significantly higher in water samples collected near AL and UA as compared with the other land-use/cover types.

With regard to the Gin River, it showed significant inter-site mean differences in the contents of Pb, As and Hg ($p < 0.05$). The mean concentration of Pb was 0.12 ± 0.07 μg·L^{-1}. The Hg content in site G1 (0.31 μg·L^{-1}) was the highest and then decreased and remained stable along the river (flow direction) with [Hg] G1 > [Hg] G2, G3, G4, G5 and G6. The mean As content was 0.12 ± 0.04 μg·L^{-1} and the differences of As among the sampling sites (to the flow direction) can be described as follows: [As] G1 > [As] G2 < [As] G3 = [As] G4 = [As] G5 < [As] G6. Therefore, the As content was significantly higher ($p < 0.05$) in the water samples collected near AL and UA than FC, while the Hg content was higher in the upper catchment. In summary, the mean contents of the HMs, considering all rivers, were observed in the order: [Hg] > [As] > [Pb] > [Cd] on site category 01 and [As] > [Hg] > [Pb] > [Cd] on site category 05. Further, categories 03 to 05 showed the highest metal concentration in As and Hg.

When the correlation matrix was considered (Figure 3), the Pb content showed a significant positive correlation coefficient with the distance of river margins covered by the urban area (coefficient = 0.285, $p = 0.0246$). However, no linear model could be fitted. The As content was positively correlated with the percentage cover of AL in the catchment (coefficient = 0.479, $p = 0.0325$). The percentage FC, on the other hand, showed a negative significant correlation with the As content (coefficient = –0.526, $p = 1.389325 \times 10^{-5}$) and the exponential regression model could be more accurately fitted (adjusted $R^2 = 0.618$, $p = 4.036 \times 10^{-5}$).

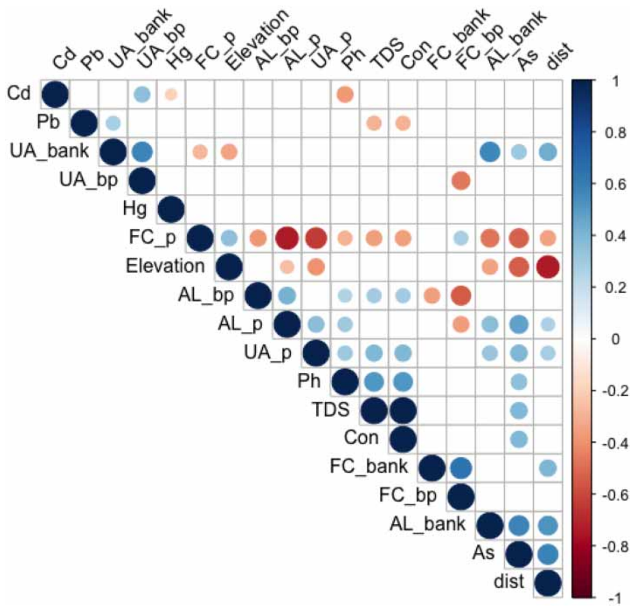

Figure 3 | Correlation matrix between HMs and land-use/cover. Cd: cadmium; Pb: lead; Hg: mercury; As: arsenic; UA: urban area; FC: forest cover; AL: agricultural land; UA_p: percentage value of urban area; UA_banks: river margin values of urban area: UA_pb: UA percentage of river margin values.

DISCUSSION

It is apparent that the landscape of the selected catchments has been extensively modified by human activities. As a result of this, ALs have become ubiquitous and predominant in the catchment areas (Gunawardhana *et al.* 2016). However, among the studied rivers, only the catchment of the Mahaweli River has a relatively large FC, which can be attributed to the fact that several areas present in the basin are well protected. However, the FC in the upper catchment has been considerably decreased due to agriculture, developmental activities and human settlements (Hewawasam 2010). This trend is common for the three studied rivers. Therefore, such kinds of anthropogenic impacts more likely result in the release of sewage, wastes and pollutants in the environment in diverse forms. These contaminations could be released from point sources such as industry effluents or from more diffuse sources such as AL run-off (Gunawardhana *et al.* 2016).

High conductivity values could be linked with sea water contamination at the river mouth. Given the fact that TDS is an indirect measure of human impact, high values could reflect the high quantity of sediments that are added to the rivers. The reasons for this external input could be land preparation for agriculture, house constructions or run-off during heavy rain. Accordingly, the fact that both TDS and conductivity increased along the Mahaweli and the Deduru Rivers towards the flow direction could be explained by the increase of water pollution due to the aforementioned factors. ALs are present on around half (50.5%) of the Gin river surface catchment and the results of Amarathunga & Kazama (2016) confirmed that the upper basin showed important FC and ALs. According to their land-cover results, the majority of the ALs in the upper catchment were dedicated to tea plantation, which is considered to be less damaging to water quality by way of preventing soil erosion. On the contrary, the Gin River had a different trend, with both TDS and conductivity remaining at low levels (except on sea water-contaminated sites) despite strong human pressure. The linear relationship between TDS and conductivity found in our results was confirmed by WHO (2017). Thus, the Gin River could be possibly used as a good reference site in making comparisons. In addition, the pH values of the Gin River were quite stable. Furthermore, Amarathunga & Kazama (2016) found similar values, supporting the fact that the pH remained stable along the Gin River. Differences shown by the Gin River could be linked to different rainfall patterns. However, none of the rivers showed the water quality parameters such as TDS or conductivity above the maximum recommended value by the WHO and SLS.

More importantly, the amount of dissolved solids is important in determining HMs since it may serve a high surface area to bind HMs. In this respect, the rivers in areas impacted by chronic kidney disease, namely, the Mahaweli and the Deduru Rivers, have a higher affinity towards HMs than the Gin River due to higher TDS. Since the HM contents are not at a risk level, running freshwater from the selected rivers can be considered safe enough to be used for drinking purposes. Moreover, one of the WHO guidelines supports the fact that the concentration of Cd in water is likely to become a health concern in environments where the pH is less than 4.5, which is not the case in the selected rivers. Supporting the results of this study, various other studies also found Cd concentration within normal limits in the rivers of NCP (Diyabalanage *et al.* 2016) and more specifically in the Gin River (Amarathunga & Kazama 2016). Our analyses showed that As was present at a level below the limit in each river and similar values were reported by Jayatilake *et al.* (2013). As received greater attention since it has been pointed out as a possible cause of chronic kidney disease in Sri Lanka after this metalloid was found in urine and hair samples in great quantity among 68% of CKD-affected patients (Jayasumana *et al.* 2013). However, the studies carried out by Chandrajith *et al.* (2011) and Jayatilake *et al.* (2013) reported greater concentrations than we did, which are still below the standard limits. When looking at the prevailing concentration of As in river systems, it is not a hazardous causative at this stage. Low levels of Pb reported in our study were further confirmed by Bandara *et al.* (2011) and Jayatilake *et al.* (2013). With regard to Hg, very low concentrations were recorded showing that these rivers are not much polluted with industrial waste. Our land-use/land-cover results have supported other results to prove that chemical industries or similar industries are not located within the UAs that are in the neighbourhood of the rivers studied. Moreover, Perera *et al.* (2016) showed that the HM concentrations of As and Cd were impacted by the rainfall regimes and, therefore, were fluctuating according to the season. It is also likely that the leaching rate of pollution is modified with the rate of precipitation. Unfortunately, studies that have been conducted in Sri Lanka rarely specify the dates of sample collection, and, thus, it is not possible to compare the results of these studies with previous results.

According to recent estimation, only a small proportion of the rural population still uses spring water directly from selected streams for drinking purposes. Since HM concentrations reported in this study are far below the recommended value given by the standard guidelines, drinking freshwater directly from streams/rivers does not impose a high health risk. Further, Herath *et al.* (2018) sampled more than 1,435 wells in the NCP where chronic kidney disease is highly prevalent and found that the levels of As, Cd and Pb were still below the WHO and the SLS set values. Thus, it is clear that not only the major rivers but also many associated wells/reservoirs are not remarkably contaminated with HMs.

Human daily water consumption increases in hot dry climates (Dissanayake 1996) and this may lead to the accumulation of a higher concentration of HMs than the expected levels. Under such conditions, trace elements can become harmful to several living organisms, including humans, when present in high quantities (Fernández-Luqueño *et al.* 2013; Huang *et al.* 2009; Van Der Hoek *et al.* 2003). In humans, especially As, Cd, Hg and Pb are are not easily biodegradable and hence accumulate in their vital organs, and this can develop to progressive toxicity (Alam *et al.* 2003). Therefore, a low level of HM in water does not always offer complete protection from HM-related diseases in the long run.

According to recent investigations, high contents of HMs were found in different phosphate fertilizers that are produced mainly from sedimentary phosphate minerals rich in trace elements, for example, As, Cd, Pb, Hg and U (Godt *et al.* 2006; Dissanayake & Chandrajith 2009). In addition, the most widely used herbicide, namely glyphosate, has been identified as a major source of HMs, which is able to destroy renal tissues when forming a complex with HMs (Gunatilake *et al.* 2019). An HM content assessment carried out with more than 450 fertilizer and pesticide samples apparently showed that agrochemicals are a major source of inorganic As in areas affected by chronic kidney disease (Jayasumana *et al.* 2015). We suggest that the correlation between As and the AL cover percentage in the catchment observed could be linked to the use of excess agrochemicals. Therefore, it is encouraged to carry out research to study the HM contents and availability in river sediments, the results of which could perhaps be different from the results of river water. Therefore, low levels of HMs at river sources clearly support the fact that human activities are the main cause of HM pollution. However, it is highly recommended to study specific sources of HMs that can be found in the neighbourhood of the studied rivers, particularly hazardous ones that are directly drained off from ALs and industries. Such a study will indeed be useful in tracing possible health issues arising out of polluted water. In addition to this, isotope analysis can be used in tracing the exact sources.

The level of HM pollution has not reached the level of risk that can create a chronic effect in the long run. Therefore, in order to prevent the extent and spread of health complications in the country arising from drinking water sources, we propose a few low-cost and low-technology methods to purify water and suggest to communities to modify their eating habits. The development of a political strategy for agrochemical management and reduction is strongly recommended. In addition, awareness programs are still essential to educate the people about the means of HM pollution and how to minimize the impacts of it.

In Sri Lanka, there are a few pollution prevention acts such as the National Environmental Act (NEA) No: 56 of 1980 amended in 1988 and 2000. This aims to protect each and every aspect of the environment and is used to make provisions for the prevention, abatement and control of pollution. The Marine Pollution Prevention Act no: 35 of 2008, which was established by the marine environment protection authority (MEPA), aims to prevent, reduce, control and manage pollution arising out of ship-based activities and shore-based maritime-related activities in the territorial waters of Sri Lanka or in any other maritime zone of Sri Lanka.

CONCLUDING REMARKS

Apparently, the HM contents of As, Cd, Pb and Hg in the Mahaweli, Deduru and Gin Rivers are not at a risk level at this point and, therefore, the waters are safe enough to be used for drinking purposes. However, the trend of the increasing levels of As, Hg and Pb from the source to the river mouth direction indicates that the sources of HM pollution originate from the surrounding environment. With a high fidelity, it is suggested that the significant differences observed in As concentration could be due to increased agricultural activities. In addition, urbanization might be linked to the rise of HMs in river waters in nearby UAs. We further suggest that the application of agrochemicals in ALs, mainly paddy fields, is the non-point source of these metal species, and the combination of their presence in diets could lead to many health issues.

*Limit of quantification for four different metal ions are as follows:

	Water samples (LoQ) – ppm	Water samples with sediments (LoQ) – ppm
Pb	0.001	0.01
As	0.001	0.001
Cd	0.001	0.001
Hg	0.001	0.0005

NIST traceable calibrations standard was used for the calibration, and this analysis was carried out at the ISO 17025 accredited facility (Residual Analysis Laboratory – RAL, Industrial Technology Institute – ITI Sri Lanka) for the above HMs. Equipment used was ICP MS – Agilent 7900.

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DATA AVAILABILITY STATEMENT

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

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