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RESEARCH ARTICLE

Ecotoxicology

Risk assessment of heavy metals in the freshwater lake sediments around Eppawala phosphate deposit, Sri Lanka

NM Batapola[1], NP Dushyantha[2*], NP Ratnayake[1], HMR Premasiri[1], H Panagoda[1], CL Jayawardena[1], R Chandrajith[3], IMSK Ilankoon[4], LPS Rohitha[1], AS Ratnayake[2], JB Koongolla[5], AMBK Abeysinghe[1], DMDOK Dissanayake[1] and RMP Ratnayake[1]

[1] *Department of Earth Resources Engineering, Faculty of Engineering, University of Moratuwa, Moratuwa, Sri Lanka.*
[2] *Department of Applied Earth Sciences, Faculty of Applied Sciences, Uva Wellassa University, Passara Road, Badulla, Sri Lanka.*
[3] *Department of Geology, Faculty of Science, University of Peradeniya, Peradeniya, Sri Lanka.*
[4] *School of Engineering, Monash University Malaysia, Selangor, Malaysia.*
[5] *Key Laboratory of Tropical Marine Bio-resources and Ecology, South China Sea Institute of Oceanology, Chinese Academy of Science, Guangzhou, China.*

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Abstract: The Eppawala area in Sri Lanka has an agricultural-based economy. As a result, the recent agricultural intensification could increase the risk of heavy metal contamination in lakes in the area as the main water canal in the area, *i.e.*, Jaya Ganga, flows across these lakes. Therefore, this study focuses on the risk assessment of heavy metals in the freshwater lake sediments in the Eppawala area and the identification of potential sources for heavy metal contamination in the lakes. Nine heavy metals (Cr, Mn, Co, Ni, Cu, Zn, As, Cd, and Pb) were investigated in surface sediments (n = 22) of the upstream and downstream lakes of the Eppawala Phosphate Deposit (EPD). The average heavy metal concentrations in the upstream lake sediments were higher than those downstream. Eppawala lake sediments were heavily polluted by As along with moderate to high Cr pollution. However, only As and Cd indicated considerable to moderate ecological risk levels to the local environment. The downstream lake sediments showed lower heavy metal contents compared to those upstream and had negative correlations between heavy metals and P_2O_5 contents. This reveals that the EPD does not contribute to the heavy metal contamination in the Eppawala lake sediments. However, the statistical analysis showed that heavy metals were mostly derived from similar pollution sources. Agrochemicals used in paddy cultivation in the vicinity might be a potential source of heavy metals. This study highlights the importance of implementing remediation to control the heavy metal pollution prevailing in the Eppawala lakes.

Keywords: Agricultural intensification, ecological risks, Eppawala, heavy metals, sediments.

INTRODUCTION

Heavy metals in aquatic ecosystems are a pressing global concern due to their adverse characteristics, such as toxicity, persistence, non-biodegradability, and bioaccumulation (Dai *et al*., 2018). In this context, heavy metal pollution in freshwater lakes has acquired significant attention as lakes are responsible for providing numerous ecosystem services (*e.g.*, water cycling, climate regulation, and habitats for aquatic flora, fauna, and microorganisms) and play a vital role in human lifestyle (*i.e.*, providing water for irrigation, drinking, and other various purposes) (Thevenon *et al*., 2013; Jahromi *et al*., 2021). Lake sediments are crucial indicators for monitoring heavy metal contamination levels in lakes since they act as both carriers and sinks for heavy metals (Li *et al*., 2013; Jin *et al*., 2021). Heavy metals get easily absorbed and accumulated in the lake sediments due to their low solubility. However, the settled heavy metals in the lake sediments could remobilize to water under various changing environmental conditions, such as pH,

* Corresponding author (nimila.dush@uwu.ac.lk; https://orcid.org/0000-0002-3871-6851)

temperature, and redox potential. It causes secondary contamination of heavy metals in the water column and aquatic organisms, posing severe risks to the ecology of the environment and to human health (Malferrari *et al*., 2009; Varol, 2020).

Heavy metals are introduced into lakes either by natural sources, such as geological weathering, volcanic eruptions, and airborne dust, or anthropogenic sources including agricultural activities (application of fertilizers/pesticides), smelting/processing of metal ores, mining, sewage discharge, industrial effluents, and urban construction (Magni *et al*., 2021). Typically, agricultural activities are overlooked as a source of heavy metal contamination in lake sediments due to relatively low content of heavy metals in agricultural runoffs (*e.g.*, Cd, Ni, and Pb contents in Wagon Train agricultural watershed, Nebraska are about 0.00045, 0.0063 and 0.0013 mg/L) (Elrashidi *et al*., 2007) compared to other anthropogenic sources, especially industrial discharges (*e.g.*, Cd, Ni, and Pb contents in an iron and steel industrial effluent from India are about 0.051, 0.303 and 0.082 mg/L) (Ladwani *et al*., 2012). However, long-term usage of agrochemicals in agricultural lands may lead to excessive accumulations of heavy metals in close-by waterbodies (Tang *et al*., 2014). Furthermore, rapid population growth and the scarcity of land have induced an agricultural intensification (*i.e.*, high levels of inputs (*e.g.*, agrochemicals, water, labour, etc.) or outputs (*e.g.*, harvest, plant growth, etc.) per unit area of land), which is significant to increase the food security in the world but causes high heavy metal accumulations in lake sediments, posing severe impacts to ecosystems and human health (Alauddin & Quiggin, 2008).

The North-Central Province of Sri Lanka has an agriculture-based economy and a centuries-old history of paddy cultivation. In the past few decades, agriculture in Sri Lanka has been intensified with an increased quantity of agrochemicals to secure the food supply within the country. However, it has been carried out improperly without following the guidelines and standards imposed by the Agriculture Department of Sri Lanka (De Costa, 2021). Eppawala in the North-Central Province is such an area where intensified agriculture is being carried out. Consequently, the ecological environment in lakes adjacent to the agricultural fields is vulnerable to heavy metal contamination through fertilizer/pesticide leaching.

Therefore, three lakes, namely Koon, Ihalahalmilla, and Kiralogama were selected to investigate the heavy metal contamination in lake sediments of the Eppawala area. In addition, these three lakes are adjacent to the Eppawala Phosphate Deposit (EPD) and since phosphate deposits contain significant amounts of heavy metals (*e.g.*, the concentrations of Cd, Cr, Ni, V, U, and Zn in the Al-Jiza phosphate deposit of Central Jordan are 15 ± 8, 109 ± 21, 34 ± 6, 211 ± 55, 142 ± 55, and 161 ± 57 mg/kg, respectively) (Al-Hwaiti *et al*., 2014; Siddique *et al*., 2018), the EPD could be a natural source for heavy accumulation in the lake sediments via geological weathering. The specific objectives of this research are (1) investigating the spatial distribution of heavy metals in surface lake sediments of the Eppawala area; (2) assessing the degree of contamination and evaluating the potential ecological risk to the environment; and (3) identifying the possible natural and anthropogenic sources for heavy metal contamination in the lake sediments.

MATERIALS AND METHODS

Study area

The EPD is the only phosphate mine in Sri Lanka with an annual production of 50,000 tonnes of rock phosphate fertilizers (Dushyantha *et al*., 2019; 2020). It lies in the Eppawala area of the North-Central province, which belongs to the Wanni Complex (Figure 1) of Sri Lanka. Moreover, a man-made water canal named 'Jaya Ganga' flows across the EPD connecting these three lakes and it distributes water over an area of 470 km^2 while feeding about 46 km^2 of paddy fields and 120 small lakes (Dushyantha, 2018). In addition, the lifestyle of people in this area is closely related to these lakes through fishing, irrigation, and agriculture.

Based on the location of the EPD, the present study area is divided into upstream (Ihalahalmilla and Koon lakes) and downstream (Kiralogama lake) regions (Figure 1b) (Dushyantha *et al*., 2017; 2021). The lakes are located in the dry climatic zone of Sri Lanka, and thus water levels of the lakes are raised dramatically during the rainy season, particularly in the north-eastern monsoon (December to February). However, during the dry season, most of the lake water volume is drastically reduced while turning some lakes into huge grasslands.

Sample collection and preparation

A total of 22 surface lake sediment samples were collected from the upstream (n = 12) and downstream (n = 10) lakes by using a Van Veen grab sampler (Figure 2). All the sediment samples were oven-dried at 105˚C for 24 hours to remove the moisture and powdered using an agate mortar and a pestle. The pulverized samples were then sieved using a 63 µm sieve to obtain undersize fractions of the samples. Finally, representative samples were prepared by the coning and quartering method for further analyses.

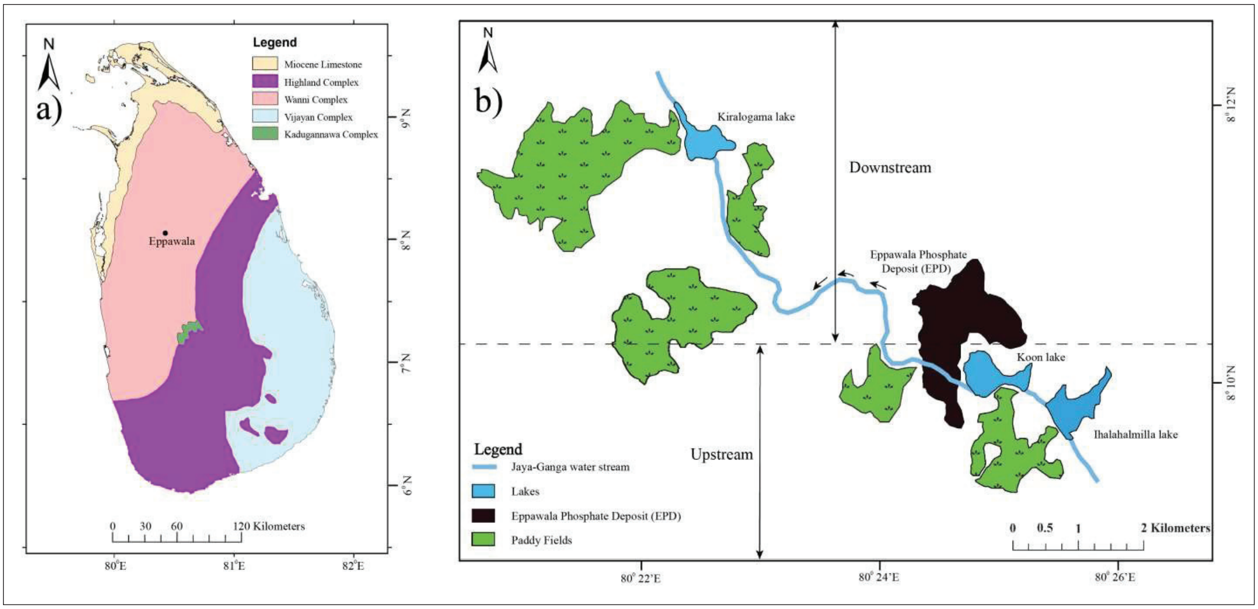

Figure 1: (a) The simplified geological map of Sri Lanka with major lithotectonic complexes (after Cooray, 1984); (b) the study area illustrating Ihalahalmilla, Koon, and Kiralogama lakes along with the Eppawala phosphate deposit (EPD) (Dushyantha *et al*., 2019).

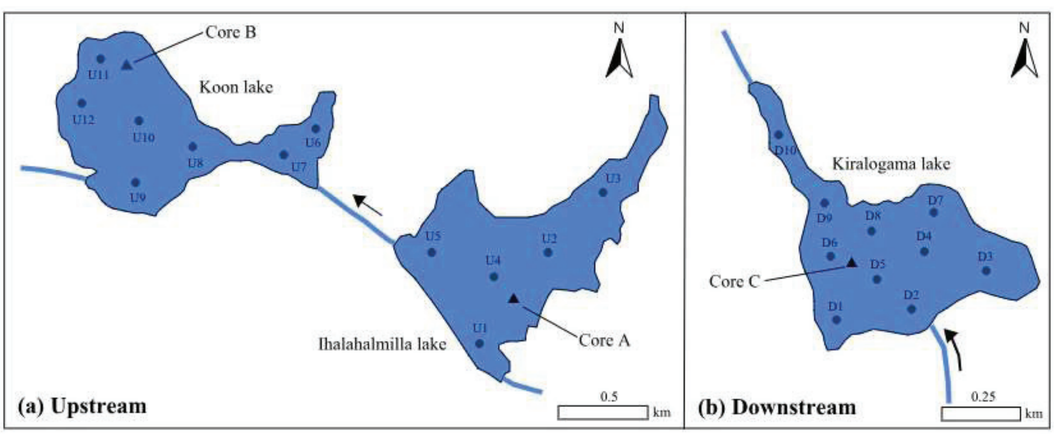

Figure 2: The surface sediment sampling locations at Eppawala (a) upstream lakes; (b) downstream lakes

Heavy metal analysis

Representative samples were digested using a mixture of HCl, HNO_3, and H_2O_2 with a ratio of 1:3:1 in a MARS-6 microwave digester (CEM; Mathews, NC) equipped with EasyPrep Plus high-pressure vessels. The digested samples were then diluted with de-ionized water and analyzed for heavy metals (Cr, Mn, Co, Ni, Cu, Zn, As, Cd, and Pb) using a Thermo ICapQ Inductively Coupled Plasma Mass Spectrometer (ICP-MS) (Thermo Fisher, Bremen, Germany), with the use of multi-elemental ICP-MS standards (Sigma-Aldrich, Germany). In addition, the certified international reference samples and blanks were used to control the accuracy and precision of the analysis.

Assessment of sediment contamination and potential ecological risk

It is not ideal to identify and compare the individual heavy metal contamination levels in the environment solely by their concentrations since the crustal abundances of each heavy metal are different. Therefore, some indices such as the geoaccumulation index are used to assess the actual contamination of heavy metals in the environment. In addition, the potential ecological risk index is used to determine the ecological risks posed by heavy metals to the aquatic environment.

Geoaccumulation index

The geoaccumulation index (I_{geo}) is used to verify the magnitude of the contamination caused by an individual heavy metal (Varol, 2011) (equation 1).

$$I_{geo} = \mathrm{Log}_2 \frac{C_s^i}{1.5\, C_n^i} \qquad ...(1)$$

C_s^i is the concentration of the element i in the lake sediment sample, C_n^i is the geochemical background concentration of the element i, and n is the number of heavy metals analyzed ($n = 9$). However, due to the absence of background data on heavy metals in the Eppawala lakes, the upper continental crust (UCC) values were used as the reference values (Wedepohl, 1995). The constant 1.5 is the background matrix correction factor due to lithospheric effects.

Potential ecological risk index

The potential ecological risk index (RI) is a quantitative measurement of the degree of ecological risks caused by the heavy metals in aquatic sediments (Hakanson, 1980) and it is obtained by equations 2 and 3.

$$E_r^i = T_r^i \times C_f^i = T_r^i \times \frac{C_s^i}{C_n^i} \qquad ...(2)$$

$$RI = \sum_{i=1}^{n} E_r^i \qquad ...(3)$$

E_r^i is the potential ecological risk factor of the heavy metal i and T_r^i is the toxic response factor of the element i, while C_f^i is the contamination factor of the element i. C_s^i and C_n^i are similar to the parameters defined in Equation 1. T_r^i for Cr, Mn, Co, Ni, Cu, Zn, As, Cd, and Pb are 2, 1, 5, 5, 5, 1, 10, 30, and 5, respectively (Hakanson, 1980).

RESULTS AND DISCUSSION

Heavy metal contents in surface lake sediments

The heavy metal concentrations in all the upstream and downstream surface lake sediment samples are presented in Table S1, whereas Table 1 shows the corresponding range and average values. According to the Table 1, the heavy metal concentrations showed a decreasing order of Mn > Cr > Zn > Ni > Cu > Co > As > Pb > Cd in the upstream surface lake sediments and Mn > Cr > Zn > Cu >Ni > As > Co > Pb > Cd in the downstream surface lake sediments. The average concentrations of all the heavy metals analysed in this study were high in the upstream surface lake sediments compared to the respective downstream values (Table 1). Therefore, it is evident that there are no point sources of heavy metals between the upstream and downstream lakes. However, based on Dushyantha *et al*. (2019, 2020), there is a phosphate-rich sediment contribution from the EPD to the downstream

Table 1: Ranges and averages (mg/kg) of heavy metals in the upstream and downstream, surface lake sediments

Heavy metal	Upstream		Downstream	
	Range	Avg.	Range	Avg.
Cr	90.42 – 427.34	263.0	69.58 – 394.38	241.3
Mn	187.61 – 2706.96	1420.7	176.62 – 2212.94	1141.1
Co	5.87 – 49.89	32.6	5.50 – 43.86	23.8
Ni	33.64 – 273.63	97.5	20.43 – 120.46	53.2
Cu	10.12 – 108.00	72.5	5.91 – 101.90	63.6
Zn	40.78 – 449.30	186.4	49.73 – 213.46	133.7
As	2.81 – 46.77	25.1	14.14 – 33.08	24.9
Cd	0.07 – 2.19	0.5	0.01 – 0.67	0.2
Pb	3.48 – 26.23	14.7	3.47 – 24.81	12.9

Avg.: Average

lake sediments, and the subsequent dilution could be the cause for the respective low contents of heavy metals. In this study, Mn displayed the highest concentration levels in the surface lake sediments. Generally, Mn is ubiquitous in the earth's crust (*e.g.*, basalt - 1300 mg/kg, gneiss - 600 mg/kg, and limestone - 550 mg/kg) and thus, it is found in higher concentrations in surface lake sediments compared to other heavy metals (Queiroz *et al*., 2021).

Table 2 presents the average heavy metal concentrations of surface lake sediments in this study compared to a few other lakes in Sri Lanka and other countries in the world. Compared to the local studies, reservoirs in Anuradhapura, Polonnaruwa, and Kilinochchi districts have reported higher concentrations of heavy metals in sediments than in the present study. Since Anuradhapura and Polonnaruwa are agriculture-based areas, especially for paddy cultivation, the extensive use of agrochemicals may be the possible reason for these high concentrations (Wijesinghe *et al*., 2018; Perera *et al*., 2021). However, heavy metal concentrations in the Bolgoda lake sediments were relatively low compared to the present study. In the global context, the reported high heavy metal concentrations of surface lake sediments in India, Egypt, and China may be due to local industrial developments, urbanization, and intense agricultural activities in the area (Suresh *et al*., 2012; Ma *et al*., 2016b; Ji *et al*., 2019). In contrast, most of the heavy metal concentrations in the surface lake sediments of Lake St. Clair and Hope Lake in the USA were found to be the lowest compared to the other lakes (Table 2). The low concentration levels in these lakes are possibly due to strict legislative frameworks in developed countries that control and monitor heavy metal discharges and emissions from industrial, urban, and agricultural sources (Dai *et al*., 2018). Therefore, legislative frameworks on heavy etals would alleviate the heavy metal pollution in freshwater lakes.

Table 2: A comparison of average heavy metal concentrations in surface lake sediments (mg/kg) in this study and a few other lakes in the world

Lake		Cr	Mn	Co	Ni	Cu	Zn	As	Cd	Pb
Local										
Eppawala	Upstream	263.0	1420.7	32.6	97.5	72.5	186.4	25.1	0.5	14.7
Lake	Downstream	241.3	1141.1	23.8	53.2	63.6	133.7	24.9	0.2	12.9
Mahakanadarawa, Anuradhapura [1]		1842	NA	NA	NA	744	1346	25.6	0.0	138.0
Iranamadu, Kilinochchi [1]		355.0	NA	NA	NA	169.0	758.0	27.3	0.0	221.0
Mahadiwulwewa, Anuradhapura [1]		1836	NA	NA	NA	207.0	894.0	188.0	26.3	231.0
Bolgoda Lake,	North Lake	109.5	NA	NA	NA	33.8	130.6	NA	2.3	36.8
Colombo [2]	South Lake	119.7	NA	NA	NA	13.4	61.8	NA	1.9	26.5
Minneriya Reservoir, Polonnaruwa [3]		97.3	NA	NA	NA	105.0	67.0	NA	105.0	722.0
Parakrama Samudraya, Polonnaruwa [3]		64.3	NA	NA	NA	55.0	41.0	NA	66.0	845.3
Global										
Veeranam Lake, India [a]		88.2	NA	NA	63.6	94.1	180.1	NA	0.8	30.1
Dongting Lake, South China [b]		70.2	781.0	NA	33.0	30.2	121.6	4.5	0.8	34.1
Baiyangdian Lake, North China [c]		30.1- 6.0	NA	NA	22.0- 44.0	16.1-204.0	41.6-263.0	5.3-24.3	0.2-2.5	25.3-99.3
Lake St. Clair, USA [d]		8.6	NA	NA	10.1	11.6	40.0	5.9	<1.0	7.9
Hope Lake, USA [e]		37.8	3903.4	20.1	38.6	21.0	128.0	NA	0.4	17.1
Lake Nasser, Egypt [f]		30.8	279.6	NA	27.6	21.8	35.4	NA	0.2	10.9

NA - Not Available

[1] (Perera *et al*., 2021); [2] (Senarathne & Pathiratne, 2007); [3] (Wijesinghe *et al*., 2018)

[a] (Suresh *et al*., 2012); [b] (Ma *et al*., 2016a); [c] (Ji *et al*., 2019); [d] (Gewurtz *et al*., 2007); [e] (López *et al*., 2010); [f] (Goher *et al*., 2014)

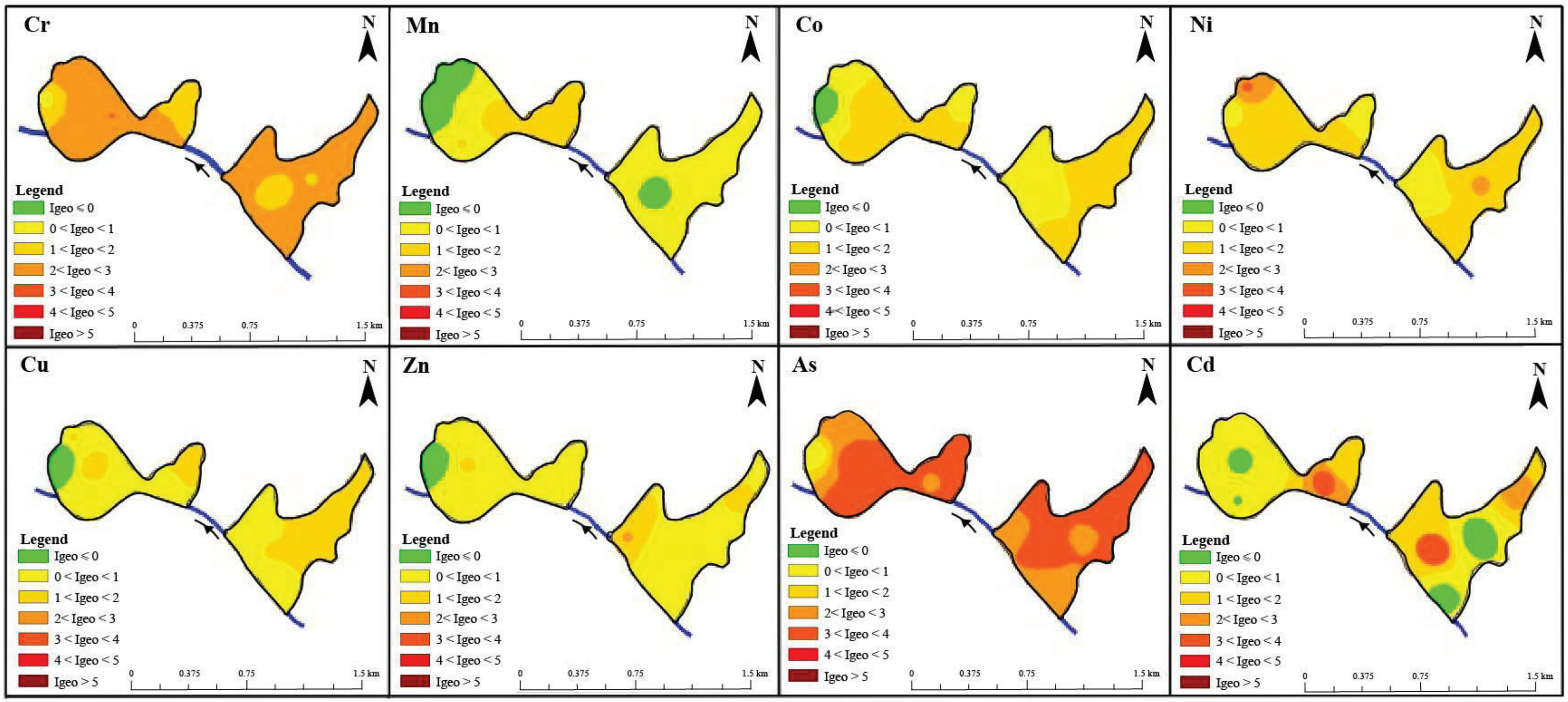

Figure 3: The spatial distribution of I_{geo} of heavy metals in the upstream surface lake sediments of the Eppawala area: $I_{geo} \leq 0$: practically unpolluted; $0 < I_{geo} < 1$: unpolluted to moderately polluted; $1 < I_{geo} < 2$: moderately polluted; $2 < I_{geo} < 3$: moderately to heavily polluted; $3 < I_{geo} < 4$: heavily polluted; $4 < I_{geo} < 5$: heavily to extremely polluted; $I_{geo} > 5$: extremely polluted

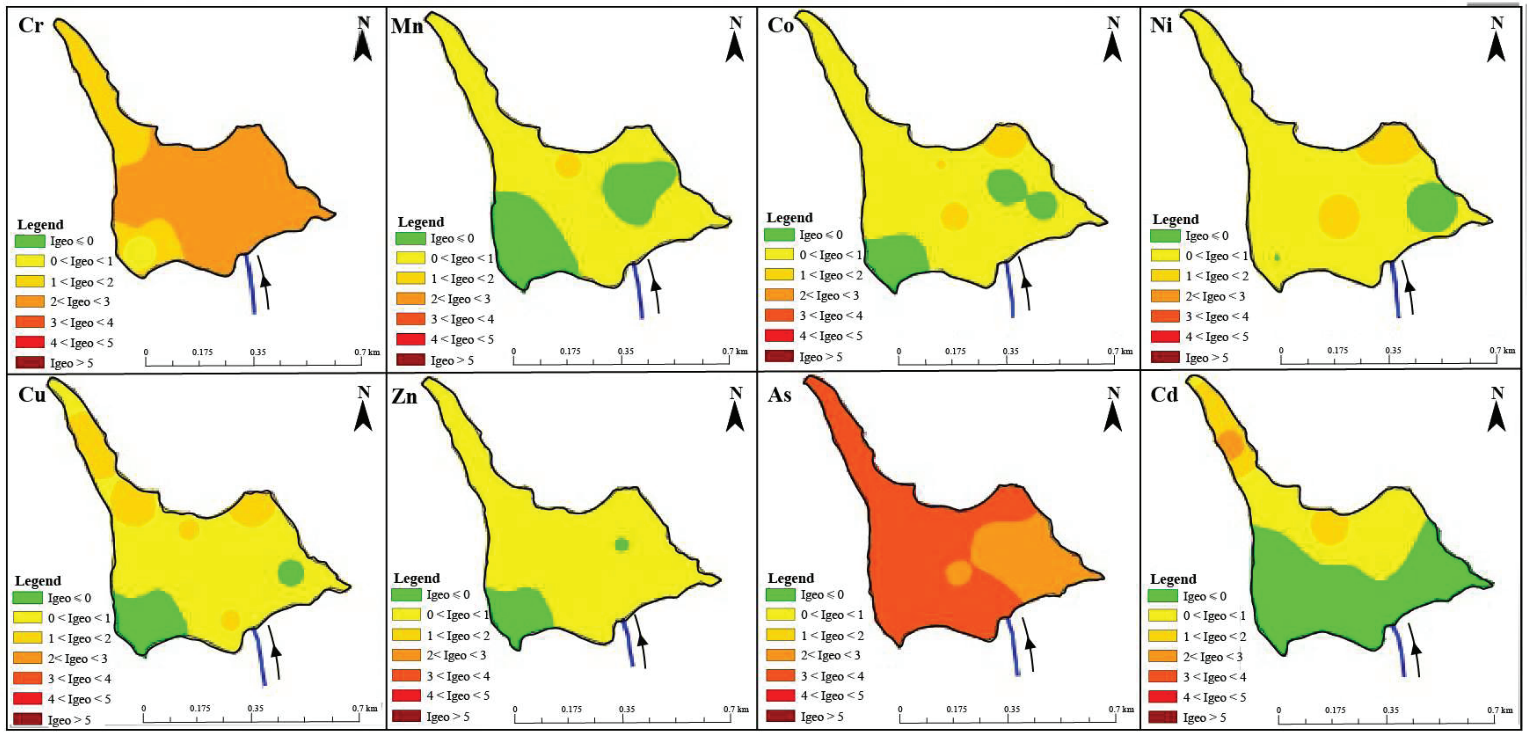

Figure 4: The spatial distribution of I_{geo} of heavy metals in the downstream surface lake sediments of the Eppawala area: $I_{geo} \leq 0$: practically unpolluted; $0 < I_{geo} < 1$: unpolluted to moderately polluted; $1 < I_{geo} < 2$: moderately polluted; $2 < I_{geo} < 3$: moderately to heavily polluted; $3 < I_{geo} < 4$: heavily polluted; $4 < I_{geo} < 5$: heavily to extremely polluted; $I_{geo} > 5$: extremely polluted

Assessment of heavy metal contamination

The classification criteria of I_{geo} are shown in Table S2. The spatial distributions of I_{geo} of each heavy metal in the upstream and downstream surface lake sediments are illustrated in Figures 3 and 4, respectively. These figures indicate that both the upstream and downstream surface lake sediments were heavily polluted by As (I_{geo} = -0.09–3.96) and moderately to heavily polluted by Cr (I_{geo} = 0.41–3.02). However, only the upstream surface lake sediments were moderately contaminated by Ni (I_{geo} = 0.46–3.19) and Co (I_{geo} = 0.10–1.73). Other heavy metals (Mn, Cu, Zn, Cd, and Pb) generally displayed unpolluted or unpolluted to moderately polluted levels in the Eppawala surface lake sediments

On the other hand, Mn, Cu, Zn, and Cd showed relatively high contamination levels in several locations at both the upstream and downstream areas (Figures 3 and 4). This heavy metal accumulation could be related to the sedimentation patterns of the lakes, and the nature of sedimentation depends on the grain size (Liang *et al*., 2019). The mean grain size distribution of the same samples in the Eppawala surface lake sediments was obtained from Dushyantha *et al*. (2019). However, Figure 5 illustrates no significant correlation between heavy metals and grain size. It elucidates that the grain size did not affect the degree of heavy metal accumulation in the Eppawala surface lake sediments, thus a local point source could be the reason for the arbitrarily high contamination levels of Mn, Cu, Zn, and Cd.

Table 3: Average potential ecological risk factors in the Eppawala surface lake sediments

Heavy metals	Upstream avg. potential ecological risk factor (E^i_r)	Downstream avg. potential ecological risk factor (E^i_r)
Cr	15.03	13.79
Mn	2.37	1.90
Co	16.30	11.82
Ni	24.38	13.31
Cu	14.49	12.71
Zn	2.63	1.88
As	125.84	124.65
Cd	137.93	66.06
Pb	3.66	3.21

Potential ecological risks

Table 3 presents the average potential ecological risk factors (E^i_r) of individual heavy metals in both the upstream and downstream surface lake sediments. E^i_r values of the heavy metals in the upstream surface lake sediments varied in the following order: Cd > As > Ni > Co > Cr > Cu > Pb > Zn > Mn. By contrast, E^i_r values of the heavy metals in the downstream surface lake sediments followed the order of As > Cd > Cr > Ni > Cu > Co > Pb > Mn > Zn. According to E^i_r classification criteria (Table S3), As and Cd posed considerable to moderate ecological risk levels in the surface lake sediments of the

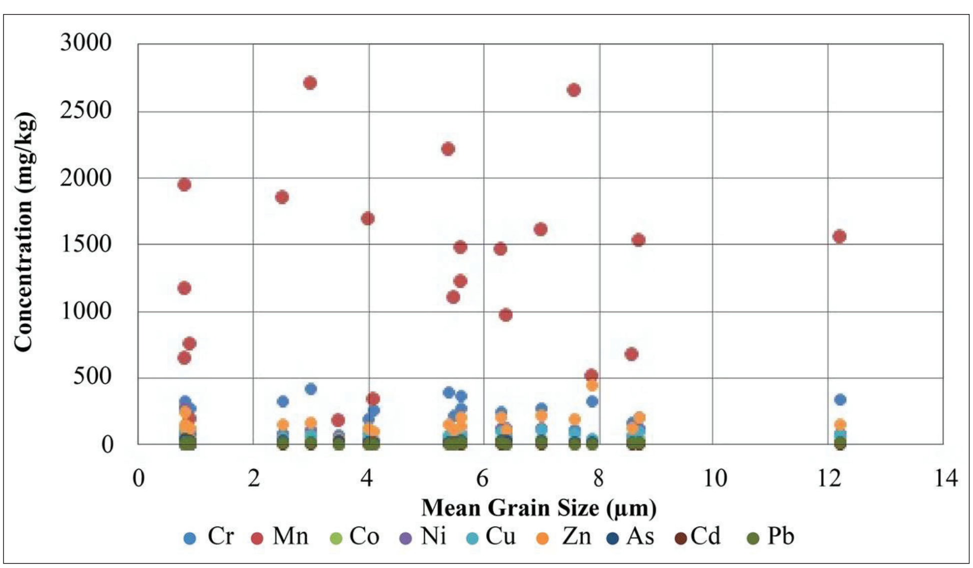

Figure 5: The variation of heavy metals with the grain size of Eppawala surface lake sediments

Eppawala area, whereas other heavy metals remained at low-risk levels. The potential ecological risks caused by As in both the upstream and downstream surface lake sediments were consistent with the I_{geo} results obtained for As (Figures 3 and 4). However, Cd showed a considerable to moderate ecological risk level despite its low degree of contamination in the surface lake sediments (Figures 3 and 4). Furthermore, despite high to moderate contamination levels of Cr, Ni, and Co in both the upstream and downstream surface lake sediments (Figures 3 and 4), they did not pose any considerable threat to the ecological environment in the Eppawala area.

The potential ecological risk of heavy metals in sediments depends on both their contents and their speciation. There are six categories of speciation of heavy metals in sediments, namely (1) exchangeable (EXC), (2) bound to carbonate (CARB), (3) bound to easily reducible oxides (ERO), (4) bound to organic matter (OM), (5) bound to residual oxides (RO), and (6) residual fraction (RES-R). Under changing environmental conditions, such as pH or redox potential, these categories behave differently with respect to remobilization. Therefore, the reason for having ecological risk levels that were inconsistent with the results of I_{geo} for Cd, Cr, Ni, and Co may be due to their speciation in the Eppawala surface lake sediments. For example, if the percentage of CARB-Cd in the Eppawala surface lake sediments were high, Cd could be released into water under low pH conditions, causing high pollution to the surrounding environment (Yang *et al*., 2009).

Figure 6 shows the spatial distribution of RI values in both the upstream and downstream surface lake sediments, which represents the overall ecological risks posed by the investigated heavy metals in the Eppawala surface lake sediments (Equation 3). The potential ecological risk levels of heavy metals in the upstream surface lake sediments were considerable compared to the downstream surface lake sediments, which had moderate risk levels. Since the heavy metal accumulation in the upstream surface lake sediments was higher, heavy metals may have yielded comparatively greater levels of ecological risk to the local upstream environment.

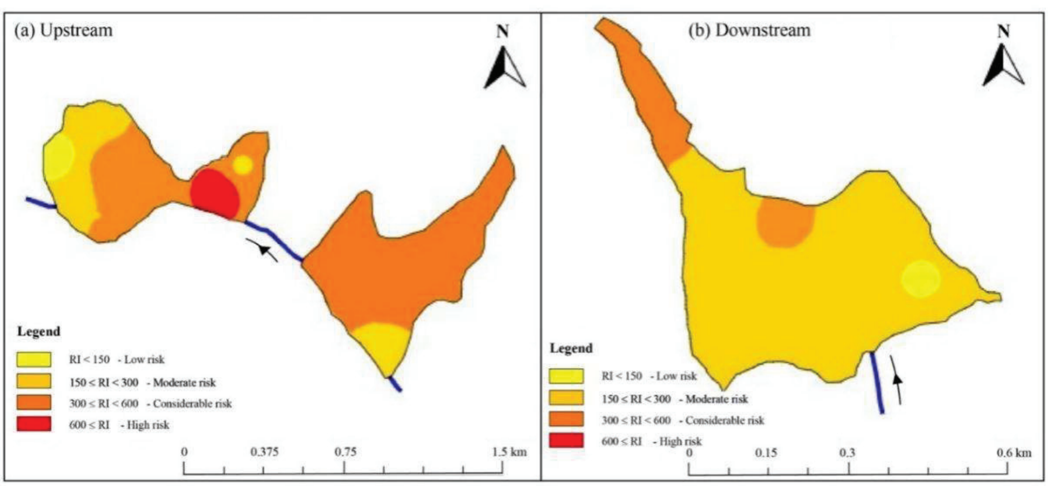

Figure 6: The spatial distribution of RI in the (a) upstream and (b) downstream surface lake sediments of the Eppawala area

Potential sources of heavy metals

Pearson's correlation coefficient matrix of heavy metals and phosphate (P_2O_5) content in the Eppawala lake sediments was derived (Table 4) since it reveals the potential sources and pathways in the environment. The P_2O_5 content of these samples was taken from Dushyantha *et al*. (2019). In the upstream surface lake sediments, significant and positive correlations were observed between Co and Cu (r = 0.773), Co and Pb (r = 0.836), Cu and As (r = 0.701), Cu and Pb (r = 0.748), and As and Pb (r = 0.765), whereas Co and As showed only a moderate positive correlation (r = 0.595). Since only Cu, As, and Pb were significantly intercorrelated, they may have derived from a similar pollution source, whereas Co could have a different origin or controlling factors in the upstream surface lake sediments (Tang *et al*., 2014).

Table 4: The correlation coefficient (r) matrix (n = 12) for heavy metals in the Eppawala surface lake sediments

		Cr	Mn	Co	Ni	Cu	Zn	As	Cd	Pb	P_2O_5
Upstream	Cr	1									
	Mn	0.267	1								
	Co	0.526	0.658	1							
	Ni	0.417	-0.062	0.384	1						
	Cu	0.132	0.409	**0.773**	0.324	1					
	Zn	0.341	-0.077	0.065	-0.135	0.199	1				
	As	0.339	0.373	0.595	-0.039	**0.701**	0.392	1			
	Cd	-0.048	0.155	0.016	-0.169	0.011	-0.019	0.065	1		
	Pb	0.363	0.417	**0.836**	-0.006	**0.748**	0.274	**0.765**	0.061	1	
	P_2O_5	-0.309	-0.511	-0.598	0.168	-0.624	-0.457	-0.648	-0.393	**-0.724**	1
		Cr	Mn	Co	Ni	Cu	Zn	As	Cd	Pb	P_2O_5
Downstream	Cr	1									
	Mn	0.578	1								
	Co	0.517	0.608	1							
	Ni	0.306	0.371	**0.890**	1						
	Cu	0.427	0.756	**0.833**	0.663	1					
	Zn	0.515	0.675	**0.952**	**0.852**	**0.792**	1				
	As	0.062	0.165	0.353	0.428	0.421	0.129	1			
	Cd	0.070	0.300	0.075	-0.065	0.431	0.113	0.074	1		
	Pb	0.523	0.223	**0.701**	**0.808**	0.372	**0.713**	0.246	-0.419	1	
	P_2O_5	0.063	0.062	0.080	-0.041	0.072	-0.116	0.571	-0.121	-0.101	1

r > 0 - positive correlation; r < 0 - negative correlation; r > ±0.7 - significant correlation (highlighted in bold)

In contrast, Co and Ni (r = 0.890), Co and Zn (r = 0.952), Ni and Zn (r = 0.852), Ni and Pb (r = 0.808), Zn and Pb (r = 0.713), and Pb and Co (r = 0.701) in the downstream surface lake sediments showed significant positive correlations. It indicated that Co, Ni, Zn, and Pb were positively correlated among themselves in the downstream surface lake sediments, thus they may be discharged from a similar pollution source. However, negative and very low positive correlations among P_2O_5 and the heavy metals suggested that the phosphate-bearing materials accumulated in these lake sediments might not be a source for heavy metals.

The application of agrochemicals (*i.e.*, pesticides and chemical fertilizers) to the regional paddy cultivations may be a potential source of As, Pb, and Cu. It is also supported by the field investigations as intense agricultural activities, especially paddy cultivations were observed in the vicinity of these three lakes. According to Jayasumana *et al*. (2015), the most commonly used chemical fertilizers in Sri Lanka contain significantly high concentrations of As, Pb, and Cu (*e.g.*, Triple Superphosphate (TSP): As = 26.5–31.9 mg/kg, Pb = 251.7–263.9 mg/kg and Cu = 14.2–16.0 mg/kg, and Muriate of Potash (MOP): As = 0.2–0.4 mg/kg, Pb = 0.8–0.9 mg/kg and Cu = 0.3–0.4 mg/kg). Moreover, despite being illegal to import, As-containing agrochemicals available in Sri Lanka contain considerable levels of As (*e.g.*, Glyphosate: 0.9–2.6 mg/kg, Dimethoate: 1.0–2.4 mg/kg, and Fenoxaprop-p-ethyl: 1.2–2.6 mg/kg) (Jayasumana *et al*., 2015). Therefore, extensive usage of TSP and pesticides in the local paddy fields may be a potential source of As contamination in the Eppawala lake sediments. However, further studies are recommended, especially for heavy metal contamination in the agricultural fields and runoffs in the vicinity of these lakes, to determine the impact of agricultural activities in the region on the observed heavy metal pollution in the Eppawala lake sediments.

CONCLUSIONS

The present study demonstrated that both the upstream and downstream surface lake sediments of the Eppawala area were moderately to heavily contaminated by As and Cr, whereas Co and Ni showed moderate contamination levels only in the upstream area. However, based on the RI analysis, only As and Cd displayed considerable to moderate ecological risk levels in the surface lake sediments, in which the highest potential ecological risk was caused by As. Based on the correlation coefficient relationships of heavy metals, the possible pollution source for heavy metals might be the intense agrochemical use in paddy cultivation. The average heavy metal concentrations were higher in the upstream lake sediments compared to the downstream, and this, together with the negative correlation between heavy metals and P_2O_5 content, suggested that the weathered materials from the EPD could not be a potential source for the heavy metal contamination in lakes. The results of this research reveal important findings that can be used to recommend proper ecological management of aquatic ecosystems while controlling the heavy metal pollution in the Eppawala lakes.

Conflict of interest statement

The authors declare that there is no conflict of interest.

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REFERENCES

Abrahim G.M.S. & Parker R.J. (2008). Assessment of heavy metal enrichment factors and the degree of contamination in marine sediments from Tamaki Estuary, Auckland, New Zealand. *Environmental Monitoring and Assessment* **136**: 227–238.
DOI: https://doi.org/10.1007/s10661-007-9678-2

Al-Hwaiti M., Al Kuisi M., Saffarini G. & Alzughoul K. (2014). Assessment of elemental distribution and heavy metals contamination in phosphate deposits: potential health risk assessment of finer-grained size fraction. *Environmental Geochemistry and Health* **36**: 651–663.
DOI: https://doi.org/10.1007/s10653-013-9587-y

Alauddin M. & Quiggin J. (2008). Agricultural intensification, irrigation and the environment in South Asia: Issues and policy options. *Ecological Economics* **65**: 111–124.
DOI: https://doi.org/10.1016/j.ecolecon.2007.06.004

Chai L., Li H., Yang Z., Min X., Liao Q., Liu Y., Men S., Yan Y. & Xu J. (2017). Heavy metals and metalloids in the surface sediments of the Xiangjiang River, Hunan, China: Distribution, contamination, and ecological risk assessment. *Environmental Science and Pollution Research* **24**: 874–885.
DOI: https://doi.org/10.1007/s11356-016-7872-x

Cooray P.G. (1984). *An Introduction to the Geology of Sri Lanka (Ceylon)*. Department of National Museums, Colombo, Sri Lanka.

Dai L., Wang L., Li L., Liang T., Zhang Y., Ma C. & Xing B. (2018). Multivariate geostatistical analysis and source identification of heavy metals in the sediment of Poyang Lake in China. *Science of the Total Environment* **621**: 1433–1444.
DOI: https://doi.org/10.1016/j.scitotenv.2017.10.085

De Costa W.A.J.M. (2021). *'Fertilizer Saga' in Sri Lanka: A Considered Opinion*. Available at *https://island.lk/fertilizer-saga-in-sri-lanka-a-considered-opinion/.*

Dushyantha N.P. (2018). Alternative phosphorus sources in lake bottom sediments around Eppawala phosphate deposit in Sri Lanka. *M.Phil thesis*, University of Moratuwa, Sri Lanka.

Dushyantha N.P., Hemalal P.V.A., Jayawardena C.L., Ratnayake A.S., Premasiri H.M.R. & Ratnayake N.P. (2017). Nutrient characteristics of lake sediments around Eppawala phosphate deposit, Sri Lanka. *Journal of Geological Society of Sri Lanka* **18**(2): 33–42.

Dushyantha N.P., Hemalal P.V.A., Jayawardena C.L., Ratnayake A.S. & Ratnayake N.P. (2019). Application of geochemical techniques for prospecting unconventional phosphate sources: A case study of the lake sediments in Eppawala area Sri Lanka. *Journal of Geochemical Exploration* **201**: 113–124.
DOI: https://doi.org/10.1016/j.gexplo.2019.02.010

Dushyantha N., Batapola N., Ilankoon I.M.S.K., Rohitha S., Premasiri R., Abeysinghe B., Ratnayake N. & Dissanayake K. (2020). The story of rare earth elements (REEs): Occurrences, global distribution, genesis, geology, mineralogy and global production. *Ore Geology Reviews* **122**:.103521.
DOI: https://doi.org/10.1016/j.oregeorev.2020.103521

Dushyantha N., Ratnayake N., Panagoda H., Jayawardena C. & Ratnayake A.S. (2021a). Phosphate mineral accumulation in lake sediment to form a secondary phosphate source: A case study in lake sediment around Eppawala phosphate deposit (EPD) in Sri Lanka. *International Journal of Sediment Research* **36**: 532–541.

DOI: https://doi.org/10.1016/j.ijsrc.2020.12.001

Dushyantha N.P., Ratnayake N.P., Premasiri H.M.R., Ilankoon I., Hemalal P.V.A., Jayawardena C.L., Chandrajith R., Rohitha L.P.S., Abeysinghe A. & Dissanayake D. (2021b). Leaching of rare earth elements (REEs) from lake sediments around Eppawala phosphate deposit, Sri Lanka: A secondary source for REEs. *Hydrometallurgy* **205**: 105751. DOI: https://doi.org/10.1016/j.hydromet.2021.105751

Elrashidi M.A., Hammer D., Fares A., Seybold C.A., Ferguson R. & Peaslee S.D. (2007). Loss of heavy metals by runoff from agricultural watersheds. *Soil Science* **172**: 876–894. DOI: https://doi.org/10.1097/ss.0b013e31814cec7b

Gewurtz S.B., Helm P.A., Waltho J., Stern G.A., Reiner E.J., Painter S. & Marvin C.H. (2007). Spatial distributions and temporal trends in sediment contamination in Lake St. Clair. *Journal of Great Lakes Research* **33**: 668–685. DOI: https://doi.org/10.3394/0380-1330(2007)33[668:SDATTI]2.0.CO;2

Goher M.E., Farhat H.I., Abdo M.H. & Salem S.G. (2014). Metal pollution assessment in the surface sediment of Lake Nasser, Egypt. *Egyptian Journal of Aquatic Research* **40**: 213–224. DOI: https://doi.org/10.1016/j.ejar.2014.09.004

Hakanson L. (1980). An ecological risk index for aquatic pollution control. A sedimentological approach. *Water Research* **14**: 975–1001. DOI: https://doi.org/10.1016/0043-1354(80)90143-8

Jahromi F.A., Keshavarzi B., Moore F., Abbasi S., Busquets R., Hooda P.S. & Jaafarzadeh N. (2021). Source and risk assessment of heavy metals and microplastics in bivalves and coastal sediments of the Northern Persian Gulf, Hormogzan Province. *Environmental Research* **196**: 110963. DOI: https://doi.org/10.1016/j.envres.2021.110963

Jayasumana C., Fonseka S., Fernando A., Jayalath K., Amarasinghe M., Siribaddana S., Gunatilake S. & Paranagama P. (2015). Phosphate fertilizer is a main source of arsenic in areas affected with chronic kidney disease of unknown etiology in Sri Lanka. *Springer Plus* **4**: 1–8. DOI: https://doi.org/10.1186/s40064-015-0868-z

Ji Z., Zhang H., Zhang Y., Chen T., Long Z., Li M. & Pei Y. (2019). Distribution, ecological risk and source identification of heavy metals in sediments from the Baiyangdian Lake, Northern China. *Chemosphere* **237**: 124425. DOI: https://doi.org/10.1016/j.chemosphere.2019.124425

Jin C., Li Z., Huang M., Wen J., Ding X., Zhou M. & Cai C. (2021). Laboratory and simulation study on the Cd (II) adsorption by lake sediment: Mechanism and influencing factors. *Environmental Research* **197**: 111138. DOI: https://doi.org/10.1016/j.envres.2021.111138

Ladwani K.D., Ladwani K.D., Manik V.S. & Ramteke D.S. (2012). Impact of industrial effluent discharge on physico-chemical characteristics of agricultural soil. *International Research Journal of Environment Sciences* **1**: 32–36.

Li F., Huang J., Zeng G., Yuan X., Li X., Liang J., Wang X., Tang X. & Bai B. (2013). Spatial risk assessment and sources identification of heavy metals in surface sediments from the Dongting Lake Middle China. *Journal of Geochemical Exploration* **132**: 75–83. DOI: https://doi.org/10.1016/j.gexplo.2013.05.007

Liang J., Liu J., Xu G. & Chen B. (2019). Distribution and transport of heavy metals in surface sediments of the Zhejiang nearshore area, East China Sea: sedimentary environmental effects. *Marine Pollution Bulletin* **146**: 542–551. DOI: https://doi.org/10.1016/j.marpolbul.2019.07.001

López D.L., Gierlowski-Kordesch E. & Hollenkamp C. (2010). Geochemical mobility and bioavailability of heavy metals in a lake affected by acid mine drainage: Lake Hope, Vinton County, Ohio. *Water, Air, and Soil Pollution* **213**: 27–45. DOI: https://doi.org/10.1007/s11270-010-0364-6

Ma R., Wang B., Lu S., Zhang Y., Yin L., Huang J., Deng S., Wang Y. & Yu G. (2016a). Characterization of pharmaceutically active compounds in Dongting Lake, China: occurrence, chiral profiling and environmental risk. *Science of the Total Environment* **557**: 268–275. DOI: https://doi.org/10.1016/j.scitotenv.2016.03.053

Ma X., Zuo H., Tian M., Zhang L., Meng J., Zhou X., Min N., Chang X. & Liu Y. (2016b). Assessment of heavy metals contamination in sediments from three adjacent regions of the Yellow River using metal chemical fractions and multivariate analysis techniques. *Chemosphere* **144**: 264–272. DOI: https://doi.org/10.1016/j.chemosphere.2015.08.026

Magni L.F., Castro L.N. & Rendina A.E. (2021). Evaluation of heavy metal contamination levels in river sediments and their risk to human health in urban areas: A case study in the Matanza-Riachuelo Basin, Argentina. *Environmental Research* **197**: 110979. DOI: https://doi.org/10.1016/j.envres.2021.110979

Malferrari D., Brigatti M.F., Laurora A. & Pini S. (2009). Heavy metals in sediments from canals for water supplying and drainage: mobilization and control strategies. *Journal of Hazardous Materials* **161**: 723–729. DOI: https://doi.org/10.1016/j.jhazmat.2008.04.014

Perera W., Dayananda M., Dissanayake D., Rathnasekara R., Botheju W.S.M., Liyanage J.A., Weragoda S.K. & Kularathne K.A.M. (2021). Risk assessment of trace element contamination in drinking water and agricultural Soil: a study in selected chronic kidney disease of unknown etiology (CKDu) endemic areas in Sri Lanka. *Journal of Chemistry* **2021**: 6627254. DOI: https://doi.org/10.1155/2021/6627254

Queiroz H.M., Ying S.C., Abernathy M., Barcellos D., Gabriel F.A., Otero X.L., Nobrega G.N., Bernardino A.F. & Ferreira T.O. (2021). Manganese: The overlooked contaminant in the world largest mine tailings dam collapse. *Environment International* **146**: 106284. DOI: https://doi.org/10.1016/j.envint.2020.106284

Senarathne P. & Pathiratne K.A.S. (2007). Accumulation of heavy metals in a food fish, Mystus gulio inhabiting

Bolgoda Lake, Sri Lanka. *Sri Lanka Journal of Aquatic Sciences* **12** : 61–75.
DOI: https://doi.org/10.4038/sljas.v12i0.2214

Siddique A., Hassan A., Khan S.R., Inayat A., Nazir A. & Iqbal M. (2018). Appraisal of heavy metals and nutrients from phosphate rocks, Khyber Pakhtunkhwa, Pakistan. *Chemie International* **4**: 1.

Suresh G., Sutharsan P., Ramasamy V. & Venkatachalapathy R. (2012). Assessment of spatial distribution and potential ecological risk of the heavy metals in relation to granulometric contents of Veeranam lake sediments, India. *Ecotoxicology and Environmental Safety* **84**: 117–124.
DOI: https://doi.org/10.1016/j.ecoenv.2012.06.027

Tang W., Ao L., Zhang H. & Shan B. (2014). Accumulation and risk of heavy metals in relation to agricultural intensification in the river sediments of agricultural regions. *Environmental Earth Sciences* **71**: 3945–3951.
DOI: https://doi.org/10.1007/s12665-013-2779-z

Thevenon F., de Alencastro L.F., Loizeau J-L., Adatte T., Grandjean D., Wildi W. & Poté J. (2013). A high-resolution historical sediment record of nutrients, trace elements and organochlorines (DDT and PCB) deposition in a drinking water reservoir (Lake Brêt, Switzerland) points at local and regional pollutant sources. *Chemosphere* **90**: 2444–2452.
DOI: https://doi.org/10.1016/j.chemosphere.2012.11.002

Varol M. (2011). Assessment of heavy metal contamination in sediments of the Tigris River (Turkey) using pollution indices and multivariate statistical techniques. *Journal of Hazardous Materials* **195**: 355–364.
DOI: https://doi.org/10.1016/j.jhazmat.2011.08.051

Varol M. (2020). Environmental, ecological and health risks of trace metals in sediments of a large reservoir on the Euphrates River (Turkey). *Environmental Research* **187**: 109664.
DOI: https://doi.org/10.1016/j.envres.2020.109664

Wedepohl K.H. (1995). The composition of the continental crust. *Geochimica et Cosmochimica Acta* **59**: 1217–1232.
DOI: https://doi.org/10.1016/0016-7037(95)00038-2

Wijesinghe H., Idroos F.S. & Manage P.M. (2018). Heavy metal contamination status in seven fish species from reservoirs of Polonnaruwa district, Sri Lanka. *Sri Lanka Journal of Aquatic Sciences* **23**(1): 95-104.
DOI: https://doi.org/10.4038/sljas.v23i1.7550

Yang Z., Wang Y., Shen Z., Niu J. & Tang Z. (2009). Distribution and speciation of heavy metals in sediments from the mainstream, tributaries, and lakes of the Yangtze River catchment of Wuhan, China. *Journal of Hazardous Materials* **166**: 1186–1194.
DOI: https://doi.org/10.1016/j.jhazmat.2008.12.034

Zhu X., Ji H., Chen Y., Qiao M. & Tang L. (2013). Assessment and sources of heavy metals in surface sediments of Miyun Reservoir, Beijing. *Environmental Monitoring and Assessment* **185**: 6049–6062.
DOI: https://doi.org/10.1007/s10661-012-3005-2

Supplementary Information

Table S1: Concentrations, ranges, and averages of heavy metals in the upstream and downstream surface lake sediments in the Eppawala area

Heavy metal	Upstream													
	U1	U2	U3	U4	U5	U6	U7	U8	U9	U10	U11	U12	Range (mg/kg)	Average (mg/kg)
Cr	343.8	208.5	277.5	168.2	324.8	115.4	264.1	427.3	325.4	282.7	327.8	90.4	(90.42- 427.34)	263.0
Mn	1556.0	1525.0	1611.1	669.4	515.3	2655.3	1949.0	2707.0	1857.0	1165.1	651.0	187.6	(187.61-2706.96)	1420.7
Co	38.8	44.6	49.9	21.9	16.0	29.2	34.2	46.7	33.5	40.9	29.5	5.9	(5.87-49.89)	32.6
Ni	83.4	131.5	120.0	56.0	34.4	33.6	99.2	111.8	87.4	98.1	273.6	41.2	(33.64-273.63)	97.5
Cu	70.5	95.5	108.0	77.6	43.1	91.5	63.3	67.9	57.2	108.0	77.0	10.1	(10.12-108.00)	72.5
Zn	159.0	203.5	217.2	121.4	449.3	193.7	132.8	161.4	149.6	249.4	158.6	40.8	(40.78-449.30)	186.4
As	22.4	23.6	31.9	25.0	23.7	26.1	23.9	24.2	35.7	46.8	16.0	2.8	(2.81-46.77)	25.1
Cd	0.1	0.1	0.7	1.0	0.6	0.3	2.2	0.3	0.1	0.1	Bdl	Bdl	(0.07- 2.19)	0.5
Pb	18.8	22.2	25.1	13.8	10.3	10.0	13.5	18.8	13.6	26.2	3.5	Bdl	(3.48-26.23)	14.7

Table S1: Concentrations, ranges, and averages of heavy metals in the upstream and downstream surface lake sediments in the Eppawala area

Downstream											
D1	D2	D3	D4	D5	D6	D7	D8	D9	D10	Range (mg/kg)	Average (mg/kg)
69.6	371.6	220.4	256.0	267.7	276.3	245.2	394.4	190.2	121.3	(69.58- 394.38)	241.3
176.6	1226.2	1103.3	336.8	1478.9	759.8	1464.5	2212.9	1687.4	964.1	(176.62- 2212.94)	1141.1
5.5	25.5	14.2	10.6	35.8	27.2	43.9	30.5	23.6	19.7	(5.50- 43.86)	23.8
29.8	57.3	20.4	35.7	76.7	59.3	120.5	51.0	41.1	40.7	(20.43- 120.46)	53.2
5.9	80.2	34.5	37.7	75.1	52.7	101.9	79.5	85.0	83.1	(5.91- 101.90)	63.6
49.7	136.4	107.6	102.1	206.1	123.8	213.5	155.7	123.6	118.1	(49.73- 213.46)	133.7
28.6	33.1	14.1	16.0	20.8	27.1	30.0	26.2	25.9	27.5	(14.14- 33.08)	24.9
0.0	0.1	0.1	0.3	0.1	0.0	0.2	0.5	0.2	0.7	(0.01- 0.67)	0.2
8.1	20.5	11.1	10.2	19.4	14.4	24.8	10.5	6.2	3.5	(3.47- 24.81)	12.9

*Bdl – Below detection limit

Table S2: Classification criteria for the degree of heavy metal contamination by geo-accumulation index (I_{geo}) (Abrahim & Parker, 2008; Ma *et al*., 2016)

I_{geo}	Class	Quality of sediment
$I_{geo} \leq 0$	0	Practically unpolluted
$0 < I_{geo} < 1$	1	Unpolluted to moderately polluted
$1 < I_{geo} < 2$	2	Moderately polluted
$2 < I_{geo} < 3$	3	Moderately to heavily polluted
$3 < I_{geo} < 4$	4	Heavily polluted
$4 < I_{geo} < 5$	5	Heavily to extremely polluted
$I_{geo} > 5$	6	Extremely polluted

Table S3: Potential ecological risk classification criteria for heavy metal contamination (Chai *et al*., 2017; Zhu *et al*., 2013)

Potential ecological risk factor (E^i_r)	Class	Level of single heavy metal ecological risk	Potential ecological risk index (RI)	Class	Level of total potential ecological risk
$E^i_r < 40$	1	Low risk	RI < 150	1	Low risk
$40 \leq E^i_r < 80$	2	Moderate risk	150 ≤ RI < 300	2	Moderate risk
$80 \leq E^i_r < 160$	3	Considerable risk	300 ≤ RI < 600	3	Considerable risk
$160 \leq E^i_r < 320$	4	High risk	RI ≥ 600	4	Very high risk
$E^i_r \geq 320$	5	Very high risk			