



# Toxic elements contamination in surface sediments of major tanks in Anuradhapura district; A CKDu endemic district in Sri Lanka

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# Abstract

Farming occupants are the major victims of Chronic Kidney Disease of unknown etiology (CKDu) in North Central region, Sri Lanka. Tanks are the main source of water for farming and they act as sinks for agricultural run-offs. Environmental toxicants are adsorbed by bottom sediments and can be released into the adjacent water column due to environmental changes as those are not permanently bound. Therefore, this study attempts to determine the contamination situation of the selected toxic metals in sediments of the tanks in a CKDu prevalence distrct (Anuradhapura) in Sri Lanka and prediction of the risk. Sixteen tanks were selected and sediment samples were randomly collected from each tank from bank to center. Digested sediment samples were analyzed for heavy metals using Inductive Couple Plasma Mass Spectrometry and Calcium and Magnesium contents were analyzed using Atomic Absorption Spectrometry. According to the statistics, average values of some toxic metals such as As, Pb, and Cr in sediments were higher than severe effect levels (SEL) while most of the mean values of Cd, As, Pb, Cr, Zn, and Cu were beyond the Lowest effect levels (LEL). Apart from that, Geo Accumulation Indexes (I<sub>geo</sub>) Indicate that almost all tanks in Anuradhapura district have moderately polluted with above mentioned pollutants. Hence, findings reveal about a risk generated on aquatic lives in the tanks as well as humans via food chain contamination with hazardous metals.

# Keywords

heavy metals; contamination; sediments; CKDu

# Introduction

Toxic elements such as heavy metals are entered into the surface water by various types of processes such as runoff, precipitation, atmospheric deposition, adsorption, point spills, etc and thereafter some of the toxic elements deposited in the sediments (Scherer et al., 2011) of different aquifers. Major reasons for the high levels of heavy metals in aquifers resulted by the economic development and growth of population (Dey et al., 2017; Hasan et al., 2007; Zhang et al., 2016). Moreover, agricultural activities may be the fundamental risk factor for the heavy metal contamination in agricultural areas. It has become a major problem, as water, soil and sediments are contaminated by various trace elements (Biswas et al., 2012; Uddin et al., 2018), which may lead to serious health impacts due to their toxicity, persistence and carcinogenic nature (Khalil et al., 2007; Naz et al., 2016; Zhu et al., 2017). Although trace elements with small quantities may help in normal functioning of the human body, excessive amounts and long term exposure to these elements may badly affect to human health (Vetrimurugan et al., 2017). Soil and sediements are heavily contaminated by pollutants from different sources such as rivers, agricultural canals, and industrial exhausts (Khan et al., 2008). However, anthropogenic sources related to the metal rich sewage sludge in agriculture, combustion, livestock manures, application of pesticides (manufacture, use, and disposal) and agricultural activities increase the pollutants which deposit in the water resources (Liu et al., 2005). Therefore, toxic metal analysis has become a subject of concern as their high persistence in the environment, ecological risk and chemical toxicity.

Previous studies reveal that Cadmium (Cd) is a major human carcinogen and excessive uptake may lead to the kidney damage, respiratory disorder and damage to the skeletal system (Bernard 2008; Belabed and Soltani 2018; Dutta et al., 2018. Apart from m that Exposure to excess amounts of Copper (Cu) causes nausea, diarrhoea and tissue injury of liver (USNRC 2000; Doza et al., 2019). As mentioned in former studies, most of the public health issues such as hypertension, cancer, vascular disease, restrictive lung disease, gastrointestinal bleeding, neurological disorder and reproductive effects may appear due to the use of contaminated water with trace metals such as Arsenic (As), Chromium (Cr), Lead (Pb) and Zinc (Zn) (Lu et al., 2015; Noubissié et al., 2016; Zhang et al., 2016; Belabed and Soltani 2018; Singh et al., 2018) for a long time with excessive amounts. Due to the requirement of large scale production of foods, agricultural practices are getting expanded throughout the world. Therefore, massive usage of pesticides and fertilizers may lead to contamination of soil, sediments, surface water and ground water (Barakat et al., 2019; Gallagher et al., 1996; Fischer et al., 2003). Water resources and soil can be contaminated with different types of contaminants due to the usage of synthetic agrochemicals in large amounts for the cultivations. Most of the toxicants which carried by the runoff may released into the existing water columns. Owing to the increase of deterioration of soil and sediment quality, assessment of sediment quality in aquifers is an important approach to determine the suitability of water resources for different purposes (Khan et al., 2008). Therefore, the study was conducted with the aims of analysis of sediment quality in major irrigation tanks in Anuradhapura district where high prevelance of Chronic kidney disease of unknown etiology (CKDu) occurs in Sri Lanka and identify of the risk related to

the toxic metal contamination. Toxic metal transport process is much important when demonstrating the heavy metal entering pathways to the human body. The transport of metals will result in increased levels of toxic metals in groundwater or surface water, thereby posing a significant environmental and health risk to the environment (Smith et al., 1996). Deposition of heavy metals occurs mainly at the bottom of areas such as streams, reservoirs and estuaries (Senesi et al, 1979). The fate of toxic metal contamination is critical because it appears to be reactive, mobile, and highly toxic.

# Materials and Methods

#### Sample collection

The total of sixteen (16) tanks were selected from Anuradhapura district in Sri Lanka including Mahakanadarawa, Lindawewa, Halmillakulama, Iranamadu, Kabithigollewa, Mahadiwulwewa, Wahalkada, Padawiya, Sadamalgama, Yakawewa, Nachchadoowa, Rajanganaya, Karuwalagaswewa, Nelugollewa, Navodhagama and Mahawilachchiya (Fig. 1) to collect sediment samples.



**Figure 1.** Distribution of the selected tanks which obtained sediment samples in Anuradhapura district, Sri Lanka.

All the locations were recorded according to the Global Positioning System (GPS) coordinates (Table 1).

Sediment samples were randomly collected from the bank to the center of the water body.

Sediment samples were obtained from Ten locations of each tank and composite samples were prepared by the combination of Five sediment samples from each location of the tank separately to ensure the sample representativeness.

	GPS Loc	ation		GN Division	
Name of Tank	Ε	Ν	Volume (m <sup>2</sup> )		
Mahakanadharawa_Tank	080.54771	08.37022	5540870	Pothana	
Lindhawewa_Tank	080.26437	08.36891	295340	Mahalindawewa	
Halmillakulama_Tank	080.21412	08.25912	406403	Halmillakulama	
Iranamadu_Tank	080.43731	09.34431	18905200	Mankulama	
Mahadiwulwewa_tank	080.51721	08.58903	664973	Mahadiulwewa	
Kebethigollewa_Tank	080.66364	08.63531	622079	Kebithigollewa	
Wahalkada_Tank	080.83713	08.72772	14934500	Wahalkada D2	
Padaviya_Tank	080.76233	08.81900	29188300	Padaviya	
Sadhamalgama_Tank	080.47546	08.42320	106956	Sandamalgama	
Yakawewa_Tank	080.39969	08.58124	No records	Yakawewa	
Naachchadhuwa_Tank	080.47030	08.25433	19991280	Nachchaduwa	
Rajanganaya_Tank	080.21471	08.13113	11909860	Rajanganaya	
Karuwalagaswewa_Tank	080.68628	08.44380	695575	Kahatagasdigiliya	
Nelugollewa_Tank	080.67986	08.48927	No records	Gonamuruwewa	
Navodhagama_Tank	080.26332	08.47509	No records	Navodagama	
Mahawilachchiya_Tank	080.19846	08.47775	4369221	Pemaduwa	

**Table 1**. GPS cordinations, volume (m<sup>2</sup>), and located Grama Niladhari (GN) división (Anuradhapura district, Sri Lanka) of the selected tanks, (Source; Ministry of Health, Sri Lanka.)

There after, samples were stored in sterile polyethylene bags and kept at 4°C during transport. At the laboratory, the samples were dried at room temperature and subsequently sieved through a 60  $\mu$ m mesh. Trace elemet analysis of the collected samples was done after 1 month from the sampling date. Triplicates from the composite sediment samples were used for the chemical analysis. All the chemical analysis was conducted in CKDu Information and Research Centre at University of kelaniya, Sri Lanka.

## Sample analysis

Heavy metal analysis of the sediment samples 0.200 g of each soil sample was digested using microwave digester (ETHOS EASY) by adding 10.00 mL of concentrated Nitric acid and digested solutions were diluted up to 25.00mL with ultrapure water and 10.00mL of each digested soil solution was filtered through 0.45 $\mu$ m nylon syringe filters and determination of metal concentrations such as Cadmium(Cd), Lead(Pb), Arsenic(As), Chromium(Cr), Copper(Cu) and Zinc(Zn ) in soil samples was done by ICP-MS technology (ICP-MS-8000-Agilent, Germany).

#### Statistical analysis

The obtained results of sediment samples were statistically analyzed by ANOVA using MINITAB software.

## Geoaccumulation index (Igeo)

Geo Accumulation value was developed for the determination of the degree of metal concentrations and the pollution which is caused by the metals in soil. The Igeo values were determined as follows (Muller, 1969):

$$I_{geo} = \log_2(C_n/1.5*B_n)$$
 [1]

where  $C_n$  is the measured concentration of the metal n in sediment,  $B_n$  is the geochemical background value of the metal n (Table 2).

In this study, the Natural Background Concentration (NBC) values were obtained from the reference table (Screening Quick Reference Table for Inorganics in Freshwater Sediment) established by the National Oceanic and Atmospheric Administration (NOAA) (Buchman 2008). Correction factor 1.5 allows to analyze natural fluctuations in the content of a given substance in the environment and to detect a very small anthropogenic influence (Espinosa et al., 2018).

le 2. Class	ses of Geo-accu	imulation index by Muller (1969).
I	I <sub>geo</sub> class	Soil/Sediment Quality
0-0	0	Unpolluted
0-1	1	Unpolluted to moderately polluted
1-2	2	Moderately polluted
2-3	3	Moderately polluted to highly polluted
3-4	4	Highly polluted
4-5	5	Highly polluted to very highly polluted
5-6	>5	Very highly polluted

 Table 2. Classes of Geo-accumulation index by Muller (1969).

# Results and Discussion

The obtained toxic element concentrations in sediment samples of the selected tanks in Anuradhapura district are listed in the Table 3. In order to evaluate the contamination due to the heavy metals and metallic elements of analyzed tank sediments, the Lowest Effect Level (LEL) and Severe Effect Level (SEL) were utilized (Pintilie et al., 2007). The LEL indicates that level of pollution has no effect on most of the species living in the reservior and the sediment or the water is proximate to slightly polluted. The SEL indicates sediment pollution stage is significant and it is likely to affect the health of sediment-dwelling species. If the level of pollution reaches the amount of severe effect then testing is required to determine whether sediment is extreamly toxic (Bala, et al., 2010).

Table 3. Mean heavy metal and metallic element concentrations in the sediments of the selected tanks in Anuradhapura.

Tank	Cd(1	ng/kg)	Pb (m	g/kg)	As (n	ng/kg)	Cr (n	ng/kg)	Cu(m	g/kg)	Zn(m	ıg/kg)
Mahakanadarawa		0	138	±_5	25.6	± 4.0	1842	± 43	744	± 32	1346	± 26
Lindhawewa		0	261	± 201	76.2	± 7.5	3594	± 533	1197	± 68	2119	± 138
Halmillakulama		0	261	± 20	64.9	± 9.0	7485	± 371	1197	± 48	1344	± 38
Iranamadu		0	221	± 24	27.3	± 4.0	355	± 38	169	± 14	758	±91
Mahadiwulwewa	26.3	± 5.8	231	±15	188	±15	1836	± 92	207	± 18	894	± 73
Kebithigollewa	7.26	± 2.15	168	±16		0	1210	± 67	258	± 49	927	± 51
Wahalkada	1.79	± 0.96	422	± 52	94.6	± 7.5	1641	± 79	530	±138	1946	± 57
Padawiya	4.62	± 1.23	338	± 37	25.0	± 4.3	4510	± 144	544	± 58	1129	± 72
Sadhamalgama		0	400	± 19	4.90	± 0.79	2299	± 158	659	± 75	1653	± 53
Yakawewa		0	280	± 36	31.5	± 4.2	3796	±284	639	± 29	2323	± 154
Naachchadhuwa		0	451	± 70	45.4	± 7.23	4529	± 203	748	± 80	1143	± 102
Rajanganaya		0	113	± 13	8.80	± 0.80	1337	± 455	171	± 12	656	± 76
Karuwalagaswewa	1.62	± 0.44	361	± 42	83.8	± 6.74	2944	± 125	742	± 47	2672	± 258
Nelugollawa		0	237	± 19	17.3	± 2.98	1545	± 85	225	± 57	456	± 17
Navodhagama		0	232	± 17	22.6	± 3.10	1267	± 162	142	± 22	1533	± 489
Mahawillachchiya	1.86	± 0.58	55.2	±3.8		0	593	± 91	180	± 30	851	± 35
LEL		0.60		35		6		37.3		35.7		123
SEL		10		250		33		110		110		820
LEL = the Lowest Effect Level - SEL = the Severe Effect Level - ND = Not Defined												

As shown in the Table 3 mean Cd levels have not been exceeded the SEL (10 mg/kg) in all the demonstrated tanks except Mahadiwulwewa where the mean Cd concentration was 26.28(±5.77) mg/kg. In addition to that, Cd contents in Kabithigollewa, Wahalkada, Padaviya, Karuwalagaswewa and Mahawilachchiya

reservoirs have reported as  $7.26(\pm 2.15)$  mg/kg,  $1.79(\pm 0.96)$  mg/kg,  $4.62(\pm 1.23)$  mg/kg,  $1.62(\pm 0.44)$  mg/kg and  $1.86(\pm 0.58)$  mg/kg respectively and show Cd levels beyond the LEL level (0.596 mg/kg). Meanwhile, almost all the selected tank sediments have exceeded the LEL (35 mg/kg) of the Pb contents while

mean Pb contents of the sediments collected from Lindhawewa (260.6( $\pm$  20.01) mg/kg, Halmillakulama (260.67( $\pm$ 20.0) mg/kg, Wahalkada (421.5( $\pm$  51.73) mg/kg, Padawiya (337.52( $\pm$  37.4) mg/kg), Sadhamalgama (399.98( $\pm$  19.3 mg/kg, Yakawewa (279.88( $\pm$ 36.4) mg/kg, Naachchadhuwa (451.44( $\pm$ 69.7) mg/kg and, Karuwalagaswewa (361.2( $\pm$  41.88) mg/kg have exceeded the SEL (250 mg/kg).

Furthermore, mean As values of sediments of six tanks Halmillakulama. (Lindhawewa, Mahadiwulwewa. Wahalkada, Naachchadhuwa and Karuwalagaswewa) have exceeded the SEL (33 mg/kg) of As. The highest mean As content was shown by the Mahadiwulwewa tank (188.24(±15.27) mg/kg). As is one of the critical metallic elements that present in all the fertilizer types in larger quantities compared to any other trace element and Previous studies (Blaise 2013, Kim et al., 2016) reveal that the metallic elements such as As can also be accumulated in fish species in the water reservoirs. When water and the sediment metal Transfer Factor (TF) is grater than 1, bioaccumination of metals such as Arsenic can occur in fish via both water and sediments. Hence, Toxic metals can also be incorporated into the food chains. (Çevik et al., 2009). Further considering the results, the mean Cr, Cu, and Zn contents in the sediments obtained from all selected tanks have exceeded the SEL. Relatively high levels of toxic metal and metallic element concentrations were recorded in selected tanks of Anuradhapura District that may be due to the agricultural activities of paddy fields. On the other side and an excessive accumulation of heavy metals in agricultural soils which lead not only to soil contamination, but also to increased crop uptake of trace metals, thus affecting food quality and safety (Muchuweti et al., 2006).

Most of the agrochemicals such as fertilizers and pesticides used in agricultural fields contain excessive amounts of toxic metals. According to the studies of (Gimeno et al., 2016) Cd concentrations in different fertilizers namely copper sulphate, iron sulphate, urea and superphosphate were 0.21 mg/kg, 0.03 mg/kg, 0.01 mg/kg and 2.22 mg/kg respectively, while mean Pb concentrations have reported as 11.0 mg/kg and 10.0 mg/kg) in copper sulphate and iron\_sulphate fertilizers respectively. Furthermore, mean Cd concetrations in Antracol, Saturn-G and 205 Ordram has reported as 1.94 mg/kg, 1.48 mg/kg, and 1.38 mg/kg respectively, while mean Pb values of above pesticides were reported as 5.0 mg/kg, 10.0 mg/kg, and 7.5 mg/kg respectively. This study has conducted in rice farming areas in

Spain (Gavrilescu, 2004). Therefore, usage of excessive amounts of fertilizers and pesticides may be a driving force to accumulation of toxic heavy metals in the sediments of tanks in the agricultural areas in Sri Lanka. Sediments serve as major sinks for heavy metals in aquatic environments, and are known as possible toxic metal sources. Furthermore, the findings of Gimeno et al., 2016 indicate that water sediment interactions in aquatic environments play an important role in the processes of bio-monitoring of accumulation of metals. According to the results of present study, some analyzed toxic metal contents of the sediments were reported in higher amounts in most of the selected tanks. Although living organisms require trace quantities of certain elements such as cobalt, copper, iron, manganese, molybdenum, vanadium, strontium, and zinc, excessive amounts of metals can be toxic to the organism. In addition, Cd, Cr, Hg, Pb, As, and Sb are non-essential heavy metals of special concern to surface water systems (Bala et al., 2010). Because of their long persistence and toxicity, toxic metals released into the aquirtic environment will harm both the diversity of fish species and the ecosystems (Persaud, 1993).

Assesement of transport and circulation of toxic metals is much significant for this study. Heavy metals can be carried by rivers that pass through of industrial and agricultural areas as soluble metals and adsorbed metals to suspended solids. Transported heavy metals can get accumulated at the bottom of the tank's sediments causing significant effects on aquatic lives and human (Monperrus et al., 2007). Apart from that Cd, Cr, Hg, Pb, As, and Sb are non-essential heavy metals of special concern to surface water systems (Khan et al., 2008). Due to the persistence of above-mentioned nonessential metals and metallic elements with elevated levels in the collected sediment samples of the tanks, a risk may be generated by transferring heavy metals in to the food chains (Zhu et al., 2017). Moreover, most of the demonstrated tanks are used for fishing industry and heavy metals can be ingested in to the body due to the consumtion of the fish.

Apart from that the geoaccumulation index (Igeo) is mainly used to assess the metal pollution in sediments to investigate the anthropogenic input to the soil or sediments (Genc and Yilmaz, 2018). In the present study, geo accumunation indexces have been calculated to evaluvate the contamination stage of the bottom sediment of the selected tanks in Anuradhapura District, Sri Lanka.

According to Muller (1969), Igeo is divided into

six categories, from uncontaminated to extremely contaminated. Obtained Igeo values (Table 4) depict that the sediments from all of the selected tanks are in the moderately contaminated stage or above the moderate level with the analyzed heavy metal and metallic elements. Considering the Igeo values obtained for Cd, Wahalkada tank and Rajanganaya tank show the highest value (1.69, 1.68) respectively.

Furthermore, Igeo values for Cr in Lindawewa, Padaviya, Yakawewa, Nachchaduwa and Karuwalagaswewa were reported Igeo values above 2.00, hence it is clear that sediments will be approaching heavier contamination with Cr, while Igeo values for As reported beyond 1 in most of the analyzed tanks except Lindawewa, Padawiya, Sadhamalgama, Yakawewa.

Studies of Espinosa 2018 reveal that the toxic metal concentrations in reservoirs mainly increase with the development activities, urbanization and

industrialization. As well as these pollutants in different acquifers are associated with natural processes and human intervention in the bio geochemical cycles of metals and metallic elements (Saleem et al., 2015). The high concentrations of the anlyzed toxic metals in the selected tanks in Anuradhapura district possibly associated with the agricultural activities because Anuradhapura is an agriculture based district. Most of the agricultural activities in these areas include the direct addition of agrochemicals over a long period of time in excessive amounts. Some toxic metals such as Cd, Cr, As, Pb, and Zn are directly related to the fertilizers and pesticides. Erosion occurred due to rain drainages initiate the migration of different harmful chemical subtstances such as heavy metals. Therefore, tanks serve as major sinks for these heavy metals, metallic elements and other chemical substances which are harmful to the human health.

	Tank	Cd	РЬ	Cr	Zn	Cu	As
1	Mahakanadarawa	0	1.64	1.71	1.55	1.70	1.62
2	Lindhawewa	0	1.63	2.71	1.71	1.66	0.47
3	Halmillakulama	0	1.63	1.72	1.67	1.68	1.60
4	Iranamadu	0	1.66	1.65	1.70	1.71	1.55
5	Mahadiwulwewa	1.51	1.59	1.70	1.59	1.61	1.39
6	Kebithigollewa	1.67	1.67	1.71	1.67	1.64	1.74
7	Wahalkada	1.69	1.61	1.70	1.69	1.71	1.41
8	Padawiya	1.09	1.53	2.34	1.54	1.72	0.60
9	Sadhamalgama	0	1.54	1.71	1.64	1.71	0.95
10	Yakawewa	0	1.64	2.13	1.64	1.71	0.96
11	Naachchadhuwa	0	1.56	2.70	1.66	1.69	1.12
12	Rajanganaya	0	1.68	1.58	1.68	1.50	1.38
13	Karuwalagaswewa	1.17	1.57	2.69	1.63	1.68	1.66
14	Nelugollawa	0	1.59	1.63	1.66	1.53	1.52
15	Navodhagama	0	1.51	1.64	1.58	1.54	1.44
16	Mahawilachchiya	0	1.64	1.60	1.43	1.60	1.54

Table 4. Geo accumulation indexes (Igeo) of the sediment of studied tanks

According the study, sediments of the selected tanks in Anuradhapura Distrit have shown the presence of toxic metals and metallic elemants. Moreover, the Geo accumilation Indexes of the tanks for selected heavy metals and metallic elements indicated that the sediments are moderately to strongly polluted as concentrations which were obtained from the present study were generally higher compared to the background values (Bn) established by the NOAA. Therefore, considering the importance of these major irrigatory tanks in Anuradapura District, it is recommended to carry out constant monitoring of heavy metals and take responsibility and actions to minimize the harm to the environmental, protect watersheds and preserve water quality and environment for current and future generations.

## **Conclusions**

According to the novel environmental pollution assessments, it is clear to note that, anthropogenic sources readily augment the risk of toxic metal contamination in soil and sediments in both industrial and agricultural areas. Intence applications of the fertilizers and pesticides for the crops may be the leading process for the contamination of the water resourses in the agricultural areas. Due to the toxic metals accumulations, even food chains can be contaminated and enhance hazardous condition on aquatic life and human. Exceeded threshold values of some toxic metals (Cd, As, Pb, and Cr) in selected tank sediments reveal that a risk might be generated to toxic heavy metal circulation in the selected area. Further, heavy metal contents of the fertilizers and pesticides which are being applied in agricultural activities in Anuradhapura district should be analyzed to investigate the sources of the toxic metals.

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# Declaration of interests

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

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