

Please cite the Published Version

Dissanayke, DMPNK, De Silva, SNT, Pathmarajah, S, Fernando, CAN, Abeynayake, NR, Kodagoda, KADA and Dassanayake, DMMNK (2023) Occurrence of heavy metals in surface water bodies in rice cultivation areas in Trincomalee district, Sri Lanka. Paddy and Water Environment, 21 (1). pp. 1-14. ISSN 1611-2490

DOI: https://doi.org/10.1007/s10333-022-00909-3

Publisher: Springer (part of Springer Nature)

Version: Accepted Version

Downloaded from: https://e-space.mmu.ac.uk/631640/

Usage rights: C In Copyright

Additional Information: his version of the article has been accepted for publication, after peer review (when applicable) and is subject to Springer Nature's AM terms of use, but is not the Version of Record and does not reflect post-acceptance improvements, or any corrections. The Version of Record is available online at: http://dx.doi.org/10.1007/s10333-022-00909-3

Enquiries:

If you have questions about this document, contact openresearch@mmu.ac.uk. Please include the URL of the record in e-space. If you believe that your, or a third party's rights have been compromised through this document please see our Take Down policy (available from https://www.mmu.ac.uk/library/using-the-library/policies-and-guidelines)

Occurrence of heavy metals in surface water bodies in rice cultivation areas in Trincomalee district, Sri Lanka

D. M. P. N. K. Dissanayke · S. N. T. De Silva · S. Pathmarajah · C. A. N. Fernando · N. R. Abeynayake · K. A. D. A. Kodagoda · D. M. M. N. K. Dassanayake

Abstract

Nonpoint source pollution from agricultural runoff which contains hazardous agrochemicals like pesticides and fertilizers threatens water bodies, posing a serious danger to aquatic ecosystems and drinking water resources. Use of agrochemicals in rice cultivation has rapidly increased in the last few decades in Sri Lanka. It has been reported that many agrochemicals contain toxic trace elements like As, Cd, Pb, Zn, Cu, Ni, Cr, Al, etc. To reduce the harmful effect of heavy metal contain-ing agrochemicals, the Government of Sri Lanka has banned the use of several toxic agrochemicals in recent years. In this context, this study was conducted after posing these restrictions to assess the occurrence of heavy metals in surface water bodies in rice cultivation areas in Trincomalee district, Sri Lanka, Ninety-three (n = 93) sample locations were randomly selected for collection of water, before and after applying agrochemicals to rice fields. This include areas from rural farm-ing communities having endemic (Padavi Sripura DS) and non-endemic (Kanthala DS and Seruvila DS) Chronic Kidney Disease of unknown etiology (CKDu). Mean and standard error of mean of As, Cd, Pb, Cu and Zn concentrations (μ g/L) before and after applying agrochemicals were as {0.048 ± 0.038 and 6.220 ± 0.912}, $\{0.014 \pm 0.013 \text{ and } 0.371 \pm 0.192\}$, {not detected and $4.421 \pm 0.712\}$, $\{1.583 \pm 0.397 \text{ and } 1.262 \pm 0.165\}$ and {not detected and 6.403 ± 0.366 }, respectively. Findings revealed that concentrations of As, Cd, Pb and Zn were significantly different (p < 0.001) before and after the application of agrochemicals. However, the observed heavy metal concentrations were far below the permissible levels for irrigation water set by Food and Agriculture Organization and United States Environmental Protection Agency, ambient water quality set by Central Environmental Authority, and drinking water quality set by World Health Organization. In addition, analyzed heavy metal concentrations in surface water samples from CKDu endemic areas were not significantly different from those from non-endemic areas (p < 0.05). To our knowledge, this is the first report of heavy metal analysis after government ban of agrochemicals. Therefore, continuous detailed research is required to fully comprehend the behavior of agrochemicals in surface water bodies in Sri Lanka.

Keywords Agriculture \cdot Agrochemicals \cdot Flame Atomic Absorption Spectroscopy (FAAS) \cdot Heavy metal concentration \cdot Water pollution

Introduction

Water pollution is a global problem that has worsened in both developed and developing countries, threatening both economic development and the physical and environmental health of billions of people. The increase in urban,

agricultural and industrial activities discharge a significant amount of wastewater has exacerbated this problem. Agriculture is considered as one of the leading causes of water pollution in the world. Surface runoff of agrochemicals used in agriculture can result in discharge to surface water bodies. As a result, dissolved pollutants will potentially end up in rivers, lakes and reservoirs. Further, the effects of deposition will last for years after the source has been eliminated or regulated. Because of the chronic toxicity of these substances or the products of their degradation, the effect of these effluents on living organisms can be harmful. Thus, protecting water bodies from agricultural pollution is a major concern. A better understanding of the causes and effects of agricultural water pollution is very important to prevent and remedy the problem.

Agrochemicals are chemical products comprised of plant-protection chemicals or pesticides and fertilizers which are composed mainly of nitrogen, phosphorous and potassium compounds. In Sri Lanka, agriculture is heavily dependent on the usage of agrochemicals (Aravinna et al. 2006), and the use of agrochemical has grown exponentially in the last few decades (Alwis et al. 2006; Karunaratne et al. 2007). Several studies reported the popularity of overusing agricultural chemicals by farmers in many parts of Sri Lanka (Padmajani et al. 2014; Selvarajah & Thiruchelvam 2007). According to Marasinghe, this situation has intensified due to the indiscriminate use of pesticides without proper awareness of their detrimental impact on the environment and human health (Marasinghe et al. 2011). Therefore, the country prohibits the use of pesticide formulations (since 1970-2015) included in the World Health Organization (WHO) hazard class Ia, Ib and the *organochlorine* pesticides including persistent organic pollutants following the WHO guideline (Department of Agriculture (DOA), 2019). Further, continuous application of relatively large quantities of permissible pesticides has led to the contamination of the ecosystem with residues of pesticides (Aravinna et al. 2017; Marasinghe et al. 2011). Moreover, continuous use of agrochemicals has harmful effects on agricultural production and reduced agricultural sustainability (Wilson and Tisdell 2001). Water pollution, resistance to pesticides and outbreaks of secondary pests are some other negative effects of agrochemicals (Dutcher 2007). Further, several researches have reported that in Sri Lanka and other countries, residues of herbicides and insecticides have been found in shallow wells, drainage streams and agricultural land reservoirs, showing the offsite movement and leaching potential of pesticides. The amounts of agrochemicals used would run off to surface water bodies and spread through the interconnected irrigation canal system or groundwater leach, and this movement is affected by climate and environmental factors, contributing to environmental pollution, including groundwater and surface water supplies (Aravinna et al. 2006). Therefore, pollution of surface water and shallow groundwater with pesticides in agricultural areas is a significant threat to vulnerable aquatic fauna including fish species, frogs, snails, snakes and aquatic plants (Aravinna et al. 2017).

The main cause of adding heavy metals to the environment is commonly considered to be agricultural activities, such as application of pesticides, fertilization and disposal of organic waste (Jiao et al. 2012). Heavy metals are considered as well-known environmental pollutants due to their toxicity, persistence in the environment, and bioaccumulative nature. Their natural sources include the weathering anthropogenic origins include mining and various industrial and agricultural activities (Ali et al. 2019). The presence of heavy metals in the environment raises the probability of the potential intake of such toxic components by the living organisms and their accumulation in many body organs, including kidneys, liver, bone, etc. Furthermore, the accumulation of these metals causes harm to a number of body systems, including the nervous, skeletal, endocrine, immune and circulatory systems (Alengebawy et al. 2021). An understanding of the current status of heavy metals in the ecosystem is therefore important to objectively determine the level of environmental heavy metal pollution as it contributes to significant environmental and health issues (Wijayawardhana et al. 2016). Numerous studies have been conducted in Sri Lanka to examine the impacts of heavy metals that are present in agrochemicals. The use of agricultural chemicals has been identified as the key anthropogenic cause of contamination of As and Cd in Sri Lanka's aquatic climate (Ileperuma 2000; Jayasumana et al. 2011). Jayasumana et al., (2015a, b) reported that phosphate fertilizers are a major source of inorganic arsenic (As) in CKDu endemic areas, based on their study of 226 fertilizer samples collected from Padaviya, Mahawilachchiya and Anuradhapura in the North Central Province of Sri Lanka. Additionally, they also stated that triple superphosphate (TSP) which was one of the first high-analysis phosphorus (P) fertilizers that became widely used in the twentieth century contained the highest concentrations of As (mean value of 31 mg/kg). Further, TSP is imported into Sri Lanka in excess of 0.1 million tons per year, containing approximately 2100 kg of As (Jayasumana et al. 2015a, b). Further, Dissanayake and Chandrajith (2009) stated that triple superphosphate (TSP) contained considerably higher concentrations of Cd, Pb, Ni, Cr and Al among the fertilizers compared to those found in urea and NPK fertilizers obtained from Medirigiriya and certain parts of Giradurukotte in Sri Lanka. Further, they have mentioned that a large amounts of Cd, Hg, Pb, U and Cr have been identified in final products of phosphate fertilizer that are ready for marketing. Premarathna et al., (2010) have researched heavy metal concentrations in some inorganic phosphate fertilizers, manures and liming materials, and the identified heavy metal concentrations of Cd were greater in imported rock phosphate (IRP) and TSP than the levels set by the SLSI. Furthermore, according to the facts described in the above sections, the route of entering of heavy metals into agricultural soils in Sri Lanka may be the application of polluted fertilizer sources to crop fields. Similarly, Jayasumana et al., (2011) indicated that 29 out of 31 brands of pesticide commonly used in Sri Lanka contain As in significant amounts ranging from $180 \pm 14 \,\mu g/kg$ (in imidacloprid collected from Colombo) to $2586 \pm 58 \mu g/$ kg (in glyphosate collected from Anuradhapura). Further,

of metal-bearing rocks and volcanic eruptions, while their

Wijesekara and Marambe (2011) identified greater As levels in some samples of pesticides obtained from sales outlets in Padaviya, Siripura, Girandurukotte and Dehiattakandiya, and detectable As levels in three pesticides, namely glyphosate, carbofuran and thiocyclam. Consequently, restrictions were imposed on the importation of pesticides including paraquat, carbofuran, carbaryl, propanil and glyphosate in recent years (Aravinna et al. 2017).

Over the past ten years, CKDu, the major health issue in the paddy farming areas in Sri Lanka, has been the focus of many scientific and political discussions. The majority of scientists have come to the conclusion that this is a toxic nephropathy even though there is disagreement among them over the etiology of the disease (Jayasumana et al. 2014). It has been speculated that heavy metals present in pesticide products and artificial fertilizers may possibly be a cause of the chronic kidney disease of unknown etiology (CKDu) (Athuraliya et al. 2011; Wijesekara & Marambe 2011). Jayasumana et al., (2015a, b) reported that CKDu patients in the Padavi Sri Pura Divisional Secretariat area of the Trincomalee District in the Eastern Province of Sri Lanka have shown irregular spotty pigmentation in the palms and soles due to chronic As toxicity; therefore, it may be the cause of CKDu. In addition, the authors stated that the most likely sources of As in the study region were pesticides and fertilizers used excessively in paddy farming. Further, As (Jayasumana et al. 2011) and Cd (Bandara et al. 2008) are the most popular heavy metals likely to cause some identified health issues in Sri Lanka. As is a class 1 human carcinogen that has been described as the second most serious global health risk of water pollution, following pathogenic organism contamination (Chappells et al. 2015). Bloom and his team in 2014 stated that As causes birth-related complications such as lower birth weight, early delivery and smaller neonatal size in exposed mothers and their fetuses. Further, it also causes defects in the cardiovascular, integumentary, reproductive, endocrine and pulmonary systems in humans (Herath et al. 2017). In addition, Averbeck and Bertin in 2006 mentioned that cadmium (Cd) is harmful to humans because of its long half-life of 15-20 years in the body and its rapid uptake and accumulation through food chains. Further, WHO in 2012 stated that Cd had been identified as one of the causes of CKDu (Herath et al. 2018). Farmers living in Trincomalee district mainly depend on paddy cultivation as their livelihood, and all of them use various forms of inorganic fertilizers and pesticides including insecticides, weedicides and fungicides. The water used in their paddy lands is diverted to the irrigation canals directly or indirectly. This study was conducted to study the occurrence of heavy metals in surface water bodies in rice cultivation areas, due to the application of agrochemicals in Trincomalee district, after banning of several toxic pesticides by the Government of Sri Lanka in recent years.

Methodology

Study area

The present study was conducted in three Divisional Secretariat divisions (DSs), namely Kanthale (Fig. 2), Seruvila (Fig. 3) and Padavi Sripura (Fig. 4) in Trincomalee district which is one of the major rice cultivation areas in Sri Lanka with extensive paddy fields that are located far from industrial zones. It is situated in the Eastern coast of the country (Fig. 1) and falls within the Northern longitudes of 8° 5' 55''- 8° 59' 52" and Eastern latitudes of 80° 44' $39''-81^{\circ}$ 27'54". There are basically two cultivation seasons, namely *Maha* and *Yala* in this area (Land Use Policy Planning Department (LUPPD) Ministry of Lands, 2016). The net harvested extent under paddy cultivation in the district during 2019/2020 *Maha* season was 35,567 hectares with an estimated paddy production of 155,592 MT (Department of Census and Statistics Sri Lanka 2020).

Sample collection

Ninety-three (n=93) surface water sampling locations were selected by using random sampling method from the three DSs during 2019/2020 Maha cultivation season to study the occurrence of heavy metals such as arsenic (As), cadmium (Cd), lead (Pb), copper (Cu) and zinc (Zn). Water samples in duplicate were collected at two times from the same location: two weeks before and after applying agrochemicals to the rice cultivation. The surface water samples from reservoirs, distribution canals, field canals and drainage canals were collected to provide adequate spatial coverage in the studied area, and the locations were demarcated by using GPS. Figure 1 shows the locations from where samples were collected for analysis. Table 1 shows the number of surface water samples collected in each DS. Collected water samples were transported to the laboratory within 4 h after storing them in a fridge by a personal vehicle. Samples were filtered through Whatman (0.5 µm) filter papers, acidified by adding concentrated nitric acid until final concentration become 2% nitric (HNO₃), pH less than 2 and stored at 4 °C until further analysis. Another separate water samples in duplicate were collected from the same sampling points into Falcon tubes (50 mL) to measure the pH value.

In addition, farmers (n = 174) were selected through a multi-stage cluster sampling method from the selected three DSs areas (Kanthale, Seruwawila and Padavi Sripura) and administered a questionnaire-based survey to collect information on the type and amount of pesticide usage, their knowledge on pesticides, adherence to recommended dosage, practices of mixing pesticides and reasons, frequency of

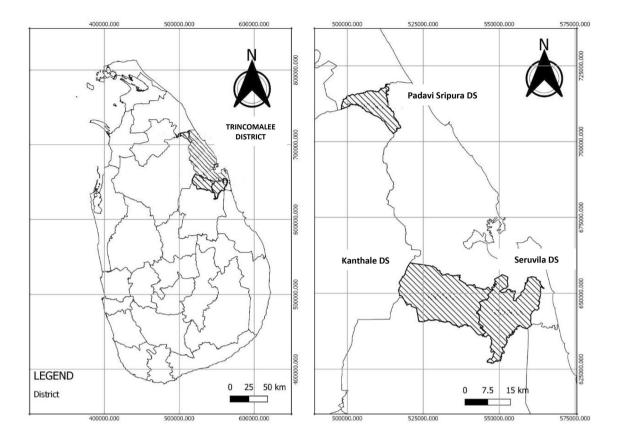
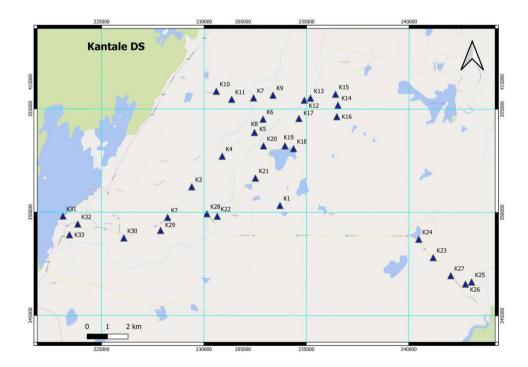


Fig. 1 Location of Trincomalee district in Sri Lanka and selected Divisional Secretariat divisions (DSs) in Trincomalee district



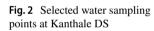
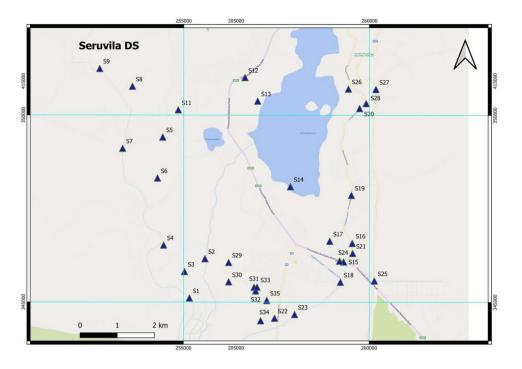
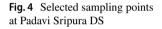


Fig. 3 Selected water sampling points at Seruvila DS





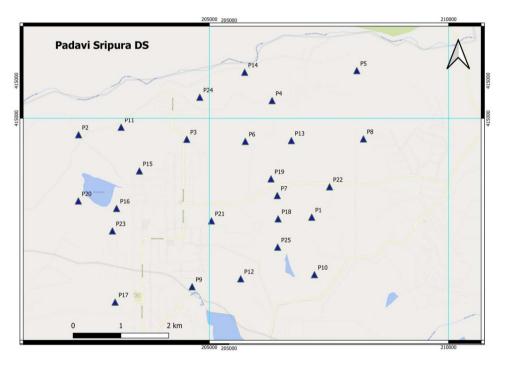


 Table 1
 Number of surface water samples collected in each selected

 DS for heavy metal analysis

DS Division	Sample point ID	No. of samples
Kanthale	К1-К 33	33
Seruvila	S1-S35	35
Padavi Sripura	P1-P25	25
Total (Trincomalee District)		93

using pesticide and method of disposal of pesticide containers and the use of local water sources for drinking.

Sample analysis

The pH value of water samples was measured by pH meter (HACH© pHC201 probe). The concentrations of heavy metals were analyzed in water samples using graphite

Table 2 Working parameters of atomic absorption spectrophotometer

Serial Number	Element	Wave length (nm)	Slit Width (nm)
1	As	193.7	1.2
2	Cu	324.8	0.8
3	Zn	213.9	0.8
4	Cd	228.8	0.8
5	Pb	217	0.8

Table 3 Graphite furnace temperature program

Step	Temperature (°C)	Hold	Ramp (seconds)
1	80	20	6
2	90	20	3
3	110	10	5
4	350	20	50
5	1100	10	300
6	1100	6	0
7	2200	4	1500
8	2450	4	500

furnace atomic absorption spectrometry (GFAAS) (*Zeenit 700P*) at the Nano Science Technology Laboratory, Wayamba University of Sri Lanka.

Instrumentation

Standard operating parameters set are given in Table 2. The hollow cathode lamps for As, Cu, Zn, Cd and Pb were used as radiation source, and air acetylene was used as the fuel. Graphite furnace temperature program is given in Table 3. All samples and standards were run in duplicate. Two milliliters of samples were added to the standard sample array which contains 108 tubes of autosampler, and readings were analyzed using Aspect LS 1.5.4.0 software. Average readings were used for further statistical analysis.

Table 4Stock solutionconcentrations for standardcurve

Standard stock solution preparation

Standard heavy metal (As, Cu, Zn, Cd and Pb) reagents of analytical grade (*Sigma-Aldrich*©) were used as stock solutions, and standards were prepared using deionized water. Before taking measurements, the highest concentration of each standard solution was inserted into sample array of the GFAAS and calibration curve was generated by autodilution procedure by the integrated dilutor. The diluents generated for As, Cu, Zn, Cd and Pb in ppb ranges are indicated in Table 4.

Statistical analysis

Data were subjected to analysis using Statistical Package for the Social Sciences (SPSS) (IBM SPSS Statistics Version 22). Frequency distributions and descriptive indicators such as mean, mode, median, minimum and maximum values were used to summarize the data. All variables were assessed for normality using the Kolmogorov-Smirnov test, supplemented with histograms. When there was no evidence to reject normality, parametric analysis was performed based on means and standard deviations. When the assumption of normality was violated, suitable nonparametric analysis was performed. Associations between categorical data were assessed using Chi-square independent test. Correlations between variables were assessed using Pearson correlation coefficients for parametric data, i.e., if both variables were normally distributed, or Spearman correlation coefficients if variables were not normally distributed. Statistical significance of all tests was assessed at 5% level.

Results and discussion

Pesticide usage pattern among farmers

Results of the survey recorded that 97%, 70% and 4% of farmers used herbicides, insecticides and fungicides, respectively. Further, none of the farmers used pesticide formulations in WHO hazard class *Ia* and *Ib*, which are prohibited in the country. However, 63% of farmers used class *II*

No Type	Туре	Concentration (ppb)						
		As	Cu	Zn	Cd	Pb		
1	Cal Zero	0	0	0	0	0		
2	Std 1	1	4	2	0.4	4		
3	Std 2	5	8	4	0.8	8		
4	Std 3	10	12	6	1.2	12		
5	Std 4	15	16	8	1.6	16		
6	Std 5	20	20	10	2	20		

Table 5 Heavy metalconcentrations ($\mu g/L$) in thesurface water before and afterapplying agrochemicals to therice cultivation

Heavy Metal	Count	Concentrat	ion (µg/L)	Maximum Permissible limits (µg/L)		
		Mean	SE	FAO (1985)	US EPA (2012)	
As						
a	93	0.048	0.038	100	100	
b	93	6.220	0.912			
Cd						
a	93	0.014	0.014	10	10	
b	92	0.371	0.192			
Pb						
a	93	ND	-	5000	5000	
b	93	4.421	0.712			
Си						
a	93	1.583	0.397	200	200	
b	93	1.262	0.165			
Zn						
a	93	ND	_	2000	2000	
b	93	6.403	0.366			

a—before applying agrochemicals to the rice cultivation, b—after applying agrochemicals to the rice cultivation, N.D.—Not Detected, SE—Standard Error of Mean

moderately hazardous category which is not highly recommended by Food and Agriculture Organization (FAO) for developing countries. Six percent (6%) used pesticides of Class III category which is slightly hazardous. Thirty-one percent (31%) used Class U which is unlikely to cause any acute hazard according to WHO pesticides hazard categories. The most commonly used herbicide, insecticide and fungicide are Pretilachlor 30% EC (34%), Carbosulfan 200 g/L SC (36%) and Tebuconazole 250 g/L EW (2%), respectively. As fertilizers urea, triple superphosphate (TSP) and Muriate of potash (MOP) were common among farmers in the study area. It has been observed that 50% of farmers violate the recommendations imposed by the DOA. During the study, it has been observed that farmers tend to use higher dosages (58%) when diluting pesticides but apply a lower amount per extent (83%) than the recommendations made by DOA in order to reduce the labor cost incurred while spraying. Study reveals that the most of the user-level problems are related to poor attitude of farmers (Dissanayake et al. 2022).

Heavy metal concentrations in surface water (As Cd, Pb, Cu and Zn)

Table 5 shows the heavy metal concentrations of As, Cd, Pb, Cu and Zn in the collected surface water samples, before and after application of agrochemicals in to the fields. Mean and standard error of As, Cd, Pb, Cu and Zn concentrations ($\mu g/L$), before and after applications, were { 0.048 ± 0.038 and 6.220 ± 0.912 }, { 0.014 ± 0.014 and 0.371 ± 0.192 }, {not detected and 4.4212 ± 0.712 }, { 1.583 ± 0.397 and

1.262 \pm 0.165} and {not detected and 6.4035 \pm 0.366}, respectively (Fig. 2). According to FAO (FAO, 1985) and US EPA (US EPA, 2012) guidelines, the recommended maximum concentrations of As, Cd, Pb, Cu and Zn for irrigation water are as 100, 10, 5000, 200 and 2000 µg/L, respectively. Further, for ambient water quality, CEA (CEA, 2019) has set the recommended maximum concentrations of As, Cd, Pb, Cu and Zn as 50, 5, 50, 100 and 1000 µg/L, respectively. The observed heavy metal concentrations in this study were far below the permissible levels set by FAO and US EPA for irrigation water as well as for the ambient water quality standards set by the CEA (2019). However, a slight increase in concentrations was observed after agrochemicals application (Table 5 and Figs. 5, 6, 7, 8, 9).

Heavy metal concentrations in water samples, before and after applying agrochemicals, were compared statistically. Firstly, the concentrations were tested for normality. According to Kolmogorov–Smirnov statistics, As, Cd, Pb and Cu (p < 0.05) were not normally distributed, while Zn (p = 0.116) is normally distributed. Hence, for those where the normality was rejected, Wilcoxon signed rank test was used for comparison, and when assumption of normality was met, data were analyzed by paired t test. The results revealed that the differences were significant for As (p < 0.001), Cd (p < 0.001), Pb (p < 0.001) and Zn (p < 0.001) and not significant for Cu (p > 0.001) (Table 6).

Use of local water sources for drinking purpose

Results from the questionnaire-based survey revealed that 55% of the farmers use local water sources for drinking.

Fig. 5 Arsenic (As) concentrations in each sampling point before and after applying agrochemicals to the rice cultivation. Sampling Points: Kanthale DS (1–33), Seruvila DS (34–68) and Padavi Sripura DS (69–93)

Fig. 6 Cadmium (Cd) concen-

trations in each water sampling

point before and after apply-

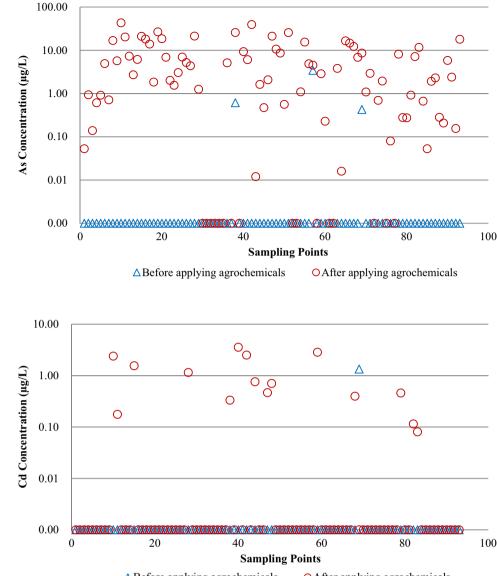
ing agrochemicals to the rice

cultivation. Sampling Points:

Kanthale DS (1-33), Seruvila

DS (69-93)

DS (34-68) and Padavi Sripura

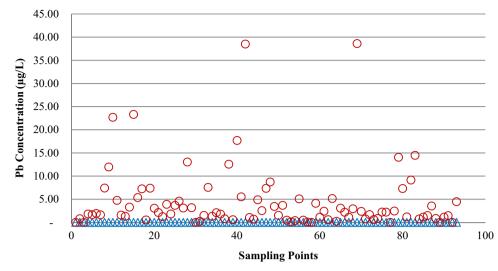


△Before applying agrochemicals OAfter applying agrochemicals

Among them, 37% use water from distribution canals for drinking purpose. It can be observed that the irrigation canal system is interconnected from one to another in the study area by distribution canals, field canals and drainage canals. Field canals and drainage canals are small waterways designed by farmers' organizations to apply and drain water from paddy lands, respectively. These field canals and drainage canals are affected by any kind of application that takes place in the paddy fields as they are directly connected to the paddy lands. Agrochemicals applied to paddy fields are mixed and drained into these canals with water. US EPA reported that contaminants that enter the flowing waters travel some distance before being thoroughly mixed throughout the flow (Perera et al. 2016). Consequently, the chemicals that are found in field canals and drainage canals can be recovered at relatively higher concentrations at the other surface water sources. Following the same phenomenon, considerable amount of heavy metals was detected in distribution canals (Table 7).

According to Sri Lankan Standards (Sri Lankan Standard Institute (SLSI), 2013) and WHO guidelines (WHO, 2011), the maximum permissible levels of As, Cd and Pb in drinking water are 10, 3 and 10 μ g/L, respectively. Further, according to Sri Lankan standards (SLSI, 2019), the maximum permissible levels of Cu and Zn in drinking water are 1000 and 3000 μ g/L, respectively. Mean and standard error of As, Cd, Pb, Cu and Zn concentrations (μ g/L) in distribution canals before and after applying agrochemicals were {not detected and 4.39 \pm 3.26}, {not detected}, {not detected and 2.00 \pm 0.87}, {0.57 \pm 0.34 and 1.83 \pm 0.44} and {not detected and 8.69 \pm 1.12}, respectively, and was less than the SLSI and WHO standards for drinking water. Cd was not

Fig. 7 Lead (Pb) concentrations in each water sampling point before and after applying agrochemicals to the ricecultivation. Sampling Points: Kanthale DS (1–33), Seruvila DS (34–68) and Padavi Sripura DS (69–93)



△Before applying agrochemicals OAfter applying agrochemicals

Fig. 8 Copper (Cu) concentrations in each water sampling point before and after applying agrochemicals to the rice cultivation. Sampling Points: Kanthale DS (1–33), Seruvila DS (34–68) and Padavi Sripura DS (69–93)

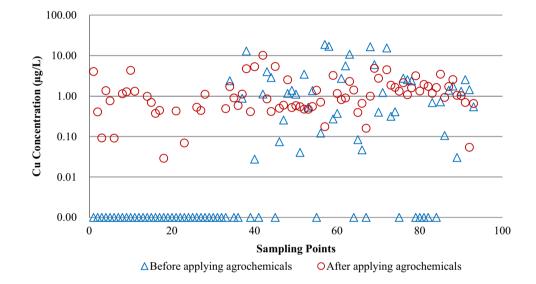


Fig. 9 Zinc (Zn) concentrations in each water sampling point before and after applying agrochemicals to the ricecultivation. Sampling Points: Kanthale DS (1–33), Seruvila DS (34–68) and Padavi Sripura DS (69–93)

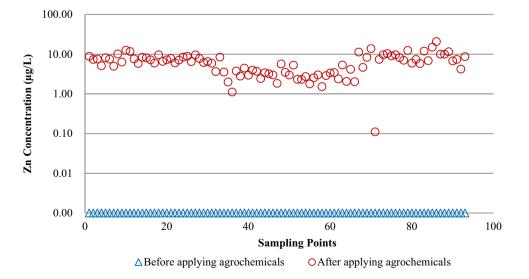


 Table 6
 Comparison of heavy metal concentrations before and after applying agrochemicals

Before and after applying agrochemicals	Asymp. Sig. (2-tailed)
a. Wilcoxon Signed Ranks Test	
As Concentration (µg/L)	< 0.001
Cd Concentration (µg/L)	< 0.001
Pb Concentration (µg/L)	< 0.001
Cu Concentration (µg/L)	0.067
c. Paired sample t test	
Zn Concentration (µg/L))	< 0.001

recorded in distribution canals (Table 7). Therefore, it can be considered under the current context that the observed heavy metal concentrations in local water sources would not pose a serious threat to the environment or human health. Due to high-prevalence CKDu in this area, the government of Sri Lanka has taken action to implemented filtered water resources for drinking. Even though the levels of concentration of heavy metals are far below the accepted levels, however, bioaccumulation over a period of time would be harmful. It has been observed that some sampling points including three distribution canals show higher levels of As and Pb. Therefore, it is recommended to use filtered water for drinking purposes. There were very few studies conducted to access the level of heavy metals in surface water bodies in Sri Lanka. Scientific evidence indicates that Sri Lankan surface water bodies, with a few exceptions, are at a safe level for daily activities of people and agricultural practices. In agreement with the results of current study, Mahagamage et al. (2018) stated that except for Al, in certain sampling areas in Kelani river basin, the concentrations of Cd, Zn, Pb, Cr and Cu remained below the SLS and WHO standards for drinking water requirements. In addition, Chandrajith et al. (2012) identified that the mean level of Cd in surface water in dry zone is 0.0003 mg/L, which is again far less than standards.

Heavy metal concentration comparison between CKDu endemic and CKDu non-endemic areas

The study revealed that there was no difference in pesticide usage in CKDu endemic areas (Padavi Sripura DS) and the CKDu non-endemic areas (Kanthale DS and Seruvila DS) (Dissanayake et al. 2022). Similarly, no significant difference in concentrations of heavy metals was detected in between the two areas (Table 8).

In a study conducted by Wasana et al. (2012) assessed the water quality using 60 water samples collected from areas with high and low prevalence of CKDu in Sri Lanka and reported that the As and Cd levels in the water were well-below the WHO guidelines of 10 μ g/L and 3 μ g/L.

Surfac	Surface water source							
Concentration (µg/L)								
	Tank		Distribution Canal		Field Ca	nal	Drainage Canal	
	Mean	SE	Mean	SE	Mean	SE	Mean	SE
As								
a	ND	-	ND	-	0.02	0.02	ND	-
b	1.63	0.89	4.39	3.26	5.82	1.79	8.54	2.17
Cd								
a	ND	-	ND	-	ND	-	ND	-
b	ND	_	ND	_	0.29	0.14	0.25	0.11
Pb								
a	ND	-	ND	_	ND	-	ND	-
b	0.86	0.49	2	0.87	4.31	1.34	6.22	1.39
Си								
a	ND	-	0.57	0.34	2.62	0.79	0.19	0.1
b	0.01	0.01	1.83	0.44	1.65	0.36	0.92	0.22
Zn								
a	ND	_	ND	-	ND	_	ND	-
b	8.01	1.1	8.69	1.12	4.86	0.57	6.01	0.64

a—before applying agrochemicals to the rice cultivation, b—after applying agrochemicals to the rice cultivation, ND—Not Detected, SE—Standard Error of Mean

Table 7Heavy metalconcentrations ($\mu g/L$) in thesurface water sources before andafter applying agrochemicals

Table 8 Heavy metalconcentrations $(\mu g/L)$ in thesurface water in each DSs,before and after applyingagrochemicals

DS Division							
	Kanthale DS**		Seruvila D	Seruvila DS**		Padavi sripura DS*	
	Mean	SE	Mean	SE	Mean	SE	
As							
a	ND	-	0.116	0.099	0.017	0.017	
b	7.977	1.754	6.847	1.591	3.024	0.901	
Cd							
a	ND	_	ND	_	0.054	0.054	
b	0.160	0.090	0.331	0.144	0.027	0.678	
Pb							
a	ND	_	ND	_	ND	-	
b	4.662	0.995	4.127	1.192	4.514	1.630	
Си							
a	ND	_	3.012	0.897	1.672	0.631	
b	0.615	0.177	1.499	0.346	1.786	0.240	
Zn							
a	ND	_	ND	_	ND	_	
b	7.566	0.322	3.359	0.297	9.131	0.789	

a—before applying agrochemicals to the rice cultivation, b—after applying agrochemicals to the rice cultivation, N.D.—Not Detected, *CKDu endemic areas, **CKDu non-endemic areas

Further, water quality assess conducted by WHO (n=234)in CKDu endemic and non-endemic water sources showed that As concentrations were borderline only in four samples in the endemic region. Further, Cd concentrations in drinking water sources used by patients with CKDu in Sri Lanka were within the usual range. In addition, a study conducted by Jayatilake and his fellow researches based on water samples collected from CKDu endemic and non-endemic areas in Sri Lanka concluded that drinking water has not been polluted by Cd which is believed to cause CKDu (Javatilake et al. 2013). Further, in a study carried out by Chandrajith, low levels of Cd in drinking water samples obtained from both CKDu endemic and non-endemic regions in the north central province of Sri Lanka were reported, which were well-below the WHO guideline of 3 µg/L (Chandrajith et al. 2011). These results coincide with the current research, which shows low concentrations of As, Cd, Pb, Cu and Zn in distribution canals.

pH of surface water

The pH of water determines the solubility and biological availability of chemical constituents such as nutrients (phosphorus, nitrogen and carbon) and heavy metals (Pb, Cu, Cd, etc.). In the case of heavy metals, the degree to which they are soluble determines their toxicity. Metals tend to be more toxic at lower pH because they are more soluble. Water samples collected in this study shows an increase in mean pH values from (7.13 ± 0.04) to (7.19 ± 0.04) before and after applying agrochemicals, respectively, and this difference is

 Table 9 Mean and standard error of mean of pH, before and after applying agrochemicals

	s- chemical ameters	Count	Mean	SE	Recommended Maximum Permis- sible limits
pН	a	93	7.13	0.04	6.5-8.4*
	b	93	7.19	0.04	

a—before applying agrochemicals to the rice cultivation, b—after applying agrochemicals to the rice cultivation

*Food and Agriculture Organization (1985) and United States Environmental Protection Agency (2012)

statistically significant (p > 0.05) (Table 9). Mean pH value lies within the permissible range of 6.5–8.4 (FAO, 1985 and US EPA, 2012) for irrigation water, ambient water quality standards of pH 6.0–8.5 (CEA 2019 and SLSI, 2013) and pH 6.0–8.5 for drinking water (WHO, 2011).

Conclusion

Agrochemicals used in the field be likely to transport through surface water bodies and spread through the interconnected irrigation canal system. In this study, ninety-three (93) water samples were collected randomly from different locations in three Divisional Secretaries in Trincomalee district. Sampling points include both CKDu endemic (Padavi Sripura) and non-endemic (Kanthala, Seruvila) areas. Random sampling method was deployed to collect water samples from the same location, duplicating each time. Two samples were obtained to represent the quality of water, two weeks before and after the application of agrochemicals to rice fields in 2019/2020 *Maha* cultivation season. Mean and standard error of mean of As, Cd, Pb, Cu and Zn concentrations (μ g/L) between two were {0.048 ± 0.038 and 6.220 ± 0.912}, {0.014 ± 0.014 and 0.371 ± 0.192}, {not detected and 4.421 ± 0.712}, {1.583 ± 0.397 and 1.262 ± 0.165} and {not detected and 6.403 ± 0.366}, respectively. Among the elements tested, the difference of concentration is statistically significant for all except for Cu.

Further, it has been noted that the observed mean heavy metal concentrations in these areas were far below the permissible levels for irrigation water set by FAO (1985) and US EPA (2012), ambient water quality set by CEA (2009) and the level set for drinking water by WHO (2011) and SLSI (2013). This study was conducted after the government regulations to ban the use of heavy metal containing fertilizers and certain pesticides, and it can be envisaged that the decrease in concentration could be a result of that. It is also worth mentioning here that the distribution of heavy metal concentrations was not significantly different in CKDu non-endemic and CKDu endemic areas.

Mean and standard deviation of measured pH levels in surface water samples were 7.13 ± 0.34 and 7.19 ± 0.35 , at two sampling occasions, respectively. These values also falls within the permissible levels for irrigation water, ambient water and drinking water set by FAO, CEA, WHO and SLSI.

It was evident from this study that malpractices such as mixing, over uses in dilution practices and lack of proper disposal mechanisms of pesticides containers are common among farmers. Since there are no legal provisions or institutional mechanisms available at the field level to deal with malpractices in the use of agrochemicals, it is suggested to raise the awareness of the community regarding the consequences through agricultural extension service. Further, it is necessary to establish regular monitoring and follow-up mechanisms at the field level to regulate the activities of the farmers, and if necessary to take legal actions against malpractices. In addition, continued monitoring of the presence of residues of agrochemicals in water bodies is proposed.

Due to high-prevalence CKDu in this area, the government of Sri Lanka has taken action to implemented filtered water resources for drinking. Even though the levels of concentration of heavy metals are far below the accepted levels, bioaccumulation over a period of time would be harmful. It has been observed that some sampling points including three distribution canals show higher levels of As and Pb. Some of the farmers (37%) were taking water samples for drinking purposes from the field distribution canals. They should be made aware of the risks and should promote to use filtered water to minimize health hazards by consuming contaminated water.

Further, national studies are required to fully assess the extent of heavy metal toxicity in water in the country. In Sri Lanka, research on variations in seasonal trace elements in the environments is rare. However, to understand the behavior of agrochemicals in the environment and to assess the seasonal trace element variance in water bodies, continuous monitoring system is required. Further, farmers should also be educated and encouraged to practice organic agriculture and integrated pest management, which will help to promote sustainable, eco-friendly development.

Acknowledgments Authors wish to express their sincere gratitude to all respondents for their valuable cooperation in responding to the questionnaire. A special acknowledgement goes to extension staff of the inter-Province, Trincomalee, for their assistance provided. Authors gratefully acknowledge the Sri Lanka Council for Agriculture Research Policy (SLCARP) for their financial support (NARP/CARP/16/WUSL/ ASC/01).

Declarations

Conflict of interest The authors declare that there is no conflict of interest.

References

- Alengebawy A, Abdelkhalek ST, Qureshi SR, Wang M-Q (2021) Heavy metals and pesticides toxicity in agricultural soil and plants: Ecological risks and human health implications. Toxics. https://doi.org/10.3390/toxics9030042
- Ali H, Khan E, Ilahi I (2019) Environmental chemistry and ecotoxicology of hazardous heavy metals: Environmental persistence, toxicity, and bioaccumulation. J Chem. https://doi.org/10.1155/ 2019/6730305
- Alwis GKH, Wijesekera RD, Jayasekera TADNAK (2006) Use patterns and residue levels of pesticides on Mukunuwenna, a leafy vegetable grown in Sri Lanka. Bull Environ Contam Toxicol 76(1):119–125. https://doi.org/10.1007/s00128-005-0897-3
- Aravinna AGP, Priyantha N, Pitawala HMTGA, Yatigammana SK (2017) Use pattern of pesticides and their predicted mobility into shallow groundwater and surface water bodies of paddy lands in Mahaweli river basin in Sri Lanka. J Environ Sci Health Part B Pestic Food Contam Agric Wastes 52(1):37–47. https://doi.org/ 10.1080/03601234.2016.1229445
- Aravinna, A. G. P., Liyanage, J. A., Mubarak, A. M., Wickramaratne, S., & Liyanage, S. (2006). Monitoring of widely used pesticides in surface water reservoirs and shallow wells in Walawe area. Sri Lanka Assassination for the Advance of Science, Proceedings of the 62nd Annual Session.
- Athuraliya NTC, Abeysekera TDJ, Amerasinghe PH, Kumarasiri R, Bandara P, Karunaratne U, Milton AH, Jones AL (2011) Uncertain etiologies of proteinuric-chronic kidney disease in rural Sri Lanka. Kidney Int 80(11):1212–1221. https://doi.org/10.1038/ki. 2011.258

- Bandara JMRS, Senevirathna DMAN, Dasanayake DMRSB, Herath V, Bandara JMRP, Abeysekara T, Rajapaksha KH (2008) Chronic renal failure among farm families in cascade irrigation systems in Sri Lanka associated with elevated dietary cadmium levels in rice and freshwater fish (Tilapia). Environ Geochem Health 30(5):465–478. https://doi.org/10.1007/s10653-007-9129-6
- Central Environmental Authority (CEA). (2019). Ambient Water Quality Standards.
- Chandrajith R, Nanayakkara S, Itai K, Aturaliya TNC, Dissanayake CB, Abeysekera T, Harada K, Watanabe T, Koizumi A (2011) Chronic kidney diseases of uncertain etiology (CKDue) in Sri Lanka: Geographic distribution and environmental implications. Environ Geochem Health 33(3):267–278. https://doi.org/10.1007/ s10653-010-9339-1
- Chandrajith R, Ariyaratna T, Dissanayake CB (2012) The Status of cadmium in the Geo-environment of Sri Lanka. Ceylon J Sci Phys Sci 16:47–53
- Chappells H, Campbell N, Drage J, Fernandez CV, Parker L, Dummer TJB (2015) Science of the total environment understanding the translation of scientific knowledge about arsenic risk exposure among private well water users in Nova Scotia. Sci Total Environ 505:1259–1273. https://doi.org/10.1016/j.scitotenv.2013.12.108
- Department of Agriculture (DOA). (2019). Pesticide Recommendations. http://doa.gov.lk/ICC/images/publication/Books/Books/ Pesticide20_08_11ver3.pdf
- Department of Census and Statistics Sri Lanka. (2020). Paddy statistics 2019/2020 Maha Season. http://www.statistics.gov.lk/Agriculture/ StaticalInformation/PaddyStatistics/PaddyStatistics/2019-20Mah aSeason
- Dissanayake CB, Chandrajith R (2009) Phosphate mineral fertilizers, trace metals and human health. J Natl Sci Found 37(3):153–165. https://doi.org/10.4038/jnsfsr.v37i3.1219
- Dissanayake DMPNK, De Silva SNT, Pathmarajah S, Kodagoda KADA, Chandimal TMR, Herath HMTD (2022) Pesticide usage pattern in rice cultivation in Trincomalee district in Sri Lanka. Trop Agric Res 33(2):173–182. https://doi.org/10.4038/tar.v33i2. 8474
- Dutcher, J. D. (2007). A Review of Resurgence and Replacement Causing Pest Outbreaks in IPM. Ciancio, A., Mukerji, K.G. (Eds) General Concepts in Integrated Pest and Disease Management. Integrated Management of Plants Pests and Diseases, Vol 1. Springer, Dordrecht. https://doi.org/10.1007/978-1-4020-6061-8_2
- Food and Agriculture Organization (FAO) of the United Nations. (1985). *Mid-decade review of food and agriculture*.
- Herath HMAS, Kubota K, Kawakami T, Nagasawa S, Motoyama A, Weragoda SK, Chaminda GGT, Yatigammana SK (2017) Potential risk of drinking water to human health in Sri Lanka. Environmental Forensics 18(3):241–250. https://doi.org/10.1080/15275 922.2017.1340364
- Herath HMAS, Kawakami T, Nagasawa S, Serikawa Y, Motoyama A, Tushara Chaminda GG, Weragoda SK, Yatigammana SK, Amarasooriya AAGD (2018) Arsenic, cadmium, lead, and chromium in well water, rice, and human urine in Sri Lanka in relation to chronic kidney disease of unknown etiology. J Water Health 16(2):212–222. https://doi.org/10.2166/wh.2018.070
- Ileperuma OA (2000) Environmental pollution in Sri Lanka: a review. J Natl Sci Found 28(4):301–325. https://doi.org/10.4038/jnsfsr. v28i4.2644
- Jayasumana MACS, Gajanayake R, Siribaddana S (2014) Importance of Arsenic and pesticides in epidemic chronic kidney disease in Sri Lanka. BMC Nephrol 15:124. https://doi.org/10.1186/ 1471-2369-15-124
- Jayasumana MACS, Fonseka S, Fernando A, Jayalath K, Amarasinghe M, Siribaddana S, Gunatilake S, Paranagama P (2015a) Phosphate fertilizer is a main source of arsenic in areas affected with chronic

kidney disease of unknown etiology in Sri Lanka. Springerplus 4(1):1–8. https://doi.org/10.1186/s40064-015-0868-z

- Jayasumana MACS, Paranagama P, Agampodi S, Wijewardane C, Gunatilake S, Siribaddana S (2015b) Drinking well water and occupational exposure to Herbicides is associated with chronic kidney disease, in Padavi-Sripura Sri Lanka. Environ Health. https://doi.org/10.1186/1476-069X-14-6
- Jayasumana, M. A. C. S., Paranagama, P. A., Fonseka, M. A., & Wijekoon, D. V. K. (2011). Presence of Arsenic in pesticides used in Sri Lanka. *Water Professionals' Day Symposium*, 18. http://www. lankaweb.com/news/items11/Arsenic-Paper.pdf
- Jayatilake N, Mendis S, Maheepala P, Mehta FR (2013) Chronic kidney disease of uncertain aetiology: Prevalence and causative factors in a developing country. BMC Nephrol 14(1):1. https://doi.org/ 10.1186/1471-2369-14-180
- Jiao W, Chen W, Chang AC, Page AL (2012) Environmental risks of trace elements associated with long-term phosphate fertilizers applications: A review. Environ Pollut. https://doi.org/10.1016/j. envpol.2012.03.052
- Karunaratne SHPP, Weerakoon KC, Nugaliyadda L, Manuweera GK (2007) Susceptibility of rice insect pests and their natural enemies to commonly used insecticides. J Natl Sci Found 35(2):97–102. https://doi.org/10.4038/jnsfsr.v35i2.3673
- Land Use Policy Planning Department (LUPPD) Ministry of Lands. (2016). Land use plan Trincomalee district. https://luppd.gov.lk/ images/content_image/downloads/pdf/llrc_trincomalee.pdf
- Mahagamage, M. G. Y. L., Chinthaka, S. D. M., & Manage, P. M. (2018). Assessment of surface water pollution by heavy metals in Kelani river basin, Sri Lanka. Symposium Proceedings, Sixth International Symposium on Water Quality and Human Health: Challenges Ahead, PGIS, Peradeniya, Sri Lanka, July
- Marasinghe, J. P., Gamage, C., Shiromi, M. G. D., & Aravinna, A. G. P. (2011). Organophosphate pesticides residues in food commodities in Sri Lanka: A review. Annals of the Sri Lanka Department of Agriculture, 13, 81–93. https://www.researchgate.net/publication/ 258227161%0AOrganophosphate
- Padmajani, M. T., Aheeyar, M. M. M., & Bandara, M. A. C. S. (2014). Assessment of pesticide usage in up-country vegetable farming in Sri Lanka. *HARTI Research Report No: 164 Hector Kobbekaduwa* Agrarian Research and Training Institute. https://doi.org/10. 13140/2.1.4241.7289
- Perera PACT, Sundarabarathy TV, Sivananthawerl T, Suranga PK, Edirisinghe U (2016) Arsenic and Cadmium contamination in water, sediments and fish is a consequence of paddy cultivation: Evidence of river pollution in Sri Lanka. Achiev Life Sci 10(2):144–160. https://doi.org/10.1016/j.als.2016.11.002
- Premarathna HMPL, Hettiarachchi GM, Indraratne SP (2010) Trace metal concentration in crops and soils collected from intensively cultivated areas of Sri Lanka. Pedologist 54(3):230–240
- Selvarajah, A., & Thiruchelvam, S. (2007). Factors affecting pesticide use by farmers in vavuniya district. *Tropical Agricultural Research*, 19, 380–388. https://pdfs.semanticscholar.org/a187/ d9eff65da2f03ca4f01b94a84dd05c64c6f5.pdf
- Sri Lankan Standard Institute. (2013). *SLS Standard 614:2013*. Sri Lankan Standard Institute, Colombo.
- Sri Lankan Standard Institute. (2019). *SLS Standard 894:2019*. Sri Lankan Standard Institute, Colombo.
- United States Environmental Protection Agency (EPA). (2012). Guidelines for Water Reuse. In United States Agency for International Development, Washington, district of Columbia. http://www.epa. gov/nrmrl/pubs/625r04108/625r04108.pdf
- Wasana HMS, Aluthpatabendi D, Bandara J (2012) Drinking water quality assessment towards "Chronic Kidney Disease of unknown etiology (CKDu)" in North Central Province (NCP) of Sri Lanka. Challenges Ahead, International Symposium on Water Quality and Human Helath

- Wijayawardhana D, Herath V, Weerasinghe A (2016) Heavy metal pollution in Sri Lanka with special reference to agriculture: a review of current research evidence. Rajarata Univ J 4(1):52–56
- Wijesekara GA, Marambe B (2011) Arsenic in the environment–an overview on global and Sri Lankan context. Ann Sri Lanka Dep Agric 13:229–243
- Wilson C, Tisdell C (2001) Why farmers continue to use pesticides despite environmental, health and sustainability costs. Ecol Econ 39(3):449–462. https://doi.org/10.1016/S0921-8009(01)00238-5

World Health Organization (WHO). (2011). Guidelines for drinkingwater quality, fourth edition.