

# 7.2.5 POLLUTION OF RIVER MAHAWELI AND FARMLANDS UNDER IRRIGATION BY CADMIUM FROM AGRICULTURAL INPUTS LEADING TO A CHRONIC RENAL FAILURE EPIDEMIC AMONG FARMERS IN NCP, SRI LANKA

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ORIGINAL PAPER

## Pollution of River Mahaweli and farmlands under irrigation by cadmium from agricultural inputs leading to a chronic renal failure epidemic among farmers in NCP, Sri Lanka

J. M. R. S. Bandara <sup>#</sup> H. V. P. Wijewardena <sup>#</sup>  
Y. M. A. Y. Bandara <sup>#</sup> R. G. P. T. Jayasooriya <sup>#</sup>  
H. Rajapaksha

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**Abstract** Chronic renal failure (CRF) associated with elevated dietary cadmium (Cd) among farming communities in the irrigated agricultural area under the River Mahaweli diversion scheme has reached a significantly higher level of 9,000 patients. Cadmium, derived from contaminated phosphate fertilizer, in irrigation water finds its way into reservoirs, and finally to food, causing chronic renal failure among consumers. Water samples of River Mahaweli and its tributaries in the upper catchment were analyzed to assess the total cadmium contamination of river water and the possible source of cadmium. Except a single tributary (Ulapane Stream, 3.9 µg Cd/l), all other tested tributaries carried more than 5 µg Cd/l, the maximum concentration level accepted to be safe in drinking water. Seven medium-sized streams carrying surface runoff from tea estates had 5.1–10 µg Cd/l. Twenty larger tributaries (*Oya*), where the catchment is under vegetable and home garden cultivation, carried 10.1–15 µg Cd/l. Nine other major tributaries had extremely high levels of Cd, reaching 20 µg Cd/l. Using geographic information system

(GIS), the area in the catchment of each tributary was studied. The specific cropping system in each watershed was determined. The total cadmium loading from each crop area was estimated using the rates and types of phosphate fertilizer used by the respective farmers and the amount of cadmium contained in each type of fertilizer used. Eppawala rock phosphate (ERP), which is mostly used in tea estates, caused least pollution. The amount of cadmium in tributaries had a significant positive correlation with the cadmium loading of the cropping system. Dimbula Tea Estate Stream had the lowest Cd loading (495.9 g/ha/year), compared with vegetable-growing areas in Uma Oya catchment with 50,852.5 g Cd/ha/year. Kendall's  $\tau$  rank correlation value of total Cd loading from the catchment by phosphate fertilizer used in all crops in the catchment to the Cd content in the tributaries was +0.48. This indicated a major contribution by the cropping system in the upper catchment area of River Mahaweli to the eventual Cd pollution of river water. Low soil pH (4.5–5.2), higher organic matter content (2–3%), and 18–20 cmol/kg cation exchange capacity (CEC) in upcountry soil have a cumulative effect in the easy release of Cd from soil with the heavy surface runoff in the upcountry wet zone. In view of the existing water conveyance system from upcountry to reservoirs in North Central Province (NCP) through diversion of River Mahaweli, in addition to their own nonpoint pollution by triple superphosphate fertilizer (TSP), this demands a change in overall upper catchment management to

J. M. R. S. Bandara (✉) · H. V. P. Wijewardena ·  
Y. M. A. Y. Bandara · R. G. P. T. Jayasooriya  
Department of Agricultural Biology, University of  
Peradeniya, Peradeniya 20400, Sri Lanka  
e-mail: jmrsb@pdn.ac.lk

H. Rajapaksha  
School of Molecular and Biomedical Sciences, The  
University of Adelaide, Adelaide, SA 5005, Australia

minimize Cd pollution through agriculture inputs to prevent CRF due to elevated dietary cadmium among NCP farmers.

**Keywords** Phosphate fertilizer·Cadmium from fertilizer·Chronic renal failure·Cadmium loading·Cadmium in tributaries·Cadmium mobility

## Introduction

Chronic renal failure (CRF) associated with elevated dietary cadmium was first reported in North Central Province (NCP) of Sri Lanka in 1993. Bandara et al. (2008) reported that the number of patients in NCP was around 5,000. CRF cases in NCP steadily increased to 7,650 in 2009, as reported by the Health Ministry of Sri Lanka (Health Fund 2009). The Government Information Department reported the number treated for CRF by October 2009 in Anuradhapura Hospital to be 9,000 (Jayamanna 2009). These cases are mainly from NCP, Uva Province (Girandurukotte and Nikewewa), and North Western Province. The total number of deaths reported from CRF in Anuradhapura, NCP General Hospital is 1,082 to date, since 1993.

It was observed that the affected patients were mostly (90–94.5%) rice farmers (Athuraliya et al. 2003) from farming communities under the major irrigation scheme established with the diversion of River Mahaweli in 1977. The average annual discharge from River Mahaweli, amounting to 8.4 billion cubic meters, is used to irrigate 140,000 ha in NCP (MAHAWELI 1988). The unique situation of water reservoirs in NCP, fed by the waters of the diverted River Mahaweli in addition to supply from their own catchment, which are fed by drainage water from rice fields, demands special study on the impact of farming on water quality. The pattern of location of reservoirs in a series of cascades and the irrigation canals are described in detail by Bandara et al. (2008). These reservoirs are the main sources of drinking and irrigation water of NCP farmers.

Several researchers have reported the impact of agriculture on the quality of irrigation and potable water in the dry zone of Sri Lanka. These reports dealt with direct contamination of water by either fertilizers

or pesticides applied in fields. Data reported were mostly on the occurrence of excessive quantities of nitrates and phosphates that led to eutrophication of reservoirs and heavy nitrate levels in reservoirs and ground water (Piyasiri 1995; Liyanage et al. 2000; de Zoysa 2002). In a more comprehensive study on water quality in NCP, Perera (2006) and Wickramarachchi (2005) reported the occurrence of higher quantities of the weedicide Propanil [1.02 mg/l in reservoirs, 1.18 mg/l in canals, in comparison with World Health Organization (WHO) recommended maximum acceptable level (MAL) of 175 µg/l] and the insecticide chlorpyrifos (2.8 and 6.77 mg/l in reservoirs and canals, respectively, compared with WHO MAL of 0.09 µg/l) in irrigation water. A bioassay based on the rate of egg yolk utilization by tilapia eggs proved the resilient efficacy of these pesticides contaminating waterways and reservoirs. Bandara et al. (2008), working on reservoirs used for drinking purposes and irrigation of rice fields, fed with either diverted Mahaweli water or precipitation in the respective catchment area, reported significantly high levels of cadmium (Cd), Iron (Fe), and lead (Pb) in reservoir waters and reservoir sediments. No arsenic (As) was detected in any of the reservoir sediments or waters tested. Bandara et al. (2008) showed that mean urinary cadmium (UCd) concentration in CRF patients ( $7.58 \pm 6.18 \mu\text{g Cd/g creatinine}$ ) and those without symptoms but exposed to elevated dietary cadmium ( $11.62 \pm 8.45 \mu\text{g Cd/g creatinine}$ ) in the same region were far above normal levels ( $2 \mu\text{g Cd/g creatinine}$ , recommended by WHO). Biopsy reports of CRF patients in Madawachchiya (NCP) show a predominant factor of tubular interstitial dysfunction with endocytosis and proximal tubular sclerosis, with no glomerular renal dysfunction (Bandara et al. 2008), with 35% of the population having creatinine clearance less than 30 ml/min. It is evident that farmers in the region are exposed to elevated dietary cadmium and also to cadmium from pesticides that contain it as a contaminant in their formulations. Cd was detected ( $0.5 \pm 0.1 \text{ mg Cd/l}$ ) in a formulation of bispyribac sodium, a weedicide very heavily used in rice farming in NCP, as reported by Bandara et al. (2008). The main source of Cd in the rice environment appears to be agrochemicals, with a major contribution by contaminated low-grade triple superphosphate fertilizer (TSP) used in rice fields. Cadmium contamination in TSP used in Sri Lankan rice

fields mainly in the dry zone varied from 23 to 71.739 mg Cd/kg of P<sub>2</sub>O<sub>5</sub> (Premarathna et al.2005; Bandara et al.2008).

The objective of this study is to determine the amount of cadmium added through the waters of diverted River Mahaweli originating in different catchment areas under various cropping systems which eventually elevate the dietary cadmium in NCP. This unique situation makes the region more prone to chronic renal failure due to elevated dietary intake of Cd compared with other regions of Sri Lanka that cultivate rice.

## Materials and methods

### Study area

The total land area of Sri Lanka is 65,525 km<sup>2</sup>, of which 59,217 km<sup>2</sup> is covered by 103 distinct river basins. Among these, the basin of River Mahaweli is the largest, covering an area of over 10,000 km<sup>2</sup>, representing one-sixth of the area of the island. Mahaweli Basin covers a land area lying across five of the nine provinces of Sri Lanka. The population in Mahaweli Basin amounts to more than 2.8 million, 15% of the total population. The average annual discharge from River Mahaweli is about 8.4 billion cubic meters, which is used to irrigate 140,000 ha in NCP, of which 75,361 ha was sowed with rice in the year 2004. In the dry zone farmlands irrigated by Mahaweli diversion schemes, the mean annual surface runoff is  $2.55 \times 10^{-6}$  ha m and the runoff rainfall ratio is 35.8%. NCP includes 4,000 small reservoirs, arranged in 280 cascade systems (Bandara et al. 2008), most of which are fed with Mahaweli water through diversions at several points of River Mahaweli in the wet zone. Tributaries of Mahaweli for sampling were selected mostly from the upper catchment area of River Mahaweli and also at points of diversion that carried irrigation water to dry zone farmlands through reservoirs located in the dry zone (Figs.1,4).

### Sample collection

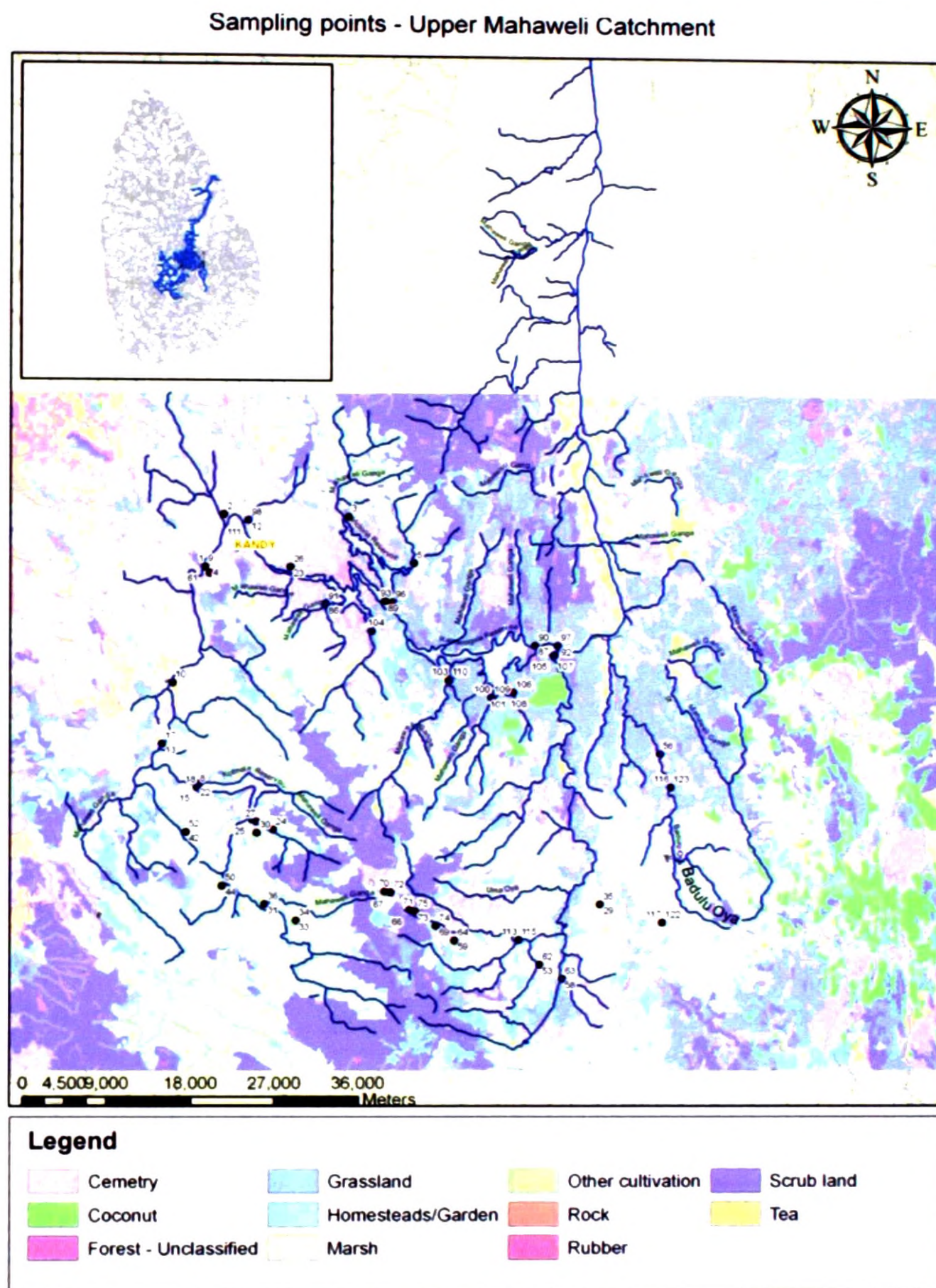
Sampling was done at 21 tributaries of River Mahaweli, namely at: (1) Pinga Oya, (2) Hulu Ganga (river), (3) Galmal Oya, (4) Mada Ela (canal), Kandy,

(5) Makandura Oya, (6) Watahena Oya, (7) Maldeniya Oya, (8) Huna Oya, (9) Pundalu Oya, (10) Badulu Oya, (11) Talatu Oya, (12) Kurundu Oya, (13) Mana Oya, (14) Belihul Oya, (15) Stream (running through) Talawakele Estate, (16) Stream Fernlands Estate, (17) Stream St Coombs TRI Estate Talwakele, (18) Stream Dimbula, (19) Stream Mount Vernon Estate, (20) Stream Katabula Tea Estate, and (21) Uma Oya. The manmade reservoirs of the upper Mahaweli catchment that were sampled were Polgolla Reservoir (at the mouth of the underground diversion canal) and Kotmale Reservoir (both left and right banks). Several samples were taken at River Mahaweli, at Gampola Township near the bridge, tributary near Ulapane, two locations near New Bridge Talawakele, and other locations along the major tributaries, represented by three-digit numbers as follows: 201, 202, 203, and 204, from Badulu Oya along Badulla Mahiyangana Road up to Badulla Rawana Falls; 205, Diganatenna Welimada Bandarawela Road; 206, 207, 208, 209, 210, 211, 212, and 213, streams leading to Uma Oya up to Welimada; and 214 and 215, Gregory Lake. Other reservoirs sampled downstream of River Mahaweli from the first diversion point at Polgolla were Victoria left and right bank, Rantambe left and right bank, and Randenigala Reservoir. In addition to these, two more sampling points in Nuwara Eliya were taken as reference from Gregory Lake situated in Nuwara Eliya in the central highlands. The total number of sampling locations amounted to 47. Water samples from the rivers and reservoirs were collected using a column sampler (2.13 m high, 76.2 mm diameter) to obtain a composite water sample from each sample point. A maximum of 2 m depth from the surface was reached wherever possible. Three samples from each location were taken, and a composite sample was made for duplicate analysis. Global positioning system (GPS) coordinates at each sampling site were recorded using a portable GPS recorder with accuracy of  $\pm 2$  m to record the point of sampling precisely for analysis of cropping systems in the catchment of the tributaries (Fig.1).

### Analytical methods

Water samples were collected in acrylic plastic containers. Containers were acid-rinsed (1:1 nitric acid) prior to use for collection of water samples. A

**Fig. 1** Upper River Mahaweli catchment and the tributaries of the river, depicting the sampling points



sample volume of 300 ml was collected in pre-cleaned acrylic plastic containers and preserved with HNO<sub>3</sub> to maintain pH < 2, then stored in dry ice until transportation to laboratory. Each water sample was well mixed by vortexing at room temperature (25±1°C), and two 20 ml working samples were obtained and stored in 25-ml acid-washed cadmium-free glass vials at 4°C until analysis for cadmium. The maximum holding time was kept at 14 days. Analyses were done as described in *Standard*

*Methods for Examination of Water and Waste Water* by Greenberg et al. (1992).

Quality control measures were practiced for cadmium extraction and analysis at all levels and also in sampling as described by the US Environmental Protection Agency (1983). Samples were analyzed using a graphite furnace atomic absorption spectrometer (GFAAS) with detection limit of 0.015 µg/l. Total cadmium in unfiltered water samples was detected using GFAAS at 228.8 nm by injecting

20 µl samples in triplicate. Cd content in HNO<sub>3</sub> used in precleaning of vials and in house standards were also assessed. Reported results for the samples are means of three replicate analyses. As an overall quality control plan, continued calibration of the instrument was practiced and performed by analyzing one mid-concentration standard after every ten analyses. Method blanks, in which deionized double-distilled water was added to precleaned sample bottles in the field and acidified with HNO<sub>3</sub>, were analyzed. Standard solutions for analytical quality assurance (VWR; VWR International Ltd., Poole, UK) with Cd (NO<sub>3</sub>)<sub>2</sub>·4H<sub>2</sub>O in HNO<sub>3</sub> (0.5 mol/l) matrix containing 1,002±5 mg Cd/l were used. It was ensured that the relative percentage difference between the initial calibration and the continuing calibration was less than 15%.

#### Assessment of cropping systems area and cadmium loading

The areas under different crops grown in the watershed of the tributaries of River Mahaweli were assessed using the detailed maps of the regions and the GIS software available at the Land Use Division of the Government Department of Agriculture, Sri Lanka. The global positioning system (GPS) values at the locations of sampling were recorded at the time of water sampling using a GPS recorder with accuracy of ±2 m. GPS used WGS84 as the default coordinate system for universal location. Although the World Geodetic System (WGS) 1984 is the world standard coordinate system, the Sri Lankan grid system makes use of an ellipsoid that goes through the crest of Mount Everest. The Sri Lankan grid system is also known as Kandawala, which is the term preferred by the European Professional Surveyors Groups (EPSG) working group on geodesy. The geographical coordinates were converted to Kandawala using Pathfinder software to identify the sheet numbers of natural resource management data sheets. Land-use pattern and area under different crops were estimated using Kandy, Gampola, Nuwaraeliya, Matale, Badulla, and Welimada region sheets available at the Natural Resource Management Centre of the Department of Agriculture, Peradeniya. The precise boundary of each catchment was identified using ArcView 3.1a. The total catchment area of the watershed and the area under each crop were estimated using ArcGIS 9.3. The

total cadmium loading into tributaries from specific cropping systems was estimated based on the rates and frequency of fertilizer application per cropping season. The fertilizer regimes practiced, type of phosphate fertilizer used, and level of cadmium contained in them, in each cropping system, were used in the estimation of cadmium loading into the catchment and eventually into tributaries. However, as the amount of cadmium added by other regular inputs in agriculture such as soil pH moderators such as dolomite (9.06 mg Cd/kg), CaO (6.53 mg Cd/kg), organic manure (0.43–0.97 mg Cd/kg), and weedicides and pesticides (0–0.5 mg Cd/l) are negligible (Bandara et al. 2008; Premarathna 2006) in the cadmium loading assessment, only cadmium added via phosphate fertilizer was considered. Sri Lanka imported triple superphosphate fertilizer (TSP) only from three major suppliers, of which 81.55% was supplied by one single foreign supplier in both 2006 and 2007 (DoC, Sri Lanka Trade Reports 2007). The mean values of cadmium contamination reported were 23.5 mg/kg of TSP (Premarathna 2006) and 71.739 mg of Cd/kg of TSP (Bandara et al. 2008). An average value of the two reported levels of Cd contamination in TSP was taken, as there is only one major supplier of TSP for the entire agriculture sector in Sri Lanka. Eppawala rock phosphate is a locally produced rock phosphate from a resource in Eppawala in NCP, Sri Lanka, and the sole supplier of ERP to tea plantations is the Government of Sri Lanka. Premarathna (2006) reported that ERP has only 1.7 mg Cd/kg. Only ERP is used in tea estates of Sri Lanka. The total cadmium loading was therefore estimated as g Cd/year based on two seasons of cropping per year in each catchment by obtaining the product of mean Cd contamination for TSP or ERP in the case of tea, the total amount of TSP or ERP added per ha in a year, and the area under each crop under consideration in a given catchment.

#### Statistical analysis

Kendall's rank correlation coefficient was estimated as a measure of association of total cadmium loading from areas of different cropping systems and the amount of cadmium pollution per liter in Mahaweli tributaries using Wessas (2009) Kendall  $\tau$  rank correlation v1.0.10 in v1.123-γ4 (free statistics software online from Software Office for Research & Development and Education).

## Results and discussion

Figure 1 presents the area under the watershed of River Mahaweli and the sampling points of its tributaries. River Mahaweli is diverted partially into the dry zone of Sri Lanka at Polgolla through an underground tunnel. Polgolla Tunnel is important as the key point of diversion of River Mahaweli to North Central Province (Fig. 4), the ancient kingdom of Rajarata, in view of the prevailing CRF epidemic occurring there. The capacity of Polgolla Reservoir is 4.1 million cubic meters, originating from catchment area of 738 km<sup>2</sup>.

River Mahaweli originates in the Mount Sri Pada Range near Hatton (Fig. 2 catchment number 5, C-5), at 2,000 m altitude. It flows 335 km north and northeast to drain into the sea at Trincomalee Bay. Seven major tributaries, namely Kurundu Oya (C-10), Belihul Oya (C-7), Mana Oya (C-2), and Kotmale Oya (C-6) from the south, and Ma Oya (C-9), Pinga Oya (C-8), and Hulu Ganga (C-1) from the north, enter Mahaweli (Fig. 2). The total upper catchment area of Mahaweli where this study was conducted is about 316,000 ha. In the lower basin there are three major tributaries that enter the Mahaweli: Amban Ganga, Ulhitiya Oya, and Kaudulla Oya (not illustrated in Fig. 2).

### Cadmium in tributaries of River Mahaweli

The mean total cadmium concentrations in the tributaries of Mahaweli, the main River Mahaweli, and its reservoirs built within the upper catchment area, categorized into four groups, are given in Table 1. Both the main river and its tributaries carried cadmium at levels considered to be unacceptable for drinking purposes based on the maximum contaminant level (MCL) of 5 µg Cd/liter or ppb to protect human health (EPA-USA 2000). Among the major tributaries tested, only a single tributary (2.1%) had acceptable levels of cadmium as recommended by the US Environmental Protection Agency (EPA). Seven medium-sized streams carrying runoff and drainage water from tea estates had mean cadmium concentration in the range 5.1–10 µg Cd/l. Among the larger tributaries (*Oyas*), 20 *Oyas*, mainly carrying drainage water from vegetable plots, carried mean total cadmium in the range 10.1–15 µg Cd/l, and 9 others had extremely high levels, reaching 20 µg Cd/l. The

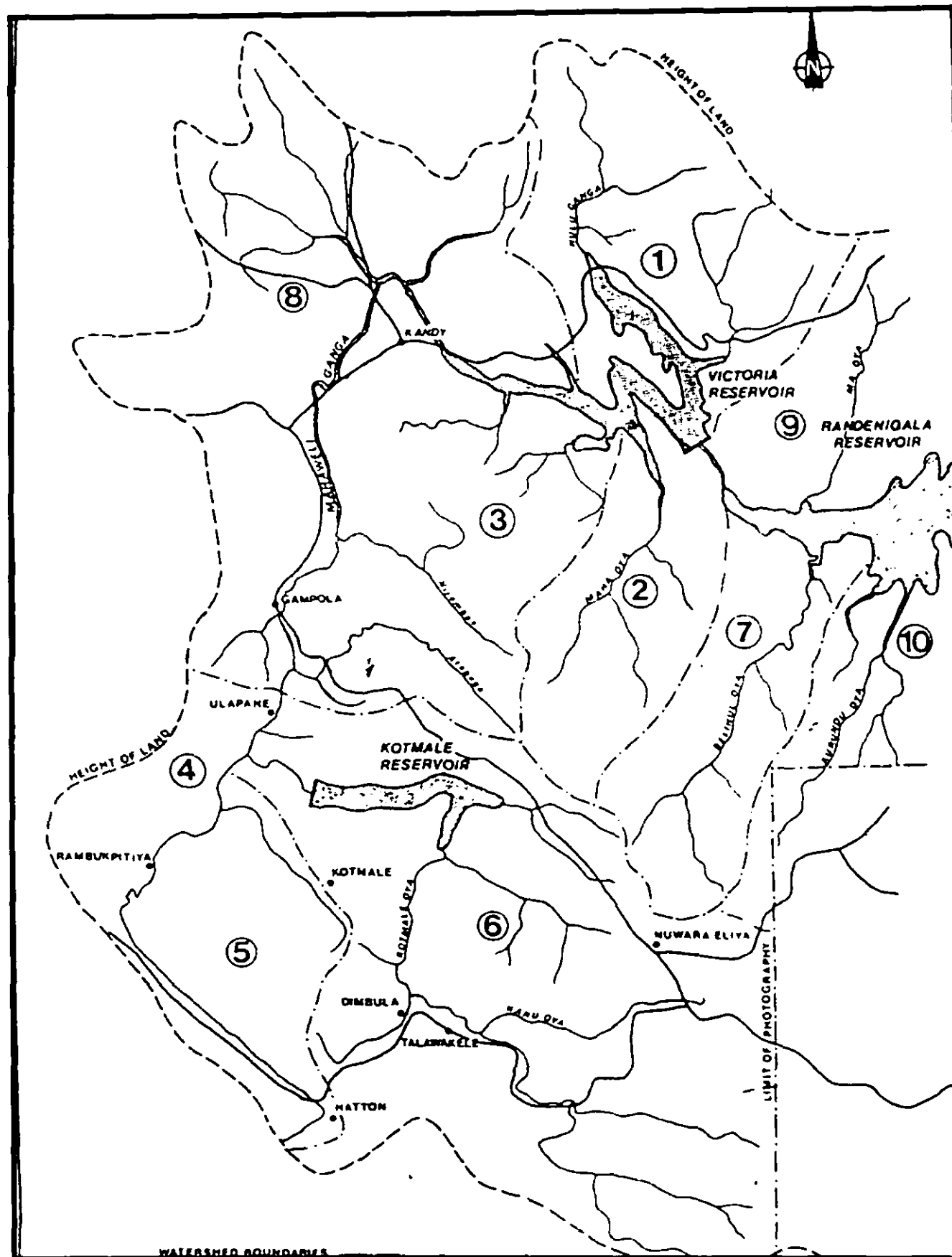
main river and the reservoirs within the upper catchment carried very high levels of cadmium (extreme observed: 23.8 µg Cd/l in Maldeniya Oya, 21.8 µg Cd/l in Rantambe Reservoir). The sampling was done during the months of November and December (21 November to 30 December, 2008) over a period of 7 weeks. The rivers and streams were rich in runoff water, as it was after the southwest monsoon and in the midst of intermonsoonal convectional rains.

The effective precipitation in the upper catchment (agroecological zones WU1, WU2, WU3, IU2 & IU3, WM1, WM2 & WM3 in Fig. 3) was found to range from 12 mm in September to 114 mm in November (UNDP/FAO 1969). The mean annual rainfall ranges from 1,650 mm in the downstream area to 5,300 mm in the upper catchment. The amount of rainfall and the resulting surface runoff depend on the intensity of the monsoon, the main climatic determinant of the island. The full force of the monsoon is received by the highest parts of the hill country, while the lowland plains and the shadow areas of the monsoon in the hill country receive lower rainfall. The upper catchment where we sampled receives the greatest rainfall during the southwest monsoon from May to September, whereas the northeast monsoon from December to January produces the least rainfall.

Premarathna et al. (2005) reported that the agricultural soil in the upcountry wet zone in agroecological zones WU3, IU2, and IU3 within the Mahaweli upper catchment region (Fig. 3) carries higher levels of Cd (total Cd 0.51–3.86 mg/kg, exchangeable Cd 0.26–1.24 mg/kg) as a result of the use of contaminated TSP as phosphate fertilizer. The low pH of upcountry soil (pH 4.5–4.8 surface soil), though rich in organic matter (3–4%), leads to easy leaching of soil Cd. Out of 39.5 × 10<sup>9</sup> m<sup>3</sup> total precipitation in the wet zone where the upper catchment is located, the total runoff per annum is 25.9 × 10<sup>9</sup> m<sup>3</sup>, amounting to 65% runoff. This heavy runoff is the reason for higher amount of cadmium in the tributaries and River Mahaweli. Blennerhassett (1998) reported the importance of the role of surface runoff in the increase in phosphate and its associated minerals in downstream tributaries (Fig. 4).

The sampling area of the upper catchment of River Mahaweli is in the region where precipitation is dominated by the intermonsoonal (IM) rain, then the northeast (NE) or southwest (SW) monsoonal rains in

Fig. 2 Watershed boundaries in the upper catchment area of River Mahaweli. Scale approximately 1:283,000 (Source TAMS Report 1980)



the order: IM > NE > SW (Fig.3). The assessment of cropping patterns and use of phosphate fertilizer sheds more light on the prevailing situation. The results of analysis of tributary cadmium levels are presented in Table 2. The effect of land-use pattern under each catchment area was estimated as cadmium loading under each cropping system in a given catchment area of the tributary. The association between cadmium loading due to the type of phosphate fertilizer applied to a specific crop and the resultant level of cadmium in the tributary of the

respective catchment was estimated using Kendall's rank correlation coefficient value and is illustrated in scatter plots in Fig. 5a-d.

#### Land use and cadmium mobility

The current land-use pattern in the upper Mahaweli watershed remains the same as it was prior to the Mahaweli diversion as reported by the Hunting Survey Corporation in 1962. Though the cropping pattern seems to be the same, the practices adopted

**Table 1** Mean total cadmium ( $\mu\text{g Cd/l}$ , both dissolved and suspended colloidal) in waters of tributaries of Mahaweli, River Mahaweli, and reservoirs in the upper catchment area from Polgolla in the mid season of northwest monsoon in 2008

Loading points (WHO <sup>a</sup> )	5.1D10 $\mu\text{g Cd/l}$	10.1D15 $\mu\text{g Cd/l}$	15.1D23 $\mu\text{g Cd/l}$
Tributaries	Ulapane St <sup>b</sup> Welimada Town St, TRI St Coombs St Talawakele Es St, Dimbula Es St, Badulu Oya <sup>d</sup> , Badulu Oya Bw Rd. Bandarawela St 204	Diganatanne, Badulu Oya Bw. Uma Oya Streams 206, 207, 208, Kurundu Oya, Mount Vernon Es St, Fernland Es <sup>c</sup> St, Katabula Es St, Uma Oya Streams 210, 211, 212, 213, Ma Oya, Beihul Oya, Uma Oya, Talatu Oya, Mana Oya, Pundalu Oya, Mada Ela Kandy	Watehena Oya, Maldeniya Oya, Pinga Oya, Badulu Oya Bad. Pass Rd, Huna Oya, Makandura Oya, Galmal Oya, Hulu Ganga
Main river	Nd New Bridge Talawakele	Polgolla Tunnel, Gampola Bridge	Gateembe Bridge
Reservoirs	Kotmale right bank	Victoria right bank, Kotmale left bank, Randenigala Res, Gregory Lake	Victoria left bank, Rantambe

<sup>a</sup> WHO recommendation as the maximum acceptable level of cadmium in drinking water

<sup>b</sup> St=streams carrying surface runoff and ground water to Mahaweli

<sup>c</sup> Es=tea estates where the streams are located

<sup>d</sup> Oya is one of the native terms used for a medium-sized river

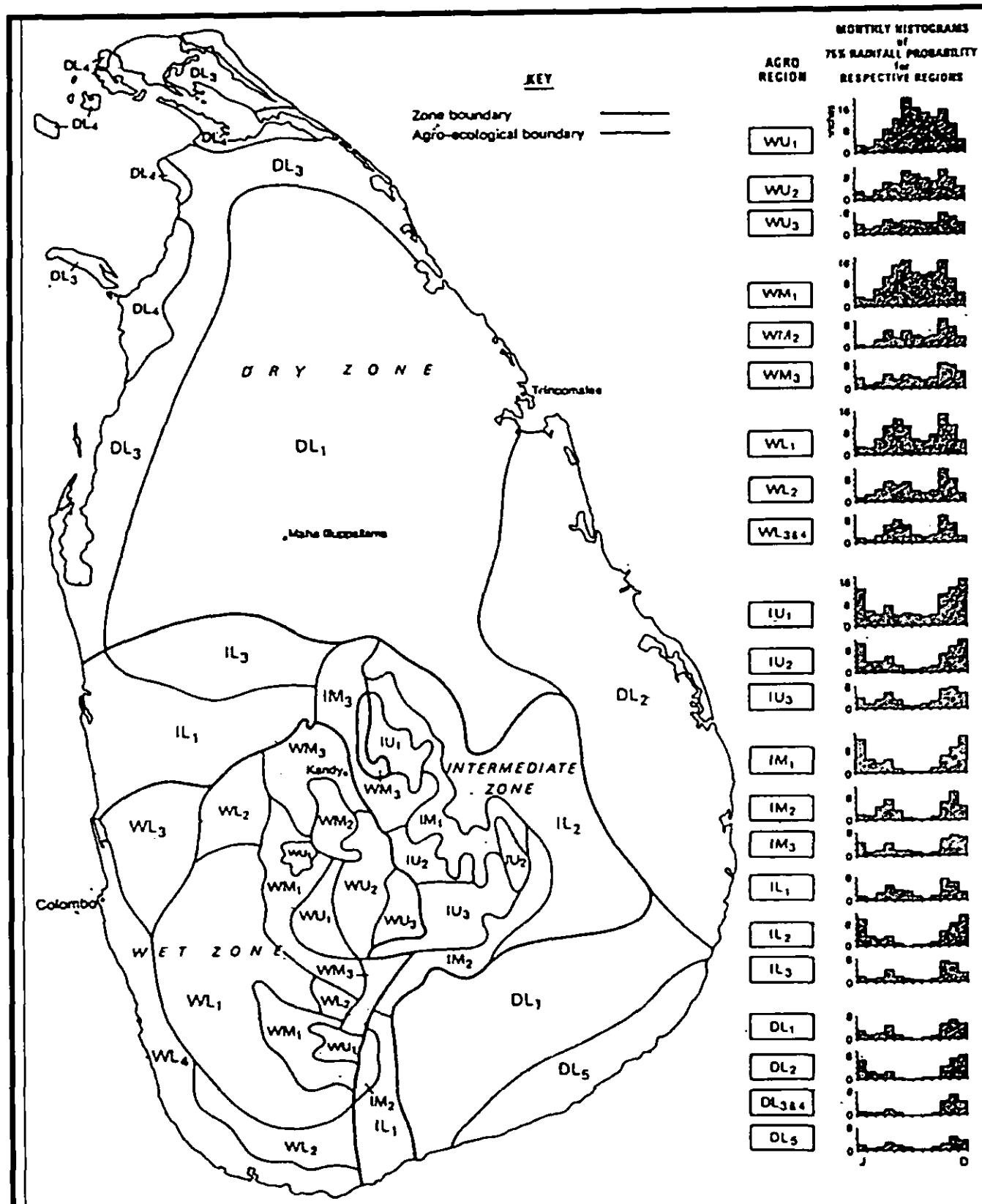
today are more chemical based. The ever-increasing use of agrochemicals and specifically excessive use of fertilizer on field crops and vegetables intensively grown under home garden systems contribute to the nonpoint pollution that we experience today (Jayatilake and Bandara 1989). The upper catchment has four major land-use groups, namely tea, field crops, home gardens with intensive vegetable cultivation, and rice. In the region we studied, 30.7% comes under intensely managed tea cultivation, 68.7% under other crops, and the balance is marshy land. However, only a single stream, which carries water from a predominantly tea area (50% of catchment area, C-4), was below the US EPA recommended MCL of  $5\mu\text{g Cd/l}$  (Table 1). It appears that all other watersheds (Fig. 2) released higher levels of Cd to runoff water. The physiographic properties of soil in the study region (WU) are undulating to rolling (Soil Science Society of Sri Lanka 1999). The amount of cadmium in uncultivated soil is  $0.51\text{ mg/kg}$ , in contrast to  $1.96\text{ mg/kg}$  in soil under vegetable cultivation (Premarathna 2006). The other major characteristics of uncultivated soil in the region are presented in Table 3.

It is evident from Fig. 5 that the heavy Cd contamination of tributary waters is due to the impact of agricultural inputs.

The minimum Cd contamination occurred in predominantly tea areas in the watershed, probably due to low input of Cd from phosphate fertilizer. Tea estates used direct unprocessed rock phosphate, namely Eppawala rock phosphate (ERP), at the rate of  $123\text{ kg/ha}$ . ERP is a rock phosphate obtained from a phosphate ore in Eppawala, Anuradhapura, Sri Lanka, and it contains only  $1.7\text{ mg Cd/kg}$ , compared with imported triple superphosphate (rock phosphate treated with sulfuric acid) that contains  $23.5\text{D}71.739\text{ mg Cd/kg}$  (Bandara et al. 2008; Premarathna 2006).

ERP is suitable for long-term crops, such as tea, in acidic soils, compared with annuals, which need to be supplied with TSP. However due to the properties of the B horizon with accumulated clay in the soil profile in the region and the rainfall pattern, the actual release of Cd may vary. The total cadmium loading that occurred in the catchment area in 1 year was therefore estimated to study the potential relationship with the Cd in the catchment, the tributaries, and the reservoirs. The total Cd loading depends on the fertilizer regimes

Fig. 3 Agroecological zones of Sri Lanka based on altitude and annual rainfall



used in each cropping system, the type of phosphate fertilizer used, and the level of Cd contained in the agrochemicals used. In the Cd loading assessment, only Cd added via phosphate fertilizer was considered, as other inputs (dolomite, organic manure, etc.) contained negligible quantities (organic manure, 0.43–0.97 mg Cd/kg; CaO lime, 6.53 mg Cd/kg; dolomite 9.06 mg Cd/kg). Average Cd content of TSP used is 47 mg/kg, based on the range 23–71 mg Cd/kg of TSP. The total loading of cadmium per ha per year based on recommended dosage of fertilizer for each crop (Lathif and Upali 2007) and the frequency of application was then calculated for the area under each

crop. Rank correlation coefficient using Kendall's  $\tau$  was assessed for total Cd loading versus Cd in the tributary waters. Due to unavailability of data on the cropping pattern and fertilizer application at all the catchment areas studied, and to maintain accuracy and clarity of information reported, only those areas where accurate agricultural input data were available were used to study correlation of Cd in tributaries and total Cd loading. A Kendall  $\tau$  rank correlation value of 0.480 (two-sided  $p=0.002$ ) was observed for the total Cd loading from tea, rice, and vegetable cultivation (including home gardening) versus Cd content ( $\mu\text{g Cd/l}$ ) in waters in tributaries. Kendall  $\tau$  values for

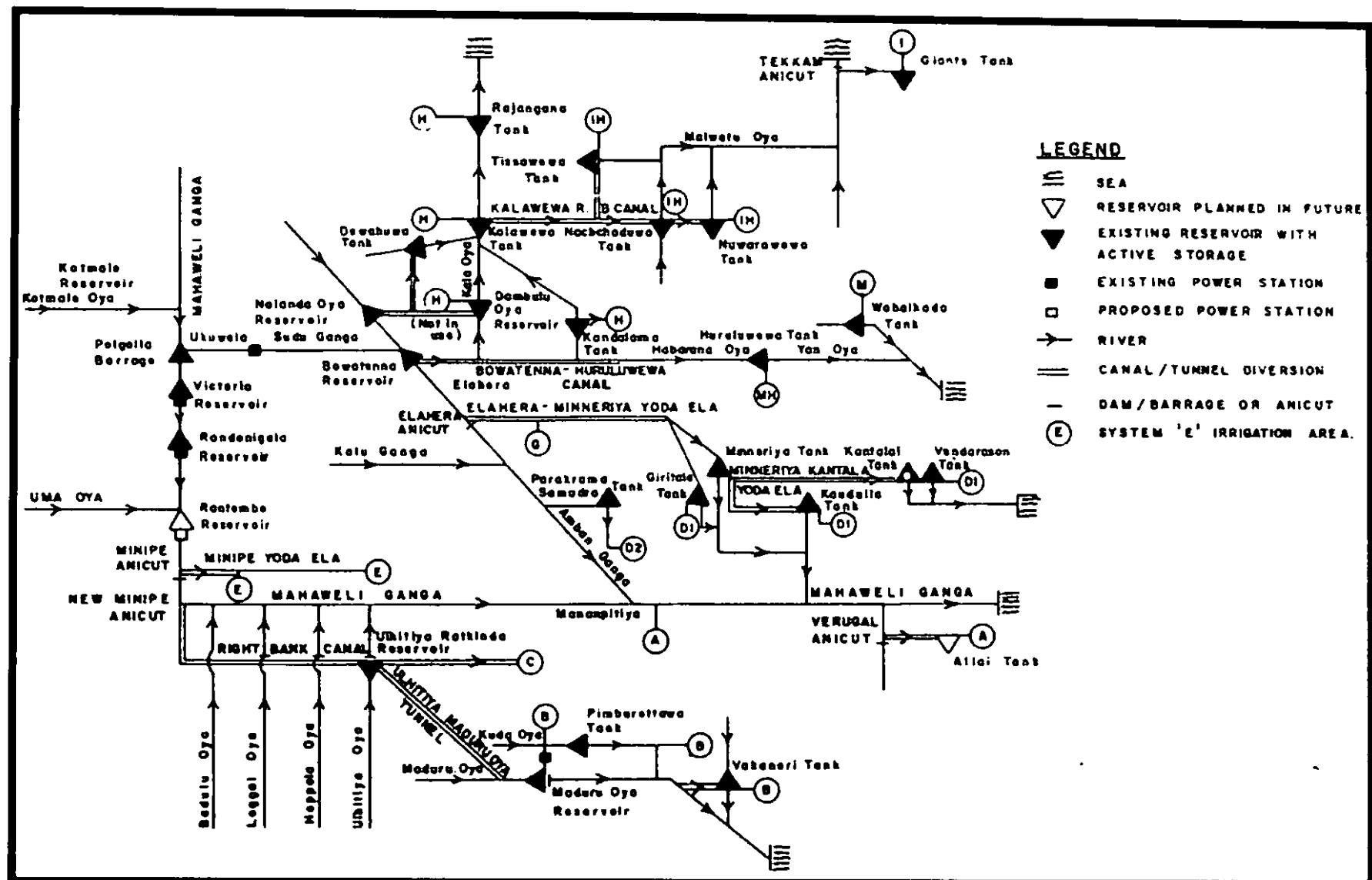


Fig. 4 Water conveyance system of the Mahaweli diversion scheme, illustrating the role of River Mahaweli upper catchment in the upcountry wet zone and eventual dispersal of Mahaweli waters throughout NCP

the association of cadmium in tributaries with cadmium loading in individual crop areas are given in Fig.5.

Figure5 presents scatter plots of correlation between cadmium loading in catchment area under different cropping systems in upper Mahaweli catchment area and the amount of cadmium in tributaries in the respective catchment. The definite positive correlation of 0.48 observed is a good indication of the impact of use of contaminated phosphate fertilizer in the upper catchment area. The ERP used was the minimum contributor to Cd in water, and as such, tea agronomy poses an insignificant threat, though vegetable and rice cultivation in the upper Mahaweli catchment, if not properly managed, could be a bigger problem. Premarathna et al. (2005) reported the occurrence of 0.59–3.86 mg Cd/kg of soil in upcountry soil traditionally cultivated with vegetables such as cabbage, leeks, carrot, and potato. In addition to TSP, another source of Cd that is regularly applied to vegetable fields and home garden farms is dolomite to bring pH to 7.0, which contains

9.06 mg Cd/kg. Sukreeyapongse et al. (2002) showed that low pH favors release of cadmium from soil, though higher organic carbon in soil favors formation of colloidal cadmium or chelation with humic substances. Sukreeyapongse et al. (2002) showed that cadmium is released easily into water when soil pH is low and organic carbon is relatively higher. Soils that showed higher gradual release of Cd to water had low pH (6.8), high carbon (1.5–1.8%) with CEC 7.3–8.5 cmol/kg, and soil Cd content of 2.3–7.0 μmol/kg. When compared with the soils of upper catchment with total C 3–4%, pH 4.5–4.8 with CEC of 18–20 cmol/kg, the ease of Cd release should be greater, based on which we should expect a higher positive correlation between high Cd content in soil in the catchment and Cd in river water. Observations by Sukreeyapongse et al. (2002) stress the fact that the relative release rates for Cd, Cu, and Pb showed uniform patterns when presented as a function of pH, except for two soil samples with different bonding strength of Cd and Cu. The relative release rates could be described with pH-dependent

**Table 2** Mean cadmium levels ( $\mu\text{g Cd/l}$ ) in waters of tributaries of River Mahaweli, the main river, and its reservoirs in the upper catchment from the Polgolla diversion

Location	Mean ( $\text{Cd}\mu\text{g/l}$ )	SD	Location	Mean ( $\text{Cd}\mu\text{g/l}$ )	SD
Ulapane	3.9	0.4	Uma Oya St 211	13.1	0.15
Welimada Town St	6.4	0.25	Uma Oya St 210	13.2	0.9
TRI St Coombs St	6.5	1.0	Ma Oya	13.2	0.5
Dimbula Es St	6.6	1.1	Belihul Oya	13.4	1
Kotmale right bank	6.8	3.1	Gregory Lake Ani 215	13.6	0.8
Talawakele Es St	8.1	2.2	Gampola Br	13.8	2.65
Bandarawela St	8.2	1.1	Uma Oya	13.8	0.65
Badulu Oya Bw. Rd	8.4	0.7	Talatu Oya	13.9	3.75
New Br Talawakele	9.1	1.65	Greg. Lake	14.3	0.2
Badulu Oya	9.7	0.2	Mala Oya	14.3	0.8
Diganatenna St	10.4	0.75	Victoria rt. bank	14.8	1.05
Badulu Oya Band.	10.6	0.7	Pundalu Oya	14.9	0.35
UO Welimada Rd 206	10.7	0.5	Rantambe	15.9	5.85
UO Welimada Rd 208	10.7	0.4	Mada Ela Kandy	16.1	0.15
Polgolla Tunnel	10.8	1.9	Makandura Oya	16.5	3.2
UO Welimada Rd 207	10.9	1	Victoria left bank	17.8	3.15
Kurudu Oya	10.9	1.96	Huna Oya	17.8	2.3
Kotmale left bank	11.2	0.8	Galmal Oya	18.8	0.3
Ferland Es St	11.6	2.45	Maldeniya Oya	19.3	4.5
Randenigala Reservoir	12.0	5.7	Hulu Ganga	19.5	0.7
Katabula Es St	12.4	1.5	Watehena Oya	20.3	3.2
Mount Vernon Es St	12.4	0.1	Pinga Oya	21.4	1.85
Uma Oya St 213	12.6	0.35	Gatembe Br	21.5	1.7
Uma Oya St 212	12.9	0.05			

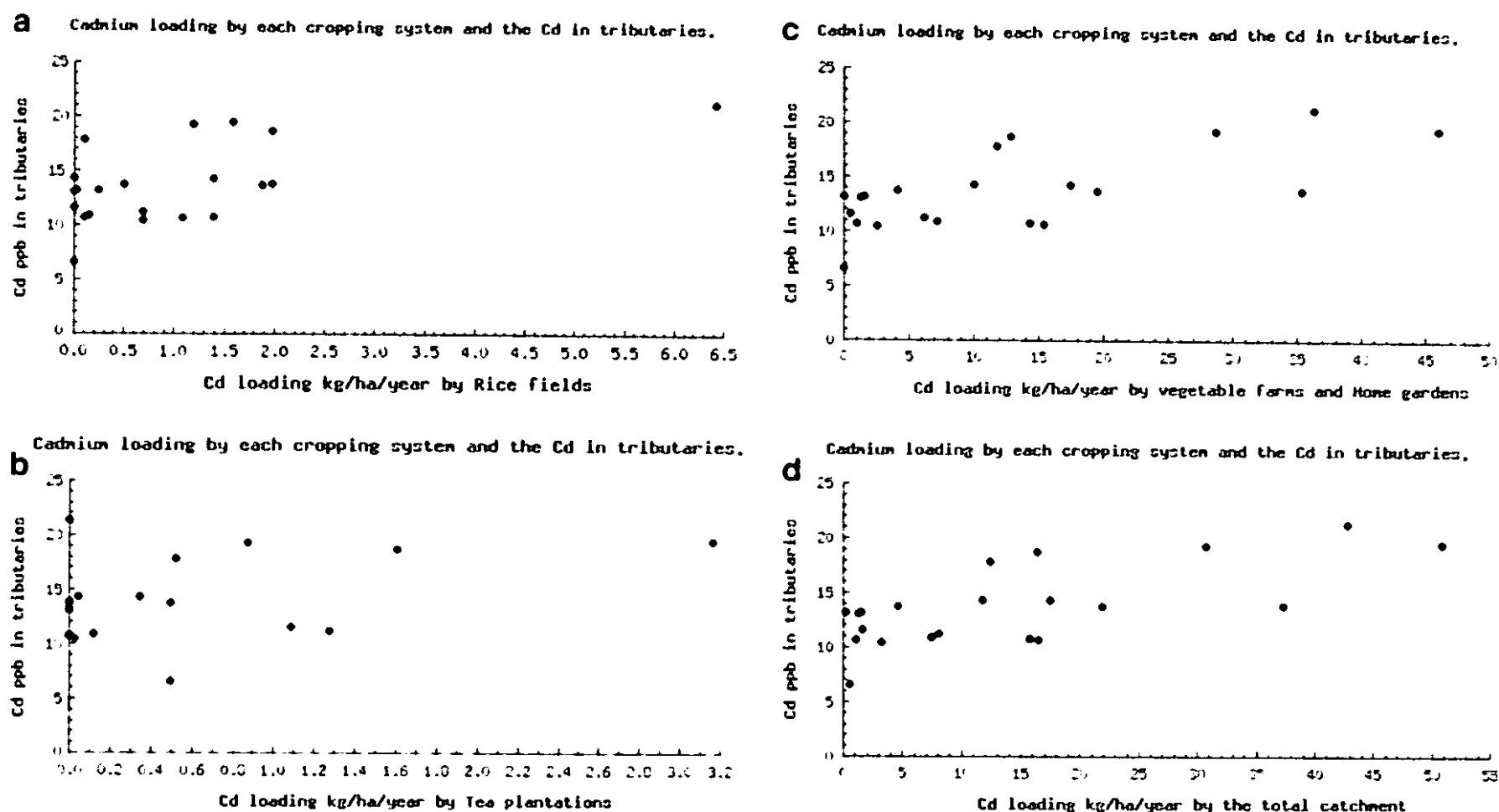
kinetics previously used to describe metal release due to proton-induced mineral dissolution. At a given pH, the relative release rates were highest for Cd, followed by Cu and Pb. Their test system provided information on how short-term (initial release), long-term (release rates), and pH-dependent release are controlled by the kinetics of the release processes. Therefore, the relative release rates of heavy metals bound to soil depend not only on pH and carbon content in soil but also on the strength of metal bonding to soil materials. In addition to the factors mentioned so far, Krachler et al. (2005) introduced a further possibility of interaction between biochelators in soil and heavy metals that affects the Pnal release of Cd from the catchment to tributaries. Therefore, it is possible to expect high but variable release of cadmium into solution with high rainfall in the catchment and that it may not exhibit a high

correlation with the fertilizer regimes practiced in the cropping systems in the watershed.

#### Cadmium mobility

The mobility and fate of heavy metals in the soil environment are directly related to their partitioning between soil and soil solution. The presence of  $\text{Cd}^{2+}$  in the solid phase is a result of precipitation and adsorption to components of the soil, processes that are highly pH dependent (Lee et al.1996).

The exchangeable Cd level in upcountry soil varied from 0.32 to 1.24 mg/kg, and the mean cation exchange capacity ranged from 11.7 in uncultivated soil to 36.4 cmol(+) per kg in soils cultivated with vegetables (Premarathna 2006). With an altitude up to 2,000 m and a total catchment area of 316,000 ha in the upper catchment and a total runoff per annum



**Fig. 5** a Scatter plots of cadmium content in tributary ( $\mu\text{g/l}$ ) and cadmium loading from rice fields in the respective catchment ( $\text{kg Cd/ha/year}$ ). Rank correlation coefficient Kendalls  $\tau=0.339$  and two-sided  $p=0.038$ . b A scatter plot of cadmium content in tributary in ( $\mu\text{g/l}$ ) and cadmium loading from Tea plantations in the respective catchment as ( $\text{kg Cd/ha/year}$ ). Rank correlation coefficient Kendalls  $\tau=0.441$  and two sided  $p=0.043$ . c A scatter plot of cadmium content in tributary in ( $\mu\text{g/l}$ ) and cadmium loading from vegetable

cultivations in large scale farms and home gardens in the respective catchment as ( $\text{kg Cd/ha/year}$ ). Rank correlation coefficient, Kendalls  $\tau=0.447$  and two sided  $p=0.003$ . d A scatter plot of cadmium content in tributary in ( $\mu\text{g/l}$ ) and cadmium loading from the whole catchment as a total of Tea, Rice, vegetable cultivations in large scale farms and home gardens in the respective catchment as ( $\text{kg Cd/ha/year}$ ). Rank correlation coefficient, Kendalls  $\tau=0.480$  and two sided  $p=0.002$

**Table 3** Chemical properties of uncultivated soil and those soils under intensive vegetable cultivation in the WU3 sampling region

Soil	pH	OM (%)	EC (dc/m)	CEC (cmol/kg)	Available (mg/kg)			Cadmium (mg/kg)	
					N	P	K	Total	Exch
Vegetable	5.2	1.47	1.50	18.0	64.8	442.8	515.6	1.96	0.48
Uncultivated	5.1	0.40	0.10	11.7	12.0	49.0	214.1	0.51	0.26

OM organic matter, EC electrical conductivity, CEC+ cation exchange capacity, Exch exchangeable, Food and Agriculture Organization (FAO)/United Nations Educational, Scientific, and Cultural Organization (UNESCO) soil taxonomy Humic Alisoils Adapted from Premarathna (2006) and Soil Science Society of Sri Lanka publications

amounting to  $25.9 \times 10^9 \text{ m}^3$  that is 65% runoff which eventually builds up to a monthly flow of water at Polgolla (457 m altitude, lowermost point in River Mahaweli prior to diversion, under consideration in this study) in River Mahaweli to 321 million cubic meters in October, preceding month from the time of sampling and 284 million cubic meters in November and 218 in December the sampling months (NEDECO 1979). Loganathan and Hedley (1997)

showed that, in areas where precipitation is heavier as in the upper catchment area of Mahaweli, 90% of the TSP applied would be leached away by runoff water. The Cd level at Polgolla Diversion Canal to NCP was found to be  $10.8 \mu\text{g/l}$ . At average flow of 6.679 million cubic meters per day and rate of  $10.8 \mu\text{g/l}$ , the potential transfer of cadmium from upper Mahaweli water to NCP from Polgolla alone is 72.13 kg/day. The total cadmium diverted to reservoirs along with

the cadmium input generated from irrigated TSP-fertilized crop fields (rice and vegetables) in NCP eventually settles in the sediments of reservoirs ( $1.77\text{--}2.45$  mg/kg) (Bandara et al. 2008). These sediments eventually release  $\text{Cd}^{2+}$  into reservoir water ( $32\text{--}57$  mg/l). In the ground water from upcountry wet zone (WU1DW3) we observed calcium ( $\text{Ca}^{2+}$ ) content of  $6.09\text{--}39.87$  mg/l, and  $310\text{--}1,200$  mg/l in the dry zone. Though Cd is retained in soil by exchange reactions, in the presence of competitive cations such as  $\text{Ca}^{2+}$ , which is more prevalent in the dry-zone soils, it competes better than Cd for adsorption sites (Gomes et al. 2001). However, with the varying water levels of the reservoir during drier periods, where reducing conditions prevail, solubility of Cd is higher under alkaline pH. Under the prevailing conditions ( $\text{F}^-$  content in dry zone waters is  $1\text{--}4$  ppm, Herath et al. 2005; we also observed that  $\text{F}^-$  content in well water in the dry zone is  $300\text{--}9,780$   $\mu\text{g/l}$ , whereas in the wet zone it is very low at  $40\text{--}700$   $\mu\text{g/l}$ ), the Cd trapped in sediments underwater forms soluble complexes with inorganic and organic ligands, particularly  $\text{F}^-$  and  $\text{Cl}^-$  (Onyatta and Huang 2008), to increase the mobility of  $\text{Cd}^{2+}$ , which results in higher cadmium levels in food crops, leading to elevated dietary cadmium (Williams and David 1973) in NCP and the rest of the island fed by their agricultural products.

The total cadmium that is carried to reservoirs in NCP from both the cadmium input from TSP-fertilized crop fields (rice and vegetables) in NCP and the diverted River Mahaweli and that eventually settles in the sediments of reservoirs, as reported by Bandara et al. (2008), amounts to  $1.77\text{--}2.45$  mg/kg on dry weight basis. These sediments then release  $\text{Cd}^{2+}$  into reservoir water, leading to high level of Cd in irrigation and drinking water. The dissolved Cd in reservoir waters ranged from  $0.03$  to  $0.06$  mg/l, which is a  $10\text{--}20$ -fold increase over the maximum contamination level ( $0.003$  mg/l) defined by the WHO for drinking water. It was observed that all sources of water were contaminated with cadmium, and the main source for all supplies is reservoir water. The geometric mean cadmium content in drinking water samples collected from domestic environments of CRF patients based on the source were reservoir water= $3.174\pm 4.658$ , shallow well= $6.931\pm 4.747$ , and agro well= $11.18\pm 2.782$   $\mu\text{g/l}$  (Bandara et al. 2010).

Bandara et al. (2010) reported that mean Cd levels in uncultivated soil in the districts of Anuradhapura and Polonnaruwa of NCP were  $0.023\pm 0.014$  and  $0.0052\pm 0.0043$  mg/kg, respectively. Mean Cd content in cultivated soil on which TSP is applied is  $0.1104\pm 0.186$  in Anuradhapura and  $0.0159\pm 0.005$  mg/kg in Polonnaruwa. Soils of NCP are not naturally high in Cd, but cadmium is added to soil through contaminated agricultural inputs. A detailed study of cadmium transfer from contaminated phosphate fertilizer used in lowland rice farming and the resultant provisional tolerable weekly intake (PTWI) by the farmer community in NCP was reported by Bandara et al. (2008). Cadmium contamination in the most common food crops grown under lowland condition, namely rice and *Nelumbo nucifera* (lotus) rhizomes, in the region where CRF is prevalent were reported to be significantly higher than in other regions of the island. The cadmium content in rice grains collected from farms of CRF patients in NCP ranged from  $0.001$  to  $0.194$  mg/kg dry weight, with mean of  $0.0404\pm 0.0196$  mg/kg, compared with a Sri Lankan background value of  $0.001$  mg/kg. Rhizomes of 120-day-old *N. nucifera* in NCP reservoirs carried mean concentration of  $252.82$  mg Cd/kg of rhizome (Bandara et al. 2008). Cadmium content in fish raised in freshwater reservoirs in the region significantly influenced the total dietary cadmium. Tilapia fish (*Oreochromis niloticus*) collected from Thuruwila Reservoir near Anuradhapura had  $0.425$  mg Cd/kg (Bandara et al. 2008). In subsequent studies on Cd residues in tilapia and lula fish (*Channa striata*) raised in NCP reservoirs fed with River Mahaweli water, Bandara et al. (2010) observed that the Cd content ranged from  $0.5$  to  $90.7$   $\mu\text{g/kg}$  with mean of  $21.8$   $\mu\text{g/kg}$  in herbivorous tilapia compared with carnivorous snakehead *C. striata* (lula) with Cd content of  $1.2\text{--}114.4$   $\mu\text{g/kg}$ .

Therefore, it is evident that long-term use of Cd-polluted TSP has contributed to excessive levels of Cd in River Mahaweli and in turn in the reservoirs in NCP. It is also possible to reduce the runoff Cd level to acceptable levels recommended by the US EPA of  $1\text{--}5$  ppb by using ERP as used in tea estates or possibly as compost prepared with ERP.

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