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GEOCHEMICAL SPECIATION AND EXPOSURE ASSESSMENT OF HEAVY METALS IN THE MAINSTREAM AND TRIBUTARIES OF THE CARONI RIVER SYSTEM, TRINIDAD: CASE STUDY FROM A DEVELOPING COUNTRY

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ABSTRACT

Trinidad and Tobago is a fast-developing country and industrial growth can impact negatively sometimes by increasing the chance of pollution if proper measures are not obeyed. Caroni River and its major tributaries flow through the most urbanized and industrialized part of this country and perhaps this increases the opportunity of polluting the river basin and its catchment area. Till now, no detail assessment has been carried out in this area to measure the accumulation indices and associated risks. This study is one of the initial efforts to investigate and the distribution and potential sources of heavy metal pollutants (Cr, Ni, Cu, Zn, As and Pb) in sediments and water from the Caroni River system. The degree of metal contamination is compared with the global standards (International Sediment Quality Guidelines, ISQG and Water Quality Guideline, IWQG). Results from the study revealed that the concentration (in µg/L): Cr (5.07-6.85), Ni (5.23-9.38), Cu (4.56-10.11), Zn (2.13-60.91), As (3.97-5.7) and Pb (4.04-9.92). Profiles of these

metals in the river bed sediment samples (Cr: 4.17-12.69, Ni: 5.0-26.86, Cu: 4.28-46.94, Zn: 49.22-161.4, As: 5.24-12.7 and Pb: 3.55-42.43; in µg/g) were below international Probable Effects Levels (PELs) at all locations, but some metal levels were above the international Threshold Effect Levels (TELs) in a few occasions (Ni, Cu, Zn and As). The geoaccumulation index order for the river sediments is As>Pb>Zn>Cu>Ni>Cr. This study also revealed that these higher concentration can be attributed to effluents from small-scale industries, unnoticed irrigation processes, and domestic wastes. Though it would be too early to make predictions, especially before more detailed study, the San Juan, St. Joesph and Arouca Rivers (three important tributaries of the Caroni River) were found to be the most polluted, and Cu, Pb, Zn and

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As were the most abundant metals in the study area. It is recommended that appropriate measures are taken by the relevant authorities to mitigate the problems at this early stage.

Keywords: Heavy metals, Trinidad and Tobago, river pollution, sediment-water quality

1. INTRODUCTION

River water quality plays an important role in the survival of aquatic life as well as of land organisms and therefore, anomalous concentrations of heavy metals in rivers (both in the sediment and water) that may have the potential to affect the river water quality should be controlled and regularly monitored (Vardaki and Kelepertsis 1999; Boszke et al. 2004; Zarcinas et al. 2004). Sediments are good indicators of heavy metal contamination as more than 90 percent of the heavy metal load in aquatic systems can accumulate in suspended particulate matter and sediments (Calmano et al., 1993). Sediments influence river water quality and can be used to determine the history of river pollution. Heavy metals are introduced into rivers naturally by rock weathering, soil erosion and dissolution of water-soluble salts or can be introduced by human activities such as mining, industrialization and urbanization.

Numerous studies on heavy metal water pollution in have been carried out in different parts of the world, such as, on the Lower Salado River Basin in Argentina (Gagneten et al. 2007), the Delimi River in Nigeria (Sabo et al. 2013), and the Gomti River in India (Singh et al. 2005). Similar studies were also carried out in some rivers of Trinidad and Tobago (Mohammed et al. 2000; Norville 2005; Surajdeo-Maharaj et al. 2007; Ramsingh 2009; Kanhai et al. 2013). Most previous studies indicated possible impacts of urbanization and industrialization on the total pollution level of this country (WRA, 2001) However, these studies were not focused on the distribution pattern or the enrichment factor or the source characterization of these metal pollution in this major river system in particular.

The purpose of this study was to: (a) investigate heavy metal contamination (Cr, Ni, Cu, Zn, As and Pb) in the river-bed sediments and river water in the Caroni River catchment; (b) assess the degree of contamination and potential risk factor using the Geoaccumulation (I-geo) Index and Contamination Factor (CF) Index; and (c) identify the possible sources of heavy metals.

2. STUDY AREA

The Caroni River is the largest river in Trinidad and Tobago and it provides potable water for over 40 percent of the country's population (EMA 2005). It is situated in the northern western section of the island, originates in the Northern Range and runs through the Caroni Plains and drains into the Gulf of Paria (Fig. 1). The basin drains both the Ranges; Northern and Central, and is estimated to be 600 km2 in catchment area (Juman and Ramsewak 2013). The Caroni

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River is drained by eighteen (18) rivers, of which twelve rivers flowing in from the Northern Range and six rivers which flow into the basin from the Caroni Plain and Central Range. Nathai-Gyan and Juman (2005) proposed that the major contribution comes from the twelve (12) northern rivers whilst the lesser contribution comes from the southern rivers.

The soil type in the study area varies from fine sandy clay and clay in the eastern part to sandy loam in the west (Table 1). Similar sequence also found in the underlying bed-rock with shale-siltstone and sandstone to calcareous phyllite to calcareous sandstone (Table 1).

3. MATERIALS AND METHODS

3.1 Sample collection

Depending on the topography, type of vegetation, soil type, industrial establishment, population density and accessibility, thirteen (13) locations were selected along the Caroni River and its seven (7) important tributaries (Fig. 1; Table 1). Sediment and water samples were collected from each location along the following rivers: Aripo River (L1), Cumuto River (L2), Tumpuna River (L3), Arima River (L5), Guanapo River (L6), Mausica River (L9), Arouca River (L10), Tacarigua River (L11), St. Joseph River (L12), San Juan River (L13) and three (3) locations along the Caroni River (L4, L7, and L8).

All samples were collected in month of January and February 2014. Thirty nine (13 locations × 3 replicates) moist top-soil samples (~ 500 gm each) were collected (from 0-20 cm depth) using a metal scoop and stored in polyethylene bags for analysis. Similarly thirty nine (13 locations × 3 replicates) river water samples (~ 500 ml each) were collected (at the depth of nearly 30 cm below the surface) using 500 ml Naglene high-density polyethylene (HDPE) bottles and 400 ml polyethylene terephthalate (PET) bottles for heavy metal analysis and physiochemical analysis respectively. All sediment samples were divided into two segments after oven-drying (at 105□C for 24 h), crushing and sieving (on a 2 mm test sieve) at the Laboratory. Water samples for metal analysis were acidified using 1.5 ml of nitric acid.

Between each sampling session, all equipment was washed with distilled water and stored in properly distilled water containers. Each HDPE bottle was stored in a cooler immediately, while all PET bottles were stored at room temperature.

3.2 Laboratory analysis

Determination of physiochemical parameters

The standard pH test was carried out using an electronic pH meter (Model 51910, Platinum Series pH Electrode, HACH, Loveland, Colorado, USA). All pH measurements were performed

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at room temperature after purifying pH probe using distillation before every measurement and the pH meter was calibrated against standard solutions. pH test of soil samples were carried out in accordance with 9.5 BS 1377: Part 3: 1990 using the electrometric method while water samples were analysed in accordance with the EPA standard testing method EPA 150.1 (USEPA 1983a).

Turbidity of the water samples was determined using a 2100N Laboratory Turbidimeter, EPA, 115 Vac (HACH, Loveland, Colorado, USA). All turbidity tests were performed in accordance with EPA Method 180.1 (USEPA 1983b).

Organic content was measured in accordance with clause 4 of BS 1377: Part 3: 1990 using the mass loss on ignition method. For all these tests, duplicate samples were tested and a mean value was obtained for accuracy (BSI 1990).

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Heavy metal analysis

Sediment and water samples were analysed according to USEPA Method 200.8 using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) by the Department of Chemistry, University of West Indies, Trinidad and Tobago. ICP-MS is an analytical technique used to determine total recoverable element concentrations in soil and liquid matrices. This analytical method is appropriate for environmental samples as it is a powerful tool for rapid determination of elements and quantification of metals present in natural waters at trace levels with excellent determination limits (Voica et al. 2012; Al-Rimawi, et al. 2013). In the present study, ICP-MS was used to determine the elemental concentrations in collected sediments for chromium, cobalt, nickel, copper, zinc, arsenic and lead. Sediment samples were homogenized, digested and diluted in 2% Nitric Acid (HNO3). To determine the dissolved analytes in the surface water sample, the acid-preserved sample was filtered and an aliquot of 20 mL was pipetted into a 50 mL centrifuge tube. A volume of (1+1) HNO3 was added to make the acid concentration an approximate 1% (v/v) HNO3 solution.

A 5 ppb solution of all the elements was ran to check the calibration of the spectrometer. The mean metal concentration was obtained for all the samples by finding the average of three readings from the spectrometer. The instrument detection limits are 0.04, 0.07, 0.004, 0.07, 0.02, 0.015, and 0.03 for chromium, nickel, copper, zinc, arsenic and lead (Creed et al. 1995).

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3.3 Contamination Factor (CF) in the sediment samples

The degree of contamination is often expressed as contamination factor (CF)

$$CF = \frac{C_{metal}}{C_{bacground\ value}}$$

Where C_{metal} is the heavy metal concentration and $C_{bacground\ value}$ represents the average background values of the element in sediment (Sabo et al. 2013). The geochemical background values in shale of the metals under investigation are (Cr – 71 µg/g, Ni- 49 µg/g, Cu –32 µg/g, Zn – 127 µg/g, As – 7.9 µg/g, and Pb – 16 µg/g; earlier reported in Earth Reference Database (https://earthref.org/ERR/361). The geo-accumulation index consists of four grades or levels; CF value of <1(practically low contamination), 1 ≤ CF ≥3 (moderate contamination), 3 ≤CF ≥ 6 (considerable (high) contamination) and > 6(very high contamination) (Nasr et al. 2006).

3.4 Geo-accumulation Index (I-geo)

This index measures the level of metal pollution in sediment. I-geo was determined by the following equation (Müller 1969; Boszke et al. 2004)

$$I - geo = log2(\frac{C_n}{1.5b_n})$$

Where C_n is the measured concentration of the element in the sediment, b_n is the background value of the trace element and 1.5 is the background matrix correction factor due to lithologic effect. The I-geo consists of seven grades or classes; I-geo values <0 (practically unpolluted), 0 ≤ I-geo ≥ 1 (unpolluted to moderately polluted), 1 ≤ I-geo ≥ 2 (moderately polluted), 2 ≤ I-geo ≥ 3 (moderately polluted to highly polluted), 3 ≤ I-geo ≥ 4 (highly polluted) and > 5 (very strongly polluted) (Müller 1969).

3.5 Statistical analysis

Basic Statistical Analysis, Principal Component Analysis (PCA) and Correlation Analysis were carried out for all observed data using the XLSTAT (2014) program. Correlation Analysis was used as a sorting mechanism of the information of a correlation matrix (Symader 1977), while Principal Component Analysis was used to analyse the correlation in heavy metal concentrations at thirteen selected locations. Pearson's Correlation analysis was applied to quantitatively analyse and determine the relationship between the heavy metals in the water and sediment samples (after Qishlaqi et al. 2007).

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4. RESULTS

Results shown in Table 2, 3 and 4 represent the mean values calculated from the three replicate samples at each location.

4.1 Water Analysis

Table 2 shows the values of pH and turbidity in the water samples collected from the study area. There was significant variation ($p \leq 0.05$) in the recorded pH level. However, all pH values varied between 6.5 – 8.5 (which is the normal range of surface water; APHA 1989). It was observed that all northern tributaries of the Caroni River showed pH levels higher than the pH levels of its southern tributaries. All measured turbidity levels of the river water were below the acceptable range of turbidity (lower than 50 NTU) for streams (North Carolina Code 2003). Moreover, statistically no significant difference ($p > 0.05$) was observed in the measured turbidity values.

Table 3 presents the values of heavy metal content in water samples obtained from the study area. No significant statistical difference was observed in Cr-concentration. The water samples collected from all locations showed similar mean (6 µg/L), except for that from from the Arouca River which had significantly higher value (12.85 µg/L). There was significant variation ($p \leq 0.05$) in Ni-values in the measured water samples. Higher concentrations (mean value of 8.4µg/L) were observed in water from the tributaries of the Caroni River flowing in the western part of the study area, while statistically low but similar values (mean 6.0 µg/L) were observed in all other locations. Difference in locations had a significant influence ($p \leq 0.05$) on measured Cu-levels. Higher Cu-values (mean 8.7 µg/L) were found in the Guanapo, St. Joseph, and San Juan Rivers, while statistically lower values (mean 5.8 µg/L) were found in all other locations. Variation in Zn-concentration in the river water samples was also observed ($p \leq 0.05$). The Cumuto and Guanapo Rivers as well as swamp locations near the Caroni River showed higher levels of zinc with a mean of 59.5 µg/L. Except for significantly lower values in the Tacarigua, Arouca and St. Joseph Rivers (mean 4.7 µg/L), all other locations showed an average level of 30µg/L. Statistically, no significant differences ($p > 0.05$) were observed in measured As-values (mean 4.9) in river water of the study area. Difference in the sampling locations had a significant influence ($p \leq 0.05$) on observed Pb-level in river water. Higher Pb-values (mean 8.4 µg/L) were found in water from nearly all northern tributaries of the Caroni River, while lower Pb-values (mean 4.8 µg/L) were found in water from its southern tributaries.

4.2 River Bed Sediment Analysis

The analysed level of pH and organic matter in the sediment samples obtained from the study area are presented in Table 2. Data analysis indicated that there was a significant difference in

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the observed pH values of sediment samples collected from the study sites. River bed samples in the Cumuto and Tumpana Rivers were acidic in nature while all other locations were observed to have slight to moderately alkaline river bed sediments (after Soil Survey Manual 1993). There was significant variation in the organic matter content in the river bed sediments ($p \leq 0.05$). Statistically low levels (mean 1.3%) were recorded in nearly all northern tributaries of the Caroni River while relatively high levels (mean 3.6%) were recorded in nearly all southern tributaries as well as the swamp locations near the Caroni River. A significantly high organic content (9.7%) was observed in the Arouca River.

The distribution of total metal concentrations in the river bed sediments obtained from the study area is presented in Table 4. Significant differences were noticed in the Cr-values in the studied rivers ($p \leq 0.05$). Low Cr-values (mean 6.2 µg/g) were recorded in Aripo, Guanapo, Tacarigua and St. Joseph rivers, while statistically highest level of Cr was observed in the sediment of the Cumuto River (18.72 µg/g) while moderately higher concentrations (10.5 µg/g) were found in the sediments of the rest of the studied rivers. Ni-content in the study area showed significant variation, with the values ranging from 5.0 µg/g (at the Aripo River) and 26.86 µg/g (at the San Juan River). Low values (mean 11.7 µg/g) were observed in the sediments from Cumuto, Tumpana, and the Caroni Rivers, whereas all other river bed sediments showed high co-concentration (mean 20.3 µg/g). Distribution of copper in the sediments of the study area was similar to the distribution of cobalt, with a high mean value of 9.2 µg/g and low mean value of 25.5 µg/g for copper. Significant variation ($p \leq 0.05$) was also observed in the Zn-concentration from various locations. High values (mean 89.5 µg/g) were observed in the sediments from the Arima, Tacarigua and St. Joseph Rivers, whereas all other locations showed lower concentration of zinc (mean 62 µg/g) except for the Aruoca and San Juan Rivers (where the mean values were 161.4 and 124.6 µg/g, respectively). There was statistically significant difference in the measured As-level ($p \leq 0.05$). Low range of metal concentrations (mean 7.0µg/g) were observed in the sediments of the Aripo and Tumpana Rivers, and in three locations of the Caroni River, while higher range of concentration (mean 10.5 µg/g) was observed in sediments from other locations of the study area. There was significant variation in the Pb-content ($p \leq 0.05$), with the minimum value recorded in the sediments from the Aripo River (3.55 µg/g) and maximum values observed in the Arouca and San Juan Rivers (mean 34.4 µg/g); all other locations showed moderate Pb-concentrations (mean 13.5 µg/g).

Contamination Factor (CF) and Geoaccumulation Index (I-geo) calculated from the metal contents in the sediment samples are given in Table 5(a) and 5(b), respectively. The least values of Contamination Factor (CF) were recorded for Cr and Ni with CF values (ranging from 0.04 to 0.5) in nearly all the locations, whereas moderate values were observed for Cu and Zn. Highest CF values found in the locations ranged from 0.6 to 2.65.Low Geo-accumulation Index (I-geo)

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values were recorded for Cr and Ni (with the values ranging from 0.02 to 0.10) in nearly all the locations and moderate values were observed for Cu, Zn and Pb. In nearly all the sampling stations, high I-geo values were recorded for As with CF values (ranging from 0.11 to 0.32). However, the maximum I-geo value was calculated from Pb in Arouca River (I-geo value 0.53) and it was the highest index in the study area.

4.3 Statistical Analysis

The Pearson correlation coefficient values (r), which shows the measure of the linear association between the river water and sediments collected from various sampling stations, are presented in Table 9. This measure of correlation showed statistically significant variations ($p \leq 0.05$). Very strong relationships (r value more than 0.9) were observed in the Cumoto, Tumpana, Guanapo, Mausica and St. Joseph Rivers as well as in all the locations along the Caroni River. Moderate to good correlation was found in the Aripo, Arima and San Juan Rivers. In the Arouca River, bed-water correlation was negative and moderate.

The Principal Component Analysis (PCA) was used with the two factors (F1 and F2) for all sample frames and the score plot was used for interpreting relations among the observations. In water samples, the factor loadings F1 and F2 (Eigenvalues 5.70 and 1.51, respectively) accounted for 69.96% of the initial variability. These factors had individual weights of 38.73% and 31.23%, respectively, and the analysis of the observations for F1 and F2 for all locations is depicted in Figure 2. The PCA chart suggests that the San Juan (L13), St. Joseph (L12), Arouca (L10), Mausica (L9), and Tacarigua (L11), Rivers were different from the other locations, as they showed elevated heavy metal concentrations in the river water. The Aripo (L1), Tumpana (L3) and Guanapo (L6) were also distinct in the PCA plot (Fig. 2) in that the water samples from these rivers were recorded with the lowest heavy metal concentrations; in these rivers, with Guanapo River showing the maximum zinc concentration. River-bed sediments F1 and F2 (Eigenvalues 4.47 and 1.68, respectively) accounted for 76.82% of the initial variability of the data, with individual weights of 55.84% and 20.98%, respectively. The PCA observations at the location points are depicted in Figure 3. The chart (Fig. 3) indicates that sediments from the San Juan (L13), St. Joseph (L12) and Arouca (L10) were different from those in the other locations due to the elevated heavy metal concentrations at these locations. The Aripo (L1) and Cumuto (L2) were also seen to be different from the other locations, as Aripo (L1) River had the lowest heavy metal concentrations and Cumuto (L2) River was observed with maximum chromium concentration on the PCA chart.

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5. DISCUSSION

The results of this study indicate that the water in nearly all northern tributaries of the Caroni River are slightly alkaline (pH more than 7.5), while its southern tributaries and the Caroni River itself have neutral pH. The level of pH in aqueous solution affects the leaching power of water and indicates its chemical reaction on sediments. The Northern Range of Trinidad, which is the source of the river and which engulfs majority of the catchment areas of the northern tributaries, consists of many limestone bodies. The alkalinity in these northern tributaries may be due the presence of these limestone bodies. The measured pH level in water samples from the present study falls within the normal range (6.5-8.4) for irrigation water (FAO 1994). Water with pH values outside the normal range of 6.5-8.4 may contain excess toxic elements posing serious threat to aquatic life (APHA 1989).

The trend of the turbidity distribution in the studied river water samples was similar to the trend of pH level in the water samples. This is due to the fact that the northern tributaries of the Caroni River have higher chances of water replenishment (by the water supply from the Northern Range), while the southern tributaries flowing in the plains have less likelihood of getting water. Turbidity indicates the presence of suspended materials in the water and a high level of turbidity can reduce the aesthetic quality of lakes and streams in a significant manner. The turbidity values from all the areas studied were below the acceptable limit.

The concentration of heavy metals in river water (Table 3) was below the upper permissible values determined by some important international recognized authorities (Smith and Carson 1981; Hamilton 1994; Vardaki and Kelepertsis 1999; CWQG 2001; EMA 2001; USEPA 2002; Nagpal 2004). Water samples from nearly all northern range tributaries showed enriched Cr-, Ni- and Cu-values, especially in San Juan, St. Joseph and Tacarigua rivers with the highest values observed in the San Juan River. Though the watershed areas which feed these rivers, do not have heavy industrial industries, there are a number of light industries there which could be contributing to these metal enrichments. These small-scale activities include chrome-plating outlets, garment industries, metal works factories, electroplating & electrical parts factories, and food-beverage manufacturing units. Significantly, different distribution patterns were observed in Zn-, As- and Pb-concentrations. Water samples from all the locations showed the maximum values of these metals except those from the Mausica, Arouca and Tacarigua Rivers. These values were also comparatively higher in the San Juan and St. Joseph Rivers, while maximum Pb-concentration was observed in the San Juan River. High concentrations of these metals may be the result of the wastes from the small-scale paint, battery recycling, electroplating, chemical cleaning industries in the catchment of these rivers. Unplanned use of agricultural fertilizers and

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pesticides, and unorganized dumping of domestic waste could also be contributing to the heavy metals in the river catchment areas.

Most of the sediment samples were slight to moderately alkaline except in the Cumoto and Tumpana Rivers, where sediments were acidic in nature (pH $\leq$ 7). The acidic nature of the sediment at the study area is an indication that certain metals can easily be desorbed from the sediment into the water, thereby increasing the chances of more bioaccumulation. The textural class of the ambient source soil of all bed sediments (except in the two above-mentioned rivers) are alkaline (rich in calcium), which could be responsible for the observed pH in these sediments. The ,northern tributaries of the Caroni River have relatively lower organic matter content in sediments, which means that these metals can easily form a metal-organic matter complex by releasing into the water column. Organic matter in sediments are supposed to control the adsorption and retention of heavy metals.

It was revealed from the sediment analysis that the recorded heavy metals have high values along the northern part of the study area, especially in San Juan River compared to other locations. The present findings of heavy metal concentration in the sediments from the Caroni River and its tributaries were compared to findings for some heavy metal concentration in the rivers of Trinidad and Tobago (Surujdeo-Maharaj et al., 2007). It was found that Cr-, Cu- and Pb-concentrations observed by the present study were below the previously recorded average values of these metals (25.12, 31.77 and 46.29 µg/g, respectively), while Ni-values (in the San Juan, St. Joseph and Arima Rivers) and Zn-values (in the Aruoca and San Juan Rivers) were higher than their previous observations (21.69 and 111.55 µg/g, respectively). No previously recorded level of arsenic from the river sediments were found for this country. To evaluate sediment chemistry data, which are frequently used to support ecological risk assessments, numerical sediment quality guidelines (SQGs) are often used, such as Probable Effect Levels (PELs), and Lowest Effect Levels (LELs) or Threshold Effect Levels (TELs) (USEPA, 2005). LELs or TELs represent chemical concentrations below which adverse effects upon sediment dwelling fauna would be expected infrequently, while PELs are concentrations above which adverse effects are likely to occur (Long and MacDonald 1998). Based on the recorded activity level from the present study and the TELs/PELs (ISQG Ontario 1993; ISQG Canada 1999) comparisons (Table 4), it was observed that all highest values of the heavy metals were above their respective LEL values but well below the PEL values. Dumping of different waste materials such as paints, electrical and electronic materials, metal scraps, and old batteries from nearby small-scale industries as well as unnoticed accumulations from irrigation water and domestic waste may be responsible for the high level of heavy metals in the sediments.

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Inter-element relationships in sediments provide information on metal sources and pathways in the geo-environment (Dragovi et al., 2008). Strong correlation was observed between all metals except chromium, which suggest common sources of these enrichments and common movement patterns (Bhuiyan et al., 2009),

The CF values for As and Pb were highest for all the analysed metals. The CF values also indicate that Cu and Zn had minor to moderate enrichment and others had minor enrichment. Generally, the sampling points which were located closer to the different industries showed higher CF values. Differences in the CF values may be contributed by the anomaly in the input factor of each metal in the river sediment and/or the variation in the leaching rate of each metal from the sediment (Ghrefat et al., 2011). All observed I-geo values suggested that the study area had zero to moderate contamination (class 1). However, possible addition in activity that caused moderate contamination might be due to the anthropogenic activities in the respective watershed areas. It is obvious that to deal with river contamination, correlation between river water and river sediment concentration of each metal needs to be verified. Very strong relationship was observed in most of the rivers (Table 6) and this is due to the alkaline nature of those rivers (pH> 7), which leads to precipitation of some metals (e.g., Cu, Zn, Pb etc.) and complexation reactions. Here, it is important to note here that the present study was undertaken during the dry season and that the process of metal precipitation may increase in the dry season (Rashed, 2001).

PCA analysis suggests that the metal enrichments were mainly by zinc (Zn), copper (Cu), lead (Pb) and arsenic (As). The distribution patterns show that heavy metal concentrations were considerably higher in the San Juan (L13), St. Joseph (L12) and Arouca (L10) rivers. A previous study has already indicated the St. Joseph as the most grossly polluted river in this area (Lucas and Alkins-Koo 2004) and present observations have added San Juan and Arouca as possible concerns.

6. CONCLUSION

Studied heavy metals in the river sediment and water samples from different catchment areas of the Caroni River and it major tributaries varied widely in concentration.

Heavy metal concentrations in sediments, which were marginally above the international guidelines, may be attributed to small-scale industrial activities, unnoticed agro-climatic conditions, and domestic waste as indicated by the measured CF values. Though the observed I-geo values showed that the study area is not contaminated, it suggested the possibility of future contamination in view of recent development in this country. Present observations indicate that San Juan River was the most polluted river followed by St. Joseph and Arouca Rivers. It is important to note here that these rivers flow through industrial estates and urban areas. However,

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more detail characterization of the catchment areas is required before reaching further conclusions.

Though the heavy metal concentration in river water was below the maximum allowable international standards, these metal concentrations need to be analysed to evaluate the variation during the wet season.

Very strong correlations between metal content in river water and sediments were observed and it would be interesting to examine these relationships during the wet season.

Furthermore, in-depth investigation of the area would be very pertinent to understand the tolerance level of the vegetation and aquatic ecosystem to these metals and to prevent hazards to humans and animals.

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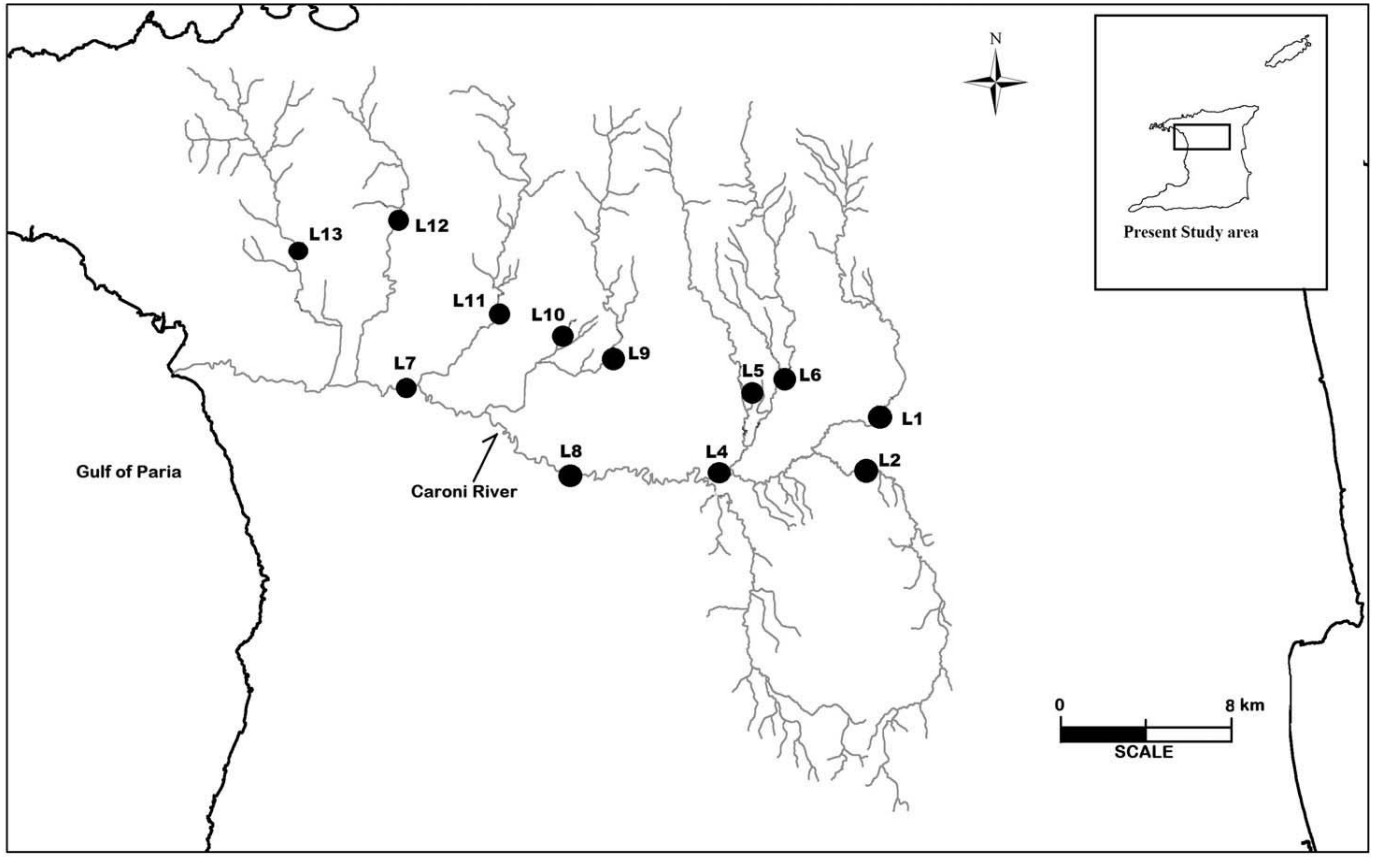

Figure. 1

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Figure 2: Principal Component Analysis of heavy metals in river water samples at the site locations

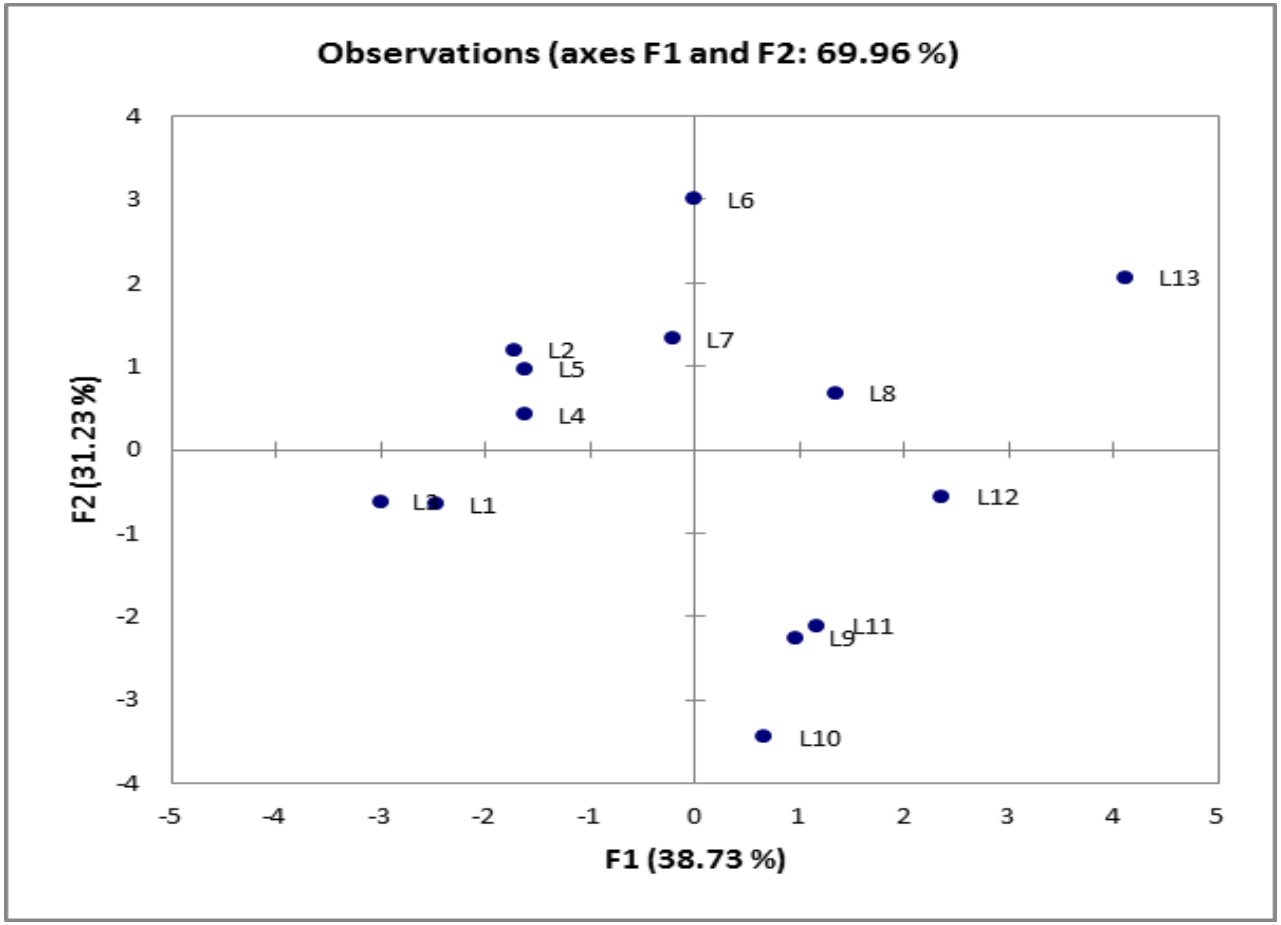

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Figure 3: Principal Component Analysis of heavy metals in river bed sediments at the site locations

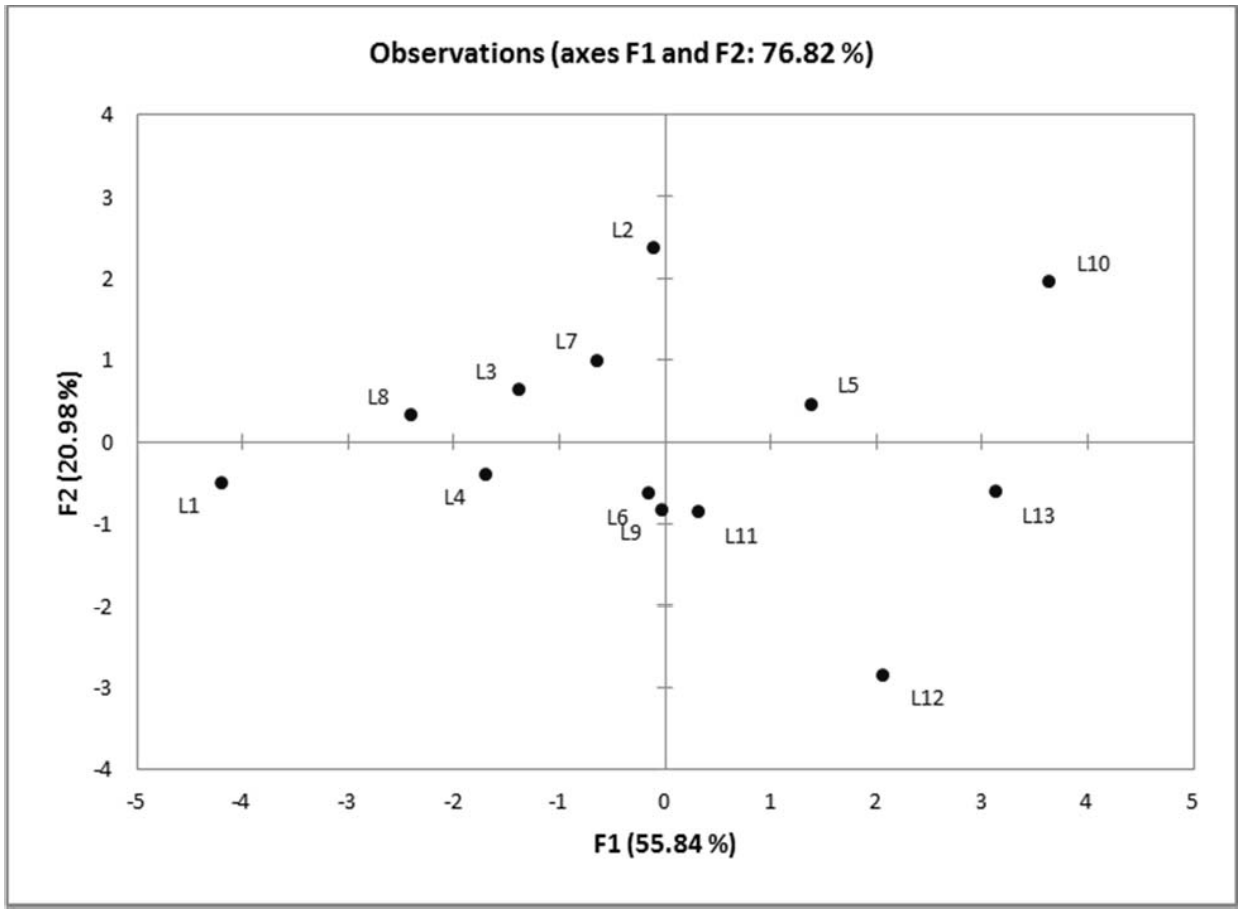

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Table 1: Province, geographic coordinates, soil classes, parent material, and textural class of the collected samples (after Trinidad Soils map, Soil & Land Capability Unit, Ministry of Agriculture, Land and Fisheries, Government of Trinidad and Tobago)

Profile	River	Coordinates	Soil class	Parent material	Textural class
L1	Aripo river	10°36'07.93" 61°12'43.25"	Valencia	Shale/ siltstone	Fine sandy clay
L2	Cumuto river	10°35'15.09" 61°13'03.06"	Ecclesville and Brasso	Shale/ siltstone	Fine sandy clay to clay
L3	Tumpuna river	10°34'30.97" 61°16'26.98"	Ecclesville and Brasso	Calcareous siltstone/ calcareous shale	Fine sandy clay to clay
L4	Caroni River	10°34'56.46" 61°16'15.01"	Ecclesville and Brasso	Calcareous siltstone/ calcareous shale	Fine sandy clay to clay
L5	Arima river	10°36'46.71" 61°15'49.02"	Piarco	Clay rich sandstone	Fine sandy loam
L6	Guanapo river	10°37'12.19" 61°15'03.97"	Piarco	Clay rich sandstone	Fine sandy loam
L7	Caroni River	10°37'03.56" 61°24'15.25"	Ecclesville and Brasso	Calcareous siltstone/ calcareous shale	Fine sandy clay to clay
L8	Caroni River	10°35'05.24" 61°20'18.17"	Ecclesville and Brasso	Calcareous siltstone/ calcareous shale	Fine sandy clay to clay
L9	Mausica river	10°37'50.98" 61°19'22.00"	Diego Martin and La Pastor	Mica rich sandstone to sandstone and limestone	Fine sandy to sandy loam
L10	Arouca river	10°38'54.60" 61°21'00.20"	Diego Martin and La Pastor	Mica rich sandstone to sandstone and limestone	Fine sandy loam to sandy clay
L11	Tacarigua river	10°38'44.84" 61°22'07.14"	Maraval	Mica rich sandstone to sandstone and limestone	Fine sandy to sandy loam
L12	St. Joseph river	10°40'56.22" 61°24'35.96"	Maracas and Matelot	Calcareous phyllite, Limestone and Micaceous phyllite	Sandy and clay loam
L13	San Juan river	10°40'13.83" 61°26'59.07"	Maracas and Matelot	Calcareous phyllite, Limestone and Micaceous phyllite	Sandy and clay loam

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Table 2: pH, Turbidity and Alkalinity in river water and river bed samples

Location	Water		Sediment	
	pH	Turbidity (NTU)	pH	Organic matter (%)
L1	6.8 ± 0.06	23.4 ± 4.1	7.5 ± 0.03	1.3 ± 0.03
L2	6.9 ± 0.24	33.2 ± 8.2	4.7 ± 0.21	3.8 ± 0.26
L3	6.9 ± 0.21	15.2 ± 2.9	5.2 ± 0.15	3.5 ± 0.34
L4	7.2 ± 0.09	6.0 ± 1.3	7.9 ± 0.14	2.9 ± 0.05
L5	8.2 ± 0.15	5.2 ± 1.2	7.9 ± 0.18	1.9 ± 0.06
L6	7.7 ± 0.28	5.9 ± 1.8	8.2 ± 0.24	0.8 ± 0.02
L7	7.0 ± 0.19	15.6 ± 2.0	7.0 ± 0.04	4.3 ± 0.04
L8	6.9 ± 0.06	16.6 ± 2.8	7.6 ± 0.04	1.5 ± 0.08
L9	7.2 ± 0.20	6.9 ± 1.7	7.6 ± 0.11	1.4 ± 0.02
L10	7.6 ± 0.07	15.5 ± 2.2	7.4 ± 0.08	9.7 ± 0.40
L11	7.5 ± 0.22	1.3 ± 0.2	7.5 ± 0.07	1.3 ± 0.08
L12	7.4 ± 0.14	34.7 ± 5.1	8.3 ± 0.04	1.7 ± 0.06
L13	7.5 ± 0.16	42.0 ± 11.5	7.9 ± 0.03	1.1 ± 0.10
Limits	**6.5 - 8.5***	**> 50 ****	**-**	**-**

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Table 3: Analyzed heavy metal concentration (µg/L) in river water sample

Location	Cr	Ni	Cu	Zn	As	Pb
L1	5.07 ± 1.01	5.94 ± 1.19	5.88 ± 1.18	32.87 ± 6.57	5.23 ± 1.05	5.15 ± 1.03
L2	6.31 ± 1.26	5.23 ± 1.05	6.37 ± 1.27	60.91 ± 12.18	5.39 ± 1.08	6.11 ± 1.22
L3	5.27 ± 1.05	5.82 ± 1.16	4.56 ± 0.91	18.07 ± 3.61	5.53 ± 1.11	4.54 ± 0.91
L4	6.17 ± 1.23	5.83 ± 1.17	5.32 ± 1.06	21.88 ± 4.38	5.04 ± 1.01	8.74 ± 1.75
L5	5.89 ± 1.18	5.97 ± 1.19	6.11 ± 1.22	34.21 ± 6.84	5.01 ± 1.00	7.14 ± 1.43
L6	6.39 ± 1.28	6.21 ± 1.24	8.56 ± 1.71	75.15 ± 15.03	4.84 ± 0.97	8.65 ± 1.73
L7	5.89 ± 1.18	7.01 ± 1.40	6.45 ± 1.29	41.13 ± 8.23	5.70 ± 1.14	5.29 ± 1.06
L8	5.54 ± 1.11	8.17 ± 1.63	8.70 ± 1.74	60.75 ± 12.15	5.36 ± 1.07	5.88 ± 1.18
L9	5.80 ± 1.16	8.25 ± 1.65	5.85 ± 1.17	4.03 ± 0.81	4.45 ± 0.89	4.70 ± 0.94
L10	5.86 ± 1.17	7.86 ± 1.57	5.33 ± 1.07	2.13 ± 0.43	3.97 ± 0.79	4.12 ± 0.82
L11	6.47 ± 1.29	7.69 ± 1.54	5.86 ± 1.17	7.93 ± 1.59	4.19 ± 0.84	4.04 ± 0.81
L12	6.50 ± 1.30	9.07 ± 1.81	7.43 ± 1.49	38.35 ± 7.67	4.53 ± 0.91	7.51 ± 1.50
L13	6.85 ± 1.37	9.38 ± 1.88	10.11 ± 2.02	35.36 ± 7.07	4.78 ± 0.96	9.92 ± 1.98
USEPA, 2002	**570**	**470**	**-**	**120**	**340**	**65**
CWQG, 2001	**10**	**C**	**C**	**30**	**100**	**C**
WRA, 2001	**500**	**500**	**500**	**200**	**-**	**100**
Other International Quality Guidelines	**50***	**50****	**100****	**100****	**50****	**50****

* - Nagpal, 2004; ** - Vardaki and Kelepertsis, 1999; *** - Smith and Carson, 1981 and Hamilton, 1994 C – Calculated by water hardness

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Table 4: Analyzed heavy metal concentration (µg/g) in river bed sediment samples

Location	**Cr**	**Ni**	**Cu**	**Zn**	**As**	**Pb**
L1	4.17 ± 1.04	5.00 ± 1.25	4.28 ± 1.07	58.17 ± 14.54	5.24 ± 1.31	3.55 ± 0.89
L2	18.72 ± 4.68	11.80 ± 2.95	9.40 ± 2.35	72.29 ± 18.07	10.30 ± 2.57	14.69 ± 3.67
L3	10.54 ± 2.63	12.99 ± 3.25	10.00 ± 2.50	76.94 ± 19.24	6.80 ± 1.70	8.81 ± 2.20
L4	6.54 ± 1.64	13.19 ± 3.30	14.15 ± 3.54	51.69 ± 12.92	7.55 ± 1.89	10.81 ± 2.70
L5	8.70 ± 2.18	21.04 ± 5.26	22.81 ± 5.70	87.58 ± 21.89	10.33 ± 2.58	17.49 ± 4.37
L6	7.23 ± 1.81	17.39 ± 4.35	16.30 ± 4.08	51.80 ± 12.95	10.42 ± 2.61	15.41 ± 3.85
L7	9.83 ± 2.46	12.14 ± 3.03	17.31 ± 4.33	70.80 ± 17.70	8.21 ± 2.05	15.14 ± 3.79
L8	6.99 ± 1.75	8.66 ± 2.17	8.19 ± 2.05	49.22 ± 12.31	7.51 ± 1.88	11.67 ± 2.92
L9	9.78 ± 2.45	18.20 ± 4.55	19.01 ± 4.75	64.63 ± 16.16	9.34 ± 2.33	11.96 ± 2.99
L10	12.69 ± 3.17	15.98 ± 4.00	46.94 ± 11.74	161.40 ± 40.35	9.24 ± 2.31	42.43 ±10.61
L11	7.17 ± 1.79	18.76 ± 4.69	22.30 ± 5.58	89.02 ± 22.26	8.97 ± 2.24	16.67 ± 4.17
L12	5.09 ± 1.27	23.82 ± 5.95	31.38 ± 7.85	92.00 ± 23.00	12.08 ± 3.02	11.58 ± 2.89
L13	11.62 ± 2.90	26.86 ± 6.72	27.61 ± 6.90	124.63 ± 31.16	12.70 ± 3.18	26.29 ± 6.57
Local mean value (Surujdeo-Maharaj et al., 2007)	**25.12**	**21.69**	**31.77**	**111.55**	**-**	**46.29**
ISQG, Canada, 1999	**TEL – 52.3 PEL – 160**	**-**	**TEL – 18.7 PEL – 108**	**TEL – 124 PEL – 271**	**TEL – 7.24 PEL – 46**	**TEL – 30.2 PEL – 112**
ISQG, Ontario, 1993	**LEL – 26 PEL – 110**	**LEL – 16 PEL – 75**	**LEL – 16 PEL – 110**	**LEL – 120 PEL – 820**	**LEL – 6 PEL – 33**	**LEL – 31 PEL – 250**

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Table 5 (a): Contamination Factor (CF) values for heavy metal concentrations in sediment obtained in the study area

Location	Cr	Ni	Cu	Zn	As	Pb
L1	0.06	0.10	0.13	0.46	0.66	0.22
L2	0.26	0.24	0.29	0.57	1.30	0.92
L3	0.15	0.27	0.31	0.61	0.86	0.55
L4	0.09	0.27	0.44	0.41	0.96	0.68
L5	0.12	0.43	0.71	0.69	1.31	1.09
L6	0.10	0.35	0.51	0.41	1.32	0.96
L7	0.14	0.25	0.54	0.56	1.04	0.95
L8	0.10	0.18	0.26	0.39	0.95	0.73
L9	0.14	0.37	0.59	0.51	1.18	0.75
L10	0.18	0.33	1.47	1.27	1.17	2.65
L11	0.10	0.38	0.70	0.70	1.14	1.04
L12	0.07	0.49	0.98	0.72	1.53	0.72
L13	0.16	0.55	0.86	0.98	1.61	1.64

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Table 5 (b): Geoaccumulation Index (I-geo) values for heavy metal concentrations in sediment obtained in the study area

Location	Cr	Ni	Cu	Zn	As	Pb
L1	0.01	0.02	0.03	0.09	0.13	0.04
L2	0.05	0.05	0.06	0.11	0.26	0.18
L3	0.03	0.05	0.06	0.12	0.17	0.11
L4	0.02	0.05	0.09	0.08	0.19	0.14
L5	0.03	0.09	0.14	0.14	0.26	0.22
L6	0.02	0.07	0.10	0.08	0.26	0.19
L7	0.03	0.05	0.11	0.11	0.21	0.19
L8	0.02	0.04	0.05	0.08	0.19	0.15
L9	0.03	0.08	0.12	0.10	0.24	0.15
L10	0.04	0.07	0.29	0.26	0.23	0.53
L11	0.02	0.08	0.12	0.14	0.23	0.21
L12	0.01	0.10	0.14	0.15	0.31	0.15
L13	0.03	0.11	0.17	0.20	0.32	0.33

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Table 7: Pearson (n) Correlation among the heavy metal concentration between the sampling frames

Location	Bed vs Water	Location	Bed vs Water
Aripo River (L1)	0.71	Caroni River (L8)	0.93
Cumuto River (L2)	0.94	Mausica River (L9)	0.91
Tumpuna River (L3)	0.96	Arouca River (L10)	-0.45
Caroni River (L4)	0.92	Tacarigua River (L11)	0.57
Arima River (L5)	0.54	St. Joseph River (L12)	0.90
Guanapo River (L6)	0.98	San Juan River (L13)	0.80
Caroni River (L7)	0.98		