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# Spatial-temporal Variation of Metallic Contamination in Sediments from Una Basin Macro-Drainage Project Area (Pará, Brazil)

**Gilmar W. Siqueira[1], Reinaldo J. de A. Grana[1], Lívia de L. Gama[2], Maria Alice do S. de L. Siqueira[1], Assad J. Darwich[3] and Fabio Aprile[3*]**

*[1]Pará Federal University, 66075-100 PA, Brazil.*
*[2]Universidade da Amazônia, 66060-902 PA, Brazil.*
*[3]National Institute of Research in Amazonian, 69060-001 Amazonas, Brazil.*

***Authors' contributions***

*This work was carried out in collaboration between all authors. All authors read and approved the final manuscript.*

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## ABSTRACT

The objective of this study was to investigate the variation of levels of organic pollution and metal contamination (Al, Ca, Cd, Cu, Cr, Fe, K, Mg, Mn, Na, Pb and Zn) in the sediments of the drainage channels from Una basin in 2002 and 2015, after implementation of the Una Basin Macro-drainage Project (PMBU). The Una basin is located within the Metropolitan Region of Belém (MRB) in the state of Pará (Amazon), Brazil. 21 samples of superficial sediments (0-20 cm) were collected during the period of low precipitation (June and July) in 2002 and 2015. From the results, the positive and negative impacts generated were identifying, including environmental, socioeconomic and cultural aspects. The analytical procedures followed known protocols for sediment extraction and acid digestion. The results were converted into environmental indexes: Contamination Factor (CF), Enrichment Factor (EF), Potential Ecological Risk (ER) and Pollution Loading Index (PLI), and presented in isovalues categories and maps. The highest rates of reduction with the implementation of the macro-drainage project were observed for metals Mg (58%), Ca (49%) and Zn (38%). The

**Corresponding author: E-mail: aprilefm@hotmail.com;*

total Fe had a reduction of 10%, and among the metals with high toxicity to the biota, the lowest reduction rates occurred for Cd (11%), Pb (25%) and Cr total (28%). Organic matter levels also had a small reduction, suggesting a recurrence of sources of pollution to the system. The analytical indices (CF) and (EF) indicated a very diversified quality of the sediments, with classification from low to high contamination, and from low to extremely high enrichment, respectively. The results suggest a state of attention and alert for sites of contamination in the basin. The lack of consciousness of the local population, coupled with the lack of public service in relation to sanitation and new irregular occupations in the region has led to the deterioration of the quality of sediments and possibly the waters of the channels.

*Keywords: Metals; contamination; pollution; environmental indicators; Amazonian.*

## 1. INTRODUCTION

Based on the National Basic Sanitation Plan (PNSB), the Una Basin Macro-drainage Project – PMBU (1992-2004), executed by the Government of the State of Pará in partnership with the Municipal Government of Belém and funded by the IDB, is one of the great urban structurings in Latin America. The PMBU provided for the installation of macro-drainage (channel rectification and coating) and micro-drainage (galleries and concrete channels, gutters and collecting nets) in several areas of the Metropolitan Region of Belém (RMB), including complementary works for grounding and paving of roads, depletion sanitation, water supply, solid waste collection and transportation, and environmental education program [1-3]. The project's sanitation activities serve more than half a million people, the most living in flood-prone areas of the RMB. The PMBU was developed to rehabilitate and recover the lowland areas (topographic dimension <4 m) of Belém, reducing flooding of houses during periods of high tide. For this, the project aimed at increasing the depth and width of the channels, to receive and transport the rainwater. It was also established the implementation of a water supply and sewage systems, reducing pollution by 60% [2,4]. It is estimated that the project, through the reallocation plan, transferred more than 4,800 families residing in the areas of the rectification works and the widening of the channels [5].

The Metropolitan Region of Belém (RMB) presents natural limitations in the use and occupation of soils because of its extensive water network. As a result, the urban infrastructure programs were accompanied by physical modifications, including alterations in the terrain expansion by landings of streams and mangroves; changes in altimetric heights; paving and waterproofing of soaked soils. Especially between 1950 and 1970, the elimination of natural obstacles resulted in changes in the water network, aggravating urban problems such as flooding, infiltration, erosion and channel siltation [4,5]. Flooding and infiltration, when associated with domestic and industrial sewage water, intensifies pollution and environmental contamination. Despite the investment in PMBU's work, it is believed that the problem of local sewage will not be fully remedied, and that part of the local population will continue to throw their tailings without treatment in the drainage channels, contributing to the contamination of the tributaries.

The sewage introduced in the RMB drainage system is of the mixed type, with varied compositions of domestic and industrial sewage. This is a very important aspect of sanitation because it increases the possibilities of contamination of the drainage basin by various organic and inorganic components such as phenolic, organophosphorous and organochlorine residues, benzenes, metallic elements and a high volume of organic matter, resulting from mixed sewage, agricultural components, clandestine waste and polluted rainwater.

Domestic sewage has the sanitary characteristic of transport and transmission of various diseases. The quality and quantity of the sanitation services offered in Brazil are quite irregular, and the Northern region is considered to be the least favored by water supply and sewage treatment services [6,7]. A more accurate analysis of the results of the National Plan for Basic Sanitation between 2008 and 2010 showed that the North presents the lowest percentage of treated water supply, supplying 54% of the total population [8,9]. The numbers are even lower for collection and treatment of sewage, with indices less than 5% of total sewage generated [9]. Consequently, the most part of the sewage produced in the municipal districts is discarding in water-bodies without any treatment [10], resulting in pollution and contamination of the aquatic ecosystems. Several diseases are strongly related to the quality of the urban environment and depend on the way this environment is structured [11]. Infections and diseases associated with sanitation problems are very common in Pará, especially in urban densities, where unhealthiness reaches alarming levels. In the periphery of the RMB, frequent cases of water-borne or dirty-water diseases such as *amebiasis*, *giardiasis*, *salmonellosis*, cholera, *shigellosis*, hepatitis and other forms of diarrhea, and water-based diseases, which include *ascariasis*, leptospirosis, and *schistosomiasis* [8,9,12-15].

Information on the incidence of water diseases by region is well known, however, as yet there is not much information on the variation of pollution levels and contamination during and after installation of urban infrastructure programs, such as PMBU. The objective of this study was to investigate the variation of levels of organic pollution and metal contamination (Al, Ca, Cd, Cu, Cr, Fe, K, Mg, Mn, Na, Pb and Zn) in the sediments of the drainage channels of the Una basin, during (2002) and after (2015) execution of the PMBU, due to the changes that occurred in the basin.

## 2. STUDY AREA

### 2.1 Physical Aspects

The Metropolitan Region of Belém (RMB) is formed by 5 municipalities: Belém, Ananindeua, Santa Bárbara do Pará, Benevides and Marituba (Fig. 1), and is situated in the equatorial belt known as the depression belt of the Central Amazon, 160 km from the Equator. The RMB occupies an area of 716 km$^2$ with an average altitude of 10.3 m above sea level. According to the classification of Köppen, the climate of the region is hot and humid *Af*, admitting also the climatic types *Am* and *Aw*, all belonging to the humid tropical forest climate. Average monthly temperatures are always above 18°C. The region presents average annual precipitation of 3000 mm, concentrating 70% of rainfall in the rainy season (December to May). The Belém city has 14 hydrographic basins, and most of the continental area of the municipality is in quotas lower than 4 meters, which allows greater risks floods [6,8]. The Una basin has an average altimetric amplitude of 22 m and a relief ratio of 4.46 m/km [16], which means a relief with a low slope (0 to 3%). The low slope associated with soil sealing (paving) and high precipitation allows greater opportunity for flooding. The local vegetation is rarefied, much modified by urbanization. Currently there are few medium and large trees located in the area, with a predominance of grasses. The natural channels have vegetation of capoeirão, mostly deforested for occupation, making difficult the flow of rainwater, as well as the return of the waters at low tide. The soil of the basin is composed of recent mud, sand and gravel alluvium, with a thickness of up to 20 m, with organic matter focus, which forms superficial deposits at the banks of the streams, close to lowlands and floodplain plains subject to periodic flooding.

### 2.2 Socioeconomic Aspects

According to the Atlas of Human Development in Brazil, Belém has a Municipal Urban Development Index (IDH-M) of 0.806 [2,7]. Despite this, the population concentration in the urban region, especially in the outskirts of the city, has generated in the last decades several problems of sanitation. The urban housing deficit in the Pará State grew 98.9% in the last decade, while the national average was 41.5% [17]. The population of RMB was estimated at 1,794,891 inhabitants, being 97.7% of urban population [6,7]. This means not only a lack of housing, but lack of basic sanitation, forcing the poorest population to live on the periphery, contributing to water pollution, exerting strong pressure of contamination on the groundwater by wastewater and contaminants, which seep into the soil until the aquifers. The RMB has a large volume of interconnected rivers, forming a large natural water network, with dozens of islands, mostly occupied irregularly (without sanitation), establishing an environment defined by [18] as a typical urban occupation in the Amazon. Since the 1980s, the annual population growth rate of RMB has been around 3%.

Most of the streams that formed the Una basin no longer exist and the population occupied irregularly the streams borders, contributing to water pollution, floods, the spread of diseases, and economic losses. In the 1970s, the first studies were carried out to evaluate the self-purification capacity of Guajará Bay. Until the early 1980s, there was no more than 75 km of

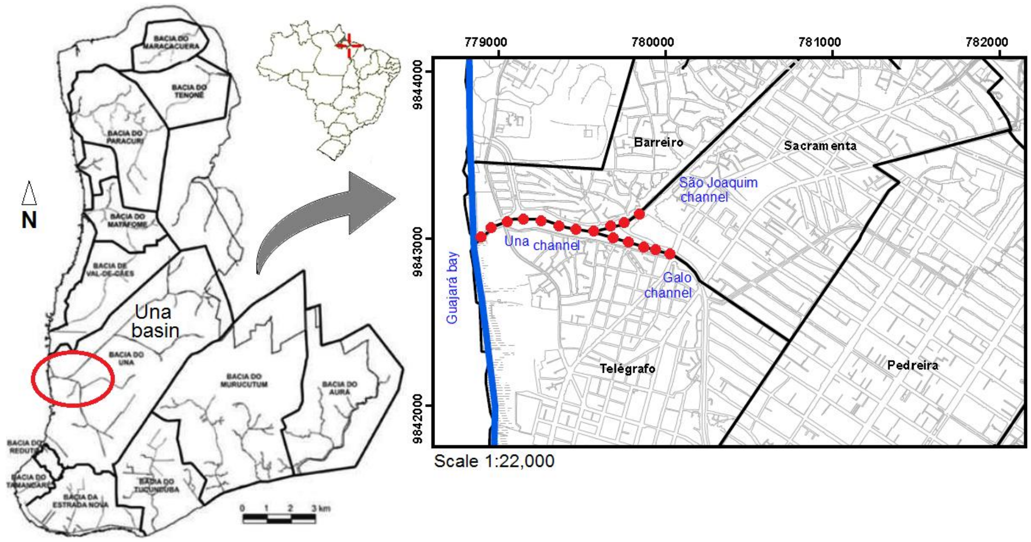


**Fig. 1. Limit of the watersheds of Belém (Pará, Brazil), with emphasis on the Una basin and the sampling sites**

*(Source: PMB/CODEM/Urban Development Board, 05/2014)*

sewerage network in the RMB, equivalent to 10% of the local population at the time [18,19]. Since then, the volume of untreated sewage has increased steadily, altering the patterns of drainage basins. The "*Drainage, Roads, Water and Sewage of the Lower Belém Project*" was characterized by the execution of basic sanitation works and services in the flooded areas of the Una basin (Fig. 1). The restructuring of the Una basin was a complex engineering project to address not only sanitary issues but also aspects of urban renewal and socio-economic promotion, aiming to improve the quality of life of 600 thousand people [2,4,20].

## 3. MATERIALS AND METHODS

In order to investigate the pattern of organic and metallic pollution in the drainage channels, field sampling (primary data) and the construction of a documental and bibliographic research base on the study area (secondary data) were established. Through observation and systematic analysis, it was possible to verify the current situation of the sediments in the basin. 21 samples of surface sediments (0-20 cm) were collected in the drainage channels during the low rainfall period (June and July), during the execution (2002) and after the works (2015) of the PMBU. The sediments were collected with dredge Van-Veen model, and sub-samples were removed with sterilized plastic spatula and stored in plastic bags preserved in a cold chamber (-22°C) [21]. The physical-chemical analyzes were carried out in the chemistry laboratories of the Faculty of Chemistry of UFPA and hydrocarbons of UEPA. In order to determine the concentration of the metallic elements, the sediments were oven dried at 600°C for 48 hours, pulverized, homogenized and sieved in a mesh of 0.063 mm (230 mesh). Sub-samples of 0.100 g (dw) fine sediment were added in 100 mL beaker containing 5 mL $HNO_3$ acid solution, 2 mL $HClO_4$ and 4 mL HF 1M, 10 mL HCl 6M and 10 mL of Milli-Q $H_2O$. After homogenization, the mixture was placed in a heating plate at 100°C until partial drying (pasty appearance). Further additions of the acid solution were added to the heating mixture. The hot acid digestion procedure was repeated several times until there was no effervescence in the samples, indicating the absence of organic matter (OM) [22]. The hot acid digestion procedure is applied to break the metal bonds, allowing the release of the metal elements to the acid solution. After digestion, the mixture was filtered in GFF and aliquots were transferred to 25 mL flasks filled with Milli-Q water. The determination of the metal fractions (Al, Ca, Cd, Cu, Cr, Fe, K, Mg, Mn, Na, Pb and Zn) was done in inductively coupled plasma atomic emission spectrometry (ICP/AES) according to [21,22] and [23 - Ref. 3120 and 3500]. For determination of the OM content, samples of moist sediments were dried in an oven at 35°C for 48 hours. After drying and sieving (≤ 63 μm), 10 g of fine sediments were placed in beaker containing excess solution of potassium dichromate in acidic medium ($H_2SO_4$) and heated to 100°C for oxidation of OM [22,24]. The heating step requires careful attention to avoid loss of material by boiling. Heating should start between 50-80°C and only gradually raise the temperature, always taking care to add distilled water to the edges of the beaker to avoid loss of material [22]. The values of the dry mass difference before and after the oxidation were used to determine the OM and organic carbon (OC) estimation, applying equation 1. According to [22], good results digestion by replacing the direct heating in electric plate by heating in a sand-bath.

$$\% OM = \% OC \times K \quad \text{; Where k= 1.8 [25]} \tag{1}$$

The values of Turekian and Wedepohl [26] were used for shale in the comparison of the obtained results. In order to identify the temporal variation of the contamination and pollution of the sediments, four analytical indexes were applied: 1) Contamination Factor (CF) or Potential Contamination Index [27,28], which relates the maximum concentration of a metal in the sample with its natural abundance (background), from the reference level determined by [26] for average shale (equation 2). The Hakanson classification [28] proposes the category scale for contamination values: <1 low contamination; between 1 and 3 moderate contamination; between 3 and 6 considerable contamination, and >6 high contamination. 2) Enrichment Factor (EF), applied using aluminum as the reference value (equation 3). The EF was calculated as a function of the metal concentration by the reference value of the element Al. Based on the sediment quality classification according to the EF established by [29], five categories of sediment quality were observed: low or natural, moderate, significant or severe, high or strong, and extremely high metal enrichment. 3) Potential Ecological Risk Factor (ER) [28], widely applied in ecological risk assessments of metallic elements, correlating CF with toxic response factor for Cr, Cu, Ni, Pb, Cd and Zn metals (equation 4). 4) Pollution Load Index (PLI) defined by [30] as index the $n^{th}$ root of the product (Π) of all calculated metal contamination factors ($CF_n$) (equation 5). Even if there is only one metallic element above the permissible levels, the PLI will indicate the presence of pollution load in the system. The result of the PLI does not identify the metal element or elements responsible for the pollution load. Thus, when possible, this analysis should be accompanied by the CF and EF indices. The indices were presented in the range of variation table (CF, EF); and isovalues maps (ER and PLI) using the kriging interpolation method (weighted-movingaverag) using Surfer® Golden Software, 9.11 (2010).

$$CF = [Metal]_{\max} / [Metal]_{RV} \tag{2}$$

$$EF = \frac{[Metal]_{sed} / [Metal]_{RV}}{[Al]_{sed} / [Al]_{RV}} \tag{3}$$

$$ER = Tr \ x \ PCI \tag{4}$$

$$PLI = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \times \ldots \times CF_n} \tag{5}$$

where: RV is the reference value based on Upper Continental Crust (UCC) data; [metal] is given in µg/g or equivalent; Tr is a toxic response factor observed in the table; $CF_n$ are the contamination factors defined by [28,30].

## 4. RESULTS AND DISCUSSION

### 4.1 Metallic Elements

The data summarized on the metallic levels in the sediments were showed in Table 1. Al content ranged from 2100 to 58,000 mg/kg (mean 22,244±15,021 mg/kg) during the year 2002, and from 1750 to 48,600 mg/kg (mean 18,613±12,589 mg/kg) in 2015. Due to urban macro and micro-drainage works, the average reduction of the Al concentration was 16% within the monitoring period. The high standard deviations were highlighted, suggesting a large spatial variation, probably due to the persistence of local sources of contamination. An analogous study developed in the São Joaquim Channel (see Fig. 1) by [31] determined Al levels between 13,400 and 46,700 mg/kg (mean 24,500 mg/kg). The mean values obtained in this study were below the average shale value 88,000 mg/kg defined by [26]. Although the sources of contamination persist, even after the macro-drainage work, reactions with $Fe^{2+/3+}$ oxides and hydroxides may be occurring in the sediment compartment, reducing the free forms, especially in the presence of OM. Al is a larger, non-reactive constituent of clay minerals, constituting the most part of the suspended matter from bottom sediments [32], which explains the high absolute values determined (Table 1). In relation to the alkali metals (Na and K) and alkaline earths (Ca and Mg), Ca was the metallic element with the greatest amplitude of absolute variation, from 4500 to 18,400 mg/kg (8150±3497) in 2002, and from 2300 to 9400 mg/kg (4156±1793) in 2015. The mean change in the monitoring period was 49%. The Ca presents strong affinity with the carbonate elements, quite abundant in the Amazonian waters, and whose association forms a compound of low solubility, remaining in the sediment. The calculated mean Ca values were well below the average shale value 38,600 mg/kg defined by [33]. The Na, K and Mg elements varied respectively from 100 to 900 (375±211); 100 to 300 (200±73) and 100 to 700 (350±186) mg/kg in 2002 and 85 to 780 (323±183); 85 to 280 (183±69) and 40 to 295 (147±78) mg/kg in 2015 (Table 1). Their percentages of average variation were 14% Na, 9% K and 58% Mg. The mean values of Na, K and Mg determined for this study represent less than 1.5% of the values estimated by [33] for the continental crust (UCC), whose values are respectively 23,600; 21,400 and 22,000 mg/kg. In general, the spatial-temporal distribution gradient for alkaline and alkaline earths was uniform, except for some point sources arising from domestic sewage. Fig. 2 shows Piper's triangular diagram for the ionic balance between the alkali metals. The results indicated the presence of sediments rich in Ca, suggesting high precipitation of Ca in the form of $CaCO_3$.

**Table 1. Variation of the metals levels (mg/kg) in the drainage channels of the Una basin (Pará, Brazil) for the years 2002 and 2015 (N = 21)**

| | 2002 | | | 2015 | | |
|---|---|---|---|---|---|---|
| | **Min** | **Max** | **Aver±SD*** | **Min** | **Max** | **Aver±SD*** |
| Al | 2100 | 58,000 | 22,244±15,021 | 1750 | 48,600 | 18,613±12,589 |
| Ca | 4500 | 18,400 | 8150±3497 | 2300 | 9400 | 4156±1793 |
| Na | 100 | 900 | 375±211 | 85 | 780 | 323±183 |
| K | 100 | 300 | 200±73 | 85 | 280 | 183±69 |
| Mg | 100 | 700 | 350±186 | 40 | 295 | 147±78 |
| Cd | 0.27 | 0.74 | 0.52±0.11 | 0.31 | 0.66 | 0.46±0.10 |
| Pb | 46.97 | 173.60 | 89.02±34.36 | 28.66 | 118.60 | 66.67±27.00 |
| Zn | 25.12 | 187.00 | 78.06±48.57 | 17.55 | 124.42 | 48.77±28.99 |
| Cr | 25.59 | 94.97 | 50.44±17.27 | 18.62 | 68.12 | 36.53±12.28 |
| Cu | 5.00 | 34.42 | 15.09±9.48 | 3.12 | 21.97 | 9.90±6.37 |
| Fe | 9300 | 38700 | 18100±7298 | 9200 | 33100 | 16300±6105 |
| Mn | 25.76 | 79.42 | 54.41±14.99 | 18.45 | 56.59 | 38.77±10.67 |

**SD= standard deviation.*

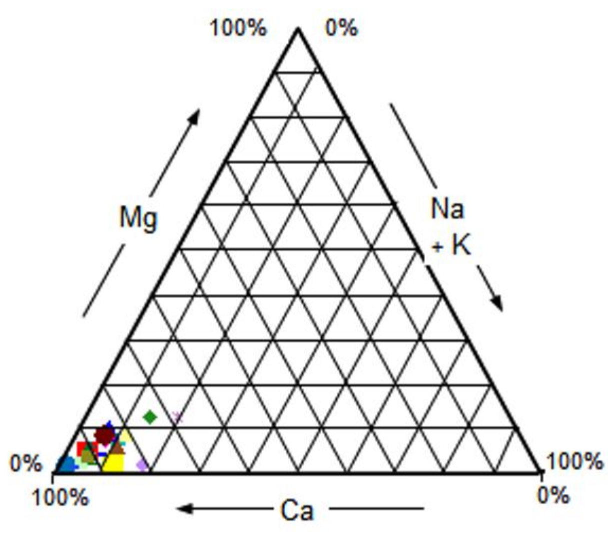


**Fig. 2. Piper type plot for the alkaline and alkaline earth metals (mean in mEq/kg) in the sediments in the drainage channels of the Una basin (Pará, Brazil)**

Three metallic elements of special interest for their contamination potentials and deleterious effects on biota (Cd, Pb and Zn) presented a pattern of divergent enrichment of the general behavior of the metals in the monitoring area. Cd had concentrations ranging from 0.27 to 0.74 (0.52±0.11) mg/kg in 2002, and from 0.31 to 0.66 (0.46±0.10) mg/kg in 2015 (Table 1), a reduction of 10% after consolidation of macro-drainage works. Despite the reduction, the Cd contents were mostly above the levels determined by [26] for the average shale in the UCC, which is 0.3 mg/kg. Several sampling sites in the drainage channels of the Una and São Joaquim presented values equivalent to levels found by other authors in modified areas, with sediments contaminated by industrial effluents. Under natural conditions, Cd is found in waters and sediments at very low concentrations (trace). Contamination of Cd in the sediments from the samplings may be occurring through solid waste deposited around the basin. The most significant residues for environmental contamination by Cd are the residues from the production of non-ferrous metals and the manufacture of articles containing Cd. These residues can come from discarded packaging in inadequate places (clandestine waste). Other potential sources include municipal waste containing Ni-Cd batteries, which usually are being disposal with household waste in landfills and dumps. The form as Cd is found in sediments is very important for determining their availability and re-mobilization. The Cd may be associated with the carbonates, precipitated as solid compounds stable or co-precipitated with hydrated Fe oxide, in this case, less susceptible to being re-mobilized by re-suspension of the sediments or by biological activity. On the other hand, Cd adsorbed to mineral surfaces such as clay and OM is more easily absorbed by biota, and released under specific conditions of environmental changes, which include pH and Eh variation of the medium, as well as physical activities such as dredging processes. The largest anthropogenic sources of Cd include mining, Cd manipulation for Ni-Cd batteries, pigments, stabilizers of products from polyvinyl chloride (PVC), manufacture of Cd alloys in ferrous (electroplating) and non- ferrous including iron and steel alloys containing Ni, Pb and Zn, the production of electronic components, as well as the burning of fossil fuels and the production of cement and fertilizers [10,34,35]. Despite the diversity of non-natural sources of Cd for the aquatic system, it is believed that in the region, due to geomorphological, socioeconomic and cultural characteristics, the main source of contamination is clandestine garbage, discarded directly in streams, landfills and drainage channels. Brazil is one of the countries with the highest concentration of cell phones per inhabitant, and the percentage of recycling of batteries and sources is very low, which suggests that most of the discards are made in common waste. Pb levels ranged from 46.97 to 173.60 with a mean of 89.02±34.36 mg/kg in 2002, and between 28.66 and 118.60 with mean 66.67±27.00 mg/kg in 2015 (Table 1). The reduction rate after completion of the macro drainage works was 25%. Despite this, Pb presents the same problems of inappropriate handling and disposal already discussed. As a result, the Pb also presented mean values higher than the reference value of 20 mg/kg for the UCC defined by [26]. This is another serious environmental problem since Pb has a high potential for contamination, high bioaccumulation rate and is quite toxic. A number of sampling sites along the Una channel presented specific levels above 100 mg/kg, an increase of 400% over the natural reference value. In the São Joaquim channel, which had continuity to the Una channel, lower levels were found between 3.66 and 38.19 mg/kg [31], but still with a maximum value above the reference value established by [26]. In addition to the inappropriate disposal of solid waste, there are sources of air pollution from the combustion of fossil fuels (gasoline). After discharging or depositing particles associated with VOCs, Pb can be adsorbed to the OM particles, forming highly toxic organic compounds. Pb is an extremely toxic trace metal, which tends to accumulate in biota, including human tissues [34,36,37]. In addition, Pb has the tendency to form anionic compounds of low solubility, such as hydroxides, carbonates, and phosphates, facilitating their absorption by benthic organisms. A significant fraction of insoluble Pb may be incorporated into the particulate material as absorbed or adsorbed ions. Once incorporated into the sediment, according to [37] the Pb associates the structure of the hydroxides, due to their strong affinity, forming complexes with the FeOOH crystals. Zn is quite abundant in solid waste and domestic sewage, which may explain its enrichment in the study area. Zn's contributions are found in plastics, glass, ferrous and non-ferrous materials, including paper and cardboard, in addition to the previously mentioned manufacturing activities [34,35]. In igneous rocks, Zn occurs as sphalerite (ZnS), and may be associated with carbonated and silicate minerals. Under natural conditions, its main oxidation state is $Zn^{2+}$, and analogously to Fe and Mn, it may form poorly soluble complexes depending on the pH of the environment. The levels of Zn varied between 25.12 and 187 (78.06±48.57) mg/kg in 2002, and between 17.55 and 124 (48.77±28.99) mg/kg in 2015 (Tab. 1). Following the same metallic behavior of Cd and Pb, it was noted that at several sampling sites the concentration of Zn was greater than 100 mg/kg, above the 95 mg/kg reference value established by [26] for the UCC average. The rate of reduction of Zn concentration after the consolidation of macro-drainage works was 38%.

The Cr values determined varied between 25.59 and 94.97 (50.44±17.27) mg/kg in 2002, and between 18.62 and 68.12 (36.53±12.28) mg/kg in 2015 (Table 1). The percentage of reduction of sediment contamination levels after the consolidation of the macro-drainage works was 28%. In comparison, all sampling sites remained below the reference value of 90 mg/kg for the UCC [26]. The Cr content determined in this study was close to the average of 48.80 mg/kg obtained by [31], for the adjacent area in the São Joaquim channel. The aggravating factor in relation to Cr is its hexavalent form ($Cr^{6+}$), which occurs from changes in physical-chemical properties (pH and Eh). The natural occurrence of $Cr^{6+}$ is rare, and its presence in water and sediment is due to industrial waste, especially effluents from the paint and pigment industries, explosives, paper, ceramics and electroplating [34]. $Cr^{6+}$ is highly toxic [34,35], with carcinogenic potential, and thus has great biological importance, especially in ecotoxicological studies. In aquatic environments, $Cr^{6+}$ is predominantly present in the soluble form. The soluble forms may be stable enough to undergo transport, however, $Cr^{6+}$ may be converted to $Cr^{3+}$ in the presence of reducing forms such as organic compounds, hydrogen sulfide, sulfur, iron sulfide, ammonium and nitrite [36,37]. In the area of macro-drainage work, one of the main planning goals was to ensure that there would be no mixing between domestic and industrial sewage. Even with the difficulties of inspection, this has apparently occurred, which explains, in part, the rate of reduction. Cu levels determined varied from 5 to 34.42 (mean 15.09±9.48) mg/kg in the construction phase (2002), and from 3.12 to 21.97 (9.90±6.37) mg/kg in 2015 (Table 1), representing a reduction of 34%. In absolute values, the Cu contents determined remained throughout the monitoring period below the reference value of 45 mg/kg for UCC [26]. Despite this, there was no homogeneity in the spatial distribution of Cu, with sampling sites along the Una channel with concentrations above 20 mg/kg. This may once again indicate local sources of pollution by discarding untreated solid waste directly into the receiving body of drainage channels. The levels of Cu determined in this study were below the mean of 58.43 mg/kg obtained by [31] for the São Joaquim channel (Fig. 1). Cu is an essential metal for biota, however, in high concentrations, it can be rendered highly toxic [34]. Cu may be present in the sediment and interstitial water as an insoluble precipitate, associated with hydroxides, phosphates or sulfides, or adsorbed to particulate matter [36,37]. The ratio between the soluble and insoluble form will depend very much on aspects such as pH and Eh, alkalinity, cation exchange capacity, the presence of other metal ions, and OM concentration [35-37].

Another element of great importance for the processes of sedimentary oxy-direction is iron. Total Fe levels ranged from 9300 to 38,700 mg/kg (mean 18,100±7298 mg/kg) in 2002, and from 9200 to 33,100 mg/kg (16,300 ± 6105 mg/kg) in 2015 (Table 1), suggesting a reduction of the total concentration of 10% after the macro-drainage works. That is considered a small reduction due to the size and high investment of the works. A study developed by [31] determined Fe contents in the São Joaquim channel (Fig. 1) between 9500 and 18,100 mg/kg (mean 13,100 mg/kg). The maximum total Fe content during the implantation phase of the macro-drainage project was close to the reference value of 43,000 mg/kg [26]. However, after the conclusion of the project, the absolute values decreased being well below the reference value (UCC). Except for some local sources, Fe contents were homogeneously distributed throughout the study area. This is explained, in large part, by the sedimentary transport that occurs in the channels with the flow of rainwater and urban residuals. The results are representations of the type and size of the sedimentary, fine and colloidal material, associated with organic compounds. There is also the possibility of autochthonous sources of Fe of minerals predominant in the region, such as hematite, goethite, and limonite. The hematite [$Fe_2O_3$] is one of the most common minerals in the bottom sediments in the region, while the occurrence of limonite [$Fe_2O_3.H_2O$] appears as a result of the hematite alteration by the formation of hydrated iron oxide [10]. In the RMB region, the soils are chemically poor in common ions, a situation compatible with the strong leaching resulting from the high precipitations (annual average of 3000 mm). Iron can be mobilized and concentrated forming laterite concretions very common in the lands of the region [38]. Due to the physical, chemical and physicochemical conditions that change in the water column, it is possible that a part of the Fe may be associated to clay minerals, which are predominant in the white or muddy waters of the region, which would facilitate its precipitation [10].

The geological characteristics of the region are an important barrier in works of the restructuring of roads, natural galleries, and channels in urban drainage projects. With typical floodplain soils, these areas present fluvio-lacustrine and bay sediments composed of clay and sandy-clay sediments, with intercalations of sand and OM. In low lands with a slope of less than 5%, there is a predominance of mud sediments with depths of up to 20 meters, which can be found on the surface of the river bed. These are typical areas of flooding, with plains and lowlands permanently flooded, or with daily variations depending on the level of the tide. The RMB has two major geomorphological features positioned in the regional physiographic: non-flooded areas denominated as ‘Terra-Firme' areas and periodically or permanently flooded areas called ‘Várzea'. These features are distinguished by their natural characteristics. However, most of the RMB relief has already been modified, with landings and paving. This de-characterization of the natural mosaic makes it difficult to establish a pattern of response to the accumulation of elements, in this case especially the metallic elements, in the drainage areas.

The Mn levels varied within the monitoring range between 25.76 and 79.42 (54.41±14.99) mg/kg in 2002, and between 18.45 and 56.59 (38.77±10, 67) mg/kg in 2015 (Table 1). The reduction rate average of the Mn was 29%. In comparison, the values between 28.60 and 50.66 (average 37.56) mg/kg for [31] were determined in the São Joaquim channel for the sediments. In relation to the levels determined by [26] for average shale in the UCC, the Mn levels determined in the drainage channels were well below the average, what confirms that the soils and sediments of the Eastern Amazon are not naturally enriched by Mn forms. The behavior of the Mn in relation to the adsorption and precipitation processes is relatively analogous to Fe [10,36]. Dissolved by the erosion and leaching of igneous rocks, the $Mn^{2+}$ ions persist in slightly acidic and not so oxidizing aqueous solution. Its precipitation will only occur under conditions of alteration of the pH of the environment, due to the influence of carbonate and silicate elements at high concentrations [36,37].

## 4.2 Organic Compounds

OM levels ranged from 0.6 to 6.1% (mean 2.8±1.6%) in 2002 and from 0.5 to 5.4% (mean 2.5±1.4%) in 2015. Proportionally, OC concentrations ranged from 0.3 to 3.6 (1.6±0.9)% and from 0.3 to 3.1 (1.5±0.8)% (Tab. 2). The reduction rate of OM, and consequently of the OC, after the consolidation of the macro-drainage project, was 10.3%, a very small value when evaluating the project dimensions and the large investment of resources applied in the work. It was observed an increase in the OM load from the central part towards the banks of the Una channel, suggesting a strong autochthonous contribution of organic waste directly into the drainage channel. Under the geomorphological aspect, the margins present smaller depths and current velocity, favoring the accumulation of sedimentary organic residues. As already suggested, socioeconomic and cultural aspects have contributed to the uncontrolled occupation of the population around streams and drainage channels. Even after the implementation of the PMBU, much of the local population remains without adequate basic sanitation. Other areas of the basin upstream of the macro-drainage work also contribute to OM mineral sources. Thus, the water and sediment of the channels continually accumulate organic compounds, making the system eutrophic, as it was before the intervention of the PMBU. The concentration of OM has great importance in the mobility and complexity of metallic elements. The increase of OM in the system may directly or indirectly be contributing to the accumulation of metals in the sediments. It is possible that new engineering interventions, including periodic dredging, may reduce the volume of organic compounds, but it is essential and urgent to implement environmental education programs, that initially was proposed in PMBU but never applied. As long as local people continue to treat rainwater drainage channels as open sewers, the system will remain polluted and sometimes contaminated. Another important factor to consider is the self-purification capacity of the system, which in this case is quite limited by the geomorphology and population density.

**Table 2. Variation of OM and OC (%) in the drainage channels of the Una basin (Pará, Brazil), for the years 2002 and 2015 (N = 21)**

| | 2002 | | | 2015 | | |
|---|---|---|---|---|---|---|
| | Min | Max | Aver±SD* | Min | Max | Aver±SD* |
| OM | 0.6 | 6.1 | 2.8±1.6 | 0.5 | 5.4 | 2.5±1.4 |
| OC | 0.3 | 3.6 | 1.6±0.9 | 0.3 | 3.1 | 1.5±0.8 |

## 4.3 Evaluation Analytical Indices

### 4.3.1Contamination Factor (CF)

For this study CF index was applied for the metals Cd, Pb, Zn, Cr, Cu and Fe (Table 3). For the Cd element, the CF index varied between 3 and 7, with most of the results being between 5 and 6, establishing for the study area a CF between the categories 'moderate' and 'significant' contamination. A similar behavior was noted for the Pb, whose results ranged from 3 to 10, with mean 5 for the index, classifying the region in the 'significant' contamination category. Based on the criterion adopted, the levels of contamination for Pb were above 10 in some sites of the monitoring area, placing these subareas at alert levels. These are the sites located in the middle section of the Una channel (Fig. 1). The metallurgical park located in the RMB is not expressive to producing large amount of metallic tailings. Activities around the basin, and especially in the monitoring area, are predominantly due to domestic activities. This suggests that the main sources of Cd and Pb are derived from discards of batteries and electronic components by the population, launched directly into the channels. The consumption and use of Cd have varied greatly, especially in recent years, and battery use has outpaced traditional productions such as pigments, paints, and stabilizers. Another possibility that may be occurring is the corrosion and wear of pipes from older water and sewage networks. Zn presented low indices, ranging from 0 to 2, with a mean of 1 in the Hakanson scale [28] and levels from 'low' to 'moderate' contamination. The metallic elements Cr, Cu and Fe presented intervals of CF between 1 - 3; 0-2; and 0-1, respectively, placing them between the categories 'low' and 'moderate' contamination. Because they are quite reactive, Cu and Fe elements may be used in the OM oxireduction and diagenesis processes (equations 6 and 7, unbalanced). In this case, the Cu, due to its high reactivity, may be being used instead of Mn. It is also possible that there is consumption of these metals in the biological processes of the benthic communities since Cu and Fe are essential metallic elements in the biochemical processes.

$$(CH_2O)_{106}(NH_3)_{16}(H_3PO_4) + Fe_2O_3 + H^+ \rightarrow Fe^{2+} + CO_2 + NH_3 + H_3PO_4 + H_2O \quad (6)$$

$$(CH_2O)_{106}(NH_3)_{16}(H_3PO_4) + MnO_2 + H^+ \rightarrow Mn^{2+} + CO_2 + N_2 + H_3PO_4 + H_2O \quad (7)$$

### 4.3.2 Enrichment Factor (EF)

The EF was calculated as a function of the metal concentration by the reference value of the element Al (equation 3), and the results are showed in Table 3. The index calculated for the Cd indicates a metal enrichment between the categories 'significant' and 'extremely high', with EF values between7.5 and 202 in 2002,

**Table 3. Evaluation indices CF and EF for the metallic elements monitored in the Una channel (Pará, Brazil), for the years 2002 and 2015 (minimum - maximum; N = 21)**

| Metal | CF | | | | EF | | | |
|---|---|---|---|---|---|---|---|---|
| | 2002 | | 2015 | | 2002 | | 2015 | |
| | Min | Max | Min | Max | Min | Max | Min | Max |
| Cd | 2.6 | 7.3 | 3.0 | 6.5 | 7.5 | 202 | 7.3 | 226 |
| Pb | 2.8 | 10.2 | 1.7 | 7.0 | 8.3 | 153 | 6.1 | 170 |
| Zn | 0.3 | 2.3 | 0.2 | 1.5 | 1.9 | 28.9 | 1.1 | 34.1 |
| Cr | 0.7 | 2.7 | 0.5 | 1.9 | 3.6 | 45.1 | 3.1 | 39.3 |
| Cu | 0.3 | 2.4 | 0.2 | 1.5 | 2.1 | 33.7 | 1.6 | 25.4 |
| Fe | 0.3 | 1.3 | 0.3 | 1.1 | 1.5 | 19.2 | 1.5 | 22.5 |

*Low; Moderate; Significant; High; Extremely high* and between 7.3 and 226 in 2015. A similar behavior was observed for Pb, which presented EF values between 8.3 and 153 in 2002 and between 6.1 and 170 in 2015. Considering the high toxic potential and high bioaccumulation capacity of these two metallic elements, the monitored area should be considered as an area of attention. Another important factor is that Cd and Pb had an increase in maximum levels in the drainage channels after a decade of PMBU consolidation (Table 3). The Zn presented indices varying from 1.9 to 28.9 in 2002 and from 1.1 to 34.1 in 2015, classifying it as 'low' to 'high' metal enrichment. Considering that Zn also has a high bioaccumulation capacity, it should also be monitored more closely, although its toxic potential is lower than that of the Cd and Pb metals. Cr ranged from 'moderate' to 'extremely high' in 2002 (EF from 3.6 to 45.1) and between the 'moderate' to 'high' categories in 2015 (EF of 3.1 and 39.3). In the specific case of chromium, as already discussed, the greatest aggravating factor is not in its total concentration, but in the concentration of the $Cr^{6+}$ fraction, which has a high carcinogenic potential [34]. Cu is an essential element for biological functions, but in high concentrations it becomes toxic. The EF index for Cu varied from 2.1 to 33.7 or 'moderate' to 'high' enrichment in 2002, and between 1.6 and 25.4 or from 'low' to 'high' enrichment in 2015. Fe is another essential element for biological and biochemical functions, and that in high concentrations can inhibit reactions and become moderately toxic. The EF for Fe was between 1.5 and 19.2 or from 'low' to 'significant' enrichment in 2002, and from 1.5 to 22.5 or 'low' to 'high' enrichment in 2015. In general, after the consolidation of the project of macro-drainage of the Una basin, a significant reduction of the concentration of metals in the sediments of the channels was expected. However, the maximum levels calculated for EF in 2015 indicate an equal or even worse condition for some elements compared to the year 2002. Again, two factors are considered to be interfering in the results: 1) the corrosion of some older pipes, releasing especially Al, Cd, Zn, Cu and Fe in the water column, and this to the sediments as a function of the pH; and 2) an increase in the amount of solid waste discarded without treatment by the population both autochthonous and allochthonous the drainage area.

#### 4.3.3 Potential ecological risk factor (ER)

The ER established for the control of aquatic pollution [28], is used as control-values associated with the toxic levels of each metallic element. The toxic potential of the elements is obtained from ecotoxicological tests and of the determination of lethal doses (e.g. $LD_{50}$, $LD_{90}$) for various aquatic organisms. The toxic potential of metals for man, especially heavy metals, is already well known [34]. Considering in the macro-drainage infrastructure plan the possibility of contamination of the sewage networks and water supply by clandestine pipelines, the ER allows establishing a state of attention and/or alerting for local contamination. Figure 3 presents the isovalues established in class by the interpolation method for the Cr, Cu, Cd, Pb and Zn metals in the sediment compartment of the Una basin. In general, in the present study the attention and alert areas were established for the initial stretch and the central sites of the channels. Cr was the metal element that presented the most variation of the index, whose values in absolute numbers ranged from 1.3 to 4.7 and average 2.5, suggesting low risk, according to the Hakanson classification [28]. Cu presented ER levels between 1.4 and 9.9 (mean 4.4), which also suggests by the classification adopted low-risk levels. Despite this, it is suggested to maintain a state of attention to the central area of the channels. The Cd presented the highest absolute values, varying between 85 and 199 (average 145), classified it between the categories 'considerable risk' and 'high risk' of contamination. This state of alert occurred for the central part of the Una channel, close to the São Joaquim and Galo channels (Fig. 1). Pb presented ER indexes between 11 and 41 (mean 23) with indications for the categories of 'low risk' and 'moderate risk' of contamination. The area classified as the moderate risk was quite comprehensive, incorporating more than 80% of the monitored area. The Zn presented a distribution pattern similar to the Cd, with local contamination, corroborating the calculations of CF and EF. The ER indexes for Zn varied between 0.3 and 1.7 (mean 0.8). It is confirmed by this analysis that despite the macro-drainage program, local sources of metallic contamination continue to reach the Una basin, probably through discards of solid waste and clandestine sewage.

#### 4.3.4 Pollution Loading Index (PLI)

In 2002, the PLI ranged from 0.6 to 2.1. In the year 2015, with the works closed, the PLI presented lower values, ranging from 0.5 to 1.4. During the entire study period, the PLI indicated local sources of pollution in the central area and

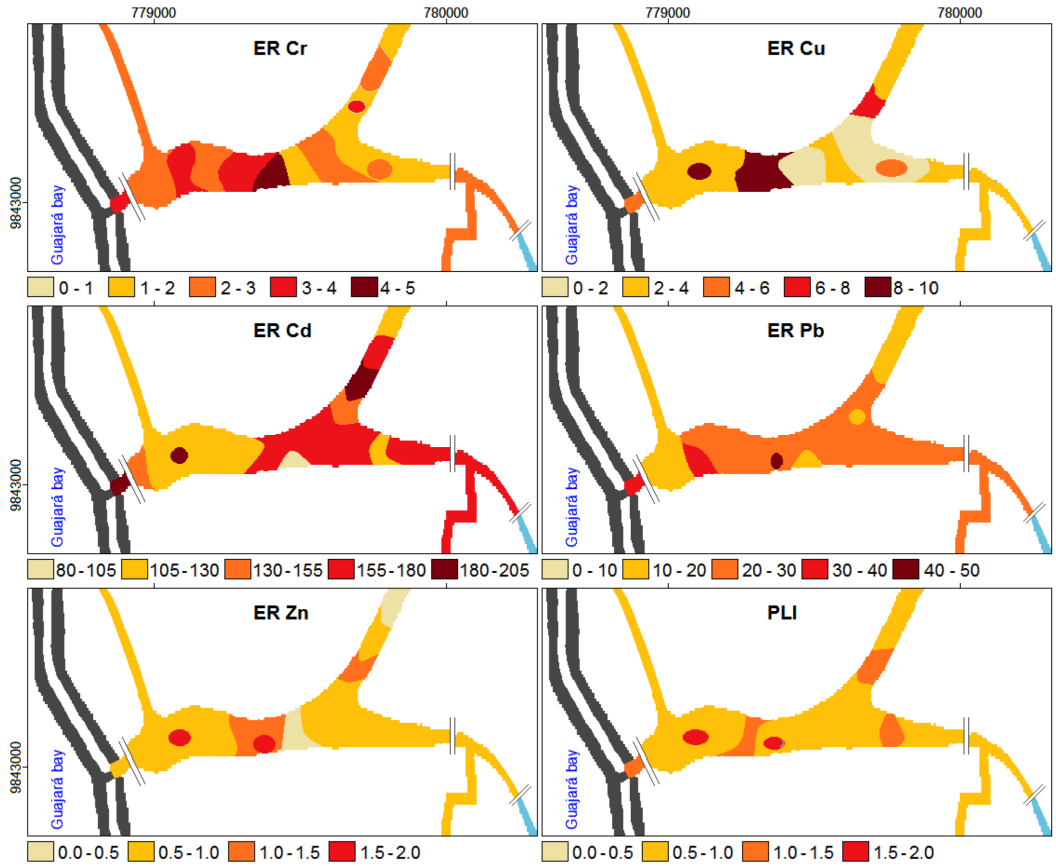


**Fig. 3. Isovalores® charts of the ER and PLI indices for the metal content in the sediments of the Una basin (Pará, Brazil). Surfer® Golden Software, 9.11 (2010)**

closer to the entrance of the Una channel (Fig. 3), confirming the results of the other indexes mentioned. This type of information can be useful not only in environmental monitoring programs but also in engineering projects for installation or restoration of urban drainage infrastructure.

## 4.4 Vulnerability of the Region

The RMB is a complex mosaic where irregular urban development caused successive transformations in the space, modifying topography, relief, soil type, water network and vegetation cover. Several infrastructure works did not have the expected result, aggravating the flooding in the areas of lowlands and erosion in the channels and rivers. The negative effects were partly due to bad planning. However, the lack of monitoring was often fueled by disordered occupation and lack of sense of preservation by the population itself.

The PMBU included in its original proposal the treatment of macro-drainage channels with environmental intervention through dredging and widening of gutters, improvement of water supply conditions, treatment of sewage, urban cleaning services, relocation of families, and restructuring of public spaces (squares and gardens). The channeling of clandestine sewage and urban cleaning should allow a reduction of the pollution loads in the waters and sediments of drainage channels. However, indicators of pollution and environmental contamination suggest that this did not occur considerably. Urban cleaning services include cleaning and collecting public and transporting and final disposal of solid residues from residences, industry, commerce, and construction. In addition, the plan included implementing a new public management model for solid waste, with participatory actions involving government and population, through environmental education programs for civil society (not implemented). This would allow the re-education of society as regards the disposal of waste. A diagnosis of the region showed the appearance of new urban densities [39]. As a result, recurrent problems such as accumulation of waste and debris in the banks and in the interior of the channels, silting, reduction of the natural infiltration area, discharge of clandestine sewage, contamination of surface and underground watercourses, and obstruction of the flow of rainwater begin to reappear in local areas. The lack of consciousness of the population of their role in society is the main source of environmental pollution. The result of the lack of interaction between public authorities and civil society, whose culture of use and irregular occupation in the region is strong, begins to bring back old problems, which include public health issues and reduction of road accessibility, making it even more difficult the services of garbage collection and urban cleaning. It forms a vicious cycle, where from time to time the government intervenes with millionaire projects, and the population re-assumes its role of irregular occupier, degrading the environment.

From the point of view of environmental health, often interventions, whose original purpose would be to improve the quality of life of the population, end up leading to new problems of insalubrities. The epidemiological and sanitary characterization of a macro-drainage area of the Una basin showed that, despite the improvements made possible by the PMBU, situations of social vulnerability, deficiencies in health services and lack of sanitation actions continued to be present in the region [39]. Poor hygiene habits maintain infectious diseases in an endemic situation. The restructuring of the environment by the works increase the local cost of living, with the obligation to pay fees for the new services. In situations such as this, it is common to observe the socio-economic pressure to reallocate the neediest population. The results are unemployment, increased urban violence, lack of public safety, school dropout and accumulation of solid waste, which will interfere with urban drainage, and again increase pollution, water contamination and increased frequency of flood events (cyclical occurrence).

Macro-drainage of the Una basin has caused evident environmental changes, affecting neighborhoods, and therefore, the environmental and sanitary problems persist along with serious socioeconomic difficulties, demanding a community involvement [39,40]. Other socio-environmental indicators should be applied to the region in order to better understand the relationship between interventions by works and urban occupation, thus enabling a better prognosis for environmental pollution and contamination.

## 5. CONCLUSIONS

The Una Basin Macro-drainage Project (PMBU) aimed not only at increasing the flow of rainwater, thus reducing flooding in the region, but also aimed at expanding treated water services, sewage collection and treatment, public cleaning, restructuring of public spaces, such as squares and gardens, and the relocation of the population in an orderly manner. A comparative analysis of the concentration of OM and metals in the sediments of the drainage channels during the work (2002) and after (2015) indicated that the reduction of pollution levels was not significant. The highest rates of reduction with the implementation of the macro-drainage project were observed for metals Mg (58%), Ca (49%) and Zn (38%). The total Fe had a reduction of only 10%, and among the metals with high toxicity to the biota, the lowest reduction rates occurred for Cd (11%), Pb (25%) and Cr total (28%). OM levels also had a small reduction (close to 10%), suggesting a recurrence of sources of pollution to the system. The CF and EF indexes indicated very diversified sediment quality with variable contamination, from low to extremely high enrichment, especially for the Cd, Pb and Zn metals. This result suggests the priority disposal of batteries of telephones and electronic components directly in the channels by the population. ER and PLI suggest diversified and local contamination along the drainage channels, with areas of attention and alertness, especially at the central sites of the Una channel. The lack of consciousness of the local population, associated to lack of public service to sanitation, and new irregular occupations in the region have deteriorated the quality of sediments and probability the waters of the channels. The accumulation of various solid wastes, including debris, along the banks of the channels, has contributed to the resurgence of old problems such as reduction of the natural infiltration area, discharge of clandestine sewage, contamination of watercourses and sediments, sedimentation and obstruction of the rainwater. The PMBU was a project of great impact and high investment for the region, but the lack of maintenance and non-commitment of civil society in preservation have deteriorated the environmental quality of the region.

## DISCLAIMER

This paper has some limitations on Statistical Analysis. Author declares that "There is no statistical analysis because in this paper the focuses were on the indexes. Indexes are values normalized (modified) data, and an Anova in this case is not adequate".

## CONSENT

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## ETHICAL APPROVAL

This section is not applicable in this manuscript.

## COMPETING INTERESTS

Authors have declared that no competing interests exist.

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