

## ANALYSIS OF THE RISK OF CONTAMINATION BY METALS IN AREAS OF SOLID URBAN WASTE DISPOSAL

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### Abstract:

The inadequate disposal of solid urban waste has been of concern, due to the possibility of environmental contamination by various toxic substances, such as heavy metals. The study aims to analyze the concentration of heavy metals in soil and water samples collected around the municipal solid waste (MSW) open dumpsite, Iguatu, Ceará, Brazil. The analysis of risk to human health was determined using Hazard Quotient (HQ). The results showed the presence of heavy metals in concentrations higher than those established by current legislation. The concentration of metals in the soil and water samples were found in the following order, respectively: Fe> Zn> Mn> Cu> Pb> Cr and Fe> Mn> Zn> Ni> Cu> Cr> Cd> Co. However, HQ values indicate that the level of these metals may have little or no adverse health effects. The results suggest the continuous monitoring of the environmental matrices, to avoid the increase of contamination and the risk to human health.

### Keywords:

Artificial aeration; constructed wetlands; nutrients removal; rural sanitation; wastewater treatment

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## INTRODUCTION

The inadequate disposal of solid urban waste has been a concern in recent years, due to the possibility of contamination of soil and water resources. Developing countries have sought strategies to implement effective waste management (Cetrulo et al., 2018; Abdel-Shafy and Mansour, 2018).

In Brazil, the National Solid Waste Policy (PNRS), Law 12.305/2010, contains important instruments for the control of the main environmental, social and economic problems arising from the inadequate handling of solid waste (Brasil, 2010). Brazilian Institute of Geography and Statistics (IBGE) related that in 2017 half of the 5570 Brazilian municipalities did not have an integrated plan for waste management (Costa & Dias, 2020).

In this context, waste management includes rules and laws that aim to minimize environmental damage. The municipal plan for the integrated management of solid waste consists of a diagnosis of the current situation of the set of waste generated in the municipality and defines guidelines, strategies and goals to be developed, related to environmental management, treatment, recycling and final disposal of waste (Brazil, 2010).

The presence of heavy metals in environmental matrices can cause several effects to ecosystems and human health (Reza & Singh, 2010; Arantes et al., 2016; Swarnalatha et al., 2013). The toxic effects of metals depend on the degree of exposure. Metals such as mercury, lead, cadmium, chromium, manganese and zinc can cause damage to the central nervous system, the hepatic, renal and skeletal systems. Manganese is essential, however, in excess it is associated with neurological diseases and behavioral disorders. High levels of copper cause gastrointestinal symptoms, such as diarrhea, abdominal pain and nausea (Cavallet, et al., 2013). A study conducted by Li et al. (2013) aimed to quantify traces of metals and to map the spatial distribution of health risk caused by these elements in the Henan-Liaocheng Irrigation Area (HLIA) in China. The results show that the average the sum of the hazard ratios ( $\Sigma$ HQs) exceeded the generally acceptable level of risk recommended by the USEPA, with a maximum of  $\Sigma$ HQs of 3.79. Horta et al., (2011) estimated the risk of contamination by cadmium (Cd) through fish ingestion by the population of Sepetiba Bay-SE, Brazil. The probability and risk due to cadmium intake was assessed using the hazard ratio (HQ). The risk for cadmium intake was 110 times greater than that found in population control, which may be related to kidney diseases.

The inadequate disposal of Urban Solid Waste can interfere with the levels of toxic metals in the ecosystem (Gouveia, 2012; Tucker et al., 2020). The main routes of exposure to these contaminants are soil dispersion and leachate percolation, reaching water resources.

Several studies have cited the contamination of toxic metals in water sources and soil (Ahmad & Goni, 2010; Souza et al., 2016; Silva et al., 2018). Silva et al. (2018) carried out research to determine the concentrations of Cd, Cu, Mn, Ni, Pb and Zn in waters used for human and animal consumption and in the irrigation of vegetables in the municipality of Camocim de São Félix-PE, Brazil. According to the results of the study, the analyzed waters presented concentrations of Cd ( $> 0.001 \text{ mg L}^{-1}$ ), Ni ( $> 0.025 \text{ mg L}^{-1}$ ) and Pb ( $> 0.01 \text{ mg L}^{-1}$ ) higher than the maximum permitted levels by Brazilian law, indicating the need for a preventive monitoring program and immediate intervention actions for sources of contamination. Sabiá et al., (2015) studied the influence of the leachate produced by the "garbage dump" on the water quality of the Salgado River Basin, Ceará, Brazil. According to the authors, the metals Cadmium, Silver and Nickel at all collection points were in concentrations above the limits allowed by CONAMA Resolution 357/05 and CONAMA 430/11. Kanmani & Gandhimathi (2013) evaluated the concentration of heavy metals in soil samples collected around the open solid urban waste dump (MSW), Ariyamangalam, Tiruchirappalli, Tamilnadu. The concentration of heavy metals in the collected soil sample was found in the following order: Mn > Pb > Cu > Cd, indicating that there is appreciable contamination of the soil by leachate migration from an open dumping site.

For about 30 years, the disposal of solid waste from the city of Iguatu, Ceara, Brazil has been done in open dumpsite, which may compromise the quality of life of the population. This work aims to expand the discussion on environmental pollution by heavy metals from the analysis of water and soil samples located around of the open dumpsite and to assess the risk to human health using Hazard Quotient (HQ).

## MATERIAL AND METHODS

### Local of study and sampling

The municipality of Iguatu is located in the Center-South Region of the state of Ceará at latitude  $06^{\circ}21'34''$  South and at longitude  $39^{\circ}17'55''$  West, with an area of 1,029,002 km<sup>2</sup> and an average population of 102,614 thousand inhabitants. The caatinga is the predominant vegetation in the region and the soils are podzolic, litolic, alluvial and vertisol (Ibge, 2017)

Since 1989, the "dump" has been installed next to the CE-282 highway, Chapadinha, CE. According to Ibge (2017), the main sources of water in the city of Iguatu are part of the Upper Jaguaribe basin, the rivers: Jaguaribe and Trussu; streams Carnaúba, Antônio, da Serra and many others. The rainfall in the municipality is 1079 millimeters per year, with rains concentrated from January to May.

**Table 1.** Soil (P<sub>S1</sub>, P<sub>S2</sub>, P<sub>S3</sub>, P<sub>S4</sub>) and water (P<sub>W1</sub>, P<sub>W2</sub>, P<sub>W3</sub> and P<sub>W4</sub>) collection points in the municipality of Iguatu-CE

Sampling	Geographic Coordinates
<b>Soil</b>	
P <sub>S1</sub>	6°22'51.20"S / 39°15'4.00"O
P <sub>S2</sub>	6°22'50.20"S / 39°15'3.90"O
P <sub>S3</sub>	6°22'49.40"S / 39°15'4.30"O
P <sub>S4</sub>	6°22'48.30"S / 39°15'4.20"O
<b>Water</b>	
P <sub>W1</sub>	6°22'44.24"S / 39°15'15.32"O
P <sub>W2</sub>	6°22'40.53"S / 39°14'56.75"O
P <sub>W3</sub>	6°23'15.51"S / 39°15'22.24"O
P <sub>W4</sub>	6°23'14.58"S / 39°15'12.68"O

Soil and water samples were collected during the period 2016/2017, in areas susceptible to contamination by solid urban waste disposed of inappropriately in the municipality of Iguatu-CE, Brazil. The soil samples (P<sub>S1</sub>, P<sub>S2</sub>, P<sub>S3</sub>, P<sub>S4</sub>) were collected in a 20 cm column. The samples were packed in plastic bags and sent for analysis. The water samples (P<sub>W1</sub>, P<sub>W2</sub>, P<sub>W3</sub> and P<sub>W4</sub>) were stored in polypropylene bottles, previously sanitized and transported to the chemistry laboratory of the Federal Institute of Science and Technology of the Ceará- IFCE and kept refrigerated until the moment of the analysis.

### Sample pre-treatment

The soil samples were first submitted to an acid digestion process following the methodology described by Method 3050b (USEPA, 1996) About 0.2 g of soil was weighed on an analytical balance and transferred to an Erlenmeyer. A 10 ml volume of 0.1 M HCl was added to the vessel. The mixture was then subjected to constant stirring, at room temperature ( $\pm 25$  °C) for about 2 hours. After acid digestion, the mixture was filtered and transferred to a 50 ml volumetric flask and the volume was measured with distilled water. All samples were analyzed in duplicate. Water samples were acidified with nitric acid (HNO<sub>3</sub>), according to the Method 200.7 (USEPA, 1982) A volume of 2.20 mL of HNO<sub>3</sub> was added in 100 mL of water sample and then identified and stored under refrigeration for later analysis of metals. All samples were analyzed in duplicate.

### Metal analysis

The determination of metals (Cd, Co, Cr, Cu, Fe, Mn, Ni, Cr and Zn) in soil samples were performed by Atomic Absorption Spectrometry - AAS (Varian-Zeeman, model 640-Z). Standard solutions containing metals in the concentration range of 0.05-5.0 mg.L<sup>-1</sup> were used for the calibration curves. The limits of quantification (LOQs) were 0.05 mg.L<sup>-1</sup> for Mn, Co, Ni, Cu, Cd, and 0.01 mg.L<sup>-1</sup> for Pb, Cr and Zn. The level of metals (Cd, Co, Cr, Cu, Fe, Mn, Ni, Cr and Zn) in the

water samples were determined by Spectrophotometry of Optical Emission with Plasma Inductively Coupled ICP-OES (model ICAP 6000 series, Thermo Scientific). The results were analyzed using the Itava Control Center software. Calibration curves were prepared in the concentration range between 0.005-2 mg.L<sup>-1</sup>. The limits of quantification (LOQs) were 0.001 mg.L<sup>-1</sup> for the metals analyzed.

### Health risk assessment

The health risk assessment study was based on the determination of the Hazard Quotient (HQ) according to the guidelines of the United States Environmental Protection Agency (USEPA, 1989). Chronic daily intake (CDI) and HQ were determined using equations 1 and 2 (USEPA 1989; Wu et al. 2009):

$$CDI \text{ (oral)} = (C_w \times IR_w \times EF \times ED) / (B_w \times AT) \quad (1)$$

$$HQ = CDI / RfD \quad (2)$$

where CDI= Chronic daily intake,  $\mu\text{g} \cdot \text{Kg}^{-1} \cdot \text{day}^{-1}$ ; C<sub>w</sub>: average concentration of trace metal in water,  $\mu\text{g} \cdot \text{L}^{-1}$ ; IR<sub>w</sub>: water ingestion rate, 2 L.day<sup>-1</sup>; EF: exposure frequency, day/year; ED: exposure duration, 70 years; BW: average body weight, 70 kg; AT: averaging time, for noncarcinogens, days (Zabin et al., 2008); RfD is the reference dose for different analytes, expressed in  $\mu\text{g} \cdot \text{Kg}^{-1} \cdot \text{day}^{-1}$  (USEPA, 2012).

## RESULTS AND DISCUSSION

### Soil Analysis

Higher levels of Fe, Zn and Mn metals were verified in the soil samples, as shown in the **Fig. 2**. The concentrations of heavy metals present in the soil samples were compared with the guiding values of soil quality regarding the presence of chemical substances established by CONAMA Resolution 420/2009, described in Table 1.

According to the Resolution, Quality Reference values (VRQ) correspond to the concentration of a certain substance that defines the natural quality of the soil. This value may vary from State to State and it is the duty of each one to present these values, however in the State of Ceará they do not yet present these estimated values. Prevention values (VP) consist of values that limit a certain substance in the soil and if exceeded, it is necessary to monitor it. Intervention (VI) values are values that determine potential risks, human health, direct or indirect, considering a standardized exposure scenario. The results in **Table 2** showed higher concentrations to Fe element, varying between 204,1-1229,3 mg/L. The Zn value found in PS3 (Zn = 402.23 mg.Kg<sup>-1</sup>) exceeded the prevention value (PV =

300 mg Kg<sup>-1</sup>). The same was verified for copper in PS2 (Cu =109.1 mg.Kg<sup>-1</sup>) and PS3 (117.1 mg.Kg<sup>-1</sup>) exceeding the prevention value VP = 60 mg Kg<sup>-1</sup>, indicating that there is a change in the soil quality. Copper is one of the most important metals industrially, an excellent conductor of electricity is currently used for the production of conductive materials.

The metals Cd, Co and Ni, were not present (<LQ) in the samples investigated in the soil near the dump of Iguatu-CE. Fe and Mn metals do not yet have limits established by the CONAMA 420/2009 legislation. The data indicate that the levels of this metal are being altered due to the human anthropic action in the garbage dump. Cu and Mn were present in all samples, except in the PS1 sample, with higher concentrations in PS3 (117.1 and 132.2 mg.Kg<sup>-1</sup> respectively). Pb was detected in all samples (5.0 - 45.0 mg.Kg<sup>-1</sup>), but within the limits established by the guiding values for soil (VP and VI residential). Lead is a highly toxic metal, in addition to damaging the environment, it is harmful to the brain and nervous system, in addition to affecting the blood, kidneys, digestive and reproductive systems.

The metal Chromium was detected only in PS3 and PS4, with levels from 8.8 to 18.2 mg.Kg<sup>-1</sup>, respectively. Marques (2011) detected a similar level of Cr in the soil (0-20 cm) of the landfill in Campo Belo-MG (21.09 mg Kg<sup>-1</sup>) and Elói Mendes-MG (20.72 mg Kg<sup>-1</sup>). Chromium is a heavy metal, has a cumulative effect and can cause various damages to the health of humans and animals.

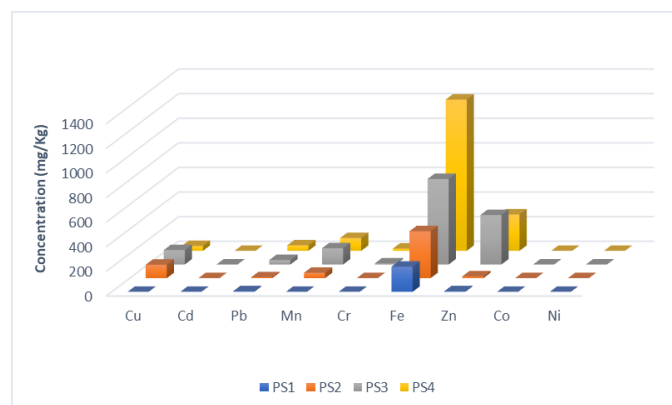


Fig. 2 Concentration of metals in mg.kg<sup>-1</sup> in soil samples

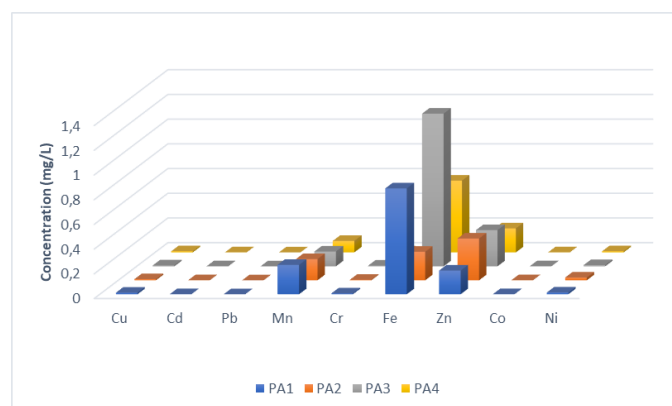


Fig. 3 Metals analyzed (mg.L<sup>-1</sup>) in water (P<sub>W1</sub>, P<sub>W2</sub>, P<sub>W3</sub> and P<sub>W4</sub>)

## Analysis of metals in water

Among the heavy metals analyzed in this study, Mn, Zn and Fe showed higher levels in water samples from mananciais located near the Iguatu dump. Figure 3 shows the concentrations verified for the elements analyzed in the water samples. **Table 1** shows the average concentration of metals obtained at each sampling point and the maximum limits established by national and international legislation (Brasil, 2005; Brasil, 2011; Eu, 1998; Epa, 2019).

The analysis of water samples from sources close to the Iguatu-CE dump showed the presence of Mn (0.093-0.238 mg.L<sup>-1</sup>) and Zn (0.192-0.338 mg.L<sup>-1</sup>), in concentrations above the maximum allowed by the resolution CONAMA 357/05 (Class 2). The levels of metals detected can be related to the infiltration of the leachate produced in the decomposition of the garbage, causing the alteration in the water quality of the water sources in the region.

A previous study conducted by Abreu (2014) detected traces of Mn (0.14 to 0.15 mg.L<sup>-1</sup>) and Zn (0.04 to 0.05 mg.L<sup>-1</sup>) in a lake close to the dump in Iguatu-CE, however the levels found still fit within the established standards of CONAMA 357/05. Comparing these results with those of the present research, it appears that there was a significant increase in contamination by these metallic elements. The detected Cd concentrations (0.0001 mg.L<sup>-1</sup>) in the water samples were within the acceptable range. For the element Ni, the highest concentration found was at point two (P2) 0.0238 mg.L<sup>-1</sup>, however these values found at the sampling points investigated were below the limit (0.025 mg. L<sup>-1</sup> Ni) (Conama, 2005). Research developed by Anjos (2015) reported contamination by Cd and Ni in the waters of the Rio Curu-CE, at levels ranging from 0.00010- 0.00103 mg.L<sup>-1</sup> and 0.0002 - 0.0038 mg.L<sup>-1</sup>, respectively.

High concentrations of Fe were found in this study, ranging from 0.231 to 1.236 mg.L<sup>-1</sup>, being above the EU acceptable value. Higher levels of Fe were observed in P1, P3 and P4, exceeding the limits of CONAMA, MS, EU and EPA. Fe is a chemical element that does not present much risk to health, but can have effects on water quality when taken together (Fe<sup>2+</sup> and Mn<sup>2+</sup> form soluble in water). These elements are considered by the United States Environmental Protection Agency (EPA), Canada, the European Union and the Ministry of Health-Brazil as secondary contaminants capable of altering organoleptic standards and water potability. Cu was detected at all points in concentrations close to the established limit of the legislation (CONAMA 357/2005) of 0.009 mg.L<sup>-1</sup>, reaching the maximum value in P1 0.013 mg.L<sup>-1</sup>. Traces of Co and Cr were detected in all samples analyzed. Pb was not detected in this study.

**Table 2.** Concentration of soil samples in mg.kg<sup>-1</sup> and Quality Reference Values-QRV, prevention values-PV and intervention values-IV (residential) established by CONAMA.

Metal	Concentration (mg/Kg)				Conama 420/2009		
	PS1	PS2	PS3	PS4	QRV	PV	IV
Cu	<LOQ	109.1	117.1	39.8	E	60	400
Cd	<LOQ	<LOQ	<LOQ	<LOQ	E	1.3	8
Pb	5.0	7.5	36.2	45.0	E	72	300
Mn	<LOQ	43.6	132.0	104.2	E	-	-
Cr	<LOQ	<LOQ	8.8	18.2	E	75	300
Fe	204.1	381.6	695.1	1229.3	E	-	-
Zn	4.6	14.7	402.2	295.8	E	300	1000
Co	<LOQ	<LOQ	<LOQ	<LOQ	E	25	65
Ni	<LOQ	<LOQ	<LOQ	<LOQ	E	30	100

\*<LOQ: Limiton of Quantification; E= to be defined by the State.

**Table 3.** Average concentration of metals analyzed in water samples in mg.L<sup>-1</sup>

Metal	Concentration (mg/L)				Conama 357/05	MS 2.914/11	EU	EPA
	PW1	PW2	PW3	PW4				
Cu	0.0132	0.0091	0.0086	0.0094	0.009	2.000	2.000	1.300
Cd	0.0017	0.0013	0.0018	0.0018	0.001	0.005	0.005	0.005
Pb	<LOQ	<LOQ	<LOQ	<LOQ	0.010	0.010	0.010	0.005
Mn	0.2380	0.1720	0.1200	0.0930	0.100	0.100	0.050	0.050
Cr	0.0079	0.0040	0.0064	0.0082	0.050	0.050	0.050	0.100
Fe	0.8570	0.2312	1.2363	0.5810	0.300	0.300	0.200	0.300
Zn	0.1926	0.3384	0.2938	0.1941	0.180	5.000	-	5.000
Co	0.0015	0.0011	0.0016	0.0010	0.050	-	-	-
Ni	0.0143	0.0230	0.0137	0.0091	0.025	0.070	0.020	-

\*<LQ: Limit of Quantification; Conama 357/05 Class 2

Contamination of heavy metals has been associated with improper disposal of solid waste in the dump. Manganese is used in the manufacture of paints, ceramics, glass, welding electrodes, they are present in the waste deposited in the dump. The effects of this metal found at high levels can cause respiratory tract infection and pneumonitis from irritation. Its systemic toxicity is more common in chronic exposure through inhalation and ingestion, where the most pronounced effects are due to prolonged exposure to contaminated environments reaching the central nervous system. High concentrations of Mn can cause serious damage to humans, such as decreased motor movements, mental and emotional disorders.

### Health risk assessment

RfD<sub>oral</sub> values for trace metals and the health risk assessment study is summarized in table 4. The calculated CDI values of metals for oral intake showed values between 0.0286 to 35.3229 µg. Kg<sup>-1</sup>.day<sup>-1</sup>. The non-cancerous effects of different metals were determined by calculating the risk quotient (HQ), which indicates the probability of adverse health effects. However, the results of the present study indicate that the level of these metals (maximum value of 0.34) may represent few or no adverse health effects (HQ <1) for the population of Iguatu-CE by oral water intake. Research conducted by Karim (2011) obtained similar results for risk assessment for metals in drinking water in the Karachi region, Pakistan.

**Table 4.** Analysis of risk of contamination by metals in water

Metal	*RfD (µg. Kg <sup>-1</sup> .day <sup>-1</sup> )	Chronic daily intake (µg. Kg <sup>-1</sup> .day <sup>-1</sup> )				Hazard Quotient			
		CDI <sub>1</sub>	CDI <sub>2</sub>	CDI <sub>3</sub>	CDI <sub>4</sub>	HQ <sub>1</sub>	HQ <sub>2</sub>	HQ <sub>3</sub>	HQ <sub>4</sub>
Cu	40	0.3771	0.2600	0.2457	0.2686	0.0094	0.0065	0.0061	0.0067
Cd	0.5	0.0486	0.0371	0.0514	0.0514	0.0971	0.0743	0.1029	0.1029
Pb	1.4	NA	NA	NA	NA	NA	NA	NA	NA
Mn	20	6.8000	4.9143	3.4286	2.6571	0.3400	0.2457	0.1714	0.1329
Cr	3	0.2257	0.1143	0.1829	0.2343	0.0752	0.0381	0.0610	0.0781
Fe	300	24.4857	6.6057	35.3229	16.6000	0.0816	0.0220	0.1177	0.0553
Zn	300	5.5029	9.6686	8.3943	5.5457	0.0183	0.0322	0.0280	0.0185
Co	NA	0.0429	0.0314	0.0457	0.0286	NA	NA	NA	NA
Ni	20	0.4086	0.6571	0.3914	0.2600	0.0204	0.0329	0.0196	0.0130

\*USEPA, 1989; Wu et al., 2009

## CONCLUSION

Analysis of soil samples from the region around open dumpsite of Iguatu-CE, Brazil, indicate high concentrations of Fe, Zn and Mn. In addition, the levels of Zn and Cu remained above the prevention values established by Brazilian legislation. The elements Pb, Mn, Cr were also detected in soil. The elements Zn, Fe, Mn, Cu and Cd showed values above that allowed by water legislation. According to the results of soil and water metals obtained, the research suggests that the level of these identified metals is likely to have little or no adverse health effects, in view of verifying that the hazard ratio is less than one, indicating low risk contamination by metals. Therefore, the data show that the dump has been contributing to the pollution of these springs. Thus, this study contributes to assist environmental agencies in controlling the quality of water in the region and in the continuous monitoring of toxic metal levels in water sources.

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