



Geochemical indices for Zn, Cu, and Co in sediment cores of Coata River, Puno Region, Peru

Índices geoquímicos para Zn, Cu, and Co em testemunhos de sedimentos no Rio Coata, região de Puno, Peru

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ABSTRACT

Urban contamination resulting from the dynamics of unplanned cities has generated interest in recent decades due to increased pollutant emissions into the natural environment. The Coata River is a complex ecosystem exposed to pollution due to the high urbanization degree and consequent industrial dynamics, as well mining activities. Significant amounts of waste and pollutant substances are discharged into the river, often without any treatment, contributing to a drastic increase in the concentration of heavy metals that cause impacts and pressures on the environment. In this context, the present study contributes to the evaluation of the metal concentration (Zn, Cu, and Co) in three short sediment cores sampled at Coata River, Puno region, Peru. The acquired concentration data was compared with guideline reference values established by international sediment quality standards and used for calculations of the geochemical indices Enrichment Factor and Geoaccumulation Index. The highest concentrations were found for zinc (639 mg/kg) and copper (96 mg/kg), whilst zinc exhibited some values above the guidelines corresponding to the probable effect level (PEL, legislation of Canada) and mean range effects (ER-M, legislation of the United States). According to the sediment quality indices, the three stations range from moderately to highly contaminated with Zn, while Cu and Co show no contamination and moderate contamination, originating from both geogenic and anthropogenic sources.

Keywords: sediment cores, geoaccumulation index, enrichment factor, Coata River, Peruvian Puno region

RESUMO

A contaminação urbana resultante da dinâmica de cidades não planejadas tem gerado interesse nas últimas décadas em virtude do aumento da emissão de poluentes no meio ambiente. O Rio Coata é um ecossistema complexo que está à poluição devido ao alto grau de urbanização e consequente dinâmica industrial, bem como atividades de mineração. Quantidades significantes de rejeitos e poluentes são descartadas no rio, geralmente sem qualquer tratamento, contribuindo para um acentuado acréscimo na concentração de metais pesados causadores de impactos ambientais. Nesse contexto, o presente estudo contribui para a avaliação da concentração de metais (Zn, Cu e Co) em três testemunhos de sedimentos coletados no Rio Coata, região de Puno, Peru. Os valores de concentração obtidos foram comparados com os de referência estabelecidos pelos padrões internacionais de qualidade dos sedimentos, bem como foram utilizados no cálculo dos índices geoquímicos correspondentes ao Fator de Enriquecimento e Índice de Geoacumulação. As maiores concentrações foram obtidas para o zinco (639 mg/kg) e cobre (96 mg/kg), tendo o zinco exibido alguns valores acima dos níveis de referência correspondentes ao efeito provável (PEL, legislação do Canadá) e efeito de alcance médio (ER-M, legislação dos Estados Unidos). De acordo com os índices de qualidade dos sedimentos, os sedimentos dos três locais de amostragem exibiram contaminação de moderada a alta para Zn, bem como moderada ou nenhuma contaminação para Cu e Co, cujas fontes possíveis poderiam ser geogênicas e antropogênicas.

Palavras-Chave: perfis de sedimentos, índice de geoacumulação, fator de enriquecimento, Rio Coata, região peruana de Puno.

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DOI:10.21715/GB2358-2812.202438006

1. INTRODUCTION

Environmental contamination is regarded as one of the most serious problems confronting society in the twenty-first century. Aquatic environments affected by contamination have garnered a lot of attention in recent decades as a result of the exponential expansion in population along their riverbanks, as well as the expanding level of industrialization and the contributions that are presented from the primary sector (CASTRO *et al.*, 2013). Heavy metals are a major component of anthropogenic sources that are harmful to aquatic biota, humans, and the general environment (ASADI *et al.*, 2017; GALLEGO-ÁLVAREZ, 2018). In aquatic ecosystems, the entry of heavy metals is very harmful due to their high toxicity, persistence and rapid accumulation by living organisms. The toxicity of certain metals is proportional to their ease of absorption by living beings; a metal dissolved in ionic form is more easily absorbed than an elemental form (PROSHAD *et al.*, 2018; ATTAH *et al.*, 2021).

According to the U.S. Agency for Toxic Substances and Disease Registry (ATSDR, 2008), the most toxic heavy metals are Sb, As, Cd, Cu, Cr, Hg, Ni, Pb, Se, and Zn. Heavy metals can have either beneficial or detrimental impacts on living beings; some of them, at concentrations of less than 0.01% of the total mass of the body, are required components for life, exactly as V, Cr, Mo, Mn Fe, Co, Ni, Cu, and Zn are for humans. However, small variations in their concentrations, both decreases and increases, can produce harmful effects, sometimes serious, chronic and even lethal, in living beings (ALI *et al.*, 2022). These metals contribute to the hydrological cycle from a variety of sources, one of which is lithological in origin. However, the present highest concentration is of anthropogenic origin through the activities mining, industrial processes and household waste that are important sources of contamination, because they contribute metals to the environment (MARIN-LEAL *et al.*, 2017).

Peru is a country with extensive natural resources and a wide variety of biodiversity in the coast, highlands, and jungle, whose environmental quality of the aquatic ecosystem has been generally compromised in various forms (HERNÁNDEZ, 2019; FERNANDES *et al.*, 2020). Puno is an important Peruvian region, where multiple socioeconomic activities have been carried out involving different productive sectors for promoting the regional

development (HERNÁNDEZ, 2019; FERNANDES *et al.*, 2020). As a consequence, environmental problems take place there, chiefly related to the discharge of untreated domestic, industrial and mining effluents, enhancing pollution in the receiving waters due to the discharged volume and types of toxic substances (GUERRERO; ZAVALA 2006; OSINERGMIN, 2016; BELIZARIO *et al.*, 2019). In this context, Coata River in Puno region is characterized as a strategic site for environmental surveys as it flows through several cities occurring there, especially Juliaca, which contains various industrial factories responsible for emissions of solids, liquids, and gaseous wastes into the environment. In addition, mining activities, domestic sewage from the population as indicated by metals such as Zn, Cu and Co, according to Chipasa (2003) and Feng *et al.* (2023), among others, has also caused profound transformations in the local environment.

Some searches have been already done at Puno region focusing the presence of heavy metals and other contaminants in sediments. For instance, Cornejo-Olarte *et al.* (2023) examined harmful metal concentrations with values of As (14.90 mg/kg), Cd (0.70 mg/kg), and Hg (0.30 mg/kg) beyond the TEL and SESS reference standards and Cr (28.41 mg/kg) above the SESS. Also, Quispe *et al.* (2019) measured maximum concentrations of Cr (28.42 mg/kg), Cd (0.70 mg/kg), and Pb (16.50 mg/kg), with only Cr exceeding the Peruvian Ministry of Environment's environmental quality guidelines for soils. In water samples, Belizario *et al.* (2019), assessed the following metal concentrations: Al (1,043 mg/L), Fe (0.856 mg/L), Mn (0.460 mg/L), As (0.029 mg/L), and P (10.287 mg/L). In wastewater from Torococha River (Coata River tributary) at Juliaca city, Asqui (2015) found high levels of Pb (0.2889 mg/L) and Zn (2.3800 mg/L) that are above the ECA limits. The aforementioned authors linked the metals inputs into Coata River sediments to anthropogenic activities, mainly related to residential and industrial activities taking place at Juliaca city.

It is important highlight that sediments are one of the main reservoirs of metals, acting as secondary contamination resources in aquatic environments (RUBIO *et al.*, 2006; ASADI *et al.*, 2017). Once contaminated, sediments represent a latent source of continued degradation of the environment, which can

deteriorate water quality after the reduction or even complete abatement of anthropogenic emissions. In this sense, the determination of metals in sediments is a good indicator of the origin of pollutants in the environment and the impacts they can produce on aquatic biota (FUENTES-HERNÁNDEZ, 2019).

Nowadays, heavy metals contamination has become one major environmental problem, originating largely from anthropogenic sources that pose a danger to aquatic biota and humans (STAMATIS *et al.*, 2019; WU *et al.*, 2019). Heavy metals generally considered toxic are those that have a density equal to or greater than 6 g/cm³ in their elemental form or whose atomic number is greater than 20 (excluding alkali and alkaline-earth metals). According to the Agency for Toxic Substances and Disease Registry in the USA, Zn and Cu, along with Pb, As, and Hg, are in the top list of the 10 major environmental contaminants of concern (ATSDR, 2008; MARIN-LEAL *et al.*, 2017).

To assess the degree of metal contamination in sediments and identify if the contamination source is natural or anthropogenic, geochemical indices are used, most frequently the Enrichment Factor (EF) and Geoaccumulation Index (Igeo) (ZHUANG *et al.*, 2018). Their calculation is based on the relationship between the total metal content and its background value (reference value), adopting comparison of current to reference values, i.e. the expected concentration level that the region would

naturally have. More recent studies have successfully applied these indices as tools for assessing heavy metal contamination in different aquatic sediments, such as Cáceres *et al.* (2013) in Peru, Jena *et al.* (2019) in India, Bonotto (2020a,b) in Brazil, and Abidi *et al.* (2022) in Spain.

Oliveira *et al.* (2015) also presented a detailed study in estuarine regions with a high industrial concentration in Brazil, where they evaluated the contents of Cr, Cu, Ni, Pb, and Zn in bottom sediments of Guajará Bay and Carnapijó River using the geochemical indices Igeo and EF to determine sediment contamination by anthropogenic sources. These authors pointed out that Igeo indicated Pb as the metal with the highest values in sediments, constituting an element that could be transported over longer distances compared to others analyzed. Thus, this finding highlighted the importance of this geochemical index as a tool for evaluating and monitoring contaminants inputs in the environment.

Based on the foregoing, this study aims to assess contamination of the heavy metals Zn, Cu, and Co in short sediment cores providing from three monitoring points at Coata River, thus, involving the determination of their total concentration, comparison with guideline reference values concentrations established by sediment quality standards, and calculation of the EF and Igeo indices.

2. MATERIALS AND METHODS

2.1. STUDY AREA

Coata River is located in the southern region of Peru, more specifically in the Department of Puno (Fig. 1). Coata River is a tributary of Lake Titicaca, crossing the Peruvian territory, particularly in the Puno region, encompassing waterbodies from its source (Nevado de Quilca), under the name of Vila Vila River, and receiving different names according to the flowing through places. For instance, in the vicinity of Lampa district, it is called Lampa River. After receiving the contribution of Cabanillas River, it forms the Coata River until reaching its mouth at Lake Titicaca, thus, covering a length of 170 km along its route. Coata River borders Ramis River northwards, Illpa River southwards, Lake Titicaca eastwards, and Lampa/Cabanillas rivers westwards (INRENA, 2007; SENAMHI, 2016).

According to Rivas (2021), the geological formations located in the study area range from the Paleozoic era to Quaternary deposits, the latter being the most notable for their lithological formation present on both banks of Coata River, such as: (a) Fluvial/glacial-composed of transitional clastic deposits and incipient stratification consisting of coarse sand, gravel, and blocks in smaller quantities; (b) Alluvial- deposits associated with valley bottoms, depressions, plains, and mountain slopes, consisting of unconsolidated clays, silts, sands, and gravels deposited by river currents and water flows, including fluvial and colluvial sediments; (c) Biogenic- consisting of clays, silts, sands, and organic debris; (d) Lacustrine-consisting of silty clays and sands; (e) Colluvial- deposits associated to geomorpho-

gical units of mountain slopes and hills, consisting of clasts with a matrix of stony sand or sandy gravel (Fig. 2). The intrusive cover can be linked to Versailles, a granodiorite body, and Collque Orco, which has a monzoquartzite composition both of which are among the most representative of the site located in the study area.

Geomorphologically, the research area belongs to the Altiplano unit, which is divided into three subunits: (a) Sedimentary Plains- the principal feature of the Altiplano, consisting of

geoforms and located to NW and SE of Puno, with altitudes ranging from 3,850 to 3,900 m; (b) Carbonate Rocky Hills- primarily composed of a mass of limestone and silt from the Ayabacas Formation, whose location is on the sedimentary plains northwards of Coata, reaching altitudes of 3,900 to 4,200 m; (c) Isolated hills of detrital and volcanic rocks- formed by Puno Group sandstones and Mitu Group volcanic sequences, located at NW of Atuncolla and SW of Juliaca, reaching an altitude of 4,050 m.

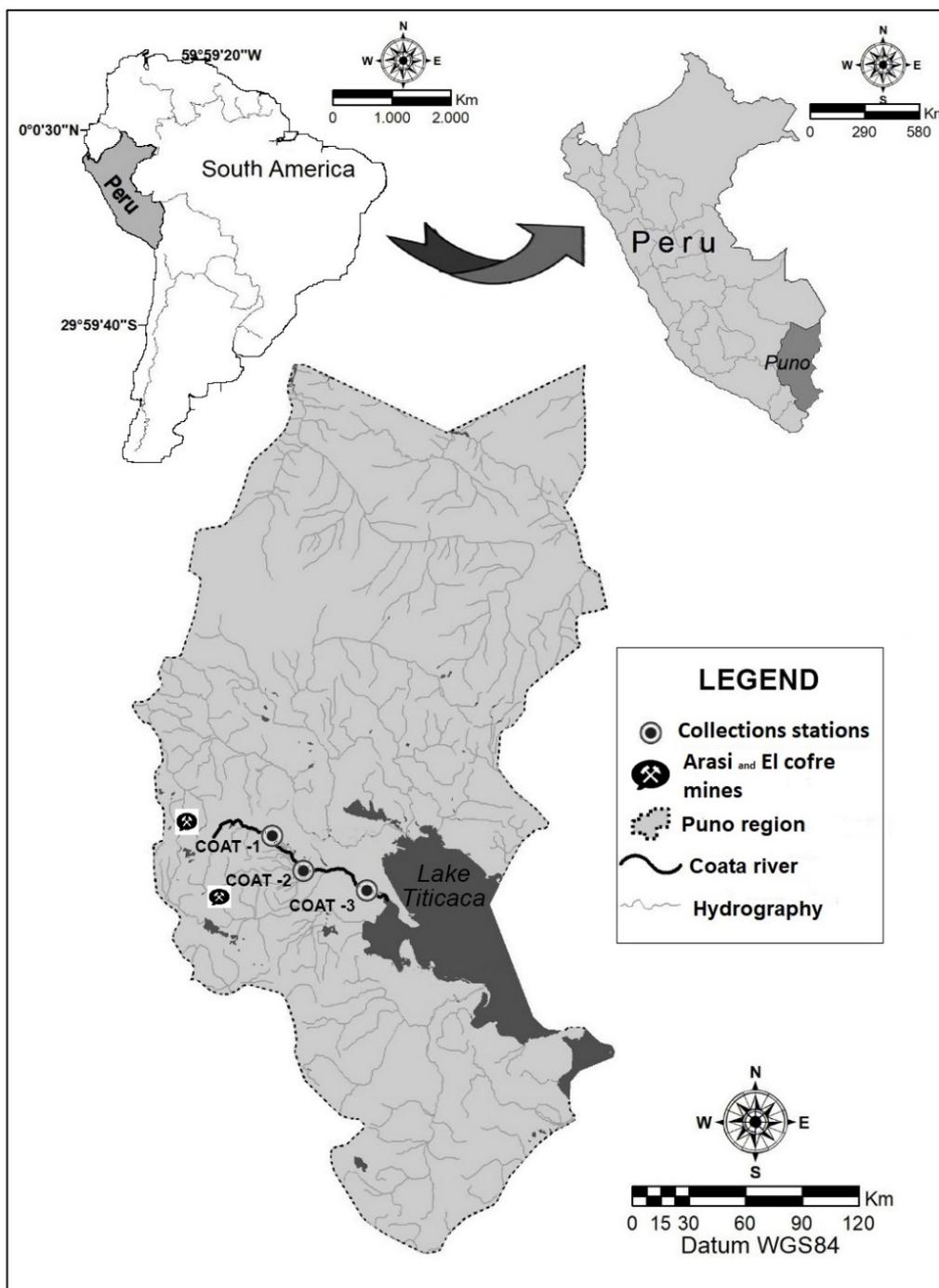


Figure 1 - Location of the study area and monitoring points at Coata River, Puno Region, Peru.

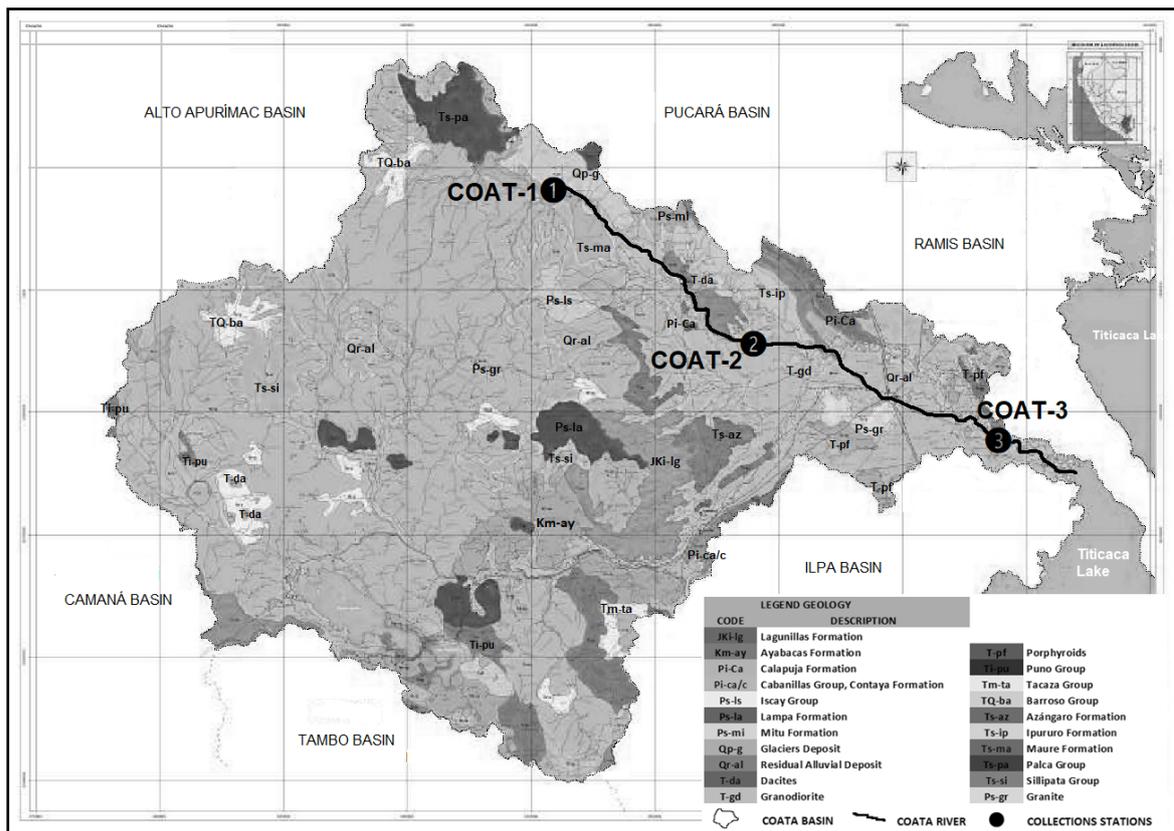


Figure 2 - Geological map of basin Coata, Puno Region, Peru (INGEMMET, 2003)

The Puno region, where Coata River is located, possesses a population of over one million inhabitants, having 17 hydrographic units that cover 64% of the regional territory. It is an area that provides a variety of ecosystems-related services, including landscape appreciation, recreation, and tourism activities, such as those found on the Islands of Amantani, Uros, and Taquile, as well as fishing and various other water sports.

Regarding environmental contamination sources in Puno area, approximately 70% of the industries are located at Juliaca city that is the most populated in the region, with 375,267 inhabitants. According to projections by the Chamber of Commerce and Production of San Román (Puno), handicrafts, mining, and commerce are the activities that have had the greatest presence in this city over the past 20 years. Another indicator of this economic growth in Juliaca city is the installation of numerous financial institutions and transnational networks with their respective shopping centers. These industries include oil refineries, metallurgical, chemical, textile, and clothing industries, among others. Juliaca city is

known as the "Capital of Andean Integration" and a major commercial hub, being considered one of the main economies in the region and Peru (INEI, 2013).

Coata River is the main water source supplying Juliaca city as exploited from Ayabacas sector that is located northeast of Juliaca. After collection, the water is treated and distributed through the public water network to the different urban sectors. According to EPS Sedajuliaca S.A, the Regional Health Directorate and Environmental Assessment and Inspection Agency responsible to propose actions for controlling the discharge of sewage, wastes, and contaminating residues, around 35% of existing properties lack a drinking water connection/installation, while 70% of the population in the region lacks sufficient sewage treatment, resulting in untreated discharges into the river. Thus, contamination has become a determining environmental problem there, as the ecosystem has been subjected to critical conditions due to high pollution levels caused by sewage discharge, industrial, mining activities, and improper solid waste disposal in the nearby cities, primarily in the upper and

middle zones (GUERRERO; ZAVALA, 2006; Diaz *et al.*, 2012; TUMI, 2014; JABBOUR, 2015; GALLEGO-ÁLVAREZ, 2018; LIPA VILCA, 2019; TOVALINO, 2019; QUISPE *et al.*, 2020).

Therefore, because there is a lack of wastewater treatment plants at Juliaca city, the discharges occur directly into Torococha River that flows through the city, becoming a source of chemical, biological, and visual contamination for the population. The majority of homes connected to the river discharge more than 9,000 m³/day of wastewater (HUACANI, 2013; PNUMA, 2015; LIPA VILCA, 2019). Untreated water from industries, as well as the Juliaca oxidation pond that is the final disposal site for the municipal slaughterhouse and solid wastes (plastic bags, disposable bottles, common garbage and sometimes even dead animals) are all dumped into Coata River. Reasons for deterioration of the Coata River water quality are similar to those already mentioned for Paratía, Palca, Vila Vila, Lampa, and Cabanillas municipalities, which do not have final control over the water quality,

2.2. SEDIMENT SAMPLING AND ANALYSIS

Three sediment cores were collected at previously selected stations (COAT-1, COAT-2, and COAT-3) during low tide along the Coata River in March and April 2019 (Fig. 3). These sampling points were chosen on the basis of previous research (INRENA, 2007) which revealed anthropogenic geochemical anomalies in the river's surface waters. The sampling stations were georeferenced using the Global Positioning System (GPS). The COAT-1 core, approximately 24 cm long, was collected at 328872/8314894 (altitude: 3,288 m; near the Palca district). The COAT-2 core, approximately 27 cm long, was collected at 381330/8289939 (altitude: 3,813 m; near the Lampa/Cabanillas districts), and the COAT-3 core, approximately 33 cm long, was collected at 402797/8275181 (altitude: 3,821 m; near the

including enough capacity to handle the wastewater volume produced by urban activities, thus, reducing the capacity to host flora and fauna, affecting biomass (DEFENSORIA DEL PUEBLO, 2014; LIPA VILCA, 2019; BROUSETT-MINAYA *et al.*, 2021).

In addition to this scenario, mining activities have been also carried out in the study area (Fig. 1), which encompasses significant reserves of metals, whose exploitation has become profitable due to the metal prices rise on the worldwide market (OSIGNERMIN, 2016). They are chiefly realized by Arasi – SAC mining unit at Ocuwiri district (altitude of 5,200 m), which exploits the open-pit gold deposit, and El Cofre – CIEMSA mining unit at Paratía district (altitude of 4,400 m), which exploits silver as the most important resource, with a production capacity of 400 TMH/day. Thus, those mines contain a large variety of minerals containing enhanced levels of lead, zinc and gold, with both companies being situated at Lampa Province in Puno region (CABOS, 2009).

Juliaca district, towards the Lake Titicaca mouth).

PVC tubes (8 cm in diameter), which were previously decontaminated in a 10% nitric acid solution and washed with local water, were used for the sediment cores collection. In each site, the PVC tube was placed vertically into the sediment layer until reaching the selected depth. This was done using a small wooden plank resting on the upper edge of the tube in order to facilitate the application of the force necessary to cause the tube penetration. Once reached the required depth, the upper end of the tube was closed with an appropriate lid. The tube removal from sediment layer was done using rotating and upward movements, allowing the cores recovery without spilling at each monitoring station. The bottom of the tube was also closed with a second lid.

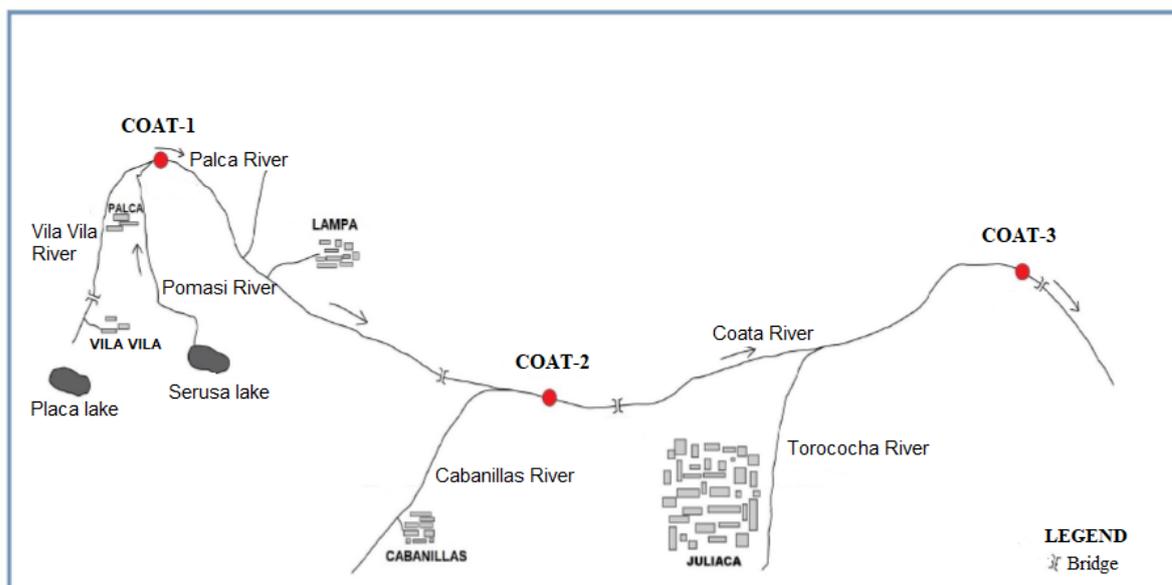


Figure 3 - Monitoring points and municipalities at Coata River, Puno Region, Peru.

Immediately after collection, the sediment-filled tubes were washed, dried with paper towels, duly identified, packaged, stored vertically inside Styrofoam boxes, kept refrigerated (at 4°C) and transported to LABIDRO - Isotopes and Hydrochemistry Laboratory (IGCE/UNESP-Rio Claro, São Paulo, Brazil). There, they were opened and sectioned at 3 cm intervals. Subsequently, the samples were placed in tightly sealed plastic bags and frozen for chemical analysis (8 segments for COAT-1 core, 9 segments for COAT-2 core, and 11 segments for COAT-3 core). All sediment samples were dried at 100°C in an oven and then crushed in a mill with agate pots.

The concentrations of Zn, Cu, and Co in the sediments were determined using the pressed pellet preparation procedure. This involved using approximately 8 g of the material to be analyzed (sediment) and 1.6 g of a binder (Cera Wax® from Merck with the chemical formula C₆H₈O₃N), which were weighed on an analytical balance. The mixtures prepared by adding the material and wax were homogenized for 20 minutes using a Turbula® mixer. After the mixing step, each sample was placed on a layer of boric acid (3.5 g) and compacted at 40

tons for 1 minute using a hydraulic press (Herzog HTP-40 model).

The concentration of Zn, Cu, and Co was determined using X-ray fluorescence spectrometry with a Bruker S8 Tiger instrument located at LARIN - Laboratory of Ionizing Radiation, UNESPetro - Center for Applied Natural Sciences - IGCE/UNESP/Rio Claro. The results of the analyses were expressed in micrograms of the element per gram of sediment (ppm). The readings were done after calibrating the equipment with standards consisting of cement, clay, feldspar, limestone, dolomite, sediment, sillimanite, magnesite, refractory, carbonatite, silica, bauxite, gypsum, superphosphate and five additional rock types (BRUKER, 2011). The concentration range of these metals in the standards corresponded to: Zn (0.5 – 3,000 ppm), Cu (0.8 – 1,000 ppm), and Co (0.7 – 2,500 ppm) (BRUKER, 2011). The analytical accuracy of these trace elements application was tested with the international certified reference material (CRM) consisting of granite MINTEK NIM-G. The data demonstrated excellent analytical performance of the S8 TIGER regarding accuracy and reliability as the absolute standard deviation based on 20 repetitions was 1.1% for Zn, 1.0% for Cu, and 1.8% for Co (BRUKER, 2011).

2.3. GEOCHEMICAL INDICES

The Geoaccumulation Index (I_{geo}) is a widely used geochemical index to quantitatively assess the contamination of marine sediments by heavy metals through the ratio of the present concentration to reference

values. The I_{geo} categorizes the degree of sediment contamination from non-contaminated to strongly contaminated. All of this is done through a scale established by Müller (1981), which assigns sediment classes

ranging from 0 to 7 based on the Igeo values and corresponding interpretation of the category, as

specified in Table 1. This index is calculated using the following formula:

$$I_{geo} = \log_2 [C_n / (1.5 \times C_b)] \quad (1)$$

Where: the factor 1.5 is applied to minimize variations in background values due to different environment characteristics. C_n is the concentration of the analyzed element, and C_b

is the reference geochemical concentration of the metal, generally adopted its mean concentration in the Earth's crust (background) as described by Turekian and Wedepohl (1961).

Table 1 - Degrees of contamination based on Igeo, according to Müller (1981).

Value	Class	Contamination degree
$I_{geo} < 0$	0	None contamination
$I_{geo} = 0 > \text{to} \leq 1$	1	None to moderate contamination
$I_{geo} = 1 > \text{to} \leq 2$	2	Moderate contamination
$I_{geo} = 2 > \text{to} \leq 3$	3	Moderate to strong contamination
$I_{geo} = 3 > \text{to} \leq 4$	4	Strong contamination
$I_{geo} = 4 > \text{to} \leq 5$	5	Strong to extreme contamination
$I_{geo} = 5 > \text{to} \leq 6$	6	Extreme contamination

The Enrichment Factor (EF) is another alternative index to assess metal contamination in marine sediments, which reduces bias caused by grain size and mineralogy using normalized data. According to Sutherland (2000), the EF can distinguish between the natural or

anthropogenic origin of metals present in the evaluated sediments, based on an assessment scale proposed by Hakanson (1980) as specified in Table 2. This is contrasted with the results after applying the following formula:

$$EF = \left[\left(\frac{Me}{Zr} \right)_{sample} / \left(\frac{Me}{Zr} \right)_{background} \right] \quad (2)$$

Where: $(Me/Zr)_{sample}$ is the ratio of the metal (Me) concentration relative to that of the chosen normalizing element in the sample and $(Me/Zr)_{background}$ represents the same ratio in the background (crust). In this study, zirconium (Zr) was used as normalizing element because it

has been considered a chemically immobile element, whose concentration has not changed over time, despite anthropogenic effects (Wang *et al.*, 2010). The background values described by Turekian and Wedepohl (1961) were used as reference values in this paper.

Table 2 - Contamination categories based on EF, according to Hakanson (1980).

Value	Class	Category
$EF < 1$	I	Background concentration
$EF = 1-3$	II	Low enrichment
$EF = 3-5$	III	Moderate enrichment
$EF = 5-10$	IV	Moderately high enrichment
$EF = 10-25$	V	High enrichment
$EF = 25-30$	VI	Very high enrichment
$EF > 50$	VII	Extremely high enrichment

3. RESULTS AND DISCUSSION

3.1. METAL CONCENTRATION

Results of the geochemical analysis are summarized in Table 3. The Zn concentration ranged from 524 to 619 ppm, much higher when

compared to Cu and Co concentration in the three monitoring stations at Coata River. In COAT-1, the metal concentration occurred in

the decreasing order Zn>Cu>Co in the top of sediment core (0-3 cm depth), but in the order Zn>Co>Cu in the remaining sediment core layers, with maximum values of 524, 46, and 36.7 ppm, respectively, for Zn, Cu and Co. For

COAT-2, the metal concentration was in the order Zn>Cu>Co, with maximum values of 639, 48, and 39 ppm, respectively. In COAT-3, the order was also Zn>Cu>Co, with maximum values of 619, 96, and 29 ppm, respectively.

Table 3 - Concentration (in ppm) for Zn, Cu and Co analyzed at COAT-1, COAT-2 and COAT-3 sediment cores from Coata River, Peru, and the sediment quality guidelines (in ppm) as proposed by Environment Canada (EC) and USEPA, as well as the Global average values (TUREKIAN; WEDEPOHL 1961).

Depth (cm)	COAT-1				COAT-2				COAT-3			
	Zn	Cu	Co	Zr ²	Zn	Cu	Co	Zr ²	Zn	Cu	Co	Zr ²
0-3	501	46	28.5	308	639	48	39.0	314	407	46	26.0	295
3-6	496	25	28.0	293	621	30	25.4	316	408	47	25.7	286
6-9	389	22	36.7	289	608	35	26.4	303	407	46	29.0	270
9-12	436	18	32.1	307	519	35	24.4	312	425	47	28.0	254
12-15	387	15	28.9	253	489	35	22.7	298	449	47	24.0	244
15-18	375	13	25.0	281	606	35	29.0	295	619	92	29.0	250
18-21	408	22	22.5	290	489	37	24.2	299	609	95	23.9	245
21-24	524	23	30.3	279	501	33	25.2	313	609	96	22.3	260
24-27					501	36	26.7	325	612	96	26.2	258
27-30									612	95	23.4	230
30-33									618	95	22.9	239
Mean	439.5	23	29.0	287	552.6	36	27.0	308	525	72.9	25.5	257
¹ SD	59.3	10.2	4.3	17.4	63.8	4.9	4.8	10.1	102	25.2	2.4	19.7
EC-ISQG ³	123	35.7										
EC-PEL ⁴	315	197										
USEPA/ER-L ⁵	150	34										
USEPA/ER-M ⁶	410	270										
Global average	95	45	19									

¹SD = standard deviation; ²Zr = normalizing element; ³ISQG = Interim Sediment Quality Guideline (TEL); ⁴PEL = Probable Effects Level; ⁵ER-L = Effects Range Low; ⁶ER-M = Effects Range Median

Table 3 also presents a comparison between the average values obtained in this study and three different international quality guidelines for sediment. This helps to identify the monitoring stations possibly contaminated by anthropogenic inputs, thus, affecting the aquatic ecosystem of Coata River. The following Interim Sediment Quality Guidelines (ISQG) guidelines of the Canadian Environmental Agency (CCME, 2002) are considered: (a) the concentrations below which there are unlikely to be any adverse biological effects (TEL, Threshold Effects Levels); (b) the concentrations above which adverse effects are expected to frequently occur (PEL, Probable Effects Level). Secondly, the following guidelines established by USEPA (2000) and described by MacDonald *et al.* (2000) and Long *et al.* (2006) are considered: lower limits, ER-L (Effect Range Low) and upper limits, ER-M

(Effects Range Median). In this case, three concentration ranges are distinguished for adverse effects: (a) rarely observed (concentration < ER-L); (b) occasionally observed (concentration between ER-L and ER-M); (c) frequently observed (concentration > ER-M). Lastly, the concentrations in sedimentary rocks (shales) taken as global averages as reported by Turekian and Wedepohl (1961) are also considered for comparison purposes.

The metals concentration as well as those for ISQG/PEL-Environment Canada, are shown in Fig. 4. Zn in all three sediment cores presented concentrations higher than ISQG (123 ppm) and PEL (315 ppm). Cu exhibited concentrations higher than ISQG (35.7 ppm) in the COAT-2 and COAT-3 cores and lower than PEL (197 ppm) in all cores.

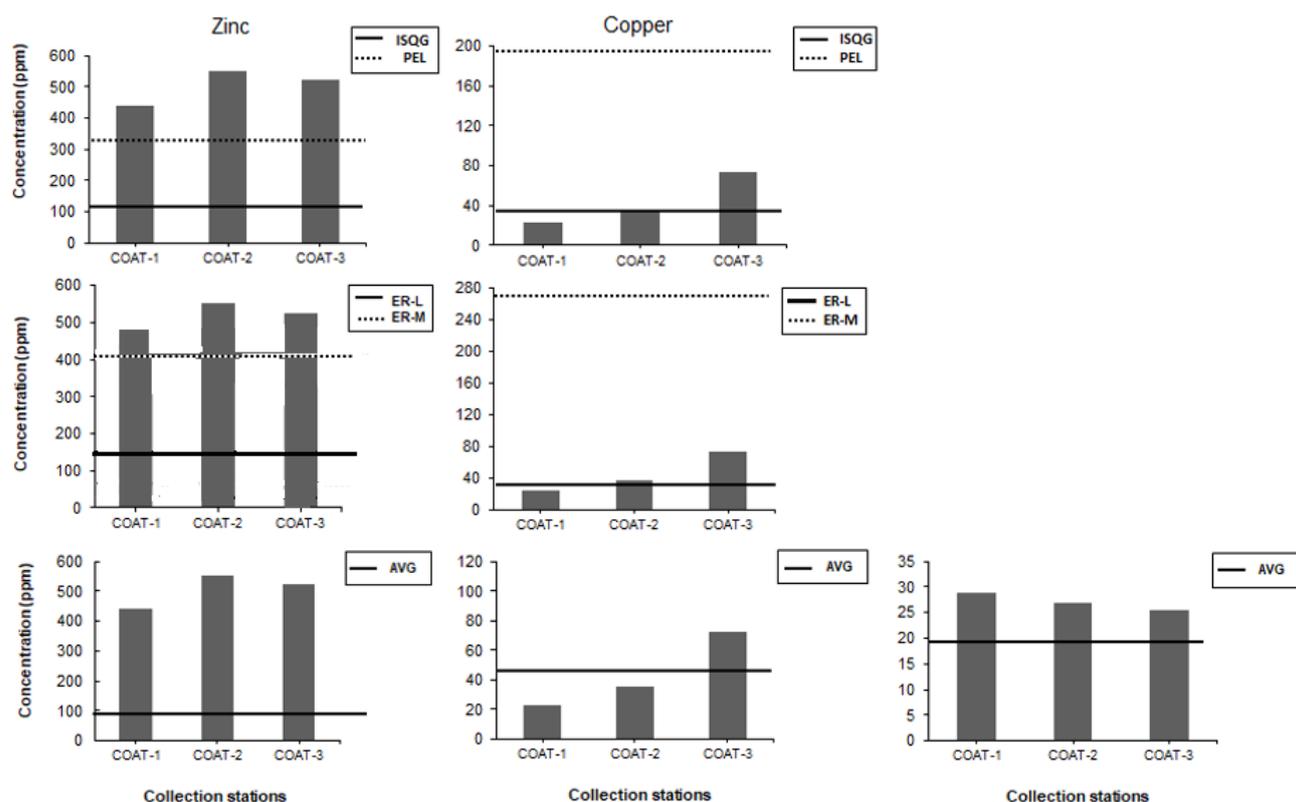


Figure 4 - Comparison of the concentrations of metals Zn, Cu and Co on COAT-1, COAT-2 and COAT-3 sediment cores collected at Coata River with the international sediment quality values: ISQG/PEL = proposed by the Canadian Environmental Agency (CCME, 2002); ER-L/ER-M = proposed by USEPA (LONG *et al.*, 2006); Global average values, AVG (TUREKIAN; WEDEPOHL, 1961).

In terms of the U.S. Environmental Protection Agency (USEPA) guidelines, Zn in the sediments exhibited concentrations higher than ER-L (150 ppm) and ER-M (410 ppm) in all three cores. Cu presented concentrations lower than ER-L (34 ppm) in the COAT-1 core, but not in COAT-2 and COAT-3 cores. On the other hand, Cu exhibited concentrations lower than ER-M (270 ppm) in all three cores. The Environment Canada and USEPA did not present guideline reference values for Co (Fig. 4).

Another criterion used to assess contamination by heavy metals in sediment is the comparison of sediment values with normal background values (AVG), as illustrated in Fig. 4. For Zn and Co, the concentrations in the sediments were higher than AVG_{Zn} (95 ppm) and AVG_{Co} (19 ppm) in all three cores. However, the Cu concentrations were lower than AVG_{Cu} (45 ppm) on COAT-1 and COAT-2 cores, but not on COAT-3 core.

In addition to mining activities, currently, the main sources of heavy metals introduction into sediments occurring at aquatic systems are the industrial wastewaters (RODRÍGUEZ,

2005; ALVAREZ-IGLESIAS *et al.*, 2007; DORIA; GÓMEZ, 2019). The metals Cd, Cu, Ni, Sb and Zn are associated with microelectronic items; Al, Sn, Ti and Zn are associated with textile products; Hg, Co, Cr and Cd are associated with plastic products and refinery products contain Cr, Ni and Pb; Ca and Mg are often associated with agricultural correctives, while As, Cu, Mn, Zn, P, U, V and Zn with fertilizers; Cd, Pb and Se may also be related with irrigation water and the metals Cd, Cu, Cr, Ni, Hg, Pb and Zn with urban effluents (BRADL, 2005). According to Zhang *et al.* (2007), atmospheric metal sources are increasingly being investigated as there is a strong link between the level of industrial growth in specific areas and the metal composition of atmospheric particulate matter. The high degree of industrialization and urbanization creates an ongoing risk of heavy metal contamination.

In this study, Zn in all sediment cores of Coata River exhibited concentrations exceeding the ISQG, PEL, ER-L and ER-M guidelines, as well as the global average values for shales (Fig. 4), indicating the possibility of exerting toxic

effects on the aquatic biota. One probable source of Zn entering into the aquatic environment may be the discharge of urban industrial wastes and untreated domestic sewage from the districts of Vila Vila, Palca, Lampa, Cabanillas, and, chiefly, from Juliaca city. According to Baptista Neto *et al.* (2006), zinc is typically linked to solid urban wastes, industrial wastes and residential sewage of urban areas.

Additionally, another potential source of Zn inputs into Coata River could be the residual discharges from Arasi and El Cofre mining areas. According to OEFA (2015), mining activities are responsible for most of the contamination of water resources by heavy metals and their migration produces harmful effects on aquatic organisms, which tend to accumulate these metals. El Cofre mining area possesses a mineral processing plant and a tailings deposit pond with a capacity of about 30,000 m³. According to the “Observatorio de Conflictos Mineros del Perú”, in 2007, part of the retaining wall of the Tailings Lagoon (2 m wide by 3 m high) collapsed, discharging approximately 15,000 m³. In 2013, there was a new episode of waste spillage with toxic wastes and heavy metals travelling 500 m to reach the Paratía River, afterwards Unocolla River, and latter Cabanillas River that flows into Coata River. ANA (2012) recorded Zn concentrations of up to 0.15 mg/L in Palca Lagoon that exceeded the ECA Peruvian guideline value, which may also have a contribution of the Arasi mining activities (Fig. 1).

The highest Cu concentration in the sediment cores was found at COAT-3 that exceeded the ISQG/ER-L guidelines and global average value for shales, but within the PEL and ER-M limits, suggesting some occasional adverse effects on benthic communities. High Cu concentrations in sediments have been often considered indicator of human interference associated to the release of urban and industrial sewage and soil occupation around rivers (SISINNO, 2012). The domestic and industrial contributions from Juliaca city through Torococha River could justify the high Cu levels found at COAT-3 as this monitoring point is located ~20 km from that municipality.

Regarding Co concentration in the sediment cores, the global average value for shales (19 mg/kg) was surpassed in all monitoring points stations at Coata River, in which the mean levels of 29.0, 27.0 and 25.5 mg/kg for cores COAT-1, COAT-2 and COAT-3, respectively, were higher than the following values reported

in the literature: 1.82 mg/kg at Cuchivero River, Venezuela (MÁRQUEZ *et al.*, 2016); 8.6 mg/kg at Ria de Aveiro Lagoon, Portugal (MARTINS *et al.*, 2017); 7.56 mg/kg at Bahia dos Santos, Brazil (NETTO *et al.*, 2022); 12.78 mg/kg at Qinjiang River, China (ZHANG *et al.*, 2022); 10.30 mg/kg at Bartın River, Turkey (GUNES, 2022); 7.40 mg/kg at Port Everglades, USA (GIARIKOS *et al.*, 2023); 1.2 mg/kg at Narmada River, India (RAHI *et al.*, 2024); 5.06 mg/kg at Korotoa River, Bangladesh (TOUFIQ HASSAN *et al.*, 2024). Despite these ecosystems belong to different biome and even continents of that studied in this paper, all they are river/lake systems in which human activities also take place as reported by Silva de Oliveira *et al.* (2018).

On the other hand, Siqueira *et al.* (2006) pointed out that the Co presence in the environment is geogenic and unaffected by contaminating sources. Alves and Della Rosa (2003) stated that cobalt occurs as a result of the combustion of fossil fuels, the use of bio-silicones and phosphate fertilizers, ore mining and smelting, and metal-containing industrial processes. Although cobalt is considered one important trace metal needed by organisms, excessive levels of this element can be harmful (WHO, 2011). It is also relevant to mention that the aquatic environment under study receives discharges from several districts and mainly from Juliaca city, which may be contributing to inputs of this metal into Coata River.

The concentrations of Zn, Cu, and Co obtained here can be also compared with the results of other studies. In Africa, Bawa *et al.* (2018) reported concentrations of Zn = 5.69 ppm and Cu = 21.2 ppm in Lagos, Nigeria. In Europe, Cieslewicz *et al.* (2018) pointed out concentrations of Zn = 278 ppm and Cu = 31.7 ppm in protected areas of Poland. In Asia, Yin *et al.* (2018) reported concentrations of Zn = 91.5 ppm and Cu = 9.95 ppm at Lake Chaohua, China, while El-Safa *et al.* (2022) related the values of Zn = 46.4 ppm, Cu = 12.3 ppm, and Co = 21.6 ppm at Gamasa estuary, Egypt. All these authors found lower concentrations than those measured at Coata River, with the greatest difference being verified for Cu on COAT-1 core.

Regarding South America, studies related to metal analysis in sediments have been conducted in Brazil by Capaleto *et al.* (2018) at Guaíba Lake, Rio Grande do Sul (Zn = 21.03 ppm and Cu = 4.17 ppm), and by Netto *et al.* (2022) at Santos Bay (Zn = 58.4 ppm, Cu = 11.14 ppm, and Co = 4.5 ppm). Also, in

Venezuela, Fuentes-Hernández *et al.* (2019) reported concentrations of Zn = 75 ppm and Cu = 58 ppm at Cariaco Gulf. Thus, generally speaking, these concentrations are well below the levels found at Coata River. Additionally, considering the study area focused in this paper, Moreno *et al.* (2018) analyzed superficial sediments from 6 sampling stations in the inner Puno bay, Lake Titicaca, reporting the

following results: Cu (6.49 mg/kg), Zn (8.20 mg/kg), Pb (0.04 mg/kg), Cd (0.04 mg/kg), As (<0.01 mg/kg), and Hg (<0.01 mg/kg). Such data are also well below the guideline reference values and concentrations measured in cores COAT-1, COAT-2 and COAT-3 for Cu and Zn as reported in Table 3, again suggesting influence of anthropogenic inputs into Coata River.

3.2. Igeo AND EF GEOCHEMICAL INDICES

To estimate Igeo, values of metal concentrations of interest and regional geochemical background are required, as

indicated in Eq. 1 (section 2.3). In this work, the geochemical global mean values proposed by Turekian and Wedepohl (1961) were adopted.

Table 4 - Geoaccumulation index (Igeo), degree of contamination, and enrichment factor (EF) of Zn, Cu and Co in COAT-1, COAT-2 and COAT-3 sediment cores from Coata River, Puno region, Peru.

Sediment core	Value	Concentration (ppm)			Igeo value			Igeo class			EF value		
		Zn	Cu	Co	Zn	Cu	Co	Zn	Cu	Co	Zn	Cu	Co
COAT-1	Minimum	375	13.0	22.5	1.40	-2.38	-0.34	2	0	0	2.2	0.2	0.7
	Maximum	524	46.0	36.7	1.89	-0.56	0.37	2	0	1	3.2	0.5	1.1
	Mean	439.5	23.0	29.0	1.62	-1.66	0.01	2	0	1	2.6	0.3	0.9
	SD	59.3	10.2	4.3	0.19	0.55	0.22	1	1	1	0.3	0.1	0.1
COAT-2	Minimum	489	30.0	22.7	1.78	-1.17	-0.33	2	0	0	2.6	0.3	0.6
	Maximum	639	48.0	39.0	2.17	-0.49	0.45	3	0	1	3.5	0.5	1.0
	Mean	552.6	36.0	27.0	1.95	-0.92	-0.10	2	0	0	3.0	0.4	0.7
	SD	63.8	4.9	4.8	0.17	0.18	0.23	1	1	1	0.4	0.1	0.1
COAT-3	Minimum	407	46.0	22.3	1.52	-0.56	-0.36	2	0	0	2.3	0.6	0.7
	Maximum	619	96.0	29.0	2.13	0.51	0.03	3	1	1	4.5	1.5	1.0
	Mean	525.0	72.9	25.5	1.86	0.02	-0.17	2	1	0	3.5	1.0	0.8
	SD	102.0	25.2	2.4	0.29	0.54	0.14	1	1	1	0.8	0.4	0.1
Global average		95	45	19									

The Igeo values and related contamination class in the Coata River sediments are shown in Table 4. For Zn, Fig. 5 shows that on COAT-1 core, an Igeo = 2 (moderate contamination) was found, while on COAT-2 and COAT-3 cores, an Igeo = 3 (moderate to high contamination) was observed. Cu presented an Igeo = 0 (none contamination) on cores COAT-1 and COAT-2, whilst Igeo = 1 was found on COAT-3 core, suggesting “none contamination to moderate contamination” for this metal. For Co, all three cores had Igeo = 1, indicating “none contamination to moderate contamination” for this metal (MÜLLER, 1981) (Fig. 5). In summary, based on the measured concentrations and Igeo index, the sediments collected at stations COAT-1, COAT-2, and COAT-3 of Coata River are in the categories of “moderate to high contamination” due to the presence of Zn, while for Cu and Co, they are in the category of “none contamination to moderate contamination”.

The Zn values on COAT-1, COAT-2, and

COAT-3 stations, with mean Igeo values between 1.62 and 1.95, exceeded those found by Espirilla and Gómez (2022) at Arequipa city, Peru, in which Igeo values were between -1.18 and -1.13, despite there is a much larger population in that city, reaching one million inhabitants. In Kerala, Southwest Coast of India, Igeo values were between -8.37 and -4.79 (VINEETHKUMAR *et al.*, 2021). In the Mediterranean Coast of Egypt, Igeo was -0.71 (EL-SAFA *et al.*, 2022) and in the Euphrates River, Iraq, the Igeo values were between 0.0019 and 0.0023 (KAMEL *et al.*, 2023). However, the Igeo values for Zn reported here are lower than 3.2 as reported by Cáceres *et al.* (2013) at Lake Titicaca, Puno, 3.04 as found by García-Martínez and Poletto (2014) at Porto Alegre, Brazil, and 2.59 as given by Dalu *et al.* (2023) at South Africa. On the other hand, they are similar to 1.63 as recorded at Çanakkale city, Turkey (ÇAVUŞ *et al.*, 2023), whose population is much smaller than that of our study area.

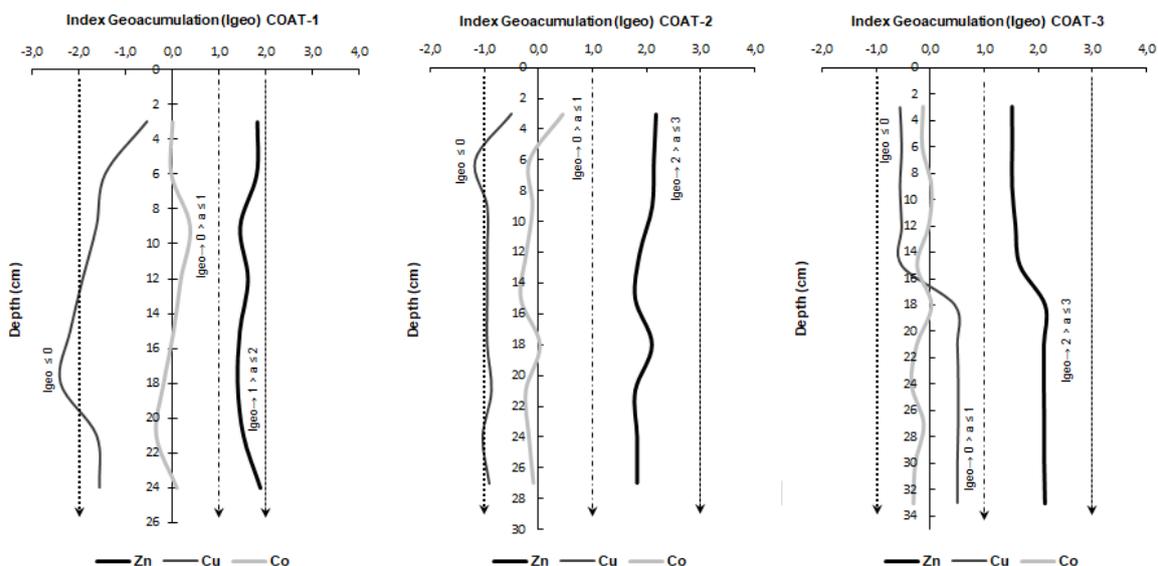


Figure 5 - Geoaccumulation Index (Igeo) for Zn, Cu and Co in the sediment cores COAT-1, COAT-2 and COAT-3 sampled at Coata river, Puno region, Peru.

The mean Igeo values for Cu (between -1.66 and 0.02) and Co (between -0.17 and 0.01) are below those described by Trujillo-González and Torres-Mora (2015) (Igeo for Cu = 2.6 and 0.9) and Nour et al. (2022) (for Cu and Co, Igeo values of 2.3 and 4.88, respectively) in southern Kuwait.

The metal concentrations (Zn, Cu, and Co) were normalized with zirconium (adopted as the normalizing element) for realizing the calculation of the enrichment factor (Eq. 2, section 2.3). Table 4 shows the minimum, mean, and maximum EF values found for each metal.

EF values ranged from 2.2 to 4.5 for Zn (mean of COAT-1 core = 2.6 ± 0.3), from 0.2 to

1.5 for Cu (mean of COAT core = 0.3 ± 0.1), and from 0.6 to 1.1 for Co (mean of COAT-1 core = 0.9 ± 0.1). According to the calculated values, Zn in the three cores had an average EF = 3.7 (moderate enrichment), suggesting possible contamination of anthropogenic origin. Cu on COAT-1 and COAT-2 cores had a mean EF = 0.5 (none enrichment), suggesting it is geogenic (indicating none contamination), whereas on COAT-3 core, Cu exhibited EF = 1.5 (low enrichment), possibly indicating anthropogenic contamination for this metal. For Co, the mean EF was 1.0 (low enrichment) in all cores, suggesting it is anthropogenic, thus, indicating some contamination of this metal in them (HAKANSON, 1980) (Fig. 6).

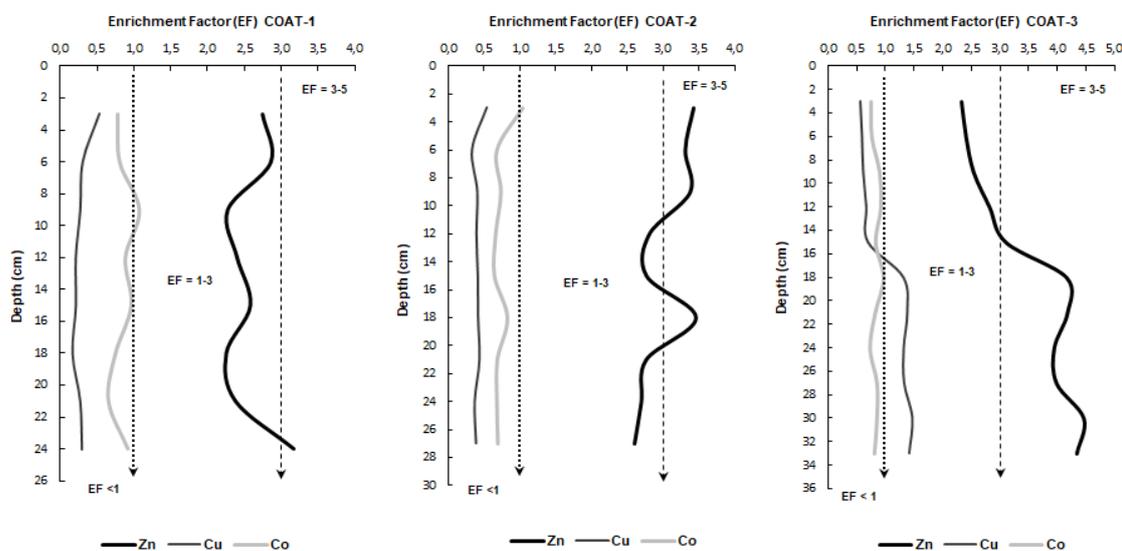


Figure 6 - Enrichment Factor (EF) for Zn, Cu and Co in the in the sediment cores COAT-1, COAT-2 and COAT-3 sampled at Coata river, Puno region, Peru.

According to Zhang and Liu (2002) and Feng *et al.* (2004), EF values between 0.5 and 1.5 suggest that metals may come from the Earth's crust or natural weathering processes, while an EF greater than 1.5 indicates that some materials can be introduced by non-natural processes, i.e. from anthropogenic sources. Under this view, in general, the EF values of Zn at all stations are above 1.5 (Table 4), indicating that its contamination may be due to anthropic inputs. However, the EF values below 1.5 for Cu and Co suggest that their concentration is geogenic.

EF values almost similar to those found in this study were reported at Venezuela (EF values of Zn = 0.97 and Cu = 0.22; MARÍN-LEAL *et al.*, 2022), and Tunis lagoon, NE Tunisia (EF values of Zn = 0.60-0.81 and Cu = 1.55-2.03; ABIDI *et al.*, 2022), whereas higher EF values were obtained at Lake Titicaca, Peru (EF value of Zn = 162.4; CÁCERES *et al.*, 2013), Aguada Blanca reservoir, Arequipa, Peru (EF values of Zn = 3.46 and Cu = 3.71; ESPIRILLA; GÓMEZ, 2022), Ria de Aveiro lagoon, Portugal (EF values of Zn = 13.2, Cu = 114.3, and Co = 2.8; MARTINS *et al.*, 2017), and Lake Badovci, Kosovo (EF values of Zn = 6.95 and Cu = 8.25; MALSIU *et al.*, 2022). Cáceres *et al.* (2013) and Espirilla and Gómez (2022) pointed out that such Zn enrichment as evidenced by the high Igeo and EF values in the sediments was due to mining activities held in their studied sites.

As mentioned earlier, COAT-3 sediments were identified as the most contaminated by Zn, Cu, and Co (highest EF values, EF > 1), possibly due to anthropogenic inputs. COAT-3 is located near Juliaca district, considered the main emitter of significant amounts of raw urban sewage and industrial wastes that are discharged into Torococha River, which drains a broad urban and industrial region (HUACANI, 2013). Maranhão *et al.* (2015) reported that sewage is known to release a wide variety of xenobiotics into the environment, including personal care products, pharmaceuticals, surfactants, PAHs, metals, among others. Godoy *et al.* (1997) analyzed metals in sediments from Guanabara Bay, Brazil, concluding that the contamination was caused by increased industrialization, also confirmed by other researchers elsewhere. For instance, Quevedo-Álvarez *et al.* (2019) pointed out that in Cuba, urban sewage and industrial wastewater are considered the main pollution source in marine environments, in which the

principal observed effect is the physical alteration of sediment texture close to the discharge site due to the fine sediment inputs.

COAT-1 and COAT-2 sites are located near urban areas (Vila Vila, Palca, Lampa, and Cabanillas) and mining zones (Arasi and El Cofre), which are potential sites for contaminants inputs into Coata River such as urban sewage, mining wastes, atmospheric emissions, and metal-containing effluents. COAT-1 and COAT-2 occupied the 2nd and 3rd position for the metal releases into the sediments, respectively, showing EF values greater than 1 for Zn and Co on COAT-1 core that indicate they may be of anthropic origin. They were classified as moderately contaminated according to the Igeo contamination class (Fig. 5 and Table 4). DRSP (2014) and OEFA (2015) pointed out that the Arasi and El Cofre mining companies are responsible for some environmental damage in the studied region, generating groundwater infiltration that affects the water table, causing the introduction of high levels of acids into the waterbodies and soils.

Furthermore, the EF and Igeo values showed contamination associated with Zn in all monitoring stations. The main sources associated with these inputs in the study area are anthropogenic since Coata River flows through highly urbanized areas, more specifically at Juliaca city, where it receives spills of fossil fuels and various types of untreated effluents. In Brazil, Abessa *et al.* (2014) reported that urban sewage is considered the main pollution source in aquatic environments.

Therefore, Igeo generally demonstrated that Zn, Cu, and Co may be responsible for the contamination of Coata River. The concentration order Zn>Cu>Co was also recorded by Cáceres *et al.* (2013) in Lake Titicaca sediments with Zn concentrations ranging from 14.9 to 940.9 ppm, Igeo = 3.2, and EF = 162.4, which are higher than those found at Coata River. These authors pointed out that such Zn enrichment in sediments was due to mining activities in that region, as the lake receives effluents from all rivers flowing into it. More detailed investigation of local geology and sediment sources variability along the depth cores could better explain the high Zn levels found at Coata River, constituting an important topic for future studies, which could better delineate whose anthropogenic influence is more relevant, i.e. if mining activities or wastewater/effluents release from urban areas.

4. CONCLUSIONS

In continental aquatic systems (rivers, lakes, reservoirs) heavy metals are present as a result of the action of natural and anthropogenic processes. This is how heavy metals are classified as natural or anthropogenic, although, in practice, it is difficult to distinguish between industrial, residential, or natural contamination from that caused by mining activities. Thus, determining the origins of metal contamination in aquatic environments is a very challenging task. Previous studies in the literature have already focused the sediments of Coata River, Peru, suggesting that the presence of the metals studied here (Zn, Cu and Co) are affected by anthropogenic influences due to mining activities or discharge of various types of untreated effluents from urban areas. In this paper, the Zn, Cu, and Co concentration was measured in three sediment cores (COAT-1, COAT-2, and COAT-3) collected at Coata River, Puno region, Peru, and compared with the reference guideline values established by CCME (Canada legislation) and USEPA (USA legislation). The comparison indicated that the metals exhibited concentrations surpassing the PEL limiting levels in all of them. Zinc was considered the most relevant contaminant because its concentration exceeded the maximum ecological risk level (ER-M) in all cores, while copper at least on COAT-3 monitoring point. Furthermore, the Zn and Co

content in the sediments exceeded the corresponding average concentration in global shales, whilst only Cu on COAT-3 core showed values above of the shales guidance level. These findings suggest possible anthropogenic influence in the studied stations at Coata River. The calculated Geoaccumulation Index (Igeo) indicated that the analyzed in the sediment cores can be classified as follows: moderately to highly contaminated due to the Zn presence on all cores; uncontaminated on COAT-1 and COAT-2 cores and moderately contaminated on COAT-3 due to the Cu presence; and moderate contamination in all cores due to the Co presence. The relative proportion of the Igeo values followed the order $Zn > Co > Cu$. Regarding the Enrichment Factor (EF) values in the sediments, they indicated the following: moderate enrichment for Zn in all cores; for Cu, none enrichment on COAT-1 and COAT-2 cores and low enrichment on COAT-3; low enrichment in Co for all cores. The relative proportion of the EF values followed the order $Zn > Co > Cu$ that is the same trend found for the Igeo values. Therefore, the sediments of Coata River have high concentrations of Zn, particularly in more urbanized areas, suggesting a possible anthropic origin, thus, confirming the results of previous investigations, and requiring the development of further studies for better characterizing its major sources.

5. ACKNOWLEDGMENTS

The authors are grateful to the technical staff of LARIN-UNESPetro-IGCE-UNESP for access to the available infrastructure and analytical support provided for performing this

investigation. One anonymous reviewer is greatly thanked for helpful comments that improved the readability of the manuscript.

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