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Study of Heavy Metal Pollution in Arid and Semi-Arid Regions Due to Mining Activity: Sonora and Bacanuchi Rivers

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Abstract

A study of the Sonora and Bacanuchi rivers was conducted to assess the mobility and bioavailability of heavy metals. These rivers were affected by a spill caused by the mining activity of the region, which is considered the most important ecological disaster in the modern history of Mexico. BCR sequential extraction was performed to determine geochemical phases in which metals are found. The evaluation of heavy metal contamination was performed using Enrichment Factor (EF) and Geoaccumulation index (Igeo). Sediments showed high concentrations (mg/kg) of Cu (8-716), Cr (8-90), Fe (7,300-52,400), Mn (80-938), Ni (6-48), Pb (14-210) and Zn (41-470). Metal concentrations in geochemical phases exhibited the following order: residual>Fe/Mn oxides> exchangeable>organic matter. The order of mobility and/or bioavailability of metals was: Mn>Cu>Ni>Pb>Zn>Fe>Cr. EF showed an anthropogenic enrichment in both rivers for Cu, Cr, Mn, Ni, Pb and Zn, mainly derived from the mining activity. Values of Igeo were classified as non-contaminated to moderately contaminated. The Bacanuchi river showed moderate to strong contamination of Cu and Pb. The quality criteria comparisons (LEL and SEL) indicate both rivers are contaminated by metals and represent a danger to biota, due to the high metal mobility and bioavailability.

Keywords: Metals; Bioavailability; Sediments; Bacanuchi and Sonora rivers

Introduction

Sediments are the result of the deposition of particles that bring along material of rocks and minerals, heavy metals, organic matter, among others. Sediment deposition occurs by entrainment of both organic and inorganic particles [1]. Sediments play an important role in transportation of nutrients, metals and other pollutants through river systems to the oceans and seas of the world [2]. Contamination of sediments by heavy metals can occur naturally or anthropogenically [3]. The most important anthropogenic activities are the agricultural, industrial, manufacturing, and mining activities. Heavy metals are among the most common pollutants and their ecological impact is due to their toxicity, high persistence and non-degradability in the environment [4,5]. Sediments concentrate metals from aquatic systems and represent a suitable and strategic means for conducting pollution monitoring studies [6].

In sediment, metals can be found in different chemical species, and depending on the species in which it is found, bioavailability, mobility and its toxicity can be determined. The determination of the total metal content can be useful for sediment characterization; However, it does not provide enough information on the bioavailability or toxicity of metals [7,8]. Sequential extraction is a technique that is widely used for the determination of chemical speciation and possible associations between metals and sediment components [9]. The sequential extraction method proposed by the Community Bureu of Reference (BCR) classifies metals into three fractions: fraction I exchangeable and acid soluble fractions; fraction II reducible fraction or Fe and Mn oxide-associated fraction; fraction III oxidizable fraction or fraction bound to organic matter [10]. If the metal corresponds to the first geochemical fractions, it will be more bioavailable, that is, it will be more available to participate in metabolic reactions of living beings and, therefore, it will be potentially more toxic or bioaccumulable, depending on the type of metal and its concentration.

In order to estimate the there is an environmental impact due to metals and the level of contamination; there are parameters such as the enrichment factor (EF) and the Geoaccumulation index (Igeo) [5]. EF establishes whether the concentration of a metal comes from a natural or an anthropogenic source. Igeo determines the extent of metal pollution in sediments, considering the concentration of that metal in baseline samples [11]. In Mexico, the mining industry is a productive sector which contributes economically, historically and culturally, and it will continue to be one of the pillars in the development and growth of the country. However, there is a strong environmental impact attributed to the mining activity, especially when accidents occur, or when improper management of the process or damages occur in any stage of the mining cycle.

In Mexico, there are documented accidents, particularly in the Sonora region, where spills of acid solutions containing metals, have been dumped and have caused a negative impact to the environment [12-14]. In August 2014, a 40,000 m^3 spill of acid waste, with high concentration of copper sulfate and low pH values, was reported from a mining dam located in the region of Cananea, Sonora affecting the Bacanuchi river stream, which is tributary of the Sonora river. The main metal contaminants contained in the spilled solution were: copper, aluminum, cadmium, chromium, iron, manganese and lead, which levels were above Mexican standards [15]. This spill is considered the most important ecological disaster in modern history of Mexico, and to our knowledge, there is no previously published information regarding the fate of such pollutants in the Sonora river system.

Based on the above, a study was carried out in the Bacanuchi and Sonora rivers, which objectives are: a) to estimate the origin and level of contamination by heavy metals (Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn) in the sediments, based on EF and Igeo; b) to estimate the distribution of heavy metals in the different geochemical fractions of the sediments; c) to study the chemical behavior (re-mobilization) of the heavy metals (Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn) in the geochemical fractions of sediments that may be available to biota and that may endanger human health, and c) to contribute to the study of aquatic ecosystems contaminated by mining activities in arid and semiarid regions in Mexico.

Methodology

Reagents and Apparatus

All reagents used in this research were analytical grade (Baker, Mexico). Deionized water (DI) was used in all the tests. All the glassware and equipment used to collect samples were prepared by soaking them in a 20 % (v/v) HNO3 solution for 3 days and then rinsed with DI water to reduce contamination risk by heavy metals. Hydrogen potential (pH) was determined using a portable pHmeter Thermo Scientific Orion 3-star benchtop. The measurement of heavy metal concentrations was determined using a Perkin Elmer atomic absorption Analyst 400 equipment. The wavelengths (nm) used for the analyses are: Cd 228.8, Cu 324.8, Cr 357.9, Fe 248.3, Mn 279.5, Ni 232.0, Pb 283.3 and Zn 213.9, respectively.

Study Area

The study area is located within the Cananea municipality, northeast of the state of Sonora, Mexico (Figure 1). In this area, one of the world's largest copper deposit is located, with an estimated production of 1.4×10^9 kg Cu, and smaller concentrations of Ag, Au, Pb, and Zn [16]. These mineral deposits can be found predominantly as pyrite, chalcopyrite, bornite, and, in a lesser extent, as galena. The total area of the mine is about 12 to 16 km^2, and the waste dumps and heap leach pads occupy an area of 1,000 hectares, which have been originated throughout the life of the mine. Arroyo Las Tinajas has its origin near Represo Tinajas 1, located inside the mining facilities. The path of the Tinajas stream flows to the south where it joins the Bacanuchi river. Then, the Bacanuchi river joins the Sonora river near the town of Arizpe, Sonora. Subsequently, the Sonora river flows into Presa El Molinito, near the city of Hermosillo, Sonora.

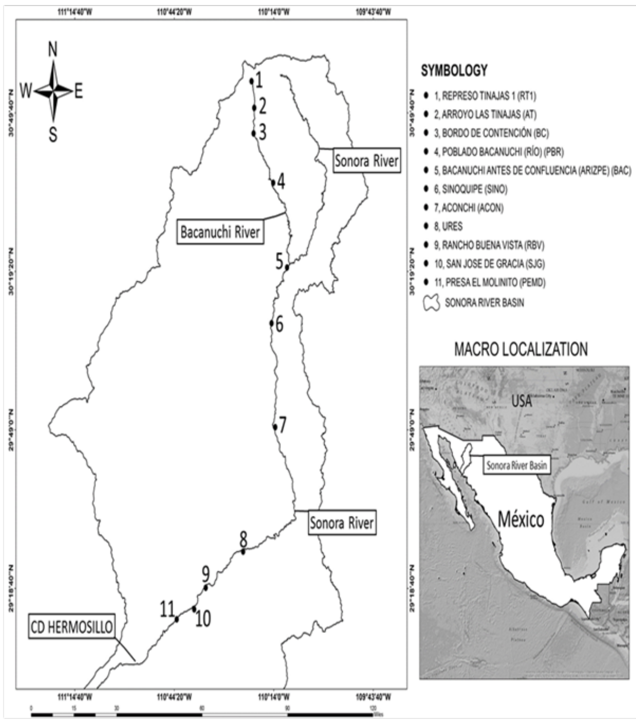

Figure 1: Location of the study area and distribution of sampling stations in Sonora and Bacanuchi rivers.

The predominant climate in the region is tempered, semi-arid climate, with a maximum, minimum, and average annual temperature of 25, 10, and 18°C, respectively. Some researchers have reported environmental pollution associated with abnormal concentrations of metals in the Bacanuchi and Sonora rivers [17]. The rivers of Bacanuchi and Sonora are a vital source of water for most economical activities of the region, i.e., agricultural and livestock activities, besides being a water supply for nearby localities, such as Hermosillo, capital of the state of Sonora [13,17].

Sample Collection and Analysis

A field sampling campaign was conducted in October 2014, on the Bacanuchi and Sonora rivers. Sediment samples were collected from a total of 11 sampling stations (Figure 1). Samples were taken at three different points within the same sampling station, to make up a composite sample. One of the points was taken at the center of the river and the other two were taken at

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each riverside, at a depth of 10 cm, using a polyethylene nucleator. Once collected, samples were put on ice and then transported to the laboratory for their further preparation and analysis. After collection, a reduction in volume was made using the cone and quartet method [18], then, they were grounded to a size smaller than 100 meshes (0.149μm) using a porcelain mortar. Hydrogen potential (pH) was determined using a Thermo Scientific Orion 3-star bench-top pH meter [18]. Sulfates were determined using the $BaCl_2$ precipitation method according to Method 980.02 [19]. Sediment texture was also determined [20]. Sediment samples were divided into four fractions: clay (<0.004 mm), lime (0.063-0.004 mm), sand (2-0.063 mm), and gravel (> 2 mm).

Analysis of Total Heavy Metals and Sequential Extraction

The sediments were totally digested with an acid mixture (HNO_3-HF-$HClO_4$) in a Teflon vessel. Residues were then dissolved with HNO_3 and boric acid (2%) and diluted with deionized water to a volume of 100 mL. Total concentrations of Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn were analyzed by atomic absorption spectroscopy. A sequential extraction procedure was performed following recommendations of the Community Bureu of Reference (BCR) [10]. Heavy metals were divided into three fractions according to the sequential extraction method: Fraction I (exchangeable and acid soluble fraction): sediment samples were extracted with acetic acid 0.11 mol L^{-1} for 16 h. Fraction II (Fe/Mn oxides): The residue from the previous extraction is then extracted by hydroxylamine hydrochloride 0.5 mol L^{-1} for 16 h. Fraction III (oxidable fraction or bound to organic matter): The residue is then oxidized with H_2O_2 8.8 mol L^{-1} and then, it is extracted with ammonium acetate 1.0 mol L^{-1}. Residual fraction. The residual metal concentrations were determined by digestion with aqua regia. After each extraction, samples were centrifuged at 3,000 g during 20 min, at room temperature. Each extract was separated and retained in a stoppered polyethylene container for analysis. The determination of the metal concentrations was carried out using a Perkin Elmer atomic absorption equipment Analyst 400.

Quality Control

Sediment samples were analyzed by duplicate. Certified reference standard NIST 2702 (Inorganic Marine Sediment) were analyzed by triplicate and were treated similarly to sediment samples, to reduce matrix interferences and to validate accuracy and precision. Blanks were analyzed by triplicate. Five standards were used to get the calibration curve for each metal in the atomic absorption analyses, and high correlation coefficients (r > 0.9990) were obtained. Metal concentrations in the reference standard NIST were within 91-105% of the reference concentrations, which is acceptable.

Determination of Enrichment Factor and Geoacumulation Index

The enrichment factor (EF) is used to evaluate whether a sediment is enriched naturally or anthropogenically and, it has been commonly used to infer anthropogenic influences [6]. The EF method normalizes the concentrations of heavy metals with respect to a reference metal, the most common being Al and Fe [21]. Some authors propose that normalization with Fe is more adequate because of its relatively high natural concentration in the Earth's crust, since its distribution is not usually associated with the appearance of other metals [22,23]. For this reason, in the present study, Fe was used as a normalizing element. EF can be calculated using the following formula [5,24].

$$EF = (M_x Fe_b)/(M_b Fe_x)$$

Where Mx and Fex are the concentrations of the metal and Fe in the sample; while Mb and Feb are the metal and Fe concentrations in a baseline sample. Igeo has been widely used to calculate levels of heavy metal contamination in sediments [25]. In the present study, the Igeo was calculated using the following equation [26].

$$I_{geo} = \log_2 [(C_n)/(1.5B_n)]$$

Where Cn is the concentration of the element in the sample, and Bn is the concentration of the same element in a baseline sample. The geoaccumulation index comprises seven classes: Igeo ≤ 0 (class 0, uncontaminated); 0 <Igeo ≤ 1 (class 1, uncontaminated to moderately contaminated); 1 <Igeo ≤ 2 (class 2, moderately contaminated); 2 <Igeo ≤ 3 (class 3, moderately to heavily contaminated); 3 <Igeo ≤ 4 (class 4, heavily contaminated); 4 <Igeo ≤ 5 (class 5, heavily to extremely contaminated); Igeo> 5 (class 6, extremely contaminated) [27]. In the present study, baseline concentrations correspond to stream sediments located within the study area; however, they have not been polluted by the mining activity of the region [28]. On the other hand, a correlation analysis was performed to investigate the relationship between the studied heavy metals (Cd, Cu, Cr, Fe, Mn, Ni, Pb, Zn) and the physicochemical properties of the sediments.

Results and Discussion

Sediment Characterization

Sediments collected in the Bacanuchi river presented, mainly, a sandy texture (67 to 85 wt%), Represo Tinajas 1, presented a gravel-sandy texture (41 to 51wt%), meanwhile, the fine fractions (limes and clay) where present in a lesser extent. The previous fraction represented less than 5% of the total amount. In the Sonora river, sediments presented the following texture types: sand (34 to 77.5 wt%), gravel (7 to 50.6 wt%), silt and clay (<12%). The textural features in both rivers are similar (except the fine fraction). In both rivers the fractions of sands and gravel are much larger than the finer fractions in the sediments of both rivers. Studies have reported similar results [16]. Previous studies report that variations in the particle size of the sediments are related to the patterns of water flow [29]. The textural analysis shows texture is strongly dependent on the water flow processes affecting the area.

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Regarding the distribution of metals in the different granulometric fractions of the sediments, the results indicate that the sand fraction plays a primordial role in the storage of metals. In this fraction, the metal levels fluctuated in the following percentages: Cu (54 - 89 %), Cr (52 - 89 %), Fe (51 - 89 %), Mn (53 - 88 %), Ni (48 - 88 %), Pb (46 - 86 %) and Zn (51 - 84 %). In the gravel fraction, significant amounts of metals were also observed: Cu (7 - 37%), Cr (6 - 39%), Fe (7 - 41%), Mn (10 - 40%), Ni (8 - 43%), Pb (11 - 46%) and Zn (12 - 41%). Results indicate most of the metals were found in the coarse fractions (sands) and in the sand and gravel fractions for Represo Tinajas 1. Other studies have reported similar behaviors [16,30]. The results obtained in the Bacanuchi river sediments are different from those reported by other studies in the literature, which shows that the content of metals in sediments is mostly associated with particle size [29].

Physical and Chemical Characterization of Sediments

In Table 1, the results of the physicochemical analysis of sediments of the Sonora and Bacanuchi rivers are presented. Regarding to pH, the stations closer to the source of contamination (Represo Tinajas 1, pH 3.1), such as Arroyo Tinajas and Bordo de Contención were the ones with the lowest pH values (4.14 to 4.4), respectively. These sampling stations received the spill of Fe/Cu acid solutions from Represo Tinajas 1 in August 2014. The rest of the sampling stations showed a pH ranging from 7.46 to 8.56. The acidic conditions favor the mobilization of metals in sediments, increasing their solubility as the pH decreases [31]. An acidic pH value in a sediment may indicate that, under certain conditions, some metals found in the site may be dissolved when pluvial runoff occurs and thus be mobilized to other areas. On the other hand, the concentration of sulfates in both rivers fluctuated in the range of 0.02 to 1.7%. The highest concentration of sulfates occurred in Represo Tinajas 1 (1.7%), which is the source of contamination. The results of sulfates in sediments of the Bacanuchi and Sonora rivers (including Represo Tinajas 1) are low compared to other studies. Previous research report high concentrations of sulfates (4.51-5.63%) in sediments of the San Pedro river contaminated by the mining activity of the region [13].

Table 1: Physicochemical parameters of surface sediments of Sonora and Bacanuchi rivers.

Sampling Sites	pH	SO_4^{-2} %	Cd mg/kg	Cu mg/kg	Cr mg/kg	Fe mg/kg	Mn mg/kg	Ni mg/kg	Pb mg/kg	Zn mg/kg
Bacanuchi River										
Represo Tinajas 1*	3.1	1.7	BDL	716	39	52400	593	48	210	470
Arroyo Tinajas	4.14	0.36	BDL	155	90	38200	612	25	124	207
Bordo de Contención	4.4	0.09	BDL	147	56	22100	560	31	130	220
Poblado Bacanuchi	7.69	0.34	BDL	94	52	26600	357	17	69	118
Bacanuchi (before confluence)	7.46	0.02	BDL	131	53	23500	297	20	69	113
Sonora River										
Sinoquipe	8.17	0.07	BDL	24	48	22000	291	23	48	77
Aconchi	7.64	0.07	BDL	18	49	21000	268	35	69	73
Ures	8.39	0.08	BDL	17	31	11700	212	6	68	56
Rancho Buena Vista	7.52	0.08	BDL	58	35	29100	732	26	79	147
San José de Gracia	8.56	0.08	BDL	8	24	7300	80	16	36	41
Presa El Molinito (discharge)	8.49	0.13	BDL	12	8	15100	938	17	14	79

* Source of contamination

BDL Below detection limit

Total Metal Determination in Sediment Samples

Total metals concentrations (mg/kg) in the Bacanuchi river fluctuated in the following ranges: Cd (<BDL), Cu (94 - 716), Cr (39 - 90), Fe (22,100 - 52,400), Mn (297 - 612), Ni (17 - 48), Pb (69 - 210) and Zn (113 - 470). In the Sonora river, total concentrations (mg/kg) were: Cd (<BDL), Cu (8 - 58), Cr (8 - 49), Fe (7,300 - 29,100), Mn (80 - 938), Ni (6 - 35), Pb (14 - 79) and Zn (41 - 147). The total metal levels in the Bacanuchi river (except Mn) were higher than those detected in the Sonora river. Represo Tinajas 1 station presented the highest values of Cu, Cr, Fe, Ni, Pb and Zn (Table 1). The Mn presented its maximum concentration (938 mg/kg) at Presa El Molinito (discharge),

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because it is located in a mineralized area. The mineralized areas (mineral deposits) influence the composition of aquatic systems, through the release of large amounts of metal ions [32]. Studies performed in the region where this reservoir is located have reported similar concentrations of Mn [33].

As for the behavior of the total metals concentration in the Bacanuchi river, the following order was observed: Fe>Cu>Mn>Zn>Pb>Cr>Ni. In the Sonora river, the order was as follows: Fe>Mn>Zn>Cu>Pb>Cr>Ni. Compared to other rivers in the world, total concentrations of Cu, Fe, Mn, Pb and Zn were much higher in the Bacanuchi river (except Paracatu river), while Cr and Ni were lower (Table 2). The Sonora river also had high concentrations of Cu, Mn, Pb and Zn with respect to sediments of the Tiaozi, Kuitum and Wei rivers, but lower than those reported in the Bacanuchi and Paracatu rivers (except Mn) [6,20,32,34].

Table 2: Comparison of metals content (mg/kg) found in sediments from different regions of the world.

Location	Cd	Cu	Cr	Fe	Mn	Ni	Pb	Zn	Reference
Tietê River	0.9 - 10.1	5.6 - 356.1	43.9-392.0	-	-	15.1-128.5	24.3 - 94.6	42.8 - 439.5	[3]
Asunle River	57.28 ±21.20	302.45 ±137.19	-	-	2411.77 ±968.50	930.60 ±539.82	34.08 ±22.98	313.09 ±88.66	[4]
El Jaralito and La Mexicana streams, Mexico	3.2 - 3.5	3.1 - 4.1	238-1090	4.12 - 6.10	678 - 1143	18 - 35	51 - 124	116 - 356	[16]
Hindon	0.29 - 6.29	21.70 - 280.33	17.48–33.70	0.41 - 1.7	49.55 - 516.97	13.90 - 57.66	27.56 - 313.57	22.50 - 288.29	[21]
Ghalechay	9.47 - 95.46	66.45 - 210.80	-	15.92 - 50.66	-	-	130.58 - 287.52	55.82 -122.28	[24]
Paracatu River	-	11 - 126	43-106	1.96 - 13.67	200 - 1900	-	10 - 159	22 - 2085	[32]
Tiaozi River	0.19	42.48	-	-	377.08	29.39	21.50	83.56	[45]
Kuitun River	-	44.20 - 56.34	42.39-129.79	-	497.92 - 629.31	20.21 - 25.51	22.31 - 34.73	1.27 - 1.43	[46]
Present study Bacanuchi river	BDL	94 - 716	39-90	2.21 -5.24	357 - 612	17 - 48	69 - 210	113 - 470	
Present study Sonora River	BDL	8 - 58	8-49	0.73 - 2.91	80 - 938	6 - 35	14 - 79	41 - 147	

Geochemical Distribution of Heavy Metals in Sediments

In Figure 2, the distribution of the metals in the different geochemical fractions of the sediment is presented.

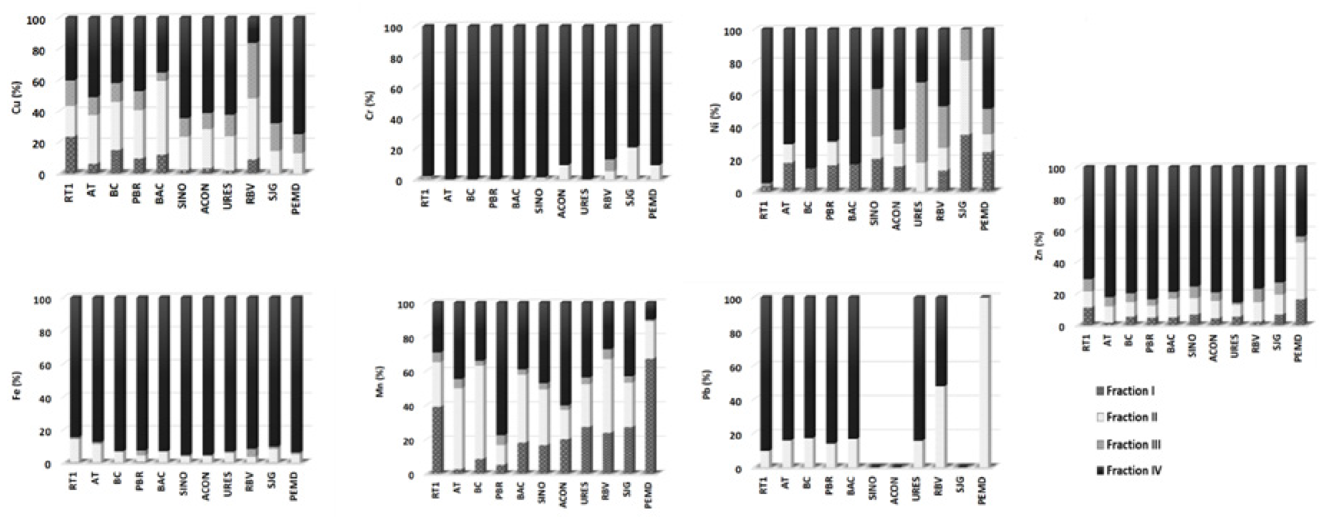

Figure 2: Distribution of Cu,Cr,Fe,Mn,Ni,Pb,Zn in sediments of Sonora and Bacanuchi rivers using sequential extraction.

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Fraction I (Exchangeable and Acid Soluble Fraction): This fraction is generally considered as the bioavailable portion of metals and, if they are present at high enough concentrations, they may be toxic to aquatic organisms [35-37]. In the Bacanuchi river, the minimum and maximum concentrations (mg/kg) were Cu (7 - 149), Fe (27 - 141), Mn (11 - 201), Ni (1 - 2.70) and Zn (3 - 23). In the Sonora river, the concentrations were as follows: Cu (<BDL - 2.82), Fe (2 - 17), Mn (20 - 602), Ni (<BDL - 3) and Zn (2 - 11). The Represo Tinajas 1 station presented the maximum values of exchangeable metals (mg/kg): Cu (149), Fe (141) and Zn (23), with percentages of 23.60%, 0.29% and 11.63%, with respect to the total metal content. Presa El Molinito (discharge), located in the Sonora river, presented the maximum values of Mn (602 mg/kg) and Ni (3 mg/kg), with percentages of 65.15% and 24.46% with respect to the total content. The highest concentrations of metals in the exchangeable fraction (except Mn and Ni) were detected in the Bacanuchi river sediments. This behavior is very similar to that reported in other studies [4,37].

Mobility and bioavailability in the Bacanuchi river were as follows: Mn>Cu>Fe>Zn>Ni; and in the Sonora river: Mn>Fe>Cu>Zn>Ni, therefore, metals accumulated in this fraction are more susceptible to be mobilized by changes in environmental conditions. Adsorption-desorption reactions, or a decrease in pH, change the ionic composition and could therefore remobilize metals in this fraction and turn them back to surface water where they may represent a potential hazard to the environment [37]. Cd, Cr and Pb presented values below the detection limit (<BDL) in both rivers, indicating that they have low potential to be remobilized and to be bioavailable in the environment.

Fraction II. Fraction related to Fe/Mn oxides: In this fraction, metals are strongly bound to Fe/Mn oxides; however, they are thermodynamically unstable under anoxic conditions [38] and, therefore, they could be dissolved in the water column and thus mobilized [39]. The minimum and maximum concentrations (mg/kg) in the Bacanuchi river were: Cd and Cr (<BDL), Cu (29 - 124), Fe (1083 - 6733), Mn (24 - 305), Ni (<BDL - 1.90), Pb (6 - 21) and Zn (7 - 21) (Figure 2). In the Sonora river had the following concentrations: Cu (1 - 12), Cr (<BDL - 1), Fe (518 - 856), Mn (25 - 302), Ni (<BDL – 3.5), Pb (<BDL - 20) and Zn (3.8 - 24). The Represo Tinajas 1 station had the highest values of Cu (124 mg/kg) and Fe (6733 mg/kg), while in Bordo de Contención had maximum values of Mn (305 mg/kg) and Pb (21 mg/kg) with percentages of 19.72, 13.87, 26.03 and 17.21, respectively. In the Sonora river, the Mn presented the highest values in the stations Rancho Buena Vista (302 mg/kg) and Presa El Molinito (discharge) (221 mg/kg), with percentages of 41.96% and 23.89%, with respect to the total metal content.

The highest concentrations of metals (Cu, Fe, Mn, Pb) were detected in the sediments of the Bacanuchi river, while in the Sonora river they were Cr, Ni and Zn. Mobility and bioavailability in the Bacanuchi river were as follows: Fe>Mn>Cu>Zn>Pb>Ni; and in the Sonora river: Fe>Mn>Pb>Zn>Cu>Ni. The fraction II presented the highest values with respect to the FI; it can be inferred that metals belonging to this fraction will have higher mobility and bioavailability than FI, and this will depend on changes in the pH and/or redox conditions of the sediment [24,38].

Fraction III. Fraction Associated with Organic Matter and Sulfides: The minimum and maximum concentrations (mg/kg) of metals in the Bacanuchi river are Cu (3 - 104), Cr (<BDL - 0.70), Fe (114 - 817), Mn (11 - 29), Ni (<BDL - 0.70) and Zn (3 - 16). For Sonora river, concentrations were as following: Cu (1 - 11), Cr (<BDL - 1), Fe (81 - 1169), Mn (5 - 45), Ni (1 - 7), Pb (<BDL) and Zn (2 - 9) (Figure 2). In both rivers Cd and Pb were not detected (<BDL). Represo Tinajas 1 presented the highest value for Cu (104 mg/kg), Fe (817 mg/kg) and Zn (16 mg/kg), with percentages of 16.49, 2.97 y 7.89 with respect to the total metal content. In the Sonora River, Rancho Buena Vista station showed the highest value for Fe (1169 mg/kg), Mn (45 mg/kg), Ni (7 mg/kg) and Cr (1 mg/kg), with percentage of 4.86, 6.29, 25.69 and 7.74, respectively.

Mobility and bioavailability in the Bacanuchi river were as follows: Fe>Cu>Mn>Zn>Ni>Cr; and in the Sonora river: Fe>Mn>Cu>Zn>Ni>Cr. It has been reported that the organic fraction (sulfides) is considered the most important component in the adsorption of metals in sediments. Some studies have reported high concentrations of metals associated with organic matter [2,38,40]. Some metals such as Cu can form complexes with organic matter, so that during their decomposition can cause their release into the environment.

Residual Fraction: Metals in this fraction are less harmful to the environment, because this fraction is chemically stable and biologically inactive [24]. Therefore, they are not likely to negatively impact surface water quality [2]. In the present study, the highest concentrations of most metals, in both rivers, were detected in this fraction. The Bacanuchi River presented the following percentages: Cu (35 - 51), Cr (98 - 100), Fe (84 - 93), Mn (29 - 77), Ni (71 - 95), Pb (83 - 91) and Zn (71 - 84). In the Sonora river, the percentages were Cu (16 - 75), Cr (79 - 99), Fe (90 - 95), Mn (10 - 60), Ni (<BDL - 90), Pb (<BDL - 52) and Zn (44 - 86). In both rivers Cd was not detected (<BDL) (Figure 2). Other studies have reported results similar to those obtained in the present research [8,13,16,41]. Metal concentrations in this fraction can be used as baseline data for the evaluation of the river system pollution. The stability of this fraction is controlled by the mineralogy and the extent of physicochemical weathering of the sediment [24].

Non-Residual Fraction: Metals in the non-residual fraction indicate the occurrence of anthropogenic contributions due to activities developed on areas nearby the river, such as mining and urban activities (untreated wastewater discharge). The non-residual metal fraction (FI+FII+FIII) was analyzed since this fraction is more bioavailable than the residual one. In the Bacanuchi river, the percentages of heavy metals in the non-residual fraction fluctuated in the following ranges: Cu (48 - 65), Cr (<BDL - 2), Fe (7 - 16), Mn (22 - 71), Ni (5 - 31), Pb (9 - 17)

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and Zn (16 - 29). In the Sonora river, they were Cu (25 - 84), Cr (<BDL - 21), Fe (5 - 10), Mn (40 - 90), Ni (38 - 100), Pb (<BDL - 100) and Zn (14 - 56). Cd presented non-detectable values (<BDL). In other studies, similar results were obtained [4,42]. The possible metal mobility in the non-residual fraction, for the Bacanuchi and Sonora rivers, is as follows: Mn>Cu>Ni>Zn>Fe>Cr and Ni>Pb>Mn>Cu>Zn>Cr>Fe, respectively. For both rivers, high metal concentrations were observed in the non-residual fraction, hence, a high mobility and bioavailability are possible. Therefore, these metals may have an impact on water quality and a harmful effect on biota.

Enrichment Factor and Geoaccumulation Index

In the present study, the enrichment factor (EF) was calculated using sediment baseline samples that are not affected by the rivers under study or by anthropogenic activities [28]. The results of EF of each metal in both rivers fluctuated in the following ranges: Cu (0.7 - 8.9), Cr (0.4 - 3.8), Mn (0.4 - 3.6), Ni (0.7 - 5.5), Pb (0.4 - 8.9) and Zn (0.7 - 2.8) (Table 3). In most sampling stations, the EF value for Cu, Cr, Mn, Ni, Pb and Zn was above 1.0, indicating an enrichment that may be attributed to the mining activity of the region, specifically in the Bacanuchi river. Regarding Igeo, in the Bacanuchi river, the Bacanuchi station (before confluence) presented an Igeo of 2.1 for Cu indicating a moderate and strong contamination; the Poblado Bacanuchi and the Bacanuchi (before confluence) stations had Igeo values of 1.4 and 2.1 for Pb, presenting moderate and strong contamination. In the Sonora river, the Sinoquipe station presented a value of 1.0 for Cr, indicating a null to moderate contamination (Table 3).

Table 3: Enrichment factors and Geoaccumulation index of the analyzed sampling sites from Sonora and Bacanuchi rivers.

Sampling Sites	Enrichment Factors					
Bacanuchi river:	**Cu**	**Cr**	**Mn**	**Ni**	**Pb**	**Zn**
Arroyo Tinajas	1.0	1.5	0.6	1.0	1.2	1.1
Bordo de Contención	2.6	1.5	1.1	2.7	3.1	2.7
Poblado Bacanuchi	3.5	0.6	0.7	0.7	5.0	1.7
Bacanuchi (before confluence)	8.9	1.2	0.7	1.9	8.9	1.9
Sonora River:						
Sinoquipe	1.6	3.8	0.4	3.1	0.4	0.7
Aconchi	0.7	1.5	0.7	4.5	1.7	0.9
Ures	3.4	1.9	0.6	1.3	5.9	2.3
Rancho Buena Vista	4.6	0.9	1.5	2.3	3.2	2.5
San José de Gracia	2.4	2.3	0.6	5.5	5.8	2.8
Presa El Molinito (discharge)	1.8	0.4	3.6	3.0	1.0	2.6
Sampling sites	**Geoaccumulation Index**					
Río Bacanuchi:	Cu	Cr	Mn	Ni	Pb	Zn
Arroyo Tinajas	-0.4	0.1	-1.2	-0.4	-0.2	-0.3
Bordo de Contención	-0.1	-1.0	-1.4	-0.1	0.1	-0.1
Poblado Bacanuchi	0.9	-1.6	-1.5	-1.5	1.4	-0.1
Bacanuchi (before confluence)	2.1	-0.8	-1.7	-0.1	2.1	-0.1
Río Sonora						
Sinoquipe	-0.3	1.0	-2.4	0.7	-2.1	-1.5
Aconchi	-1.8	-0.6	-1.8	1.0	-0.4	-1.3
Ures	-0.4	-1.2	-2.8	-1.8	0.4	-0.9

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Rancho Buena Vista	1.4	-1.0	-0.2	0.4	0.9	0.5
San José de Gracia	-1.5	-1.6	-3.4	-0.3	-0.2	-1.3
Presa El Molinito (discharge)	-0.9	-3.1	0.1	-0.2	-1.7	-0.4

Spearman Correlation Analysis

To analyze the association between Physical parameters and total metal content, a Spearman correlation analysis was used, since most parameters do not have a normal distribution. Results indicate there was a positive correlation among metals such as Cu, Fe, Ni, Pb, Zn and the gravel fraction (r = 0.29 to 0.36); while sand fraction had negative correlations with Cu, Fe, Ni, Pb and Zn (r = -0.28 to -0.43) (Table 4). The fraction of silt had a positive correlation with Cu, Fe, Mn, Ni, Pb and Zn (r = 0.30 to 0.70), while the clay fraction was positively related to Fe, Mn and Zn (r = 0.51 to 0.64). Similarly, other studies report a strong correlation between heavy metals and particle size, being silt and clay the fractions most associated to metals due to their large specific surface area in which metals are adsorbed [43]. In general, most metals had a positive correlation between each other (r = 0.46 to 0.91), which may indicate a strong mineralogical association coming from the same origin, that is, the mining activity of the region. This agrees with data reported by other studies [43,44]. On the other hand, Cu, Fe, Mn, Pb and Zn showed a positive correlation with sulfates (r = 0.33 to 0.59), indicating that they may be associated as sulfates. Regarding pH, all metals (including sulfates) had negative correlations with pH values (r = -0.32 to -0.97).

Table 4: Spearman's correlation matrix among physical and chemical parameters, grain size and total metals of surface sediment.

	pH	SO_4^{-2}	Cu	Cr	Fe	Mn	Ni	Pb	Zn	Gravel	Sand	Silt	Clay
pH	1.00												
SO_4^{-2}	-0.32	1.00											
Cu	-0.94*	0.4	1.00										
Cr	-0.69*	-0.01	0.70*	1.00									
Fe	-0.86*	0.48**	0.87*	0.52**	1.00								
Mn	-0.47**	0.53**	0.46	0.17	0.66*	1.00							
Ni	-0.70*	0.06	0.56**	0.36	0.48**	0.29	1.00						
Pb	-0.97*	0.33	0.90*	0.61*	0.80*	0.4	0.69*	1.00					
Zn	-0.85*	0.59*	0.87*	0.50**	0.91*	0.78*	0.54**	0.82*	1.00				
Gravel	-0.32	-0.35	0.33	-0.01	0.36	0.05	0.29	0.32	0.3	1.00			
Sand	0.36	0.29	-0.39	-0.05	-0.43	-0.1	-0.28	-0.36	-0.33	-0.98*	1.00		
Silt	-0.49**	0.79*	0.47	0.05	0.70*	0.65*	0.3	0.50**	0.69*	0.05	-0.16	1.00	
Clay	-0.34	0.37	0.27	-0.05	0.64*	0.61*	0.13	0.3	0.51**	0.37	-0.46	0.81*	1.00

*Significant at 0.05 level; **significant at 0.1 level

Sediment Quality Criteria

Total metal concentrations were compared to the Low Effect Level (LEL) and the Severe Effect Level (SEL) criteria, [45,46]. If metal concentration is higher than the LEL, that metal may moderately affect biota, and if the metal exceeds the SEL criterion, the metal may severely affect biota. Most of the sampling stations in both rivers exceeded the LEL quality criterion for Cr, Cu, Fe, Mn, Ni, Pb and Zn, thus a moderate effect on biota is possible (Table 5). The SEL criterion was only exceeded for Cu in the Bacanuchi river (Arroyo Tinajas, Bordo de Contención and Bacanuchi (before confluence)), and Fe in the Poblado Bacanuchi Station. This suggests that there could be a severe effect caused by Cu and Fe, to the biota. Previous research has reported similar results in San Pedro river sediments contaminated by mining activity in the state of Sonora, Mexico [16].

Table 5: Concentration comparison (mg/kg) of total metals (except Fe, %) with respect to the Sediment Quality Criteria LEL and SEL [43,46].

Sampling Sites	Cr	Cu	Fe %	Mn	Ni	Pb	Zn
Bacanuchi River							
Represo Tinajas 1	39	716*	5.24*	593	48	210	470
Arroyo Tinajas	90	155*	3.82	612	25	124	207
Bordo de Contención	56	147*	2.21	560	31	130	220
Poblado Bacanuchi	52	94	2.66	357	17	69	118

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Bacanuchi (before confluence)	53	131*	2.35	378	20	69	113
Sonora River							
Sinoquipe	48	24	2.20	291	23	48	77
Aconchi	49	18	2.10	268	35	69	73
Ures	31	17	1.17	212	6	68	56
Rancho Buenavista	35	58	2.91	732	26	79	147
San José de Gracia	24	8	0.73	80	16	36	41
Presa El Molinito (discharge)	8	12	1.51	938	17	14	79
Low Effect Level (LEL)	26	16	2.0	460	16	31	120
Severe Effect Level (SEL)	110	110	4.0	1100	75	250	820

*Sampling stations that exceeded LEL and SEL levels.

Conclusion

A study was carried out to evaluate the mobility and bioavailability of heavy metals in sediments of the Sonora and Bacanuchi rivers, affected by the mining activity. The Bacanuchi river presented the highest levels of heavy metals, which concentrations had the following descending order: Fe>Cu>Mn>Zn>Pb>Cr>Ni>Cd. The Sonora river had the following order: Fe>Mn>Zn>Pb>Cu>Cr>Ni>Cd. Sediments in both rivers presented a sandy texture, with high concentrations of most of the analyzed metals. The sequential extraction study showed that the predominant order of metals was as follows: residual>Fe/Mn oxides>exchangeable>organic matter. However, a significant percentage of metals were associated with the non-residual fraction, which represents anthropogenic contributions due to the region's mining activity. The potential mobility of metals in the non-residual fraction had the following order: Mn>Cu>Ni>Pb>Zn>Fe>Cr.

EF showed an enrichment of anthropogenic origin in both rivers for Cu, Cr, Mn, Ni, Pb and Zn, indicating an enrichment that may be derived from the mining activity of the region, specifically in the Bacanuchi river. Most Igeo values were within the classification of non-contaminated to moderately contaminated. Bacanuchi station (before confluence), showed moderate to strong contamination of Cu and Pb. In both rivers, the LEL criterion was exceeded for Cr, Cu, Fe, Mn, Ni, Pb and Zn, and therefore, a moderate effect on biota is expected. The SEL criterion was only exceeded for Cu and Fe in the Bacanuchi River. This may represent a danger to the biota of both rivers, due to their high mobility and bioavailability.

This study has generated important information on the concentration of metals, their mobility/bioavailability, and their possible effect on biota. However, future studies on water and sediment chemistry (including biota) are required to fully assess the Bacanuchi and Sonora rivers. These studies are particularly important due to the spill occurred in August 2014, in the Cananea region of Sonora, Mexico, which consequences have not yet been fully evaluated.

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