Journal of Ecohumanism
2025
Volume: 4, No: 1, pp. 861 – 871
ISSN: 2752-6798 (Print) | ISSN 2752-6801 (Online)
https://ecohumanism.co.uk/joe/ecohumanism
DOI: https://doi.org/10.62754/joe.v4i1.5888

Assessment of Heavy Metal Contamination in Water Resources of the Andean Mining Zone: A Decadal Analysis of the Santa Rosa River Basin, Ecuador

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Abstract

The vast majority of mineral deposits that exist in Ecuador are located in the Andean zone. It is important to carry out permanent monitoring of heavy metals present mainly in the water, es-pecially in regions where mining is one of the economic activities. The objective of this work is to evaluate the possible contamination of the water that supplies the community and that uses it for human consumption, due to the mining activity by potentially toxic elements: Mercury, Ar-senic, Lead and Cadmium, and its comparison. with the analyzes carried out in the last 10 years in this same sector of the Santa Rosa river basin, The analysis of the samples was carried out by Atomic Emission Spectroscopy with Inductively Coupled Plasma (ICP-AES), the results obtained show contamination of the water resource, according to the United States Environmental Protec-tion Agency (EPA), and the World Health Organization (WHO) , and from the surveys applied to the community, the use of this water in different activities, both domestic and agricultural, is evidenced, with this a better perspective of the influence of anthropogenic activity on the influ-ence of water quality can be obtained.

Keywords: *Water Quality, Toxic Effects, Heavy Metals, Mining, Public Health, Anthropogenic Activity.*

Introduction

The activities of man have always interacted with the environment either to a greater or lesser extent, this is due to the great growth of the world population, causing a greater increase in the needs of resources and food, causing damage to the environ-ment, some of which cannot be solved, such as water, soil or air pollution, depletion of non-renewable resources, generation of greenhouse effect, etc. [1].

According to the Central Bank of Ecuador (BCE), between January and November 2020, the export of mining products reflected a growth of 206% compared to the same period in 2019, in this context, until November 2020, the Minerals represented 4.40% of the country's total exports, they ranked as the sixth most exported product after oil, shrimp, bananas, canned fish and cocoa, the increase in mining exports in 2020, despite The stoppage of activities due to the COVID 19 pandemic was mainly due to the con-tinued growth of small-scale mining activities, as well as the results of large-scale pro-duction from the Mirador (copper) and Fruta mines. North (gold); Another relevant re-sult in this sector is reflected in Foreign Direct Investment (FDI) that this industry at-tracted until the third quarter of 2020. These data, published by the ECB, position min-ing as the activity that continues to be the one that extracts the most foreign currency. -jeras has captured, occupying 41.73% of the national total of FDI. Despite the health crisis caused by the spread of COVID-19, mining continues to consolidate itself as a strategic and necessary axis for the social and productive development of Ecuador, since these currencies energize and contribute to the economic stability of the country [2,53]

The environment and society have become fundamental elements for decision-making and the development of operations in the mining industry. The impacts registered by the mining activity are: environmental, such as water resources, air quality and wild-life; and social impacts such as human displacement, access to livelihoods, public health, and cultural resources, which puts society and the entire world on alert [3]. This activity is traditionally carried out without observing biosafety measures both in the

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mine, in the narrow and hot tunnels, in the transport of the material, and also in the beneficiation plants, where gold, silver and other metals are processed, becoming a danger constant.[4].

The most significant impact that mining produces on its environment is the effect on the availability and quality of water resources, since when materials such as tailings, debris, and leachates are exposed to oxygen and water, an acid is created that dissolves the metals and other contaminants found in mining materials and form a high sulfate acid solution rich in metals, including possible high concentrations of copper, cadmi-um, lead, zinc, arsenic, etc.), which is generally thrown directly into the riverbed with-out receiving any prior treatment, thus causing contamination of the water resource due to the presence of a high content of heavy metals according to the EPA [5,6].

The presence of heavy metals in the environment and food as described can trigger various poisoning causing irreparable damage to human health such as teratogenic ef-fects, cancer and even death, it is important to consider that high concentrations of said metals in the body of living beings alter biochemical and physiological processes, causing various pathologies. According to the EPA, high-density metallic elements (for example, chromium, mercury, arsenic, cadmium, and lead); they can be toxic to living beings in small concentrations and tend to accumulate in the trophic chain", [6,7]. Ac-cording to the WHO, we can mention some toxic effects in humans caused by heavy metals, such as chronic toxicity with arsenic causes skin lesions (keratosis, hyperkera-tosis, hyperpigmentation) and vascular lesions in the nervous system and liver. Acute complications appear due to exposure to high doses and can be lethal. Their first ef-fects are usually fever, hepatomegaly, melanosis, cardiac arrhythmia, peripheral neu-ropathy, anemia, and leukopenia [8,9]. Arsenic is classified in group I of carcinogenic substances by the International Agency for Research on Cancer (IARC), the types of premature skin cancer (basilioma and squamous cell carcinoma), lung (bronchogenic carcinoma), hemangiosarcoma liver, lymphoma, and cancer of the bladder, kidney, and nasopharynx [9,10].

Chronic mercury poisoning presents tremors, thyroid hypertrophy, tachycardia, gingi-vitis, personality changes, erethism, memory loss, severe depression, delusions and hallucinations. The three most used traits to recognize occupational disease in industry are excitability, tremors, and gingivitis [10]. In addition, there is kidney damage from chronic exposure to mercury, reversible effects, which cease when the person avoids exposure, the main effects of methylmercury in adults are neurological, while exposure during pregnancy causes the appearance of congenital lesions of the nervous system [11]. Neurotoxicity manifests with tremors and loss of sensation in the fingers of both extremities, ataxia, loss of vision and hearing, spasms, and finally coma and death. [12].

Poisonings caused by pesticides represent an important public health problem, given the diversity of chemicals that constitute them, the high number of active principles and the multiple applications in practically all activities of daily life. Acute intoxica-tions (AI) by organophosphates (OF) are especially frequent in agricultural areas, where these toxins are commonly used; occupational exposure being an important cause of toxicity [13]

When mining is for gold, soil contamination by anthropic activities has gradually worsened in recent times, coming to be considered a serious environmental problem on a planetary level [14]. In relation to health, the impacts of gold mining are concen-trated in the exposure of mining workers to toxic substances such as mercury and cya-nide, especially in the manual and rudimentary practices of artisanal mining, in which workers they perform without hygiene conditions and labor protection [15, 16, 17]. But there are also health problems in the inhabitants of the towns surrounding the gold mining deposits and in those located downstream, due to high levels of toxic substanc-es such as mercury, cyanide and manganese present in the tributaries. water and food obtained directly from rivers and subsistence agriculture, such as fish, bananas, rice, among others [15]

The mining activity dedicated to the extraction of gold in Ecuador is officially framed, through the National Secretariat for Planning and Development, in various medium and large-scale projects: 1) Río Blanco, located in Molleturo and Chauca , Cuenca can-ton, Azuay province; 2) Fruta de Norte, located in Los Encuentros, Yantzaza, Zamora Chinchipe; 3) Viewpoint, developed in Tundayme, El Pangui canton,

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Zamora Chinchipe; 4) Loma Larga, located in San Genaro; 5) Chumblín and Victoria del Portete, in the Cuenca canton, Azuay province; 6) Cascabel Project, located in Lita, Ibarra can-ton, Imbabura; and 7) Cangrejos Project, located in Bella María, Santa Rosa canton, El Oro (Central Bank of Ecuador, 2019). Currently, some of these projects are paralyzed due to legal actions taken by the communities for environmental protection, such as the case of Río Blanco. Others are in the exploration phase and close to starting their extraction activities, such as the Loma Larga project, whose production is expected to start in 2021. [17].

International estimates allow us to assert that in Ecuador there are approximately 60,000 artisanal mining workers, ranking fourth in the Latin American region, after Brazil with 250,000, Colombia with 200,000 and Bolivia with 100,000, which is a conse-quence of the traditional dominance of artisanal mining over large-scale mining. Ap-proximately 6,000 artisanal miners are concentrated in the town of Portovelo - Zaruma alone [15,17]

In addition to this medium- and large-scale mining activity, small-scale and artisanal mining extractive activities are also carried out, which contribute approximately 89% and 11%, respectively, of Chinchipe gold production (Ministerio de Minería de Ecua-dor, 2016). They are concentrated in the south of the country, such as Portovelo-Zaruma, El Oro (figure 1 (a)), Azuay and Zamora.[18]

The Province of El Oro leads the percentage of mineral extraction in Ecuador by 85% [21], and according to the cadastre of the Mining Regulation and Control Agency (ARCOM) Machala Regional Coordination, it is registered that in the canton Santa Ro-sa 22,140.46 hectares in which mining works are being carried out, of which, an area of 17,841.46 hectares, which represents 80.58%, are of metallic mineral resources (mainly gold); 3,347 hectares, that is, 15.12% are non-metallic mineral resources; and 952.00 hec-tares, which is equivalent to 4.30%, is made of stone material such as gravel, sand and ballast. These farms are located in the parishes of Bella María, Torata, Victoria and La Avanzada. It is important to mention some of the large-scale projects that are being developed in the Province of El Oro, such as the Crab Project, which is located in the Bella María and San José de Cedro Azul parishes, in the cantons of Santa Rosa and At-ahualpa in the province of El Oro; and, it is concessioned to Odín Mining del Ecuador S.A., which is a Quito-based mining exploration and development firm focused on gold and copper projects in Ecuador, a subsidiary of Canada's Lumina Gold Corpora-tion. Until the third quarter of 2020, the Cangrejos project has not yet defined the ex-traction method and is in the advanced exploration period. The life of the project has been calculated at around 20 years and the start date for the construction of the mine has not been determined either. The Cangrejos Project is made up of 10 concessions: Los Cangrejos, Los Cangrejos 11, Cangrejos 10, Cangrejos 20, Crabs A, Crabs B, Crabs C, Crabs D, Casique and Canarias. The Ministry of the Environment granted the ad-vanced exploration license to the Los Cangrejos concession; to the Cangrejos 20 con-cession, initial exploration license and the advanced exploration license is pending. For its part, the Secretariat of Water granted authorizations for the use and exploitation of water and for industrial use to the Los Cangrejos concession; and, the permit for water use and water use and for not affecting water sources to the Cangrejos 20 concession. Finally, this project until the third quarter of 2020 generated 380 jobs of which 95 are direct (46 are generated by the company and 49 by the contractor) and 285 are indirect jobs [22,23,24,25]. The fluctuation of the direct and indirect employees of the mining sector that work in the province of El Oro fluctuates depending on the needs, in rela-tion to the different projects. Most of these are made of precious metals such as gold, silver, and copper, although there are also a smaller number of projects related to lime-stone, clay, pumice, etc. [20]

Santa Rosa (figure 1 (b)) is an Ecuadorian city; cantonal head of the Santa Rosa Canton, as well as the third largest and most populated city in the province of El Oro. It is lo-cated to the south of the coastal region of Ecuador, on an extensive plain, on the right bank of the Santa Rosa River [19], the Santa Rosa canton has two hydrographic sources: the Caluguro River, which has the Chico River and the Biron River as tributaries; and that of the Santa Rosa River whose tributaries are born from the La Chilca, El Guayabo and Sabayán hills, located in the Dumarí Mountain Range, which is part of the Andes Mountains. It also has the Buenavista River, which is the geographical feature that borders the Machala and Pasaje canton, has as tributaries the Caluguro River, Negro River, San Agustín River and Dumarí River. The union of the Santa Rosa and Buenavis-ta rivers gives rise to the Pital river [19]. The Santa Rosa River comprises 6,838

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DOI: https://doi.org/10.62754/joe.v4i1.5888

ha. and they are found specifically in the Torata and La Avanzada parishes [26,27}. The water collected from this micro-basin by the company EMAPASR S.A. it supplies the inhab-itants of the urban parishes of Santa Rosa and Nuevo Santa Rosa; and the rural parish-es of Bellavista, La Avanzada and Puerto Jelí. Around this space are 257 divided fami-lies located in eight neighborhoods.

The upper basin of the Santa Rosa River is an area where its main economic activity is mining, therefore, the objective of this study is to evaluate the existence of contamina-tion resulting from mining activity, and to analyze the evolution of contamination at the point of discharge of mining waste without prior treatment during the last 10 years.

Materials and Methods

To achieve the objective, an analysis of the presence of heavy metals is carried out: mercury, cadmium, arsenic and lead in the water coming from the upper basin of the Santa Rosa River.

The location where the river water samples were collected is shown in Figure 1(d), located in the Province of El Oro, southwestern Ecuador, to be transported to the city of Cuenca for their respective analysis.

At each sample collection point, it was georeferenced using GPS; In total there were 10 sampling points along 81 km along the entire length of the upper basin of the Santa Rosa River, different zones were established that surround the sampling points established based on different economic or social criteria such as: agricultural area, mining area, populated area, drinking water supply area[30,31,32]

The ArcGIS geographic information system software was used to perform the georeferencing of the study area. Figure 1(a-c) shows the georeferencing of the points in detail and Table 1 the coordinates and description of the ten sampling points.

Figure 1. (a) Map of Ecuador and its respective provinces; (b) Map of the Province of El Oro with its respec-tive cantons; (c) Upper Basin of the Santa Rosa River (d) Sampling points for heavy metals.

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Tabla 1. Heavy Metal Sampling Points with Their Respective Coordinates in the Upper Santa Rosa River Basin

Name of the sampling place	Sampling point	Zone Description
Santa Rosa River, Guayabo site:	1	Location of most mining concessions.
the ravine: The Panteón	2	Unloading point for mining waste without prior treatment. *
the junction of the ravine the Panteón with the Santa Rosa River	3	Union of two rivers.
Kilometers downriver from the Guayabo site	4	agricultural zone
Kilometers downriver from the Guayabo site	5	Agricultural zone and beginning of populated zone
Playón-Torata Parish	6	populated area
Limón Beach-Torata Parish	7	populated area
Remolino place, La Avanzada parish	8	populated area
Collection of drinking water from the canton of Santa Rosa.	9	Drinking water collection
Santa Rosa Canton drinking water treatment plant	10	Water treatment plant

After the sampling, the physicochemical tests of the water samples were carried out, the containers were treated with a previous washing, the same one that was car-ried out with nitric acid diluted to 10% to eliminate any type of substance that could interfere with the tests, the chemical analysis was carried out in the Inductively Cou-pled Plasma (ICP-AES) equipment [33,34,35], the results are obtained in ppm[36,37], the technique used for the sampling of the last 3 years was the same as that used by the municipality through of his company EMAPASR-EP (Empresa Pública de Agua Po-table y Alcantarillado del Cantón Santa Ro-sa) and that provided us with the results from 2012 to 2019, since the results obtained from 2020 to 2023 were those obtained by the researchers of the present document, and that it can be indicated that the results are comparable since in both cases the technique used to obtain the results is the same.

The samples for monitoring heavy metals were distributed as follows: the first 8 samples correspond to river water from the upper basin of the Santa Rosa River, and samples 9 and 10 correspond to the water that will be used for the control. -Human supply after entering the treatment plant where the previous disinfection process is carried out and then distributed to the network.

The trend change analysis was performed using joinpoint regression and the free software Joinpoint Trend Analysis Software version 5.0.1 developed by the Surveil-lance Research Program of the US National Cancer Institute. (https://surveillance.cancer.gov/joinpoint/)

Results

The results of the analyzes of the samples obtained in the upper basin of the Santa Rosa River compared with the respective regulations can be seen in the Table 2.

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Tabla 2. Results of Heavy Metal Analysis Along the Santa Rosa River

*	**EPA**	**OMS**	**Legislation Ecuatoriana**	**m 1**	**m2**	**m3**	**m4**	**m5**	**m6**	**m7**	**m8**	**m9**	**m10**
Lead	0.01	0.01	0.05	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Arsenic	0.05	0.01	0.05	0.004	0.0493	0.008	0.008	0.0093	0.0085	0.0154	0.02	0.0099	0.0155
Cadmium	0.003	0.003	0.01	0.001	0.0075	0.022	0.022	0.0164	0.00	0.00	0.00	0.00	0.0106
Mercury	0.001	0.006	0.001	0.013	0.0088	0.001	0.001	0.0001	0.0009	0.00	0.00	0.0478	0.0471

Comparison of the results of the analysis of heavy metals from 2012 to 2023 in the up-per basin of the Santa Rosa river, Quebrada El Panteón zone-El Guayabo zone.

In the upper basin of the Santa Rosa River, analyzes of heavy metals in the water have been carried out by the public drinking water company of the Santa Rosa canton (EMAPA SR). Table 3 shows the values of the analyzes from 2012 to 2023 at a specific point such as the Quebrada El Panteón-El Guayabo sector; In this place, the wastewater from the mines without any previous treatment is discharged, the same one that joins the Santa Rosa River.

Mercury at sampling point 2 corresponding to Quebrada El Panteón from 2012 to 2018 complied with the maximum permissible limits, however, after 2019 it exceeded the permissible limits according to our analyzes of recent years [38,39,40]]

It is observed that arsenic exceeds the permissible range established in the stand-ard, being harmful to human health, lead with a value that meets the maximum per-missible limits.

Cadmium exceeds the maximum permissible limit established in the standard in the last few years, as does mercury that exceeds the permissible ranges in the analyzes of the last 4 years. [41,42,43]

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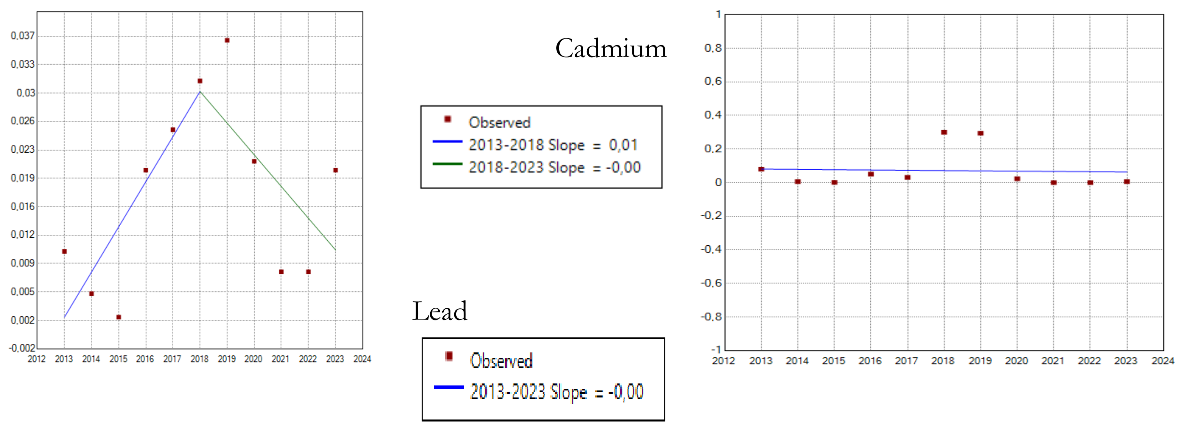
Tabla 3. Comparison of the Analyzes of Heavy Metals from the Year 2012 To 2022 in the La Quebrada Zone the Pantheon

	Arsenic (mg/l)	lead (mg/l)	Cadmium (mg/l)	Mercury (mg/l)
Year	**LMP 0.1**	**LMP 0.01**	**LMP 0.03**	**LMP 0.006**
2012	0.08	0.08	0.01	0.001
2013	0.081	0.0054	0.0048	0.0001
2014	0.0053	0.0008	0.0019	0.00068
2015	0.01	0.05	0.02	0.005
2016	3.7	0.031	0.025	0.0002
2017	0.0026	0.3	0.031	0.005
2018	0.03	0.294	0.036	<0.002
2019	0.2	0.0225	0.0211	0.0114
2020	0.049	<0.001	0.0075	0.00876
2021	0.055	<0.001	0.0075	0.0112
2022	0.075	0,0054	0.02	0.00896
2023	0.068	0.0048	0.02	0.0986

The results of the trend analysis did not indicate trend changes in three of the four metals studied, only Cadmium showed a trend change in 2018, where it began a downward trend after a prolonged upward trend. Lead decreases during all the years of the study and the other two metals remain practically constant. The results are shown in figure 2.



(a) Cadmium (b) Lead

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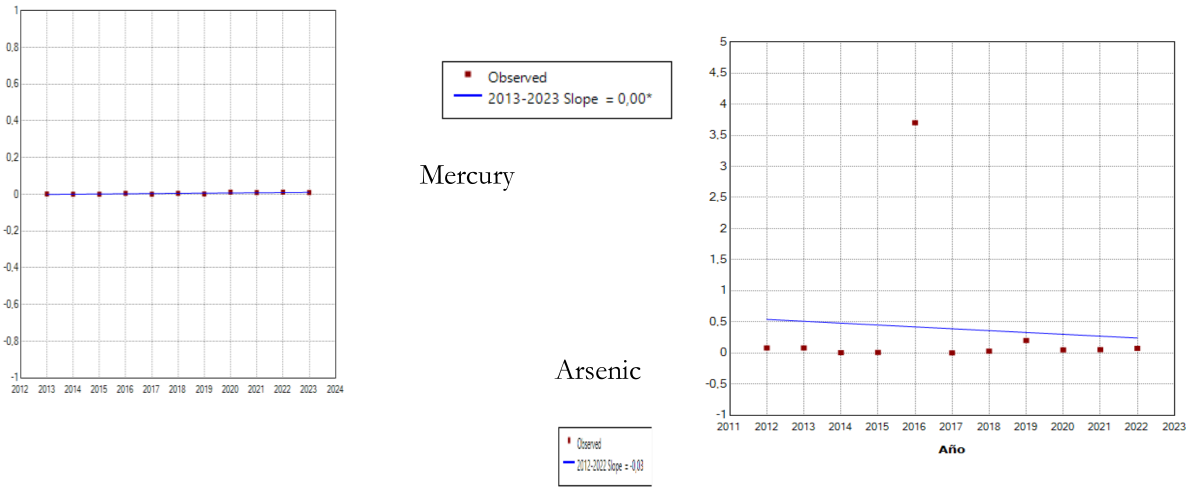

(c) Mercury

(d) Arsenic

Figura 2. Trend Analysis Using Joinpoint Regression for Metals (A) Cadmium, (B) Lead, (C) Mercury, (D) Arsenic.

Discussion

From the results obtained from the heavy metals analyzed such as: mercury, lead, cadmium and arsenic, of the eight samples taken from the river water, not all the sam-ples comply with the maximum permissible limits in accordance with the provisions -d in the regulations, at the sampling points, the values that exceed the permissible lim-its due to anthropogenic activity can be related specifically to the mining activity that occurs in this area, since this contamination is This could be due to the fact that sam-pling points 1 and 2 contain mining concessions that spill leachate into the Santa Rosa River without any prior treatment.

Sample 10 corresponds to treated water, obtained at the drinking water plant, whose values exceed the maximum limits of Arsenic, Cadmium and Mercury; allowed for drinking water, which can cause several effects on the health of the inhabitants of the sector.

On the other hand, the two samples taken both at the drinking water plant to the incoming water and the sample taken at the household tap, do not meet the maximum permissible limits in accordance with the provisions of the regulation on drinking wa-ter criteria, this result may be due to an accumulation of these heavy metals in drink-ing water storage tanks [44,45,46].

Routine analyzes monitor water quality based on physical, chemical and microbio-logical analyses, but it is necessary to link the water quality analysis to the different economic activities that the community engages in, in the case at hand, mining areas, heavy metals should be included in their routine analysis or, for example, in cultivated areas, chemical analyzes for the presence of pesticides should be included. Therefore, these routines analyze are insufficient [47,48,49].

Regarding the trends in the presence of heavy metals during the period studied, mercury has barely changed, lead and arsenic tend to decrease slowly, something that can be encouraging if it continues over time, although it has been It might have to speed up that decline. Cadmium has registered a large decrease since 2019, however, we should be cautious since it could be due to the inactivity of mining activity during the

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confinement caused by COVID-19, since the data for 2023 increases again and would have to wait for future data that confirms this downward trend that began in 2019.

From the foregoing, it is determined that there is contamination of the water re-sources of the upper basin of the Santa Rosa River by mining activity, especially in the El Guayabo sector, due to the fact that most of the mining concessions that discharge the leached without any prior treatment into the river, which is detrimental to aquatic life, agriculture, livestock, and human health in the area; It was also verified that there are heavy metals in the drinking water plant, being harmful to the population of the Santa Rosa canton that uses this water for human consumption.

In order for mining to develop sustainably, it requires foundations that establish criteria for making the right decisions, communicating to society, and correcting the sustainability gaps that we see in industrial or artisanal mining.

Conclusions

In the upper basin of the Santa Rosa river there is a presence of heavy metals and a possible accumulation of mercury, arsenic and cadmium in the water storage tank in-tended for human consumption, which would be harmful to health. Routine tests are incomplete. In order to avoid contamination of the water resources of the upper basin of the Santa Rosa River, we suggest some recommendations to government agencies; carry out periodic inspections of mining concessions and verify compliance with Envi-ronmental Management Plans and the application of treatments to eliminate heavy metals, in addition to adapting periodic analyzes according to economic activity. Measures of good environmental practices among artisanal mining should be promot-ed through training that allow certifying a responsible mining activity.

Author Contributions

- Conceptualization, methodology, research, writing—original draft preparation: Angélica Geovanna Zea Cobos.
- Software, resources, data curation, project management: Angélica Geovanna Zea Cobos.
- validation, formal analysis, writing—review and editing, visualization, monitoring, fund-raising: Angélica Geovanna Zea Cobos, Pablo Caballero.

Funding: "This research was funded by the Research Group Research in Biotechnology and En-vironment - INBIAM https://www.ups.edu.ec/inbiam, of the Salesian Polytechnic University.

Data Availability Statement

The data generated and analyzed during the current study are available in the public repository [https://orcid.org/0000-0002-6293-0735/] and [https://inbiam.blog.ups.edu.ec/investigadores/geovanna-zea] (https://inbiam.blog.ups.edu.ec/investigadores/geovanna-zea) and can be requested from the corresponding author via institutional mail at azea@ups.edu.ec. No specific code was generated for this stud.

Acknowledgments: The execution of this project is carried out thanks to the Universidad Politécnica Salesiana-Ecuador through its research groups INBIAM, and to the contribution and support of the population of the canton of Santa Rosa-Ecuador, thanks also to the University of Alicante for carrying out this work in an inter-institutional way, which contributes with a per-spective of a reality in Latin America. The author AGZC thanks the rest of the authors for their contribution to this work, which is part of her doctoral thesis.

Conflicts of Interest: "The authors declare no conflict of interest.

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Journal of Ecohumanism
2025
Volume: 4, No: 1, pp. 861 – 871
ISSN: 2752-6798 (Print) | ISSN 2752-6801 (Online)
https://ecohumanism.co.uk/joe/ecohumanism
DOI: https://doi.org/10.62754/joe.v4i1.5888
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