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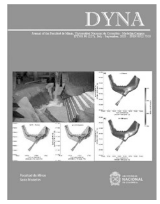

UNIVERSIDAD NACIONAL DE COLOMBIA

Mercury and human health: probabilistic risk characterization in one of the oldest gold mining areas in Ecuador

Daniela Paz-Barzola, Kenny Escobar-Segovia, Bryan Salgado-Almeida, Cindy Goyburo-Chavez & José Moreno-Chavez

Facultad de Ingeniería en Ciencias de la Tierra, Escuela Superior Politécnica del Litoral, Guayaquil, Ecuador. dpaz@espol.edu.ec, kescobar@espol.edu.ec, bryjosal@espol.edu.ec, cgoyburo@espol.edu.ec, josmoren@espol.edu.ec

Received: Mach 13th, 2023. Received in revised form: June 29th, 2023. Accepted: July 19th, 2023.

Abstract
Artisanal and small-scale gold mining is an important contributor to global human emissions of Hg. This study assesses the probabilistic human health risk for two receptors groups (residents and workers) of Portovelo mining area through exposure to Mercury (Hg) in the air at two sites: (a) the central urban area; and (b) El Pache sector. A preliminary risk assessment was performed using Hg concentrations from previous studies. The human health risk in the workplace scenario showed unacceptable levels of non-carcinogenic risk in rainy and dry seasons. Hazard Quotient (HQ) showed that 75% of the workers receptors were exposed to harmful effects on the nervous, digestive, respiratory, and immune systems. Regarding the residential scenario, no human health risk is presented. The study provides information on the levels of risk to the health of the inhabitants living in mining communities to improve public management strategies to minimize risks.

Keywords: hazard quotient; probabilistic risk; mercury pollution; gold mining.

Mercurio y salud humana: caracterización probabilística del riesgo en una de las zonas mineras auríferas más antiguas del Ecuador

Resumen
La minería del oro artesanal y a pequeña escala contribuye de forma importante a las emisiones globales de Hg. El presente estudio evalúa el riesgo probabilístico para la salud humana de dos grupos de receptores (residentes y trabajadores) de la zona minera de Portovelo por exposición al mercurio (Hg) en el aire en dos sectores: (a) la zona urbana central; y (b) el sector de El Pache. Se realizó una evaluación preliminar del riesgo utilizando concentraciones de Hg de estudios anteriores. El riesgo para la salud humana en los lugares de trabajo mostró niveles inaceptables de riesgo no cancerígeno en las estaciones lluviosa y seca. El cociente de peligrosidad (HQ) mostró que el 75% de los trabajadores receptores estaban expuestos a efectos nocivos en el sistema nervioso, digestivo, respiratorio e inmunitario. En el escenario residencial, no se presenta ningún riesgo para la salud humana. El estudio proporciona información sobre los niveles de riesgo para la salud de los habitantes de las comunidades mineras con la finalidad de mejorar las estrategias de gestión pública para minimizar los riesgos.

Palabras clave: cociente de peligrosidad; riesgo probabilístico; contaminación por mercurio; minería del oro.

1. Introduction

In many rural areas in developing countries, it is common to see how workers are not skilled to extract and process gold in a technical and environmentally friendly way [1], mainly due to poor and rough mechanization as well as social and financial barriers in the artisanal and small-scale gold mining (ASGM) [2].

Anthropogenic activities have nearly increased the amount of atmospheric mercury (Hg) threefold, which is rising at a rate of 1.5% per year [3]. ASGM is considered as the largest contributor to global human Hg emissions [4]. It is estimated that worldwide,

How to cite: Paz-Barzola, D., Escobar-Segovia, K., Salgado-Almeida, B., Goyburo-Chavez, C. and Moreno-Chavez, J., Mercury and human health: probabilistic risk characterization in one of the oldest gold mining areas in Ecuador. DYNA, 90(227), pp. 110-116, July - September, 2023.

inadequate mining practices release about 400 tons of Hg [5]. Despite, the use of Hg is illegal, it's still used frequently in developing regions, including areas of Latin America [4], [6]. Studies allude the presence of heavy metals (specially Hg) in mining areas to the processes of mineral extraction activities [7]. Although the environmental destination of Hg released during artisanal gold mining remains undetermined, there is strong evidence to date of Hg pollution in air, soil and sediments related to artisanal gold mining workplaces and urban areas where gold is traded and refined [8-9].

Mercury occurs naturally in three primary forms: elemental (Hg^0), inorganic and organic. Hg exists at workplace environments as a result of its extensive use for gold processing, mostly in the informal and illegal sector. Elemental mercury has been employed in ASGM to isolate gold from other non-target minerals by amalgamation from placer ores or mineral deposits [8]. When the Hg-Gold amalgam is formed the gold particles are separated by adding nitric acid or by roasting [10]. Usually ASGM miners do the second case and roast the amalgam in open-air pans [11]. In this way, the Hg vapors emitted are inhaled by the miners and by the inhabitants of the mining communities as well [12]. Thus, most human exposure pathways to mercury are the inhalation and ingestion [13].

An example of mercury pollution and exposure occurs in the Amazonian ecosystems, where there is evidence of methylmercury (MeHg) contamination. This situation has aggravated the health risk of indigenous people and gold prospectors [14]. Another case takes place in Colombia, where miners have been forced to extract the gold ore and ship it to a center located in an urban area to be processed without any type of filtering system [15]. As a result, both rural and urban areas have been affected due to the lack of measures to prevent and reduce contamination from mineral extraction.

In Ecuador there is a long tradition of artisanal and informal mining activities, associated with environmental impacts and socio-environmental conflicts because of heavy metal contamination [16–19]. Therefore, with the purpose of absolutely banning the use of Hg in mining activities, the Ecuadorian government signed the Minamata Convention on Mercury in October 2013 and implemented the nation-wide initiative called "Zero Mercury",

Nonetheless, mercury remains used to extract gold despite the fact that its use can pose serious health risks to miners [20-21], due to its relative accessibility and affordability. In this line, in 2015, 65% of the mineral processing plants in Zaruma-Portovelo mining area, were still performed amalgamation, using almost 1.63 tonnes per year of Hg according to [22-23]. Velásquez et al. [23], estimated that the treatment plants released 1.5 tons of total Hg annually, of which approximately 70% was due to evaporation during amalgam combustion and 30% was discharged to rivers in the form of Hg-contaminated waste. Furthermore, Gonçalves et al. [22] revealed that processing centers in Zaruma-Portobelo released 1.9 million tonnes per year of tailings, including 222 kg $year^{-1}$ of Hg, and that the burning of gold amalgams released 303 kg $year^{-1}$ of Hg to the atmosphere. Miners in the Zaruma-Portovelo area burn amalgams in their own residences, which represents a potential health risk for inhabitants mainly for children [22]. The main route of direct exposure to Hg was through inhalation, which implicates severe damage to population health.

Hg in vapor form can easily enter the circulatory system and remain for years [24]. Hg can also remain in the environment, bioaccumulate in the food chain and cause toxicity to the central and peripheral nervous system of humans despite low levels of exposure [25-27]. The effects of mercury on health rely on the chemical state of Hg [28]. Chronic and prolonged exposures may cause neurotoxicity that affects the central nervous system. Most vulnerable and affected organs are the brain and kidneys [29]. Physical disorders such as headache, body aches, memory loss and muscle cramps are common symptoms among miners directly exposed to Hg [30]. In this sense, the World Health Organization (WHO) considers Hg a potentially harmful trace element and one of the chemicals of greatest concern [27], [31].

In this regard, this research aimed to assess the human health risk among population living in the mining area of Portovelo due to exposure of Hg in the air. The results obtained from this work present information on human health risk levels in mining areas, contributing insights to improve public management strategies in order to minimize health risks for the inhabitants living in the mining communities.

2. Materials and methods

2.1 Study area

The study area corresponds to one of the oldest gold mining districts, Zaruma-Portovelo [6], exploited since pre-colonial times. It is located in the El Oro Province, in the Puyango-Tumbes River Basin (Fig. 1). The district is considered one of the traditional small-scale gold mining areas in Ecuador, with milling plants capable of processing up to 300 tons/day of ore [32]. Gold ore processed in Zaruma-Portovelo comes from mines of different provinces from southern Ecuador and Northern Peru [22].

This study focuses on the risk assessment of the El Pache sector, where 27 benefit processing plants are located, and in the central urban area of Portovelo city[33].

2.2 Data collection and analyses

Because the use of Hg in mining is an illegal activity, which is often carried out in residences, there is no updated information on the concentration of Hg in air. Therefore, for a preliminary risk evaluation, the concentration of Hg used in this study was obtained from González-Carrasco et al. [34]. The sample treatment and analytical protocols from the above-mentioned study can be consulted therein. Concentration of Hg in air was evaluated in two sites of Portovelo: a) the central urban area; and b) El Pache sector.

In order to compare the risk outcomes variations according to the season, the data analyzed correspond to the dry season and the rainy season. Information from Table 1 was used to determine the probability distributions of Hg concentration in air, and then, to evaluate the probabilistic human health risk.

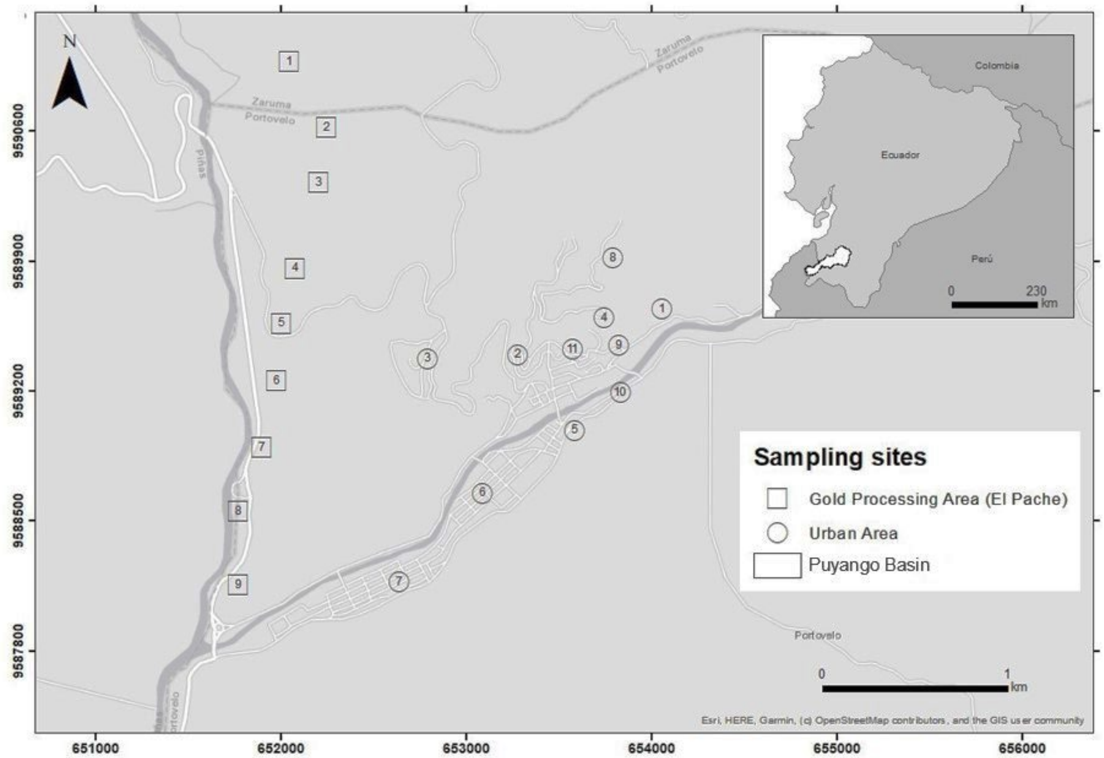

Figure 1. Location of evaluated sites in the urban area and the mining processing area.
Source: Authors

Table 1.
Concentrations of Hg in outdoor air and workshops' atmospheres in the study area

Place	Dry season	Rainy season
Outdoor air - Portovelo urban area	574.2 ± 72.8 ng m^{-3}	214.0 ± 43.7 ng m^{-3}
Workshop atmosphere - Pache sector	3,699.5 ± 1,225.3 ng m^{-3}	2, 356.7 ± 1,807.6 ng m^{-3}

Source: Authors

2.1 Probabilistic human health risk

The human health risk assessment was carried out according to the Environmental Protection Agency guidelines for non-carcinogenic health effects [35], using a probabilistic approach. In this method, statistical sampling techniques are used to obtain probability distributions of the variables [36-37].

Fig. 2 shows a graphical description of the human health risk assessment process carried out in this study. Two different exposure scenarios were considered: a) miner workers (adults) with occupational exposure; and b) residents adults of the urban central area of Portovelo. The exposure pathway included in the risk assessment was inhalation of elemental Hg because it is the dominant route of concern [1].

To quantify the potential risk, HQ, was estimated using eq. (1) as the ratio between the exposure concentration in air (C_{air}: mg m^{-3}) and its inhalation reference concentration (RfC: mg m^{-3}). The C_{air} was calculated using eq. (2) and the RfC (0.0003 mg m^{-3}) was gathered from the Risk Assessment Information System (RAIS) website [38].

Following United States Environmental Protection Agency (USEPA)'s standard recommendation, when HQ> 1 the non-carcinogenic risk is considered unacceptable [39]. The equations to compute C_{air} and HQ were implemented in R language.

$$\mathrm{HQ} = \frac{\mathrm{C_{air}}}{\mathrm{RfC}} \tag{1}$$

$$\mathrm{C_{air}} = \frac{\mathrm{C} \times \mathrm{EF} \times \mathrm{ET} \times \mathrm{ED}}{24 \times \mathrm{AT}} \times 10^{-6} \tag{2}$$

where C: concentration of Hg in air (ng m^{-3}), EF: annual exposure frequency (day $year^{-1}$), ET: exposure time (hours day^{-1}), ED: lifetime exposure (30 years), AT: averaging time (10,950 days).

The probability distributions of the mean concentration of Hg in air were determined by re-sampling 1,000 times. The probabilistic human health risk assessment was conducted in R free software. For Hg vapor inhalation, it was assumed that the average mine worker spends 8 hours per day and 300 days per year exposed to the workplace [12]. On the other hand, the average resident was assumed to spend 6 hours day-1 and 350 days year-1 exposed to the outdoor air.

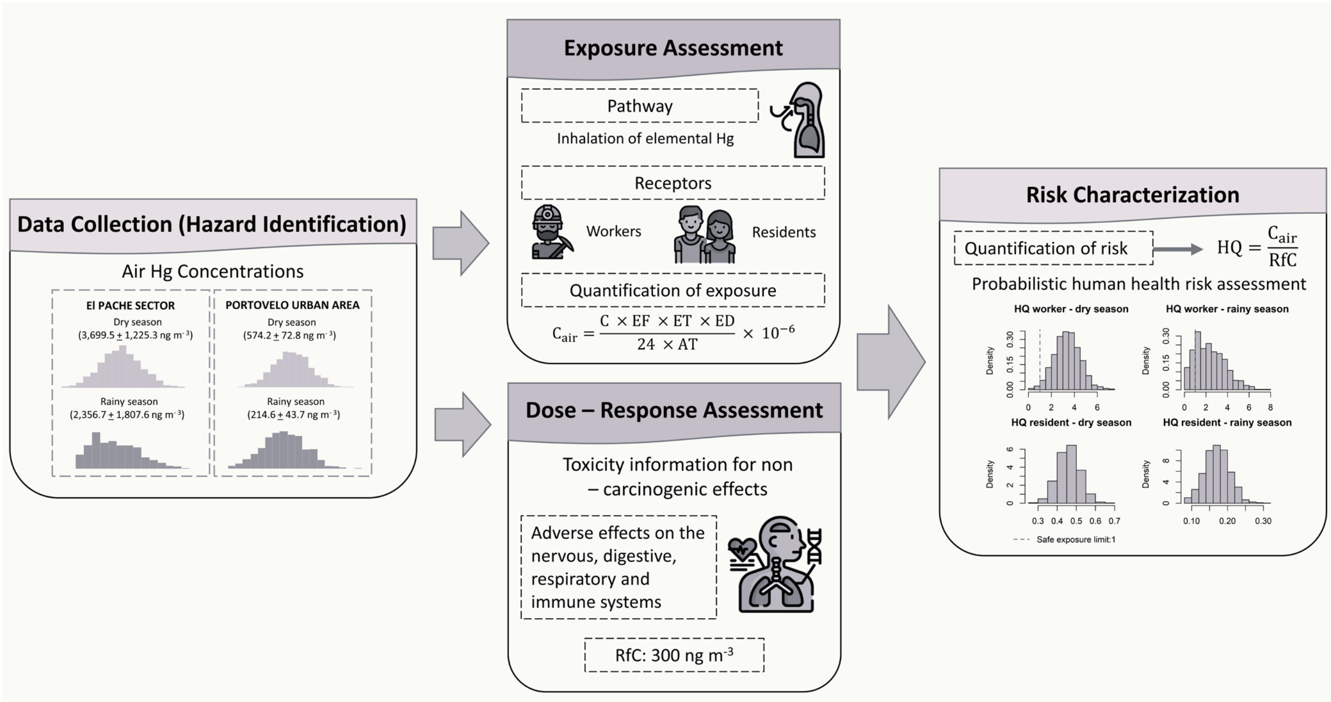

Figure 2. Graphic scheme of the human health risk assessment process.
Source: The authors

1. Results and discussion

1.1 Probability distributions of Hg concentration in air

The probability distributions of the concentration of Hg in air for the Pache and the urban area of Portovelo for both seasons are shown in Fig. 3. The range of Hg exposure concentration for the urban area in the dry season was between 315.68 and 845.13 ng m^{-3}, while in the rainy season ranging from 101.13 to 379.34 ng m^{-3}. In the Pache sector, Hg concentration ranging from 162.08 to 8152.65 ng m^{-3} in the dry season and 4.60 to 8291.92 ng m^{-3} in the rainy season. As expected, workers of the Pache sector are exposed to inhalation of higher concentrations of Hg than the residents of the urban area. Nevertheless, the average concentrations of Hg for both workplace and residential scenario exceed the international minimal risk levels for Hg in air (200 ng m^{-3}) recommended by the Agency for Toxic Substances and Disease Registry [40].

In addition, in the field research it was possible to observe the natural resources degradation around mining sites as a result of the illegal ASGM processes. Despite the Hg ban in mining operations in Ecuador since 2010, the substance is distributed and used in the sector through the black market [22]. An important finding in the field is the potential risk of the Zaruma – Portovelo district for the ecosystem and the population, due to that it's one of the areas where the largest amount of extracted mineral from the country is processed. The Hg problem is not only reported in Ecuador but also globally in other countries such as Philippines [41], Indonesia [42], Niger [43], Nicaragua [44], Ghana [27] and Colombia [15].

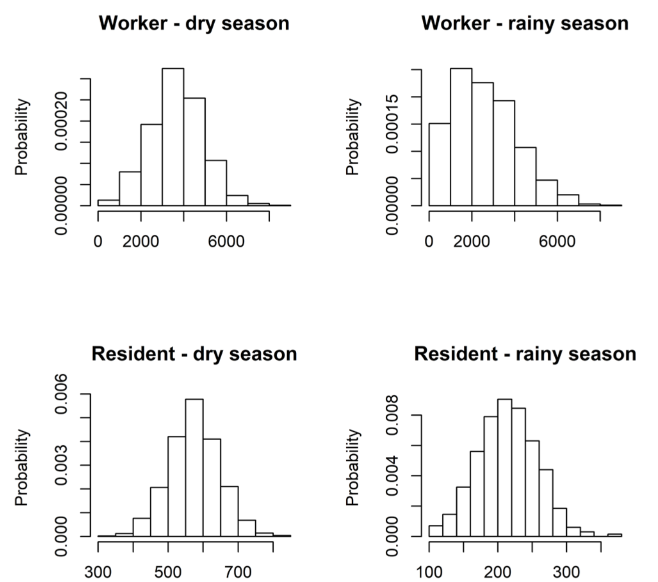

Figure 3. Probability distribution of Hg in air for both residential and workplace scenario.
Source: The authors

1.2 Probabilistic human health risk assessment

The outcomes for the probabilistic human health risk assessment by both workplace and residential scenario are given in the Table 2.

Table 2.
Statistical summary of the HQ. Values in bold exceed the safe exposure threshold

	Worker		Resident	
	Dry Season	Rainy Season	Dry Season	Rainy Season
Min.	0.148	0.004	0.252	0.080
p25	2.616	1.227	0.423	0.147
Median	3.368	2.177	0.459	0.170
Mean	3.378	2.354	0.460	0.171
p75	4.107	3.301	0.499	0.195
Max.	7.445	7.272	0.675	0.303

Source: The authors

The human health risk in workplace scenario showed an unacceptable level to non-carcinogenic risk in both rainy and dry season, with the highest values was detected in dry season.

Fig. 4 shows the histograms of Hazard quotient (HQ) for worker for both dry and rainy season. HQ values were above the safe exposure limit for the 25th percentile, indicating that 75% of the receptors in the studied zone were exposed to harmful effects on the nervous, digestive, respiratory and immune systems [45]. These values are like those reported by Pavilonis et al. [1] in two mine processing centers located in the Bolivian Andes where it was found Hg vapor levels 30 times higher than the EPA reference concentration (300 ng m^{-3}).

In the same way, Castilhos et al. [45] reported a high potential risk for artisanal gold miners from Sao Chico (Brazilian Amazon) due to the Hg vapor inhalation and MeHg exposure; miners presented high concentrations of Hg in blood and urine partially related to Hg vapor exposure.

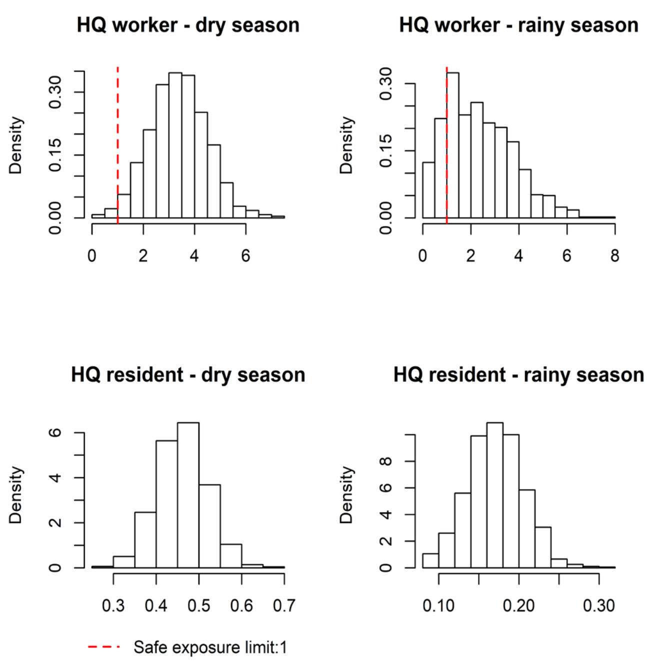

Figure 4. Histograms of Hazard quotient (HQ) for workers and residents in dry and rainy season.
Source: The authors

Furthermore, in Burkina Faso (West Africa) 78% of the Artisanal and Small-scales Gold Mining workers who are present at the burn of Hg amalgams exceed the permissible exposure limit of 100,000 ng m^{-3} established in occupational standards (OSHA) [47].

In addition, research conducted in 19 different countries in South America, Asia and Africa has shown concentrations in the hair and urine of people who live within or near artisanal gold mining sites to be well above the values recommended by the WHO [48].

Regarding the residential scenario, no risk to the human health of the residents exists. The HQ values were below the safe exposure limit of 1 (Fig. 4). Similar results were reported by Jiménez-Oyola et al. [48] in a study conducted in the Hg mining district of Almadén (Spain), for the inhalation of indoor and outdoor air. On the other hand, in the Northeast of Antioquia (Colombia), air Hg levels are extremely high, with values ranging from 300 ng Hg m^{-3} to 1 million ng Hg m^{-3} inside gold shops, and around 10,000 ng Hg m^{-3} in residential areas [15] .

2. Conclusions

The present research is a preliminary report on the health risk of Hg to residents and workers in the Portovelo mining area. Probabilistic risk results showed that Hg-exposure in workplace scenario showed an unacceptable level to non-carcinogenic risk in both rainy and dry season; 75% of workers are exposed to developing adverse effects on their health due to continued exposure to Hg. Otherwise, in the residential scenario, the risk remained within acceptable limits in the dry season and the rainy season. However, risk assessment is recommended for vulnerable people such as children and pregnant women. In addition, adequate regulatory strategies and continuous environmental monitoring must be implemented to eradicate the use of Hg and therefore reduce risks to human health.

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D.M. Paz-Barzola, received the BSc. Eng. in Engineering in Geology in 2022, since 2023 she has been working for the Escuela Superior Politécnica del Litoral, Ecuador, as a research technician of the Faculty of Engineering in Earth Sciences, her research areas include: geology and environmental sciences.
ORCID: 0000-0002-9966-6632

K.F. Escobar-Segovia, is a BSc. in Geological Engineering in 2009, from the Escuela Superior Politécnica del Litoral, Ecuador., MSc. in Productivity and Quality Management in 2011, from the Escuela Superior Politécnica del Litoral, Ecuador. He specializes in biostatistics, occupational health, geology, mining and petroleum.
ORCID: 0000-0003-1278-7640

B.J. Salgado-Almeida, lecturer and director of community projects at the Geoscience Faculty from the Escuela Superior Politécnica del Litoral (ESPOL), Ecuador, since 2021. His research areas of interest are: environmental risk assessment, mining waste management, mining environmental liabilities and engineering education.
ORCID: 0000-0002-7188-8081

C.S. Goyburo-Chávez, is a BSc. Eng. in Engineering in Mine in 2022, since 2023 she has been working for the Escuela Superior Politécnica del Litoral, Ecuador as a laboratory technician of the Faculty of Engineering in Earth Sciences, her research areas include: treatment water, mining and environmental sciences.
ORCID: 0000-0003-2314-5619

J.R. Moreno-Chavez, is a BSc. Eng. in Mining Engineer in 2015, from Escuela Superior Politécnica del Litoral, Ecuador, and MSc. in Applied Science in Mining Engineering in 2018, from the University of British Columbia. He professionally works in subway mining and open pit mining.
ORCID: 0000-0001-7955-4679