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Levels of radium-226 in the rainbow trout, macroinvertebrates-substrates and water adjacent to the mining concession Loma Larga, Azuay-Ecuador

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ABSTRACT

Mineral processing has been identified among the twelve main industrial processes that must be under control due to the expose to natural radioactive sources. The modification and, generally, the increase in the concentration of Naturally Occurring Radioactive Materials (NORM) in the earth's crust cause an imbalance in the ecosystem. Imbalance that affects the fauna, which are able to bio accumulate these radioisotopes and introduce them into the trophic chain. The main purpose of this work was to establish a radio-ecological baseline. For this purpose, the concentration levels of ^{226}Ra in rainbow trout, in macro invertebrate matrices-substrates and water from the Irquis and Portete rivers were evaluated. A study area adjacent to the Loma Larga mining concession was selected. The measurements were made using the Lucas cells coupled to a Pylon AB6A counter and the "Can Technique", using LR-115 passive detectors. The results indicated that rainbow trout bio accumulated in bones and organsa mean of 94.7% of the total ^{226}Ra detected, which values varied from 4.69 to 2.46 Bq/kg, while macro invertebrate-substrate matrices showed a concentration that ranged between 0.7 and 1.3 Bq/kg in the Irquis River, while in the Portete River was between 1.1 and 19.3 Bq/kg. The water samples, in the sampling points of the Irquis River, showed concentrations that ranged between 37.3 and 119.1 mBq/l and in the Portete River between 43.5 to 146.4 mBq/l.

Keywords: ^{226}Ra, rainbow trout, pylon AB6A, LR-115, mining.

Niveles de radio 226 en la Trucha Arcoíris, macroinvertebrados-sustratos y agua adyacente a la concesión minera Loma Larga, Azuay, Ecuador

RESUMEN

El procesamiento de minerales se ha identificado entre los doce principales procesos industriales que deben estar bajo un control a la exposición de fuentes radiactivas naturales. La modificación y, por lo general, el aumento de la concentración de NORM en la corteza terrestre causa un desequilibrio en el ecosistema. Desequilibrio que afecta a la fauna, la cual es capaz de bioacumular estos radioisótopos e introducirlos en la cadena trófica. La finalidad principal de este trabajo fue establecer una línea base radio-ecológica; para ello se evaluaron los niveles de concentración de ^{226}Ra en la trucha arcoíris, en matrices macroinvertebrados-sustratos y el agua de los ríos Irquis y Portete, ubicados en la provincia del Azuay, Ecuador. Se seleccionó una zona de estudio adyacente a la concesión minera Loma Larga. Las mediciones se realizaron haciendo uso de las celdas Lucas acopladas a un contador Pylon AB6-A y la "Can Technique", utilizando detectores pasivos LR-115. Los resultados indicaron que la trucha arcoíris bioacumuló el 95.2 % del total del ^{226}Ramdetectado, en los huesos y órganos, las matrices macroinvertebrados-sustrato presentaron una concentración que osciló entre 0.7 y 1.3 Bq/kg en el río Irquis, mientras que en el río Portete resultó entre 1.1 y 19.3 Bq/kg. Las muestras de agua en los puntos de muestreo del río Irquis, reflejaron una concentración variable entre 37.3 y 119.1 mBq/l y en el río Portete entre 43.5 a 146.4 mBq/l.

Palabras clave: Radio 226; trucha arcoíris; pylon AB6-A; LR-115; minería.

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Introduction

The US Environmental Protection Agency (US-EPA) acknowledged in 1987 that the problems related to mining waste can be described as surpassed only by global warming and depletion of stratospheric ozone in terms of ecological risk. The release to the environment of mining waste can cause a profound, and generally irreversible, destruction of ecosystems (Durand, 2012).

According to the International Atomic Energy Agency (IAEA, 2006), any mining operation or activity in which a mineral is involved has the potential to increase the effective dose that a person can receive from natural sources as a result of exposure to Naturally Occurring Radioactive Materials (NORM) contained in industrial waste.

The aquatic systems among others contain NORM related to the human activity, and therefore are responsible for a fraction of the dose that the human being receives from the environment. The global average internal dose due to radionuclides other than radon and its decay products is 0.29 mSv/y of which 0.17 mSv/y comes from ^{40}K, 0.12 mSv/y comes from the uranium and thorium series, and 12 µSv/y come from ^{14}C (United Nations Scientific Committee on the Effects, 2008).

The natural radioactivity of water comes essentially from the ^{40}K and the disintegration series of the ^{238}U and ^{232}Th. For groundwater, the main radioactive isotopes found are those mentioned above, and their daughters; within the hazardous elements are found ^{226}Ra, ^{228}Ra and ^{222}Rn, also their progeny. These elements when inside the body move to the bone tissue and concentrate (Cox, 1993). The prolonged exposure can cause bone cancers and other bone irregularity; their concentrations vary depending on the geochemical history of the water and the industrial activities of the environment.

The bioaccumulation of natural or artificial radionuclides in aquatic organisms is the focus of radio-ecological studies, because they act as an additional source to the radiation dose; of particular importance for the aquatic flora and fauna that form food chains used by the population (Stewart et al., 2008).

Environmental problems associated with NORM and solid mineral mines occur during the process of drilling, leaching, handling, storage, transportation of minerals and related equipment (Innocent et al., 2013). In Ecuador the mining is very active, because the regions have several minerals, such is the case from the southern region of Ecuador, known as Austro, where we found an important variety of minerals such as gold, silver, copper, lead and zinc (Rea-Toapanta, 2017). Therefore, it is important to carry out baseline studies regarding radioactive elements, such as ^{226}Ra, in different media near the mines, in order to be able to calculate the radiological impact, considering that the exploitation mining activity in most cases significantly alters the natural levels of these. As expected, they also increase the concentration levels of the NORM in the biota of the rivers.

In this study, the concentration levels of ^{226}Ra were evaluated in three matrices, the rainbow trout (*Oncorhynchus mykiss*), the macroinvertebrate and the water of the Irquis and Portete rivers. The rainbow trout is an ichthyic species of the Salmonidae family, present in South America, where its habitat is generally located in cold waters of rivers and lakes, tolerating a temperature range between 0 to 30 °C, are a very resistant species, capable of displacing endemic species. The macroinvertebrate are bio indicators of the water quality with biological indexes, giving a significant importance to these matrices.

Materials and methods

The technique used in this study for the quantification of the content of ^{226}Ra in the organic matrix (fish) was the *Can Technique* making use of solid-state nuclear track detectors (SSNTDs), specifically the LR-115 detector (Mohamed Abd-Elzaher, 2012).

Selection of sampling areas

This investigation was carried out in the mining project called Loma Larga which covered an area of 7,960 ha, where three concessions operate, known as Cerro Casco, Cristal and Río Falso. It is located in the south of Ecuador 15 km northwest from the town called Girón and 30 km southeast of Cuenca City, at an altitude between 3 500 and 3 900 mamsl (Jarrín-Jurado & Morán-Reascos, 2017). The area of study was Río Falso concession with an area of 3 168 ha, located in the parishes Baños, Victoria del Portete, San Gerardo, Chumblín, San Fernando and Girón, belonging to the province of Azuay (Fig. 1) (INV Minerales Ecuador SA, 2016).

The physico-chemical properties of the site analyzed in this study were characterized by presenting strata of andesitic porphytic lavas of very fresh acicular pagioclase and amphibole, which are volcanic or igneous extrusive rocks containing between 52% and 63% silica (SiO2), in addition 5% iron (FeO) and 7% calcium (CaO), on average. In relation to hydrology, the drainage network is a radial dendritic type constituted by a series of streams. Drainage to the southeast occurs through two main rivers, the Irquis and the Portete. These effluents, being also transport vectors of matter of the mining exploitation, have been analyzed in this study to determine the ^{226}Ra levels.

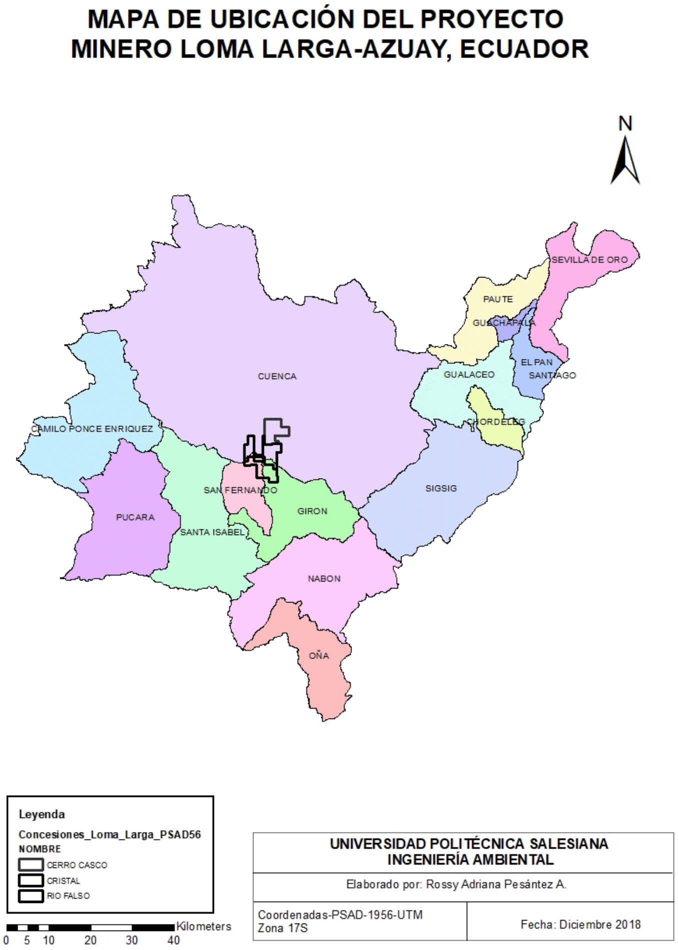

Figure 1. Location map of the Loma Larga mining project, in the Province of Cuenca, Ecuador. Coordinates-PSAD-1956-UTM, Zone17S, (December 2018).

In this work, sampling points for the rainbow trout, macroinvertebrate matrices and water, were selected as established by the World Health Organization (WHO, 2006). It considers that the number of sampling points depend on the characteristics of the water quality under analysis. For components whose concentrations tend to vary during distribution, the number of strategic points should be three, distributed along the region of studies (See Table 1). In this work these were situated in the headwater of the river, in the middle course of the region of interest (adjacent to the mining activity) and at the exit of the concession (See Fig. 2).

Water samples

Collection of surface water samples

Obtaining water samples was carried out by the immersion method, following the protocol established for the Pylon AB6 detector (Pylon Electronics Inc., 2015). To take the liquid sample, a 250 ml glass container was used, ensuring the absence of bubbles, which was hermetically sealed and each of the containers identified with the necessary data.

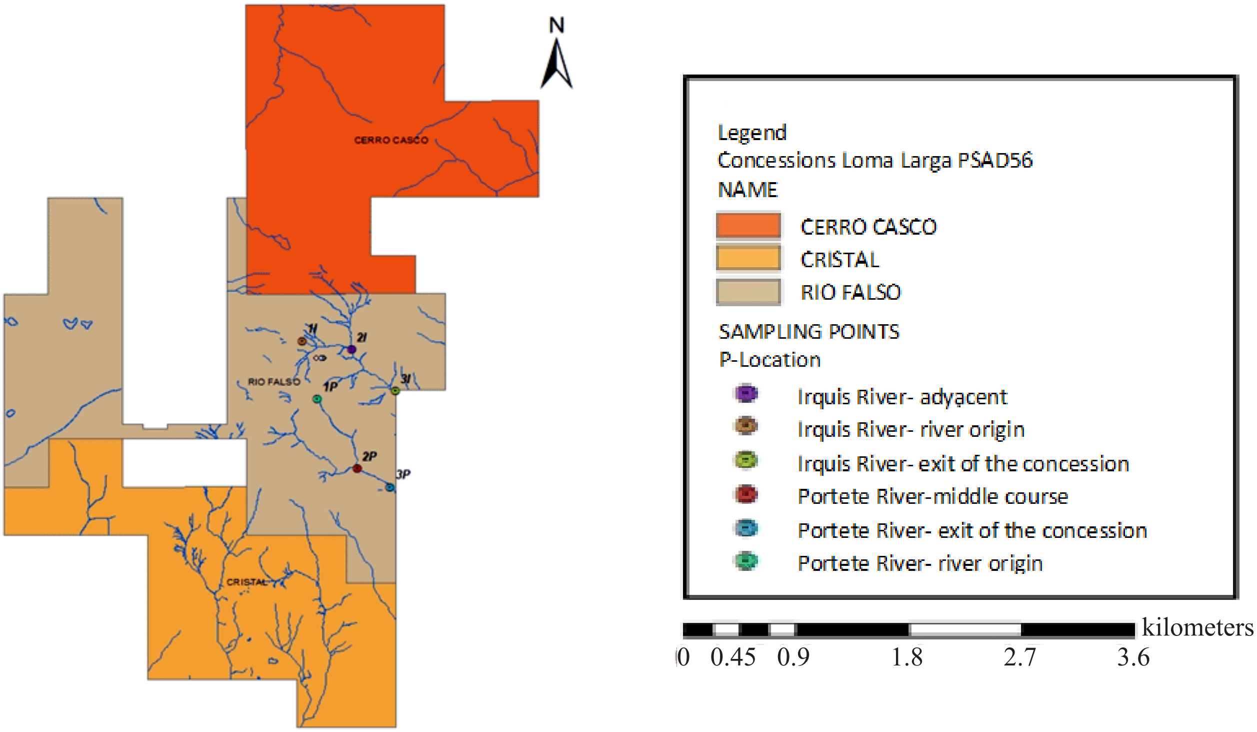

Figure 2. Map of the location of the sampling points in the Irquis and Portete rivers. Coordinates-PSAD-1956-UTM, Zone17S (December 2018).

Table 1. Coordinates of the sampling points in the study area where the investigation was conducted.

CODE	COORDINATES-PSAD-1956-UTM-Zone 17S	SAMPLING AREA	RIVER
1I	698141 9664023	-	Irquis-headwater
2I	699137 9663840	Quebrada high Quinuahuaycu	Irquis-adjacent to entrance of the mine
3I	700018 9662981	Quebrada Quinuahuaycu	Irquis-exit of the concession
1P	698448 9662815	-	Portete-headwater
2P	699249 9661368	Quebrada Kalluancay	Portete-middlecourse
3P	699916 9660990	Quebrada Kalluancay	Portete-exit of the concession

Measurement of ^{226}Ra activity concentrations in water.

To measure the ^{226}Ra activity concentrations in the water sample, the ^{222}Rn was evacuated from the latter, flowing nitrogen through it for a period of approximately 15 minutes; the samples were stored for a period of 3.8 days, after this time, they were degassed, transferring the gas to a Lucas cell model 600-A, according to the Pylon AB6 protocol (Pylon Electronics Inc., 2015).

The formula used to calculate the Ra activity concentrations in water was:

$$Conc_{Ra} = \frac{NCPM}{nAE * CF * Eff_c * V_s * A_2 * Eff_D * G}\left(Bq/l\right) \qquad (1)$$

Where, *NCPM* is the number of counts per minutes, *nAE* is the number of alpha emissions (for ^{222}Rn is 3), *CF* is the conversion factor (for this calculation is 60 dpm to dps), Eff_c is the cell efficiency (0,745 cpm/dpm), V_s is the sampling volume (0,19 l), Eff_D is the ^{222}Rn extraction efficiency (for the Lucas cell 600A is 90 ± 5%) and *G* is the growth correction factor.

The growth correction factor is calculated by the following equation,

$$G = 1 - e^{-\left(\lambda T_{Delta1}\right)} \qquad (2)$$

Where,

$$T_{Delta1} = t_1 - t_{0A}\left(min\right)$$

And t_{0A} is the time in which the sample was initially degassed, t_1 is the time in which the sample was degassed for measurement.

After each measurement the Lucas cells were flushed with nitrogen, at a flow rate of 5 l/min for 3 minutes, in order to prevent contamination and reduce background count buildup that could affect future measurements. The cells were stored for a minimum of 24 hours, before being reused again.

Biological samples.

Collection of samples.

The macro invertebrates and the substrate in which they live, such as sediments, algae and macrophytes, were collected with Surber sampler nets, located at the three selected points of each river. The rainbow trout were collected at the midpoint of the Irquis River, adjacent to the mine mouth of the concession denominated Río Falso by gillnets.

Preparation of the trout samples

Each of the individuals was dissected, preparing with them two matrices, one of muscle tissue, and another of bones and organs. Organic matrices of both macro invertebrates and rainbow trout were dehydrated in an oven at 110 °C for 24 hours. And then crushed to obtain a finer granular consistency for favoring the exhalation rate of ^{222}Rn; after this the weighing was carried out.

Taking into account that the radon exhalation of ^{222}Rnfrom the matrices is conditioned by the physical properties of it, among which the porosity is one of the most important, and considering that the porosity is not controlled by grain size, as the volume of between-grain space is related only to the method of grain packing, but the emanation of radon depends on the grain size and the shape, because they determine in part how much radium is near enough to the surface of the matrix grain to allow the newly formed radon to escape into a pore space, we considered appropriated to make a fine matrix to reduce the probability of ^{222}Rn to remain embedded in the grain.

Content measurement of ^{226}Ra in macro invertebrates-substrate matrices.

The samples of macro invertebrates and rainbow trout previously processed were placed in PVC cylindrical containers, of 10 cm high and 7 cm diameter in which a passive detector was positioned. In this case, 1.5 cm x 2 cm LR-115 type II (Kodak-Pathe, France) detectors were used, located in the internal upper part of the container, in such a way that the sensitive part of the detector is oriented towards the sample, at approximately 4 cm from the sample surface (Khan et al., 1992).

In order to avoid the registration of alpha particles emitted in the decay of the isotopes of radon, prior the ^{226}Ra and the ^{222}Rn reached the secular equilibrium within the cylindrical containers, the detectors were covered for four weeks with a metal foil that was held in place with the help of a small magnet on the outside of the chamber. After that time, the metal sheet was released, exposing the detector for the following 60 days (Sonkawade et al., 2008). After this time, the detectors were removed and etched, placing each detector in a 2.5 N NaOH solution for 90 minutes, at a temperature of 60 °C in a laboratory water bath (Zubair et al., 2012). The nuclear tracks density was quantified with the help of an optical microscope with a magnification of 400X.

The content measurement of ^{226}Ra was calculated by the following equation.

$$C_{Ra} = \frac{\rho * h * A}{K_{Rn} * T_{eff} * M}\left(Bq/kg\right) \qquad (3)$$

Where, $\rho$is the radon track density, h is the distance between the surface of the sample and the detector in meters, A is the cross-sectional area of the container in m^{-2}, K_{Rn} is the calibration factor of LR-115 plastic track detector (0.033 track cm^{2} d^{-1}/ Bq m^{-3}), T_{eff} is the effective exposure time, and M is the mass of the sample in kg.

The effective exposure time can be calculated using the formula

$$T_{eff} = t - \frac{1}{\lambda}\left(1 - e^{-\lambda t}\right) \qquad (4)$$

Where, t is the actual exposure time and λ is the decay constant for ^{222}Rn.

Results

The ^{226}Ra concentrations in the matrices of macro invertebrates-substrate of the Irquis River ranged from 0.90 ± 0.07 Bq/kg to 1.30 ± 0.08 Bq/kg, while in the Portete River the minimum concentration was 1.10 ± 0.08 Bq/kg and the maximum of 19.30 ± 0.08 Bq/kg (Fig. 3).

Because of the difficulty to fish in others sampling points along the rivers, only two sampling were done at only one sampling point, the midpoint of the Irquis River analysis region (adjacent to the mine mouth); in the first sampling the average ^{226}Ra levels in the muscle tissue of the rainbow trout was 0.17 ± 0.04 Bq/kg, and the corresponding to the bone-organs matrix was 4.69 ± 0.51 Bq/kg. While in the second sampling the muscle tissue concentration was 0.19 ± 0.03 Bq/kg, and the bone-organs sample showed a ^{226}Ra concentration of 2.46 ± 0.31 Bq/kg. All values correspond to dry weight (see Fig. 4). It is worth to noting that, the difference between the ^{226}Ra concentration in the bone-organs matrices in the first and second sampling was 47.74 %.

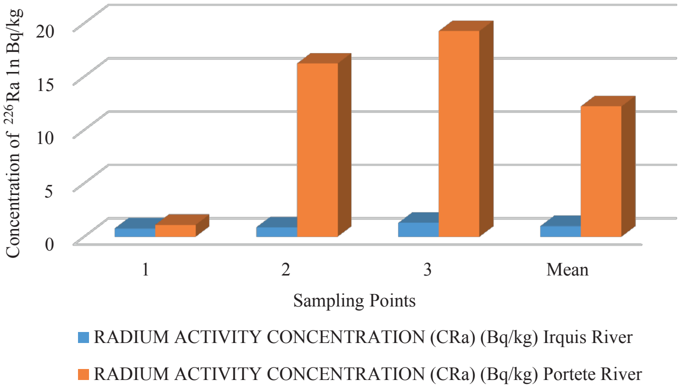

Figure 3. Concentration of ^{226}Ra in macro invertebrates of the Irquis and Portete rivers (1 corresponds to the source of the two rivers, 2 represents the midpoint of both the Irquis and Portete rivers, and 3 are the sampling points at the exit of the concession)

The original purpose of the two samplings was to obtain a better statistic, but that noticeably greater difference between the values of the ^{226}Ra concentrations in the bone-organs matrices, makes a comment on it worthwhile, even though it is not part of the main objective of this work. The bioaccumulation factor depends on different factors, such as the duration of exposure, the rate of metabolism, the age and the diet of the species, among others.

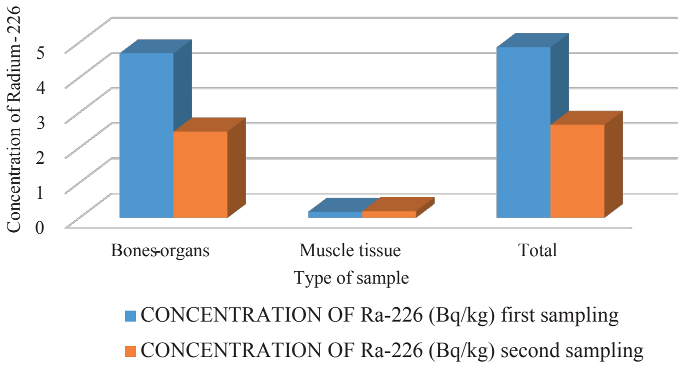

Figure 4. Concentration of ^{226}Ra in the rainbow trout of the Irquis River in the area adjacent to the mine entrance.

The variation of the radium levels in both rivers, have a similar behavior, they increase as the measurements are done downstream. The levels of radium in the headwater of the Irquis River and the Portete River were 39.3 and 57.5 mBq/l respectively, while in the lowest points downstream were 119.1 and 146.39 mBq/l for the Irquis and Portete River respectively (see Fig. 5).

Discussion

The IAEA (2014) reports that in uncontaminated surface waters the concentration range of radioisotope ^{226}Ra in fish (whole body) ranges between 0.68 and 1.12 Bq /kg (fresh weight). Swanson S. M. (1983) reports concentrations of ^{226}Ra in fish, among them trout, in uncontaminated surface water, with levels between 0.296 and 3.400 Bq/kg in dry weight, values similar to those obtained in this investigation, in which was obtained an average concentration for rainbow trout of 3.75 Bq/kg (dry weight).

The concentration levels of ^{226}Ra in the bone-organ matrices were higher than those in the muscle tissue matrices, this is due to the fact that both calcium and radium belong to the same group in the periodic table, so both elements have very similar chemical properties, and that is why in human and animal organisms, radium is assimilated as calcium and deposited in the bones.

Clulowet al. (1998), in their study in lakes adjacent to non-industrialized zones, found levels of ^{226}Ra in the bones of lake trout (*Salvelinus namaycush*),

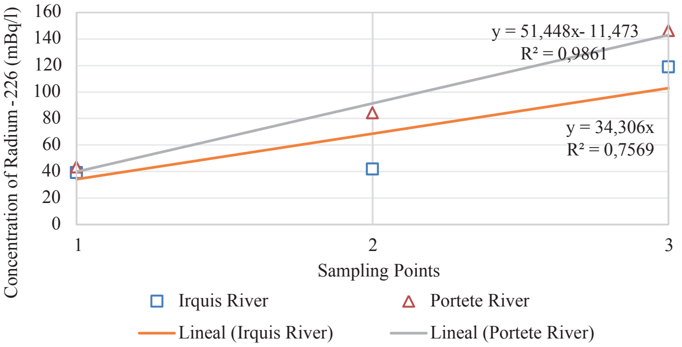

Figure 5. Average concentrations of ^{226}Ra in the Irquis and Portete rivers in the three strategic points (1 corresponds to the point of the source of the rivers, 2 is the adjacent point of the mine entrance in the Irquis River and the middle point of the Portete River, and 3 is the point at the exit of the concession in each river respectively)

with average values less than 20 Bq/kg, as well as Skipperud, et al. (2013) in their work claim that the concentration of radionuclides bio accumulates in large quantities in bones rather than in gills, more than in kidneys, and more than in the liver, while in smaller amounts it is in the muscle, agreeing satisfactorily with the result of this study, which was that of the total ^{226}Ra measured, 95.21% was accumulated in bones and organs, and only 4.79% in muscle tissue.

The results reported by Siddeeg, et al., (2014), and Sethy, et al., (2011), on ^{226}Ra concentrations in sediments of rivers that are considered affected by mining activity, are in a range between 15. 0 and 179.0 Bq/kg, while in the works of Sethy, et al., (2011) and Rowan, et al., (2013) in rivers not affected by mining activity, concentrations of ^{226}Ra in the sediments are between 1.99 Bq/kg and 4.50 Bq/kg; these last values are comparable with those obtained in this work, which were between 0.90 ± 0.07 Bq/kg and 1.30 ± 0.08 Bq/kg for the Irquis River, and 1.10 ± 0.08 Bq/kg and 19.30 ± 0.08 Bq/kg for the Portete River.

The levels of dissolved ^{226}Ra activity in river waters are related to the composition of the soil and the physical and chemical properties of the waters. Siddeeg (2013) asserts that the physical properties of the bedrock along the river bed play an essential role in the contribution of the concentration of ^{238}U, ^{232}Th and ^{226}Ra in natural waters. The geochemical processes of ^{238}U, ^{232}Th and ^{226}Ra, such as the adsorption by clay minerals, insoluble oxides such as iron oxide, play a key role in the understanding of the behavior of these radioisotopes and in the analysis of their concentration levels present in both water and in the soil. On the other hand, Porcelli, et al., (2001), affirm that these levels of concentrations vary, when "the streams drain limestone, phosphates and rocks rich in ^{238}U", factor that influences so that the concentration of this radionuclide is greater. When comparing the results obtained by Hameed et al.,(1997) (0.07-314 mBq /l) and De Lama & Osores (2002) (2.1-175.4 mBq/l) whose works were carried out in areas with lithological similarities to the present work, where predominate the andesitic strata from volcanic (igneous) rocks, which contain relative high percentage of silica, iron oxides and calcium oxides, we found that the values obtained are within the ranges mentioned above; in the Portete River the activities of the dissolved ^{226}Ra ranged between 43.5mBq/l and 146 mBq/l, while in the Irquis River the values were between 37 mBq/l and 119 mBq/l.

Some authors, such as De Oliveira, et al., (1996) and the International Atomic Energy Organization (2014), state that ^{226}Ra at low or high pH is more soluble than at medium pH. Szabo et al. (2012) analyzed the correlation of the physical-chemical parameters of water with the ^{226}Ra content and affirm that this alkaline earth metal has a significant correlation with pH, dissolved oxygen (DO) and total dissolved solids (SDT), as well as with some metals. High concentrations of ^{226}Ra occur mainly in anoxic waters (low DO) with high content of iron, manganese, calcium, magnesium, barium, strontium and eventually with total dissolved solids, potassium, sulfates (SO_4) and carbonates (HCO_3), as well as when the water is acidic (low pH) and contains high concentration of nitrates (NO_3) and aluminum. Both the waters of the Irquis and Portete rivers, which were analyzed in this study, differ completely from the aforementioned characteristics.

Conclusions

The values obtained in this work, concerning to the amount of ^{226}Ra measured in trout bone-organ matrices, the trout muscle-tissue matrices, and water of the rivers Irquis and Poetete, were very similar to values obtained by other authors who measured the same magnitudes in uncontaminated waters.

It should be noted that the values obtained in this work are the first of their kind and could serve as a reference database for future works whose main objective will be to measure the radiological impact due to the mining activity in Loma Larga.

It is suggested to expand the sampling area around the Loma Larga concession, the number of samples, and the sampling period and calculate the bioaccumalation factor (BAF).

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