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HEAVY METALS PROBLEM IN MICRO-BASIN THAT FEEDS A DRINKING WATER DAM, MILLUNI - BOLIVIA CASE

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The water problems generated by the negative impacts of mining in Bolivia have international recognition. The micro-basin of Milluni, mining activity territory, is an important freshwater source for the department of La Paz. The water treatment plants that treat the Milluni water do not have specific treatments to remove heavy metals; water quality is not guaranteed. Identifying the heavy metals present in the area is the first step in seeking solutions to this contamination. This study makes an evaluation of the current monitoring program in Milluni, compiling and analyzing historical data since 2006 in order to identify persistent heavy metals in the area.

Keywords: *Heavy metals; micro basin; mining; water problems.*

PROBLEMÁTICA DE METALES PESADOS EN UNA MICRO CUENCA DE ALIMENTACIÓN DE REPRESA DE AGUA POTABLE, CASO MILLUNI - BOLIVIA

La problemática hídrica generada por los impactos negativos de la minería en Bolivia tiene reconocimiento internacional. La micro-cuenca de Milluni, territorio de actividad minera, es una fuente de agua dulce importante para el departamento de La Paz. Las plantas potabilizadoras que tratan las aguas de Milluni, no cuenta con tratamientos específicos para la eliminación de metales pesados; la calidad del agua no está garantizada. Identificar los metales pesados presentes en el área es el primer paso para buscar soluciones a esta contaminación. Este trabajo realiza una evaluación del programa de monitoreo vigente en Milluni, recopilando y analizando datos históricos desde el año 2006 con el fin de identificar los metales pesados persistentes en el área.

Palabras clave: *Drenajes ácidos de mina; metales pesados; tratamientos de agua.*

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1. Introduction

Multiple magmatic intrusions beneath the Eastern Cordillera and folded-thrust belt of the Andes Mountains has led to widespread mineralization in Bolivia (Miller & Villarroel, 2011). This has attracted large and medium-scale mining activities which developed intensively in several regions of Bolivia, causing significant environmental damage (Chatelain & Wittinton, 1992). Despite this, the mining of silver, gold, tin, lead, zinc, and antimony, among others, continues to be an important component of the Bolivian economy (Miller & Villarroel, 2011).

Mining areas are known to be the major anthropogenic sources of heavy metal pollution. In these areas, rainwater can cause metals to leach into water bodies (Institute of Environmental Conservation and Research [INECAR], 2000). Heavy metals are transported by water flows, either dissolved or as part of entrained sediments. These metals can accumulate in sediments and enter groundwater (Duruibe et al. 2007).

In Bolivia, a number of scientific studies have evaluated specific cases of mining pollution at high-altitude locations, such as: Cerro Rico of Potosí (Garrido et al. 2017), Pilcomayo river in Potosi (Hudson-Edwards et al. 2003), the Chayanta river in PotosÍ (Oporto, Smolders & Vandecasteele, 2012), Poopó and Uru-Uru lakes in Oruro (Fernández et al. 2014), Apolobamba in La Paz (Muñoz et al. 2013), and Sajama Glacier in La Paz (Ferrari et al. 2001).

While not considered in previous studies, Milluni is an important region because its lagoons supply more than 18% of the total drinking water demand of the department of La Paz (Public Social Company of Water Sanitation [EPSAS] according to its name in Spanish, 2013). The department of La Paz has an approximate population of 2.8 million inhabitants (National Institute of Statistics of Bolivia [INE] according to its name in Spanish, 2018), over half of which live in the cities of La Paz and El Alto. Unfortunately, some Milluni lagoons are affected by mine water run-off, and little is known about the extent or severity of this pollution (Salvarredy-Aranguren et al. 2008).

Please consider changing this to “Heavy metal contamination of water sources, like in the case of Milluni, is a serious issue. Contamination of drinking water sources with heavy metals can compromise the quality of water for human consumption because heavy metals, depending on their concentration, are considered toxic or dangerous to human health (World Health Organization [WHO], 2006).

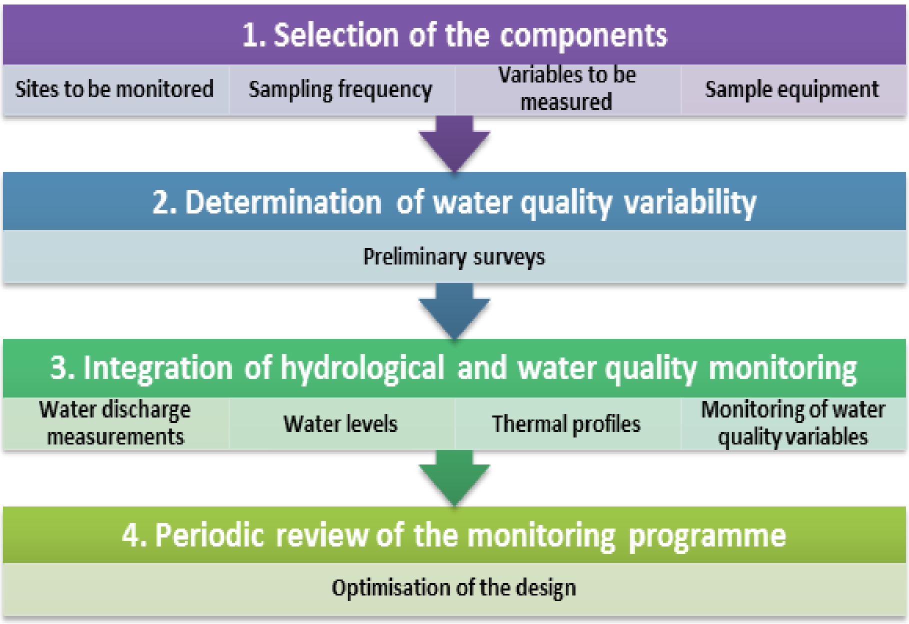
The water quality of lagoons in the Milluni area has been monitored since 2006. However, a specific evaluation of the heavy metal data to identify metals with dangerously high concentrations has not yet been carried out. An evaluation of the current monitoring program in Milluni, compiling and analyzing historical data since 2006, is needed to identify persistent heavy metals in the area. The result of this evaluation will be the first step in identifying measures to eliminate heavy metals from surface water which will reduce public health risk by reducing the likelihood of the consumption of contaminated water.

2. Water quality monitoring program

There could be as many types of monitoring program as there are objectives, water bodies, pollutants, and water uses. In practice, assessments are limited to about ten different types of programs. It should be noted that many countries or water authorities have installed multi-purpose or multi-objective monitoring programs without conducting the necessary preliminary surveys (Chapma & WHO, 1996).

In the design of a water quality monitoring program, the first requirement is to establish an objective to define the components of the program. Once the objective has been identified, there are four steps in the program development (Figure 1).

Figure 1: Steps for the development of a water quality monitoring program



Note: Own elaboration adapted from Chapma and WHO (1996) & Bazán et al. (2014).

The selection of sites and the frequency of monitoring depend to a large extent on the morphology, hydrodynamics, sources of pollution, and resources available for monitoring. The variables that will be measured will vary according to the available resources and the objectives of the program (Bazán et al., 2014).

The handling of large datasets obtained monitoring programs is often a complex task which is made more difficult by the range of different stakeholders involved in the process of water quality assessment. The values obtained must allow different types of conflicts to be solved such as the use of water and the ecological integrity of aquatic systems, as well as others, which also involve socioeconomic aspects (Samboni, Carvajal & Escobar, 2007).

3. Case study: Milluni

Large scale mining activities in the Milluni area stopped around 20 years ago, but the impact of mining waste on water quality remains a serious environmental problem. Another problem that affects water quality in this area is the small-scale and often illegal mining activities, for which, no accurate information is available (Salvarredy-Aranguren et al., 2008).

3.1. Location and geomorphology

Milluni is a glacial valley that is within the Murillo province of the department of La Paz (16° 08' to 16° 10' south latitude and from 68° 17' to 68° 21' west longitude), approximately 25 km north of the urban area of the city of El Alto and about 30 km from the city of La Paz. It has an altitude from 4,500 to 4,700 m.a.s.l and is part of the Cordillera Real. It is located on the south wall of Cerro Huayna Potosí (6,088 m.a.s.l), who's snow-capped mountains supply drinking water, after treatment, to the cities of La Paz and El Alto. (Miranda, Arancibia & Quispe, 2010).

The valley contains several small lakes, which, along with other geomorphological features, help to control drainage in the catchment. For example, the moraines and sheet flow have been noted to effect the drainage in the Zongo Valley located in the northern part of the Milluni. A network of diffuse runoff was observed during the wet season in the Milluni Valley. Several sectors of the basin, particularly the upper basin, are draped with peat bogs and high mountain pasture land (Salvarredy-Aranguren et al., 2008).

The Milluni Valley, located in the high altitude Eastern Andes, covers an area of 40 km^2 and constitutes part of the Altiplano basin system. Lithological units can be summarized from north to south as:

1. Granitic terrain partially covered by glaciers (Huayna PotosÍ granite, HPG)
2. Slightly metamorphosed fine-grained sandstone with bedded black shales (Cambrian to Ordovician)
3. Mineralized Silurian sandstone (Catavi Formation). The Catavi Formation is mainly composed of sandstone deformed by faults and folds, which allowed mineralization to occur (Ahlfeld, Schneider-Scherbina & Bolivia, 1964).

Lehmann (1978) performed a number of geological and mineralization investigations. The primary minerals in the region are: pyrite (FeS_2), marcasite (FeS_2), pyrrhotite ($Fe_{(1-X)}S$), sphalerite (ZnS), arsenopyrite (FeAsS), cassiterite (SnO_2), galena (PbS), wolframite (Fe,Mn $[WO_4]_2$), and stannite (Cu_2FeSnS_4). Associated minerals include quartz (SiO_2), siderite ($FeCO_3$), hematite ($Fe2O_3$), apatite ($Ca_5(PO_4)_3F_2$), and monazite ($[Ce,La,Nd]PO_4$).

Underground mining was used to exploit this mineralization, and a maximum production of 110,000 T/a of rough mineral extracted was achieved between 1970–1980s (Ríos, 1985). A large amount of the mining waste produced, after mineral extraction by gravimetric and flotation procedures, was spread hazardously on slopes in the Milluni Chico sector (4.6 km^2).

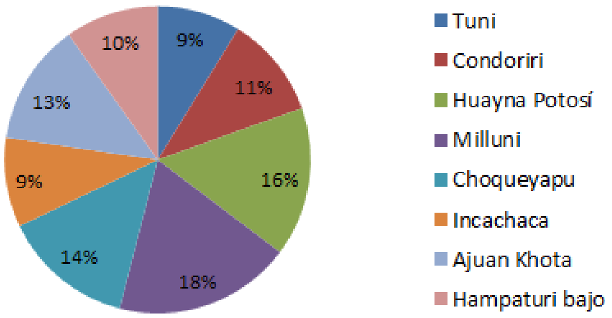
3.2. Surface water bodies in Milluni

In the upper Milluni basin there are four lagoons (Iltis, 1988):

- **Lagoon Pata Khota:** A natural lagoon with an irregular shape which receives water from the snow-melt from Huayna Potosí. It is located at 4665 m.a.s.l., and contains no evident water pollution.
- **Lagoon Jankho Khota:** A natural lagoon with an irregular shape which receives water from the Pata Khota lagoon. It is located at 4560 m.a.s.l., and contains no evident water pollution.
- **Lagoon Milluni Chico:** An artificial lagoon, at an altitude of at 4550 m.a.s.l., with an irregular shape, its tributaries are natural springs and mines. The artificial lagoon aims to capture drainage from the mines to prevent it from entering the Milluni Grande lagoon.
- **Lagoon Milluni Grande:** Located at 4530 m.a.s.l. the Milluni Grande lagoon receives effluents from natural springs, the Milluni Chico lagoon, and possibly drainage from mines. It also receives water from the Jankho Khota lagoon through a "bypass system," which consists of a pump that draws water from the Jankho Khota lagoon, through an open cement channel, to the Milluni Grande dam.

The surface water bodies in Milluni contribute to the storage dam located in the Milluni Grande lagoon. It has a capacity of 10,000,000 m^3 and an area of 2,450,000 m^2 (Raffailac, 2002). Figure 2 shows the contribution of water by basin for the department of La Paz, where the Milluni basin is the one with the greatest contribution for water supply.

Figure 2: Water contribution by basins for the department of La Paz.



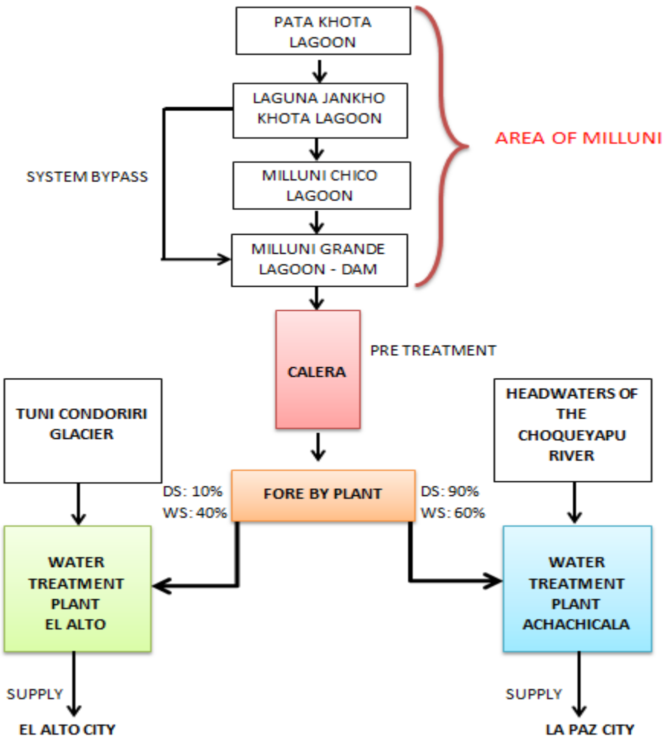
Note: Data from EPSAS (2013).

3.3. Milluni water treatment

The growing demand for drinking water for the cities of La Paz and El Alto, and the lower volume of glacial water as a result of climate change, have led to the contaminated waters of Milluni (acidic water) being used to satisfy the current demand. Although these waters receive treatment, it does not mean that Milluni's water quality is safe for supply.

The water that comes from the surface bodies of the Milluni basin is stored in the Milluni Grande lagoon by a dam. This water which is contaminated with mining waste is mixed with uncontaminated water from the Jankho Khota lagoon. This mixed water receives pre-treatment, consisting of the manual addition of lime (in the Calera), before arriving at the Fore By plant. In this plant, the total flow is distributed to two water treatment plants of the Public Social Company of Water and Sanitation - EPSAS Figure 3, is a diagram of the process that follows the Milluni water from source to tap.

Figure 3: Outline of water flow through water treatment plants in Milluni



Note: DS= Dry Season; WS= Wet Season. Data from EPSAS (2013).

The distribution of the flow through the Fore By plant is not uniform throughout the year. In the dry and wet seasons, there are distributions of 10% and 90%, and 40% and 60% to the El Alto and Achachicala plants, respectively.

During periods of maintenance of any of the plants, 100% of the flow goes to the plant in operation. Both plants have a maximum input flow of 1000 L/s. The line process that each plant has is listed below with a brief description.

Table 1: Processes of the water purification plants, El Alto and Achachicala

Processes	Description	Water purification plants	
		El Alto	Achachicala
Homogenizer tank	Decreases the kinetic energy of water.	✔	✔
Pre-sedimentation tank	Removes the coarse and fine suspended particles in the water		✔
Sedimentation tank	Removal of solids in suspension.	✔	✔
Aeration waterfalls	Increases the dissolved oxygen in the water.	✔	
Coagulant hoppers and feeders	Aluminum sulfate and lime are used as coagulants.	✔	✔
Flocculation tanks	Sulfate of aluminum or lime is added to generate the formation of flocs to separate the solid particles from water.	✔	✔
Rapid gravity filters	Have layers of sand and gravel to retain floccules and impurities.	✔	✔
Sludge tank	Storage of sludge coming out of the fast filter		✔
Filtered water pond	Stores filtered water for chlorination.	✔	✔
Chlorine dispenser	Injects chlorine gas into filtered water for disinfection.	✔	✔
Storage tank	Tanks where the total water accumulates before supply.	✔	✔
Quality sensor	Online measurement of pH, temperature, turbidity, frees chlorine and conductivity.	✔	✔

Note: Data from EPSAS (2013).

The Table 1 shows the processes that each water treatment plant has. Both plants have similar processes, although, the El Alto plant does not have a pre-sedimentation tank or sludge tank, and the Achachicala plant does not have aeration waterfalls. The total storage of the El Alto and Achachicala plants is 15,000 m^3 and 10,000 m^3, respectively.

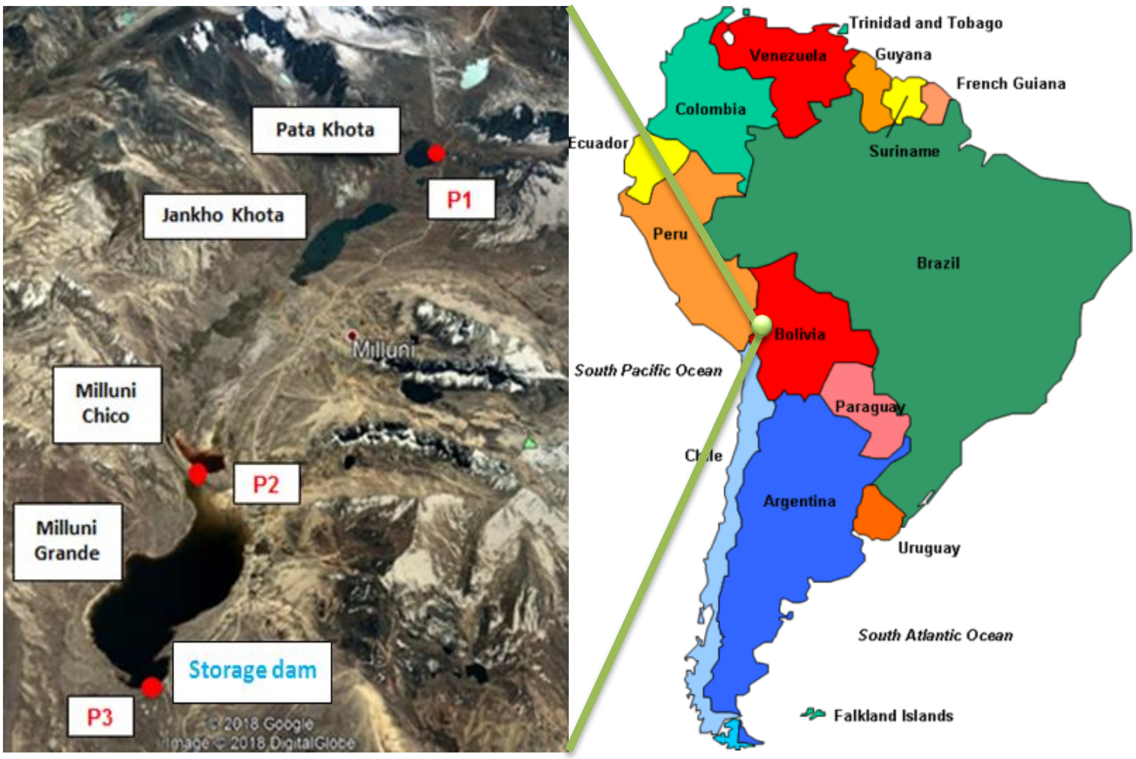
3.4. Water quality monitoring program of Milluni

From 2006 to date, the Katari Basin Management Unit (UGCK), belonging to the Ministry of Environment and Water (MMAyA) of Bolivia, monitors the Katari Basin, one of the biggest in the country. This macro-basin, which is labeled strategic by the National Watershed Plan, houses the Milluni area, which stands out in its importance due to the pollution generated by the mining exploitation carried out for more than 50 years (Salvarredy-Aranguren et al., 2008). Currently, illegal, small-scale mining continues in and around the Milluni basin (Miranda, Arancibia & Quispe, 2010).

Initially, yearly water quality monitoring in the Milluni basin was planned. However, from 2015 the frequency of monitoring increased to twice per year to gather information during both the wet and dry seasons. These periods are considered critical because there could be a change in the composition of the water caused by the increase or decrease in rainfall.

The monitoring program at Milluni was created to give a general overview of the water quality in the basin, in the beginning; it just provided information for certain projects that were being developed in the area. As of 2016, the monitoring program achieved its main objective, to "Generate a database on water quality in Milluni". Following this, the final monitoring points were defined in 2017; these are shown in Figure 4. The monitoring points of surface water bodies in Milluni are located: In the headwater of Pata Khota lagoon (P1), after effluent of the Milluni Chico lagoon, the most polluted sector (P2) and in the storage dam at the exit of the Milluni Grande lagoon (P3).

Figure 4: Sampling points and water bodies in the Milluni area



Note: Data from UGCK, 2018.

4. Results and discussion

The mixed water containing mine drainage (acidic water) is treated in the water treatment plants of El Alto and Achachicala, that do not have specific processes for the removal of heavy metals. The Milluni water should, therefore, be free of heavy metals before entering the water treatment plants, to prevent interference in the performance of the plants. For this reason, it is essential that dangerous and persistent pollutants are identified to prioritize and take measures for their elimination. To identify dangerous and persistent pollutants, an evaluation of the current monitoring program in Milluni is presented based on compiled and analyzed historical data from 2006-present.

4.1. Basic procedure

The basic procedure for an information selectivity analysis should begin with an initial and approximate examination of the data (Dahmen, Hall & International Institute for Land Reclamation and Improvement, 1990). From the analysis of the monitoring data set of the Milluni area from 2006 to 2018, the following points stand out:

- **The monitoring was not homogeneous in its frequency.** Between 2006 and 2015 a single mandatory monitoring event occurred each year, however from 2015 to present, two mandatory monitoring events occurred each year, one in the dry and one in the wet season.
- **The monitoring points were not maintained.** The points in some cases changed their position over time. This makes analyzing the variability of the data over time difficult; in addition, comparisons between monitored locations cannot be made if their positions have changed.
- **The variables which were monitored were changed**. This makes an analysis of how certain parameters change through time difficult.
- **In 2016 a detailed study of the Milluni area was carried out.** Eight points were monitored, which gave a way to validate the three points that would be maintained in future monitoring
- **2017 began with the monitoring of the three final points for future monitoring.**

Considering the above points, there is some inconsistency and heterogeneity in the Milluni monitoring data, which is common in time-series of hydrological data (Yevjevich & Jeng, 1969). These weaknesses suggest optimization of the monitoring program is necessary to generate complete and reliable information on the water quality in the Milluni area.

The lack of data makes it impossible to perform a statistical study of historical data in this area. Therefore, a rough data screening exercise was undertaken with this limited data to identify the presence of heavy metals and their persistence over time.

4.2. Rough data screening

The points used for rough data screening were those identified as more homogeneous throughout the monitoring period and important for their strategic location. Figure 4 shows the selected points.

Using rough data screening it was possible to identify the heavy metals with occurred most frequently at the monitored points. In addition, two other parameters (pH and conductivity) were analyzed due to their direct relationship with the presence of heavy metals. At this stage of the study, due to the framework and the stakeholders involved, no values are presented for parameters. However, a comparison of the monitoring data compared to the Bolivian Standard 512 for drinking water, used as a reference, is shown in Tables 2, 3, and 4.

Point 1. The headwater of Pata Khota lagoon

At the highest point of the basin, the lagoons feed directly from the thaw of the Huayna Potosi glacier. This first point does not appear to be affected by mining.

Table 2: Measured parameters at sample point 1.

Year	pH	CE	As	Cd	Fe	Zn	Mn
2011 (WS)	✔	✔	✔	-	-	✔	✔
2011 (DS)	✔	✔	x	✔	✔	✔	✔
2012 (WS)	✔	✔	-	-	✔	✔	✔
2014 (DS)	✔	✔	x	✔	✔	-	-
2015 (WS)	✔	✔	x	x	x	-	-
2015 (DS)	✔	✔	✔	✔	x	✔	✔
2016 (WS)	✔	✔	x	✔	✔	✔	✔
2017 (DS)	✔	✔	✔	✔	✔	✔	✔
2018 (WS)	✔	✔	✔	✔	✔	✔	✔

Note: CE= Conductivity; x= values that exceed the regulation; ✔= values that are within the regulation; - = values not available; DS= Dry Season; WS= Wet Season. Data from UGCK, 2018.

In all years, the pH and conductivity values were within the acceptable limits of the Bolivian standard 512 for drinking water. In 2015, three metals, arsenic (As), cadmium (Cd) and Iron (Fe), exceeded the acceptable limits. This phenomenon could be due to a small exploration or mining activity that happened over a short time-span. Arsenic is the metal that most often exceeds regulatory values, which implies that As should be prioritized in future monitoring efforts

Point 2. The effluent of the Milluni Chico lagoon

This point has only been monitored since 2016, which provides limited information. The pollution from mining waste is evident in the reddish-orange color of water and sediments in this area.

Table 3 shows that the second point is the most polluted part of the Milluni area, due to the mining effluents that reach this waterbody. The pH and conductivity values mostly lie outside the recommended ranges (Bolivian Standards 512). The direct relationship between pH, conductivity, and the presence of heavy metals is evident, because concentrations of arsenic (As), cadmium (Cd), Iron (Fe), Zinc (Zn) and manganese (Mn) all exceed the permitted limits (Bolivian Standard 512), for the monitored years.

Table 3: Measured parameters at sample point 2.

Year	pH	CE	As	Cd	Fe	Zn	Mn
2016 (WS)	x	✔	x	x	x	x	x
2017 (DS)	x	x	x	x	x	x	x
2018 (WS)	x	x	x	x	x	x	x

Note: CE= Conductivity; x= values that exceed the regulation; ✔= values that are within the regulation; - = values not available; DS= Dry Season; WS= Wet Season. Data from UGCK, 2018.

Point 3. Storage dam, the exit of the Milluni Grande lagoon

In the third monitored point provided longer-term and more homogeneous data due to the presence of the water storage dam. The contamination of the Milluni Grande lagoon was expected because it receives water from the Milluni Chico lagoon (the most contaminated water body in Milluni). The pH is outside the acceptable range of Bolivian Standard 512, except in 2011. Finally, the concentrations of arsenic (As), cadmium (Cd), Iron (Fe), Zinc (Zn), and manganese (Mn), regularly exceed the permitted limits.

Table 4: Measured parameters at sample point 3.

Year	pH	CE	As	Cd	Fe	Zn	Mn
2006 (WS)	x	✔	x	-	x	x	-
2007 (WS)	x	✔	x	x	x	x	x
2007 (DS)	x	✔	x	x	x	x	x
2008 (WS)	x	✔	✔	✔	x	✔	✔
2011 (WS)	✔	✔	✔	-	-	✔	✔
2011 (DS)	✔	✔	x	x	✔	✔	✔
2014 (DS)	x	✔	x	x	✔	-	-
2016 (WS)	x	✔	x	x	x	x	x
2017 (DS)	x	✔	x	x	x	x	x
2018 (WS)	x	✔	x	x	x	x	x

Note: CE= Conductivity; x= values that exceed the regulation; ✔= values that are within the regulation; - = values not available; DS= Dry Season; WS= Wet Season. Data from UGCK, 2018.

In summary, the rough data screening revealed that heavy metal contamination can be observed in concentrations which exceed permitted limits in the most important lagoons of Milluni (Milluni Chico and Milluni Grande lagoons). To validate this first evaluation, an in-depth study should be carried out in the area to verify the presence of arsenic, cadmium, iron, zinc, and manganese. An in-depth study will allow appropriate measures to be identified and taken to remove persistent heavy metals from the surface water bodies of Milluni.

5. Conclusions

This study dealt with the contamination of mining waste at high altitudes, a topic which has received little attention, particularly in this geographical area, until now. The Milluni basin is an important source of drinking water for the department of La Paz. It is located 4665 m.a.s.l. and includes the water storage dam for the cities of La Paz and El Alto.

An evaluation of the current monitoring program was carried out in Milluni, which consisted of compiling and analyzing historical data, since 2006, to identify persistent heavy metals in the area. Heavy metals with concentrations exceeding values set out in the Bolivian Standard 512 for drinking water, and those which were persistent over time, were: arsenic, cadmium, iron, zinc, and manganese. Further research is necessary to validate these preliminary results. An in-depth study of water quality in Milluni is recommended because a statistical assessment of the data was not possible due to the limited information.

Some inconsistencies and a lack of homogeneity were evident in the development of the Milluni water quality monitoring program; among the most important were a non-uniform monitoring frequency, variation in the locations of monitoring points and the variables measured. An area with a high probability of contamination by heavy metals, which is also a water storage area for the supply of drinking water, such as Milluni, must have an adequate monitoring program to reduce risk to public health. In addition, a functional monitoring program generates valid information for the exact identification of the contaminant, which makes it possible to choose the type of treatment required for its elimination.

Treating the water from the Milluni lagoons will not only make the water supply for the cities of La Paz and El Alto safer, but it will prevent the contamination of soil and groundwater in the area. Also, the vegetation and living things in the region will not be at risk of bioaccumulation of heavy metals. Water from the Milluni lagoons continues to be a direct source of water for small populations that are not connected to the drinking water network, which further increases the need to find a solution to this problem.

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