https://doi.org/10.17221/152/2019-SWR

Potentially toxic metals and metalloids in surface water intended for human consumption and other uses in the Mantaro River watershed, Peru

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Citation: Custodio M., Álvarez D., Cuadrado W., Montalvo R., Ochoa S. (2020): Potentially toxic metals and metalloids in surface water intended for human consumption and other uses in the Mantaro River watershed, Peru. Soil & Water Res., 15: 237–245.

Abstract: The concentration of Cu, Fe, Pb, Zn and As in the surface water intended for human consumption and other uses in the Mantaro River basin were analysed using multivariate methods. The water samples were collected from seven water bodies in the Junín region in June 2019, in the low discharge period. In each body of water, a sector with six sampling sites was established. The Cu, Fe, Pb, Zn and As contents were determined by the flame atomic absorption spectrophotometry method. The correlation analysis revealed positive and significant correlations ($P < 0.05$) for the Zn/Pb, Pb/Fe and Zn/Fe pairs with a good association, higher than 0.80 and for the Pb/Cu, Fe/Cu, As/Pb and As/Zn pairs a weak degree of association ($P < 0.05$). The analysis of the main components showed three components with their own values > 1. The hierarchical grouping analysis classified the evaluated water bodies into three groups according to the concentration of the Cu, Fe, Pb, Zn and As. The high concentrations of heavy metals and arsenic recorded in the CIMIRM and MERIS irrigation channels reveal that the Mantaro River continues to be a sink for mine wastewater discharges and runoff from mining liabilities at the headwaters of the Mantaro basin. It is, therefore, necessary to implement urgent management policies to control and reduce the levels of contamination by potentially toxic metals and metalloids in the Mantaro River.

Keywords: arsenic; heavy metals; human consumption; Mantaro River; water quality; watershed

Water is essential for the support of life. Variations in its quality are the result of natural and anthropogenic pressures that it has endured. Among the natural pressures, weathering and bedrock erosion, volcanoes and atmospheric transport are sources of significant contributions of heavy metals to the aquatic environment (Liu et al. 2018). In the last decades, the anthropogenic pressures that these environments have been supporting have increased rapidly. This increase is mainly due to the mining and metallurgic industries, the use of herbicides and pesticides, the combustion of fossil fuels, and the discharge of industrial and domestic effluents (Goher et al. 2014; Jia et al. 2017). The pollution of water bodies is of a great global concern, due to the high volumes of municipal and industrial wastewater they receive, from accelerated population growth, urban expansion and industrialisation and agricultural modernisation (Akinbile et al. 2016).

Heavy metals are the group of pollutants with the greatest interest due to their potential toxicity, persistence and bioaccumulation (Kara et al. 2017). In aquatic environments, these toxic elements can be released from the sediment through processes of adsorption and desorption, which prolongs the residence time of the heavy metal contamination, constituting a major source of pollution for the water column (Hassan et al. 2015). The degradation of the water quality by these metals leads to serious risks to human health and ecosystems, the loss of biodiversity and the deterioration of environmental quality (Hou et al. 2019).

Peru is rich in mineral resources and one of the world's largest producers of silver, copper, zinc, tin, lead and gold. Therefore, mining is the main economic activity in Peru. In addition, it has one of the most important metallurgical complexes in South America, where most of these metals are melted. The operation of various mining units in Peruvian territory has led to the contamination of numerous bodies of water with serious implications for human and animal health (Mirzabeygi et al. 2017). Also, with the exception of the Amazon region, the availability of water is one of the current priority problems, due to the fact that many of the regions have a large deficit of water destined for human consumption. Water pollution by heavy metals in various river basins has generated conflicts (Guittard et al. 2017); since it constitutes a potential threat to a human's well-being, ecological security and economic development.

In the Central Andes of Peru, the Cerro de Pasco region has one of the most intensely exploited silver deposits in the world. Although the exploitation of metals in colonial times is well known from historical accounts in this region, large-scale mining began in the late 16th century (Hunefeldt 2004), with Pb and Hg being the main pollutants related to the pre-colonial foundries (Cooke et al. 2009). Cerro de Pasco and the numerous mines that surround it exploit polymetallic (Pb-Zn-Cu-Ag) ore deposits, the primary minerals of which are enargite (Cu3AsS4), arsenopyrite (FeAsS), aramayoite (Ag[Sb, Bi]S2), argentiferous galena ([Ag, Pb]S), argentiferous tennantite ([Ag, Cu, Fe]12[Sb, As]4S13), gratonite (Pb9As4S15), sphalerite ([Zn, Fe]S), bismuthinite (Bi2S3), and native silver (Ag) (Einaudi 1977). Lake Junín is the body that receives the mine water runoff through the various tributary rivers (Rodbell et al. 2014) and the origin of the Mantaro River.

Water quality studies of the Mantaro River in 2007 showed high contents of Pb, Cd, Hg, Fe, Cr, Zn, As and cyanide (Environmental Dialogue Bureau 2007), < indicating that the metals are the tracers of the mining activity in this basin. The Ministry of the Environment reported 32 mine water discharge sources in the upper Mantaro River (MINEN 2009). Meanwhile, the National Water Authority identified 17 mine waste water discharge sources from operating mining units and 72 contaminating sources of mining liabilities (NWA 2014); despite the monitoring and control carried out by Peruvian auditing organisations.

In the Mantaro River watershed, a major part of the bodies of water are used for energy, agriculture, population, fishing and mining, and are of great importance in the national and regional economy. However, the use of the waters of the main river in this basin, the Mantaro River, has been restricted for most uses due to the high concentrations of heavy metals and metalloids it possesses, which exceed the national environmental quality standards-EQS (MINEN 2017). Within the framework of Peru's environmental policy, the National Water Authority conducts bi-annual and in some cases annual assessments of the water quality of the Mantaro River and some of its important tributaries. However, the health status of many water bodies in the central region of Peru remains unknown, as the water use has intensified and untreated wastewater discharges increase year after year, affecting the water quality to meet basic human needs.

The assessment of the water quality has been carried out for several decades by conventional methods mainly using physical and chemical parameters. At present, there are various methods for assessing water quality, ranging from water quality indexes, bioindication methods to multivariate statistical methods. The integration of these methods provides a detailed understanding of the water quality through the interpretation of complex data matrices (Bengraïne & Marhaba 2003), facilitating the identification of potential sources of pollution through the interpretation of chemical data. In this sense, and considering the importance of the implications of water quality in a human's well-being and in the ecosystems, this preliminary study was carried out with the objective of analysing the spatial concentration of heavy metals and arsenic in the surface water destined for human consumption and other uses in the Mantaro river basin by means of multivariate methods.

MATERIAL AND METHODS

Study area. The Mantaro River Watershed is located between 10°30' to 13°30'S latitude and between 74°00' to 76°30'W longitude, in an area consisting largely of moderately folded and faulted Mesozoic-era sedimentary miogeosynclinal rocks. This central

Andean Mesozoic belt contains most of the mineral deposits of the Central Andes of Peru (Petersen 1965), many of which are associated with the Tertiary intrusive hypabyssal rocks whose composition ranges from granite to diorite (Nagell 1960). The Mantaro River originates in Lake Junín, at an altitude of 4 080 m a.s.l., and runs 735 km up to its confluence with the Apurímac River, at 480 m a.s.l.

The study area is limited to the Junín region and includes the rivers in the provinces of Junín, Jauja, Concepción and Huancayo, at an altitude ranging from 3 240 to 4 400 m a.s.l, with an extension of 15 118.80 km^2 (Figure 1). The average rainfall is 600 mm and the average temperature is 11 °C. However, the rainfall has great seasonal variability, with the dry season between May and August and the rainy season between September and April (Avalos et al. 2013).

The bodies of water considered in the study are sources of water for human consumption (previously going through disinfection processes), fish farming, irrigation and other uses. In each body of water, sectors were established with their respective sampling sites. In Lake Junín, the sampling sector was located in front of the community of San Pedro de Pari, on the Mantaro River, in two sectors; one in the irrigation canal on the left bank of the Mantaro River (CIMIRM) and the other in the irrigation canal of the irrigation improvement plan in the Sierra (MERIS) on the right bank. A representative sampling sector was defined in the Cunas River, which originates in the western mountain range at 5 180 m a.s.l., the Shullcas, Chía and Chanchas Rivers, which originate in the Huaytapallana snow-capped mountain range (Table 1).

Collection and analysis of the water samples. The water samples were collected from seven bodies of water in the Junín region in June 2019, on sunny days during the dry season. In each body of water, a sector was established with six sampling sites with a distance of 100 m between them. The sampling in the rivers and irrigation canals was carried out in the opposite direction to the flow of the current at a depth of 20 cm, in two-litre plastic bottles, previously treated with a 10% nitric acid solution for 24 h and repeatedly rinsed with distilled water. The conservation of each sample was achieved by adding 1.5 ml of concentrated nitric acid to one litre of water (APHA/AWWA/WEF 2012). Prior to the sample collection, the bottles were rinsed three times with water from the lake, river or irrigation canal. (Hassan et al. 2015). The samples, under refrigerated conditions, were sent to the laboratory for the determination of the Cu, Fe, Pb, Zn and As contents by atomic absorption spectrophotometry.

The digestion of the samples was carried out from 250 ml of the water in a precipitation glass, which was

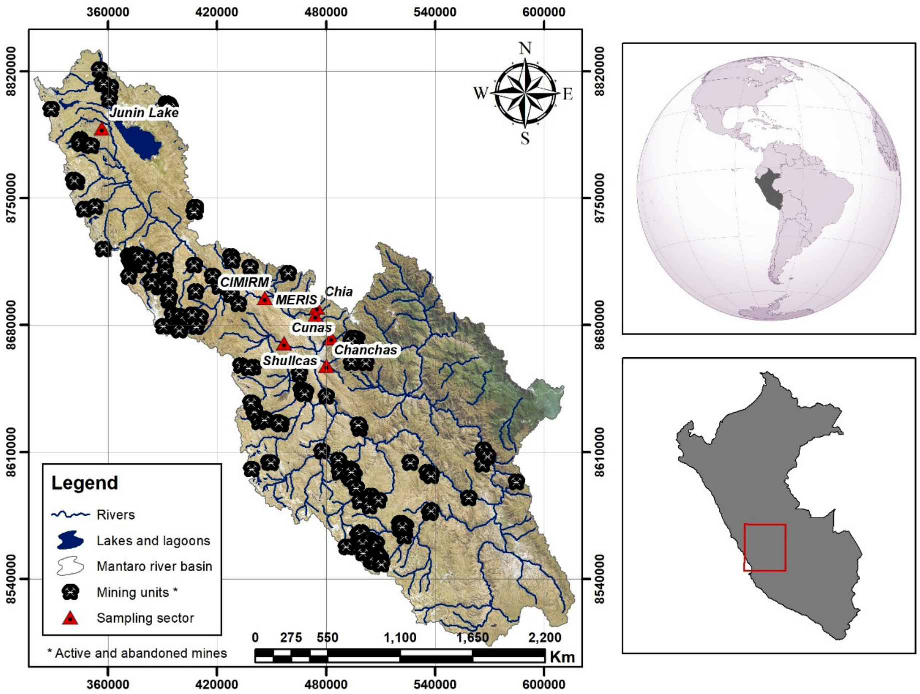

Figure 1. Location of the surface water sampling sectors in the upper and middle Mantaro watershed, the Junín-Perú region

brought to boiling point, until 100 ml was obtained. Immediately, 5 ml of nitric acid and 5 ml of concentrated hydrochloric acid were added for the destruction of the organic matter and again it was brought to the boil (until consuming the water and obtaining a residue of pasty consistency). It was allowed to cool and then 10 ml of distilled water was added, filtered and gauged in a 100 ml pan with 1% nitric acid. The concentration of Cu, Fe, Pb, Zn and As (mg/L) was determined by the method of atomic absorption spectrophotometry by flame, according to the methodology recommended by the FAO (1983), using an AA-6800 Atomic Absorption Spectrophotometer, Varian AA240.

Data analysis. The statistical differences in the concentrations of the heavy metals and arsenic in the surface water of the sampling sectors were analysed by a one-way ANOVA (analysis of variance) and the post-hoc comparisons were made using Tukey's Honestly Significant Difference Test (HSD), considering the P-value < 0.05 as significant. Prior to the standardisation of the data, the analysis of main components (PCA) with Varimax rotation was applied in order to identify the potential sources of the elements registered in the study area. The number of the factors was selected taking its own values greater than 1 into account. The factorial loadings of each element, with values higher than 0.6, were considered as influential in each factor. To complement the PCA, the analysis of the conglomerates was carried out with Ward's method and the square Euclidean distance to identify similarities between the groups of elements based on the concentration levels. All the statistical analyses were performed with R CRAN free software, (Ver. 3.5.2, R Core Team et al. 2016).

RESULTS AND DISCUSSION

Characterisation and distribution of the heavy metals and arsenic. Table 2 shows the ANOVA descriptive statistics and results for the copper, iron, lead, zinc and arsenic in the surface water bodies of the Mantaro River basin intended for human consumption, fish farming and irrigation. The highest concentrations of these elements were recorded in the sectors of the CIMIRM and MERIS irrigation canals, derivations of the Mantaro River; as well as in the sectors of Junín Lake in front of the San Pedro de Pari community. The release of these types of pollutants into the aquatic ecosystems affects their functioning, accumulating in different compartments of the trophic chain (Howladar et al. 2018). The distributions of the mean concentrations of Cu, Fe, Pb, Zn and As in the different bodies of water showed significant differences ($P < 0.05$).

Iron was the element that presented the highest concentrations in the CIMIRM, MERIS, Lake Junín-Pari and the Chanchas River water bodies, with ranges of 5.799, 0.6672, 0.124, 0.223 and 0.0238, respectively. In most water bodies, the concentrations of iron exceeded the permissible values for human consumption. This result would be influenced by natural factors such as the abundance of iron in the

Table 1. The geographical coordinates, description and sources of the contamination of the surface water bodies in the Mantaro River watershed, Junín region

Surface water bodies	Latitude	Length	Altitude	Description of the study area	Sources of the contamination
Junín Lake (Pari)	−10.99	−76.21	4 086	Lake Junín gives rise to the Mantaro River, one of the main Andean tributaries of the Amazon basin. For many years it has been the recipient of effluents from the mining units operating in the area.	Acid mine drainage Tailings dam overflow Toxic liquid dumps Mining liabilities Discharge of domestic water
CIMIRM Canal	−11.83	−75.39	3 320	The Mantaro River receives contaminated water from the rivers that run through the areas with abandoned and operating mining units. As well as, from the polymetallic metallurgical complex of La Oroya, which has dumped toxic liquids into it for decades.	Discharge of liquid waste from La Oroya Metallurgical Complex Dumping of non-metallic mining Hydroelectric power plant discharge Discharge of domestic water
MERIS Canal	−11.96	−75.30	3 245		
Chanchas River	−12.17	−75.19	3 560		
Chía River	−11.88	−75.24	3 734		
Cunas River	−12.13	−75.43	3 656		
Shullcas River	−11.98	−75.10	3 994		

https://doi.org/10.17221/152/2019-SWR

Table 2. The descriptive statistics and ANOVA of the Cu, Fe, Pb, Zn and As (in mg/L) concentrations recorded at the surface water sampling stations in the upper and middle zone of the Mantaro River watershed, Junín, Peru

Chemical element and descriptive statistics	Surface water bodies							ANOVA	Drinking water guidelines (mg/L) WHO	Environmental quality standards Peru (mg/L)		
	Canal CIMIRM	Canal MERIS	Junin-Pari Lake	Chanchas River	Chía River	Cunas River	Shullcas River			drinking water	water for fish culture	water for irrigation
Cu												
Mean	0.0153^{a}	0.0153^{a}	0.00602	0.00257^{b}	0.0149^{a}	0.00217^{b}	0.00160^{a}	< 0.05*	2	2	0.2	0.2
SD	0.0094	0.0003	0.0002	0.0007	0.0013	0.0002	0.0004					
Max	0.0300	0.0159	0.0062	0.0034	0.0165	0.0025	0.0021					
Min	0.0045	0.0150	0.0058	0.0012	0.0132	0.0020	0.0010					
Fe												
Mean	2.40	0.471^{a}	0.415^{a}	0.241	0.00953	0.0164	0.0534	< 0.05*	0.3	0.3	NA	5
SD	2.1200	0.2850	0.0486	0.0758	0.0095	0.0023	0.0342					
Max	6.5600	0.7240	0.4640	0.3470	0.0241	0.0200	0.1220					
Min	0.7610	0.0568	0.3400	0.1240	0.0003	0.0139	0.0307					
Pb												
Mean	0.0152^{a}	0.0111^{a}	0.0296	0.00101^{b}	0.00107^{b}	0.00112^{b}	0.000385	< 0.05*	0.01	0.01	0.0025	0.05
SD	0.0105	0.0012	0.0206	0.0003	0.0007	0.0010	0.0003					
Max	0.0322	0.0128	0.0547	0.0013	0.0019	0.0027	0.0010					
Min	0.0026	0.0097	0.0050	0.0006	0.00003	0.00018	0.00011					
Zn												
Mean	0.0152^{a}	0.0159^{a}	0.00384^{b}	0.000375^{b}	0.000237	0.000162	0.000705	< 0.05*	3	3	1	2
SD	0.0190	0.0074	0.0034	7.29e-05	0.000101	8.64e-05	1.05e-05					
Max	0.0524	0.0229	0.0093	0.00049	0.00036	0.00027	0.00072					
Min	0.0003	0.0045	0.00021	0.0003	0.00009	0.00001	0.00069					
As												
Mean	0.0837^{a}	0.0689^{a}	0.0341	0.0143	0.0116	0.0107	0.00685	< 0.05*	0.01	0.01	0.1	0.1
SD	0.0787	0.0565	0.0213	0.0017	0.0027	0.0012	0.0037					
Max	0.229	0.138	0.0581	0.0165	0.0156	0.0128	0.0109					
Min	0.0132	0.0071	0.0087	0.0123	0.0087	0.0094	0.0026					

***Significant at the probability level of 0.05; means followed by the same letters (a, b) are not significantly different (Tukey multiple comparisons of means, $P < 0.05$)

earth's crust and in the natural waters. However, in aquatic environments, the chemical behaviour of iron is controlled by the pH, redox potential, inorganic and organic complexing agents (Khan et al. 2017), but when concentrations are higher than the recommended value, it can affect the biological communities, both in their diversity and abundance. The mean concentrations of iron in the CIMIRM, MERIS and Junín-Pari Lake water bodies exceeded the WHO drinking water guidelines (0.3 mg/L) (WHO 2011) and the EQS-water for drinking water production (0.3 mg/L) of Peru (MINEN 2017). However, they did not exceed the EQS-water for irrigation (5.0 mg/L). Comparatively, the mean iron concentration in the CIMIRM channel in this study was higher than those obtained by PADES (2006), who recorded a mean iron concentration of 0.59 mg/L for this irrigation canal.

The mean lead concentration in the study was lower (0.0152 mg/L) than that obtained by PADES (0.032 mg/L). The reason for the lower concentration of this heavy metal recorded is due to the temporary closure of the metallurgical centre in La Oroya for more than a decade (one of the most polluted cities in the world). Lead in nature is found as Pb^{2+} and Pb^{4+}, it has no useful biological function in humans. In aquatic environments, the salt of Pb^{2+} is slightly soluble and its high water content is toxic to aquatic biota (Has-Schön et al. 2015). The maximum concentrations of iron and lead were recorded in the CIMIRM canal (6.56 and 0.0322 mg/L, respectively) exceeding, by far, the WHO guidelines for drinking water and the EQS-water intended for the production of drinking water (0.3 mg/L) and irrigation (5.0 mg/L).

The mean and maximum concentrations of this metal recorded in all the evaluated surface water bodies did not exceed the WHO drinking water guidelines (2.0 mg/L) or Peru's environmental quality standards for water (EQS-water) intended for drinking water production, fish farming and irrigation (0.2 mg/L). Cu is an essential micronutrient for humans, but an excess amount of Cu can be harmful to humans, animals and aquatic organisms. The mean zinc concentrations in the seven surface water bodies did not exceed the WHO guidelines for drinking water (3.0 mg/L) and the EQS ones for drinking water production (3.0 mg/L), fish farming (1.0 mg/L) and irrigation (2.0 mg/L). Zinc is one of the oldest trace metals and a frequent environmental pollutant that is widely distributed in the aquatic environment. The high concentration of this metal is toxic to aquatic organisms. However, high concentrations of this metal can also cause harmful problems to human health. Arsenic behaved dissimilarly to the heavy metals. The mean concentrations of 85.71% of the water samples exceeded the reference values of the WHO drinking water guidelines and the Peruvian EQS-water ones (0.01 mg/l). The highest concentrations of this metalloid were recorded in the CIMIRM and MERIS canals, with values of 0.229 mg/L and 0.138 mg/L, respectively. In the study area, the main sources of arsenic are lead smelters, the exacerbated pesticide use, and mining. Arsenic can cause strong toxic effects in ecosystems, can enter the aquatic food chain through biological absorption and can be a risk to animal and human health (Asare-Donkor et al. 2015).

Multivariate statistical analysis. Table 3 shows the matrix of Pearson correlation coefficients of the heavy metals and arsenic in the surface water of the upper and middle Mantaro River basin. The pairs Zn/Pb, Pb/Fe and Zn/Fe showed positive and significant correlations ($P < 0.05$) with a good association, superior to 0.80 according to the scale proposed by Martinez et al. (2009). The positive and significant correlations ($P < 0.05$) between the pairs Pb/Cu, Fe/Cu, As/Pb and As/Zn, showed a weak degree of association whose range is between 0.26 and 0.50, revealing that these metals are related to the main mining and smelting activities of the study area. The concentrations of As did not correlate with the Cu or Fe. This result is supported by numerous studies that show that the environmental pollution of a region is not only related to the anthropogenic impact, but also the geochemical effects of the mineralised zones (Demková et al. 2017).

The results of the KMO (Kaiser-Meyer-Olkin) test = 0.8 and Barlett's sphericity test [χ^2 approx. (df = 4) = 1083.3, $P < 2.2x10^{-16}$] validated the application of the PCA. There were three components with their own values > 1 for the entire data set (Figure 2A). The selected components explained a total variance of 92.82% (Table 4). Component 1 represented 62.22% of the total variability in the dataset,

Table 3. The Pearson Correlation of heavy metals and arsenic in the surface water of the upper and middle Mantaro River watershed

	Cu	Pb	Zn	Fe	As
Cu	1	0.406**	0.372*	0.403**	0.122
Pb	0.406**	1	0.859***	0.814***	0.412**
Zn	0.372*	0.859***	1	0.763***	0.479**
Fe	0.403**	0.814***	0.763***	1	0.336*
As	0.122	0.412**	0.479**	0.336*	1

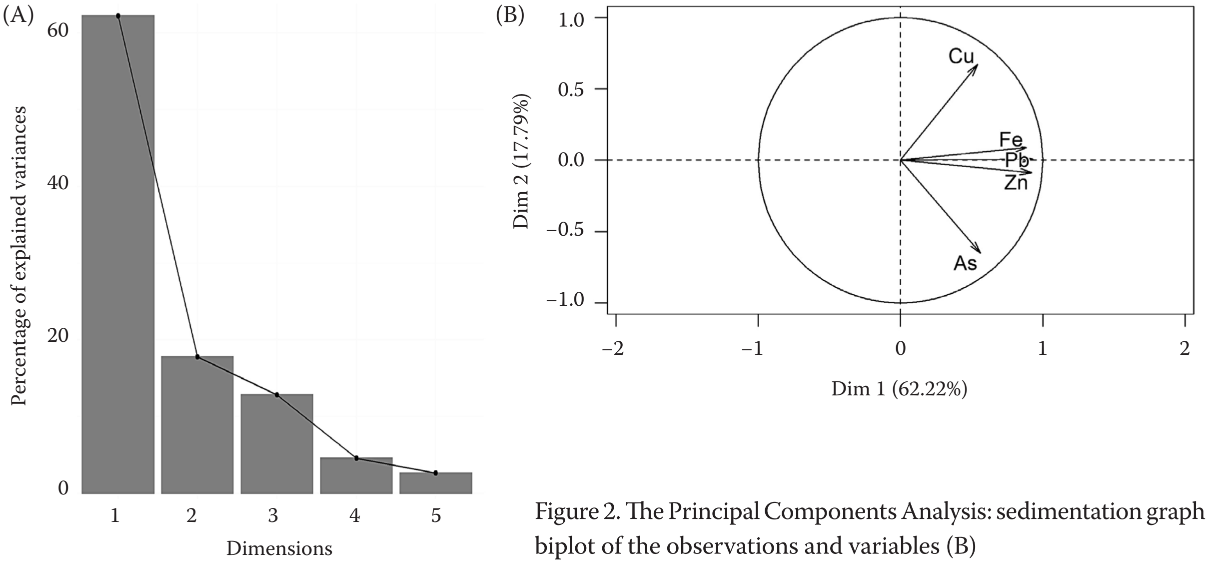

Figure 2. The Principal Components Analysis: sedimentation graph (A), biplot of the observations and variables (B)

with high loads of Pb, Zn and Fe trace elements of anthropogenic activities associated with mining and smelting. Component 2 represented 17.79% of the total variability, with high Cu levels. Component 3 represented 12.81% of the variability revealing important loads of As, which would have come from natural and anthropogenic sources (industrial discharge sources and municipal waste) given its significant correlation with Pb ($r = 0.412$). The representation of the axes is shown in Figure 2B.

Figure 3 shows the results of the hierarchical clustering analysis of the surface water bodies evaluated and the concentrations of the heavy metals and arsenic, according to Ward's method using the Euclidean squared distance. The surface water bodies were classified into three groups according to their mean concentrations (Figure 3A). In group I, the Chia River shows a clear tendency to separate from the other bodies of water. This behaviour would be influenced by the low concentration of the heavy metals and arsenic in its water bodies. In group II, the Cunas River maintains the tendency to separate from the water bodies that present high concentrations of the contaminants conglomerated in group III. The results of the hierarchical cluster analysis indicate that the sampling sites belonging to the same group of water bodies have similar concentrations of heavy metals and arsenic. The hierarchical clustering analysis for the metals and arsenic yielded three groups (Figure 3B). Group I was formed by concentrations of Cu, group II by As and group III by Fe, Pb and Zn.

The results reveal that the concentrations of the heavy metals and arsenic vary among the water bodies of the Mantaro River watershed. This variability would be associated with the discharges from the mining industry in operation, to the dragging of metals from abandoned mining dams due to heavy rainfall, to industrial and domestic discharge sources from

Table 4. The analysis of the main components with the Varimax rotation of the heavy metals and arsenic in the surface water of the upper and middle Mantaro River watershed

	Component 1	Component 2	Component 3	MSA
Cu	0.24	**0.97**	0.04	0.94
Pb	**0.91**	0.19	0.20	0.75
Zn	**0.87**	0.16	0.30	0.78
Fe	**0.90**	0.20	0.09	0.85
As	0.24	0.03	**0.97**	0.86
Proportional variance (%)	62.22	17.79	12.81	
Cumulative proportion (%)	62.22	80.01	92.82	

MSA – measure of sampling adequacy; elements with high loads (>0.8) are in bold (those that show a high correlation with the respective component)

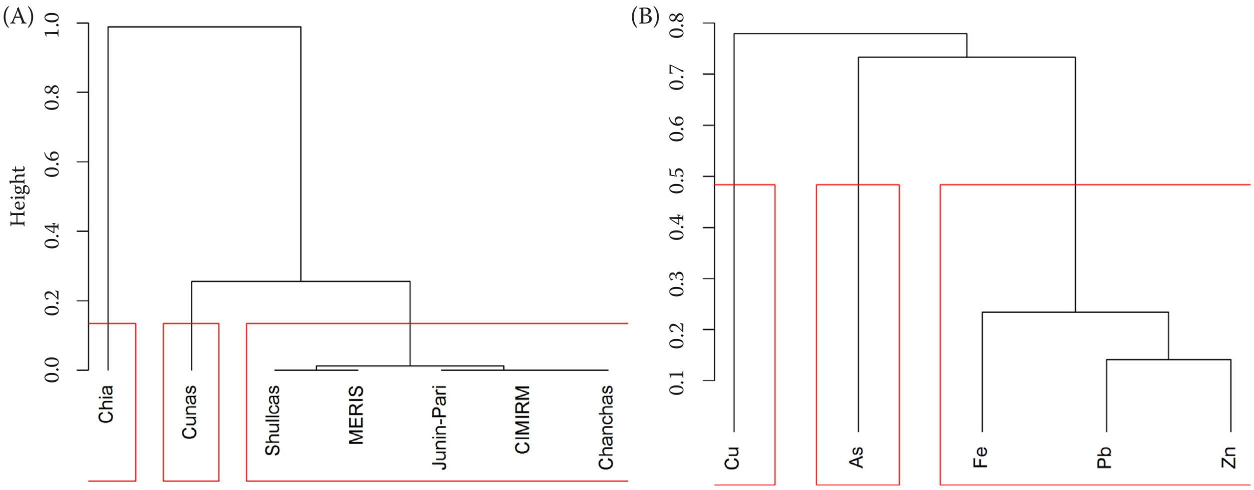

Figure 3. The hierarchical grouping analysis of the surface water bodies (A) and concentrations of the heavy metals and arsenic (B), according to Ward's method using the Euclidean squared distance

the population settled in this basin, as well as natural processes such as the dissolution of geological deposits, biological degradation of organic matter and atmospheric deposition (Guittard et al. 2017; Kilunga et al. 2017). The spatial distribution of Cu, As, Pb and Zn indicates that the quality of the water bodies is mainly influenced by the waste and effluents from the mining and metallurgical activities that are developed in the study area in comparison to Fe that would mainly be coming from natural sources (Akinbile et al. 2016). The importance of the distribution of these potentially toxic chemicals in the water bodies is evidenced by the cluster analysis.

CONCLUSION

The water bodies of the Mantaro River watershed are exposed to pollution from a variety of natural and anthropogenic sources. The high concentrations of heavy metals and arsenic recorded in the CIMIRM and MERIS irrigation canals reveal that the Mantaro River continues to be a sink for the mine wastewater discharges and runoff from the mining liabilities at the headwaters of the Mantaro watershed. As evidenced by the loads of these chemical elements determined through the analysis of the main components. However, variations in the chemical characteristics of the water are also influenced by the natural processes. The behaviour that is established by the hierarchical cluster analysis classifies the water bodies into three groups according to their mean concentrations of the heavy metals and arsenic. The results obtained reveal that it is necessary to implement urgent management policies to control and reduce the levels of the contamination of the potentially toxic metals and metalloids of the Mantaro River, since its waters are used for irrigation in much of the basin.

Acknowledgements. The authors would like to express their gratitude to the General Research Institute of the Universidad Nacional del Centro del Perú for funding the study, to the Water Research Laboratory for facilitating the use of the equipment and materials for this study, and to engineer Irving Lizárraga for his participation in the sampling.

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Received: December 9, 2019
Accepted: March 24, 2020