

RESEARCH ARTICLE

Artisanal gold-mining in a rural environment: Land degradation in Kenya

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Abstract

Artisanal gold-mining (AGM) is one of the most important activities in the districts of Migori and Transmara (Kenya). Gold-mining, however, is known to release vast quantities of arsenic and metals (some of which are very toxic like As, Hg, Cd, or Pb), which poses a serious threat to not only miners but also to the ecosystem and local populations. We, herein, determine the concentrations of arsenic and some heavy metals in several sample types (i.e., ore, soil, river sediment, and mine tailing) collected from the districts of Migori and Transmara. We also employ lichens and mosses as pollution bioindicators. Geostatistical tools and canonical correlation analysis were used to identify the relevant factors that affected arsenic and metal concentrations in the analysed samples. The following concentration ranges were reported in topsoil: As (1–17,250 mg kg⁻¹), Cd (0.01–15.10 mg kg⁻¹), Cu (7–9,238 mg kg⁻¹), Cr (1–214 mg kg⁻¹), Ni (5–766 mg kg⁻¹), Pb (3–1,149 mg kg⁻¹), and Zn (22–1,271 mg kg⁻¹). It was concluded that the ecosystem in both districts was highly polluted by heavy metals whereas the arsenic concentrations in topsoil were among the highest reported worldwide. The results of this study provide new evidence on the impact of AGM on the environment and may further contribute to the design of policy measures with the aim of reducing environmental and human health risks associated to AGM activities.

KEYWORDS

arsenic pollution, bioindicators, environmental degradation, Kenya gold-mining, spatial variation

1 | INTRODUCTION

Gold production in Kenya is a major driver of the country's economic growth and development and contributes \$10.3 million per year to the nation's economy. In the Migori and Transmara districts (SW Kenya), gold-mining started in the 1930s (Odumo et al., 2014), whereas industrial mining has recently become a relevant activity (in 2008) as newly explored gold-bearing reefs were discovered in the Lolgorien region. As several companies have been licensed for gold prospecting, it is quite likely that industrialised gold-mining will spread to other areas. Additionally, artisanal gold-mining (AGM) undertaken by local populations (Figure S1) has become one of the most important sources of income in this area (Odumo et al., 2014).

Apart from being a relevant source of income for local and national economy, gold-mining may be responsible for several environmentally harmful effects, including pollution from heavy metals and metalloids (HMMs), tailing dumping, soil erosion, and subsequent river damage or landscape deterioration (Gutiérrez et al., 2016). Hg is among the most relevant environmental contaminants released from AGM. During AGM, gold is manually extracted from open pits using hand tools and is then crushed. Afterwards, mercury amalgamation, which is used for gold extraction purposes, has been reported as being responsible for increasing Hg concentrations in habitats near pits (Odumo et al., 2014; Veiga et al., 2009). Other metals and metalloids, for example, Ni, As, Pb, Cd, Cr, Zn, and Cu, can be released to the environment after gold-mining activities (Cobbina, Myilla, & Michael,

2013). For instance, in the Migori area, arsenopyrite (FeAsS) and galena (PbS), which are primary mineral phases of gold-sulphide deposits, can be released to the environment from roaster stacks after amalgam combustion. Indeed, As is oxidised to arsenite to then precipitate during roasting from vapours to become As_2O_3 . Several studies report an association of As with gold deposits containing sulphide minerals (Antwi-Agyei, Hogarh, & Foli, 2009; Bisone et al., 2016; Chakraborti et al., 2013; Houben, D'Onofrio, Kokelj, & Blais, 2016; Nyanza, Dewey, Thomas, Davey, & Ngallaba, 2014; Ollson, Smith, Scheckel, Betts, & Juhasz, 2016; Ono, Tappero, Sparks, & Guilherme, 2016; Toujaguez et al., 2013).

The HMMs released from mining by-products are usually deposited in surrounding habitats, where soil is one of the most relevant reservoirs (Antwi-Agyei et al., 2009; Kpan, Opoku, & Gloria, 2014). Concentrations of HMMs in soil strongly depend on bedrock composition (Alloway, 1995; Mirzaei, Esmaili-Sari, Hemami, Rezaei, & Rodríguez Martín, 2014; Nanos & Rodríguez Martín, 2012), which is generally very low, hence, ensures optimum ecological functioning (Campos-Herrera et al., 2016; Mirzaei, Ghorbani, Hafezi Moghaddas, & Martín, 2014; Rodríguez Martín et al., 2014). Nonetheless, human activities resulting in the release of HMMs have drastically increased the natural background in soil (Rodríguez Martín, De Arana, Ramos-Miras, Gil, & Boluda, 2015). Therefore, several authors suggest that the soil HMM concentration close to the pollution source may be used as an archive of historic emissions of pollutants into the environment (Rodríguez Martín, Carbonell, Nanos, & Gutiérrez, 2013; Rodríguez Martín & Nanos, 2016). Apart from soil, mosses and lichens have also been utilised as bioindicators of HMM pollution since the 1980s (Conti & Cecchetti, 2001). Vegetation not only absorbs gaseous compounds but also accumulates airborne particles. It is well-accepted that mosses and lichens (not influenced by their substrate) can be employed as biomonitors of HMM pollution (Rossini Oliva & Mingorance, 2006). These bioindicators are long-living perennials with an immense geographical distribution and tolerant to toxic metals. This facilitates biomonitoring programmes over extensive areas (Loppi & Bonini, 2000; Odumo et al., 2014).

The objective of the present study was to assess the extent of environmental pollution in Migori and Transmara districts of Kenya

due to As and certain heavy metals (Pb, Cu, Cd, Ni, Cr, and Zn) released into the environment from gold-mining activities. Moss and lichen samples were applied to provide further evidence of the effect of AGM on the local environment. Finally, the potential negative repercussions from the continuous releases of pollutants to the environment from AGM activities were reported, and the spatial extent of land degradation in the study area was assessed applying geostatistical techniques.

2 | MATERIALS AND METHODS

2.1 | Soil sampling

In 2013, 123 samples (35 soil, 15 ore, 17 tailing, 22 river sediment, 15 lichens, and 19 mosses) were collected from georeferenced locations (Figure 1). Approximately 1 kg of soil samples was collected randomly at a 30-cm depth in undisturbed positions in the surrounding areas of rivers and mines. Samples of river bed sediment were taken from central parts of flowing rivers, wet tailings from panning pond bottoms, and dry tailings from heaps near these ponds (see Odumo et al., 2014).

2.2 | Chemical analyses

Sediment, soil, and tailing samples were air-dried and then sieved by a 2-mm grid. Samples were pulverised before being sieved by a 90/100- μ m sieve. Although the study aim was to estimate the direct effect of atmospheric pollution, it was not possible to clean the moss and lichen samples because the cuticle layers and leaf morphological ultrastructure differed in eight of the analysed species. The concentration of As and other metals (Cd, Cu, Cr, Ni, Pb, and Zn) in the soil, sediment (tailing and river), as well as in lichen and moss samples were established by Atomic Absorption Spectrometry (PerkinElmer, Shelton, CT, USA 06484-4794) applying Flame Atomic Absorption Spectrometry equipment for the As and Zn analyses, whereas Graphite Furnace Atomic Absorption Spectrophotometry was run for Cu, Cr, Cd, Pb, and Ni (Carbonell, Imperial, Torrijos, Delgado, & Rodriguez, 2011). All the samples were analysed in triplicate. Reference samples (river sediment, BCR-320; olive leaves, BCR-62; and calcareous loam

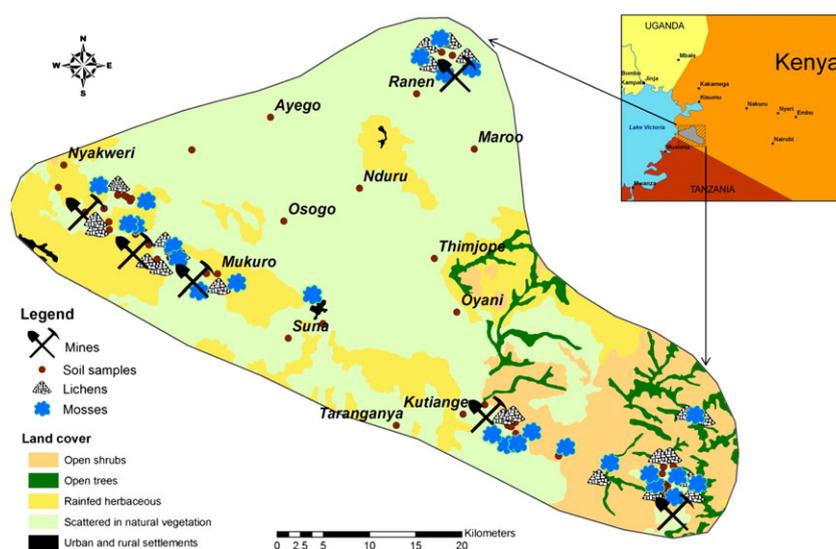


FIGURE 1 General map of the study area, which shows the Migori–Transmara gold-mining areas and sampling sites [Colour figure can be viewed at wileyonlinelibrary.com]

soil, BCR-141R) were obtained from the European Commission Community Bureau of Reference and were used to validate the analytical procedures. The percentage recoveries of metals in the reference soil were as follows: 105% for Cr, 97% for Ni, 99% for Cd, 98% for Pb, 95% for Cu, and 101% for Zn. The recoveries regarding the olive leave reference material were 99% for Cu, 97% for Pb, 101% for Cd, and 108% for Zn.

2.3 | Statistical and geostatistical analyses

Statistical analyses (summary statistics, analyses of variance, and correlations) were ran to describe the correlation structure among the trace elements in the moss, lichen, soil, sediment, and tailing samples. Non-parametric Kruskal–Wallis tests were used to find any statistical significant differences among means (land cover, soil texture, landforms, and regional slope).

A simple geostatistical analysis was used to generate kriging maps of the elements analysed in soil and sediments (Nourzadeh, Hashemy, Rodríguez Martín, Bahrami, & Moshashaei, 2012; Rodríguez Martín et al., 2016; Takoutsing et al., 2016; Zamani-Ahmadmahmoodi, Bakhtiari, & Rodríguez Martín, 2014). We computed experimental variograms and adjusted spherical variogram models for this purpose. Ordinary kriging was utilised to estimate the concentrations of HMMs at unsampled locations (a grid of 3 km × 3 km was used for this purpose). The unknown HMMs concentrations at the locations where lichens and mosses were collected were also estimated by the aforementioned kriging method. Kriging cross-validation was employed to quantify kriging prediction errors (Goovaerts, 1997).

We resorted to canonical correlation analyses (CCorA) to examine the relationship among the concentration of HMMs in soil and bioindicator tissues. This method was followed to study the correlation pattern between several explanatory and response variables. We did statistical analyses with XLSTAT (version 2012.2.02 of Addinsoft), the Geostatistical Analyst for ArcGis 10 and ISATIS v10.0.

3 | RESULTS AND DISCUSSION

3.1 | Arsenic and trace element contents in soil

High As levels can be found in the Migori–Transmara area (Table 1). The mean As content in the soil samples was 545.9 ± 491.7 mg kg⁻¹, and the maximum recorded value (above 17,000 mg kg⁻¹) was observed close to the Macalder mine (this is one of the highest As concentrations to be found in the literature for topsoil samples). At this level, the soil As concentration can be considered a human health and environmental risk because a normal As soil concentration falls within the 1–40 mg kg⁻¹ range (Alloway, 1995). The average arsenic concentrations in the other sample types were also very high. The respective average values were $1,102.2 \pm 507.1$ mg kg⁻¹, 793.1 ± 349.6 mg kg⁻¹, and 141.9 ± 57.7 mg kg⁻¹ for ore, tailing, and river sediments (Table 1). Arsenic generally has the highest concentrations in the gold mine tailings (Antwi-Agyei et al., 2009). The gold mine tailings in Obuasi (Ghana) contained a maximum of 1,885 mg kg⁻¹ of As concentration (Antwi-Agyei et al., 2009).

The highest As concentration is observed in the north-western area (Figure 2) and affected the following localities near Lake Victoria: Ranen, Ayego, Macalder, or Nyakwer. The high As levels in Macalder could be due to gold being mined for long periods by the amalgamation technique. The high levels found around Ranen and other areas with no active mining could be owing to the dry and wet depositions of As released to the environment from AGM. This argument can also be used to explain the high As values recorded in Tasmania (Nyanza et al., 2014) and Ghana (Antwi-Agyei et al., 2009), where AGM was implicated in land degradation, which poses serious human health concerns for local African communities. Gold-mining activity effects were observed in soil As at a distance of over 20 km away from mines (Houben et al., 2016). Elsewhere, gold mines have been implicated as a source of trace element environmental contamination, including soils, river, and sediments (Antwi-Agyei et al., 2009).

The concentration range of heavy metals in soil from Migori–Transmara area was quite wide (Table 1). Soil enrichment with Pb, Zn, and Cu was often associated with gold-mining activity, but the spatial extent of mining activities on soil HMMs reportedly reduced at distances of 400–700 m from one deposit of gold-base metal (Shumlyansky, Ivantyshyna, Makarenko, & Subbotin, 2005), which was also the case of Migori–Transmara (Figure 2). The average Pb value in soil (244.4 ± 197.9 mg kg⁻¹) exceeded the reference values for unpolluted soil (Rodríguez Martín et al., 2014). The maximum concentrations herein observed were, respectively, 6,952.6 and 11,148.9 mg kg⁻¹ in soil and ore samples (Table 1). These Pb contents suggested that this soil is polluted. The majority of average soil Pb contents reported in the literature are lower than those described in the present study. Indeed, the average Pb content was 39 mg kg⁻¹ in Europe (Adriano, 2001), 14 or 17.1 mg kg⁻¹ in Spain (Díez et al., 2009; Nanos & Rodríguez Martín, 2012), and 26 mg kg⁻¹ in Portugal (Inácio, Pereira, & Pinto, 2008).

The high average Zn concentrations in tailing (1,270.8 mg kg⁻¹), soil (1,123.8 mg kg⁻¹), and ore (618.2 mg kg⁻¹) were also related with gold-mining activities. The maximum permitted level for most countries is 300 mg kg⁻¹ (Alloway, 1995). For Zn contents, we indicated mean values of 124 mg kg⁻¹ in soil, 253.9 mg kg⁻¹ in tailing, and 165 mg kg⁻¹ in ore (Table 1). In the study area, the copper concentration ranges between 13.8 and 9,237.8 mg kg⁻¹ in soil, 8.3 and 6,916.8 mg kg⁻¹ in ore, and 27.9 and 2,329.3 mg kg⁻¹ in tailing. Although these constitute a wide range, 90% of the Cu values were estimated between 8 and 174 mg kg⁻¹. Other authors have reported that normal variation for Cu in soil was 5–50 mg kg⁻¹ (Bloemen, Markert, & Lieth, 1995). Such a high copper level justifies its mining alongside gold in Macalder from 1920 to 1966 (Ogola, Mitullah, & Omulo, 2002).

In the Migori–Transmara area, the mean total Cd content in soils was 0.294 mg kg⁻¹, which is a relatively low concentration compared with the values obtained for other countries, where the total soil Cd concentrations were reportedly 1–3 mg kg⁻¹ (Adriano, 2001). Nevertheless, the maximum values were very high: 5.50 mg kg⁻¹ in the soil, 15.10 mg kg⁻¹ in ore, or 4.14 mg kg⁻¹ in tailing samples. The Cd levels in agricultural soils are dangerous if in excess of 3 mg kg⁻¹ as they can have toxic effects (Kabata-Pendias & Mukherjee, 2007). Ni and Cr, which are known for their mainly natural (i.e., geogenic) origin (Nanos, Grigoratos, Rodríguez Martín, & Samara, 2015), had relatively low

TABLE 1 Statistical summary of the As and heavy metals contents in the soil, ore sediment (tailing and river), lichen, and moss samples

		Soil	Ore	Tailing	River sediment	Lichens	Mosses
As	N	35	15	17	22	15	19
	Min	2.1	18.3	2.1	4.7	1.5	0.5
	Max	17,250.4	7,523.9	5,830.0	977.9	797.7	234.4
	Mean	545.9	1,102.2	793.1	141.9	90.7	19.9
	SE	491.7	507.1	349.6	57.7	56.0	12.4
	<i>H. Groups</i>	<i>ab</i>	<i>d</i>	<i>cd</i>	<i>bc</i>	<i>ab</i>	<i>a</i>
Cd	Min	0.01	0.00	0.07	0.02	0.12	0.09
	Max	5.50	15.10	4.14	0.71	1.49	0.47
	Mean	0.29	1.60	0.84	0.21	0.33	0.17
	SE	0.15	1.02	0.25	0.04	0.09	0.02
		<i>H. Groups</i>	<i>a</i>	<i>abc</i>	<i>c</i>	<i>ab</i>	<i>bc</i>
Cu	Min	13.8	8.3	27.9	9.8	11.2	7.3
	Max	9,237.8	6,916.8	2,329.3	373.1	66.5	27.7
	Mean	316.0	647.7	350.9	82.8	25.3	13.6
	SE	262.7	464.5	144.1	18.7	4.1	1.2
		<i>H. Groups</i>	<i>bc</i>	<i>cd</i>	<i>d</i>	<i>cd</i>	<i>b</i>
Cr	Min	16.7	0.8	17.0	26.5	9.8	5.1
	Max	169.1	203.6	138.6	150.0	63.7	30.8
	Mean	61.8	101.2	84.9	68.5	21.9	11.3
	SE	5.8	13.1	8.3	7.2	3.9	1.3
		<i>H. Groups</i>	<i>c</i>	<i>d</i>	<i>cd</i>	<i>cd</i>	<i>b</i>
Ni	Min	17.4	5.3	10.8	8.9	9.3	6.3
	Max	105.2	112.5	766.1	254.7	36.3	18.3
	Mean	40.3	51.2	229.0	78.2	18.1	9.5
	SE	3.1	7.7	63.4	15.3	2.0	0.7
		<i>H. Groups</i>	<i>c</i>	<i>cd</i>	<i>d</i>	<i>cd</i>	<i>b</i>
Pb	Min	9.5	3.1	4.5	13.7	3.5	3.1
	Max	6,952.6	11,148.9	3,238.8	1,528.3	102.6	33.1
	Mean	244.4	1,048.4	371.3	130.2	24.6	12.1
	SE	197.9	742.8	195.4	69.0	6.8	2.0
		<i>H. Groups</i>	<i>bc</i>	<i>c</i>	<i>bc</i>	<i>bc</i>	<i>ab</i>
Zn	Min	48.7	22.4	41.2	33.1	43.8	31.4
	Max	1,123.8	618.2	1,270.8	230.3	164.8	113.2
	Mean	124.4	156	253.9	99.7	89.5	60.4
	SE	30.2	45.2	84.9	10.3	9.9	4.4
		<i>H. Groups</i>	<i>b</i>	<i>ab</i>	<i>b</i>	<i>b</i>	<i>ab</i>

Note. Concentration in mg kg⁻¹.

concentrations in the Migori–Transmara area. Anthropogenic Cr and Ni inputs were generally below existing soil concentrations of 34 and 20 mg kg⁻¹ reported for Cr and Ni, respectively, in the United Kingdom (McGrath & Loveland, 1992). These represent mean values obtained from a broad analysis of 6,000 samples in the United Kingdom (England and Wales).

From the geostatistical analysis, it was observed that those regions with high concentrations of HMMs roughly covered the same area (Figure 2). The most elevated level of these trace elements was located near Lolgorien (Transmara), and this trend generally descended towards the gold-mining areas of Migori. Greater quantities were indicated around Lolgorien and Macalder, with intensive mining activity. Hence, such high levels probably resulted from the dry/wet depositions of emitted metals from burning gold mercury amalgam. It might

also be due to leaching from metal-rich tailings. The low contents noted around Kitere were possibly because the mining there was not as intense as in other areas. However, the spatial distribution of Cr and Ni differs substantially from the rest of HMMs (Figure 2) supporting the conclusion that, most probably, the concentration of these heavy metals is not really altered by AGM activities.

A correlation analysis was ran (Table 2) to explore possible relationships among the soil trace elements. The correlation matrix indicated some high and significant correlations between As/Cd ($r = 0.905$), As/Cu ($r = 0.999$), As/Pb ($r = 0.998$), and As/Zn ($r = 0.974$) in both the soil and sediment samples. We can also see significant correlations between trace elements in the ore and tailing samples (As/Cd [$r = 0.731$], As/Cu [$r = 0.871$], As/Pb [$r = 0.923$], and As/Zn [$r = 0.814$]). This indicated a common origin of trace elements

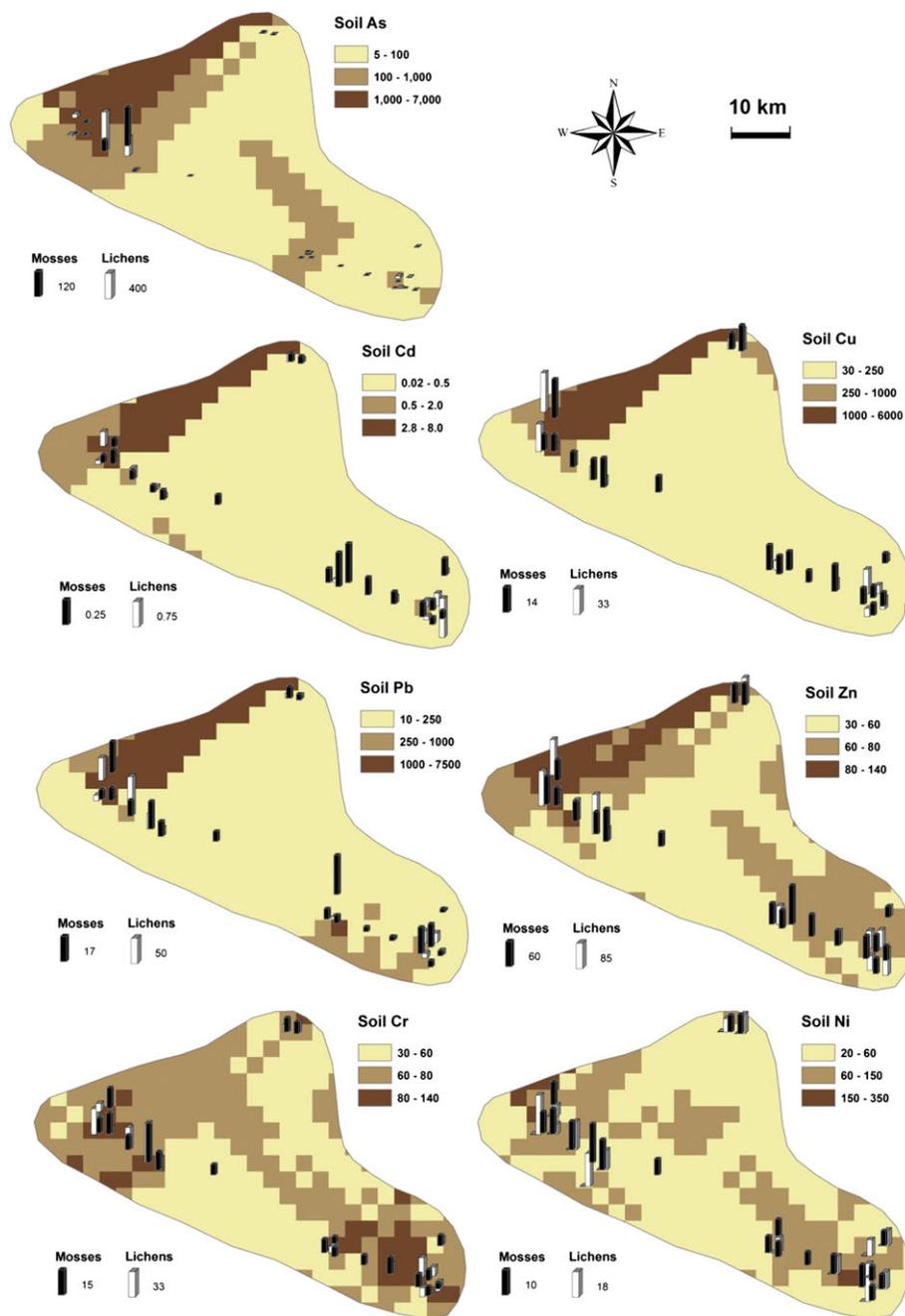


FIGURE 2 Kriging map of As and heavy metals in the soil from the Migori-Transmara area, and the sample distribution trace element contents of mosses and lichens (mg kg^{-1}) [Colour figure can be viewed at wileyonlinelibrary.com]

in the soil, which perhaps were released, dispersed, or deposited from adjacent mining sites. A more serious health and environmental risk could be associated with the simultaneous presence of several HMs in soil, especially when elements were deposited and dispersed to nearby farming soil, as recently reported in Namibia (Mileusnić et al., 2014). Precautions ought to be taken because As and HM levels in soil may be hazardous, which has unfavourable implications for agriculture and also for human health via the food chain.

3.2 | Arsenic and heavy metal contents in lichens and mosses

In the study area, the concentrations of HMMS in lichens are generally higher than in mosses (Table 1). Mosses usually could tolerate

phytotoxic atmospheric pollutants better than foliose lichens (Bargagli, Monaci, Borghini, Bravi, & Agnorelli, 2002). The mean concentration of metals can be placed in decreasing order as $\text{Zn} > \text{Pb} > \text{Cu} > \text{Cr} > \text{Ni} > \text{Cd}$ for lichen and $\text{Zn} > \text{Pb} > \text{Cr} > \text{Cu} > \text{Ni} > \text{Cd}$ for moss. Table 3 presents a correlation analysis between the soil trace elements in the moss and lichen samples. Although the variability of HM contents in lichens and mosses in the bibliography is wide, the HM contents (Cd, Cr, Cu, Ni, Pb, and Zn) in Migori-Transmara are consistent with those reported in several studies (Garty, 2001; Ite et al., 2016; Saat, Talib, Harun, Hamzah, & Wood, 2016; Szczepaniak & Biziuk, 2003).

The spatial distribution of As observed in the mosses and lichens is evidently affected by mining activity (Figure 2). The highest As values corresponded to the samples collected from the area around Macalder, where intensive mining took place and mining activity existed in this

TABLE 2 Pearson's correlation matrix for the As and heavy metals contents in soil, ore, river sediment, and tailing sediment

		As	Cd	Cu	Cr	Ni	Pb	Zn	
As	Sediment <i>n</i> = 22		0.993	0.999	0.276	-0.018	0.998	0.974	Soil <i>n</i> = 35
Cd		0.194		0.995	0.232	-0.040	0.994	0.980	
Cu		-0.049	0.542		0.257	-0.025	0.999	0.980	
Cr		-0.111	0.280	-0.162		0.860	0.262	0.262	
Ni		0.173	0.226	0.230	0.306		-0.022	-0.017	
Pb		-0.090	-0.113	-0.101	0.213	0.336		0.982	
Zn		-0.090	0.690	0.911	0.007	0.168	-0.007		
As	Ore <i>n</i> = 15		0.731	0.871	0.435	0.610	0.923	0.814	Tailing <i>n</i> = 17
Cd		0.842		0.713	0.101	0.590	0.778	0.681	
Cu		0.849	0.906		0.463	0.692	0.959	0.940	
Cr		-0.103	-0.207	-0.194		0.337	0.408	0.414	
Ni		0.393	0.221	0.256	0.426		0.733	0.717	
Pb		0.860	0.918	0.927	-0.149	0.232		0.926	
Zn		0.643	0.883	0.761	-0.094	0.243	0.690		

Note. Blonde font, correlation significant at $p < 0.05$.

TABLE 3 Pearson's correlation matrix for As and heavy metal contents in lichen and moss samples

		As	Cd	Cu	Cr	Ni	Pb	Zn	
Moss <i>n</i> = 19	As		0.002	0.027	0.583	0.449	0.881	0.254	Liquen <i>n</i> = 15
	Cd	-0.217		0.044	-0.205	-0.208	0.119	0.177	
	Cu	0.065	-0.109		0.220	0.110	0.363	0.671	
	Cr	0.837	-0.358	0.390		0.783	0.436	0.615	
	Ni	0.777	-0.309	0.378	0.870		0.252	0.514	
	Pb	0.340	0.285	0.316	0.318	0.393		0.338	
	Zn	0.070	0.384	0.313	0.073	0.301	0.585		

Note. Blonde font, correlation significant at $p < 0.05$.

area since the 1920s. In these areas, the highest concentration of As in the lichens and mosses was approximately 797 and 234 mg kg⁻¹ dry wt, respectively. No other studies has reported such high values. Both the lichen and mosses as bioindicators revealed that contamination from As decreased with increase in distance from mining sites. The mean As content herein was 90.7 mg kg⁻¹ (lichens) and 19.9 mg kg⁻¹ (mosses). The mean As level in lichens in the study area was roughly 20-fold higher than those recorded in Italy (0.36 mg kg⁻¹; Bargagli et al., 2002) or Cameroon (0.5 mg kg⁻¹; Saat et al., 2016), whereas the mean As content in mosses is 14- to 50-fold higher when compared with mosses concentrations recorded in Italy (0.4 mg kg⁻¹; Bargagli et al., 2002) or Cameroon (1.43 mg kg⁻¹; Saat et al., 2016).

3.3 | Relationships between soil trace elements and bioindicators

The CCorA (Figure 3) provided evidence of relationships between the HMM contents in soil and bioindicators, which respectively accounted for 60.43% and 64.58% of total variance in the lichens and the mosses analysis. In the lichen CCorA, As, Pb, and Zn were related on axis F1 (Quadrant Q1) and were present in polluted areas, Quadrants Q3 and Q4 (Figure 3) were associated with polluted areas. The As, Pb, and Zn contents in lichens were also represented in Quadrant Q1 and were associated with high As and Pb levels in soil. The CCorA

analysis for mosses provided the dichotomy between the polluted areas on Axis 2 (Quadrants Q1 and Q2), whereas the As content in mosses was closely associated with soil As content from the impact of high values. This could be interpreted in the light of arsenic having an influence on a local scale and also because lichen and moss arsenic concentrations were influenced positively by proximity to mines.

Local-scale anthropic influences are related with the environmental pollution that results from extracting gold by amalgamation. As formerly reported, gold-mercury mining is the major source of As (and other HM) locally in the environment (Houben et al., 2016; Ono et al., 2016) and contributes to a bigger proportion of a given environmental As type (Nyanza et al., 2014). So it is plausible that the effect of As released to the environment on biological indicators (mosses and lichens) was limited and on a small spatial scale. Houben et al. (2016) showed that the Yellowknife gold mines released 237,000 tonnes of waste arsenic dust and revealed that gold ore extraction/roasting processes can affect a 25-km radius from mines. In tailings from a gold-mining area in Brazil, Ono et al. (2016) showed high As levels (up to 2,666 mg kg⁻¹) and stressed the need to establish reference values for trace elements in suspended air particulates. The 55 mg kg⁻¹ Netherlands intervention value hence poses a grave environmental health risk (Antwi-Agyei et al., 2009).

AGM in many parts of Africa magnifies the environmental As contamination level. Such activities lead to a far-spreading As release and

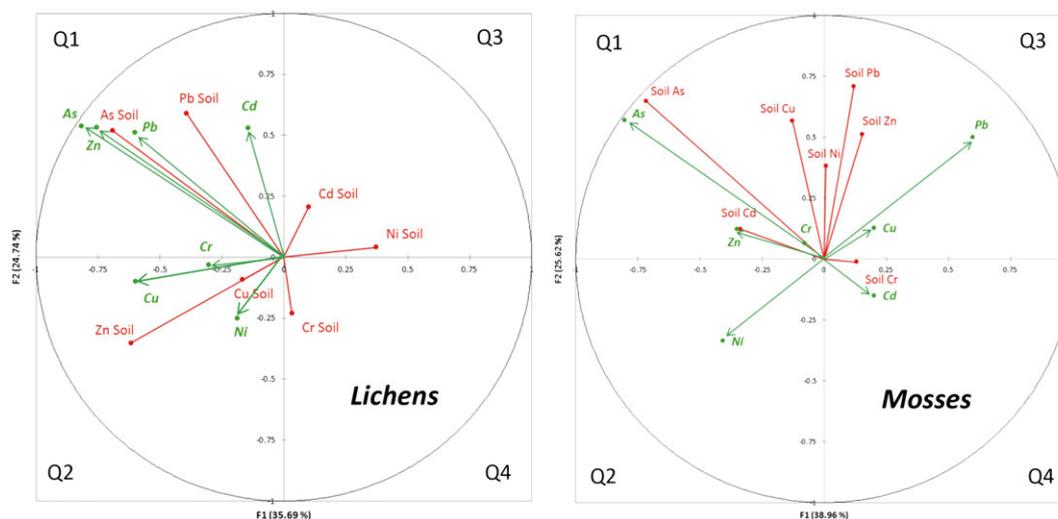


FIGURE 3 Ordination diagram based on the canonical correlation analyses of As and heavy metals versus trace element abundance of lichens and mosses [Colour figure can be viewed at wileyonlinelibrary.com]

poses major human health concerns for local communities. Nyanza et al. (2014) showed high As levels (between 180 and 20,000 mg kg⁻¹) in the Geita District (Tanzania), and increasing levels in water, including the Lake Victoria Goldfields, a problem that affects many parts of Africa. In Obuasi (Ghana), one of the longest running mines on the African continent has an average As level of 8,305 mg kg⁻¹ (Antwi-Agyei et al., 2009). The World Bank reckons that the number of artisan miners in Africa has grown from 10 M in 1999 to perhaps some 30 M today (“In praise of small miners”, 2016). Although this sector offers effective poverty relief by providing jobs and ensuring profits to small-scale artisanal gold miners, it contributes to land degradation by raising contamination and adversely affects local populations. By studying the effects of traditional gold-mining activities in Karnataka (India), with a topsoil As content over 2,000 mg kg⁻¹ in the nearby community, Chakraborti et al. (2013) demonstrated arsenical skin lesions, which were confirmed in three of every four individuals, including children. Toujaguez et al. (2013) indicated high gastrointestinal arsenic bioaccessibility, with the highest risk associated with ingestion (particularly for children) in a gold-mining area in Delita (Cuba), with an As concentration of around 8,000 mg kg⁻¹ in tailings. Presence of very high As levels (which exceeded 17,000 mg kg⁻¹) in artisanal gold-mining soil from Migori–Transmara (Kenya) warrants their relative toxicity contributions. Traditional gold mine roasting processes in Migori–Transmara significantly contributed to land degradation from pollution. This could also be the case in other gold-mining regions that applied similar gold-processing techniques. Therefore, it is crucial to identify communities at risk and to remedy the health effects that stemmed from their degradation.

4 | CONCLUSIONS

High HM and As contents were reported in the Migori and Transmara districts. From the spatial distribution patterns, it was concluded that the highest As levels in topsoil were found near mines. We provide evidence for substantial arsenic accumulation in soil, with values that exceeded 17,000 mg kg⁻¹ around some mines. Furthermore, As

concentration in lichens and mosses was remarkably high, especially in proximity to mines. Soil concentration for other HMs was also very high. There is inherently a wide spatial variability in Pb, Zn, Cu, and Cd contents in relation to gold-mining activity. Some trace element distribution anomalies were linked to emissions from neighbouring mining areas.

According to our results, the Migori and Transmara districts are highly polluted areas; hence, a potential human-health risk should be considered. The artisanal mining sector cannot only adversely affect mining communities but can contribute to land degradation by increasing contamination and negatively affecting local populations. Alternative gold-mining methods should be considered to lower the quantity of environmentally released As because this practice results in land degradation. Further research is necessary to study As levels in the food crops growing in these and surrounding areas because As soil concentrations in some locations exceeded the permissible levels that international organisations recommend.

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REFERENCES

- Adriano, D. C. (2001). *Trace elements in terrestrial environments: Biogeochemistry, bioavailability, and risks of metals*. New York: Springer Verlag.
- Alloway, B. J. (1995). *Heavy metals in soils*. Netherlands: Blackie Academic & Professional.
- Antwi-Agyei, P., Hogarh, J., & Foli, G. (2009). Trace elements contamination of soils around gold mine tailings dams at Obuasi, Ghana. *African Journal of Environmental Science and Technology*, 3, 353–359.
- Bargagli, R., Monaci, F., Borghini, F., Bravi, F., & Agnorelli, C. (2002). Mosses and lichens as biomonitors of trace metals. A comparison study on *Hypnum cupressiforme* and *Parmelia caperata* in a former mining

- district in Italy. *Environmental Pollution*, 116, 279–287. [https://doi.org/10.1016/S0269-7491\(01\)00125-7](https://doi.org/10.1016/S0269-7491(01)00125-7)
- Bisone, S., Chatain, V., Blanc, D., Gautier, M., Bayard, R., Sanchez, F., & Gourdon, R. (2016). Geochemical characterization and modeling of arsenic behavior in a highly contaminated mining soil. *Environmental Earth Sciences*, 75, 1–9. <https://doi.org/10.1007/s12665-015-5203-z>
- Bloemen, M. L., Markert, B., & Lieth, H. (1995). The distribution of Cd, Cu, Pb and Zn in topsoils of Osnabrück in relation to land use. *Science of the Total Environment*, 166, 137–148. [https://doi.org/10.1016/0048-9697\(95\)04520-b](https://doi.org/10.1016/0048-9697(95)04520-b)
- Campos-Herrera, R., Rodríguez Martín, J. A., Escuer, M., García-González, M. T., Duncan, L. W., & Gutiérrez, C. (2016). Entomopathogenic nematode food webs in an ancient, mining pollution gradient in Spain. *Science of the Total Environment*, 572, 312–323. <https://doi.org/10.1016/j.scitotenv.2016.07.187>
- Carbonell, G., Imperial, R. M., Torrijos, M., Delgado, M., & Rodriguez, J. A. (2011). Effects of municipal solid waste compost and mineral fertilizer amendments on soil properties and heavy metals distribution in maize plants (*Zea mays* L.). *Chemosphere*, 85, 1614–1623. <https://doi.org/10.1016/j.chemosphere.2011.08.025>
- Chakraborti, D., Rahman, M. M., Murrill, M., Das, R., Siddayya Patil, S. G., Sarkar, A., ... Das, K. K. (2013). Environmental arsenic contamination and its health effects in a historic gold mining area of the Mangalur greenstone belt of Northeastern Karnataka, India. *Journal of Hazardous Materials*, 262, 1048–1055. <https://doi.org/10.1016/j.jhazmat.2012.10.002>
- Cobbina, S., Myilla, M., & Michael, K. (2013). Small scale gold mining and heavy metal pollution: Assessment of drinking water sources in Datuku in the Talensi-Nabdam District. *International Journal of Scientific & Technology Research*, 2, 96–100.
- Conti, M. E., & Cecchetti, G. (2001). Biological monitoring: lichens as bioindicators of air pollution assessment—A review. *Environmental Pollution*, 114, 471–492. [https://doi.org/10.1016/S0269-7491\(00\)00224-4](https://doi.org/10.1016/S0269-7491(00)00224-4)
- Díez, M., Simón, M., Martín, F., Dorronsoro, C., García, I., & Van Gestel, C. A. M. (2009). Ambient trace element background concentrations in soils and their use in risk assessment. *Science of the Total Environment*, 407, 4622–4632. <https://doi.org/10.1016/j.scitotenv.2009.05.012>
- Garty, J. (2001). Biomonitoring atmospheric heavy metals with lichens: Theory and application. *Critical Reviews in Plant Sciences*, 20, 309–371. <https://doi.org/10.1080/20013591099254>
- Goovaerts, P. (1997). *Geostatistics for natural resources evaluation*. Oxford: Oxford University Press.
- Gutiérrez, C., Fernández, C., Escuer, M., Campos-Herrera, R., Beltrán Rodríguez, M. E., Carbonell, G., & Rodríguez Martín, J. A. (2016). Effect of soil properties, heavy metals and emerging contaminants in the soil nematodes diversity. *Environmental Pollution*, 213, 184–194. <https://doi.org/10.1016/j.envpol.2016.02.012>
- Houben, A. J., D'Onofrio, R., Kokelj, S. V., & Blais, J. M. (2016). Factors affecting elevated arsenic and methyl mercury concentrations in small shield lakes surrounding gold mines near the Yellowknife, NT, (Canada) region. *PLoS One*, 11, e0150960. <https://doi.org/10.1371/journal.pone.0150960>
- In praise of small miners (2016). The economist. Retrieved on March 4th 2018 from <https://www.economist.com/news/middle-east-and-africa/21698262-boom-artisanal-mining-offers-lessons-development-praise-small-miners>
- Inácio, M., Pereira, V., & Pinto, M. (2008). The soil geochemical atlas of Portugal: Overview and applications. *Journal of Geochemical Exploration*, 98, 22–33. <https://doi.org/10.1016/j.jgexplo.2007.10.004>
- Ite, A. E., Ubong, U. U., Etesin, U. M., Nsi, E. W., Ukpog, E. J., Ekanem, A. N., ... Udo, A. (2016). Heavy metals in epiphytic lichens and mosses of Oil-Producing communities of Eket and Ibeno, Akwa Ibom State-Nigeria. *American Journal of Environmental Protection*, 4, 38–47. <https://doi.org/10.12691/env-4-2-1>
- Kabata-Pendias, A., & Mukherjee, A. B. (2007). *Trace elements from soil to human*. Berlin: Springer Verlag.
- Kpan, J. D., Opoku, B. K., & Gloria, A. (2014). Heavy metal pollution in soil and water in some selected towns in Dunkwa-on-Offin District in the Central Region of Ghana as a result of small scale gold mining. *Journal of Agricultural Chemistry and Environment*, 3, 40–47. <https://doi.org/10.4236/jacen.2014.32006>
- Loppi, S., & Bonini, I. (2000). Lichens and mosses as biomonitors of trace elements in areas with thermal springs and fumarole activity (Mt. Amiata, central Italy). *Chemosphere*, 41, 1333–1336. [https://doi.org/10.1016/S0045-6535\(00\)00026-6](https://doi.org/10.1016/S0045-6535(00)00026-6)
- McGrath, S., & Loveland, P. (1992). *The soil geochemical atlas of England and Wales*. New York: Blackie Academic & Professional.
- Mileusnić, M., Mapani, B. S., Kamona, A. F., Ružičić, S., Mapaure, I., & Chimwamurombe, P. M. (2014). Assessment of agricultural soil contamination by potentially toxic metals dispersed from improperly disposed tailings, Kombat mine, Namibia. *Journal of Geochemical Exploration*, 144, 409–420. <https://doi.org/10.1016/j.jgexplo.2014.01.009>
- Mirzaei, R., Esmaili-Sari, A., Hemami, M., Rezaei, H., & Rodríguez Martín, J. (2014). Levels and variability of metals in soils of the Province of Golestan (Iran). *Archives of Environmental Contamination and Toxicology*, 67, 617–629. <https://doi.org/10.1007/s00244-014-0043-5>
- Mirzaei, R., Ghorbani, H., Hafezi Moghaddas, N., & Martín, J. A. R. (2014). Ecological risk of heavy metal hotspots in topsoils in the Province of Golestan, Iran. *Journal of Geochemical Exploration*, 147, 268–276. <https://doi.org/10.1016/j.jgexplo.2014.06.011>
- Nanos, N., Grigoratos, T., Rodríguez Martín, J., & Samara, C. (2015). Scale-dependent correlations between soil heavy metals and As around four coal-fired power plants of northern Greece. *Stochastic Environmental Research and Risk Assessment*, 29, 1531–1543. <https://doi.org/10.1007/s00477-014-0991-3>
- Nanos, N., & Rodríguez Martín, J. A. (2012). Multiscale analysis of heavy metal contents in soils: Spatial variability in the Duero river basin (Spain). *Geoderma*, 189–190, 554–562. <https://doi.org/10.1016/j.geoderma.2012.06.006>
- Nourzadeh, M., Hashemy, S. M., Rodríguez Martín, J. A., Bahrami, H. A., & Moshashaei, S. (2012). Using fuzzy clustering algorithms to describe the distribution of trace elements in arable calcareous soils in north-west Iran. *Archives of Agronomy and Soil Science*, 59, 435–448. <https://doi.org/10.1080/03650340.2011.636356>
- Nyanza, E. C., Dewey, D., Thomas, D. S., Davey, M., & Ngallaba, S. E. (2014). Spatial distribution of mercury and arsenic levels in water, soil and cassava plants in a community with long history of gold mining in Tanzania. *Bulletin of Environmental Contamination and Toxicology*, 93, 716–721. <https://doi.org/10.1007/s00128-014-1315-5>
- Odumo, B., Carbonell, G., Angeyo, H., Patel, J., Torrijos, M., & Rodríguez Martín, J. (2014). Impact of gold mining associated with mercury contamination in soil, biota sediments and tailings in Kenya. *Environmental Science and Pollution Research*, 21, 12426–12435. <https://doi.org/10.1007/s11356-014-3190-3>
- Ogola, J. S., Mitullah, W. V., & Omulo, M. A. (2002). Impact of gold mining on the environment and human health: A case study in the Migori gold belt, Kenya. *Environmental Geochemistry and Health*, 24, 141–157. <https://doi.org/10.1023/A:1014207832471>
- Ollson, C. J., Smith, E., Scheckel, K. G., Betts, A. R., & Juhasz, A. L. (2016). Assessment of arsenic speciation and bioaccessibility in mine-impacted materials. *Journal of Hazardous Materials*, 313, 130–137. <https://doi.org/10.1016/j.jhazmat.2016.03.090>
- Ono, F., Tappero, R., Sparks, D., & Guilherme, L. (2016). Investigation of arsenic species in tailings and windblown dust from a gold mining area. *Environmental Science and Pollution Research*, 23, 638–647. <https://doi.org/10.1007/s11356-015-5304-y>
- Rodríguez Martín, J. A., Álvaro-Fuentes, J., Gonzalo, J., Gil, C., Ramos-Miras, J. J., Grau Corbí, J. M., & Boluda, R. (2016). Assessment of the soil organic carbon stock in Spain. *Geoderma*, 264, 117–125. <https://doi.org/10.1016/j.geoderma.2015.10.010>

- Rodríguez Martín, J. A., Carbonell, G., Nanos, N., & Gutiérrez, C. (2013). Source identification of soil mercury in the Spanish Islands. *Archives of Environmental Contamination and Toxicology*, 64, 171–179. <https://doi.org/10.1007/s00244-012-9831-y>
- Rodríguez Martín, J. A., De Arana, C., Ramos-Miras, J. J., Gil, C., & Boluda, R. (2015). Impact of 70 years urban growth associated with heavy metal pollution. *Environmental Pollution*, 196, 156–163. <https://doi.org/10.1016/j.envpol.2014.10.014>
- Rodríguez Martín, J. A., Gutiérrez, C., Escuer, M., García-González, M. T., Campos-Herrera, R., & Águila, N. (2014). Effect of mine tailing on the spatial variability of soil nematodes from lead pollution in La Union (Spain). *Science of the Total Environment*, 473–474, 518–529. <https://doi.org/10.1016/j.scitotenv.2013.12.075>
- Rodríguez Martín, J. A., & Nanos, N. (2016). Soil as an archive of coal-fired power plant mercury deposition. *Journal of Hazardous Materials*, 308, 131–138. <https://doi.org/10.1016/j.jhazmat.2016.01.026>
- Rossini Oliva, S., & Mingorance, M. (2006). Assessment of airborne heavy metal pollution by aboveground plant parts. *Chemosphere*, 65, 177–182. <https://doi.org/10.1016/j.chemosphere.2006.03.003>
- Saat, A., Talib, M. S., Harun, N., Hamzah, Z., & Wood, A. K. (2016). Spatial variability of arsenic and heavy metals in a highland tea plantation using lichens and mosses as bio-monitors. *Asian Journal of Natural & Applied Sciences*, 5, 10–21.
- Shumlyanskyy, V., Ivantyshyna, O., Makarenko, M., & Subbotin, A. (2005). Environmental pollution around the Muzhievo gold-base metal deposit, Ukraine. *Management of Environmental Quality: An International Journal*, 16, 593–604. <https://doi.org/10.1108/14777830510623664>
- Szczepaniak, K., & Biziuk, M. (2003). Aspects of the biomonitoring studies using mosses and lichens as indicators of metal pollution. *Environmental Research*, 93, 221–230. [https://doi.org/10.1016/S0013-9351\(03\)00141-5](https://doi.org/10.1016/S0013-9351(03)00141-5)
- Takoutsing, B., Weber, J., Aynekulu, E., Rodríguez Martín, J. A., Shepherd, K., Sila, A., ... Diby, L. (2016). Assessment of soil health indicators for sustainable production of maize in smallholder farming systems in the highlands of Cameroon. *Geoderma*, 276, 64–73. <https://doi.org/10.1016/j.geoderma.2016.04.027>
- Toujaguez, R., Ono, F. B., Martins, V., Cabrera, P. P., Blanco, A. V., Bundschuh, J., & Guilherme, L. R. G. (2013). Arsenic bioaccessibility in gold mine tailings of Delita, Cuba. *Journal of Hazardous Materials*, 262, 1004–1013. <https://doi.org/10.1016/j.jhazmat.2013.01.045>
- Veiga, M. M., Nunes, D., Klein, B., Shandro, J. A., Velasquez, P. C., & Sousa, R. N. (2009). Mill leaching: A viable substitute for mercury amalgamation in the artisanal gold mining sector? *Journal of Cleaner Production*, 17, 1373–1381. <https://doi.org/10.1016/j.jclepro.2009.03.012>
- Zamani-Ahmadm Mahmoodi, R., Bakhtiari, A. R., & Rodríguez Martín, J. A. (2014). Spatial relations of mercury contents in Pike (*Esox lucius*) and sediments concentration of the Anzali wetland, along the southern shores of the Caspian Sea, Iran. *Marine Pollution Bulletin*, 84, 97–103. <https://doi.org/10.1016/j.marpolbul.2014.05.030>

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