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# Small-scale Gold Mining in the Puyango River Basin, Southern Ecuador: A Study of Environmental Impacts and Human Exposures

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Abstract: This study addresses the impact of small-scale gold mining on the environment and communities of the Puyango River basin in the southwest of Ecuador between 1999 and 2001. Our primary objectives were to measure mercury, manganese, and lead in the river, to identify pathways of population exposure, and biological indicators of human exposure. A multi-method design at the catchment scale was applied to the analysis of water, sediments, and particulate matter for mercury, lead, and manganese over two different seasons. A household survey was undertaken, and individuals were assessed for lead (blood samples), and for mercury (urine and hair samples). River water samples had high concentrations of particulate matter (700 mg/L). Mercury and lead levels in water at the gold processing plants were very high in both seasons (250 ng Hg/L and 160 µg Pb/L). Mercury and lead, there was a downstream gradient with the areas adjacent to the gold processing plants having the highest levels. In Portovelo, the Upper Basin city, 14% of the population reported occupational exposure to inorganic mercury. Although no one in Portovelo consumes water from the river, 10% of the population consume local fish. This contrasts the Lower Basin where 98% of the population consume fish, and 100% use river water for drinking and cooking. Lead blood levels over 20 µg/dl were found in 39.4% of the study population and blood mercury over 10 µg/L only in 10.0%. Urinary mercury over 4 µg/L was found in 66.5% of the examined persons. Hair methylmercury did not exceed the safe limit of 2 µg/g, the mean concentration was 1.2 µg/g. Mining activity and erosion contributes to heavy-metal contamination (mercury, lead, and manganese) throughout the Puyango Basin. The relation between environmental contamination is complex and further research is being conducted to understand these relations.

Key words: mining, environmental impacts, mercury, lead, manganese, Ecuador

#### INTRODUCTION

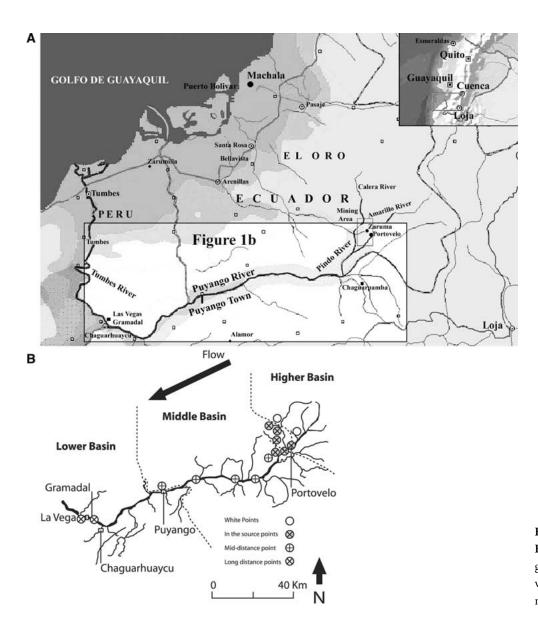
Mining activity in Zaruma-Portovelo, Ecuador, dates back more than 500 years. More recently, since the 1980s, there

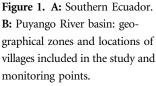
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has been a boom in small-scale gold mining and it is responsible for high discharges of pollutants into the Puyango River and tributaries (CENDA, 1996; Fredrikson, 1997; Tarras-Wahlberg, 2001; Torres, 1991; von-Trümpling, 1994, Prodeminca, 1999).

Previous research concentrated on the assessment of environmental impacts near the gold-mining areas in the

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Upper Basin of this ecosystem (CENDA, 1996; Prodeminca, 1999). Tarras (2001) found high levels of lead and cadmium in the larvae of insects several kilometers below the processing plants. Recent studies, especially in South America, have shown that many riverside ecosystems and fish are contaminated with mercury (Lebel et al., 1997; Malm, 1998; Roulet et al., 1998a, 1999, 2000).

The present study examines the impact of gold mining on the environment, human exposures, and biological indicators, using an ecosystem perspective and other catchment-based approaches to analyzing the links between biophysical and social dimensions of health (Lebel, 2003; Parkes et al., 2004; Rapport and Megler, 2004). The study covers the entire Puyango River basin (110 km in length), targeting not only the area of mining activity located in the Upper Basin (Zaruma and Portovelo), but also small downstream communities (Puyango Viejo, Gramadal-Las Vegas, and Chaguarhuaycu) (Fig. 1).

## **M**ethods

#### **Study Site and Population**

The Puyango River traverses southern Ecuador from northeast to southwest from the Andes' western foothills to the border with Peru, where it changes its name to the Tumbes River, before finally reaching the Pacific Ocean (Fig. 1). Our study includes only the Puyango River basin.

Based on characteristics such as altitude and geological conditions, the Puyango River basin can be divided into three zones (Fig. 1): Upper, Middle, and Lower Basins. The Upper Basin (Zaruma and Portovelo) is a temperate humid region. It is the most important gold mining district in Ecuador. The Middle (Puyango Viejo and Marcabelí) Basin is a sub-tropical humid region, while the Lower Basin (Gramadal-Las Vegas and Chaguarhuaycu) is a sub-tropical dry region. Zaruma and Portovelo counties (Upper Basin) has 41,375 inhabitants but the middle and lower parts of the Basin are thinly populated. The river is an important source of fish and water for communities living in the Lower Basin. Agriculture and cattle are the predominant economic activities in the Lower and Middle Basin, aside from artisanal and corporate small-scale gold mining.

#### **River Sampling Methods**

An environmental sampling network comprising 15 points was established along the river (Fig. 1). Pristine Stations, free of mining activity and contaminant emissions, were located upstream of the mining activities as control or reference points.

One sample of sediments, particulate matter and filtered water were collected twice annually at each station: in March (high water levels during the rainy season) and again in November 2000 (lowest water levels at the end of the dry season).

Water was sampled following the protocol described by Roulet et al. (2001). In brief, two fractions were considered at each site: filtered water and suspended particulate matter (Fig. 1). Following the clean handsdirty hands protocol (Montgomery et al., 1995), filtered water was sampled using a manual peristaltic pump and an in-line filter holder equipped with a glass fiber filter (0.2-µm pore size, EPM2000). Filtered water was collected in a 60-mL Teflon flask for mercury analysis and in a 125-mL polypropylene flask for lead and manganese analysis. Samples were then preserved with the addition of ultra pure nitric acid and refrigerated. Suspended particulate matter (SPM) was collected on combusted (300°C) and pre-weighed glass fiber filters (0.7 µm; Whatman GFF, Brentford, London, UK) and on Polycarbonate membrane (0.45 µm). Samples of bottom sediments were collected at the same sites. The surface sediment (about 1-cm depth) was sampled using a 10-cm diameter petri dish and then passed through nylon 250µm sieve. Suspended particulate matter and sediment were dried on an oven at 45°C.

#### Laboratory Analyses

Mercury in the filtered water was assessed at the University of Quebec in Montreal (UQAM) by cold vapor atomic fluorescence spectrophotometry (CVAFS), using a modified version of the method developed by Bloom and Fitzgerald (1988) and described by Pichet et al. (1999). Mercury in filtered water samples, operationally called dissolved mercury (Hg-D), was measured by direct injection of a 4-ml sub-sample following a 30-minute photochemical (Ultraviolet Violet light) digestion in the presence of potassium persulfate.

Lead in the filtered water was analyzed by graphite furnace atomic adsorption spectrophotometry (GFAAS) at the UQAM laboratory. Manganese in filtered water was analyzed at the XRAL–SGS laboratory (Toronto, Canada). All of the SPM and sediment samples were dried in an oven at 45°C.

Lead and manganese in both sediment and SPM were analyzed by ICP-MS following Aqua Regia digestion at the XRAL–SGS laboratory. Mercury was analyzed by CVAFS at the same laboratory.

#### Household Survey

A household survey was conducted involving the participation of 241 households (families): 178 from Portovelo City, representing 11.6% of the total families in this city, and the 100% of families from Puyango, Gramadal-Las Vegas, and Chauarhuaycu (32, 19, and 12 families, respectively) (Table 1). The sample size sought in Portovelo was estimated by the EPI-INFO program (Centers for Diesease Control, Atlanta, GA; version 6.04) using the following parameters: 1,538 estimated families, 53% of them use river water for cooking and drinking (i.e., the prevalence of exposure), a 5% level of significance, and an 80% power. This resulted in an estimated sample of 119 families. In the Middle and Lower Basin communities, all who agreed to participate in the study were recruited (n =318 persons in 63 families).

#### **Bioindicators of Exposure**

Exposure was studied in a sub-sample of 215 persons ranging from 18 to 65 years of age. At Portovelo, 95 families were sampled from the 178 that participated in the household survey. One person was recruited from each family. In the Middle and Lower Basins, all the inhabitants who agreed to participate were included in the household study (Table 1).

Community			Household	survey	Individual exposure study		
	Estimated population			Persons	Persons	Persons	
	Families	Persons	Families	(all ages)	(18-65 years)	(18–65 years)	
Portovelo	1538	6290	178	728	402	95	
Puyango Viejo	32	150	32	150	84	62	
Gramadal/Las Vegas	19	93	19	93	45	24	
Chaguarhuaycu	12	75	12	75	46	34	
Total	1601	6608	241	1046	577	215	

Table 1. Population and Samples of Household Survey and Individual Exposure Study by Communities

There are some differences between individual and household survey, such as in the size of the population by community, occupation, and education levels, but both samples are similar by age and sex. Performing this same comparison for Portovelo, differences were found between age and occupation. The percentage of miners is less in the household survey and farmers did not participate in the individual study. No differences were found for duration of residence in both comparisons (Table 2).

Two blood specimens were drawn by venipuncture from 215 adults: 10 ml in a plain Monoject® tube (Tyco Healthcare, Mansfield, MA) for lead concentration measurement, and 5 ml in a Vacutainer<sup>TM</sup> tube (Becton Dickinson, Franklin Lakes, NJ) with K<sub>3</sub>EDTA for total mercury concentration measurement (T-Hg). Hair strands, 5–10 cm-long, were collected from 117 adults and stored in a zip-lock plastic bag for methylmercury analysis (difference between total and inorganic mercury). Urine samples were collected from 215 persons using sterile plastic receptacles and kept refrigerated. Total mercury was measured in each of these specimens.

The number of valid samples for lead in blood and mercury in urine were 213 and 214, respectively. Only 84 samples were analyzed for mercury in blood. After analyzing the first batch of samples of mercury in blood, these 84 samples were randomly selected from all the samples, since very low levels were found: 23.8% were below the detection limit: (3  $\mu$ g/L), and 10.0% were above 10  $\mu$ g/L (WHO, 1991). Mercury in hair samples was determined in 117 inhabitants of the Middle and Lower Basins. These tests were not performed on those from Portovelo because of the eventual external exposure to inorganic mercury from the processing plants, and because there is not river-fish consumption.

Lead in blood and mercury in urine were analyzed at the laboratories of the Ministry of Public Health in

Quito, Ecuador. The spectrophotometry-vis method was used with ditizone complex levels of 0.003 mg/100 ml for the detection of lead, and 0.003 mg/L for mercury. Mercury in blood and methylmercury in hair strands was analyzed at the laboratory of the Federal University of Rio de Janeiro, Brazil. Blood samples were digested by microwave, and hair strands by hot water bath. Determinations of blood and hair mercury were conducted by cold vapor atomic absorption spectrophotometry with the FIMS-400 (flow injection system of Perkin-Elmer, Wellesley, MA) for total mercury and by gas chromatography for methylmercury.

#### Statistical Analysis

EPI-INFO (version 6.04) statistical software package program was used to process the household survey data and that generated from the individual specimens. Percentages for qualitative variables were used.

## RESULTS

#### Characteristics of the Population

According to the household survey, 50.1% of 1046 persons studied were men. The mean age was 39.9 years; 51.4% of this population was aged between 19 and 65 years and only 5.4% were older than 65 years old.

According to household survey and the individualized study, 25% had more than 6 years of formal education (Table 2), with big differences among communities.

Agriculture and cattle are the prevalent economic activities in the Lower and Middle Basin (51% of the population), only 5% combines agriculture and mining. In

	Total communities <sup>a</sup>				Portovelo <sup>b</sup>			
	Household survey (n = 577)		Individual study (n = 215)		Household survey (n = 402)		Individual study (n = 95)	
Variables	No.	%	No.	%	No.	%	No.	%
Community								
Portovelo	402	69.7	95	44.2	402		95	
Puyango Viejo	84	14.6	62	28.8				
Gramadal	45	7.8	24	11.2				
Chaguarhuaycu	46	8.0	34	15.8				
Sex								
Female	295	51.1	110	51.2	213	47.0	51	53.7
Male	282	48.9	105	48.8	189	53.0	44	46.3
Age (years)								
18–25	159	27.6	48	22.3	106	26.4	18	18.9
26–45	283	49.0	101	46.9	210	52.2	55	57.9
46–65	135	23.4	66	30.7	86	21.4	22	23.2
Education								
Illiterate	4	0.7	3	1.4		0.3		2.1
Incomplete elementary	70	12.1	115	20.0	24	6.0	10	10.5
Complete elementary	218	37.8	43	53.5	121	30.1	35	36.8
Incomplete high school	97	16.8	24	8.4	80	19.9	15	15.8
Complete high school	22	3.8	12	5.6	70	17.4	22	23.2
Undergraduate school	24	4.2	18	11.2	42	12.4	11	11.6
Occupation								
Mining	73	12.7	32	14.9	72	17.9	32	33.7
Agriculture and mining	16	2.8	28	13.0	8	2.0	_	
Other and mining	14	2.4	13	6.0	14	3.5	12	12.8
Agriculture	96	16.6	32	14.9	14	3.5	_	
Housekeeper	203	35.2	90	41.9	137	34.1	34	35.8
Other	175	30.3	20	9.3	157	39.1	17	17.9
Time of residence, mean (SD)	20.3	(15.0)	27.4	(14.4)	16.9	(15.3)	24.6	(15.8)

 Table 2.
 Demographic and Socio-economic Characteristics of Household and Individual Exposure Study Population by Total Communities and Portovelo

<sup>a</sup>Portovelo is included in total communities.

<sup>b</sup>This column highlights the demographic differences of Portovelo (Upper Basin) from Middle and Lower Basin communities.

Portovelo, mining is predominant (31.1% of the overall population are involved in mining), only 4.3% of the population is in the agriculture and cattle activities.

# Mercury, Lead, and Manganese along the Puyango River System

Suspended particulate matter (SPM) showed different patterns in the high-water season as compared with the low-water season. Figure 2 shows a remarkable contribution of ore processing plants (tailings) to the concentration of particulate matter in water, which in the dry season (November 2000) diminishes abruptly downstream. The concentration of SPM in the mining zone is very elevated (up to 700 mg/L). It increases steeply in comparison to the pristine zone upstream (3 mg/L). Downstream, at the confluence of the Amarillo and Calera Rivers, the SPM decreases progressively from 300 mg/L to 20 mg/L in the Gramadal sector. During the rainy season (March 2000) the pattern is reversed: there is an increase in concentration with distance from 140 mg/L to 430 mg/L. As seen in November 2000, a rapid increase of particulate matter

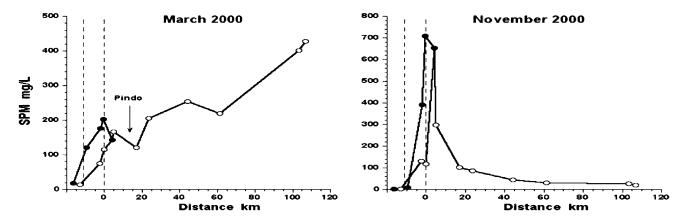


Figure 2. Suspended particulate matter (SPM) in the Calera (closed circles) and Amarillo (open circles) Rivers, during rainy (March) and dry (November) seasons.

concentrations from the upstream pristine zone (15 mg/L) to the mining zone (200 mg/L) is observed. But between the confluence of the Calera and Amarillo Rivers and the Gramadal sector, particulate matter continues to increase progressively from 140 mg/L to 430 mg/L.

Because particulate matter contains 70%–100% of the total load of manganese, lead, and mercury in water (Roulet et al., 2001), the spatio-temporal variation of SPM determined the transport of metals in the river system (Fig. 3). During the dry season, concentrations of particulate mercury, lead, and manganese follow those of suspended particulate matter. Concentrations decrease exponentially from the mining sectors to attain 110 ng/L for mercury, 50  $\mu$ g/L for lead, and 75  $\mu$ g/L for manganese, in the Gramadal sector 100 km downstream. The increase downstream of the confluence of the Calera and the Amarillo Rivers during the rainy season is responsible for the same increase in manganese concentrations (160–340  $\mu$ g/L) and for high concentrations of mercury (80–250 ng/L) and lead (75–160  $\mu$ g/L) in water of the Puyango River.

Lead and manganese reach levels above the limits considered safe for human consumption 10  $\mu$ g Pb/L (WHO, 1995) and 50  $\mu$ g Mn/L (Drinking Water Services, 2005). Except for a peak in the Calera River, near processing plants, mercury was found within safe limits (Fig. 3)

#### Human Exposure Pathways

Results from the household survey showed that none of the families from the Portovelo and Chahuarhuaycu consume river water in any form, while in Gramadal-Las Vegas, 100% of the families consume river water. In the dry season, 67.8% of the families downstream drink the river water without

any treatment. The fish consumption pattern is similar to the water consumption pattern. For occupationally related exposure, 12.7% work as miners and 5.2% combine other activities with mining; 73% of these persons manipulate mercury in the amalgamation process.

For the individual survey, 55.3% of surveyed persons consume fish from Puyango River or its tributaries, 11.6% consume river water (drinking or cooking), 31.2% are exposed to mercury in ore processing, 27% are exposed to pesticides and 14.9% and 31.6% consume tobacco and alcohol, respectively (Table 3).

#### **Biological Indicators of Exposure**

The means of lead blood, blood mercury, and urinary mercury were 23  $\mu$ g/dl (SD = 22), 5.5  $\mu$ g/L (SD = 5.09), and 12  $\mu$ g/L (SD = 14), respectively. Lead blood levels over 20  $\mu$ g/dl (WHO, 1995) were found in 39.4% of the study population and blood mercury over 10  $\mu$ g/L (WHO, 1991) only in 10.0%. Urinary mercury over 4  $\mu$ g/L was found in 66.5% of the examined persons. Hair methylmercury did not exceed the safe limit of 2  $\mu$ g/g (WHO, 1990). The mean concentration was 1.21  $\mu$ g/g (SD = 0.87) and the maximum concentration was 6.09  $\mu$ g/g.

#### DISCUSSION

# Heavy Metals and the Dynamics of the Puyango River Basin

The findings of this study demonstrated that the mining activity and erosion (which varies depending on the dry or rainy season) modify the composition of the river

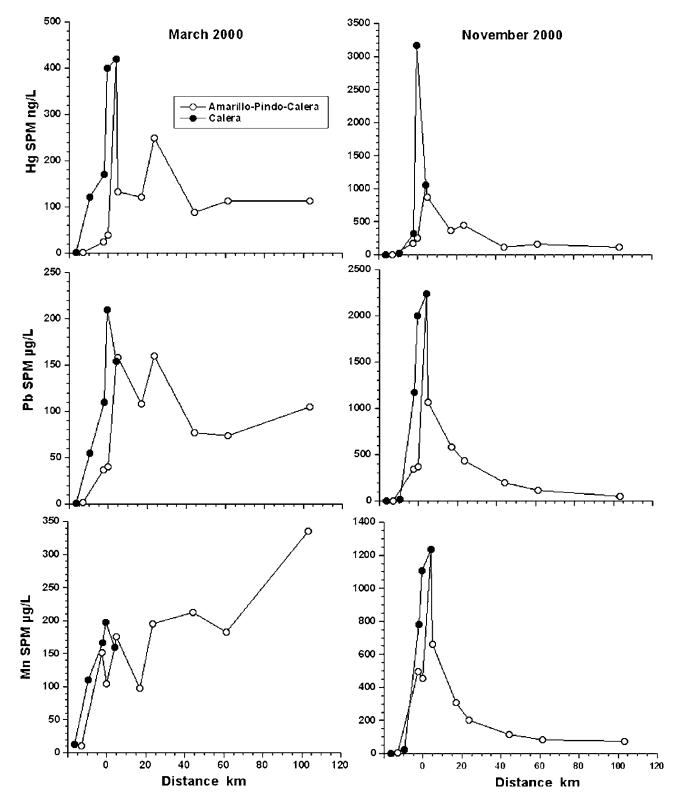


Figure 3. Mercury (Hg), lead (Pb), and manganese (Mn) concentrations in nonfiltered water.

water, including heavy metals. The proposal that mining activity and soil erosion on the Puyango River are drivers of particulate matter in the river is consistent with other studies in South America (Roulet and Lucotte, 1995; Roulet et al., 1998a, 2000; Guimarães et al., 2000).

This study shows that during both seasons, the concentrations of mercury and lead in the particulate matter

Exposure factors	No.	%	
Fish consumption			
Yes	119	55.3	
No	96	44.7	
Water consumption			
Never or rare	181	84.2	
Daily year or seasonal	25	11.6	
Missing value	9	4.2	
Exposure to mercury amalgamation			
Yes	67	31.2	
No	148	68.8	
Exposure to pesticides			
Yes	58	27.0	
No	157	73.0	
Tobacco consumption			
Yes	32	14.9	
No	183	85.1	
Alcohol consumption			
Yes	68	31.6	
No	147	68.4	

**Table 3.** Exposure Factors of Individual Exposure Study Population, Puyango Basin, 2002 (n = 215)

are high (mercury: 80–250 ng/L, and lead: 75–160  $\mu$ g/L). High lead concentrations are particularly alarming as they often exceed the reference level for human water consumption (10  $\mu$ g/L) (WHO, 1995).

Average water flows at the Puyango Viejo hydrological station of the National Institute of Meteorology and Hydrology (INAMHI) are 416.9 m<sup>3</sup>/s and 13.7 m<sup>3</sup>/s for rainy and dry seasons, respectively, for the year 2000 (INAMHI, 2000). During the rainy season, transport of mercury, lead, and manganese at Puyango Viejo increases by a factor of 20 for mercury and lead, and 70 for manganese in comparison to the dry season (Roulet et al., 2001).

The tributaries of the Puyango River appear to make an important contribution to the concentrations of mercury, lead, and manganese in the Middle and Lower Basin during the rainy season. This suggests a sweeping effect of rainfall and run-off from soils with a high lead content—similar to those observed in the upper part of the basin (Valarezo et al., 1998; Prodeminca, 1999). There appears to be an important environmental contribution to the load of mercury, lead, and manganese into the Puyango River, brought about by the soil-usage patterns prevailing in this region (i.e., mining and agriculture). This is consistent with findings for mercury in the Amazonian tropical forest (Roulet et al., 1998b, 1999). To date, however, studies of heavy metals performed in the Amazonian basin have not included testing for lead contamination (Lebel et al., 1997; Malm, 1998; Roulet et al., 1998a,b, 1999, 2000; Guimarães, 2000). The new study design and findings presented in this study have guided the new investigations in the Puyango River basin, to determine heavy metals (mercury, lead, and manganese) in the soil of micro-basins (FUNSAD, 2003).

The high levels of lead in the Puyango River found in this study are similar to those found in the Tarras report (Tarras, 2001). The results of this study suggest that high levels of lead are derived mainly from natural geological sources. (CEN-DA, 1996; Valarezo, 1998; PRODEMINCA, 1999).

### Heavy-Metal Exposure Events in Puyango River Basin Communities

Since the population in Portovelo (Upper Basin) does not consume fish or water from the river, it appears that heavymetal exposure is mostly related to mining activity. In the Lower Basin, were the river is a vital resource for the inhabitants, the people consume local fish and river water, especially in summer.

In this study, high levels of mercury in blood and hair were not found—which contrasts with other studies in Latin America (Lebel, 1996; Lebel et al., 1997; Dolbec et al., 2000). The absence of high levels of mercury in hair strand could be explained by several hypotheses such as:

- The Puyango River basin does not offer adequate conditions for the methylation of mercury, in contrast to what regularly occurs in the Amazonian rivers where relatively high levels of methylation occur in the floodplain; in this hot, dry, and mountainous region, apparently, there are not conditions for methylation of inorganic mercury (low organic matter and low microbial activity).
- Fishes in Puyango River are smaller than Amazonian rivers.
- There is less fish consumption in this area than Amazonian region.

Each of these hypotheses are currently being tested in a follow-up study being conducted by FUNSAD. This research has been limited by the absence of in-country, heavy-metal, laboratory analysis capabilities for environmental and human samples. Sending samples to laboratories abroad presents challenges to maintaining the quality of the samples, and limits comparisons of results among laboratories.

#### CONCLUSIONS

This study has focused on a distinct ecological system defined by a river basin to investigate possible links between a critical natural resource-based economic activity (mining and agriculture), river ecological processes (i.e., erosion and sedimentation, heavy-metal uptake by the food chain, etc.) and human health. The apparent relationships between the river basin's variable hydroecological dynamics, heavy-metal fluxes, and human health has proven most intriguing and potentially significant. However, the complexity of relationships between these catchment ecosystems, including their social processes and the health effects requires further investigation to clarify the mechanisms and public health significance.

The sources of the metal and exposure pathways appear to differ between catchments. For example, contrary to literature from other areas that informed the initial study planning and design, the presence of lead in the Puyango River basin requires further study. The high levels of blood lead suggest other sources of river contamination. Possibilities include smoke and dust produced in different mining processes, as well as occupational exposures from welding and the domestic use of pottery with lead.

Our investigation of the social and ecological factors responsible for the heavy-metal contamination problem in the Puyango River basin has produced additional benefits besides increased scientific information. It has fostered the mobilization of local and national stakeholders of Ecuador. This has been a result of the many workshops held involving the participation of mining with health and mining authorities for the purpose of reviewing the study results and identifying remediation strategies based on environmental management approaches. Among the most important results has been a plan to reduce the mining pollution and the Environmental Management Units (UGAMs) creation in Zaruma and Portovelo counties.

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