Technical Communication

Heavy Metals in Stream Sediments from the Coquimbo Region (Chile): Effects of Sustained Mining and Natural Processes in a Semi-arid Andean Basin

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Abstract. Active sediments from the Elqui River in Chile were sampled 4 times at 10 sites during 2000. Concentrations of Ag, Ba, Cd, Co, Cr, Hg, Mn, Mo, Ni, Pb, Sr, Ti, V, Al, Ca, Fe, K, Mg, Na, P, and S were normal. Zinc levels were clearly high, and those of Cu (hundred to thousands ppm) and As (tens to hundreds ppm) were highly anomalous. Dissolved Cu (0.1-12.7 ppm) and Zn (0.1-2.2 ppm) levels were also very high. The anomalies of the upper tributaries are due to the El Indio-Tambo Au-Cu-As district and large hydrothermal alteration zones at altitudes between 3500-4500 m. Lower on the river, old and active tailing waste deposits and on-going mining operations in the Talcuna Cu (Pb) district are responsible. Partially eroded tailing deposits in the alluvial plain of the Elqui River and its tributaries, and especially in the El Indio-Tambo district, after mine closure in 2000, warrant special attention.

Key words: Arsenic; contamination; copper; Elqui, Chile; gold-copper mining; stream sediments; watershed; zinc

Introduction

Drainage water and sediment geochemical studies began in Chile in the 1960s (c.f. de Grys 1961, 1962) and proliferated during the following decades. Most of these studies were carried out by mining companies for exploration purposes, though some are available and can provide useful baseline information for environmental studies. In contrast, a great deal of geochemical research has been performed in Poland and published at different scales for environmental assessment and land use purposes (e.g. Lis and Pasieczna 1995, 1999a, 1999b). This communication describes the results of a joint research program in the Elqui River basin, Coquimbo, Chile, which was carried out by the University of La Serena, Chile, the Geological Institute of Poland, and the Academy of Mining and Metallurgy of Cracow (Poland). The main purpose of this research was to assess the level of heavy metal pollution in active drainage sediments of the Elqui River and its tributaries, an area that was mined for Cu, Ag, Au, and Hg during the 19th and 20th centuries. This information is relevant to the

various users of these water resources (agriculture, municipalities, etc.) and may constitute a useful baseline for future studies. The latter is important because the Puclaro irrigation dam, in the middle course of the Elqui River, with a capacity of 200 Mm³, was inaugurated in 1999, and the El Tambo (Au) and El Indio (Au-Cu-As) mines, in the NE part of the Elqui watershed, began their closure-plan activities in 1999-2000. Both actions have the potential to induce significant changes in the geochemistry of the Elqui River.

The Elqui Watershed

The Elqui watershed encompasses an area of 9,794 km² and is located in the so-called Transversal Valleys segment (28-33° S) of the Chilean territory, which lacks the presence of a central N-S tectonic basin. The Elqui River flows westward from the Andes Mountains to the Pacific Ocean, while its main tributaries, the Claro and Turbio Rivers, channel water coming from the north, south, and east. The Chilean territory in this realm is narrow and the high peaks of the Chilean-Argentinean border, attaining over 5000 m, are only some 130 km from the coast. Consequently, the Claro and Turbio Rivers present steep longitudinal profiles and very narrow alluvial plains. In contrast, the Elqui River plain and terraces are about 3 km wide near the town of Vicuña, and 5 to 6 km wide for the last 25 km of the river course.

Precipitation (including snowfall) in the Elqui watershed is about 100 mm/year. However, in years affected by El Niño oceanic temperature disturbances, precipitation may increase by a factor of 2 or 3 (INE 1998). Precipitation is greater in the high Andes, with a mean of 180 mm over a period of 20 years, and a maximum of 740 mm in 1987, an El Niño year (H. Zavala, pers. comm.). Except for the Andean sector, where some rain may occur during summer, rain or snowfall in the Elqui basin is restricted to the April – September period (autumn-winter). During the year 2000, the main precipitation events occurred in May (6%), June (56%), July (7%) and September (30%), totaling 103 mm at the La Florida meteorological station (La Serena).

The geology of the Elqui basin (Figure 1) is dominated by calc-alkaline intermediate volcanic rocks of Mesozoic and Cenozoic age, intercalated with sedimentary rocks of a similar clastic lithology. Tonalitic to granodioritic intrusive rocks of Mesozoic to Tertiary age intrude the stratigraphic sequence (SERNAGEOMIN 1982). The chemical composition of the rocks is rather homogeneous for the whole basin. These rocks host hydrothermal ore deposits of a number of metals, although only copper, gold, silver, and, to a lesser extent, manganese, have had economic importance. Silver deposits were mined during the 19th Century and the first half of the 20th Century. After the recent closure of the El Indio gold(+Cu-As) district and the manto-type gold mines of Andacollo, copper is the only metal currently being mined in the region (Andacollo and Talcuna districts) (Figure 1). However, two centuries of intensive and extensive mining and metallurgical operations in the Elqui watershed have left a heritage of abandoned mines and piles of mineral wastes. Tailing deposits left by the copper operations are especially significant. The waste deposits are typically located

close to rivers or ravines, and are subjected to erosion and mass movement during flooding events induced by strong El Niño episodes (once or twice every 10 years). The Talcuna copper mining operations (Boric 1985) (Figure 1) are located within a very narrow valley (Marquesa Gulch) and as such, are very vulnerable to these episodes, each time contributing large amounts of tailings to the Elqui River. The chemical composition of the Talcuna district wastes (Table 1) is also worrisome, since about 1.5 Mt of unprotected tailing wastes are deposited in the valley. In addition to contamination episodes related to flood events. mismanagement introduces additional environmental problems. For example, on the 22nd of September and the 8th of November 2002, several thousands tons of tailings from the Marquesa Gulch polluted the Elqui River, forcing a drastic reduction in the water supply to La Serena. These incidents also affected the irrigation of the rich alluvial agricultural terrain of the Elqui River's lower course and damaged valuable plantations in the valley. Additional environmental threats in the Elqui watershed are provided by the El Indio - Tambo

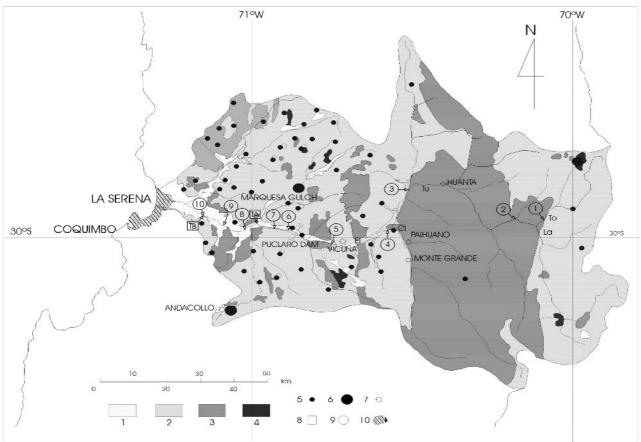


Figure 1: Lithological units, mineral deposits, sampling stations, and towns of the Elqui River basin:
1. Quaternary sediments; 2. Mesozoic and Cenozoic volcanic and sedimentary rocks; 3. Paleozoic and Mesozoic granitic to dioritic batholiths; 4. hydrothermal alteration zones; 5. small mineral deposits; 6. medium to large mineral deposits; 7. town. 8. sampled tailing deposit; 9. sampling station; 10. cities of Coquimbo-La Serena; Rivers: To=Toro; La=Laguna; Tu=Turbio; Cl=Claro; El=Elqui. Geographical base after IGM map (1983), lithological units according to SERNAGEOMIN (1982), and mineral deposits from Ulriksen (1990)

Table 1: Average annual trace elements contents of the sediment at the 10 sampling stations (SS) and reference data from Chilean and Polish streams in mg/kg (ppm)

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SS	Ba	Cr	Mo	Mn	Co	Ni	Cu	Zn	Cd	Hg	Pb	As
1	82	9	2	657	12	6	350	105	< 0.5	0.062	20	209
2	89	10	1	1196	21	14	1086	369	1.1	0.084	18	178
3	116	13	2	1311	27	21	1956	610	1.8	0.088	19	174
4	103	11	1	1309	24	17	1731	508	1.6	0.125	20	145
5	110	12	<1	1242	22	15	1794	465	1.3	0.113	19	130
6	159	16	<1	1122	19	13	1258	353	2.0	0.125	13	68
7	127	14	<1	1286	18	11	1025	290	1.8	0.145	12	56
8	730	15	<1	1103	14	10	664	229	0.8	0.107	56	41
9	644	13	<1	1304	13	9	498	171	0.7	0.163	43	29
10	663	17	<1	978	11	8	405	161	0.8	0.104	44	26
						_						_ v
Av	282	13	1	1151	18	12	1077	326	1.2	0.112	26	106
			-				10,,			*****		100
TA	345	68	4	2729	13	23	8209	1515	15	1.42	760	72
TB	342	5	<1	987	21	14	2216	130	< 0.5	0.153	34	31
12	5.2	Ü	•	507	_ 1		2210	150	0.0	0.100	٥.	31
CH-1							40	25			25	
CH-2	46	63			19	27	135	55			3	
CH-3	.0	62		1300	18	24	50	105			5	
PO	80	18		506	4	11	21	247	2.8	0.06	68	7
Wav	460	72	2	770	14	52	33	95	0.2	0.19	19	8
	700	12		, , , ,	1.7	32			0.2	0.17	(7)	

SS: Sampling station; Av: total average content (40 analyses); TA and TB: tailing deposits (Figure 1); CH-1: average content for Chilean Andean rivers between 34° and 41° S (de Grys 1961); CH-2: average content for desert creeks sediments in the Coastal Cordillera, between 22° 41' and 23° 21' S (Arias et al. 1989); CH-3: average content of Mesozoic and Cenozoic volcanic rocks of Chile (Oyarzún 1971); PO: average content of Polish river sediments (Lis and Pasieczna 1995); WAV: worldwide average content of sediments (Bowen 1979, in Sparks 1995).

gold-copper-arsenic district (Jannas and Araneda 1985), located in the headwaters of the Rio del Toro in the high Andes, above 3500 m of altitude. The small Malo River drains the district and flows into the Toro River, and is characterized by low pH and high dissolved and suspended As, Cu, and Zn. This is consistent with: 1) the enargite- rich nature of the El Indio ores; 2) the advanced argillic alteration of the volcanic rocks of the district; and 3) the existence of hot, As-rich (20 ppm) thermal waters in the center of the El Indio area (Baños del Toro). In addition to the natural Cu and Zn enrichment and the acidity of the Malo and Toro rivers, mining in the district is responsible for a huge increase in the rock-water-air contact surface area (e.g., 42 km of tunnels were constructed at the El Indio mine from 1976 to 1984). The interactions responsible for acid drainage have thus been greatly enhanced, a worrisome factor considering that pH values of 1.0-2.0 were normal in the El Indio mine water during the operation of the underground mine. The geotechnical nature of the rock massif has also been damaged. Furthermore, the district hosts ~22 Mt of tailings (El Indio) and ~12 Mt of heap leaching wastes (Tambo). These are low grade or barren, but are chemically active. Although the company that owns the El Indio-Tambo mines (Barrick Gold) has invested large amounts of money (>50 million US\$) in closure plans, a number of factors are worrisome. Among them are the structural

and lithological characteristics of the host-rocks, the natural conditions for acid generation, the presence of As in the ore minerals and barren rocks and soils, and the fact that the district is at high altitude, at the head of an Elqui River tributary, the Toro River.

Gold has been mined in the Andacollo district (Reyes 1991; Oyarzun et al. 1996) since pre-Colonial times, and copper was mined from 1950 to 1970. Later, small-scale mining decreased, leaving behind tailings and copper leaching wastes, many of them in the town of Andacollo. Two medium-sized open pit operations (10,000 t/day) were opened in Andacollo in 1996: the Dayton gold and the Carmen copper mines. Both operations use hydrometallurgical processes: sodium cyanide at Dayton and sulfuric acid at the Carmen mine (followed by SX-EM). Dayton closed its mine in 2000 due to low gold prices, though metal recovery continues from the leaching piles. Carmen is still active.

The third important mining district is Talcuna, where two companies produce copper sulfide concentrates from ores with minor Pb contents, at a rate of about 2000 t/day. The tailings are deposited in the narrow and steep Talcuna Gulch, where periodic flooding caused by heavy rains and poor tailings management practices have been responsible for many episodes of contamination in this tributary of the Elqui River.

Apart from the mining districts already mentioned, a large number of ore deposits (Cu, Ag, Au ores, with minor contents of Zn, Pb, As, etc.) were mined during the 19th and 20th centuries. Most of these former operations, and the associated metallurgical facilities, were located in the western half of the watershed (Figure 1). These operations have left a heritage of barren and low-grade material, tailings of the sulfide concentration processes, acid (Cu) and alkaline cyanide (Au) bearing leach pile wastes, and minor residues of mercury related to gold recovery. Also, the cavities left by mining collect groundwater, enhancing water-rock interaction, the transport of soluble salts containing heavy metals, and in some cases, the generation of acid drainage.

It is important to highlight the fact that the mining industry coexists with a very important agricultural sector. Also, the Elqui River provides the water for three major towns, with a total population of about 350,000 inhabitants.

Methods

The Elqui River and its principal tributaries were sampled at 10 stations, selected according to regular spacing, accessibility, and presence of active fluvial sediments, rich in the silt fraction. Each point was sampled four times during the year 2000, on January 13th and 14th (Summer), April 14th (Autumn), July 14th (Winter), and October 18th (Spring), producing a total of 40 samples. Each location was carefully spotted and photographed in order to take the sample at the same place every time. The samples (1-2 kg each) were taken along a 50 m line of the riverside, to avoid the effect of local concentrations. Also, water samples were collected at each locality, for pH, conductivity, and Cu and Zn analysis. Apart from the stream sediments, two tailing deposits from former sulfide copper minerals concentration plants were sampled (Figure 1; TA and TB).

The samples were dried on protected porcelain plates and then sieved at the -60 mesh fraction, using a fiber sieve. 100 g of each of the previously quartered material were sent to the Polish Geological Institute in Warsaw and analyzed by ICP (inductively coupled plasma) (Lis and Pasieczna 1995). This paper reports and discusses the results obtained for Ba, Cr, Mo, Mn, Co, Ni, Cu., Zn, Cd, Hg, Pb, and As. Detection limit for these elements is 1 ppm, except for Hg (0.03 ppm), Cd (0.5 ppm), and Pb and As (5 ppm). Cu and Zn were also analyzed in the water samples by the Geoanalítica laboratory (Chile), using AA (atomic absorption), with a detection limit of 0.01 ppm for both elements.

Results and Discussion

The majority of the elements were present in the stream sediments at concentrations close to that of Chilean igneous rocks (Oyarzún 1971; Oyarzún et al. 1993). However, the average Zn content for the 10 stations was 326 ppm, and those of Cu and As, 1077 and 106 ppm, respectively. These figures are very high when compared to average concentrations in stream sediments of Chile, Poland, or the world in general (Table 1).

Copper and zinc display a similar distribution pattern, defined by an increase from stations 1 to 3, followed by sustained high levels at stations 4, 5 and 6 for Cu, and 4 and 5 for Zn. After station 6, both elements decrease; however, they still display higher than normal values (Cu: 350-752; Zn 105-231). The pH values, together with that of dilution, and the possible role of the Puclaro Dam, may explain this trend. As shown in Table 2, Cu and Zn content in the sediments seem to be controlled by the local pH of the stream waters. As the river water pH increases, Cu and Zn are transferred to the solid (sediment) phase. In consequence, pH is the first geochemical control for the high Cu and Zn content in water draining the El Indio district and neighboring hydrothermal alteration zones. Since both elements are linked to the silty fraction of the fluvial sediments, the water used for irrigation purposes probably introduces significant amounts of these metals to agricultural soils, which at present cannot be detected because water is usually filtered for chemical analyses. Except for the hydrolytic transfer of Cu and Zn from water to sediments, expressed by their increase in the solid phase between stations 1 to 3 (Figure 2), both elements present a parallel trend to that of As,

Table 2: Cu and Zn in sediments (s) (mg/kg) and water (w) (ppm), pH, and conductivity (mmhos/cm) at the 10 stations, summer and winter sampling

	1	2	3	4	5	6	7	8	9	10			
Summer:													
Cu (s)	361	612	1022	995	827	873	279	686	579	318			
Zn(s)	111	243	393	404	297	266	171	225	178	137			
pH(s)	5.2	8.2	8.3	8.1	8.1	7.9	8.1	8.0	7.9	8.0			
pH (w)	4.7	7.6	7.7	7.9	7.8	7.9	7.9	8.1	8.0	8.0			
Cond.	1.56	.47	.45	.45	.44	.54	.54	.54	.59	.79			
Cu (w)	9.1	0.7	1.0	0.6	0.4	0.9	0.9	0.3	8.2	3.0			
Zn (w)	2.2	0.2	0.3	0.2	0.2	0.1	0.2	0.1	0.2	0.1			
Winter:													
Cu (s)	352	1798	3301	2098	1630	111	5 166	0 578	3 508	406			
Zn(s)													
pH(s)	5.2	7.3	7.9	7.9	7.9	7.6	7.5	8.3	7.9	8.1			
pH (w)	4.7	5.7	5.9	6.4	6.6	6.8	6.8	6.9	7.0	7.1			
Cu (w)	12.7	4.5	1.8	1.6	0.8	0.2	0.1	0.2	0.2	0.2			
Zn (w)	2.0	0.9	0.6	0.4	0.3	0.1	0.2	0.1	0.2	0.4			

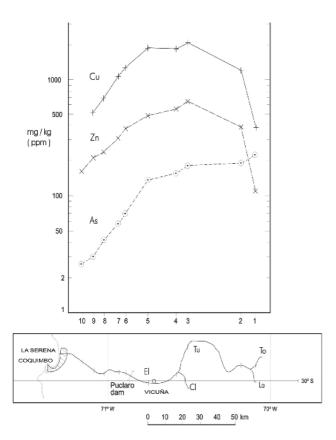


Figure 2: Average concentrations of Cu, Zn, and As, on a logarithmic scale, for the 10 sampling stations; To, La, Tu, Cl, and El indicate rivers (see Figure 1)

indicating a common principal source. The seasonal variation in Cu and As in the sediments (Figure 3) is remarkably parallel, despite their important differences in chemical behavior, which also suggests a common source.

The Puclaro reservoir appears to be a physical barrier and a sink for metals linked to the sediments. In fact, a considerable decrease in active sediments and suspended material occurs in the Elqui River downstream of the Puclaro dam. However, although Cu and Zn were relatively low at stations 6 to 10, there are other sources in this section. The main ones are the Talcuna district (Marquesa Gulch) and associated tailings and mineral dumps, which are exposed to fluvial erosion (see composition of the TA tailing deposit, in Table 1). It is likely that the high Cu content detected in the summer sampling at station 9 is related to this type of contamination. Note that due to the polymetallic affinities of the Talcuna ores, Pb and Ba contents increase by a factor of 2 and 5 respectively at stations 8, 9, and 10, downstream of Marquesa Gulch.

The As content of the sediments exhibit a gradual decrease from station 1 (average 209 ppm) to station

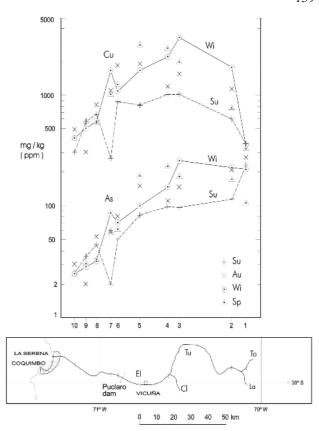


Figure 3: Seasonal variation of Cu and As on a logarithmic scale; Su: Summer; Au: Autumm; Wi: Winter; Sp: Spring.

10 (26 ppm), although a major break (from 130 ppm to 69 ppm) is observed between stations 5 and 6, which coincides with the Puclaro reservoir, which acts as an important sink for particulate As compounds. Although dissolved As was not analyzed, according to unpublished information provided by the DNA (National Water Authority), the average monthly content of this element is 0.320-0.380 ppm at station 1; 0.040-0.100 ppm at stations 2 and 3; 0.020-0.028 at stations 4 and 5 and 0.007-0.013 at stations 6 to 10. Again, arsenic decreases downstream, although by a factor of 30-40, instead of the factor of 8 registered for the average As content in the stream sediments between stations 1 and 10. According to the same source (DNA), the dissolved SO₄ ranges from 20.1 to 15.8 mg L⁻¹ at stations 1 to 3, then decreases to 5.2 at station 4 and to 0.9-1.0 mg L between stations 5 and 9. Finally, slight increases to 2.8 mg L⁻¹ are recorded at station 10.

Although dilution does not appear to play a role in explaining the variations of Cu and Zn in sediment of the Elqui River and its tributaries, the case could be different for As. For instance, assuming that most of the As is transferred by the Toro River, we should have lower values at station 2, i.e. after the La Laguna River meets the Toro. The latter has an average flow

of 0.8 ms⁻¹ (209 ppm As) and the La Laguna, flows at 1.7 ms⁻¹ (55-60 ppm As). If dilution was important, then we should expect As values of about 105 ppm at station 2; however, the measured value was 178 ppm. Furthermore, at station 3, where the river flows at about 6 m/s (after input from three other tributaries), the As average content is 174 ppm, practically the same as at station 2. Thus, dilution does not appear to play an important role for As, indicating multiple particulate sources of this element, at least in the Andean sector of the Elqui River basin.

Regarding the natural and anthropogenic sources of and Zn in the Toro hydrogeochemical survey carried out in 1974 before the opening of the El Indio mine detected dissolved As and Cu concentrations of 0.5 and 1.02 ppm respectively in a location corresponding to our station (Canut de Bon, unpublished report). geochemical maps produced during the exploration stage of El Indio district (Siddeley and Araneda 1986) also revealed the presence of extensive As anomalies (over 3500 ppm) close to the Malo River (a tributary of the Toro River). However, given the mining wastes and cavities left behind, it appears likely that ambient concentrations may be elevated after mine closure.

Conclusions

Of the 12 elements considered in this study, only As, Cu, and Zn attained very high levels. The El Indio hydrothermal alteration zone is a major source of As, Cu, and Zn for the Elqui River sediments and water, but it is not the only one. Although As, Cu, and Zn content in sediments vary significantly seasonally (with a minimum in summer and maximum in winter), their concentrations are always high to very high. Dissolved Cu and Zn are exceptionally high at station 1, and are also higher in winter for the first five stations; however, the opposite is observed for stations 6 to 10. Downstream from the Puclaro Dam (between stations 7 and 8), the Elqui River is under the influence of the Talcuna district and its tailing deposits. Thus, the Pb content doubles at stations 8, 9, and 10, downstream of the Marquesa Gulch. Chemical analysis of one of its tailing deposits at the Marquesa Gulch – Elqui River confluence (see TA, Table 1) revealed high to extremely high Cu, Zn, Pb, Cd, and Hg levels. This is important because these tailings are often eroded by heavy flooding during El Niño episodes, or undergo destabilization due to poor mining practices, e.g., two spills during 2002.

Considering the data obtained from this research, careful and rigorous monitoring and protection of the tailings should be implemented to assess the risk posed by their heavy metal content. Some of them,

like TB (see Figure 1, Table 1) may involve little or no risk; however, others such as TA, can be harmful and require remediation or protective barriers.

Regarding mine closure in the El Indio—Tambo district, special care is necessary, given the As-rich hydrothermal alteration, which is also a natural source of acid drainage. Mining has left surfaces for rock-air-water interaction, and a high risk of surface and ground water contamination. Finally, there are deposits of different types of mining wastes (tailing, low-grade ore piles, etc.) at low or high altitude within the Elqui watershed, which in turn sustains an important agricultural sector. Thus, there is a high potential to aggravate an already serious risk caused by natural and anthropogenic heavy metal sources.

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