Environ Geol (2007) 52:1243–1251
DOI 10.1007/s00254-006-0562-0

ORIGINAL ARTICLE

Mineralogy and heavy metal content in sediments from Rio Gato, Carelmapu and Cucao, Southern Chile

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Received: 27 July 2006 / Accepted: 17 October 2006 / Published online: 14 November 2006

Abstract In southern Chile, with a mild and rainy climate, high levels of heavy metals have been detected in many gold placer deposits. Many of the contaminants pose risks to human life, and consequently damage the trofic chain in this environment. The study zones selected correspond to the handicraft gold-bearing sands of Rio Gato (Los Muermos), Carelmapu and Cucao. These are all located in the X Region of "Los Lagos" in Chile. The type of methodology used in the characterization of the associated mineralization consists of testing each representative sample with a grain size distribution, statistical parameter analysis and a mineralogical analysis, using a binocular magnifying glass, a petrographic microscope, XRD and SEM/EDX. The chemical composition was defined by means of X ray fluorescence and micro-chemical analysis. The results of the study about sediments in southern Chile are presented. The major concentrations of heavy minerals are generally located in areas of dynamic river energy. In the samples, more than 70% of the heavy minerals were distributed among grain sizes corresponding to thin sand with good grain selection (meshes of 0.25–0.06 mm). The main mineral phases present in the analysed samples were gold, zircon, olivine, ilmenite, hornblende, hypersthene, hematite, garnet, chromite, chlorite, augite, amphibolitic-epidote, etc. The main heavy metals found as a result of the study were mercury, lead, cadmium, chrome, tellurium, indium, zinc, cobalt, copper, platinum, gold, etc., and as well some less common elements like cerium, praseodymium, gadolinium, neodymium, samarium and lanthanum. This research work is only a starting point for carrying out a risk probability mapping of heavy metals and the mineralogy of the Cucao, Carelmapu and Rio Gato gold-bearing sands.

Keywords Gold · Heavy metals · Mineral classification · Sediment contamination · Mining activities

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Introduction

Like any productive activity, the exploitation of mineral resources produces negative impacts upon the three elements in the environment: water, atmosphere and soil. Pollution levels, in the case of the handicraft mining industry, prove in many cases to have high repercussions

Fig. 1 Map of the study area locations of Rio Gato, Carelmapu, Cucao and Los Muermos (southern Chile)

on the ecosystem despite lighter production levels and possessing smaller quantities of equipment. The repercussions fundamentally take root in the great number of pollutants involved. In this way, we manage to get an idea of the importance that adequate environmental management Chilean mining projects are acquiring, on an industrial and semi-industrial scale, as well as handicraft levels (González et al. 1999a, b).

Toxic sludge contaminates large quantities of water, especially in the exploitation of placers in young riverbeds and river mouths of lakes and tidelands. No subsequent purification of the water is generally carried out. The suspended sediment causes long-lasting alterations that are detectable more than 30 km downstream (Oyarzún et al. 2004). The water destined for consumption is directly collected from rivers, especially in valleys, and its quality is greatly damaged because it does not undergo purification processes (Higueras et al. 2004). On the other hand, a change or destruction of the fauna and flora originating near the river is caused by the alteration of its aquatic environment. This brings negative consequences, not only just for fishermen but also for the food supply with animal proteins. The contaminated elements enter the biological cycle and produce irreversible damage because their physical and chemical characteristics cannot be destroyed, but rather can only transform from one compound into another. These tend to accumulate in soils or sediments along the river ways and in living organisms (González et al. 2000; Quiroz 2001).

Although handicraft mining intensively employs contaminating elements like mercury and cyanide, the net quantities are comparatively similar to those employed and exposed into the environment by dredges and tailings in cyanide plants of traditional mining (Olivo 2000). The environmental impact of handicraft mining fundamentally occurs due the dispersion of many small operations, which hinder the application of preventative or corrective measures, and due the inconvenient location of these installations in urban locations or rural settlements that have high population densities (Prieto 2000).

The contamination levels in handicraft mining, despite less production and possessing less equipments, have high repercussions upon the environment in many cases. Their root cause is fundamentally the large number of contaminants involved. This is due to the contamination from escaping gases, used oils, noise, alterations in the river ways, loss of topsoil and vegetative cover, etc., that are abundant (González et al. 1999a). Chilean small-scale mines employed some 1,700 workers in 2000 (5.1% of total mining workers), and plants and production capacity of less than 50 tpd accounted for 0.97% (44,603 t) of Cu, 6.5% (3,484 t) of Au, and 1.1% (13,665 kg) of the Ag output in the country (Castro and Sanchez 2003). High-grade ore extraction, flotation, gravity concentration, acid leaching cementation and mercury amalgamation are the main metallurgical technologies employed (Castro and Sanchez 2003).

Some of the mining districts in Chile have historic importance and have been exploited almost continuously since Spanish colonial time (sixteenth to eighteenth centuries). Gold recovery involving milling and amalgamation appears to be the most contaminating source of mercury. Higueras et al. (2005) report data of atmospheric mercury from northern Chile. Oyarzún et al. (2004) studied sediments from the Elqui watershed river system that was highly enriched in arsenic. The river system initiates in the high altitude domain of the Andes (3,500–4,000 m) and drains important hydrothermal alteration zones and epithermal deposits, including the copper and arsenic-rich gold veins of the well-known El Indio-Tambo district. Sillitoe (2003) studied the iron oxide–copper–gold deposits in Chile in 2003. Higueras et al. (2004) measured the concentration of Cu, As, Cd, Zn and Hg in samples from stream and mine wastes, stream sediments, soils, flotation tailings and mine wastes in Andacollo (Cu, Au, Hg) and Punitaqui (Cu–Au, Hg). The gold dispersion in Andean desert environments was studied by Herail et al. (1999). Similar studies were carried out by Parada et al. (1997) in Chilean Patagonia and by Johnson and McQueen (2001) in gold-bearing sediments in Western Australia.

Other recent studies (González et al. 1999a, b, 2000) were able to detect the existence of contaminating heavy metals associated with gold in gold-bearing sands located in southern Chile, where dense vegetative cover and a temperate rainy climate permit good characterization of the materials that make up the hydrographical basin, and as such, the geologic environment. Complementing these studies, a study orientated with the investigation of the mineralogical components of different fractions of auriferous sediments is attempted, emphasizing the toxic elements that constitute a risk for the health of human beings.

Fig. 2 Handicraft gold-bearing sands of Rio Gato

The present study reports some of the preliminary results about the distribution and content of heavy elements in gold-bearing sand sediments in southern Chile.

Geographic location of the study area

The areas selected for the study (Fig. 1) correspond to the handicraft gold-bearing sands of Rio Gato (41°19′ to 41°32′ S and 73°20′ to 73°45′ W, Los Muermos), Carelmapu (41°44′35″ S and 73°44′17″ W) and Cucao (42°39′08″ S and 74°07′12″ W, Isla Grande de Chiloé). All are located in the X Region of Los Lagos, Chile.

Access to these areas is difficult and done so over poor dirt trails from the city of Puerto Montt. An example of this type of handicraft installation, the Rio Gato gold-bearing sands (Los Muermos), is shown in Fig. 2.

A great quantity of existing secondary deposits in the X Region is located on the coastal mountain range on both its western and eastern slopes (Quiroz 2001). The lithological units that are found in this range correspond with metamorphic rocks of a Palaeozoic crystalline base that present an auriferous content liberated from the successive erosive processes suffered. Metasedimentary and metavolcanic rocks also make up part of this range, but their auriferous load is much smaller.

Carelmapu

The Carelmapu auriferous placer is located in Llanquihue province at Brava beach, a costal sector of Carelmapu. Brava beach is 10 km long and the majority of the gold-bearing sands of the area are concentrated within it. It is bordered to the north by the Amortajado hills, and to the south by the Chonqui and Chocoi hills and the town of Carelmapu.

The predominant rocks consist of sands, loams and conglomerates. The sand on the beach is made up of quartz and feldspar, and deeper down contains black layers of magnetite and ilmenite (González et al. 2000; Quiroz 2001).

Three types of gold-bearing sands stand out at Carelmapu: (i) ancient gold-bearing sands that make up the strip between the Chacao channel and the Carelmapu plain; (ii) a mantle of black sand located on the beach; and (iii) a plain covered by dunes.

In accordance with the historical record, deposits of auriferous sand all along the extensive coast of Brava beach have been exploited for several years in an artisan form. These deposits primarily accumulate within the margins of high and low tide and are characterized by their constant renewal from the surf activity and erosion of ancient cliffs; the gold is in dust form and concentrated by the wave activity.

Rio Gato

Rio Gato is located in the Los Piques sector of Los Muermos, in Llanquihue province. It lies on the southeastern slope of the coastal range and reaches maximum altitudes of 280 m.

The lithologic units show evidence from two eras: one of deposits developed during the Palaeozoic and another from modelled erosion of the landscape, generated during the Tertiary and Quaternary. The zone corresponds with an extensive and narrow ravine with a N-NE orientation, limited by moderately sloped terrain with numerous water currents standing out. These currents are the principal erosion factor of the sides and of the landscape in general, where the erosion of auriferous material and the slope of the area complement each other so this material arrives due to gravity at the river channels and estuaries to be deposited. Consequently, the auriferous placer corresponds with a fill of fluvial and alluvial sediment of an extensive valley formed by the Cordillera River. The auriferous content is distributed in the gravels, increasing the concentration with the depth, the greater size of the gravels and the proximity to the base.

Cucao

This auriferous placer is located at Cucao Bay on the western coast of Chiloé island, in Chiloé province. The eastern portion of the coast is an extensive plain, while the coastal range expands in the central sector, descending gradually on its western slope, featuring some terraced levels of plains. Cucao beach is approximately 20 km long, with a maximum width of 1.5 km in its central portion. The Cucao River ends here after flowing 2.5 km to drain water from a lake of the same name.

The gold-bearing sands on the beach extend all along the coast. The best auriferous stratum is 1–2 m below the surface in an area of approximately 30 m just beyond the reach of the high tides, where small rivers join the sea. The gold comes from the gravels of the auriferous rivers.

Materials and methods

The areas selected for the study were considered primarily due to their significant content of heavy metals. In the case of the gold-bearing sands, the study was carried out by relying upon the historical record of where gold extractions either existed or exist today. This is in accordance with the geological distribution of these secondary deposits in Chile (Ruiz and Peebles 1988).

A surface and deep sample were performed in each study area with the intention of acquiring representative samples. The samples taken at Carelmapu and Cucao beaches were extracted from approximately 30 cm under the surface. The samples from the dunes near Carelmapu beach were taken per stratum, obtaining: (i) an upper stratum sample 20 cm depth; (ii) a middle stratum sample 30 cm depth; and (iii) a lower stratum sample 60 cm depth. The samples taken at Rio Gato (Los Muermos) were also sampled following the same methodology.

The samples were processed at the Natural Resources and Processes Laboratory of the Metropolitan Technological University, subjecting them to sieve (phi ½) on W. Styler Standard Sieve Series from 2.00 to 0.05 mm meshes. A magnetic separation was subsequently performed, where the non-magnetic fractions were subjected to segregation with heavy liquids (bromoform and methylene iodide), with the intention of obtaining heavy fractions with specific weights greater than 3.32 that correspond to those that the required minerals possess.

The characterization and identification were carried out by binocular magnifying lens, petrographic microscope, X-ray diffraction (XRD) and SEM/EDX. X-ray fluorescence (XRF) was conducted to determine the chemical composition of the sediments, while the mineralogical identification was corroborated by XRD and chemical micro-analysis (SEM/EDX).

The registry of the X-ray diagrams was made by a Siemens D-5000 powder diffractometer equipped with Bragg–Bretano geometry, graphite monochromer and sparkling detector. The spectra were taken with Cu radiation, K_α (1.54052) at 40 kV, 20 mA for the total sediment and middle-sized fractions (0.5–0.25 mm), and 30 mA for small-sized fractions (0.25–0.05 mm) (Davis 1980). A normal diagram of 2θ was made from 3° to 65°, data taken every 0.02° and a step-time of 3.2 s. The combination of incidence and divergence slights for middle-sized fractions was 2 and 0.2 mm, respectively. The incidence and divergence slights for small-sized fractions were 1 mm and 0.6 mm, respectively, with a 0.1 mm detector with continuous register modality (Davis 1981). The diffractometer was operated by a Pentium 100 using the Siemens Diffract plus v. 2.0 software program.

The identification of the mineral phases in each sample was made using the Socabim EVA computer

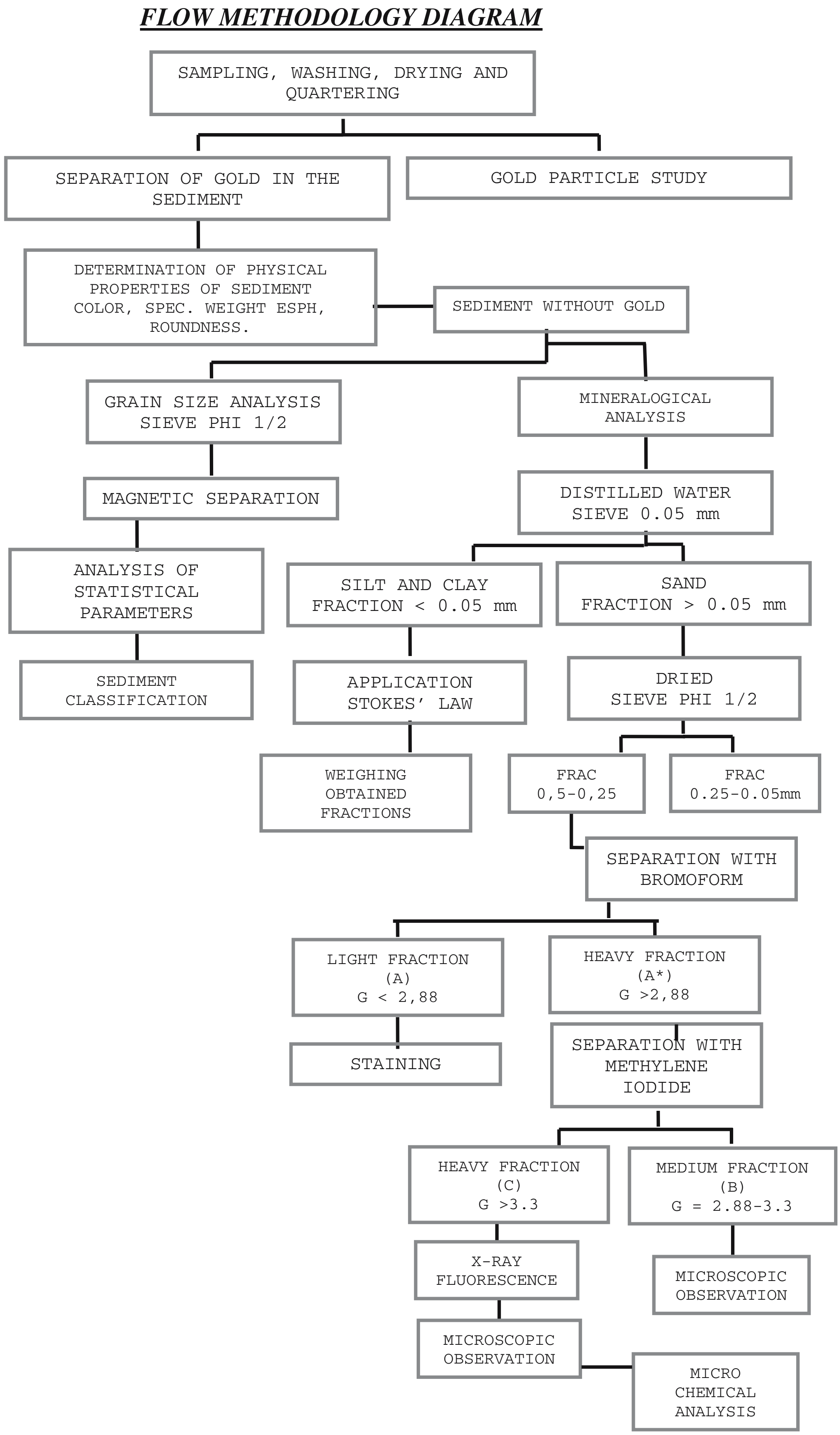

Fig. 3 Flow methodology diagram

program and the JCPDS files (Davis 1981, 1984). A background removal corresponding to the diffractometer, a selection of the diffraction peaks, and the search for phases were done by the above-mentioned program. It was subsequently completed by consulting the JCPDS diffraction files.

The scanning electronic microscope (SEM) was used as a complementary technique with XRD. It allowed the analysis of the non-internal reticulated order materials, and the morphological and micro-analytic research (EDX). The SEM analysis was carried out by a SEM (Leo model 440) with Oxford EDX micro-analysis equipment incorporated into the microscope. The working conditions were the following: acceleration voltage 20 kV, energy range 0–10 keV, acquisition time 60–100 s (according to samples) and the ZAF method for semi-quantitative analysis with virtual samples.

The present study comprises 22 representative samples of each of the studied gold-bearing sands. Figure 3 shows the flow methodology diagram.

Results and discussion

The sediments analysed with XRF (Table 1) contain an ample range of elements in their composition, going

Table 1 Results of semi-quantitative analysis of heavy metals performed by X-ray fluorescence

Chemical element	Concentration (%)					
	M 1-A	M 2-A	M 1-B	M 2-B	M 1-C	M 2-C
Iron	2.53	40.60	24.80	41.30	0.599	27.00
Titanium	0.444	12.70	11.90	11.60	0.0512	1.89
Zirconium	0.0602	2.20	0.377	0.183	0.0019	0.438
Gold	–	–	–	0.0106	0.0003	0.0162
Platinum	–	–	–	0.0105	0.0002	–
Mercury	0.0005	1.80	–	–	–	0.0111
Lead	–	0.0543	–	–	0.0001	–
Rhenium	–	1.73	–	–	–	–
Sulphur	0.0046	1.13	0.0336	–	0.003	0.0102
Manganese	0.115	0.312	0.303	0.324	0.0082	0.36
Arsenic	0.0008	0.261	–	–	–	–
Magnesium	37.70	0.252	43.40	0.514	24.4	2.92
Nickel	0.0012	0.202	–	–	0.0004	0.0338
Bromine	–	0.145	–	–	–	–
Vanadium	0.0043	0.130	–	–	0.0011	0.0291
Antimony	–	0.112	–	–	–	–
Chromium	0.0052	0.0893	0.0874	0.0755	0.001	0.031
Caesium	0.0044	0.0609	–	0.0578	–	–
Cobalt	0.0029	0.0495	–	–	–	0.0405
Germanium	–	0.0616	–	–	–	–
Zinc	0.0014	0.0427	0.025	0.0365	0.0005	0.0464
Niobium	–	0.0245	–	–	–	–
Copper	0.0004	0.0199	–	–	0.0003	–
Scandium	–	0.017	–	–	–	–
Strontium	0.0003	0.0176	–	–	0.0008	0.075
Thallium	0.0004	0.0131	–	0.0126	0.0002	–
Yttrium	0.0013	0.0051	–	–	0.0001	0.0149
Rubidium	–	0.0034	–	–	0.0001	–
Tungsten	0.0109	–	0.0127	0.0206	–	0.0253
Cerium	0.0088	–	–	–	–	0.108
Praseodymium	0.0074	–	–	0.162	–	0.100
Samarium	–	0.077	–	–	0.0013	–
Gadolinium	–	0.0472	0.0445	–	–	0.0537
Lanthanum	0.0008	–	–	–	0.0002	–
Neodymium	–	–	–	–	0.0009	–
Indium	0.005	–	0.044	–	0.0022	–
Cadmium	0.0001	–	–	–	–	–
Thorium	0.0006	–	–	–	–	–
Palladium	0.0006	–	–	–	0.0003	–
Iridium	0.0006	–	–	–	0.0005	–
Tin	0.0063	–	–	0.0554	–	–
Ruthenium	–	–	–	–	0.0002	–
Tellurium	–	–	–	–	0.0033	–

Samples M1-A (fraction 0.5–0.250) and M2-A (fraction 0.250–0.05) correspond to the Cucao auriferous placer; samples M1-B (fraction 0.5–0.250) and M2-B (fraction 0.250–0.05) to Rio Gato; samples M1-C (fraction 0.5–0.250) and M2-C (fraction 0.250–0.05) to Carelmapu from iron, magnesium, gold and platinum, all the way to some rare metals including samarium, gadolinium and lanthanum.

Table 1 shows the preliminary results of the variation of the content of the trace elements in the study area. The analysis of the mean contents for some traces in the sediments of Rio Gato, Carelmapu and Cucao shows greater concentrations of iron, magnesium, titanium and manganese with important differences among the three gold-bearing sands. Sediments from Cucao have high amounts of mercury (1.8%), lead (>0.05%), vanadium (>0.1%) and zirconium (>2%). The sulphur content in the analysed heavy fraction is also very elevated, reaching values superior to 1% at the Cucao auriferous placer. The analysed elements form their respective mineralogical species, like for example zinc in ZnS, zirconium in zircon, magnesium in olivine and hypersthene, titanium in ilmenite, lead in galena and mercury in cinnabar. These elements are present at Rio Gato and Carelmapu in concentrations that can be considered relatively low.

The concentration of Hg and Pb in the studied sediments is related to texture and grain size variations and is higher in the heavy fraction than the light one (Table 1). Mineralogically, they are primarily found in the form of sulphurs (cinnabar and galena, respectively).

In general, it was observed that greater concentrations of heavy metals are located in the area of greater energy. It was determined in the samples of fine sediments with good grain selection that more than 70% of the heavy minerals were distributed between the grain sizes corresponding to meshes of 0.250–0.05 mm. This indicated that a strong tendency exists for heavy minerals to be concentrated within the finest grain size classes.

In this way, the semi-quantitative analysis of heavy metals by XRF allows us to only know the percentage of elements and not the mineralogical species that they make up. Concentrations of rare elements (REE) do also exist, making a more extensive and detailed study necessary. This is because, just like in the previous case, there are no historical records indicating their existence or exploitation. If the information regarding these minerals associated with the platinum group is used, it allows the establishment of what can begin to constitute a good prospecting guide for this precious metal.

For this the observation and separation were performed under binocular magnifying glass and transmission optic microscopy (applied to thin mineral sections) of the different species that made up the sediments. These were recognized together by microchemical analysis that allowed recognizing the following mineral phases:

Rio Gato gold-bearing sands	Gold, zircon, olivine, ilmenite, hornblende, hypersthene, hematite, garnet, chromite, chlorite, augite, amphibolitic-epidote, magnetite and platinum in an undetermined form
Carelmapu gold-bearing sands	Cinnabar, zircon, chromite, epidote, galena, garnet, olivine, hypersthene, augite, hornblende, ilmenite, magnetite, gold and pyrite
Cucao gold-bearing sands	Cinnabar, zircon, chromite, epidote, cellular pyrite, galena, garnet, hematite, olivine, hypersthene, hornblende, ilmenite, magnetite, gold and pyrite

From Table 1, and even though numerous data and studies are missing in these sediments, it was deduced that major components like iron, magnesium and titanium are present in the minerals found. For example, Fe is mainly present in the crystalline phases of magnetite and hypersthene at Rio Gato, Mg in garnet at Carelmapu, and Ti in ilmenite at Cucao.

Heavy minerals susceptible to being mechanically concentrated in nature are generally numerous, although only a few possess any economic value. In our study area, these primarily correspond to gold (Figs. 4, 5), platinum, zircon, magnetite and ilmenite. The others, like pyrite, epidote, olivine, etc., normally accompany those aforementioned and primarily occur due to the denudation processes acting more rapidly in these plots than those of oxidation.

The presence of REE in the gold-bearing sands studied results being important as an element indicator of platinum (found to be present in the areas in unidentified species), as well as the economic value that these have (González et al. 2002).

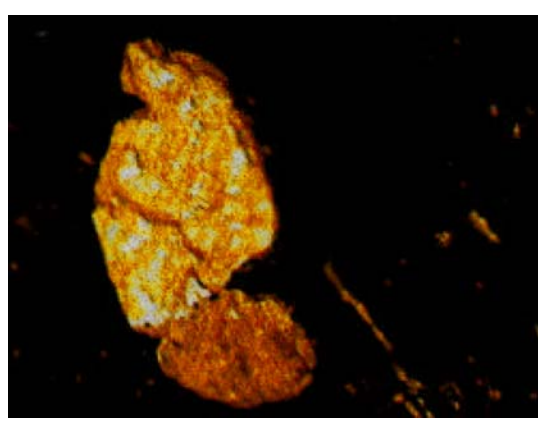

Fig. 4 Microphotograph of metallic gold particles (30×)

The lead content at Carelmapu and Cucao allows verifying important contamination levels, just as the presence of mercury in cinnabar, whose formation is due to the handicraft extraction of gold (Figs. 5, 6, 7, 8). Sediment metal concentrations found in these gold-bearing sands were comparable with aquatic systems from other regions in the world classified as contaminated.

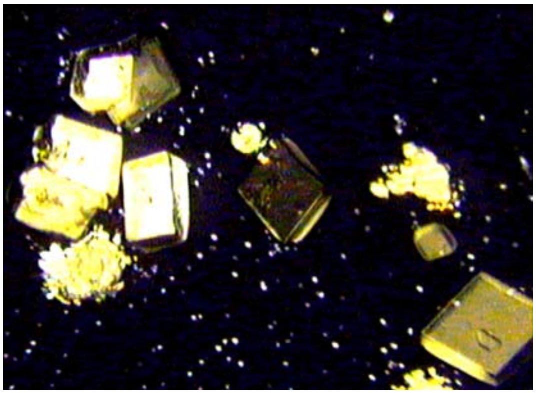

Fig. 5 Microphotograph of acicular and octahedral Au^0 and TlCl crystals (650×)

Fig. 6 Microphotograph of cinnabar particles

Fig. 7 Microphotograph of blue dentrital $CoHg(SCN)_4$ crystals (250×)

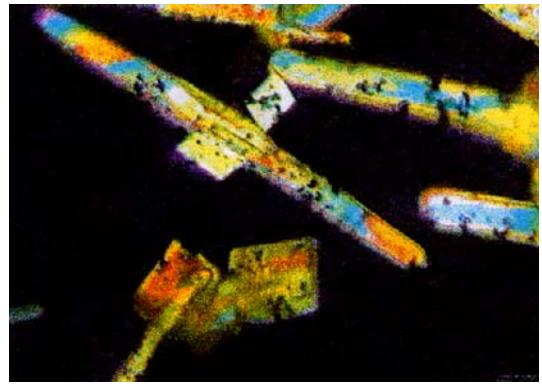

Fig. 8 Microphotograph of rhombohedron $PbHg(SCN)_4$ crystals (500×)

One of the most important results of this persistence is their increase in trofic chains. Because of this process, the heavy metals in the upper members of this chain can reach greater levels than what is found in the water or air. The intake of these by many plants and animals can constitute genuine risks for human health.

Moreover, the recurring presence of lead, sulphur and mercury, among others, in mineralogical species like galena and cinnabar reveal accumulation indices, a product of the contaminating action of human beings. This is notable since no records exist of natural deposits of these minerals that can justify their presence, and records were utilized from semi-industrial exploitations for the extraction of gold where mercury is utilized in the amalgamation processes.

Conclusions

Artisan gold mining at the Cucao, Carelmapu and Rio Gato areas of Chile has resulted in extensive contamination of the fluvial, or beach environment, by mercury (Hg) and potentially toxic ore-related elements including lead (Pb) and zinc (Zn). This principally occurs through the fluvial dispersion of contaminated mineral processing wastes and recycling of Hg sublimated during the burning of amalgam. Significant releases of mercury at Cucao are associated with inefficient amalgamation techniques. The present situation will otherwise continue or even worsen with no oversight or control of the global Hg trade, in which the transfer of excess European Union Hg to artisan miners is favored because of the low Hg prices relative to that of gold. However, not all of the samples analysed show contamination with these heavy metals.

Darker sediments contain a greater percentage of heavy minerals. This is due to the presence of magnetite, which in general surpasses 50% of the heavy mineral fraction. In some of them, cinnabar does not abundantly exist in order to attribute it to anthropogenic contamination with mercury.

The content of heavy minerals like magnetite, ilmenite and zircon in the fine fractions of the sediment also constitutes a good guide for gold prospecting.

From both, the study performed and the economic point of view, Los Muermos–Rio Gato is the area that presents the greatest interest. Sand exists there with contents of zircon, gold and platinum. Then the Carelmapu and Cucao areas follow, whose sand contains gold and zircon. The method of separating heavy minerals in heavy liquids (bromoform and methylene iodide) is only efficient on a laboratory scale as it is slow and expensive. Moreover, large quantities of sediments cannot be utilized. There exists the advantage, however, that all of the heaviest grains of a determined specific weight were separated.

This research work in only a starting point for carrying out a risk probability mapping of heavy metals and the mineralogy of the Cucao, Carelmapu and Rio Gato gold-bearing sands. This would provide better knowledge about the vulnerability of the soil to Hg, Pb and other heavy metal contamination. To achieve this purpose, the initial variable was transformed into an indicator variable using the risk-based standards (intervention values) for soils as thresholds. A risk probability mapping will also be done to assess the vulnerability of the soil towards mining operations in the studied areas.

Acknowledgments The present study makes known part of the information obtained during the realization of Projects 108/98 "*Prospección de Yacimientos Secundarios de Oro y Evaluación de la Contaminación Producto de su Explotación*" and 269/04 UTEM-Empresa, financed by the Universidad Tecnológica Metropolitana de Chile (UTEM).

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