

Mineralogy and geochemistry of sulfide-bearing tailings from silver mines in the Taxco, Mexico area to evaluate their potential environmental impact

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Received: December 11, 2002; accepted: February 4, 2004

RESUMEN

Seis depósitos de jales y un terrero resultantes de la explotación de los depósitos epitermales de Pb-Zn-Ag-Cu de la región de Taxco, Guerrero han sido caracterizados mineralógicamente (microscopía óptica, MEB y XRD) y geoquímicamente (EDS, metales totales y solubles en agua) para estimar su potencial para liberar metales al medio ambiente. Las fases primarias están dominadas por el cuarzo y los fragmentos de roca con escasos feldespatos, barita y ferromagnesianos. La calcita sólo ha sido reconocida en los jales menos oxidados. Los sulfuros primarios incluyen piritita y esfalerita con escasa pirrotita, galena, calcopiritita, sulfosales de plata y arsenopiritita, los cuales sólo se conservan en los depósitos no oxidados. En los jales oxidados, los sulfuros están ausentes o aparecen como relictos. Las fases secundarias están ampliamente distribuidas e incluyen al yeso, calcocita, keoheita, nimita, hetaerolita, jarosita, smithsonita, bernalita, eponita, rozenita, pickeringita, hexahidrita, hematita, escorodita así como precipitados amorfos y criptocristalinos y oxihidróxidos de hierro pobremente cristalizados. Análisis por EDS indican que la adsorción en las fases secundarias es un importante proceso que influye en la retención de metales.

Todos los depósitos de jales y terreros de Taxco se caracterizan por presentar altas concentraciones totales de Ag (9.5-74.2 mg/kg), Cd (1.0-780 mg/kg), Cu (71.8-1320 mg/kg), Fe (2.49-25.1%), Mn (18.6-13 800 mg/kg), Pb (780-43 700 mg/kg), V (2.0-127 mg/kg), Zn (380->10 000 mg/kg) and As (19.0-11 800 mg/kg) y todos se encuentran por encima de los niveles regionales de fondo de los suelos de cultivo. Los contenidos de metales solubles en agua indican que los jales son muy heterogéneos y que liberan cantidades importantes de metales tóxicos, (e.g. Cd = <0.005-19.2 mg/L; Cu = <0.025-63 mg/L; Fe = >0.025-1105 mg/L; Pb = <0.025-0.655 mg/L; As = <0.010-1.79 mg/L) particularmente los niveles oxidados, y que la liberación de estos metales está controlada esencialmente por el pH. Los datos indican que la oxidación de los sulfuros y la generación de AMD están acompañados por procesos de disolución de otras fases, de neutralización por la calcita, la reprecipitación de fases secundarias y la adsorción, y que estos procesos son los principales responsables de la movilidad y disponibilidad de metales en los desechos mineros de Taxco.

PALABRAS CLAVE: Taxco, jales, sulfuros, metales pesados, drenaje ácido de mina, biodisponibilidad, toxicidad.

ABSTRACT

Six sulfide-bearing, flotation tailings dams and one waste-rock dump from epithermal Pb-Zn-Ag-Cu deposits in Taxco, Guerrero have been mineralogically and geochemically characterized to evaluate their potential environmental impact. Primary phases are dominated by quartz and rock fragments with scarce feldspar, barite and ferromagnesian. Calcite is found in the less oxidized tailings. Primary sulfides are pyrite and sphalerite with some pyrrothite, galena, chalcopyrite, Ag-sulfosalts and arsenopyrite. In oxidized zones, sulfides are low or absent. Secondary phases are widespread and include gypsum, chalcocite, keoheite, nimite, hetaerolite, jarosite, smithsonite, bernalite, eponite, rozenite, pickeringite, hexahydrite, bassanite, boyleite, hematite, scorodite, cryptocrystalline and amorphous precipitates and poorly crystallized Fe-oxyhydroxides. EDS analysis suggests that adsorption is important in the retention of metals.

All Taxco tailings and waste-rock dumps show high concentrations of total Ag (9.5-74.2 mg/kg), Cd (1.0-780 mg/kg), Cu (71.8-1320 mg/kg), Fe (2.49-25.1%), Mn (18.6-13 800 mg/kg), Pb (780-43 700 mg/kg), V (2.0-127 mg/kg), Zn (380->10 000 mg/kg) and As (19.0-11 800 mg/kg) exceeding the regional background concentrations in crop soils. Contents of water-soluble metals are heterogeneous but contain significant amounts of toxic metals (e.g. Cd = <0.005-19.2 mg/L; Cu = <0.025-63 mg/L; Fe = >0.025-1105 mg/L; Pb = <0.025-0.655 mg/L; As = <0.010-1.79 mg/L), particularly from highly oxidized zones. The release is primarily controlled by pH. Sulfide oxidation and generation of AMD are coupled by dissolution, neutralization via calcite, reprecipitation and adsorption.

KEY WORDS: Taxco, tailings, sulfide, heavy metals, acid mine drainage, bioavailability, toxicity.

INTRODUCTION

Environmental impact of mines may severely affect ecosystems. In most cases, metal contents exceed the regional backgrounds (e.g. Fergusson, 1990; Hudson, 1998; Plumlee, 1999). Tailings contain high levels of Pb, Cd, Zn, As, Cu, Fe and other metals of environmental concern and may be the most important sources of toxic metal pollution on earth (Bylinsky, 1972; Plumlee, 1999). Tailings are dumped frequently along streams near mines, where interaction with air and rainfall oxidizes sulfides, particularly pyrite, thus generating acid mine drainage (AMD; Ritcey, 1989; Nordstrom and Alpers, 1999; Dold, 1999). Further AMD generation may be favored by the action of *thiobacillus ferrooxidans*, a bacteria that increases substantially the capacity of pore water to dissolve metals (Ritcey, 1989; Evangelou and Zhang, 1995). In carbonate-bearing deposits or calcareous terrains, AMD is often neutralized to substantially reduce dispersal of metals, but dispersion may occur in secondary phases (e.g. Jambor, 1994; Dold and Fontboté, 2001; Dold and Fontboté, 2002). In such mining sites, sediments and soils are severely affected, whereas the effect on surface and groundwater is more limited (Ritcey, 1989; Fergusson, 1990).

The main concern of metal pollution is the adverse effect on biota, including people around mines (e.g. Furst, 1971; Bylinsky, 1972; Fergusson, 1990; Montgomery, 1992; Keller, 1992; US Department of Health and Human Services, 1993; Smith and Huyck, 1999). The neurotoxic and phytotoxic effect of metals caused the United Nations Environmental Program, and the World Health Organization, to state that the physical and chemical processes occurring in solid and liquid mining waste are topics of high priority.

Information on the mineralogical and geochemical composition of tailings in terms of potential pollution is essential to better understand the processes controlling the dissolution of metals, their dispersion, persistence, availability and toxicity as well as by pathways of uptake animals and humans. The purpose of this study is to provide data on the mineralogical (primary and secondary) composition and total and water-soluble metal concentrations of tailings in the mining district of Taxco, Guerrero in southern Mexico. Data are compared with background levels to estimate the potential risk for releasing metals to the nearby environment and to identify the major process controlling their mobility.

GEOLOGICAL SETTING AND ECONOMIC GEOLOGY

A simplified geological map of the Taxco region is shown in Figure 1. The location of studied tailings is also shown. For a description of the stratigraphy and structure of

the Taxco area, the reader is referred to Fries (1960); Campa et al. (1974); Campa and Ramírez (1979) and De Cserna and Fries (1981). Geological units in the Taxco area include the Taxco Schist, the Morelos, Mexcala, Balsas and Tilzapotla formations, which range in age from lower Cretaceous to Oligocene (Campa, 1974; Morán-Zenteno et al., 1999; Campa et al. 2003).

In Taxco, mineralization mainly appears in hydrothermal veins, replacement ores and stockworks hosted in limestones of the Morelos and Mexcala formations, and more rarely in the Taxco Schists and Balsas formations (IMSA, 1978). Mineralized structures are 1 to 3 m wide although a few reach 10 m, and 700 to 2000 m long. Main ores include galena, chalcopyrite, sphalerite, native silver, argentite and Ag-bearing (polybasite, proustite, pyrargirite) sulfosalts whereas gangue phases include pyrite, pyrrhotite, quartz, barite, calcite, rhodochrosite, hematite, tremolite, epidote and ilvaite (CRM, 1999).

MINING BACKGROUND AND MINE-WASTE DUMPS

The Taxco region has been one of the most important precious metal producers in Mexico since prehispanic times. During the colonial (1521-1810) epoch, Taxco mines produced large quantities of silver and gold and were considered among of the richest mines in America. Since fire reduction was the main method used to obtain silver at the beginning of colonial times, native silver-rich veins were intensively exploited. Sulfide-rich ores were often discarded and together with residual scoria were dumped near mines. During the mid-eighteenth century, the method of amalgamation was introduced in Taxco leading to an increase in the efficiency of silver and gold recovery. This method was used until the beginning of the twentieth century. Unfortunately, this method produces large quantities of granular waste containing not only high levels of trace metals but also mercury increasing substantially the risk of environmental pollution. In some places, waste was dumped directly into rivers but in others they were accumulated near mines. The latter, together with sulfide-rich waste generated during the beginning of the ore exploitation were characterized by high level of base metals and silver and gold. Most of this waste was reworked during the mid-twentieth century (CRM, 1999).

Modern mining in Taxco began in the 1940's with the introduction of the selective flotation method that increased substantially the capacity of production. Until 1976, ore processing was performed individually separately in several mines and the resulting tailings were accumulated in dumps in the mine surroundings. Since 1976, production of all mines is underground concentrated in the Solar Plant and

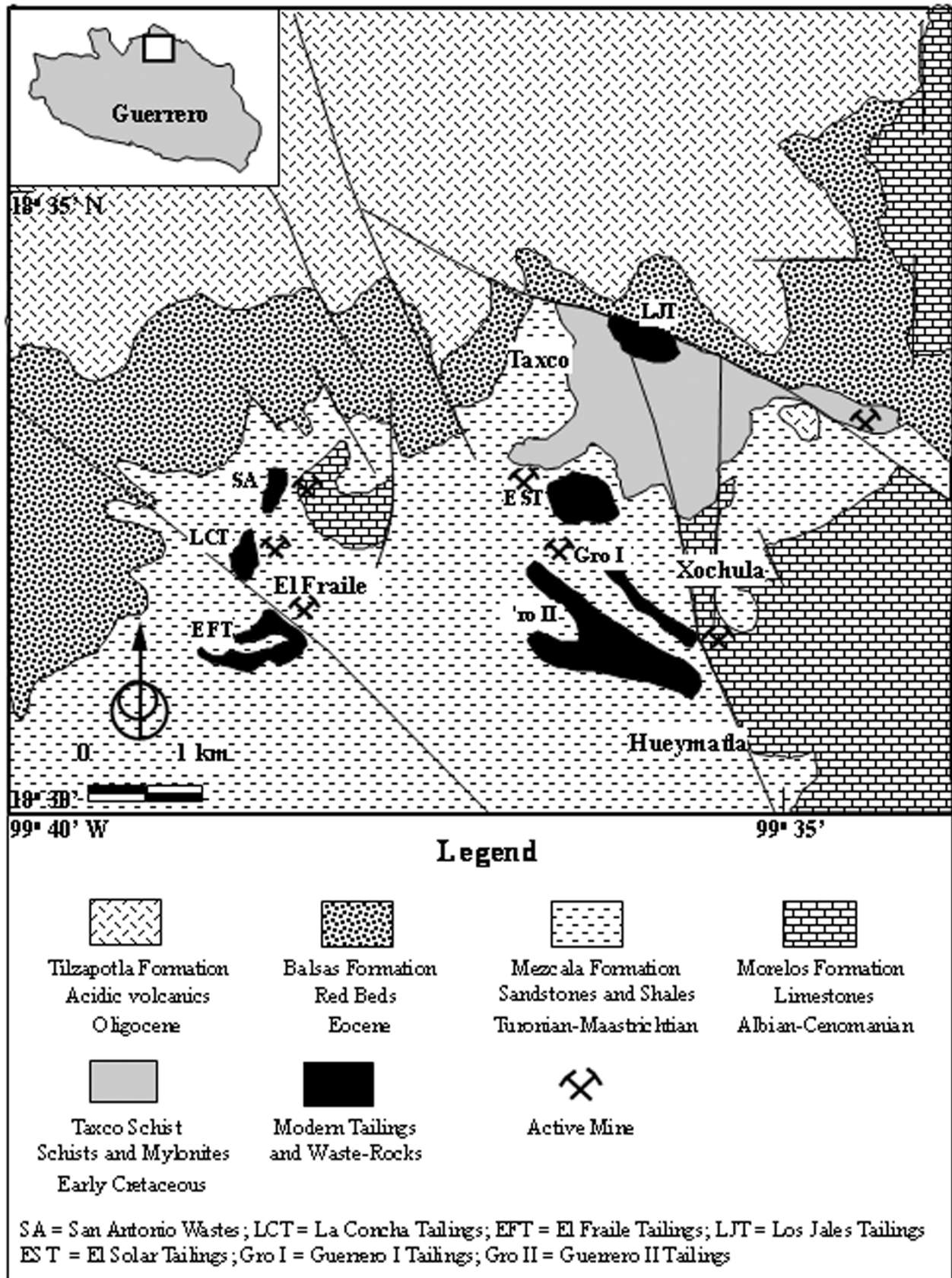


Fig. 1. Simplified geologic map of the Taxco Mining District showing the location of the studied sulfide-bearing, flotation tailings and waste-rock dumps (after Campa and Ramírez, 1979).

resulting tailings are accumulated in the current (here named Guerrero II) tailings dump.

All studied tailings and waste-rock dumps were deposited on rocks of the Mexcala and Taxco Schist formations (Figure 1). These formations are considered essentially impermeable units, although secondary porosity can be locally important. These characteristics prevent the infiltration of large volumes of AMD to aquifers, but enable surface drainage of metal-rich leachates affecting the quality of effluent water. From the seven dumps studied here, one (San Antonio) is a coarse waste-rock accumulation containing appreciable amounts of sulfides and oxides and the other six (El Fraile, La Concha, El Solar, Guerrero I, Guerrero II and Los Jales) are tailings dumps (Figure 1). All tailings except the upper levels of the Guerrero II and La Concha ponds were generated before 1976 and correspond to exploitation of individual mines in the beginning of the modern period. Although there is no documented evidence, people who worked in the mines in the beginning of the modern epoch of exploitation (1940-1960), affirm that the lower levels of La Concha tailings were generated during the 1940-1950 period, whereas El Fraile, El Solar and Guerrero I dumps were produced during the 1950's. The upper levels of the Guerrero II tailings were generated by the El Solar concentration plant from the exploitation of all mines since 1976. The upper part of La Concha impoundment seems to be non-oxidized tailings added to avoid dispersal of highly oxidized material. Conservative calculations indicate that waste-dumps contain in total more than 25 million tons of tailings and waste rock, being the current, Guerrero II pond the biggest accumulation with approximately 20 million tons and San Antonio the smallest with 0.3 million tons. With the exception of San Antonio, all impoundments show typical structures (nearly horizontal banks, gradational, parallel and crossed lamination) of accumulations deposited as oversaturated muds.

SAMPLING AND ANALYTICAL METHODS

One hundred and thirteen samples were studied, 49 samples are from El Fraile, 31 from La Concha, 9 from El Solar, 6 from Guerrero I, 6 from Guerrero II, 7 from Los Jales and 5 from the San Antonio mine waste-dumps. Samples were taken from the uppermost 5 centimeters, except in highly oxidized levels, which developed a 0.5 to 10 cm ochreous crust of Fe-Mn oxyhydroxides. In such levels, samples were taken from the oxidized zone immediately below crust. Pre-washed, disposable polyethylene tools were used for sampling to avoid cross contamination. Each sample consisted of about 1 to 2 kg of tailings, stored in plastic bags and immediately transported to the laboratory. Regional background concentrations (RBC; Table 2) of soils were calculated by analyzing pristine soils derived from the dif-

ferent formations present in the area of study, with the exception of the Taxco Schist, which shows evidence of modification by mining activities.

Samples were air dried during 72 hrs in a dust-free environment and sieved through a 2 mm stainless steel mesh. Approximately 250 g of the <2 mm fraction was further sieved through No 40 and 60 US Standard meshes to recover medium-size sand particles. After complete removal of adsorbed clay by strong washing in an ultrasonic bath, this fraction was impregnated with epoxy resin and polished thin sections were prepared using standard techniques. Representative thin sections of the La Concha, El Fraile, El Solar and Guerrero II tailings were studied under the scanning electron microscopy (SEM). Chemical phases were identified using energy-dispersive spectroscopy (EDS). Another subsample of the <2 mm fraction was sieved through a No 220 US Standard mesh to recover the fine-size sand, loam and clay fraction. This fraction was used for X-ray diffraction with a Phillips X-Pert 50 diffractometer from the Universidad Autónoma del Estado de Hidalgo, Mexico and a Bruker AXS D8 Advance diffractometer from the Universidad de Sonora, Mexico.

Total metal contents were determined using an Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES) after $\text{HNO}_3 + \text{H}_2\text{O}_2 + \text{HCl}$ extraction in 0.200 gram of sample in Teflon bombs following the 3050b USEPA method. Two HP, metal-rich solid reference standards (CRM-SOIL-B and CRM-LOAM-B) were analyzed with samples and recovery in both standards was better than 90% in all studied elements except Ag, whose recovery was around 70%. Physical-Chemical (electrical conductance, pH) and water-soluble metal contents were determined in two tailings impoundments (El Fraile and La Concha) in a 1:2.5 extract according to the methodology proposed by Nogales *et al.* (1997). Water-soluble contents were checked against three HP aqueous reference standards (CWW-TM-H; CWW-TM-A and CWW-TM-E). The lowest concentrations determined in these certified standards were: Ag and Cd 0.005 mg/L; Cu, Fe, Mn, Pb, V and Zn 0.025 mg/L and As 0.010 mg/L, which are considered as the working detection limits of our method.

RESULTS

Mineralogy

Primary and secondary phases observed in the studied dumps as well as their relative abundance are shown in Table 1. Figure 2 shows selected X-ray diffraction patterns of samples of non-oxidized and oxidized tailings from different dumps as well as secondary precipitates. As a whole, significant differences exist in modal composition between

Table 1

Properties and main mineralogy (primary and secondary) recorded in the Taxco, Guerrero tailings and waste-rock dumps.
 X Scarce; XX Slightly abundant; XXX Moderately abundant; XXXX Very abundant

	El Fraile	La Concha	El Solar	Guerrero I	Guerrero II	Los Jales	San Antonio
<i>General Characteristics</i>							
Waste-type	Tailings	Tailings	Tailings	Tailings	Tailings	Tailings	Waste-Rock
Operation time	1950-1960	1940-1950	1950-1960	1950-1960	1976-	---	---
Aprox Volume (Millions of Tons)	2.0	0.6	1.0	0.6	20.0	0.5	0.3
<i>Mineralogy</i>							
<i>Primary</i>							
Quartz	XXXX	XXXX	XXXX	XXXX	XXXX	XXXX	XXXX
Rock Fragments	XXXX	XXXX	XXXX	XXXX	XXXX	XXXX	XXXX
Feldspar	XXXX	XXXX	XXXX	XXX	XXX		
Ferromagnesian	XXX	XXX	XXX	XXX	XXX		
Calcite	X	XX	X	XXX	X	X	X
Dolomite		X					
Barite	X	X	X	XX			
Pyrite	XX	XXX	XX	XXXX	XX	XX	XXXX
Pyrrotite	X						XX
Galena	X	X		XXX		X	XXXX
Chalcopyrite	XX	XX	XX	XXX	X	X	XXX
Sphalerite	XXX	XXX	XXX	XXX	XXX	XXX	XXX
Magnetite	X	X	X	X	X	X	X
Ag-Sulfosalts		X					
Arsenopyrite		X					
<i>Secondary</i>							
Saponite	XX						
Montmorillonite	XX						
Chalcocite	X	X	X	X			
Gypsum	XXXX	XXX	XXX	XXX	XXXX		
Hexahedrite	XX						
Basanite	XX						
Epsonite	XXX				XX		
Bernalite	XXX						
Hematite	XXX						
Smithsonite	X	XXX					
Keoheite	XX	XX		X			
Nimite				X			
Hetaerolite				X			
Jarosite	XXXX	XXXX	XXXX		XXXX		
Boyleite					X		
Starkeyite					X		
Scorodite		X					
Pickeringite	X			XX			
Rozenite	XX		X	XX			

Fig. 2. Representative X-ray diffraction patterns of non-oxidized (a, b), partly oxidized (c) tailings and secondary precipitates (d, e, f) of selected samples from the Taxco flotation tailings.

oxidized and non-oxidized samples particularly in the relative abundance of sulfides (mainly pyrite), calcite and secondary phases, which suggest that the mineralogy in tailings

and waste-rock dumps is mainly controlled by oxidation-neutralization processes as has been determined in many other sulfide-bearing tailings impoundments (e.g. Ritcey, 1989;

Jambor, 1994; Dold and Fontboté, 2001; Dold and Fontboté, 2002). These mineralogical differences were even observed in samples from the same dump indicating great mineralogical heterogeneity. Primary non-metallic phases are largely dominated by quartz and rock fragments (mainly schists and volcanics) with minor amounts of feldspar, barite and ferromagnesian (amphiboles and pyroxenes). Calcite is ubiquitous in the more recent, non-oxidized samples but it is rare or absent in the older, highly oxidized samples. Primary metallic phases (sulfides and oxides) are generally in the range of 2-7 % vol and consist of variable amounts of pyrite, chalcopyrite, sphalerite, galena and crystalline Fe-oxides. Rare arsenopyrite and Ag-sulfosalts were only observed in La Concha and El Fraile impoundments. Secondary phases include chalcocite (Cu_2S), gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), bassanite ($\text{CaSO}_4 \cdot \text{H}_2\text{O}$), epsomite ($\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$), hexahydrate ($\text{MgSO}_4 \cdot 6\text{H}_2\text{O}$), starkeyite ($\text{MgSO}_4 \cdot 4\text{H}_2\text{O}$), Pickeringite [$\text{MgAl}_2(\text{SO}_4)_4 \cdot 22\text{H}_2\text{O}$], Boyleite [$(\text{Zn,Mg})\text{SO}_4 \cdot 4\text{H}_2\text{O}$], jarosite [$\text{KFe}^{3+}(\text{SO}_4)_2(\text{OH})_6$], rozenite ($\text{FeSO}_4 \cdot 4\text{H}_2\text{O}$), nimite [$(\text{Ni, Mg, Al})_6(\text{Si, Al})_4\text{O}_{10}(\text{OH})$], hetaerolite ($\text{Zn Mn}^{3+}\text{O}_4$), smithsonite (ZnCO_3), hematite (Fe_2O_3), bernalite [$\text{Fe}(\text{OH})_3$] as well as cryptocrystalline or amorphous precipitates and poorly crystallized Fe-oxyhydroxides. Keoheite, a cryptocrystalline mixture of quartz + gypsum + sphalerite was identified in some samples from El Fraile, La Concha and Guerrero I impoundments.

Sulfides generally appear as free grains but are also present as inclusions in quartz and other non-metallic phases (Figure 3). In oxidized tailings, all primary sulfides show partial or nearly total transformation to secondary phases, whereas in non-oxidized, sulfides are weakly altered or fresh (Figures 3a, b, c). Pyrite is largely the sulfide phase that shows the most intense alteration as reported by several authors in other tailings (e.g. Ritcey, 1989). Some grains have been completely altered to poorly crystallized Fe-oxyhydroxides and in other cases grain alteration is limited to the rim (Figures 3c, e, f). Chalcopyrite, sphalerite and galena also show partial alteration, although it is systematically less penetrative. Chalcopyrite is transformed to a secondary Cu-phase, probably chalcocite, whereas sphalerite and galena are transformed to Zn- and Pb-oxides, respectively (Figures 3d, f).

SEM observations and EDS analyses show that secondary Fe oxyhydroxides either around pyrite or as free precipitates contain significant amounts of S, As, Pb, Mn and Zn probably as adsorbed elements. On the other hand, Zn and Pb oxides produced by alteration of sphalerite and galena, respectively contain Mn, Fe and As as minor elements.

Total contents

Environmentally available base metals (Ag, Cu, Pb and Zn) and related elements (As, Cd, Mn and Fe) are presented

in Table 2. Arithmetic mean and standard deviation (SD) of each dump are also shown. Bulk average concentrations and its calculated SD including all the studied samples are presented at the bottom of Table 2. In this study, metal concentrations of tailings are compared to regional background concentrations (RBC) of crop soils in order to determine enrichment ratios of tailings relative to regional pristine soils. This approach is based on the following observations: (i) tailings are fine-grained particulate, which is similar in several physical aspects to soils; (ii) some tailings impoundments are located at or near communities and inhabitants are currently in physical interaction with them; (iii) most soils around tailings are used for crops; (iii); and, (iv) tailings particulate is constantly added to nearby soils via aerial deposition and the use of tailings-derived polluted water for crop irrigation. The range of enrichment factors for each studied dump is shown in Table 2.

All tailings and waste-rock accumulations show high concentrations of all studied metals, although concentrations are variable suggesting high heterogeneity among the different mine waste impoundments. Silver contents range from 9.50 to 74.20 mg/kg with a bulk average (\pm SD) of 38.07 (\pm 14.60) mg/kg. Concentrations of Cu are more variable and range from 71.8 to 1320 mg/kg with a bulk average of 374 (\pm 322) mg/kg. Lead contents show the widest variation recorded in tailings and range from 780 to 43 700 mg/kg. The bulk average of lead is 8761 (\pm 9842) mg/kg. Zinc contents have also a great variability ranging from 380 to >10 000 mg/kg with a bulk average of 5581 (\pm 3820) mg/kg. Arsenic, which is associated to silver sulfosalts and arsenopyrite, ranges from 19 to 11 800 mg/kg with a bulk average of 1727 (\pm 1742) mg/kg. Similarly, cadmium is commonly associated with zinc ores and varies between 1.0 and 780 mg/kg with a bulk average of 143 (\pm 209) mg/kg. Iron, which is a major constituent of pyrite and crystalline Fe-oxides and it is also present in chalcopyrite and arsenopyrite, shows a significant variability. It ranges from 2.49 to 25.1% with a bulk average of 8.9 (\pm 4.5) %. Finally, manganese, commonly associated to Fe-oxides, varies from 18.6 to 13 800 mg/kg with a bulk average of 2483 (\pm 3129) mg/kg.

Using average concentrations of each dump, it can be stated that La Concha pond contains the highest concentration of Cd, Cu, Pb and Zn whereas the Guerrero II tailings contain the lowest contents of Cu, Pb and Zn. The San Antonio waste-rock dump contains the highest contents of Ag, Fe and Zn and the Guerrero I the lowest levels of Ag, Cd and As. The highest values of Mn and V are observed at Los Jales impoundment and the lowest Fe and Mn concentrations in El Fraile.

When compared to the RBC of crop soils, Taxco tailings are highly enriched in most metals, although enrichment ratios vary from one dump to another (Table 2). If only

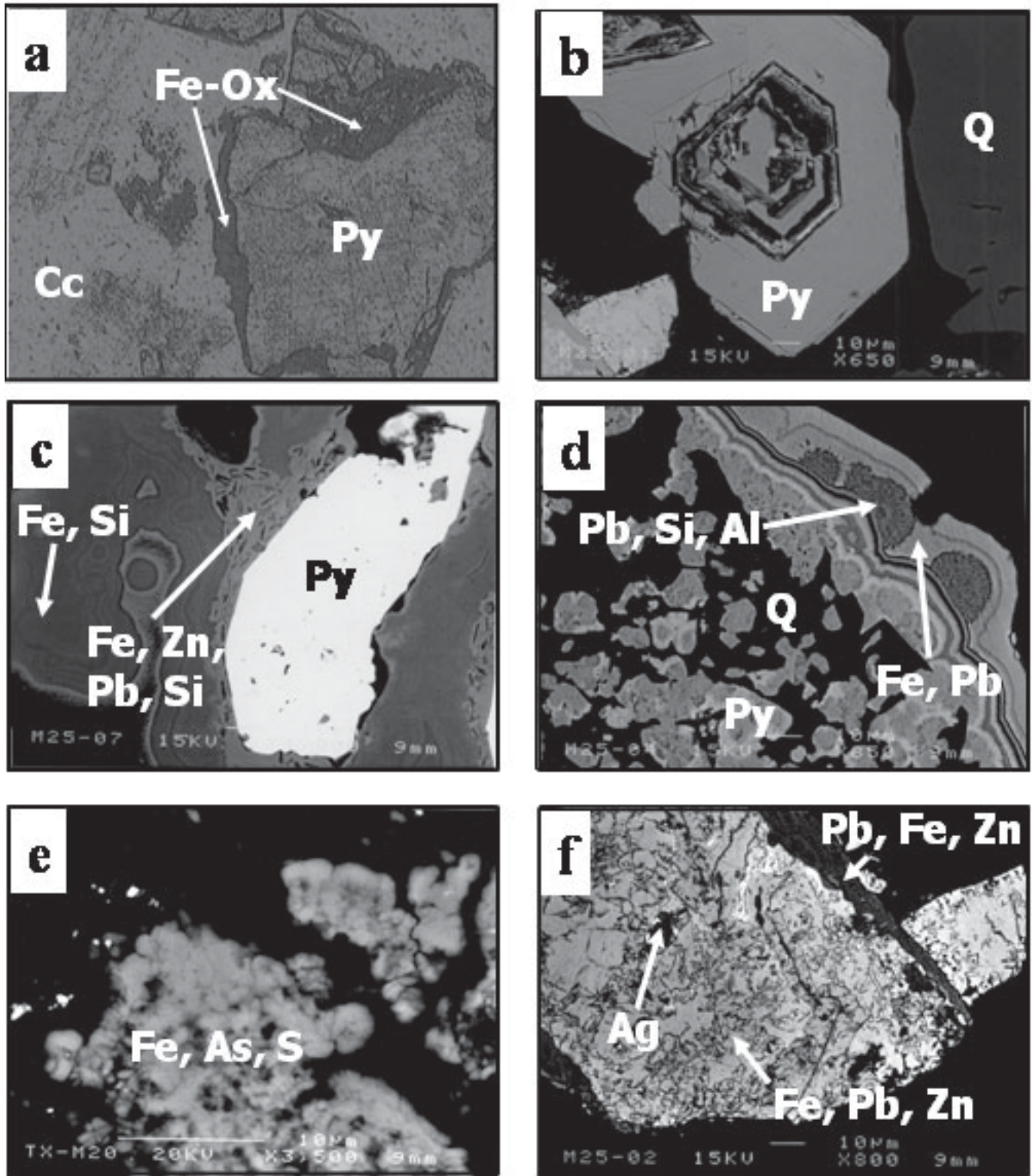


Fig. 3. Optical and backscattered photographs of representative samples of tailings from Taxco, Guerrero impoundments. (a) Partly altered pyrite and calcite from a non-oxidized layer in the (current) Guerrero II tailings dump; (b) fresh pyrite showing concentric growth and quartz from a non-oxidized layer in the La Concha tailings; (c) partly altered pyrite showing a corona of complex Fe-oxyhydroxides containing significant amounts of Pb, Zn and Si; (d) corona of Pb oxides and Fe-oxyhydroxides around a quartz grain containing inclusions of partly oxidized pyrite; (e) free Fe-oxyhydroxide grain containing trace amounts of As and S; (f) secondary Fe-oxide containing relicts of Ag-rich phases and trace of Pb and Zn. A Pb rich secondary oxide containing Fe and Zn can be observed in the upper border of the Fe oxide.

Table 2

Concentrations of total metals in the Taxco, Guerrero tailings and waste-rock dumps

Sample	Ag (mg/kg)	Cd (mg/kg)	Cu (mg/kg)	Fe (%)	Mn (mg/kg)	Pb (mg/kg)	V (mg/kg)	Zn (mg/kg)	As (mg/kg)
RBC	2.20	1.00	25	2.70	409	26	40	64	30
El Fraile									
M-01	27.40	37.00	149	4.0	440	2910	16.00	3820	1360
M-02	32.20	86.00	220	5.5	1320	1780	16.00	7830	2120
M-03	31.40	10.00	124	6.4	420	2230	40.00	880	1920
M-04	20.90	13.00	151	3.8	157	3110	7.00	1680	580
M-05	33.50	8.00	107	4.0	220	3690	11.00	1160	840
M-06	21.30	2.00	220	3.4	68.00	4250	5.00	380	420
M-07	31.50	13.00	170	3.7	220	4370	4.00	1360	780
M-08	26.10	9.00	260	5.0	260	6620	15.00	1140	860
M-09	29.40	16.00	320	6.4	164	1070	< 2.00	1780	2270
M-10	35.50	13.00	280	6.0	340	4900	17.00	1700	920
M-11	34.50	4.00	189	5.2	122	6040	14.00	780	800
M-12	36.10	11.00	182	7.8	360	3440	14.00	1400	580
M-13	18.90	150.00	420	4.7	5070	2980	49.00	>10000	720
M-14	44.70	181.00	420	8.4	4160	4120	10.00	>10000	2720
M-15	33.70	40.00	166	7.8	780	3640	24.00	2860	2970
Mean	30.50	39.50	225	5.5	940	3677	16.30	3118	1324
STD	6.80	55.60	98	1.6	1534	1481	12.90	3329	846
AEF	13.86	39.50	9.08	2.04	2.30	144.18	0.41	48.49	44.88
La Concha									
M-16	55.10	520	1020	12.3	6110	19700	8.00	>10000	2150
M-17	34.90	8.00	71.80	6.0	57.00	20300	2.00	780	1140
M-18	66.30	560	1080	11.9	5780	29900	4.00	>10000	1960
M-19	49.90	300	840	10.7	2850	14300	4.00	1140	2380
M-20	55.90	65.0	146	11.7	114	37900	2.00	8040	11800
M-21	34.10	76.0	320	10.5	1360	10100	2.00	>10000	2240
M-22	59.90	780	1020	8.3	6630	43700	13.00	>10000	4590
M-23	41.20	540	860	9.5	7020	11200	5.00	>10000	1440
M-24	42.90	500	800	9.4	7950	12500	5.00	>10000	1420
M-25	58.00	680	920	12.0	5580	21400	2.00	>10000	2580
M-26	53.00	720	1320	11.3	7250	17700	2.00	>10000	1680
Mean	50.10	432	763	10.3	4609	21700	4.50	8178	3035
STD	10.50	277	405	1.9	2953	11045	3.40	3617	3050
AEF	22.77	432	30.78	3.81	11.27	850.98	0.11	127.19	102.86
El Solar									
M-27	25.00	171	280	9.7	1220	1580	38.00	>10000	1600
M-28	37.80	46.00	134	11.2	3590	1900	5.00	5360	1360
M-29	28.30	30.00	158	4.3	780	1780	15.00	3230	800
M-30	35.60	99.00	195	16.1	1610	1260	19.00	10000	1780
M-31	26.70	39.00	150	6.6	4360	980	37.00	4300	1340
M-32	55.00	20.00	166	8.1	400	1200	8.00	1960	2360
M-33	63.50	8.00	143	6.5	176	1260	7.00	1200	1320
M-34	18.00	25.00	118	8.9	1030	840	51.00	3130	1320
M-35	23.10	71.00	460	6.4	920	5850	28.00	4290	440
Mean	34.80	56.60	200	8.6	1565	1850	22.00	4830	1369
STD	15.30	51.10	108	3.5	1442	1540	15.50	3185	547
AEF	15.82	56.60	8.08	3.2	3.83	72.55	0.55	75.12	46.41

Table 2, Continued...

Sample	Ag (mg/kg)	Cd (mg/kg)	Cu (mg/kg)	Fe (%)	Mn (mg/kg)	Pb (mg/kg)	V (mg/kg)	Zn (mg/kg)	As (mg/kg)
RBC	2.20	1.00	25	2.70	409	26	40	64	30
Guerrero I									
M-36	22.30	10.00	340	8.24	1060	4050	25.00	4150	520
M-37	52.10	9.00	440	10.50	1120	>10000	28.00	5540	560
M-39	22.50	22.00	400	9.87	2030	>10000	28.00	7370	460
M-40	40.80	1.00	380	6.41	300	7010	16.00	2380	500
M-41	17.50	11.00	260	9.07	420	3150	34.00	4020	700
M-42	9.50	1.00	76.80	19.40	3970	3340	7.00	8290	220
Mean	27.50	9.00	316	10.60	1483	6258	23.00	5292	494
STD	15.90	7.80	132	4.50	1366	3213	9.80	2226	157
AEF	12.50	9.00	12.75	3.93	3.63	245	0.58	82.30	16.75
Guerrero II									
M-43	48.50	15.00	121	7.0	1240	2280	9.00	1660	1200
M-44	31.10	1.00	105	8.2	480	780	2.00	600	800
M-45	31.70	4.00	72.30	8.0	960	820	11.00	1120	1400
M-46	29.80	17.00	107	8.1	740	1300	8.00	940	3800
M-47	47.70	6.00	95	8.9	740	1560	4.00	1040	1060
M-48	55.40	78.00	180	10.6	98.20	1740	4.00	7830	3030
Mean	40.70	20.20	113	8.5	2330	1413	6.30	2198	1882
STD	11.10	29.00	36.40	1.2	3678	574	2.50	2780	1228
AEF	18.50	20.20	4.57	3.1	5.70	55.41	0.16	34.18	63.80
Los Jales									
M-50	37.30	320	546	8.3	7220	11700	20.00	>10000	1440
M-51	35.50	277	595	13.9	13000	32700	32.00	>10000	5210
M-52	18.80	107	150	6.2	13800	8090	127.00	4590	898
JA1	26.70	113	178	3.8	3160	9130	14.00	>10000	431
JA2	20.90	41.00	76.50	2.5	18.60	3680	35.00	3560	277
JA3	38.80	66.00	875	10.8	64.00	12500	13.00	6700	2034
JA4	74.20	32.00	1220	25.1	3340	17100	46.00	>10000	408
Mean	36.00	137	520	10.1	6063	13557	41.00	7836	1672
STD	18.60	115	423	7.7	5059	9404	39.80	2853	1832
AEF	16.36	137	20.97	3.7	14.82	532	1.03	122	56.68
San Antonio									
T-01	69.10	367	566	16.0	2460	3610	12.00	>10000	3660
T-02	41.40	545	522	6.3	4570	7160	5.00	>10000	1410
T-03	48.40	58.00	415	24.1	174	30700	24.00	9310	4100
T-04	52.20	387	578	10.1	6310	17100	7.00	>10000	2220
T-05	52.60	121	172	11.1	313	6920	8.00	>10000	19.00
Mean	52.70	296	451	13.5	2765	13098	11.20	9862	2460
STD	10.20	202	169	6.9	2676	11060	7.60	209	1024
AEF	23.95	296	18.19	5.0	6.76	514	0.28	153	83.39
Bulk Mean	38.07	143	374	8.9	2483	8761	17.55	5581	1727
STD	14.60	209	322	4.5	3129	9842	19.54	3820	1742

average enrichment factors (AEF) are considered, Pb shows the widest range of enrichment ratios ranging from 55.4 times the RBC found in the Guerrero II tailing to 851 times the RBC in La Concha impoundment. Enrichment ratios of Cd are also variable ranging from 9 times the RBC recorded in the Guerrero I tailings to 431.7 times the RBC in La Concha dump. Zinc is another metal that shows one of the highest enrichment factors, from 34.2 times the RBC reported in the Guerrero II tailings to 153.4 times the RBC recorded in the San Antonio waste-rock dump. Arsenic shows lower enrichment factors, although it is one of the metals with of the highest ecotoxicological concern. It shows enrichment ratios ranging from 16.8 times the RBC in the Guerrero I tailings to 102.9 times the RBC in La Concha dump. The other metals (Cu, Ag, Fe and Mn) are also enriched, although the enrichment ratios are substantially lower and within the following ranges: Cu = 4.6 - 30.8 times the RBC; Ag = 12.5 - 24.0 times the RBC; Fe = 2.0 - 5.0 times the RBC; and, Mn = 2.3 - 14.8 times the RBC.

Water-soluble metals

Physical-chemical parameters (electrical conductance, pH) and water-soluble metal contents were determined in samples from two impoundments (La Concha and El Fraile) in order to evaluate their capacity for releasing metals through simple washing processes. These impoundments were selected because they are located near or at communities where inhabitants are in continuous interaction with tailings and because leachates are used by some inhabitants as tap water, particularly during dry seasons. Results are reported in Table 3.

Samples from both impoundments show a wide range of pH values from slightly basic (8.4) to highly acidic (1.5). In the detail, La Concha samples show either neutral to slightly basic (pH = 7.0-8.4) or acid (pH < 2.9) values with a wide gap between 2.9 and 7.0, whereas samples from El Fraile span over all the observed range. With the exception of a few samples, electrical conductance, although variable, is systematically high in both impoundments ranging from 100 to 5180 $\mu\text{S}/\text{cm}$.

Water-soluble contents of metals show a wide variation. Ag contents are very low in both impoundments and most samples contain less than 0.005 mg/L. A few acidic samples contain somewhat higher levels but they are usually low (< 0.035 mg/L). Soluble Pb shows a similar behavior than Ag, with most samples containing less than 0.025 mg/L and only a few samples show more elevated concentrations (0.043-0.655 mg/L). On the contrary, contents of soluble Cd, Mn and Zn are mostly high in both impoundments and increase systematically with decreasing pH (Figure 4). In El Fraile samples, Cd ranges from 0.025 to 19.2

mg/L and in La Concha dump Cd contents fluctuate from < 0.025 to 11.8 mg/L. Contents of soluble Mn go from < 0.025 to 205 mg/L in El Fraile whereas that in La Concha samples contents are significantly higher ranging from < 0.025 to 393 mg/L. Soluble Zn ranges from 0.048 to 397 mg/L in El Fraile and from < 0.025 to 393 mg/L in La Concha samples. Contents of Cu show a contrasting behavior in samples from both impoundments. With a few exceptions, in near neutral or slightly acidic (pH > 5.0) samples, Cu concentrations are relatively low (< 0.283 mg/L) but in moderately to highly acidic (pH < 5.0) samples, concentrations are very variable ranging from < 0.025 to 63 mg/L with most samples falling in the range of 1.0 to 63 mg/L. Despite this high variation, there is a good negative correlation of soluble Cu with pH (Figure 4). A similar behavior is observed with soluble Fe. In samples with pH values higher than 4.4, Fe contents are low (< 0.025 mg/L) whereas in more acidic samples (pH < 4.4) soluble Fe increases substantially with concentrations in the range of 84 to 1105 mg/L (Figure 4). Finally, soluble As contents are low (< 0.010 mg/L) in most samples from both impoundments regardless of pH values. However, some few samples with either near neutral (7.4-7.7) or acidic pH (2.5-3.5) contain higher concentrations of soluble As (0.264-0.746 mg/L) indicating that As release is possible at a wide range of pH.

DISCUSSION AND CONCLUSIONS

Mining in Taxco is a tradition of more than 450 years. During this period, large amounts of metals, mainly silver but also lead, zinc and copper have been exploited and Taxco became one of the largest silver producing area in Mexico. Currently, it accounts for more than 90% of metals produced in the State of Guerrero. Large amounts of tailings and waste-rock dumps have been deposited in the area. However, only tailings and waste-rocks generated during the modern period (1940 to present) have been recognized around Taxco. Older wastes were mostly reworked and thus, colonial tailings and waste-rocks are rare.

The primary mineralogy recognized in the tailings is typical of epithermal Pb-Zn-Ag Cu deposits with pyrite as the dominant sulfide phase accompanied with sphalerite, galena, chalcopyrite, pyrrothite, rare Ag-sulfosalts, arsenopyrite and crystalline Fe-oxides. Secondary phases are conspicuous in all dumps and include a variety of Fe-oxyhydroxides, hydroxide sulfates and carbonates. Although a real quantification of the sulfides (particularly pyrite) as acid source and of the carbonates (particularly calcite) as neutralization phases was not performed in this study, field observations together with optical and SEM observations indicate that oxidation of sulfide phases is strong and has occurred everywhere in the older tailings. High electrical conductance and the presence of secondary minerals like gypsum and jarosite in some samples from the Guerrero II and the upper part of La Con-

Table 3

Physical-chemical parameters and concentrations of water-soluble metals in El Fraile and La Concha tailings impoundments in Taxco, Guerrero

El Fraile Tailings											
	pH	Cond (microS/cm)	Ag (mg/L)	Cd (mg/L)	Cu (mg/L)	Fe (mg/L)	Mn (mg/L)	Pb (mg/L)	V (mg/L)	Zn (mg/L)	As (mg/L)
JF-01	7.4	2070	<0.005	0.091	0.033	<0.025	4.48	0.060	<0.025	7.29	<0.010
JF-02	7.4	1078	<0.005	0.116	<0.025	<0.025	6.63	<0.025	<0.025	3.28	0.746
JF-03	7.7	336	<0.005	1.13	<0.025	<0.025	1.23	<0.025	<0.025	1.11	0.522
JF-04	2.9	1810	<0.005	2.02	13.9	579	38.9	0.237	0.314	123	<0.010
JF-05	3.2	1619	<0.005	1.18	5.55	522	29.4	0.237	0.263	92	<0.010
JF-06	2.7	3950	<0.005	4.40	17.9	585	87	0.274	0.246	179	<0.010
JF-07	3.4	1501	<0.005	0.525	4.95	623	31.9	<0.025	<0.025	56	<0.010
JF-08	4.2	2360	<0.005	0.251	1.27	154	8.70	<0.025	0.039	23.1	0.019
JF-09	7.4	1070	0.016	0.125	<0.025	<0.025	10.3	<0.025	<0.025	5.48	<0.010
JF-10	3.4	1574	0.029	1.13	1.93	458	166	<0.025	<0.025	152	<0.010
JF-11	3.4	4020	0.009	3.78	9.31	396	98	<0.025	<0.025	162	<0.010
JF-12	7.6	2090	<0.005	0.025	0.027	<0.025	4.06	<0.025	<0.025	1.07	0.011
JF-13	2.5	4460	<0.005	1.80	20.0	929	37.6	<0.025	0.345	94	0.573
JF-14	3.5	2840	<0.005	2.35	4.08	449	51	<0.025	<0.025	120	<0.010
JF-16	7.9	1699	0.013	0.525	0.038	<0.025	0.069	<0.025	<0.025	0.048	0.032
JF-17	3.1	2850	<0.005	0.138	1.71	144	5.19	0.069	0.036	16.4	<0.010
JF-18	5.3	2570	0.059	4.14	0.065	<0.025	179	<0.025	<0.025	171	<0.010
JF-19	3.5	2520	0.035	0.060	0.782	275	10.3	<0.025	<0.025	13	<0.010
JF-19A	4.4	2480	0.051	3.26	0.688	<0.025	152	<0.025	<0.025	154	0.011
JF-20	2.4	5180	<0.005	2.64	16.7	762	83	<0.025	<0.025	180	0.017
JF-20A	3.7	2660	<0.005	0.440	0.678	410	24.7	<0.025	<0.025	64	<0.010
JF-21	3.8	2740	<0.005	0.506	0.237	553	26.3	<0.025	<0.025	66	<0.010
JF-22	3.7	3610	<0.005	0.602	8.96	1052	15.1	0.090	0.346	44.3	<0.010
JF-23	3.8	2340	<0.005	0.040	0.134	210	3.26	0.047	0.045	6.24	<0.010
JF-24	3.1	2850	<0.005	0.175	2.68	638	4.33	0.253	0.173	10.6	<0.010
JF-25	3.3	3280	<0.005	0.666	4.09	516	34.4	0.057	0.137	88	<0.010
JF-26	3.6	1235	<0.005	0.050	0.658	237	1.09	<0.025	0.058	2.70	<0.010
JF-27	3.3	1559	<0.005	0.425	9.88	540	40.0	0.043	0.150	54	0.045
JF-28	2.7	3850	<0.005	19.2	63	1105	<0.025	0.314	0.534	397	0.264
JF-29	7.5	1080	<0.005	0.157	<0.025	<0.025	7.81	<0.025	<0.025	3.06	<0.010
JF-30	3.4	2730	<0.005	0.200	2.82	483	7.17	0.068	0.131	17.1	<0.010
JF-31	3.3	1431	<0.005	0.318	5.45	310	34.0	0.278	0.083	39.0	<0.010
JF-32	3.1	2160	<0.005	7.93	0.092	655	205	0.363	0.200	268	0.088
JF-33	3.5	1380	<0.005	0.236	5.29	391	25.1	0.106	0.133	29.3	<0.010
JF-34	3.3	1360	<0.005	0.206	6.33	355	7.16	0.100	0.109	13.9	<0.010

Table 3, Continued...

La Concha Tailings											
	pH	Cond (microS/cm)	Ag (mg/L)	Cd (mg/L)	Cu (mg/L)	Fe (mg/L)	Mn (mg/L)	Pb (mg/L)	V (mg/L)	Zn (mg/L)	As (mg/L)
JC-01	7.0	100	<0.025	<0.005	<0.025	<0.025	0.093	<0.025	0.305	<0.025	<0.010
JC-02	1.5	1670	<0.025	11.80	11.20	842	257	0.655	<0.025	257	1.790
JC-03	7.7	1476	<0.025	<0.005	<0.025	<0.025	<0.025	<0.025	<0.025	<0.025	<0.010
JC-04	2.3	1340	<0.025	3.79	<0.025	<0.025	166	0.130	<0.025	166	<0.010
JC-05	8.4	1123	<0.025	<0.005	0.069	<0.025	<0.025	<0.025	<0.025	<0.025	<0.010
JC-06	1.7	1837	<0.025	3.59	3.27	84.0	267	<0.025	<0.025	267	<0.010
JC-07	7.9	929	<0.025	<0.005	<0.025	<0.025	<0.025	<0.025	<0.025	<0.025	<0.010
JC-08	1.5	2030	<0.025	7.91	7.69	283	393	<0.025	<0.025	393	0.060
JC-09	7.8	927	<0.025	<0.025	0.067	0.067	<0.025	<0.025	<0.025	<0.025	<0.010
JC-10	2.9	1293	<0.025	1.68	0.403	0.259	152	-	<0.025	152	<0.010
JC-11	7.4	520	<0.025	<0.005	<0.025	<0.025	<0.025	<0.025	<0.025	0.255	<0.010
JC-12	7.8	493	<0.025	<0.005	0.055	<0.025	<0.025	<0.025	<0.025	0.245	<0.010
JC-13	7.9	742	<0.025	<0.005	<0.050	<0.025	<0.025	<0.025	<0.025	0.874	<0.010
JC-14	7.6	519	<0.025	<0.005	0.067	<0.025	0.886	<0.025	<0.025	1.46	<0.010
JC-15	7.9	1614	<0.025	<0.005	0.055	<0.025	<0.025	<0.025	<0.025	0.572	<0.010
JC-16	7.8	102	<0.025	<0.005	0.052	<0.025	<0.025	<0.025	<0.025	0.658	<0.010
JC-17	7.8	222	<0.025	<0.005	<0.050	<0.025	<0.025	<0.025	<0.025	2.57	<0.010
JC-18	7.3	540	<0.025	0.239	<0.050	<0.025	<0.025	<0.025	<0.025	6.99	<0.010
JC-19	7.8	1676	<0.025	0.042	0.283	<0.025	<0.025	<0.025	<0.025	2.02	<0.010
JC-20	7.7	927	<0.025	0.030	0.07	<0.025	<0.025	<0.025	<0.025	2.24	<0.010

cha impoundments suggests that oxidation has also occurred locally in a moderate extent even in the recent tailings impoundments. Measurements by EDS of secondary phases indicate that important amounts of As, Pb and Zn are fixed to Fe-oxyhydroxides, likely as adsorbed ions (Smith, 1999; Dold, 1999; Dold and Fontboté, 2001).

All studied metals (Ag, Cd, Cu, Fe, Mn, Pb, V, Zn and As) appear to be highly enriched in all dumps relative to RBC of soils, although total contents are variable even within a single dump indicating that tailings and waste-rocks are a very heterogeneous media. With the exception of vanadium, all metals are well above the RBC in several orders of magnitude. Lead, cadmium and arsenic, considered of high environmental and toxicological concern are among the metals with the highest enrichment factors and represent a major source of metal pollution to the environment and constitute a risk to the nearby resources. This is of prime importance in El Fraile and La Concha sites where tailings are located within communities and people are in permanent interaction with them. Some inhabitants around these tailings use leachates

for domestic usage or for crop irrigation. In these two impoundments, pH of tailings is very variable spanning from slightly basic (8.4) to highly acidic (1.5) illustrating the great heterogeneity of tailings. Contents of water-soluble metals are also very variable and indicate that release of metals occurs particularly at low pH (<4.5). At higher pH values, concentrations of metals decrease drastically falling in most samples below the method detection limit. Such a behavior would suggest that at pH values between 4.5 and 8.4 metal concentrations are mainly controlled by the precipitation of Al-hydroxides, Fe-oxyhydroxides and carbonates (particularly calcite) and that metals are essentially immobilized by co-precipitation or by sorption onto surfaces (Jambor, 1994; Drever, 1997; Dold and Fontboté, 2002). In most samples, arsenic concentrations seem to be also controlled by precipitation of these phases. However, the relative high concentrations of arsenic recorded in some near neutral samples (pH=7.4-7.7) from El Fraile impoundment suggest the involvement of other mechanisms that control the mobility of arsenic, including probably desorption and complexation processes.

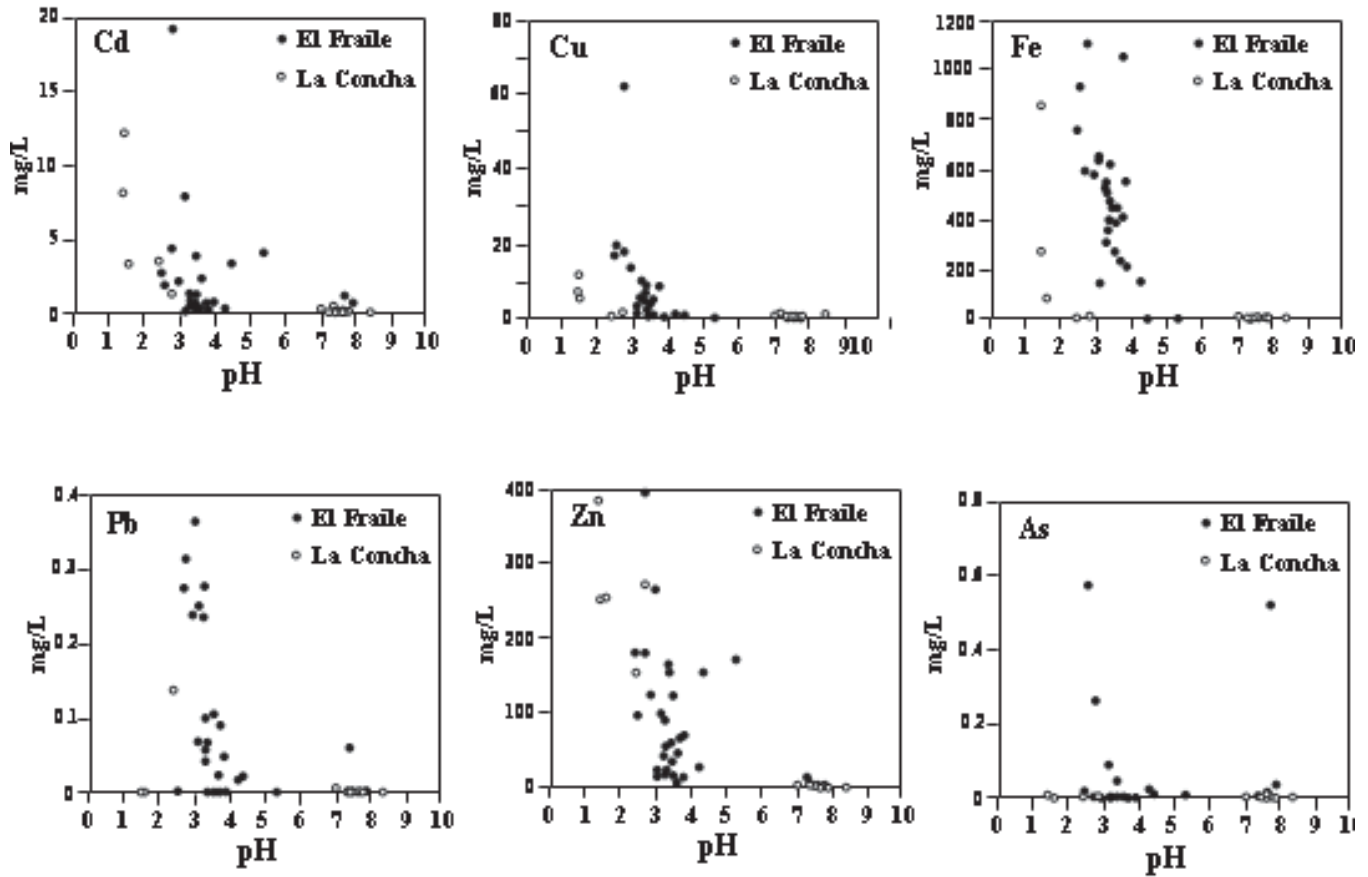


Fig. 4. Graphs of pH versus water-soluble contents of selected metals in samples from El Fraile and La Concha tailings dumps.

Physical-chemical characteristics indicate that Taxco tailings have a large capacity for generating AMD with large amounts of toxic metals and sulfate. However, the presence of calcite in some samples, particularly in the less oxidized ones, indicate that there is still an important capacity of the system for neutralizing AMD and, consequently, for retaining metals (Jambor, 1994; Paktunc, 1998; Walton-Day, 1999; Dold, 1999). Data reported by García Abundis (2002) on leachates from El Fraile indicate that despite the degree of oxidation of tailings, leachates are essentially neutral solutions with moderate concentrations of sulfate, Zn, Fe, Mn and As, which are distinctively lower than typical AMD. These data evidence the bulk capacity of system for neutralizing acid solutions. Similar characteristics were recorded by Armienta *et al.* (2000) in leachates from the Guerrero I tailings. Jambor (1994) pointed out that neutralization of AMD by calcite typically reaches pH above 6.5 whereas neutralization by Al- and Fe-oxyhydroxides generates slightly (pH = 4.5-6.5) acid leachates. According to this author, when the capacity of these phases for neutralizing AMD is depleted, clays and hydroxide silicates are the only buffer-controlling minerals leading to the generation of very acidic (pH < 3.0) solutions. At this point, mobility of metals is at maximum. Thus, according to

the data reported by García-Abundis (2002), Armienta *et al.* (2000) and our data, it is highly probable that calcite still acts as the main pH-controlling buffer in Taxco tailings. This process enables the precipitation of secondary phases, mainly Fe-oxyhydroxides and sulfates, which fix metals either as major or minor constituents or as adsorbed ions as recorded by Nordstrom and Alpers (1999). However, extracts with pH values between 1.5 and 6.5 suggest that the neutralization capacity of calcite is locally depleted and that neutralization by other phases including Al- and Fe-oxyhydroxides and clay actually occurs. Moreover, the data strongly suggest that, in a short period of time, most oxidized tailings in Taxco would lose their capacity for neutralizing AMD and mobilization of toxic metals will increase dramatically.

Thus, the degree of oxidation of tailings together with the widespread presence of secondary phases and the physical-chemical characteristics of tailings, strongly suggests that the generation of AMD coupled with dissolution, neutralization and reprecipitation processes occur normally in all tailings. These processes are considered the main factors controlling the mobility and availability of metals in the Taxco tailings.

ACKNOWLEDGMENTS

Authors are very grateful to Elvia Díaz V. for invaluable help and encouragement along all the stages of the research. We thank F. Barra for useful comments and English corrections. Funding was provided by SIBEJ (Sistema de Investigación Benito Juárez) grant NRMA-043 and by CONACYT (Consejo Nacional de Ciencia y Tecnología) grant G28975T. Detailed studies of La Concha and El Fraile impoundments constituted the bachelor thesis of ADA and NFM, respectively at University of Guerrero. Discussions with M. A. Armienta and F. Romero are gratefully acknowledged. Critical comments and suggestions by O. Morton and two anonymous reviewers greatly help to improve the manuscript.

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