



Mechanism of removal and retention of heavy metals from the acid mine drainage to coastal wetland in the Patagonian marsh



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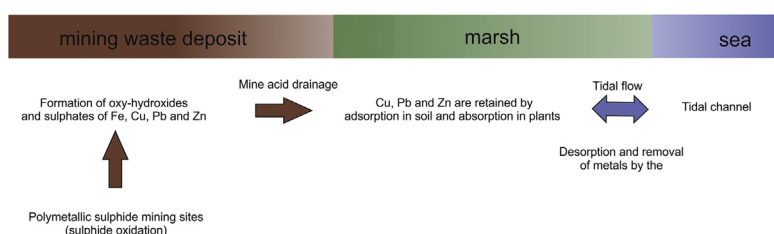
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HIGHLIGHTS

- Mobilization mechanisms of metals from the mining waste deposits were determined.
- Metal are retained by adsorption in salt marsh soils.
- Also, metals are absorbed by salt marsh plants.
- Marsh is an active environment in mitigating pollution caused by acid drainage.

GRAPHICAL ABSTRACT



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ABSTRACT

The attenuation of the acid mine drainage is one of the most important environmental challenges facing the mining industry worldwide. Mining waste deposits from an ancient metallurgical extraction of heavy metals were found near to the San Antonio marsh in Patagonia. The aim of this work was to determine which mechanisms regulate the mobilization and retention of metals by acid drainage. A geological and geomorphological survey was carried out and samples from the mining waste deposits and the marsh were collected to determine soil texture, Eh pH, organic matter, Cu, Pb, Zn and Fe content, and soil mineralogical composition. Metals in marsh plants were determined in above- and below-ground structures. In the mining waste deposits polymetallic sulphides were recognized where the oxidation and formation of oxy-hydroxides and sulphates of Fe, Cu, Pb and Zn occurs. Then, by the alteration of those minerals, the metals enter in solution and are mobilized with the surface drainage towards the marsh where adsorption in the soils fine fraction and organic matter and/or by plants occurs. Locally, in the mining waste deposits, the precipitation/dissolution of Cu, Pb, and Zn sulphates take place in small centripetal drainage basins. In topographically lower portions of the marsh desorption and removal of metals by tidal flow could also be happen. The results allow to concluding that the marsh adjacent to the mining waste deposits is a geochemically active environment that naturally mitigates the contamination caused by acid drainage.

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1. Introduction

The attenuation of the acid mine drainage is one of the most important environmental challenges facing the mining industry worldwide. The acid mine drainage occurs when the sulphide

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mineral phases on oxidant conditions contact and react with water resulting in the formation of sulphates and soluble metals as well as protons which generate a decrease in pH. A the insidious feature of acid mine drainage is that its fonts can remain active for decades or even centuries after mine closure (Modis et al., 1998). Both operational and abandoned polymetallic sulphide minerals are often active sources of acid mine drainage. The metal load is of greater concern than the acidity in the terms of environmental damage (Sheoran and Sheoran, 2006).

Natural wetlands can reduce the load of pollutants contained in wastewaters through different processes such as plant uptake, bacterial degradation, oxidation of contaminants, sedimentation and adsorption of particles and substances dissolved in the residue on the substrate (Tam and Wong, 1994; Matagi et al., 1998; Mays and Edwards, 2001; Hwang et al., 2006; Reboreda and Caçador, 2007; Sun et al., 2016). Given these features, in the last decades wetlands have begun to be used for treatment of wastewater in order to reduce the possibility of harmful impacts on humans (Sheoran and Sheoran, 2006). Wetlands were used also for the treatment of acid mine drainage derived from metalliferous and coal mining (Groudev et al., 2001; Ye et al., 2001; Johnson and Hallberg, 2005).

The salt marsh surrounding the San Antonio Bay is located in the littoral of the Argentinean Patagonia (Fig. 1) and constitutes a wetland that was designated as Natural Reserve in 1993. Although natural areas are conserved in this salt marsh, there are also areas affected by anthropic action. Examples of these affectations are the mining wastes originated by the metallurgical extraction of polymetallic sulphide that were deposited more than four decades ago in the proximities of San Antonio Oeste City (Idaszkin et al., 2015, 2017). The aim of this work was to determinate which mechanisms regulate the mobilization and retention of the heavy metals from the mining waste deposits by acid drainage toward the intertidal wetland located in the salt marsh next to the deposits. To this end, the present mineralogy of the mining waste deposits and the contents of Cu, Pb, Zn and Fe in the soils and vegetation of the salt marsh are analyzed together with some physicochemical soil properties. The results obtained allow as to establish the environmental conditions of this area of the natural reserve as well as to evaluate the wetland capacity to mitigate the effects of the acid drainage.

2. Study area

The San Antonio salt marsh is located surrounding the San Antonio Bay (40°44'S, 54°68'W; Río Negro, Argentina; Fig. 1). Beach ridges and coastal spits, composed by sands and gravels with mollusc shells, limits the marsh area that it is composed by silty clay loam to sandy loam (Angulo et al., 1981). The tide have a semidiurnal macro-tidal regime (tidal amplitude: ~9 m) and periodically flood the active tidal channels recognized in the marsh. Towards the more continental sectors old tidal channels that are actually out of the tidal cycle were also recognized. Beach ridges and coastal spits are positive geomorphological features where the superficial drainage tends to be collected by the tidal channels and drained towards the marsh. The high permeability sediments that constituted the beach ridges and coastal spits facilitate the rain-water infiltration that then discharges with the underground flow towards the marsh.

The climate is semi-arid, the mean annual precipitation is around 250 mm, and the mean annual air temperature is 15.1 °C, fluctuating between mean extremes of -7.7 °C in July to 41.4 °C in February (Firstater et al., 2016). The mean annual wind speed is 18 km h⁻¹, reaching an average of 25 km h⁻¹ in the hotter seasons (Firstater et al., 2016). The salt marsh is dominated by *Spartina*

species, being *Spartina alterniflora* the only species present in the lower marsh zone, and the higher zones are inhabited by *Spartina densiflora*, and the shrubs *Limonium brasiliense*, *Sarcocornia perennis* and *Atriplex* spp (Bortolus et al., 2009; Idaszkin et al., 2015).

3. Methodology

Based on the geomorphological features of the area, a geological survey of the mining waste deposits located in the vicinity of the salt marsh of San Antonio Oeste was done. Soil samples were taken to identify the mineralogy and to determinate pH in saturated paste. Sampling was performed according to the drainage traits observed within the deposits, such as the raised, the slop and the depressed areas associated with surface water accumulation. The minerals were observed under magnifying glass and the mineralogical composition determined on total rock by X-ray diffraction (XRD) using a Phillips X'pert Pro. The samples were also observed in a Jeol JSM 6460 LV scanning electron microscope with an EDAX PW7757/78 X-ray energy-scattering micro-analyzer (SEM-EDS), which was used to determine the qualitative composition of certain minerals.

Based on the surface runoff and drainage from the mining waste deposits areas recognized in the field, three sample sites within the salt marsh were choice (Fig. 1). Site 1 is located in the higher topographic sector of the salt marsh that receives the surface runoff from the mining deposits drainage. Site 2 is located nearest the above site but in a topographically lower sector of the salt marsh. Site 3 is located in the same channel than sites 1 and 2, but in an external sector of the salt marsh with marked tidal influence. Within each site, fifty soil samples (15-cm-diameter and 15-cm-depth) were collected at low tide. The redox potential (Eh) and pH of the soil samples were determined in the field using a portable pH/Eh (ORP) meter and an electrode system Termo/pH meter Altronix TPA-IV. Samples were stored in polyethylene bags, transported to the laboratory, and frozen to -20 °C until analyzed. All soil samples were dried at 80 °C until constant weight and sieved through a 2 mm mesh to remove large stones and dead plant material. Electrical conductivity (EC) was measured with a conductivity meter in a 1:5 soil water extract. Percentages of sand, silt, and clay were estimated using the Pipette method (Day, 1965). Organic matter (OM) was determined by the loss on ignition method (4 h at 450 °C) (Davies, 1974). As well as in the mining waste deposit samples, the mineralogical composition in the soil salt marsh samples was determined by X-ray diffraction (XRD) analysis using a Phillips X'pert Pro.

Thirty samples of *Spartina densiflora* and *Sarcocornia perennis* (15-cm-diameter and 15-cm-depth) were collected within the same sites of the salt marsh (five per plant specie in each site). These samples were carefully washed with tri-distilled water and separated into below-ground tissues (roots and rhizomes) and above-ground tissues (stems and leaves). All samples were dried at 80 °C until a constant weight was reached and pulverized in a mill until the powder was fine enough to pass through a 1 mm sieve.

For the analysis of metals in all sampled matrixes, 1 g of dried and sieved soil or 0.5 g of dried plant material was digested in 2 ml of HNO₃ (Merck) ultrapure using microwave oven MARS-5, (CEM, 2011), and was then diluted to a final volume of 15 ml with nitric acid (EPA, 2000). Copper (Cu), iron (Fe), lead (Pb), and zinc (Zn), in both matrixes were then measured by inductively coupled plasma (ICP-AES) spectroscopy (Shimadzu 9000). In all cases, the average uncertainty of metal ion determination was <2%. All extractions were carried out in duplicate and blanks were processed as the samples. Results are reported on a dry weight. Reagents of analytical grade were used for the blanks and for calibration curves. Quality assurance of soils and plants was done through analysis of

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