



Geochemical processes controlling the distribution and concentration of metals in soils from a Patagonian (Argentina) salt marsh affected by mining residues



Yanina L. Idaszkin^{a,b}, María del Pilar Alvarez^{a,b}, Eleonora Carol^{c,*}

^a Instituto Patagónico para el Estudio de los Ecosistemas Continentales (IPEEC –CONICET), Boulevard Brown 2915, U9120ACD Puerto Madryn, Chubut, Argentina

^b Universidad Nacional de la Patagonia San Juan Bosco, Boulevard Brown 3051, U9120ACD Puerto Madryn, Chubut, Argentina

^c Centro de Investigaciones Geológicas, Consejo Nacional de Investigaciones Científicas y Técnicas - Universidad Nacional de La Plata (CIG – CONICET – UNLP), Diagonal 113 # 275, CP1900 La Plata, Argentina

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ABSTRACT

Heavy metal pollution that affects salt marshes is a major environmental concern due to its toxic nature, persistence, and potential risk to organisms and to human health. Mining waste deposits originated four decades ago, by the metallurgical extraction of heavy metals, are found near to the San Antonio salt marsh in Patagonia. The aim of the work was to determine the geochemical processes that control the distribution and concentration of Cu, Fe, Pb and Zn in the soils of this Patagonian salt marsh. A survey of the mining waste deposits was carried out where three dumps were identified. Samples were collected to determine soil texture, Eh pH, organic matter and metal contents and the soil mineralogical composition. The results shows that the soils developed over the mining waste deposits are predominantly reddish constituted mainly by iron oxide, hydroxide and highly soluble minerals such as Zn and Cu sulphates. The drainage from these deposits tends to move towards the salt marsh. Within the salt marsh, the highest concentrations of Cu, Pb and Zn occur in the sectors closest to the mining wastes deposits. The sulphide oxidation and the dissolution of the Cu, Pb and Zn sulphates could be the mainly source of these metals in the drainage water. The metals in solution that reach the salt marsh, are adsorbed by the organic matter and the fine fraction of the soils. These adsorbed metals are then remobilized by tides in the lower sectors of the marsh by desorption from the cations present in the tidal flow. On the other hand, Fe tends to form non soluble oxides, hydroxides and sulphates which remain as altering material within the mining waste deposit. Finally, the heavy metal pollutants recorded in the San Antonio salt marsh shows that the mining waste deposits that were abandoned four decades ago are still a source metal contamination.

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1. Introduction

Soils play an important role in the mobility of trace metals in coastal systems, acting occasionally as sink and/or source (Du Laing et al., 2009). The presence of metals in coastal and marine environments, caused both by natural processes and anthropogenic activities, is a major environmental concern worldwide due to its toxic nature, persistence, and potential risk to human health and to organisms (Rainbow et al., 2006). Several studies of metal accumulation rates in salt marshes, with different impact levels of human activities, have been carried out in different regions of the world (e.g., Lee and Cundy, 2001; Duarte et al., 2010; Suntornvongsagul et al., 2007; Hung and Chmura, 2007; Sundaramanickam et al., 2016).

Soil pollution associated with heavy metals from mining residues still being an important problem worldwide (Sheoran and Sheoran, 2006). Dumps and foundry slags of polymetallic rich-sulphide deposits are sources of acid drainage in which high concentrations of metals are mobilized (Nash, 2005). The metal load is of greater concern than the acidity in the terms of environmental damage (Kuyucak, 2002). An insidious feature of acid mine drainage (AMD) is that its sources may remain active for decades or even centuries after mine closure (Modis et al., 1998). Both operating and abandoned polymetallic sulphide mining sites are often active sources of metal.

Slags originated four decades ago, by the metallurgical extraction of heavy metals from an ore mainly composed by galena, sphalerite, pyrite and chalcopyrite, are found in deposits near to the San Antonio salt marsh in Patagonia (Fig. 1). The aim of the work was to determine the geochemical processes that control the distribution and concentration of Cu, Fe, Pb and Zn in soils of the above mentioned salt marsh.

* Corresponding author.

E-mail address: eleocarol@fncym.unlp.edu.ar (E. Carol).

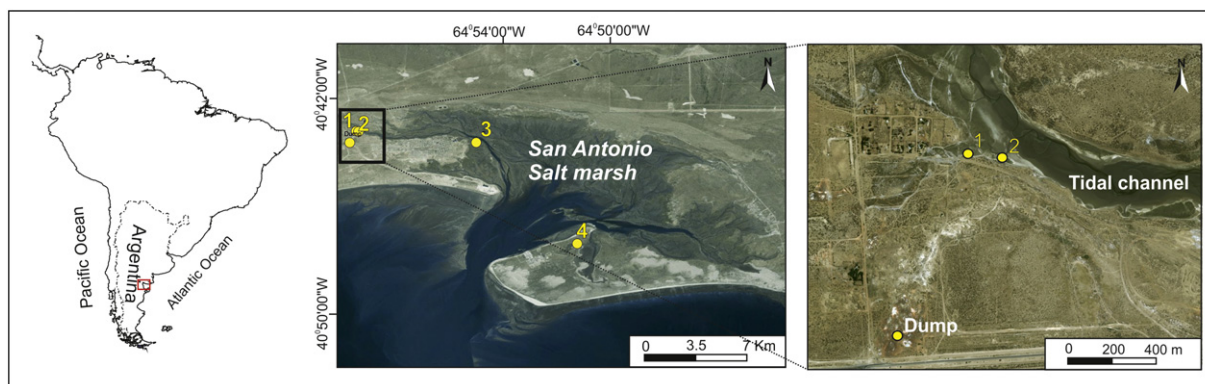


Fig. 1. Location map showing the study area and the sampling sites.

2. Materials and methods

2.1. Study area

The study was carried out in the salt marsh located surrounding the San Antonio Bay ($40^{\circ}44'S$, $54^{\circ}68'W$), in a Natural Protected Area (Río Negro, Argentina; Fig. 1). This is a strictly marine salt marsh with a semi-diurnal macro-tidal regime (tidal amplitude: ~ 9 m) crossed by several tidal channels. The climate is semi-arid, the mean annual precipitation is around 250 mm, and the mean annual air temperature is $15.1^{\circ}C$, fluctuating between mean extremes of $-7.7^{\circ}C$ in July to $41.4^{\circ}C$ 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 vegetation is characterized by *Spartina alterniflora* in the lower marsh zone, and the higher zones are commonly dominated by *Spartina densiflora*, accompanied by the shrubs *Limonium brasiliense*, *Sarcocornia perennis* and *Atriplex* spp. (Bortolus et al., 2009; Idaszkin et al., 2015). The outcropping geology that limits the marsh area, it is constituted by quaternary deposits. These deposits are mostly represented by beach ridges and coastal spits, composed by sands and gravels with mollusc shells. Overlaying them some aeolian landforms constituted by dunes and sandy layers are developed (Angulo et al., 1978).

2.2. Methodology

A survey of the mining waste deposits located in the vicinity of the San Antonio salt marsh was carried out, where samples were collected to measure pH in saturated soil-paste and to determinate the mineralogy. The mineralogical composition was determined by X-ray diffraction analysis (DRX) using a Phillips X'pert Pro. According to the surface runoff and the drainage from the waste dump deposits area recognized in the field, soil samples were extracted at four points within the marsh (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. Finally, site 4 is located in the northern sector of the salt marsh where does not receive drainage from mining deposits, with tidal characteristics similar to site 3.

At each site ten soil samples from the upper 15 cm were collected, always 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 $-20^{\circ}C$ until analyzed. The soil samples were dried at $80^{\circ}C$ 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 after diluting 10 g of dried and sieved soil with 50 ml of distilled water. Organic matter (OM) was determined by the loss on ignition method (4 h at $450^{\circ}C$) (Davies, 1974). Percentage of sand, silt, and clay were estimated using the Pipette method (Day, 1965). For the analysis of metals, 1 g of dried and sieved soil was digested in 2 ml of HNO_3 (Merck) ultrapure using microwave oven MARS-5, CEM Corporation, USA (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 were reported on a dry weight. Reagents of analytical grade were used for the blanks and for calibration curves. Quality assurance of soils was done through analysis of standard reference freshwater sediment CNS392-050. The recovery was 87% for Zn, 89% for Pb, 90% for Cu, and 98% for Fe. Also in these soil sample mineralogical composition was determined by X-ray (DRX) diffraction analysis using a Phillips X'pert Pro were determined.

3. Results

Three mining waste deposits were identified, which showed abundant erosion and drainage features identified mainly in the higher sectors where the gully development dominates (Fig. 2a and b). The drainage from these three deposits tends to move towards the salt marsh. Although locally the surface runoff also accumulates in the lower sectors within these areas. The soils developed over the mining waste deposits are predominantly reddish (Fig. 2b), with acid pH values between 4.0 and 5.7. The DRX mineralogical determinations showed that soils are composed mainly by iron oxides and hydroxides such as hematite (Hm), magnetite (Mt) and iron sulphates of jarosite (Jrs) type. Superficially, the formation of yellowish to whitish crusts, consisting of gypsum (Gy), carminite (Crt), roemerite (Ro), anglesite (Ang), beaverite (Bv), zincosite (Zc), halite (HI) (Fig. 2 e and f) was observed. Locally, blue to greenish aggregates composed of malachite (Mal), chalcantite (Chal), azurite (Az) and linarite (Li) (Fig. 2 g and h) were also observed. Within the residues some ore minerals such as pyrite (Py), sphalerite (Sph), chalcocite (Chlc) and quartz (Qtz) were identified. On the other hand, salt marsh soils are composed mainly by quartz (Qtz), plagioclase (Pl), feldspar (F and KF), dolomite (Dol) and calcite (Cal) (Fig. 3). Within sites 1 and 2 the presence of gypsum (Gy), hematite (Hm) and halite (HI) were identified, being this last one more abundant in site 1 (Fig. 3).

Table 1 resume texture, Eh, pH, EC, OM, and metals content in the soils samples from the four salt marsh sampled sites. In site 1, which is located in the topographically higher part of the salt marsh, nearest to the mining waste deposits, the soil is dominated by fine textures,

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