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Pedogenesis in mine tails affects macroporosity, hydrological properties, and pollutant flow

Sergio Pellegrini [a,*], Gregorio García [b], Jose Matías Peñas-Castejon [b], Nadia Vignozzi [a], Edoardo A.C. Costantini [a]

[a] CREA-ABP, Consiglio per la Ricerca in Agricoltura e l'Analisi dell'Economia Agraria, Centro di Ricerca per l'Agrobiologia e la Pedologia, Piazza D'Azeglio 30, I 50121 Firenze, Italy
[b] Soil Science and Agriculture Chemistry Unit, Agriculture Science and Technology Department, Regional Campus of International Excellence 'Campus Mare Nostrum', Universidad Politecnica de Cartagena, Spain

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ABSTRACT

Mining activities take a direct impact on the environment, particularly when this activity is focused on metal-ore exploitation. Abandoned metal mine areas contain different types of residues from ore-processing operations that are typically characterized by high concentrations of heavy metals. One consequence is the generation of acid-mine drainage, caused by the oxidation and hydrolysis of metal sulphides. Mine tail soils have been studied under several points of view, but not on how crack formation affects their properties and functioning, before irreversible hardening. The Cartagena–La Union mining district is one of the more ancient Spanish mining regions. This region is placed at the south-eastern part of Spain, and contains one of the largest lead and zinc ore deposits in South Europe, which was exploited for Au, Ag, Pb, Zn, Fe and Cu from the Phoenician and Carthaginian times. The aim of this research work was to characterize the pedogenesis that occurred in mine tailings of different age and properties, to highlight the physical and hydrological changes that occurred as a consequence of pedogenesis, and their consequences on the environmental pollution risk. Soil profile morphology was studied in the field likewise the surficial pattern of cracks, shear strength, bulk density, and erosion. Laboratory analyses were conducted to characterize physical, chemical, elemental, mineralogical, and hydrological differences between the soil mass in the macro aggregates and in the fissures. Macroporosity was studied through image analysis of thin sections of undisturbed soil samples. X-ray diffraction (XRD) was performed on the clay fraction. Studied soils resulted strongly polluted, especially as for Zn, Pb, Cd, Cl, Mn, and Ni and were classified as different kinds of Spolic Technosol. Crack formation significantly affected shear strength, bulk density, macroporosity, saturated hydraulic conductivity, and element distribution. Gully and tunnel erosion was also driven by crack formation. The mineralogy of the clay fraction revealed that smectites are present, favouring crack opening. Crack formation deeply affected soil features and behaviour, namely hydrological properties and flow of pollutants, which in turn condition environmental health and possible soil stabilization and reclamation strategies.

1. Introduction

The study of soil formation processes on mine waste deposits is not only of great importance from a scientific standpoint, but also because it may give useful suggestions about the possible monitoring strategies and measures to be introduced to counteract threats to environmental health. Mining waste often contains high concentrations of toxic metals which may contaminate the surrounding environment upon leaching (Martìnez-Frias, 1997; Meza-Figueroa et al., 2009; Mudd, 2007). Subject of recent research includes the effect of natural attenuation processes on the extent of contaminant leaching from mining waste. Natural attenuation may result in a restructuring of a mining waste dump when more stable mineral phases precipitate forming cemented layers or hardpans (Blowes et al., 1991; Gilbert et al., 2003; Grissemann et al., 2007; Kassahun and Rammlmair, 2005; Rammlmair, 2002; Rammlmair et al., 2008; Regenspurg et al., 2005). Many of the above authors have hypothesized that cemented layers act as hydraulic barriers inhibiting transport of pore water, and as diffusion barriers inhibiting transport of pore gases such as O_2 and CO_2 (DeSisto et al., 2011; Graupner et al., 2007; Kohfahl et al., 2010).

Mobilisation of Cd, As, Zn, and Pb from metallic mine tailings is known to be induced by oxidative weathering of metal sulphides (Blowes et al., 2014). Because in mining soils, hardpans and cemented layers may consist of reactive secondary minerals, they are expected to play a crucial role in contaminant attenuation. Cemented, indurated layers in sulphide-bearing mine tailings have been studied for their physical, chemical and mineralogical properties (Blowes and Jambor, 1990; Gilbert et al., 2003; Graupner et al., 2007; Gunsinger et al., 2006; Johnson et al., 2000; Kohfahl et al., 2010; McGregor and Blowes, 2002; Moncur et al., 2005). Field studies on hardpans have reported (Gunsinger et al., 2006; McGregor and Blowes, 2002; Moncur et al.,

* Corresponding author.
E-mail address: sergio.pellegrini@entecra.it (S. Pellegrini).

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2005) that the highest concentrations of originally dissolved metals were observed directly above and within a hardpan layer, thus the hardpans may have restricted the movement of dissolved metals through the tailings and may have acted as a zone of metal accumulation.

Indurated layers are zones at the capillary fringes where agglutination of particles is basically due to processes driven by capillary transport within an O_2 dominated environment (Graupner et al., 2007). Supersaturation results in the precipitation of secondary phases and gels, which may coat particles, agglutinate them, and reduce the porosity. Cemented layers on the other hand, can be observed at the transition between oxidized and reduced zones, which often occurs at the transition between saturated and unsaturated zones. At these transitions, variations in master geochemical variables, such as Eh and pH, occur (Bourg and Loch, 1995).

Despite these cemented layers may play a pivotal role in the natural attenuation of unwanted metals that may originate from sulphide-bearing mine tailings, their formation processes have not been thoroughly studied. In particular, the hydrological role played by the cracks that form along with the indurated soil horizons has not been investigated yet. Actually, it can be hypothesized that the development of cracks negatively counterbalances the natural attenuation processes provided by the firm soil horizons, and enhances contaminant leaching from mining wastes.

The open cast mining of metallic sulphides under the semiarid weather conditions of Southeast Spain created significant areas of degraded land that need remediation (Castillo et al., 2013; Navarro et al., 2008). In the province of Murcia, SE Spain, the presence of about 85 abandoned mining waste ponds poses a potential risk for soil and water pollution, also threatening plants, animals, infrastructures and human health (Martinez Pagàn et al., 2009).

In this paper, a multi-disciplinary approach based on physical, micromorphological, geochemical, and mineralogical methods were applied to study the surface indurated horizons and related cracks of mining soils under semiarid climate conditions. The aim of this research work was to characterize the pedogenesis that occurred in mine tailings of different age and properties, and to highlight the physical and hydrological changes that occurred as a consequence of pedogenesis.

2. Materials and methods

2.1. The study area

The Cartagena–La Union Mining District (0–392 m above sea level; 37°37′20″ N, 0°50′55″ W – 37°40′03″ N, 0°48′12″ W) is placed at the most south-eastern part of Spain (Cartagena, Murcia) and its area is approximately 50 km^2 (Garcia, 2004). The region contained one of the largest Pb–Zn accumulations in the Iberian Peninsula. This zone was one of the most important mining areas in Spain in the last centuries. This region has been exploited for Au, Ag, Pb, Zn, Fe and Cu from the Phoenician and Carthaginian times (Orejas and Antolinos, 2001) until 1991. During these 2500 years, the area suffered important environmental stresses such as the sedimentation of millions of tons of post-flotation wastes (sludge) in Portman Bay and Mar Menor coastal lagoon. Metal contamination in this area has been recently confirmed by some authors (Garcia, 2004; García et al., 2008; Gómez Ros et al., 2013; Robles-Arenas et al., 2006).

The main geochemical process recognised is sulphide-mineral oxidation and later-generated sulphate dissolution by groundwater and runoff. Runoff and wind are the major mechanisms of metals and sulphate transport in the study area and adjacent zones (Moreno Brotons et al., 2009; Robles-Arenas et al., 2006). According to the concentration of sulphate and heavy metals in sediment, soil, rainwater, surface water and groundwater samples, it is possible to conclude that the impact of mine activities occurs not only in the immediate mining area (100 km^2), but also in the surrounding areas, for a total surface of 1000 km^2 approximately (Robles-Arenas et al., 2006). Contamination affects local creeks and costal lagoons (Marín-Guirao et al., 2007), but also groundwater (Robles-Arenas and Candela, 2010).

The climate in Sierra de Cartagena–La Union is typically semiarid Mediterranean, characterized by an annual mean precipitation of approximately 300 mm, with a range between 250 and 350 mm year^{-1}, distributed in a few intensive rainfall events concentrated during spring and autumn. The area shows a temperature range between 5.4 (January) and 40 °C (August). The annual average temperature is 17 °C. Wind is always present in the study area, mainly the "Levante" (a wet wind with an E-W trend). The lowest average wind speed is registered in autumn (15.6 km h^{-1}) and the fastest average in spring (22.4 km h^{-1}). The potential evapotranspiration is approximately 900 mm year^{-1} (Garcia, 2004). The natural vegetation is scarce and mainly based on pioneer species of shrubs and thicket plants with xerophytic characteristics, with some of these species growing on mining waste deposits (Arocena et al., 2010).

The "Sierra de Cartagena" mountains are part of the Betic Mountains that arose in the Miocene epoch. At the end of the Miocene, a great post-orogenic magmatic activity happened, with an associated hydrothermal activity, which produced important polymetallic ore deposits (Manteca and Ovejero, 1992). Mineral paragenesis has formed greenalite, chlorite, magnetite, pyrite, marcasite, sphalerite, galena, siderite, mixed carbonates of Fe, Zn, Mn, and quartz, and as accessory minerals pirrotine, arsenopyrite, chalcopyrite, tetrahydrite, stannine, etc. In the oxidation zones, or gossan, the main minerals are goethite, hematite, jarosite, alunite, smithsonite, anglesite, cerusite, gypsum, jasp, etc. Locally, some Mn minerals can be abundant, which is the case of pyrolusite, romanechite, psilomelane, etc., or Ba minerals, such as barite (Manteca and Ovejero, 1992).

From the point of view of mining, the area shows the existence of 12 open-pits, 1902 mining wells, 2351 waste deposits, including 89 tailing dams and waste rock derived from mining processes (Garcia, 2004; Robles-Arenas et al., 2006). Mine wastes occupy an area of 9 km^2 and have an approximate volume of 200 Mm^3 (Garcia, 2004). Mineralogical, physical and chemical data distinguish nine different types of mine and metallurgical waste (Garcia, 2004). Since 1940, the mineral separation process was performed by using differential flotation and various reagents, while prior to it by gravimetric techniques. The mining wastes, or tailings, with a very fine size of particle were deposited inland (tailings dams) and, since the 1950 decade, huge releases were made on the sea coast, Mediterranean and Mar Menor.

Due to the climate and the nature of parent material, soil development is scarce in the area. Soil types that are often found in the territory are Haplic Calcisols, Petric Calcisols, Calcaric and Haplic Cambisols, Gipsiric and Calcaric Regosols, and some types of Leptosols. In many cultivated areas they have been replaced by Terric and Hortic Anthrosols, Spolic Anthric Regosols, etc. (Hernández et al., 2005). However, on the surface of mining waste deposits, only incipient processes of soil formation are occurring, because of the limited biological activity and vegetation.

Within the study area, 5 mine tails of similar age and mineralogy, but different geomorphological position, were sampled. The sampling point distribution is shown in Fig. 1.

2.1.1. History of the sites

2.1.1.1. La Esperanza (site 1). Geographically, the deposit of waste sludge from mining called "La Esperanza" is located in the Southwest Sector of the Sierra Minera de Cartagena–La Unión (Murcia, Spain) and has been built as a mining pond. Its construction dates from the mid-50's, with the aim of treating the material from mines that exploited the minerals of galena and sphalerite. Since its closure to the present, the mining pond has not undergone any restoration work, being very strongly affected by the action of erosive agents, in addition to having undergone a process of collapse in the 60's. The UTM coordinates of the sampling