Metal pollution in hydrographic networks of abandoned mining basins: The case of Linares Mining District (SE Spain)

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Abstract

The extractive mining activity and the associated concentration and smelting industries produce different types of solid waste that are accumulated in dumps, flotation tailings dams and slag heaps. These mining waste have high contents of metals and semi-metals, which can cause an environmental risk if they reach the ground and drainage networks. Aware of this risk, administrations have been addressing the problem in recent years, in such a way that many of these tailing dams are being restored and sealed.
In this field trip, we will focus on the area known as Adaro, within the Linares mining district. A review of the geology of the district and types of deposits, the mining techniques used and the concentration processes will be carried out. Based on this knowledge, the hydrogeochemical works carried out for the characterization of surface and groundwater in the studied sector are indicated, analyzing the condition they have evolved since the mining activity ended. The geophysical techniques used for the characterization of mining dams are also described and the effectiveness of the sealing carried out in some of these restored dams is evaluated.

Resumen

La actividad extractiva minera y las industrias de concentración y fundición asociadas producen diferentes tipos de residuos sólidos que se acumulan en escombreras y presas de flotación y lavado. Estos residuos mineros tienen altos contenidos de metales y semimetales, lo que puede ocasionar un riesgo ambiental si llegan al suelo y a las redes de drenaje. Conscientes de este riesgo, las administraciones han venido abordando el problema en los últimos años, de tal forma que muchas de estas presas están siendo restauradas y selladas.
En esta salida de campo nos centraremos en la zona conocida como Adaro, dentro del distrito minero de Linares. Se realizará una revisión de la geología del distrito y los tipos de yacimientos, las técnicas mineras utilizadas y los procesos de concentración. Se muestran los trabajos hidrogeoquímicos realizados para la caracterización de las aguas superficiales y subterráneas en el sector de estudio, analizando su evolución desde que finalizó la actividad minera. También se describen las técnicas geofísicas utilizadas para la caracterización de presas mineras y se evalúa la eficacia del sellado realizado en algunas de estas presas restauradas.

Key-words: Linares, Mining activity, Waste mining materials, Metal pollution, Solis, Water.

1. Introduction

The metallogenic district of Linares-La Carolina (SE Spain, province of Jaén) is characterized by the presence of phyllonian deposits with sulfide metallizations, basically galena (PbS) in Paleozoic granites (Azcárate et al, 1977; Lillo 1992a). These mining waste have high contents of metals and semi-metals, which can origin an environmental risk if they reach the drainage networks This extractive activity yield an important accumulation of mining waste material that was deposited in the proximity of the exploitation work. On the other hand, in mineral concentration processes waste was also produced, specifically, the rejection generated in the flotation process was deposited in dumps, flotation tailings dams and slag heaps. In these areas is possible to find contents of metal sulfides, which behave unstable under the oxidizing conditions, which can give negative effects on the environment. In addition, in cases where there is generation of leachates, these have high contents of sulfates and metallic elements, as has been described both in this sector (Hidalgo et al., 2006; 2010; Cortada et al, 2019).

2. Description of the study area

In the mining district of Linares, hydrothermal vein deposits are hosted in a Palaeozoic basement and fossilized by a Triassic sedimentary cover. Bedrock is composed of metamorphic rocks (mainly phyllites with alternating quartzites), intensely deformed during the Hercynian orogeny and affected by a granitic intrusion. This Palaeozoic basement is severely fractured and characterized by the presence of a dense dyke network associated with the granitoid massif (Azcárate, 1977; Lillo, 1992). The exploited ore consists of Pb-Ag and Cu-Fe sulfides. Dominant ore minerals are galena, sphalerite, iron sulfides (pyrrhotite, pyrite, marcasite) and chalcopyrite. Gangue minerals are ankerite, quartz, calcite, amorphous silica, barite, and minor amounts of kaolinite.
For this activity, the waste deposits of an old flotation plant (Adaro washery, Fig. 1), in the mining district of Linares, were chosen. There are two tailing ponds, one located along the left bank of the Guadiel River (restored in 2011) and another (unrestored) on the right bank (Fig. 2). The aim of this filed trip is to visit these dams and showing the characterization of these waste materials and the effectiveness of the sealing techniques employed.

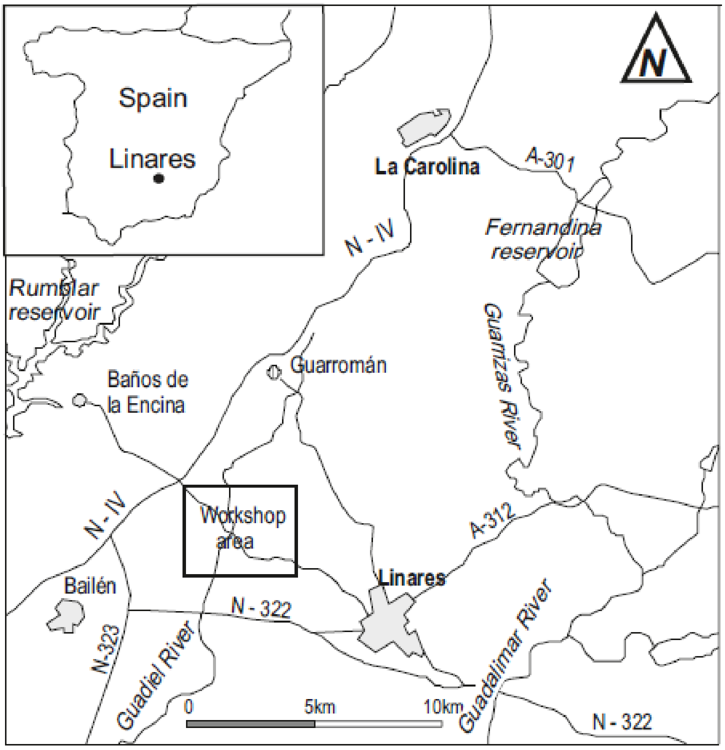

Fig.1. *Location of the field trip in the area of Linares.*

The mineralogy of the tailings (De la Torre et al., 2012) was studied using XRD analysis. The results of the semi-quantitative analysis indicate that these tailings are mainly composed of quartz, which is very abundant (30–35%), accompanied by phyllosilicates (25–30%), and feldspar, calcite and ankerite, classified as abundant (10–15%) in the tailings samples. Galena, cerussite and lepidocrocite also appear, but only as trace minerals (< 5%). The total contents of metal(loid)s were also analyzed in surface and bottom (Rojas et al., 2012) showing that concentrations were also quite similar in surface and bottom samples, and significant values were found for Fe (25,600–26,900 mg/kg,), Pb (3400–1200 mg/kg), Zn (150–100 mg/kg), and As (35–38 mg/kg).

3. Hydrochemistry

Based on the work carried out by the RNM-374 research group and the results obtained in Cortada et al. (2017), in a hydrogeochemical study was made in this area in order to detect water pollution. Table 1 show the physicochemical characteristics of surface and groundwater in the environment surrounding the studied dams. The maximum annual mean values (AA) are also included, in accordance with the environmental quality standards of the European Union for priority toxic elements in surface water. In the case of the Guadiel River, there are analytical data obtained from periodic sampling carried out for more than a decade at point R1 (Fig. 2). The first column of Table 2 shows the mean values obtained for water sampled before 2011, the time when the restoration of the main Adaro dam took place. At that time, the river had a calcium bicarbonate-sulfate facies, which reflected the effects of the oxidation of the sulfides present in the mining wastes. In the same way, the electrical conductivity values (968 µS/cm on average) indicated an increase in the mineralization of the surface waters near the mining area, with notable sulfate contents

(130 mg/L on average) and some trace elements. This is the case of iron (mean value of 0.5 mg/L), manganese (mean value of 0.6 mg/L) and lead (38 µg/L), which, downstream of the dams, exceeded the maximum limits established by environmental laws. During that period, the concentrations of these elements reached particularly alarming maximum values around the Adaro flotation tailing dams (1.3 mg/L Fe, 1.9 mg/L Mn and 0.2 mg/L Pb).

These hydrochemical controls at R1 continued at four-month intervals between 2012 and 2014, after the dam restoration. The average value of the electrical conductivity of the water was 735 µS/cm, which is significantly lower than the value recorded before carrying out the restoration work, also observing an increase in the pH of the water to values above 8, typical of alkaline conditions. In addition, the average sulfate content has been reduced by almost half. A decrease in the mean value of the dissolved concentrations of all the contents of metallic and semi-metallic elements, except As, can be observed (Table 1). Despite this apparent improvement in the chemical quality of the Guadiel river waters, it should be noted that the average Se and Pb contents are still above the annual average values (AA) set by the European Union for surface waters. Furthermore, Fe, Mn and Zn have reached high maximum values in some samples (0.7 mg/L, 0.6 mg/L and 0.6 mg/L, respectively). These results suggest that, despite sealing the landfill, during certain times of the year leachate leaks are generated that can be incorporated into the river, both from the environment surrounding the restored dam and from the unrestored pond on the right bank.

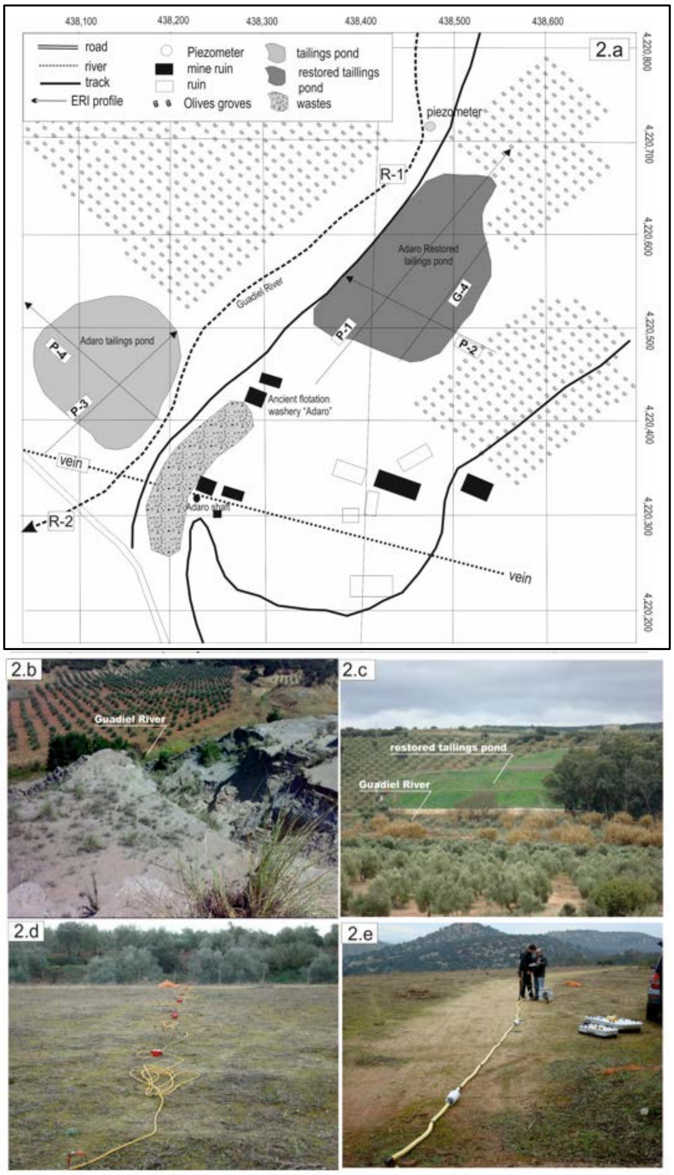

Fig.2. *Location of the studied tailings ponds with the positions of the piezometer, ERI and GPR profiles (P.1, P.2, P.3, P.4 and G4). R-1 and R-2: surface water sampling points in the Guadiel River (2.a). Tailings pond before restoration affected by a landslide (2.b). Restored tailings pond (2.c). Electrical resistivity imaging profile 2 (2.d). Ground penetrating radar equipment used in this study: 30-MHz Rough Terrain Antennas (RTA) and 100 and 250 MHz screened antennas (2.e) (Cortada et al. 2017).*

To analyze the role that abandoned dams and ponds currently play in the chemical quality of the waters of the Guadiel River in the Adaro environment, a new sampling campaign was carried out in the spring of 2016 (Table 2) that included the river in R1 next to the restored dam (Adaro) and a second sampling point downstream of a second restored pond (R2) (La Carlota). The results obtained were compared with the quality of the groundwater from the old Adaro mining well (Well B), flooded after the closure of the mine, and with the waters of a piezometer with a depth of 90 m, located between the restored dam and the riverbed. In the mine water, calcium bicarbonate-sulfate hydrofacies were found with a conductivity greater than 900 μS/cm, a low content of dissolved oxygen (4 mg/l) and high concentrations of Fe (0.3 mg/l), Pb (0.05 mg/l), Sr (0.5 mg/l) and Zn (0.2 mg/l).

Table 1: *Physical-chemical characteristics of surface and ground waters. Sample locations shown in Fig. 2. Maximum allowable annual average (AA) values for selected elements are included. Contents exceeding the AAs are in bold.*

Parameters and metal(oid)s	**Guadiel River - R1**		**Guadiel River**		**Spring 2016**	
	before 2011	**2012-2014**	**R1**	**R2**	**Piezometer**	**Adaro shaft**
Electrical Cond. (μS/cm)	968	735	690	712	645	932
Temperature (ºC)	18,1	19	10,5	10,2	19,0	18,6
pH	7,6	8,3	8,2	8,4	6,5	7,2
O_2 (mg/L)	8,7	11,5	8,9	10,5	4,3	4,0
Major constituents (mg/L)						
Ca^{2+}	96	78	66	71	66	102
Mg^{2+}	32	24	27	21	17	28
Na^+	58	42	34	40	34	45
K^+	6	5,3	2	6	3	3
Cl^-	67	48	53	55	32	45
SO_4^{2-}	133	69	65	81	36	126
HCO_3^-	345	272	287	269	236	337
Trace elements (μg/L)						
Al	172	31	46	32	27	29
As (50 μg/l AA)	8	14	26	13	30	13
Ba	256	169	226	202	204	137
Co	1,0	0,6	1,5	0,9	0,5	0,4
Cr (50 μg/L AA)	2,6	0,7	1,5	0,5	0,7	10
Cu (40 μg/L AA)	20	3,5	7	6	10	15
Fe	475	395	353	275	219	324
Ga	10	37	18	8	11	7
Mn	632	339	390	482	20	21
Ni (20 μg/L AA)	**31**	3,2	5,0	2,8	1,7	3,5
Pb (7.2 μg/l AA)	**38**	**23**	**20**	**10**	**16**	**47**
Rb	4	2	2	2	8	8
Se (1 μg/l AA)	**1,4**	**1,2**	**1,5**	**1,6**	**1,4**	**2,6**
Sr	428	256	290	243	288	460
Zn (300 μg/l AA)	218	161	201	147	148	198

The increase in sulfate content in this hydrogeological context must be linked to an oxidation process of the metal sulfides present in the old mining galleries. The existence of an acid drainage, as could be inferred from this process, is not detected at any point in this area, probably due to the presence of carbonate gangues (ankerite) that balance the pH (Hidalgo et al., 2006). On the other hand, the water collected in the river piezometer is of the calcium bicarbonate type and has the lowest electrical conductivity (645 µS/cm) and pH (6.5) measured in the study area. Its chemical composition is representative of a subsurface flow, which, in periods of high water, can reach the river bed in the vicinity of the dam. The metal(oid) concentrations analyzed in this water sample are, in almost all cases, lower than those detected in the mine water, as expected, with the exception of As and Ba.

For the waters of the Guadiel River, it should be noted that in this last sampling campaign a calcium bicarbonate facies and a moderate conductivity (690-712 µS/cm in R1 and R2) are maintained, which are very similar to the average values of the 2012 period. 2012-2014. Despite this, the concentrations of dissolved metals are higher than those obtained for the piezometer water in most cases, and for Fe, Mn and Ba, they even exceed those of the mine water analyzed on the same date. A slight increase in the degree of mineralization of the water is recorded between R1 and R2, associated with an increase in the sulfate content. Additionally, in this section of the river, a significant increase in manganese content is observed, reaching 0.5 mg/l, a particularly high value when compared to the concentrations measured in groundwater samples (0.02 mg/l, both in the mine water and in the piezometer water). Therefore, it is reasonable to assume that the origin of the Mn content does not come from underground or subsurface flows towards the river bed.

4. Characterization of dams by geophysical techniques

4.1. Electrical Resistivity Imaging (ERI) and Induced Polarization (IP)

This geophysical prospecting technique is based on determining the distribution of a physical parameter in the subsoil, based on a very high number of measurements made from the ground surface. In this case, the difficulty (resistivity) offered by a material to the passage of electric current through it is measured (Telford et al., 1990; Store et al., 2000). The different geoelectric behavior allows obtaining 2D profiles, turning out to be a very effective non-destructive tool for the study and characterization of subsurface discontinuities (Sasaki, 1992; Store et al., 2000). In this investigation, profiles have been executed with spacing between electrodes of 5 m in the restored dam and 3 m in the non-restored one (Fig. 2 a and d). The electrical tomography equipment used is the RESECS model, Deutsche Montan Technologie (DMT).

In total, 4 electrical tomography profiles have been made with 64 electrodes (Cortada et al., 2017) located in Figure 2. Two of them have been carried out in the restored pond (P-1 and P-2 in Fig. 2) with a length of 320 m each, and another two in the unrestored pond (P-3 and P-4 in Fig. 2), of 192 m per profile. The interpretations of the electrical tomography profiles have been made from the apparent resistivities obtained in the field work, treated by means of the specific resistivity and induced polarization software RES2DINV (Griffiths and Barker, 1993; Loke and Barker, 1996; Loke and Dahlin, 2002). Figure 3 shows the two profiles made on the restored dam, both with apparent resistivity values (A and B in Fig. 3) and real resistivities (C and D in Fig. 3).

The profiles have been designed to cover the entire dam, in order to characterize the residue and the contact with the substrate. Profile 1, SW-NE direction, was made in one of the berms of the dam. From the resistivity values, two large sets can be differentiated (C in Fig. 3): a superficial one, with low values (less than 150 Ω·m) that is associated with the sludge dam, and another set, with high values. increasing resistivity (which can exceed 2000 Ω·m) associated with granite. Within the granite assemblage, in the highest part, the lowest resistivity values appear, which is associated with the high degree of alteration that this rock presents on the surface. At depth, the resistivity values increase associated with a progressively healthier granite.

In the longitudinal profile of the structure (D in Fig. 3), as in the previous case, two sets could be considered: the granite and the mining deposits. A very irregular topographical base is detected, on which the sludge was deposited, filling two depressions, with changes in power in the fillings that range between 5 and 7 meters in the crest area up to 35 m in the middle part of the slope. In the fill, a surface level with somewhat higher resistivity values is observed in a discontinuous way, especially in the berm areas and in the lower part of the slope, which suggests areas in which the waterproofing has not been as effective. Bearing in mind that the granulometry variations in this dam are not very significant, ranging between silt and fine sand, the resistivity variations observed in this fill (between 30 and 150 Ω·m) are related to changes in humidity and not with lithological changes, as occurs in other dams in the sector (Martínez et al. 2014; 2016; 2021). On the other hand, the granitic basement is characterized by presenting high resistivities, with important and abrupt decreases in the values (up to 50 Ω·m) that are difficult to explain. These could be attributed either to alteration zones associated with fractures or even to old mining operations currently filled with mud and water. Therefore, in profile 1, a higher level with a lower degree of humidity could be considered, which could be related to the restoration and waterproofing work carried out. However, this level of medium resistivity (between 100-150 Ω·m) is not continuous, being lost at both ends of the mining dam, in contact with the granite. From this it can be deduced that the waterproofing process was not completely achieved in the perimeter area of the pool, in contact with the substrate. For this reason, this perimeter zone continues to be a recharge zone for leachates.

Figure 4 shows the two profiles (P3 and P4) made in the unrestored dam (A and B, apparent resistivity values, C and D real resistivities).

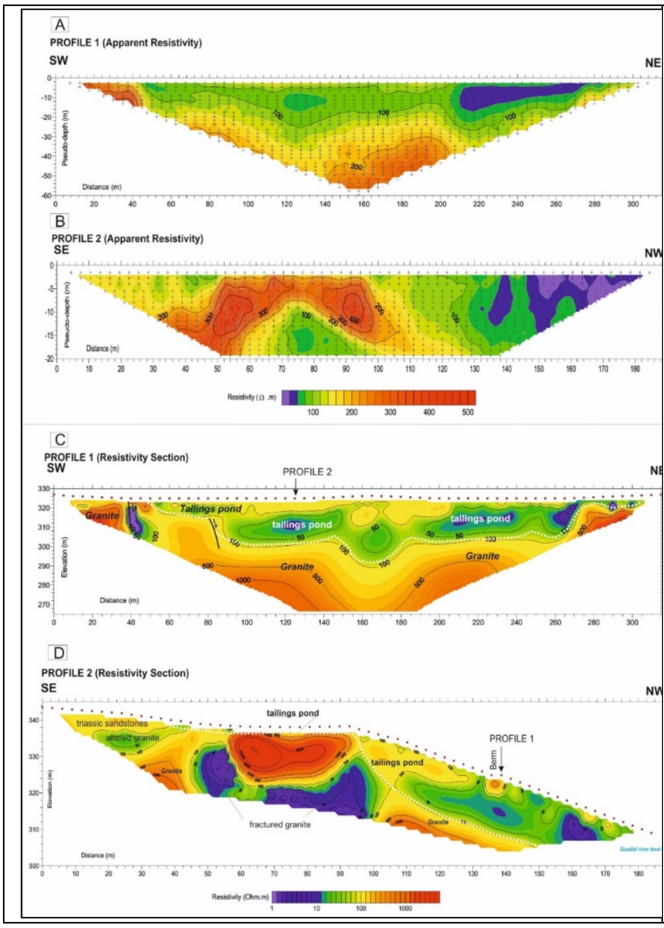

Fig.3. *Electrical resistivity imaging profiles (profiles 1–2). The position of each profile is shown in Fig. 2. (Cortada et al., 2017).*

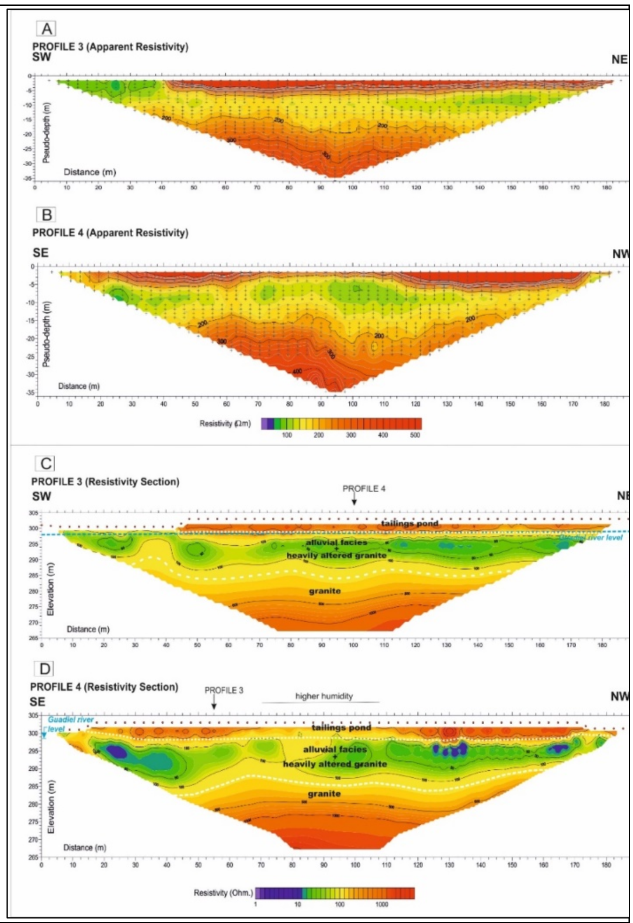

Fig.4. *Electrical resistivity imaging profiles (profiles 3–4). The position of each profile is shown in Fig. 2. (Cortada et al., 2017).*

Profile 3, SW-NE direction, was made on the mud pond, parallel to the Guadiel River. In it, from the resistivity values, three large sets can be differentiated (C in Fig. 4), one superficial, of about 2-3 m of power, with medium-high values (between 500-1500 Ω·m), which is associated with the waste from the sludge dam. The intermediate set, which outcrops in the vicinity of the Guadiel River, offers low resistivity values (between 50 and 100 Ω·m) and is related to the shales associated with the old floodplains of the river. Finally, in the deepest position, granite appears, first very altered (with resistivity values less than 500 Ω·m and healthy in depth (with values that can exceed 2000 Ω·m). Profile 4 is perpendicular to the previous one and passes right through the center of the old structure (Fig. 2). On this occasion, the presence of the three levels (granite, fluvial shale and mud dam) can also be deduced. However, a difference is noted compared to the previous profile: in the central part of the sludge dam since the resistivity values drop considerably to values of 150 Ω·m (Fig. 4, D). This geoelectric behavior could be associated with the model of construction and growth of the pond since the system discharge was through a perimeter gutter and the drainage system was centered, with a slightly conical structure morphology. Therefore, the central part of the structure will retain the highest humidity, which translates into the lowest resistivities.

Other works carried out later, in which Electrical Resistivity and Induced Polarization have been measured, at the foot of the restored dam next to the Guadiel River (Rey et al, 2020) (Fig. 5 and 6), reveal the presence of possible leachate that would be taking place at the foot of the structure, as can be seen in figure 6D, where we assimilate the chargeability values between 5 and 8 mv/V measured superficially to these leachates.

In Fig. 6, it is represented: A) real resistivity in a profile 395 meters long (80 electrodes every 5 meters). B) real resistivity of a profile 315 meters long (64 non-polarizable electrodes every 5 meters). C) Chargeability obtained in the Induced Polarization profile (315 meters).

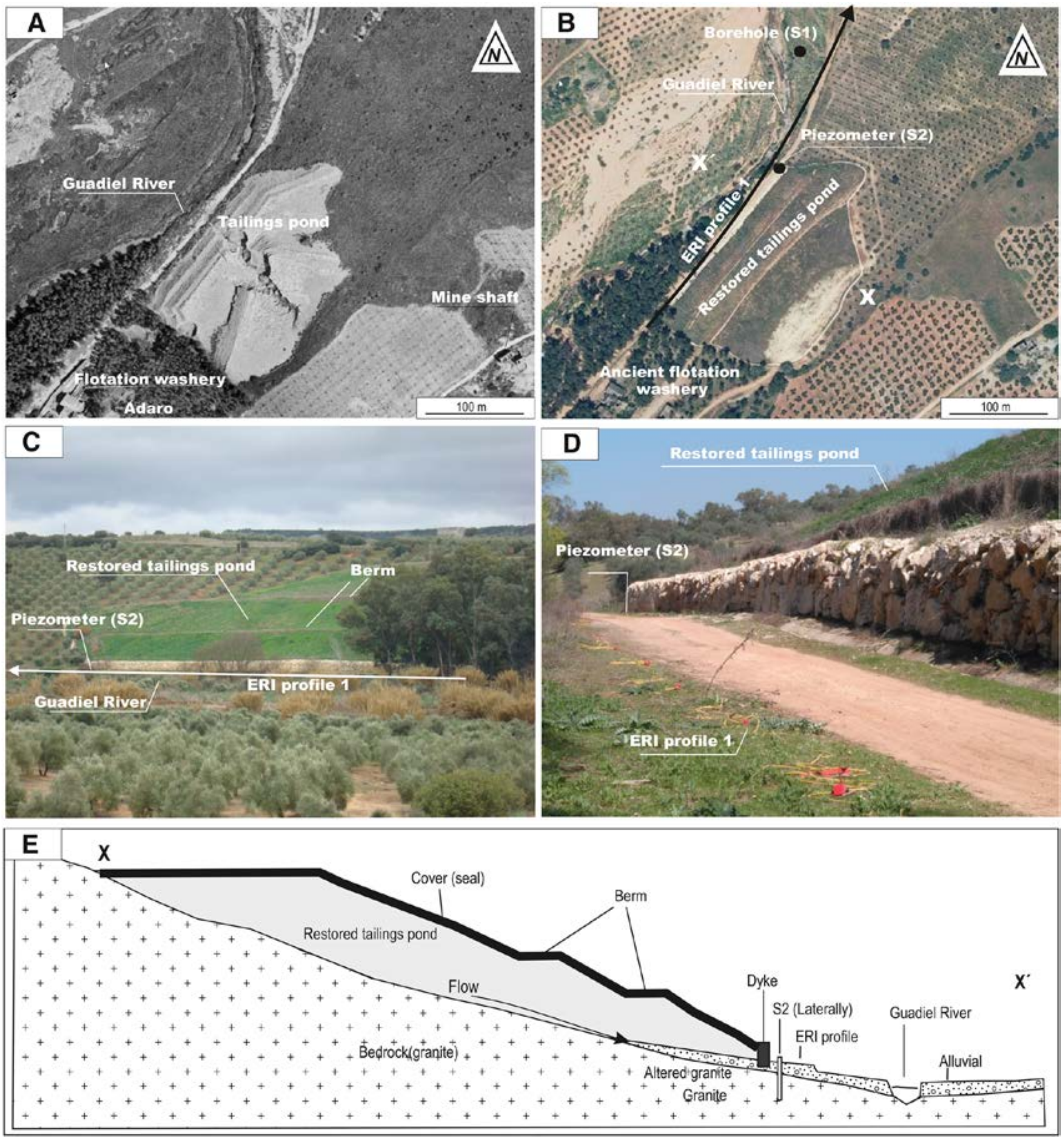

Fig.5. *Position of the Resistivity and Induced Polarization profile made at the foot of the Adaro dam. A) dam before restoring. B and C) restored dam and profile position. D) Layout of the wiring at the foot of the dam. E) cross-sectional profile of the dam and situation with respect to the Guadiel river (Rey et al, 2020).*

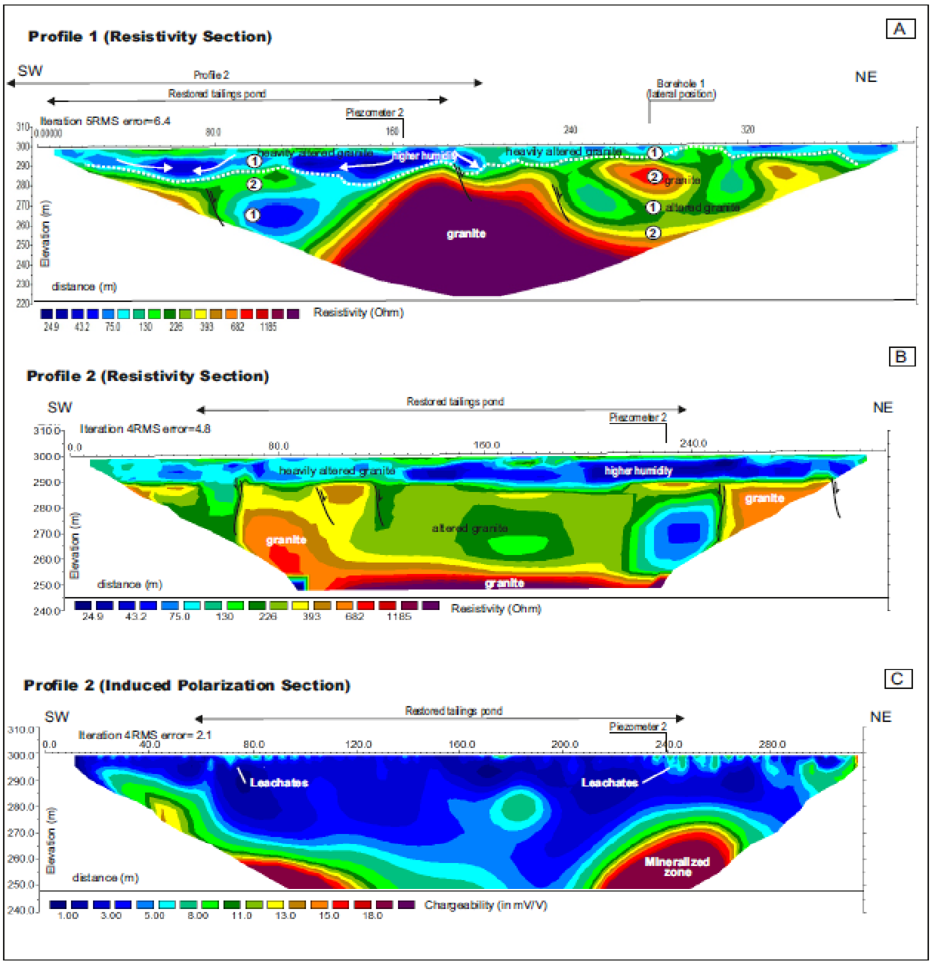

Fig.6. *A) Real resistivity of the 395 m profile. B) Real resistivity of the 315 m profile. C) Chargeability of the Induced Polarization profile (315 meters) made at the base of the Adaro dam (Rey et al, 2020).*

5. Conclusions

The hydrochemical study carried out on the surface and groundwater of the Adaro area allows the identification of high contents of sulfates, carbonates and metals(oids) dissolved in the water, with a signal of sulfated hydrofacies. It is significative the high concentrations of Fe, Mn, Pb, Zn and Se. These elements are associated with the oxidation process of metallic sulfides of the mining dams, so they could be related to local processes of generation of leachates from these waste deposits. The comparison of the hydrochemical studies carried out in the sector before and after sealing can also inform us about the effectiveness of the sealing and, therefore, the elimination of contamination vectors. In this way, although the hydrochemical parameters have improved substantially after encapsulation, high metal(oid) values continue to be measured, which are associated with leaks from the sludge deposit associated with local leachate infiltration processes from these residues towards the substrate. all this facilitated by the intense fracturing of the granite plinth. Geophysical prospecting methods, Electrical Resistivity Imaging (ERI), were used in the characterization of mining dams. In this way, the ERI technique allows reconstructing the morphology of these mining structures, it is possible to identify the contact of the residue with the substrate and detect the areas of greatest humidity. Thus, areas with higher humidity could be related to failures in the insulation of the structure during the sealing stage. The possible leakage of leachate at the foot of the structure was also made clear by the results obtained in the Induced Polarization profile carried out.

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