

**Geomorphic-based mine rehabilitation coupled with AMD chemical stabilization in sulphide-rich ore deposits and soils: Insights from a pioneering intervention at the Lousal mine, Iberian Pyrite Belt**

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# **Geomorphic-based mine rehabilitation coupled with AMD chemical stabilization in sulphide-rich ore deposits and soils: Insights from a pioneering intervention at the Lousal mine, Iberian Pyrite Belt**

The Lousal mine rehabilitation project proactively planned strategic milestones around key local geomorphic landforms and geochemical characteristics. GeoFluv™ method was used to design a rehabilitation topography mimicking natural landforms, on which a built-up soil cover with chemical buffering capabilities and open limestone channels were implemented across the intervention areas. Once these landform restoration works were completed, positive effects are observed in March 2022, with the native vegetation cover (the third main component of the restoration plan) established in most of the recovered area and a visible water quality improvement to the AMD treatment system water. Potentially Toxic Elements (PTEs) concentrations have reduced significantly after the reclamation actions, especially Fe (404 to 34 mg L<sup>-1</sup>), Zn (65.7 to 15.8 mg L<sup>-1</sup>) and Cd (122 to 0.8 µg L<sup>-1</sup>) concentrations. Minor problems detected have been processes of rill erosion associated with the inflow of adjacent watersheds, and the creation of ephemeral reducing conditions resulting from the leaching of the organic amendment of the topsoil. The Lousal mine reclamation project is the first built example in Europe where the concept of geomorphic reclamation has been combined with geochemical restoration in an area with intense acid mine drainage formation. Detailed monitoring of this project's restorative progression throughout 2024 and beyond, should offer learning opportunities and innovations which will benefit future rehabilitation projects, with comparable underlying features.

Keywords: mine rehabilitation; geomorphic rehabilitation; geochemical remediation; metal mining; acid mine drainage.

## 1. Introduction

As an essential communal activity, mining provides those fundamental materials and elements which support contemporary human life and its development. Whereas it impacts less than 0.5% of the earth's surface [1], critical environmental impacts associated with mining activities include:

- Generation of extensive volumes of solid and/or liquid waste
- Variably pronounced terrain transformation (on-site effects).
- Waste and contaminant emission / dispersion (off-site effects) [2].

In developed countries with wide-ranging mining infrastructure and histories, environmental legislation acknowledges the need for rehabilitation, including physical and chemical stabilization of individual mine sites. In Australia, there is a requirement to construct post-mining topographies which are safe, stable and non-polluting for humans and wildlife, and capable of sustaining land use after mining is completed [3]. Mining legislation in Canada requires the reconstruction of 'self-sustained' landforms and ecosystems that are compatible with a healthy environment and the maintenance of other human activities [4]. In the United States, a reclaimed mine area must be stable against erosion, thereby minimizing off-site effects and avoiding potential changes in the hydrological balance [5], and seamlessly integrate with and complement the drainage network of the surrounding land [6]. In Spain, RD 975/2009 [7] legislation requires guaranteeing the stability of mining waste, while preventing soil, surface and groundwater contamination. Finally, Chile's legislation requires guaranteeing the physical and chemical stability of all sites where mining activities have taken place [8].

To comply with these regulations, mining companies need to design closure plans which include rehabilitation of disturbed land. Conventional approaches in mine

rehabilitation such as outslope/berm systems, rectilinear slopes, and drainage ditches, aim to maximize material storage with a minimum footprint, convey water away from the disturbed areas, and maintain geotechnical stability of unconsolidated spoils [9, 10]. However, studies of mine waste erosion demonstrate that conventional rehabilitation landform design and practices do not guarantee long-term waste stability, nor are they necessarily self-sustaining [11, 12]. Although geotechnically stable, rehabilitation landforms are rarely planned employing geomorphic principles [13]. The consequences are that, without constant maintenance, these landforms gradually develop severe water erosion problems, which often pose into costly environmental hazards [14].

Consequently, there is a growing need for innovation in mining rehabilitation and remediation, to minimise environmental and ecological impacts, from both physical (accelerated erosion, topographic modification, contribution of sediments to river courses), and chemical perspectives (increase in water acidity, contribution of metals and metalloids, sulphates). Increasingly, the extractive industries will be required to abandon mining management and rehabilitation techniques which are incapable of sustaining ecosystems in degraded areas, by adopting those based on ecological and geomorphic principles.

At present, the technical understanding exists to design and implement mine rehabilitation solutions which respect those principles. Over the past decade, many such geomorphic rehabilitation approaches have been implemented in US, Australia, Canada and Spain [14 – 20]. These solutions employed fluvial-geomorphic design principles, capable of building ecologically focused catchments and rehabilitation topographies. Their morphologies are comparable to natural and mature landscapes resulting from thousands of years of active topographical development.

This paper describes the design and construction process of the Lousal mine (Portugal) rehabilitation project, within the LIFE funding program framework of the European Union (LIFE RIBERMINE; LIFE18 ENV/ES/000181). This novel mine rehabilitation project jointly applies geomorphic rehabilitation solutions with soil reconstruction techniques and chemical neutralization of Acid Mine Drainage (AMD), to finally revegetate the area with autochthonous species. As the first example of a combined approach to metal mining rehabilitation, the information and expertise provided by the implementation of these methods will undoubtedly offer significant innovations for future mine rehabilitation projects, particularly for those operations with comparable features.

## **2. Regional setting and characteristics of the mining area**

### ***2.1. Physical environment and climate***

The Lousal polymetallic massive sulphide mine is located in the Grândola municipality (Setúbal District, Portugal), in the northwest sector of the Iberian Pyrite Belt (Figure 1), a volcanogenic massive sulphide province that extends from Seville to the Portuguese western coast [21 – 23]. The mining area is bordered to the north by the Espinhaço do Cão stream and to the south by the Corona stream, both tributaries of the Sado River [24]. Regional geomorphology is influenced, at the northwest, by the presence of the Palaeozoic basement rocks of the South Portuguese Zone, and to the southeast, by Tertiary sediments of the Alto Sado Basin [25 – 28].

This region is characterized by a Mediterranean climate, with moderately dry and cold winters, with warm and dry summers. Average annual temperatures of 16.5°C and rainfall of 310 mm are recorded. Regionally, soil profiles are characterized by Leptosols [29], with pH values ranging from 5 - 5.5 and low organic matter concentrations. Lousal

mine site soils are classified as Technosols [29], composed of soil, rock fragments, and tailings material [30, 31].

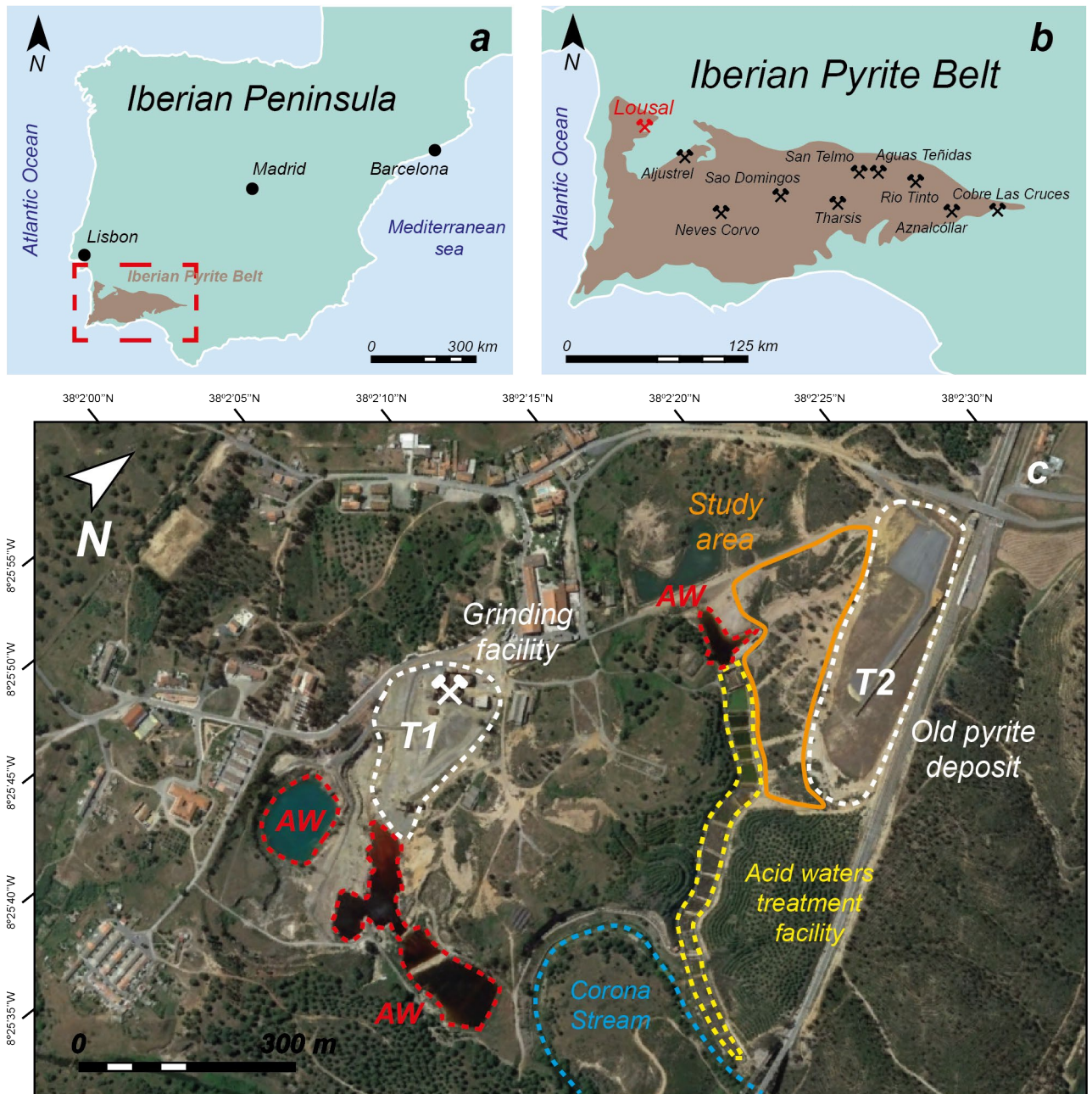


Figure 1. a) Location of The Iberian Pyrite Belt in the Iberian Peninsula; b) Iberian Pyrite Belt and some of its most outstanding metal mines. The Lousal pyrite mine is located at the northwestern border of the Iberian Pyrite Belt; c) Detailed aerial view of the Lousal mine perimeter. T1 and T2: tailings and mine waste piles locations. Pyrite deposits in T2 were removed in reclamation actions in 2010 – 2011 and 2014 – 2015; AW: acid waters dams and ponds.

## ***2.2. The Lousal Mine***

The Lousal mine was exploited between 1900 and 1988 primarily for pyrite to extract sulphur, via underground workings [24, 28, 32]. Mining operations generated large quantities of waste, including overburden material, mineral waste and tailings, with cumulative tonnage estimated at >1 Mt [30, 33].

Nowadays, one principal waste stockpile is observed including material proximal to mine shaft and mill infrastructure, in addition to an old fine-grained pyrite deposit area at the northeast of the mine site [31, 33]. Despite the fact that these areas have recorded extensive weathering, historically no measures were taken to either reduce erosion or to prevent the occurrence of AMD conditions.

With the rapid social, economic and environmental degradation, following post-closure in 1988, an integrated rehabilitation program (RELOUSAL, Rehabilitation and Integral Development of the Lousal Mine) was promoted by the historic mine operator (SEPEC) and the local municipality (City Hall of Grândola). Starting in 2010 – 2011 (first phase) and, later on, in 2014 – 2015, an environmental rehabilitation project was developed as part of the RELOUSAL program by EDM (Empresa de Desenvolvimento Mineiro) which is the Portuguese state company responsible for the environmental rehabilitation of old mine sites in Portugal. Under the scope of that project, a system of artificial wetlands and open limestone channels was constructed in the northeastern complex to mitigate dispersion of potentially toxic elements (PTEs) and to increase the pH of surface waters draining into the Corona stream [34] (Figure 1c). Most of the pyrite residues present on the old pyrite stockpile were removed, and to further encapsulate local waste and reduce the impact of weathering, geotextile with topsoil-cover was placed on a section of the fine-grained old pyrite stockpile.

By the time they were made, EDM's interventions in Lousal were ambitious and innovatives and, overall, well designed and constructed. However, in the absence of ongoing maintenance, these rehabilitation activities have undergone significant deterioration (Figure 2). Unfortunately, signs of accelerated erosion were observed on pyrite stockpile slopes where geotextile was applied. Without adequate maintenance, limestone blocks in open channels became encrusted with iron oxide, while the wetland system lost virtually all riparian vegetation.



Figure 2. Current state of rehabilitation activities implemented by Empresa de Desenvolvimento Mineiro (EDM) in the RELOUSAL rehabilitation project; a) Degraded artificial wetland system after several decades; b) Open limestone channel exhibiting iron oxide encrustation.

Soil contamination from mining activity across the Lousal area has undergone previous investigation. Local studies in Technosols report Contaminant Factors (CF) and Pollution Load Indexes (PLI) for PTEs (Pb, Cd, As, Zn and Cu), representative of high to very high contamination, and especially elevated PTE concentrations in areas nearest the encapsulated pyrite deposit [35]. In 2017, surficial waters were sampled from the wetland treatment system and mine ponds across the mining area. Samples collected from AMD



waters and ponds showed low pH ranges (2.36 - 2.46) and elevated conductivities (4888 - 8600  $\mu\text{S cm}^{-1}$ ). Samples collected from the wetland treatment system exhibited slightly higher pH values, varying from 3.45 - 5.80 and lower conductivities (2900 - 3250  $\mu\text{S cm}^{-1}$ ). Water samples collected from the AMD ponds and the proximal wetland treatment system were analysed for PTEs, exhibiting the highest values in Fe, Cd, As, Pb and Zn (Fe: 404  $\text{mg L}^{-1}$ ; Cd: 0.122  $\text{mg L}^{-1}$ ; As: 0.007  $\text{mg L}^{-1}$ ; Pb: 0.006  $\text{mg L}^{-1}$ ; Zn: 65.7  $\text{mg L}^{-1}$ ).

### **3. Materials and Methods**

#### ***3.1. Rehabilitation actions planned in the LIFE RIBERMINE Project***

To reduce AMD generation and erosion problems across the Lousal mine area, the LIFE RIBERMINE rehabilitation project was implemented in 2019. Its aim was to heal downstream aquatic habitats by introducing best available rehabilitation techniques to both physically and chemically stabilise local mining waste. In particular, disturbed soils proximal to the pyrite deposit were targeted for a geomorphic-based rehabilitation approach (Figure 3). This required adoption of holistic methods combining: a) topographic regrading through geomorphic rehabilitation solutions; b) design and construction of a soil cover to neutralize lateral subsurface water flows and minimize AMD; and c) revegetation of the reclaimed area.

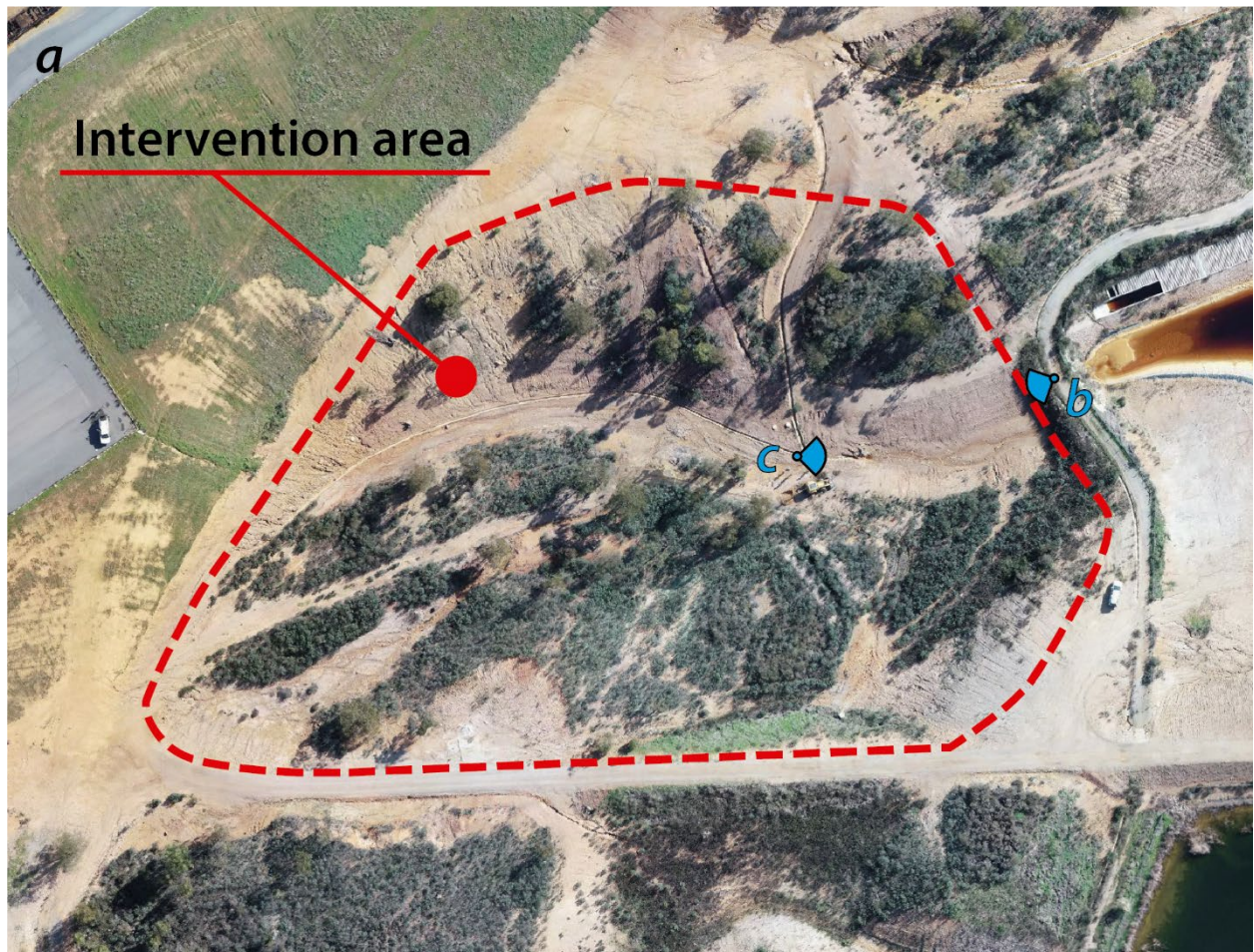


Figure 3. Intervention area; a) General view (the outlook points (b) and (c) are indicated); b) and c) Detailed views of the intervention area before the implementation of the geomorphic-based rehabilitation actions.

The justification for the holistic approach of the rehabilitation methodology used rests on the fact that a total movement of earth and neutralisation of the pyrite deposit was

not economically feasible. Therefore, the option of combining a geomorphic regrading with a neutralising soil cover and open limestone meandering channels adapted to the new 'natural' catchment topography, was thought to be the most efficient solution to manage the subsurface and surficial water flows. We recognise, however, that these actions will not totally solve the chemical contamination problem. The main aim here was to help reduce the AMD levels yield at the head of the Corona watershed to the artificial wetland system built by RELOUSAL. It was also recognised that the open limestone meandering channels for amelioration of AMD are effective for only a limited time, before the limestone lining becomes clogged with iron precipitates, which renders them ineffective. Therefore, regular replacement with fresh limestone blocks will be necessary, to preserve their functionality.

### ***3.2. Geomorphic-based mine rehabilitation***

The GeoFluv-Natural Regrade method [36] was used to design a rehabilitated topography throughout the Lousal mine intervention area. GeoFluv™ is a fluvial-geomorphic method for land rehabilitation which provides users design functionality to create landforms which develop naturally under specific climatic and physiographic conditions [14, 17, 20] (Figure 4). To start, suitable and stable reference areas must be identified to provide rehabilitation design criteria.

Natural Regrade software facilitates making GeoFluv designs in a Computer Aided Design (CAD) format [14, 19, 20, 35, 37]. This method enables mature and natural landform creation, with the formation of steady-state landscapes which seek to balance erosive and resistance forces. Mining areas reclaimed using a geomorphic rehabilitation approach not only provide long-term stability, but also maximise the recovery of local

biodiversity and visually re-integrate faster, thereby achieving a high level of community and administrative acceptance [38].



Figure 4. The GeoFluv rehabilitation method aims to design and construct landforms similar to those found in mature landscapes, including scalloped morphologies and convex-concave hillslopes. Upper image: Agricultural landscape in the Lousal – Faleiros area (Grândola, Portugal) in October 2021. Lower Image: the same landscape in February 2022.

Design inputs were derived from landscape benchmarks using ‘natural’ landform analogues sourced from comparable climatic and physiographic domains, with runoff adjusted to minimise erosion rates. Average Recurrence Intervals (ARI) are required for precipitation input to generate bankfull and flood-prone discharges, as well as maximum stream velocities [39, 40].

GeoFluv-Natural Regrade designs are created by methodically combining all input criteria. The most suitable are those that meet waste material volumes, operational conditions, together with specific project characteristics and constraints from which erosion is stabilized. As ephemeral channels had started to form naturally at Lousal, preliminary designs were adjusted to ensure the primary topography was not unduly eroded / altered.

### ***3.3. Acid Mine Drainage neutralization***

To reduce the impact of AMD and PTE dispersion at Lousal, chemical remediation solutions were integrated with the geomorphic-based rehabilitation activities. A soil cover was designed for the new geomorphic-based surface, coupled with the construction of open limestone channels. This cover was introduced to control AMD generated by runoff and subsurface waters in contact with mine waste materials, with immobilization (adsorption and co-precipitation) of potential PTEs in leachates. In addition, such soil cover would help to develop an edaphic substrate on the new topography, suitable for the re-vegetation of native plants and shrubs. Open limestone channels remain cost-effective means of treating AMD by raising pH and thereby promoting immobilization of dissolved metals, as part of the precipitated Fe and Al oxides and hydroxides.

Owing to their ready integration into the geomorphic design, such channels were chosen at Lousal over alternative options for neutralizing the AMD. Although their service life is usually limited, periodic replacement of spent limestone blocks can be undertaken to preserve their functionality.

### **3.4. Revegetation of the reclaimed mine soils**

To guarantee establishment of vegetation cover across the intervention area, pioneer plants with a wide ecological range, capable of surviving in different environmental and substrate conditions were chosen. A variety of native leguminous, herbaceous and grass species were selected, with a 'freehand' sowing technique employed for seeding. The species selected and their respective proportions (percentages) included: *Brisa maxima* (20%), *Brachypodium phoenicoides* (15%), *Lupinus luteus* L. (10%), *Lupinus angustifolius* L. (7%), *Trifolium pratense* L. (5%), *Papaver rhoeas* (5%), *Calendula arvensis* (4%), *Chrysanthemum coronarium* (4%), *Echium plantagineum* (4%), *Scabiosa atropurpurea* (4%), *Erophaca baetica* (4%), *Cichorium intybus* (3%), *Foeniculum vulgare* (3%), *Lavatera trimestris* (3%), *Raphanus raphanistrum* (3%), *Lupinus micranthus* (1%), *Allium ampeloprasum* (1%), *Gladiolus italicus* (1%), *Piptatherum miliaceum* (1%), *Hyperium perforatum* (1%), *Verbascum sinuatum* (1%). Sowing was carried out in early November 2021, coinciding with the local rainy season.

### **3.5. Monitoring activities**

To monitor the evolution of water quality and riparian ecosystems near the reclaimed area, two new piezometers (P1 and P2) were installed in lower sections of the intervention area, with intakes designed from previous geophysical studies. Piezometer P1 was drilled to nine meters (60 mm diameter), with its perforated screen located between meters seven and eight. Piezometer P2 was shallower (six meters), had the same diameter and its perforated screen was located between meters four and five.

In May 2022, the first water quality monitoring sampling was carried out, collecting a composite groundwater sample from each of the new monitoring piezometers

(P1 and P2), and two pre-existing piezometers (EDM – 1 and EDM – 2) (Figure 5). To ensure the collection of representative samples, all piezometers were purged 24 hours in advance. Groundwater was collected using a PVC Bailer, which was thoroughly cleaned with distilled water between samples. In addition, two surface water samples were also collected from the AMD pond located at the outlet of the reclaimed area (AMD pond) and from a clean water pond located upstream (Clean pond) (Figure 5). All water samples were kept refrigerated until they were sent for analysis.

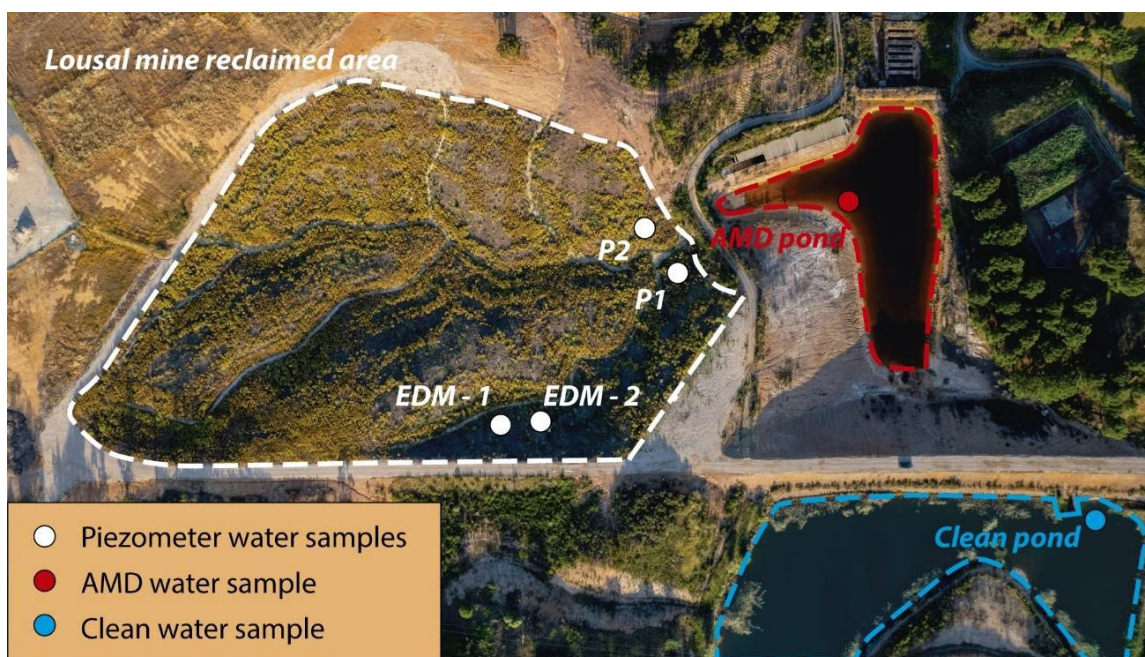


Figure 5. Location of the water samples collected as part of the monitoring actions in Lousal.

All physicochemical parameters were measured *in situ*, using a portable multiparametric probe, while cations, anions and PTEs contents were analysed in external laboratories, using ion chromatography and atomic absorption spectrometric techniques.

## 4. Results

### 4.1. Geomorphic-based mine rehabilitation

#### 4.1.1. Design

Geomorphic reference areas in the vicinity of the Lousal mining area were examined. Stable design inputs were completed from reference areas from across the Iberian Pyrite Belt [35]. These formed the basis for the GeoFluv-parameters (Table 1) used to design 'A' to 'Aa+' and 'Cb' type channels, according to Rosgen's fluvial morphological classification system [39, 40]. Then, the fluvial channels and related scalloped hillslopes were modelled into functional catchments, consistent with the hydrological, topographic and volumetric conditions across the intervention area. Wherever possible, these inputs were adapted to the existing topography, respecting most of the recent surface runoff paths, spontaneously generated throughout the intervention area. Once an optimal alternative was set, the geomorphic restoration plan was finalised, the schematic design views of which are presented in Figure 6.

Table 1. GeoFluv design inputs for the Lousal mine rehabilitation project

<i>Input/setting</i>	Description	Units	Value
Topographic conditions of the reclaimed area	Base level elevation	m.a.s.l.	60
	Slope at the mouth of the main valley bottom channel	%	-1
	'A' channel reach length – slope higher than 4%, according to Rosgen [39, 40]	m	12



Morphometric characteristics from the reference area	Sinuosity of channels with slope higher than 4% ('A' type channels [39, 40])	m	1.12
	Sinuosity of channels with slope lower than 4% ('Cb' type channels [39, 40])	m	1.15
	Maximum distance from the ridgeline to channel's head	m	10
	Target drainage density	m ha <sup>-1</sup>	30
	Angle from subridge to channel's perpendicular, upstream	degrees (°)	10
Precipitation and hydrological data	2-yr, 1-h precipitation	cm	1.79
	50-yr, 6-h precipitation	cm	5.57
	Runoff coefficient	-	0.3
	Maximum channel flow velocity	m s <sup>-1</sup>	1.37

The GeoFluv design selected was based on a single catchment with an area of approximately 1.7 hectares, comprising three main fluvial channels, including an 'A' to 'Aa+' morphology in their headwater and middle reaches connecting downstream to an AMD treatment system. Two 'A' to 'Aa+' channels merge in the middle section of the GeoFluv design to form a single 'A' to 'Aa+' channel, while in its final section, the channel is seen to meander ('Cb' channel type). The third GeoFluv channel runs parallel to the western margin of the rehabilitation design, and is composed of an 'A' to 'Aa+' type channel section (upper and middle reaches of the channel), and a meandering section. This channel joins the central channel in its lower section, before reaching an outlet point.

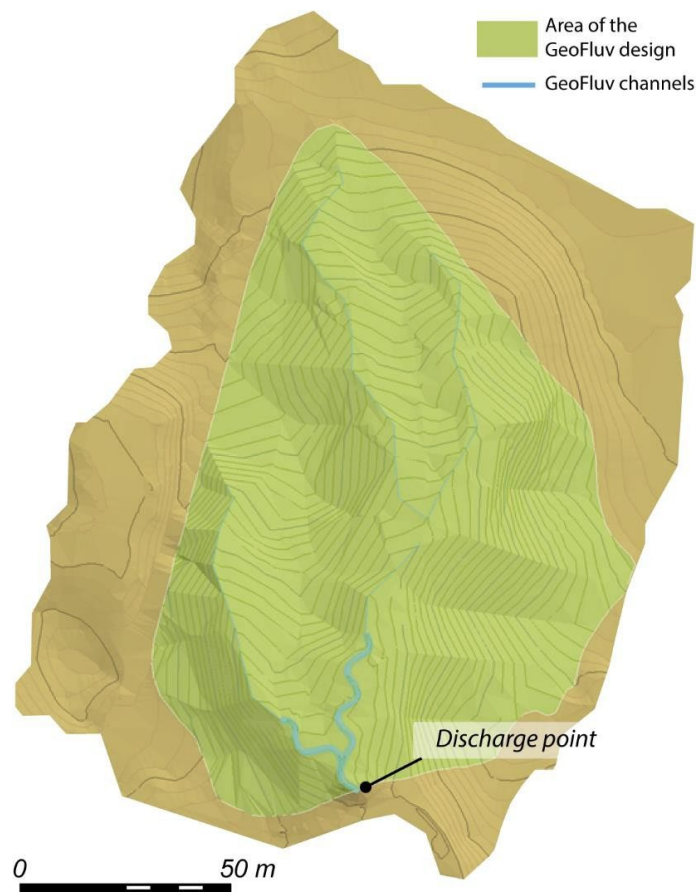


Figure 6. Geomorphic design of the intervention area in Lousal mine.

The position and trajectory of the GeoFluv-designed channels were designed to follow natural drainage patterns within the intervention area. The interfluvial areas between channels were set on main ridges (divides) with rounded scalloped hillslopes on both sides, with alternating swales and subridges in each slope. This scalloped morphology improves surface water flow by distributing runoff over smaller micro-catchments and reduces the impact of surface runoff erosion processes.

#### *4.1.2. Execution and earthworks actions*

The Lousal mine geomorphic-based rehabilitation was undertaken between September

and October 2021 (Figure 7). Prior to the initiation of the program, shrubs (mainly *Cistus ladanifer*) were cleared and invasive eucalyptus trees in the intervention area were uprooted.



Figure 7. Geomorphic regrading actions: a), b), c) and d) Bulldozer and excavator constructing new rehabilitation topography; e) Back-up tractors used for earth moving; f) View of the intervention area during late-stage of topographic regrading.

Design landforms were then surveyed using differential GPS and the earthworks plan was laid out respecting local topography, roads, and operational cycles.

A week of specialised training was required for heavy equipment operators. Thereafter, topographic regrading and construction was undertaken using bulldozers (D6), with the construction of the GeoFluv channels and other landforms using multiple excavators. The earthworks achieved cut – fill balance (no exporting or importing materials were needed). Despite minor deviations to plan were observed after heavy rainfall at the end of November 2021, periodic inspections confirmed satisfactory runoff management across the intervention area, although drainage from the encapsulated old pyrite stockpile led to local rilling (Figure 8).

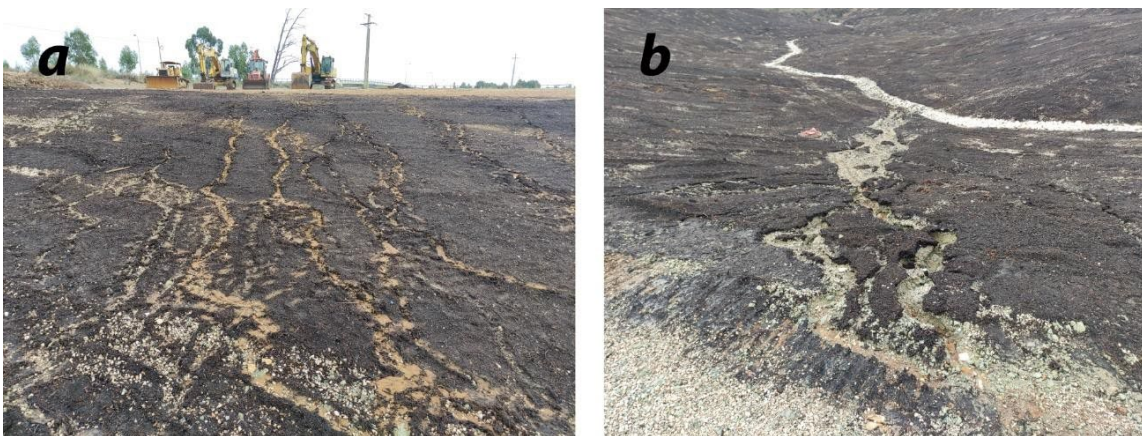


Figure 8. Rilling processes in the reclaimed area near the encapsulated pyrite deposit; (a) Small rills of 5 – 10 cm width and 2 cm depth; (b) Rills of 20 – 30 cm width and 10 – 20 cm depth.

#### ***4.2. Acid Mine Drainage chemical stabilization***

Soil cover consisted of two horizons: a) a shallower organic horizon (horizon A); and b) a subsurface mineral horizon (horizon B) (Figure 9).

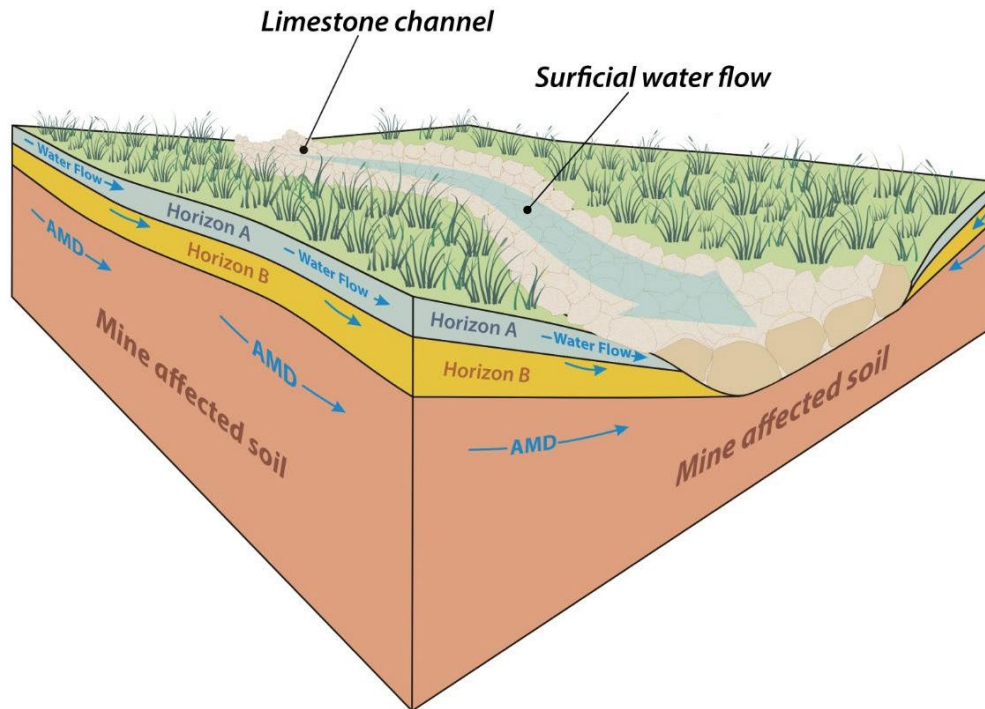


Figure 9. Diagram of the soil cover designed and constructed in the Lousal rehabilitation project.

Horizon A attained thicknesses of approximately 5 -10 cm (486,67 m<sup>3</sup> total volume), and was composed of a homogeneous mixture of poultry and horse manure (220 m<sup>3</sup>, 45% of total Horizon A volume), together with topsoil (266.67 m<sup>3</sup>, 55% of total Horizon A volume). Given the right moisture and temperature conditions for plant germination and development, the manure fraction provided sufficient organic matter and carbon to develop the shrub cover.

Horizon B had thicknesses of approximately 10 - 15 cm (1866.77 m<sup>3</sup> of total volume), and was composed of a mixture of clay (<10% smectite; 750 m<sup>3</sup>, 40% of total Horizon B volume) and limestone gravel (12/20 mm diameter, 1116.77 m<sup>3</sup>, 60% of total Horizon B volume). This underlying layer was introduced to: a) help neutralise water; b) facilitate the circulation of subsurface flows; and c) aid in water retention to promote plant growth.

The thickness of each of the soil horizons is shown as a range of values, between a maximum and a minimum, because the heavy machinery methodology used for the extension of each of the horizons is not precise enough to allow for an invariable thickness to be applied throughout the entire reclamation site. Furthermore, during the application of the soil cover, it was considered appropriate to increase the thickness of the Horizon B in some areas where there was a greater risk of AMD generation.

As designed, this rehabilitation program was intended to link the open limestone and geomorphic channels including: a) 'A' to 'Aa+' type channels; and b) meandering 'Cb' type channels. The limestone lining designed for 'A' to 'Aa+' type channels was 50 cm in width and 20 cm in height. For the 'Cb' type meandering channels, the lining was designed with a width of 2 m and a height of 20 cm.

#### *4.2.1. Program Execution*

Remediation began in October and was completed in November of 2021. It commenced with the creation of the soil layers using heavy equipment for batch mixing (Figure 10), with soil spread mechanically by backhoe and where required, manual labour. As uniform thicknesses were difficult to achieve, additional limestone gravel and clay materials (Horizon B) were added where AMD was anticipated. In headwaters and elevated ground, Horizon A was intentionally thickened to accommodate rain, wind and gravity settling. After implementation of the soil cover, the limestone neutralization channels were constructed as described above, using both heavy machinery and manual labour.

#### *4.3. Monitoring activity*

Six months after completion of the rehabilitation program, monitoring chemical analysis

of groundwater and surface water samples was undertaken (Table 2). These values are intended to provide a reference point against which future analyses can be compared to evaluate the success of the program.



Figure 10. Execution of soil cover and AMD remediation actions: a) and b) Materials used in the construction of the soil cover, limestone gravel and clays (a), and animal manure and topsoil (b); c) Extension of the materials over the rehabilitation area with a backhoe; d) View of the intervention area after the extension of the soil horizon B; e)

View of the first stages of the extension of the soil horizon A; f) Lining of limestone blocks over the GeoFluv designed channels.

Table 2. Chemical characterisation of groundwater and surface water samples collected from the monitoring piezometers, AMD pond and Clean water pond near the intervention area.

Sample	pH	EC ( $\mu\text{S cm}^{-1}$ )	ORP (mV)
P1	7.0	1770	-10.5
P2	6.3	>8000	31.0
EDM – P1	4.3	4750	124.5
EDM – P2	3.9	3840	152.5
AMD pond	2.6	3018	226.0
Clean pond	7.8	950	5.0

Sample	Cl <sup>-</sup> (mg L <sup>-1</sup> )	NO <sup>2-</sup> (mg L <sup>-1</sup> )	NO <sup>3-</sup> (mg L <sup>-1</sup> )	HPO <sub>4</sub> <sup>2-</sup> (mg L <sup>-1</sup> )	SO <sub>4</sub> <sup>2-</sup> (mg L <sup>-1</sup> )
P1	141	<0.4	<2	<2	195
P2	80	<0.8	22.1	<4	6031
EDM – P1	120	<0.2	1.1	<1	2818
EDM – P2	59	<0.4	<2	<2	2400
AMD pond	117	<0.3	5.9	<1.5	1474



Sample	Na <sup>+</sup> (mg L <sup>-1</sup> )	NH <sub>4</sub> <sup>+</sup> (mg L <sup>-1</sup> )	K <sup>+</sup> (mg L <sup>-1</sup> )	Ca <sup>2+</sup> (mg L <sup>-1</sup> )	Mg <sup>2+</sup> (mg L <sup>-1</sup> )
P1	135	1.8	61.6	58	67
P2	714	<4.0	2.4	286	954
EDM – P1	344	<1.0	1.7	192	414
EDM – P2	146	<2.0	1.9	223	368
AMD pond	71	2.07	26.8	162	95
Clean pond	99	<0.5	6.0	32	36

Sample	Fe (mg L <sup>-1</sup> )	Zn (mg L <sup>-1</sup> )	Pb (mg L <sup>-1</sup> )	Cd (µg L <sup>-1</sup> )
P1	5.03	0.06	<0.01	<0.3
P2	0.08	3.89	<0.01	83.1
EDM – P1	0.26	2.19	<0.01	<0.3
EDM – P2	3.69	3.47	<0.01	<0.3
AMD pond	34.45	15.89	<0.01	0.8

Values of pH measured from P1 and P2 locations are close to neutral, comparable to those observed in the Clean water pond near the intervention area. Conversely, values from samples EMD – 1 and EMD – 2 are more acidic, mirroring values measured from the AMD pond (pH = 2.6). Both EDM piezometers are located on the slope of the entrance

road to the old pyrite storage area which is underlain by sulphide-bearing rock, which may be a local AMD source.

In terms of conductivity, all water samples show relatively high EC values, except for a Clean water sample. The extremely high value at P2 may be associated with  $Mg^{2+}$  and  $Ca^{2+}$  concentrations and local dissolution of carbonate. ORP values are typical of an oxidizing environment, with the exception of a negative value obtained in P1. High organic carbon content related to organic matter (leached from the organic layer of the soil cover) and bacterial activity, as well as low recharge, likely explain that negative ORP value, which typify reducing environments.

High Fe concentration values are observed in the AMD pond ( $34.45 \text{ mg L}^{-1}$ ), and in P1 ( $5.03 \text{ mg L}^{-1}$ ), linked to low ORP values. Zn values are higher in piezometers exhibiting lower pH, while Pb values are not uniformly detected. Concentrations of Cd, the element with the highest toxicity, are only significant in P2 ( $83.1 \text{ } \mu\text{g L}^{-1}$ ) and in the AMD pond ( $0.8 \text{ } \mu\text{g L}^{-1}$ ).

Concerning the remediation strategy on the water quality of the AMD pond, the comparison of data from 2017 to 2022 offers few differences in terms of pH (2.2 to 2.6), ORP (740 to 226 mV) and EC ( $4800$  to  $3018 \text{ } \mu\text{S cm}^{-1}$ ). The differences in some PTEs concentrations are more significant, especially in Fe ( $404$  to  $34 \text{ mg L}^{-1}$ ), Zn ( $65.7$  to  $15.8 \text{ mg L}^{-1}$ ) and Cd ( $122$  to  $0.8 \text{ } \mu\text{g L}^{-1}$ ). The Cd and Pb are lower than those established by the Portuguese Legislation for acceptable water quality in surface waters (Pb –  $0.05 \text{ mg L}^{-1}$ , Cd –  $0.01 \text{ mg L}^{-1}$ , Zn –  $0.5 \text{ mg L}^{-1}$ ) [41], which may indicate marginal improvement in water quality after remediation.

## **5. Insights and lessons learned from a pioneering mine rehabilitation project**

In this innovative study combining chemical neutralization of Acid Mine Drainage (AMD) with geomorphic reclamation designs (Figure 11), the planning and construction of the edaphic cover was subject to ongoing adjustments throughout the execution phase of the project. One of the main threats to the soil cover in the intervention area was the impact that high rainfall might have on soil chemistry linked to premature erosion. Inspections made after the single intense event recorded in late November 2021 suggest that, as constructed, the current soil cover should remain intact, with erosive runoff unlikely to occur. However, some localized rill formation ranging from 1-2 cm to 20 cm in depth and wide was observed after heavy precipitation (Figure 12a). Another component contributing to erosional instability was observed as surface runoff from the adjacent pyrite deposit and nearby roads entered the area of intervention.

This erosional issue has been previously identified in other geomorphic mine rehabilitations in the Iberian Peninsula [14, 20]. Although there are constraints that prevent the inclusion of upper drainage sources into the rehabilitation project, external drainage sources require careful and continued management. Thus, rill development was corrected by extending limestone channels using blocks to moderate surface runoff and its erosive potential (Figure 12).

The rehabilitation actions implemented in the Lousal mine have significantly improved the ecological value and water quality of the reclaimed mine area. After five months, the acidic water lagoon located at the drainage point of the reclaimed area has changed from a reddish AMD colour, to a turquoise green, comparable to nearby clean water lagoons (Figure 13). Moreover, a vegetation cover has already begun to establish over the new reclaimed topography, including numerous herbaceous, gramineae and leguminous species, which is consistent with the revegetation plan (Figure 14). The

invasive and exotic species that were removed prior to the initiation of the program (*Cistus ladanifer* and eucalyptus trees) were not established again in the intervention area.

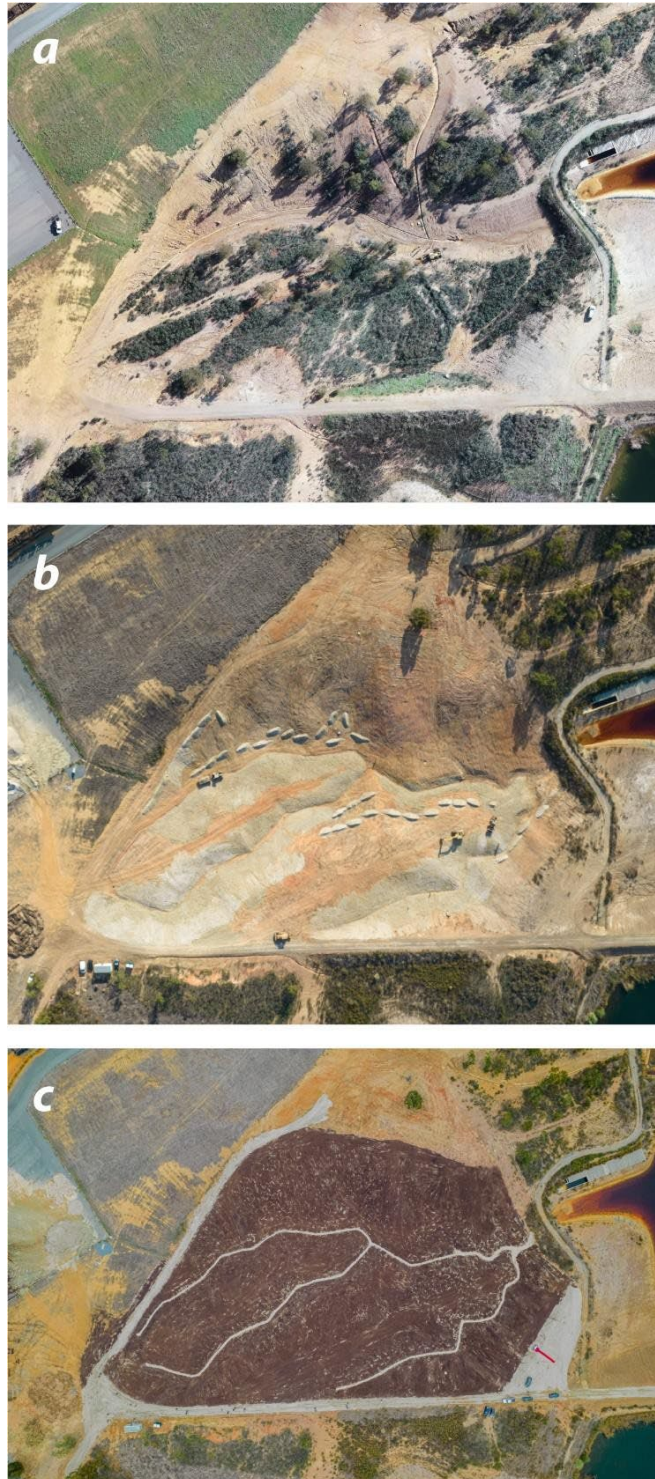


Figure 11. Comparative view of the Lousal mine pilot area during different stages of the implementation of the rehabilitation project; a) View of the intervention area prior to the

start of the rehabilitation actions, January 2020; b) Aerial view after the conclusion of the topographic regrading actions, October 2021; c) Aerial view of the intervention area once all the rehabilitation actions were implemented, November 2021.

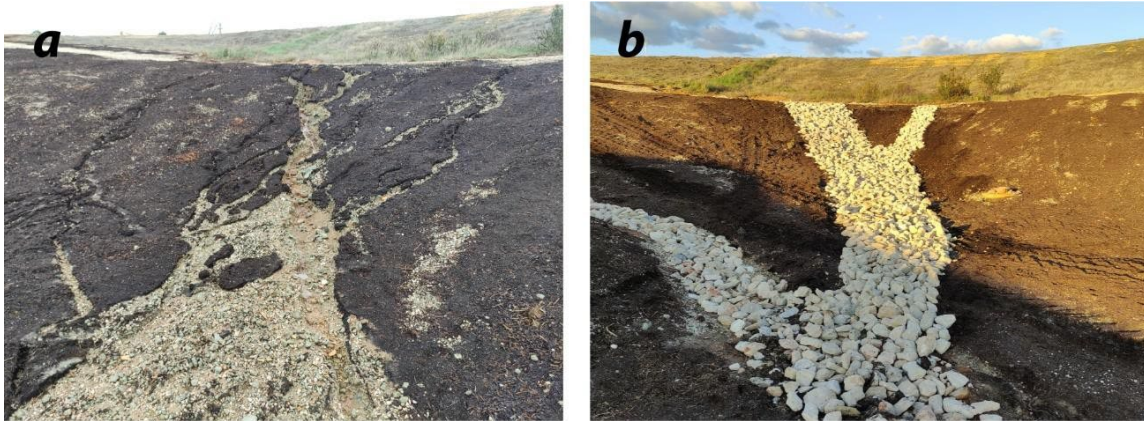


Figure 12. a) Rills developed in the slopes as a consequence of external runoff entering the rehabilitation area; b) New limestone open channels constructed in rill eroded areas in order to create additional channels to manage the incoming surface runoff.



Figure 13. AMD drainage pond located down waters from the intervention area. Left, view of the AMD drainage pond in January 2020. Right, view of the AMD drainage pond in April 2022.

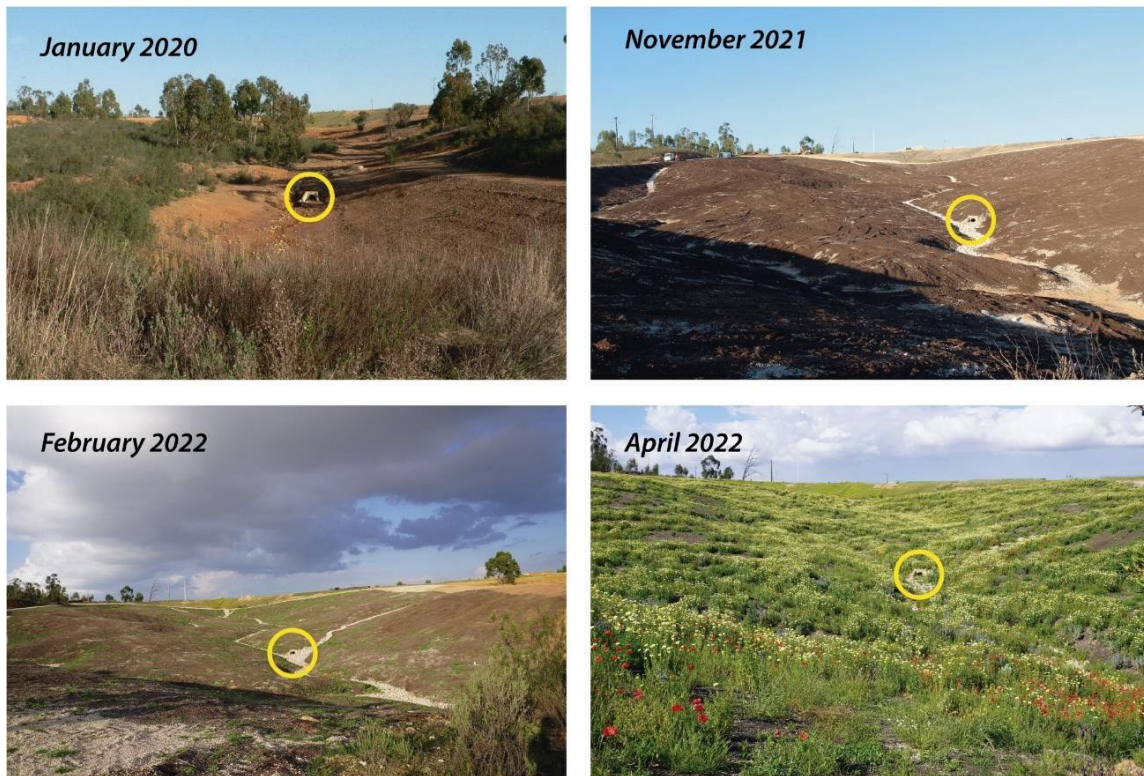


Figure 14. Comparative view of the reclaimed area of the Lousal mine in January 2020 (before the rehabilitation actions), November 2021 (after all the rehabilitation actions were implemented), February 2022 and April 2022 (after seed germination). A vegetation cover started to grow in the intervention area due to the combined reclamation approaches. In the yellow circle is an artificial outlet that serves as reference point.

Although acid leachates do not yet reflect the effects of the rehabilitation on pH and EC values, such effects are observed in both ORP and PTEs concentration. For the duration of the LIFE RIBERMINE project, periodic analysis of surface and groundwater and riparian vegetation will continue to monitor water quality and riparian ecosystem recovery.

## 6. Conclusions

The rehabilitation of the Lousal mine, within the framework of the LIFE RIBERMINE project, is the first project in Europe to combine geomorphic-based rehabilitation methodologies with chemical remediation solutions. To reduce erosion of soils affected by the mining activity, a new rehabilitation topography, mimicking natural landforms, was designed and constructed, using the GeoFluv method. This project has been based on the synergies between the beneficial effects of each rehabilitation action. Thus, the distribution of surface runoff in small sub-basins, and the scalloped landscape composed of ridges and swales, reduces the erosive potential of surface runoff. This, in turn, makes the soil cover stable and remains mostly unaffected by erosion. The effects of the integrated physical and chemical rehabilitation actions became visible in March 2022, with the establishment of a vegetation cover over most of the reclaimed area and a visible improvement of the water quality in the AMD treatment system. Only few localized areas of the new rehabilitation topography have experienced minor rill erosion, which is associated with water ingress from adjacent watersheds.

Regarding the geochemical conditions on the site, minor localised reducing conditions (negative ORP values) were detected near the outlet of the reclaimed area, possibly related to leaching of organic matter from the soil cover and low water recharge. Potentially Toxic Elements (PTEs) concentrations have reduced significantly after the reclamation actions, especially Fe (404 to 34 mg L<sup>-1</sup>), Zn (65.7 to 15.8 mg L<sup>-1</sup>) and Cd (122 to 0.8 µg L<sup>-1</sup>) concentrations. The results obtained from chemical analysis of water samples suggest that open limestone channels constructed on the site are effective for the amelioration of AMD. However, regular replacement of the limestone lining with fresh limestone blocks will be necessary to preserve their functionality over time. Detailed monitoring of this restoration progression through 2024 and beyond should offer learning opportunities and

innovations which will benefit future rehabilitation projects with comparable environmental features.

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### **Disclosure statement**

The authors report there are no competing interests to declare.



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