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Review

Do Old Mining Areas Represent an Environmental Problem and Health Risk? A Critical Discussion through a Particular Case

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Abstract: A bibliographic review was carried out to establish the state of knowledge of a mining area with several centuries of exploitation and currently abandoned. The selected case study, the Sierra Minera de Cartagena-La Union (Spain), has a long history of mining activity, ending in 1990. The area is rich in metallic sulphide (lead, zinc and iron), with underground mines and quarries. The zone is very close to important populations and affects protected sites of special ecological value. It is also adjacent to areas dedicated to agriculture and important centres of tourist interest. It is a territory that meets the requirements to be classified as a critical area, as it is in a state of unstable physical and geochemical equilibrium, giving rise to possible risks to human health and ecosystems. A literature review was carried out according to the PRISMA (Preferred Reporting Items for Systematic Reviews and Meta-Analyses) methodology criteria, consulting a large number of related publications. The results obtained using the Source-Pathway-Receptor model make it possible to identify the main impacts caused by the contamination sources, the main routes of contamination, as well as the transfer to the biota and the influence on adjacent agricultural soils. In this study, lead, cadmium, zinc, arsenic, copper and manganese were considered as potential toxic elements (PTEs), and data were obtained on concentrations in soil, water and air as well as in fauna and flora. Finally, once the receptors and the associated risks to the ecosystem and human health were identified, a conceptual model of the contamination was drawn up to consider a management proposal to tackle the problems associated with this area, which would also be applicable to critical mining zones.

Keywords: mining areas; contamination source; transfer; risks; management

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1. Introduction

Abandoned mining areas present some common characteristics associated with the degradation of the original ecosystem. These include impacts on the relief, changes in the fluvial network, effects on fauna and flora, and transformations of physico-chemical properties and chemistry of soil, stream waters, groundwater and biota related to the distribution and dispersion of contaminants in the surface environment. They are difficult to manage, have no defined legal framework and can be considered as orphan areas [1], representing Critical Abandoned Mining Areas (CAMAs) [2].

This is a global problem that requires countries to manage post-mining waste. It is estimated that there are more than one million abandoned mining areas worldwide, highlighting the importance of this environmental problem in countries such as Japan and France, with 4000 and 6000 abandoned mines, respectively. Other areas of the world with a large number of abandoned mining sites include South Africa with an estimated 6150, New South Wales (Australia) with 2000, Sweden with 10,000 and the United States with approximately 550,000 [3].

The problems encountered in the management of CAMAs are related to different factors, such as the legal complexity that regulates them, the establishment of prioritised areas of influence and the absence of global recovery projects. Many areas have been extensively studied in a large number of publications, with the main impacts of each site being identified; however, in most studies, the area of influence has not been clearly defined, and there are no comprehensive management proposals [4].

This paper addresses the current environmental problems represented by CAMAs as seen in the case of the Sierra Minera de Cartagena-La Unión (SM), located in south eastern Spain, analysing the influence on the local population, agricultural soils and nearby coastal areas. To this end, a conceptual model of the current state of contamination of the SM is proposed, identifying contaminant sources, transfer routes, impacts of Potentially Toxic Elements (PTEs) contamination on adjacent receiving areas and the transfer to the flora and fauna of the receptor. In this way, a graphical representation of a system that includes the different elements that affect both local and diffuse contamination in a given area is obtained. This allows the identification of the risks of contamination in adjacent receiving areas and their transfer to the flora and fauna of the receptors (ecosystems (terrestrial and aquatic), crops and public health) (Figure 1).

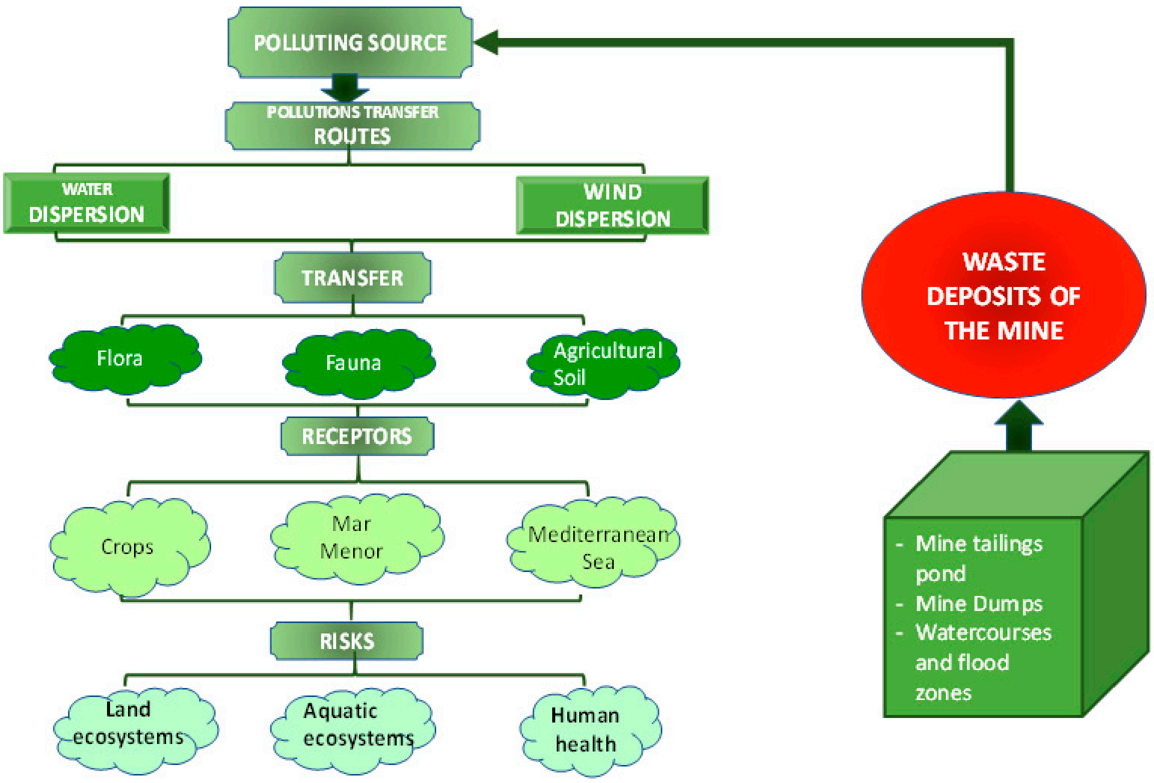

Figure 1. Conceptual model of the current state of contamination of Critical Abandoned Mining Areas (CAMAs): application to the Sierra Minera of Cartagena-La Unión case.

An extensive literature review of scientific articles published from 1973 to March 2021 and a classification according to the type of published work and the subjects covered (Figure 2) were carried out. For this purpose, the PRISMA methodology was used, and a total of 220 research articles, 15 books and book chapters and 34 doctoral theses were considered.

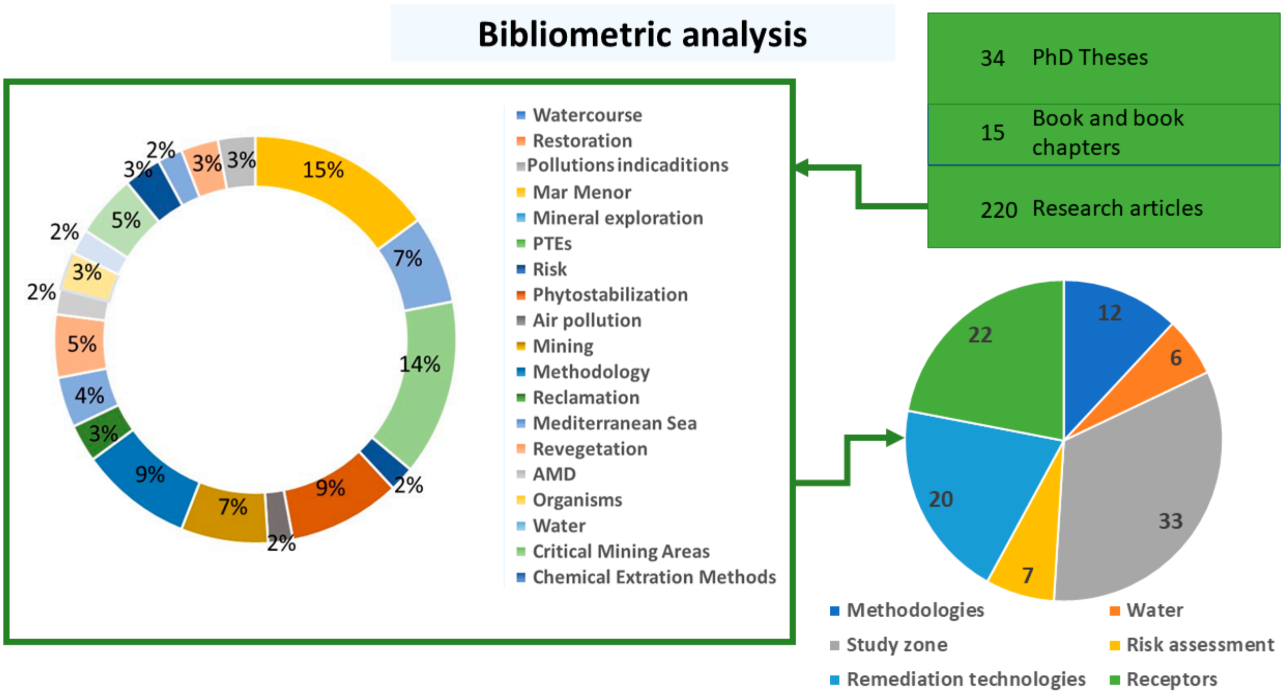

Figure 2. Summary of bibliometric analysis.

2. Protected Areas Affected by the Mining Activities of the Study Zone

The contamination resulting from abandoned mining activities carried out in the study area of the SM affects numerous sites that are protected by a variety of environmental measures. This protection may seem somewhat contradictory, in the sense that the underlying aim is to protect biodiversity by means of protection instruments established by Law 42/2007, of 13 December, on Natural Heritage and Biodiversity [5]. Most of the mining district of Cartagena-La Unión has been declared as being of Cultural Interest [6].

There is no single regulation governing this type of site; the current legislative sequence is based primarily on European directives and also on national transpositions of different categories relating to water, waste, mining waste, and the Natura Network. Through the different European sectoral policies on environmental protection, the legislation that indirectly affects soil protection can be summarized as follows:

- EC Directive on Prevention and Control of Pollution (1996) [7]
- Hazardous Waste Directive (91/689/EEC) [8]
- Environmental Impact Assessment Directive (85/337/EEC) [9]
- Water Framework Directive (60/2000/EC) [10]
- Directive on Environmental Liability in relation to the prevention and remedying of environmental damage (Directive 2004/35/EC) [11]
- Directive for the protection of groundwater against pollution (2006/118/EC) [12]
- Proposal of Directive for the protection of soil (2006/0086 (COD)) [13]
- Waste Framework Directive 2008 [14]
- Integrated Pollution Prevention and Control Directive ((2008/1/EC) [15]
- Industrial Emissions Directive (IPPC) (2010/75/EC) [16]

The transposition into Spanish law of Directive 2006/21/EC (OJEU, 2006) [17] was done through Royal Decree 975/2009 of June 12 (BOE, 2009) [18] on the management of waste from extractive industries and established that within four years an inventory of closed mining waste facilities should be prepared. The aim of this legislation was to identify those installations that have a serious negative environmental impact or that could in the medium or short term become a threat to human health or the environment; however, it did not include any methodology for carrying out the necessary risk analysis that should accompany it.

This left a regulatory vacuum that led to the consideration of mining activity as a cause of soil contamination and sites as potentially contaminated. Therefore, although mining was not directly included in the Royal Decree (RD) 9/2005 of January 14 (BOE, 2005) [19], which established the list of potentially soil-polluting activities, it is related to polluting activities with very diverse effects, including toxic risk to human health and ecosystems.

The great environmental importance that all the protected areas affected by this mining contamination represents should be borne in mind, as should the specific protection afforded to area of the Mar Menor lagoon, by the Law 3/2020 of 27 July [20]. This regulation addresses the problems derived from the transport of heavy metals from abandoned mining areas into the salt lagoon and explains the need to facilitate the restoration of mining waste facilities and the recovery of sites affected by metal mining (Article 75). This law also states that it is the State that has the power to issue the basic legislation, while it is up to the Autonomous Community to develop the legislation and implement the laws related to mining (Article 11.4 EARM).

Without entering into debate about who holds the power to respond to the problem of the heavy metals left from past mining activities, this law does not establish specific protection measures for immediate application, as has happened in the case of agriculture, livestock rearing and urban development. This means that the situation has not changed, and this Mining Critical Zone continues without a plan, without specific actions to execute in a given time limit in order to prevent or at least decrease the amount of PTEs reaching the Mar Menor lagoon.

3. Geological Characteristics of the Study Zone

The SM and its area of 21 mining influence is a coastal mountain range, bordering the Mediterranean Sea to the south and the important salt lagoon, Mar Menor, to the north. Figure 3 depicts the SM and its area of influence, which covers a surface of approximately 100 km^2, and shows the lithology of the SM and its areas of influence, indicating the mining area in its central part. The materials located to the north of the SM are red soils (luvisols), which reach as far as the Mar Menor lagoon. On the southern slope towards the Mediterranean Sea, carbonate rocks, schist, quartzite and micaschist are abundant.

The study zone belongs to the Betica s.s. zone or internal domain and is characterised by the structural stacking of three tectonic complexes, affected by decreasing degree of metamorphism from bottom to top, both within each complex and in the entire series. These units were intensively folded and thrust during the Late Oligocene and Early Miocene and subsequently underwent extensional collapse through large landslide systems in the Mddle-Late Miocene [21,22]. This latter episode was accompanied by important calc-alkaline and shoshonitic volcanism (andesites, dacites, rhyolites) along the Almería-Cartagena volcanic belt (ACVB), while sedimentation took place within restricted marine sedimentary basins. Volcanism triggered hydrothermal activity, leading to the formation of important ore deposits of Pb (Ag), Zn and Sn [23]. Mineralisation is strata bound and the stockwork and vein type of PSG (pyrite, sphalerite and galena) [24].

In the Mining District of Cartagena-La Unión, the total area occupied by mine tailings is approximately 9 km^2, with a volume of 175 Mm^3 on land and a further 25 Mm^3 on the seashore (Bays of Portman, Gorguel and Playa la Galera) [25].

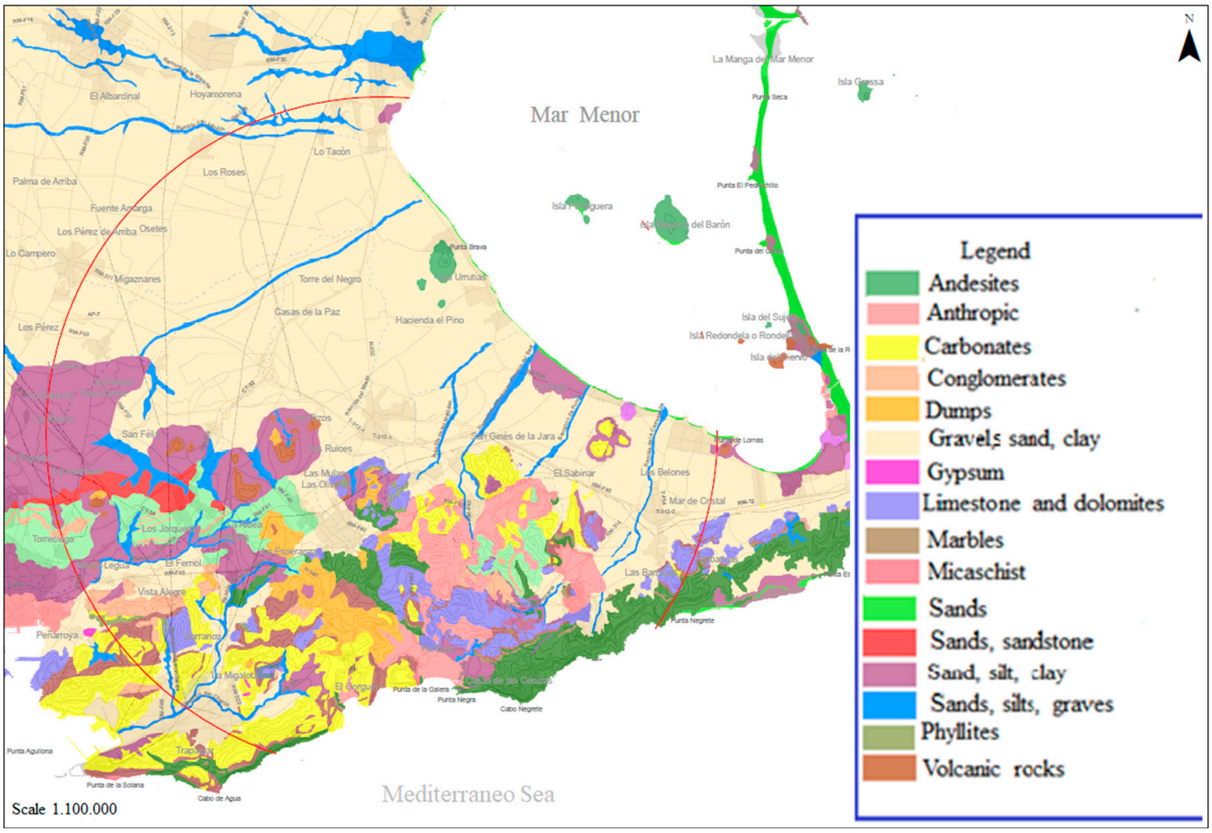

Figure 3. Lithology of the study zone.

4. Contamination Sources

Figure 4 shows the location of the main abandoned mining facilities and the Protected Areas affected by mining contamination. Table 1 summarizes the main areas protected by the different protection laws, which, due to their proximity and/or the transfer of contaminated materials resulting from mining wastes, present high concentrations of PTEs.

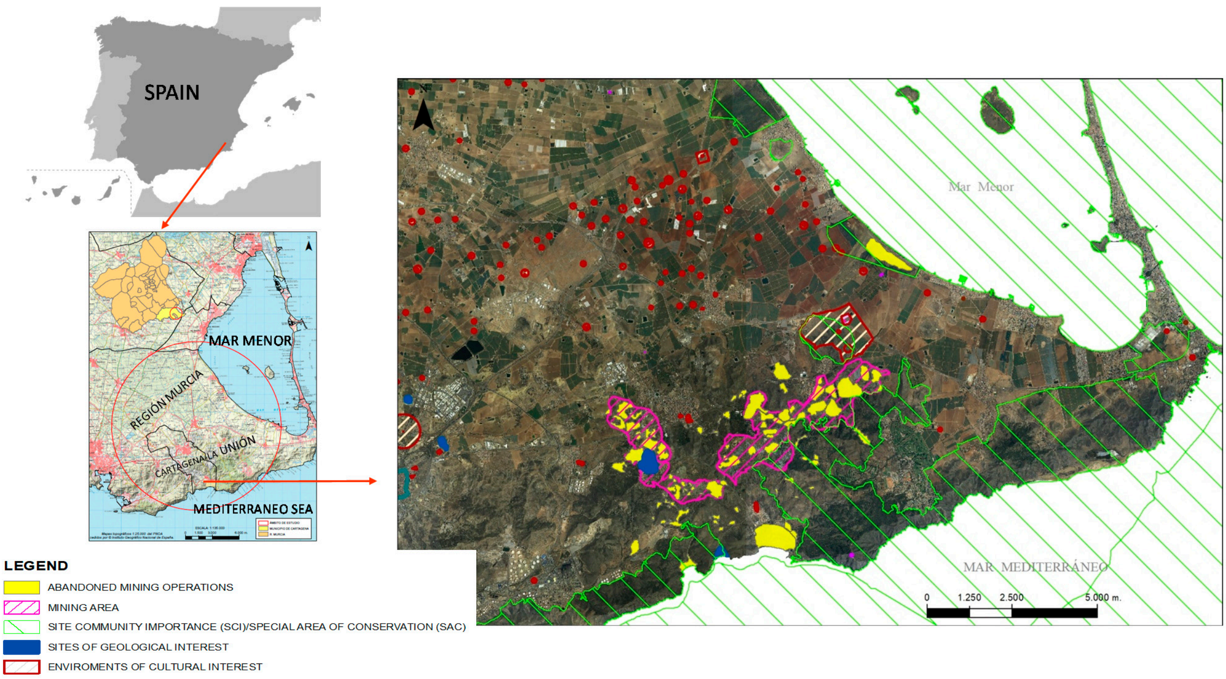

Figure 4. Location and characteristics of the study area.

Table 1. Protected areas affected by the mining activities of the study zone.

ZONE: Mar Menor Lagoon	ZONE: Mediterranean Sea
Protected Natural Spaces at Región of Murcia Level	
Regional Park: Salinas & Arenales de San Pedro del Pinatar [26] Protected Landscape: Open Spaces and Islands of the Mar Menor [26] Protected Landscape: Cabezo Gordo [27] Protected Landscape: Islands and Islets of the Mediterranean Littoral [27]	Regional Park: Calblanque, Monte de las Cenizas and Peña del Águila [26]
Protected Natural Spaces at Community Level (Natura 2000)	
SAC ES0000175 Salinas y Arenales de San Pedro del Pinatar [27] SAC ES6200006 Open Spaces and Islands of the Mar Menor [27] SAC ES6200007 Islands and Islets of the Mediterranean Littoral [27] SAC ES6200013 Cabezo Gordo [27] SAC ES6200029 Submerged Coastal Belt of the Region of Murcia [27] SAC ES6200030 Mar Menor [27] SPAB ES0000175 Salinas y Arenales de San Pedro del Pinatar [27] SPAB ES0000200 Grosa Island [27] SPAB ES0000256 Hormigas Islands [27] SPAB ES0000260 Mar Menor [27] SPAB ES0000270 Cueva de Lobos Island [27] SPAB ES0000271 Palomas Island [27]	SCI ES6200029 Submerged coastal strip of the Murcia Region [28] SCI ES6200025 Sierra de La Fausilla [28] SPAB ES0000199 Sierra de la Fausilla [29]
Natural Spaces Protected by International Instruments	
Wetland of International Importance Mar Menor [30] Specially Protected Areas of Importance for the Mediterranean Sea (SPAMI) [31]	
Sites of Geological Interest	
Sierra Minera de La Unión [32] Cabezo Rajao [32] Corta Brunita and laguna ácida [33]	

SAC: Special Areas of Conservation; SPAB: Special Protection Areas for Birds; SCI: Sites of Community Importance; SPAMI: Special Areas of Mediterranean Importance.

The mining tailings found in the SM are materials remaining from the mechanical and metallurgical treatment of the beneficiary ores. These materials consist of undisturbed bedrock, altered bedrock, materials of primary mineralisation (metallic sulphides) and secondary mineralisation (hydrothermal alteration), products resulting from the supergenic alteration of sulphides, and waste/tailings resulting from ore processing [34]. The focal points corresponding to the contamination sources present materials of a varied nature and heterogeneous granulometry. Different types of contaminating sources or focal points, both of a soluble and particulate nature, that behave differently as a result of their dispersion by water can be established [35–39]:

- Type 1 focal points: Tailing ponds of mine wastes. These include ponds in the middle of the slopes, where erosion caused by water is very high, as well as others located in piedmont areas, where leachates are accumulated, forming large ponds.
- Type 2 focal points: Mine dumps. These are accumulations of mixed materials (waste dumps, smelter tailings, smelting pits) that have undergone different degrees of supergenic alteration and very highwater erosion and show a high degree of stoniness.
- Type 3 focal points: Watercourses and flood zones. These make up an extensive network in the study area and act as the main dispersion pathways of mining wastes, both in the form of particles and in soluble form.
- Type 4 focal points: Deposits of mining wastes or mineral flotation sludge located in areas far away from the SM, which have been transported to or dumped directly in these areas. Examples of such sources are the Portman Bay, the Lo Poyo salt flats (located in Mar Menor lagoon) and other small beach areas.

These foci represent a source of contamination of Potentially Toxic Elements. The characteristics of mining wastes have been extensively studied. They are mainly characterised by a low plant cover, or even the absence of the same, as well as a paucity of associated fauna and soil microorganisms, evidence by substantial supergenic alteration processes. Chemically, they are characterised by their acidity, salinity and high content of PTEs.

Table 2 provides a statistical summary of the specific characteristics of the abandoned mining area studied. It summarises the results of several focal points in the study area and shows that most of the mining residues studied have acidic or very acidic pH values, a medium or high salt content and PTE content.

The highest values of PTEs correspond to the tailing ponds and to the deposits by the sea of Portman and Lo Poyo. Portman bay accumulates two types of waste, one washed and reclassified by seawater, with a sandy texture and dark colour (Portman BS, Table 2), and the other yellow, with a silty-clay texture, which corresponds to periods of direct discharge of flotation sludge (Portman YS, Table 2). Both materials are characterised by a high content of PTEs, including carcinogenic elements such as As and Cd, with very different mobilities, high in yellow-coloured materials and very low in dark-coloured ones. This fact corresponds to the alteration processes to which the material not affected by the grain selection is subjected, which are the same as those of the rest of the mining ponds in the SM [40]. The Lo Poyo site, next to Mar Menor lagoon, only presents a type of material similar to that of a mining pond, which in this case is located at a great distance from the source and which received, for a period of time, mineral flotation sludge.

Table 2. Fe_2O_3 total, PTE content, pH and Electrical Conductivity (EC) values of the abandoned residues in the study area (median values).

		pH (dS m^{-1})	E.C. (dS m^{-1})	Pb (mg/kg)	Zn (mg/kg)	Cu (mg/kg)	Cd (mg/kg)	As (mg/kg)	Fe_2O_3 (%)	Mn (mg/kg)	Ref.
Mine Dumps	Brunita (Depth 1 m)	2.79	7.99	2810	6500	119	33.1	102	39.3	-	[41]
Mine Dumps	Mining tailing “Belleza”	3.0	4.47	7240	5420	379	9	-	-	-	[42]
Mine Dumps	Cabezo Rajao	5.4	4.5	8	12.5	332	41	315	88.2	1508	[34]
Tailing ponds of mine wastes	Llano del Beal	7.84	1.25	4650	12772	72	31	-	-	-	[43]
Tailing ponds of mine wastes	La Unión	3.4	4.1	4199	2473	95	6	190	10.23	1052	[44]
Tailing ponds of mine wastes	El Llano del Beal	4.9	2.5	1422	7539	265	28	637	96.2	147	[45]
Tailing ponds of mine wastes	El Gorguel	3.45	10.76	5190	8406	235	9	650	30.41	-	[46]
Deposits	Portman (YS)	1.66	2.35	2978	2415	35	0.7	178	29.05	-	[46]
Deposits	Portman (BS)	2.2	9.6	5086	54,366	233	71	4610	22.12	2264	[45]
Deposits	LoPoyo	5.3	16.8	8317	50,405	645	74	3627	21.12	3668	[45]
Watercourses (stream sediments)	W. Llano del Beal	6.8	1.9	1972	5882	141	31	572	20.16	3114	[45]
Watercourses (stream sediments)	W. Portman	4.4	2.5	1954	5223	195	19	550	10.04	2850	[45]

Table 3 shows the average mineralogical compositions of different SM sites, grouping chlorite, mica, kaolinite and greenalite as phyllosilicates and copiapite, bianchite, rozenite, etc. as hydrated sulphates forming efflorescences [38].

Table 3. Mineralogical characteristics of the abandoned residues of the study area [38].

		Mineralogical Composition (%)												
		fsp	**phy**	**qtz**	**gp**	**jar**	**gt**	**hm**	**ak**	**py**	**alu**	**ca**	**dl**	**hs**
Mine Dumps	El Gorguel	9	4	15	15	26	4	3	8	4	-	-	-	5
Deposits	Lo Poyo	18	2	31	10	11	4	4	12	8	1	-	-	5
Tailing ponds of mine wastes	Llano del Beal	23	3	23	8	17	6	8	14	3	-	2	-	6
Mine Dumps	Cabezo Rajao	18	5	22	17	14	4	3	11	2	1	-	-	6
Deposits	Portman Bay	14	7	21	9	23	6	5	14	3	2	-	-	8
Watercourse (stream sediments)	El Beal	19	3	32	5	9	4	4	13	5	-	22	2	2
Watercourse (stream sediments)	Portman	17	9	32	6	6	13	6	7	2	-	3	-	2

Fsp = feldspar; phy = phyllosilicate; qtz = quartz; gp = gypsum; jar = jarosite; gt = goethite; hm = hematite; ak = akaganeite; py = pyrite; alu = alunite; ca = calcite; dl = dolomite; hs = hydrated sulphates.

In general, there are differences between the materials from the different sources, which are related to the degree of supergene alteration, the degree of contribution of the mined material and the presence of volcanic and/or carbonate rocks. A higher content of soluble sulphates and jarosite can be related to materials from ponds with a high degree of alteration, the presence of greenalite with gangue from volcanic rock mineralisation (trachytes–andesites) and iron oxyhydroxides with a more evolved state of alteration [39].

Figure 5 summarises the main supergene alteration processes affecting these materials, their implications for biogeochemical cycles and the resulting products [29,35–38,47–65].

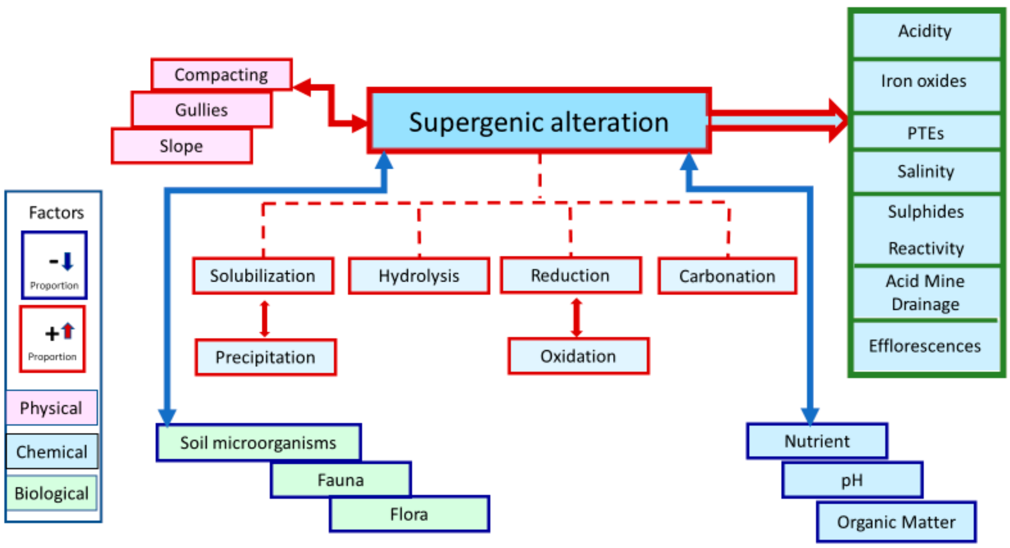

Figure 5. Processes involved in the mining tailings deposits.

The minerals neoformed in these processes can retain different amounts of PTEs and release them when environmental conditions change. Natural or potential mobilisation studies are based on the use of sequential and selective extractions using different media that simulate different environmental conditions [35,36,40,66].

The most commonly used selective extraction media are water (rain), citrate-ditionite (reducing and complexing medium) and HNO_3, H_2O_2 and ammonium acetate (oxidising medium). Selective extractions are used less often and refer to the BCR procedure. The PTEs also vary according to the mineral composition of the sample, but in general, Zn is the most mobilisable element, followed by Pb; As the least mobilisable. The mineralogical composition and crystallinity, presence of soluble salts and granulometry, are the most important factors affecting the mobilisation of PTEs [37,39,40,66].

5. Mining Contamination Transfer Routes

Areas containing abandoned mining deposits often lack restoration projects and have a poor, or even total lack of, plant cover protection, which makes them very vulnerable to erosive processes, with a high risk of contaminant dispersion. The study area is located in an arid and/or semi-arid climate zone, where torrential rains are usually the most common form of precipitation; this means that a strong erosive action (water and wind) is exerted on the ponds and deposits of mining waste, depending on the degree of compaction of the materials.

The degree of affectation is a function of the distance to the contamination source and is influenced by topographical and climatic factors, such as location (hillside, plain, river network, etc.), prevailing winds, water courses, rainfall regime, etc. Figure 6 shows aspects of the degrees of local contamination that occur in the SW and NE area of the SM [53].

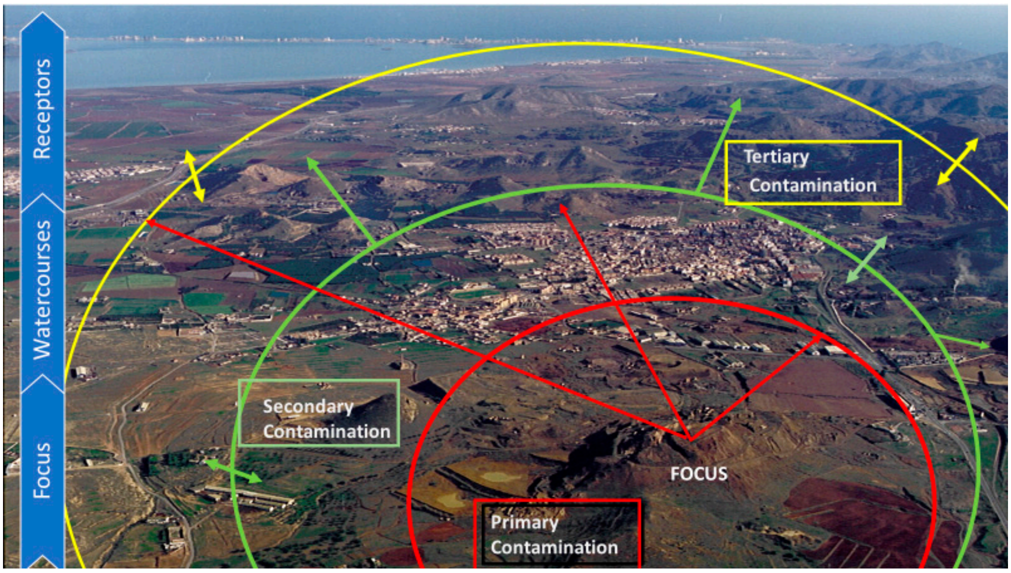

Figure 6. Focus and levels of contamination in the study area.

5.1. Water Dispersion

The dispersion of pollutants through water erosion of abandoned mining waste has been extensively studied. For example, after a period in which 60.5 mm of rain fell [19], a soil loss of 2.14 kg/m^2 was quantified for one of the mining deposits in the study area.

Ramblas (temporary watercourses) are identified as the main routes of surface dispersion and PTE transfer, especially in the Mar Menor area, where concentrations similar to those determined in mining waste piles may occur. In addition to material erosion, the runoff triggers the generation of acid mine drainage (AMD) water. The acidic waters of mine drainage are mainly characterised by their high content of dissolved metals and sulphates, acidic pH, and suspended particles [34,61,66–71]. The concentration of iron has been seen to vary from 56 to 558 mg L^{-1}, and sulphate can reach > 20,000 mg L^{-1} [58]. In internal areas of the Sierra Minera and in the vicinity of the ponds, PTEs can reach high concentrations, with values of As > 100 mg L^{-1}, Zn > 5000 mg L^{-1} and Pb > 400 mg L^{-1}.

The acid mine drainage waters enable chemical alteration of minerals (including sulphides with dispersion of PTEs), mainly due to their acidic pH, and represent one of the most important impacts generated by the intense activity was carried out for more than 2500 years in the District of Cartagena-La Unión. The resulting environmental problems are considerable.

Sources that generate acid drainage include leachates from dumps, tailings ponds, drainage from underground galleries, short or open pit mines, residues of treatment and concentration plants, and any material that has sulphides susceptible to oxidation [36,38,55,66,72].

The following general reaction is established for the formation of acid mine drainage:

$$FeS_2(s) + \frac{7}{2}O_2\ (g) + H_2O \rightarrow Fe^{2+} + 2SO_4^{2-} + 2H^+ \tag{1}$$

$$Fe^{2+} + \frac{1}{4}O_2(g) + H^+ \rightarrow Fe^{3+} + \frac{1}{2}H_2O \tag{2}$$

$$Fe^{3+} + 3H_2O \rightarrow Fe(OH)_3(s) + 3H^+ \quad pH > 3.5 \tag{3}$$

$$FeS_2 + 14Fe^{3+} + 8H_2O \rightarrow 15Fe^{2+} + 2SO_4^{2-} + 16H^+ \quad pH < 3.5 \tag{4}$$

Pyrite (FeS_2) is an iron sulphide of particular importance in metal mining areas. It is often found in mining waste, being the most common sulphide in these mining areas. Pyrite oxidation is the main cause of acidic drainage [40,42,43,60–62]. Other sulphides present also react and form new minerals, but only those containing Fe (II) contribute to the formation of AMD (Figure 7) [66,73–76].

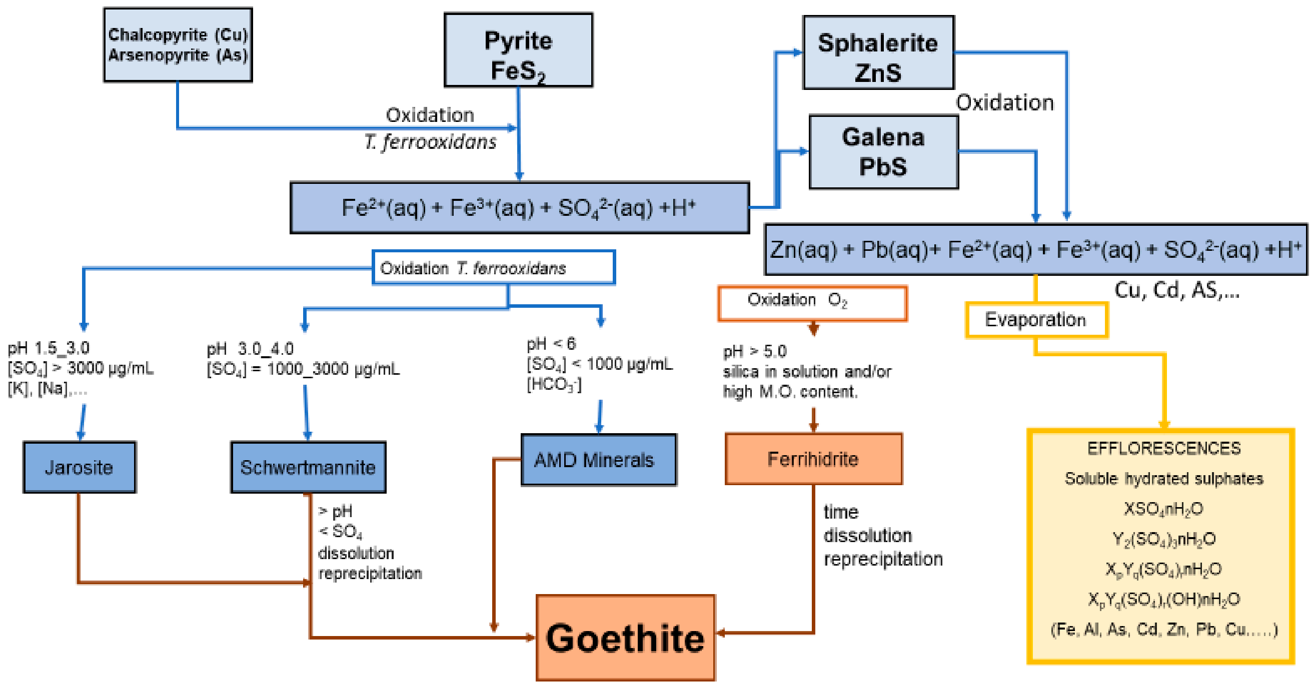

Figure 7. Biogeochemical model of supergene alteration of polymetallic mining (PSG).

Table 4 summarises the average values obtained from rambla waters at four strategic points in the SM. In all cases, these are waters with acid pH and high PTEs and sulphate content, which highlights the importance of AMD production from the materials in the area [67].

Table 4. Average concentrations of PTEs (mg/L, except As in μg/L) in superficial water samples [28].

	pH	Zn	Pb	Cd	Fe	As	Cu	Mn	SO_4
Lo Poyo	3.0	11	8.6	5	19	21	12	5040	61,326
El Llano del Beal	3.0	9883	9.5	7	7	14	14	2099	32,224
Cabezo Rajao	2.1	905	5.8	100	5900	11,676	13	175	53,650
Portman-Gorguel	1.9	430	0.6	16	1860	15,319	76	270	21,555

5.2. Wind Dispersion

Another contamination factor present in the Sierra Minera is the impact of air on the contaminated areas that are devoid of vegetation; further erosion is suffered as a result of the action of wind. These areas are subjected to the full intensity of the wind, which is a key factor in determining wind erosion risk. The characteristics of flotation sludge ponds of fine granulometry and the oxidation of sulphides into sulphates increase their vulnerability to being eroded by wind action. The very-low-density crusts that form in sulphide oxidation processes can be eroded by wind and transported over long distances (Table 5) [77]. This phenomenon occurs throughout the year, but especially in the summer months, when the dehydration of the crusts is total [29,78–80].

Table 5. Particulate matter and heavy metals in the atmospheric aerosol of the study area: average values over a period of 9 years [77].

Site	TPS ($\mu g\ m^{-3}$)			Pb ($\mu g\ m^{-3}$)			Cu ($\mu g\ m^{-3}$)		
	Mean	**Standard Deviation**	**Number of Measurements**	**Mean**	**Standard Deviation**	**Number of Measurements**	**Mean**	**Standard Deviation**	**Number of Measurements**
P1	118.8	±75.1	1634	1.01	±2.81	1927	0.03	±0.02	309
P2	200.1	±86.5	1639	0.69	±0.64	1947	0.05	±0.04	322
P3	88.5	±36.6	1601	0.25	±0.23	1885	0.03	±0.03	303

Site	Zn ($\mu g\ m^{-3}$)			Cd ($ng\ m^{-3}$)		
	Mean	**Standard Deviation**	**Number of Measurements**	**Mean**	**Standard Deviation**	**Number of Measurements**
P1	1.36	±0.89	309	10.03	±19.05	309
P2	2.10	±1.43	322	9.23	±9.30	322
P3	1.54	±1.19	303	5.45	±4.77	303

The presence of saline efflorescences is very important, due to the high concentration of heavy metals and metal trace elements they contain. Their impact, too, is closely related to wind erosion because their very fine texture and low density characteristics confer on them a high degree of erosionability. They are also closely related to the generation of acid drainage water (Figure 7) [50,51,53,55,60,61,81–84].

6. Receptors

Three areas adjacent to the study zone that are influenced by mining contamination are identified in this paper. Some work and results obtained are outlined in the following sections.

6.1. Campo de Cartagena

The reception areas include the arable flood zones of the Campo de Cartagena, where the PTEs are deposited when the flow of water slows down; both soluble and particulate forms may even reach the Mar Menor lagoon in times of torrential rains.

In the vicinity of the Mining Zones there is a large expanse of limestone soils, with a long agricultural tradition. This area is dedicated to open-air horticultural crops and greenhouses and is greatly favoured by the climate close to the Mediterranean Sea. These soils, which are located in the flood area of the watercourses, have levels of heavy metals that can sometimes be very high, with a large spatial dispersion of values. In the area feeding these watercourses, which form short and discontinuous water channels when it rains, there is a large contribution from lateral flows consisting of materials belonging to the mining area [85].

6.2. Mar Menor Lagoon

The transport of PTEs in the watercourses from the SM to the Mar Menor lagoon after rainy episodes is very important and has been extensively studied. This was highlighted in 1973 by Simonneau [86], who considered the Mar Menor lagoon as a mining site. The study concludes that this aquatic system is affected by the entry of PTEs from adjacent areas of the SM. In the same sense, the recent promulgation of Law 3/2020 of 27 July [20] on the recovery and protection of the Mar Menor lagoon mentions that runoff or infiltration phenomena resulting from old, unrestored mining areas through the drainage systems that make up the watercourses allow sediments and PTEs to reach the Mar Menor lagoon.

6.3. Mediterranean Sea

The impact on the Mediterranean side of the SM caused by past mining activity should also be mentioned. It is estimated that 25 Mm^3 of sludge has been dumped into the Mediterranean Sea, forming artificial beaches, such as Portman Bay with an area of 0.8 km^2 and Gorguel Bay (0.2 km^2) [29]. The accumulation of mining wastes with very high concentrations of PTEs in the vicinity of the Mediterranean Sea is so great that it has come to be considered a contamination black spot.

7. Transfer

To learn more about mobilisation of the PTEs contained in abandoned mining waste, several research studies and analyses have been carried out to determine the concentrations of metal contaminants in the organisms of the receiving environments, which include both flora and fauna.

7.1. Transfer to Flora

Plant species have been found in the abandoned mining area of Cartagena-La Unión that support high concentrations of metals in their rhizosphere and are considered hypertolerant. Figure 8 presents a summary of naturally growing plants in the critical area studied [28,33,36,38,42,44,49,58,87–100]. The same figure also differentiates between species that have high concentrations of PTEs in their roots but do not transfer them to their aerial part, or do so in very low concentrations (Bioconcentration Factor (BF) > 1; Transfer Factor (TF) < 1), and other species in which PTEs are translocated to their green and/or edible parts (TF > 1). However, it should be noted that there is no consensus and that the behaviour of plant species may differ depending on the area where they grow [90] and also on the heavy metal or trace element being studied.

Species such as *Dittrichia viscose* and *Zygophyllum fabago*, among others, are of particular interest, having a high BF and low TF, which makes them highly recommendable for phytostabilisation programmes, although others such as *Atriplex halimus* or *Helichrysum decumbens*, with a high TF, are discouraged for such programmes. [100] found that Cymodocea nodosa in the Mar Menor accumulates metals from the sediments and emphasised the bioavailability of this seagrass.

Species selected for phytostabilization programs must be tolerant to high concentrations of heavy metals in the soil and to acidic and hypersaline environments. In addition, the species used will not be those used for phytoextraction, which at the end of their function of extracting and accumulating heavy metals, will be uprooted and treated as hazardous waste because of their high PTE contents, but will be species that will be part of the restoration project and remain in the rehabilitated area. They must be species with a low or no translocation rate, especially to their aerial part, thus preventing the transfer of PTEs to the surrounding fauna, with the associated danger of entry into the trophic chain and toxicity to humans.

7.2. Transfer to Fauna

Research was carried out in the study area to determine the extent to which PTEs are transferred to the fauna that inhabit the mining-influenced areas. For example, Auernheimer et al. [101] determined that the shells of *Cerastoderma edule* and *Veneryis aurea* contain higher concentrations of Pb (influenced by Rambla del Beal) than found in samples of the same species in the northern section of the Mar Menor (Lo Pagan) and control samples. [102] found a gradient of toxicity with depth in tests carried out with sea urchin larvae *(Arbacia lixula* and *Paracentrotus livids*) and amphipods (*Gammarus aequicauda* and *Microdeutopus grillotalpa*) in Portman Bay.

Other studies have also been carried out: Marín-Guirao et al. [103] found a certain level of disturbance in the benthic invertebrate communities. Cervantes et al. [52] determined that the content of PTEs significantly increases in the soft tissue of *Hexaplex trunculus* when their concentrations in sediments increase. Dassenakis et al. [104] observed contamination by heavy metals in the flesh of the mollusc *Ostrea edullis* due to the contaminated ecosystem of the Mar Menor lagoon. Navarro García et al. [105] studied several tissues (bone, feather, kidney, liver and muscle) of large juvenile examples of *Phalacrocorax carbo sinensis*, found dead in the Mar Menor, and found high levels of Cr and Mn in the feathers, while the kidney and liver had the highest accumulation of Se and other metals. The same authors concluded that the use of *Phalacrocorax carbo sinensis* is suitable for heavy metal biomonitoring. Campos-Herrera et al. [106] carried out a study with nematodes, in which the authors suggest that the high content in Pb in soils affected by mining activity decreased the soil's biodiversity and that these taxa were especially useful as biological indicators of soil mining contamination. Muñoz Vera et al. [62] used two species of jellyfish to determine the bioavailability of trace elements in the Mar Menor and observed extremely high levels of bioconcentration of metals such as Al, Ti, Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, Sn and Pb.

7.3. Transfer to Crops

If heavy metals and trace elements are transferred to crops, there is a potential risk of PTEs entering the trophic chain. Aukour et al. [108] conducted a comparative study of the parameters of crop soils that had been exposed to heavy metals and soluble salts, such as those in the Campo de Cartagena, and other soils of similar soil characteristics that had not been thus exposed—in this case the soils of northern Jordan. Interesting positive correlations between rhizosphere and root were reported for Pb, Cd and Zn, with low values for TF and a very high sensitivity that prevented development for some plant species such as tomato.

An experimental study in a greenhouse to model the processes that can take place in restored soils of mining origin, using several plant species grown for consumption, including baby lettuce, leek, onion, broccoli, alfalfa, iceberg lettuce and chard, has been reported [94,109]. The author used a natural soil and Technosols (with different proportions of mining soil and other materials such as plant soil, construction waste and limestone filler). It concluded that contamination studies are needed for agricultural land use influenced by mining contamination to reduce the level of exposure of As in the diet. It resembles the effect of Technosols, with limestone filler and RCDs, with a natural attenuation of contamination that exists in mining sites when there are soils with calcium carbonate and clay.

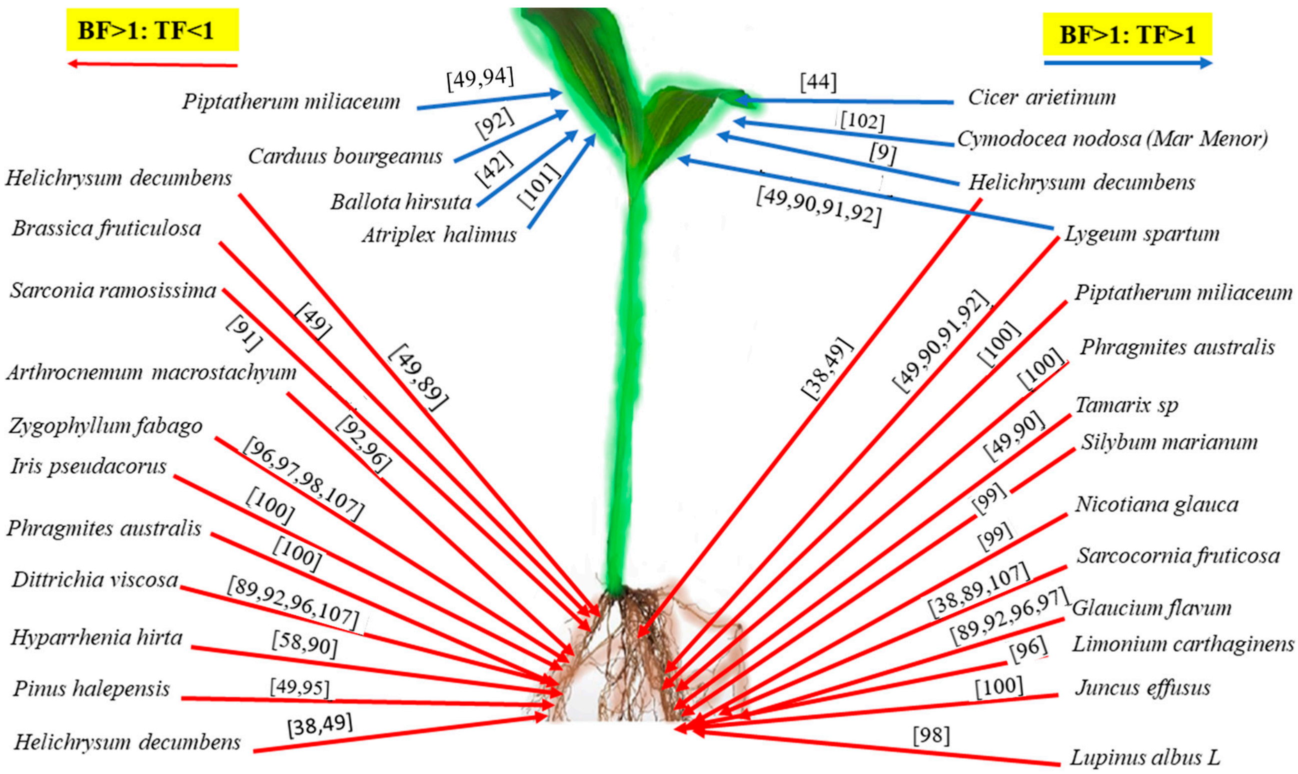

Figure 8. BF and TF of the main plant species related to the study area [9,38,42,44,49,58,87–90,92–100,107].

8. Risks

Several researchers have pointed out that an assessment of the total concentration of a pollutant is not sufficient for the determination of toxic effects or for the characterisation of contaminated sites. They suggest that other methods, such as biological methods, as a complement to chemical methods should be used in any determination of the risks posed by contaminants.

8.1. Risks to Land Ecosystems

To evaluate the entry of arsenic (As) into the trophic chain, Caparrós Ríos et al. [91] studied the transfer of arsenic to the trophic chain via the plants and fauna that inhabit the soils contaminated by heavy metals of abandoned mining areas. To do this, they used 165 soil samples and the same number of plants, while sheep and mice represented mammals. By studying bioconcentration and displacement factors, it was seen that As transfer depends on the plant species and soil characteristics (pH, content in carbonate, metal concentration). The authors concluded that plants such as *Dittrichia viscose* do not pose a risk of As intake when consumed by the fauna studied and can therefore be recommended for soil recovery programmes. Other species such as *Arthrocnemum Macrostachyum* and *Glaucium flavum* can be used but with caution. In addition, the use of *Helichrysum decumbens*, *Carduus bourgeanus*, and *Lygeum spartum* for these purposes is discouraged. It is concluded that a phytoremediation programme should study the contribution of As that a plant makes to the diet of animals. This indicator can be used to select suitable plant species.

8.2. Risks to Aquatic Ecosystems

Ecotoxicity bioassays have been used by researchers, such as those cited, below for the evaluation of potentially toxic elements of the Sierra Minera and the Mar Menor; the good correlation found with other methods for determining acute toxicity confirms that bioassays are suitable tools for assessing toxicity.

García Lorenzo et al. and Pérez Sirvent et al. [36,110] used bioassays (Microtox, Phytotoxkit and Ostracodtoxkit) to assess the toxicity of the Sierra Minera and its area of influence and concluded that toxicity tests can be used for screening soils contaminated by mining activities and that a mixture of sediments with limestone filling could be applied for their remediation.

Benhamed et al. [111,112] studied the effect of metals from contaminated sediments in Portman Bay (black sands and yellow sands) on samples of gilt-head bream (*Sparus aurata*). The results showed that differences in the expression of genes from antioxidant enzymes and genes related to the immune system vary depending on the organ and genes studied. The authors concluded that biomarkers studied in gilt-head bream could be useful for assessing the impact of contamination by mining waste in coastal environments.

In a study of PTEs entering the Mar Menor, Martínez López et al. [113] found that the As, Fe and Mn present in the sediments enter the lagoon through the watercourses and are susceptible to mobilisation, so that they could be incorporated into the aquatic environment if there are changes in environmental conditions (such as acidification, oxidation or reduction).

8.3. Risks to the Human Population

There are in vitro assays, such as extractions in the gastrointestinal tract, which are performed by chemical extractions that simulate the gastrointestinal conditions of the stomach in humans and with which the bioaccessible fraction can be determined [114]. Such studies have shown that direct exposure to soil, including soil ingestion, skin absorption and inhalation exposure, is an important route of intake of potentially hazardous trace elements by humans and, particularly, children. These in vitro methods are comparable to in vivo studies but are faster and cheaper and thus are highly recommended for calculating bioaccessibility. According to the analysed literature, this technique has been widely used by researchers from the soil contamination group of the University of Murcia [35,36,38,60,85,90,115–118]. In vitro methods have also been used by several authors to determine the bioavailability elements in the soil following the use of element immobilisation techniques in the recovery of contaminated soils [119–121].

The determination of extractable metals, together with a number of other parameters, form the basis of risk analysis in soils. This involves the identification, measurement and comparison of various parameters through which the potential and real risks that contaminated soils may pose for humans can be identified and evaluated, suggesting possible scenarios for current and future uses. Four stages are contemplated: (1) hazard identification, (2) analysis of contaminant toxicity, (3) exposure analysis, and (4) risk characterisation [122].

Arsenic is a carcinogenic metalloid, and therefore the risk of its accidental intake should be evaluated when studying areas with high levels of this element. Cesar et al. [102] studied the arsenic bioaccessibility of the mining-influenced soils of southeast Spain. They determined the potential risk of soil intake in two fractions (<2 mm and <250 μm), taking into account the possible uses of soils (residential/agricultural) and possible receptors (adults and/or children) as well as the properties and mineralogical composition of the soils. The results suggested that using of a conservative approach to calculate the Chemical Daily Intake (CDI) based on total concentrations of arsenic may overestimate the risk and thus lead to problems in the management of contaminated soils. The authors concluded that it is very important to consider the amount of bioaccessible As in the <250 μm fraction before allowing the use of land close to temporary watercourses for residential use near beaches or for agriculture.

Martínez Sánchez et al. [121] studied the implications of risk analysis for human health in decision-making for the risk management of contaminated soil and suggested that acceptable/unacceptable risk should be decisive in the selection of soil recovery technologies for a given use. The example they presented was that of Portman Bay. The results show that the receptors most affected by using the bay are children, the most important route of exposure being the intake of solid particles, given the characteristics of the material, followed by through the skin and inhalation. Recovery may be/is possible through the ad hoc manufacture of Technosols, depending on the risk detected.

García-Lorenzo et al. [107] studied the ecological risk posed by PTEs in the same bay and concluded from the results that in an alkaline medium, such as the intestinal phase that occurs in the process of digestion of humans, Zn and Pb are highly bioaccessible. For the stomach phase of digestion, i.e., that occurring in an acidic medium, the highest availability would be As, Cu, Fe and Pb.

9. Final Remarks

The bibliographical review that was carried out provides a diagnosis of the problems associated with the current situation in the mining district of Cartagena-La Unión and its adjacent areas, especially the Mar Menor lagoon. The main risks were also identified, following an assessment of evaluation of the impacts, threats, functions and socio-ecological services affected, which permits recommendations to be proposed for drafting a protocol for the action that should be taken for treating degraded soils in the current scenario.

There are a number of recommendations that would be appropriate for addressing the necessary methodology for the development of a protocol of action for the management of this critical mining area.

1. Identification of mining critical zone hazards.

The main hazards identified in the abandoned mining district of Cartagena-La Unión are contamination of SACs, sites of geological interest, heritage, the high concentration of PTEs, high reactivity, acidity and little vegetation of fine easily erodible material from abandoned wastes, acid drainage from mines, the presence of efflorescences, and landscape impacts.

In marine areas (Mar Menor lagoon and Mediterranean Sea), contamination may occur from SPAMIs and SCIs as well as flora and fauna.

In cultivated areas of the Campo de Cartagena, the soils with PTEs have a risk of transfer to crops.

In accordance with the results obtained, the characterisation of the study area has been sufficiently complete and adequate, with abundant and contrasted data available. However, for a restoration plan for the Sierra Minera, it would be advisable to obtain detailed maps and use integrated methodologies that allow prioritisation of the areas with the greatest problems. Another aspect that is underdeveloped in the consulted research is the use of risk indicators and pollutant dispersion models, which in certain cases can be useful for the management of these areas.

2. To prevent or limit the transfer of PTEs.

Any contaminated soil management strategy should prioritise actions that limit and reduce the transfer of PTEs to crop areas, aquatic systems and population centres located in areas of influence. Such actions, in our case, must be directed at the headwaters of the watercourses.

3. An appropriate risk analysis and assessment.

The management of soils contaminated with PTEs will depend on risk analysis and assessment, with PTE mobility and bioaccessibility studies, rather than the total concentration of PTEs. Risk analysis is presented as a fundamental tool that allows zoning for the management of affected areas. It represents a new management paradigm for decision-making in the implementation of recovery/rehabilitation/decontamination projects at the local level, following priority orders established based on the risk and land uses.

4. To avoid or decrease the main impacts of altered areas.

To limit or slow down the main impacts of these altered areas, vegetation is essential. Phytostabilisation actions are a priority in restoration projects, and plant species adapted to the difficult conditions of these areas should be used.

5. To select a suitable management strategy for decision-making.

There has been much research, providing a sufficiently large body of results to establish a management strategy that addresses the problems of the Abandoned Mining Critical Areas of the Sierra Minera. However, it is clear that the whole area and its area of influence has been already well characterized. Consequently, it does not seem necessary to conduct any more specific research or to test more methodologies, since the extensive research carried out to date has yielded good results for in situ application.

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