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Decontamination and Circular Economy of Dredged Material and Mining Waters Using Adiabatic Sonic Evaporation and Crystallization (ASEC) Technology

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Abstract: Dredged material is a common environmental and economic issue worldwide. Tons of highly contaminated material, derived from cleaning the bottoms of bays and harbours, are stored until depuration. These volumes occupy huge extensions and require costly treatments. The Ria of Huelva (southwest Spain) receives additionally high metal contamination inputs from the Odiel and Tinto Rivers which are strongly affected by acid mine drainage (acid lixiviates with high metal content and sulphates). These two circumstances convert the port of Huelva into an acceptor/accumulator of contamination. The current study proposes an alternative active treatment of dredged material and mining residues using ASEC (Adiabatic Sonic Evaporation and Crystallization) technology to obtain distilled water and valuable solid conglomerates. Different samples were depurated and the efficiency of the technology was tested. The results show a complete recovery of the treated volumes with high-quality water (pH~7, EC < 56 µS/cm, complete removal of dissolved elements). Also, the characterization of the dried solids enable the calculation of approximate revenues from the valorization of some potentially exploitable elements (Rio Tinto: 4 M, Tharsis: 3.7 M, dredged material: 2.5 M USD/yr). The avoidance of residue discharge plus the aggregated value would promote a circular economy in sectors such as mining and dredging activities.

Keywords: dredged material management; environmental remediation; circular economy; critical raw materials; Adiabatic Sonic Evaporation and Crystallization

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1. Introduction

Sediments are dredged from water bodies to ensure proper water levels or restore aquatic ecosystems worldwide. This dredging activity produces large volumes of sediments that reach the land and need adequate disposal management. According to SedNet (the European Sediment Network), Europe annually produces more than 200 million m^3 of dredged materials [1]. Dredged sediments are commonly disposed of in open oceans or landfills. However, traditional methods are restricted by legal and environmental concerns. On the one hand, open ocean disposal is banned in several countries due to its potential contamination of the surrounding environment [2,3]. On the other hand, landfilling is associated with a high demand for space and the production of risky by-products [4]. However, both methods face restrictions due to legal and environmental concerns. One example is the European Landfill Directive (1999/31/EC) [5], which has caused some landfills to close, resulting in a reduction in the space available for landfills and higher landfilling costs [6].

A recent assessment (2008–2014) of European dredged material by the OSPAR (2017) [7] estimated that 20–40% of dredged material [8] is highly polluted by human activity, mainly those related to fine grain size [7]. Incorrect management would adversely affect the ecosystem, so strict regulations enforce that the material must be treated before the sediments can safely be sent back into the environment or landfilled. Current treatments are not cost-efficient; they have high energy consumption and do not effectively separate water from contaminated solids that must be transported to landfills with a very high moisture content (>30%), which increases transport and disposal costs exponentially [9,10]. More importantly, these technologies do not allow solids and water cost-efficient valorization, which might bring important environmental and economic benefits (circular economy) [11]. As a result of the high treatment costs and the lack of treatment in some highly contaminated cases, many coastal and fluvial areas need to stop dredging, which may have a negative economic impact on the area (e.g., reducing the number and draught of ships the port can accommodate) [9,12].

However, several environmental, economic, social and technological factors determine the viability of implementing the proposed practices. Social aspects require attention, particularly the end-user acceptance of using sediments. Investment and operational costs are also barriers because using sediments in industries could require new sources of income for the development of technologies, adjustments and upgrades of existing practices, as well as the implementation of cleaning-up technologies, among others [13].

The composition of the dredged material is a decisive factor in determining the viability of using sediments for beneficial purposes. Typically, sediments contain organic matter, sulphides, chlorides, nutrients, organic compounds and trace elements [11,14–16]. When the concentration of toxic compounds (metal(loid)s) exceeds the concentration limits of the regulations, it is necessary to decrease environmental concentrations. Furthermore, compounds with a high value on the market could potentially be recovered to be commercialized [17,18]. Hence, sediment component extractive technologies are strongly needed to enhance and promote beneficial uses [19].

Elevated metal concentrations in sediments are a crucial problem, due to their persistence in the sediments and potential negative effects on aquatic organisms [20,21]. Metals such as Sn, Cu and Zn, and organotin compounds such as tributyltin (TBT) are often found in elevated concentrations in aquatic sediments [22–24]. These contaminants and especially metals emanate from natural and anthropogenic origins. Industrial pollution sources include shipping (including paints) and harbour facilities, agriculture, mining, and chemical factories [25]. Metal pollution in sediments poses an extended environmental issue worldwide because the toxicity of elements affects public health and natural ecosystems [26,27]. Investigating metal extraction technologies for sediments is crucial to decrease the amount of metal pollution released into the environment. The obtained elements could potentially be recycled in metallurgical industries, contributing to reintroducing the material into production cycles instead of landfilling valuable resources [28]. Other treatments include stabilization and solidification methods, which enable the use of dredged materials in construction (concrete materials, construction products, roadway construction, habitat building, landfill liner/cap, etc.) and are a common strategy internationally [29–31].

Huelva province, located in the southwest of Spain (Figure 1), is a mining region with ancient mining activity (since Roman times) due to the massive sulphide composition of the territory. This particular composition (pyrite- FeS, arsenopyrite -FeAsS, chalcopyrite- $CuFeS_2$, galena- PbS, spharalite- ZnS, among others) promotes the natural and forced formation of acid lixiviates from mining residues and minerals exposed to oxidation. These acid lixiviates, known as acid mine drainage (AMD), are highly acidic fluids with high concentrations of dissolved metals (and metalloids) and sulphates which flow into the water courses of the Rio Tinto and Odiel River basin and eventually reach the Huelva

Estuary, where the Port of Huelva lies. Therefore, mining contributes significantly to the input load of pollution in this region [32–34].

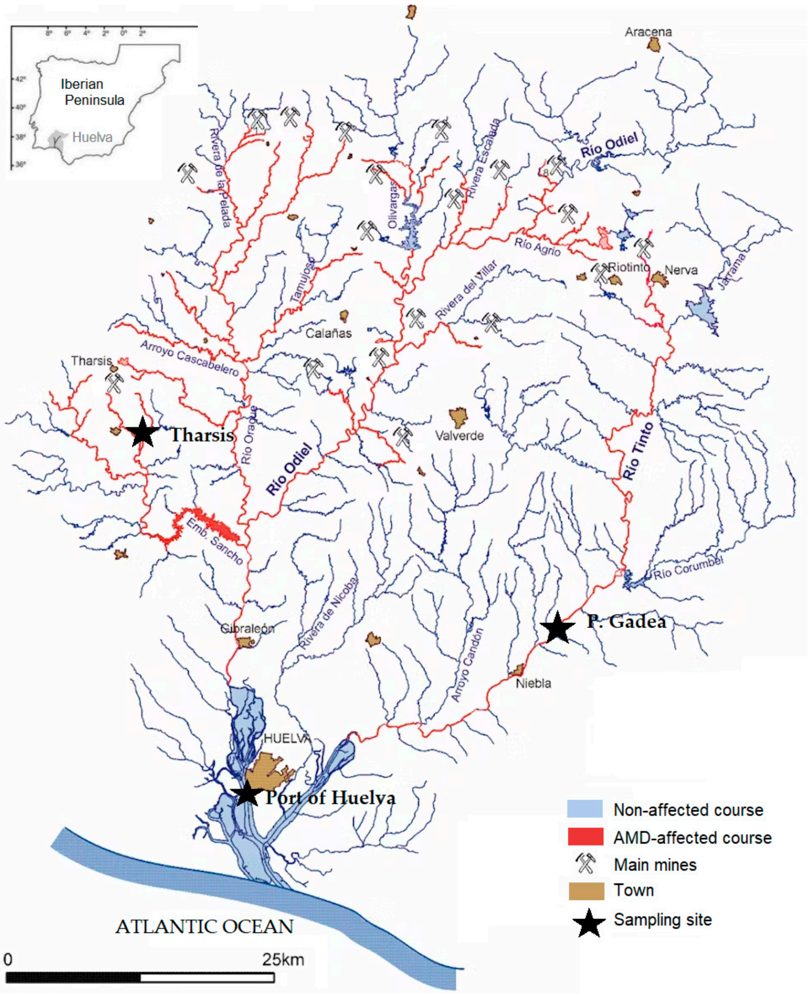

Figure 1. Odiel-Rio Tinto River watershed in the province of Huelva (southwest Spain), the main affected courses according to [35], and the location of the sampling points (Tharsis mine, Puente Gadea/Rio Tinto River and the Port of Huelva-supplier dredged material).

The Port of Huelva is on the southern Atlantic coast of the Iberian Peninsula (Figure 1). The port underwent its most recent major transformation in the 1960s. At that time, there was a sharp upsurge in the development of the chemical and oil industries, with the creation of its industrial zone and the growth of the port towards the exterior, beyond the Punta del Sebo. At present, bulk solids and liquids are the main goods used by the Port of Huelva. In 2005, port traffic had a turnover of 21 Mt. Since the inception of the Junta Especial de Comercio y Puerto de Huelva (Special Trade Board and Port of Huelva) in 1874, it was considered essential to dredge in the sand bar area and the inner harbour, and these tasks were undertaken and are still carried out today.

Dredging activities can be classified into the following two types based on their aims: dredging of the first order, to make the navigable zones deeper or wider, and maintenance dredging, to alleviate the effects of the periodical silting-up processes. In recent years, major dredging work of the first order has been undertaken.

The two most outstanding actions were the widening and deepening of a channel in 1995 when approximately 6 Mm^3 were extracted, and the activities carried out in 2000 and 2001, when a volume approaching 4 Mm^3 was removed, this being carried out to improve safety conditions for the larger methane carriers entering the port. Maintenance dredging has proved necessary because the sediments that build up within the harbour water amount to an average dredging volume of about 0.5 Mm^3 per year.

Sediment pollution is mainly associated with fine particles [36]. In the case of the Port of Huelva, the contamination of certain zones by the materials lying on the bed of the Ria can be attributed to the following causes [37]: (i) Natural origins, due to phenomena such as leaching and sediments being washed down and discharged by two mining-impacted rivers such as the well-known Rio Tinto and Odiel rivers. (ii) Ancient anthropic origins, owing to mine workings in the upper basins of these two rivers. (iii) Recent anthropic origins, due to industrial discharges into the river from industrial zones and loading and unloading operations.

As previously mentioned, the main source of metals, metalloids and most of the critical raw materials (CRMs) in the sediments of the port of Huelva come from mining activity in the area through the Rio Tinto and Odiel rivers [38] and in some cases, exceed the alert levels of CEDEX [39]. Knowledge of the hydro-geochemistry of metal(loid)s, rare earth elements (REE) and other CRMs in AMDs and their distribution using normalized patterns would help discriminate the most potentially marketable AMD sources. Particularly, the Rio Tinto and Tharsis mines were catalogued as the most promising facilities with the highest recovery potential [40].

Due to high annual metal loads in the Iberian Pyrite Belt (IPB, where the Port of Huelva and its associated ecosystems are located) of elements such as Al (6600 ton), Zn (1600 ton), Cu (600 ton), Co (26 ton), Ni (10 ton), LREE (10.7 ton/yr), MREE (2.1 ton/yr), HREE (1 ton/yr), Y (3.7 ton) or Sc (0.7 ton), AMDs of the IPB would have an economic potential of 24.1 MUSD/yr (e.g., REE being 22.6% of this potential) [41]. So, the Ria of Huelva might be considered as a reservoir of CRMs, although technical and economic limitations would impose a more realistic value of 4.2–10.3 M USD/yr. The magnitude of this economic potential cannot be compared with active mines; however, the longevity of the AMD generation processes and the need to achieve an environmental improvement render the valorization of these leachates an interesting option to recover metals, and other CRMs, which would help treatment plants costs, notably improving the quality of the dredged material in the Port and its dredging activities, as well as the water bodies in abandoned mining sites.

On the other hand, wealth is not only an economic measure but also includes other immaterial considerations such as the economy's long-term sustainability. These considerations are included in the World Bank's new wealth indicators and include educational investments, natural resources depletion, and ecosystem degradation. Furthermore, it also includes the restoration of the ecosystem, which has a significantly greater impact than other revenues. Ecosystem services play a strong role in social consciousness and human well-being, and despite not being included in public finance, they directly affect the environment positively (e.g., quality of water, greenhouse gas emissions, etc.) [42]. Ecosystem services are estimated to cost up to 16–54 trillion USD yearly for the entire biosphere.

The main objective of this study is to assess the efficiency of ASEC in purifying contaminated fluids from dredging activities, the dredging itself, and mine waters that are the source of contamination in the dredging area (Ria of Huelva). As a secondary objective, the study aims to visualize the scope of a complete circular economy including the commercialization of water and dried solids as possible CRMs.

2. Materials and Methods

2.1. Sampling Collection and Analytical Procedures

Figure 1 shows the water sample location from mining areas affected by AMD: at Puente Gadea (Rio Tinto river tributary), and the immediacy of Tharsis Mine (Odiel River basin). Samples of dredged material, characterized as highly contaminated and managed as waste products, were provided by the port authorities of the Port of Huelva. Some aliquots of samples were collected in plastic bottles and transported under cool and dark conditions to laboratories where they were analyzed; their physicochemical parameters were determined as outlined in [43].

2.2. Adiabatic Sonic Evaporation and Crystallization Technology (ASEC) Depuration

Water Challenge has patented an energy-efficient (<20 kWh/m^3) Zero Liquid Discharge (ZLD+) technology that purifies fluids, delivering freshwater and crystallized solids known as ASEC (International Patent: EP3135635). It is an environmentally friendly solution that does not require chemicals or consumables, which reduces pollution and preserves biodiversity through a one-step process for the total separation of solids that are dissolved or undissolved in water, achieving 100% clean water (EC < 200 µS/cm) and dry solids. Further details of the design and technology used can be found in [43–45], where it was tested with other industrial fluids, including mining waters. Briefly, it uses a physical approach to purify the contaminated fluids, obtaining fresh water and dry solids by promoting adiabatic changes and the acceleration of the input fluids. In this paper, dredged material and acid mining waters were used as input fluids. The energy consumption is the total amount of power required for internal pumping and the heat exchanger. It depends on the energy source consuming around 19 kWh/m^3 if only an electrical power supply is used. If a residual heat source can be used, then only about 3–4 kWh/m^3 is needed to boost the water in the system circuit. Furthermore, it will reach almost 0 kWh/m^3 if the ASEC system is run using a hybrid solar thermal–photovoltaic system.

A volume of 50 L of the dredged material samples and acid mining fluids was collected in the field and treated using ASEC technology. This yielded almost 50 L of pure water and crystallized solids, which were collected as outlined in [44].

2.3. Sample Characterization

The elemental composition of the water samples from the sampling points supplied to the ASEC system (input), besides the water samples collected after the treatment (output), were analyzed using Inductively Coupled Plasma-Mass Spectroscopy (ICP-MS, Thermo Electron Corporation©, X-series II, Thermo Fisher, Karlsruhe, Germany). Analyses were carried out at certified laboratories at the ICMAN/CSIC (Spain).

On the other hand, the dried solids and crystallized salt samples collected as the output of the ASEC technology were semi-quantitatively characterized using X-ray fluorescence spectrometry (XRF, in a Panalytical Epsilon 1 model spectrometer equipped with an Ag target X-ray tube and a Be detector window that can be operated at a maximum voltage of 50 kV) and X-ray diffraction (XRD, using a Panalytical X'Pert Pro diffractometer with CuKα radiation over an angular range (2θ) from 10° to 90°). Acid digestion of the same aliquots enabled the determination of the multi-elemental composition using ICP-MS at the ICMAN/CSIC laboratories [44].

3. Results

By-Products Characterization

The ASEC treatment of the three samples resulted in the complete removal of the dissolved elements from the liquid samples yielding pure water and crystallized solids with a moisture content of <1% (Figure 2). The whole volume of liquid sample treated was collected at the output of the ASEC system as clean/transparent water for each one of the samples. On the other hand, solids were also collected as dried conglomerates (Figure 2). The solid samples were weighted as 27.3 g/L of dried solids for the Rio Tinto river sample, 30.21 g/L for the Tharsis sample, and 68.73 g/L of dried solids for the dredged material sample.

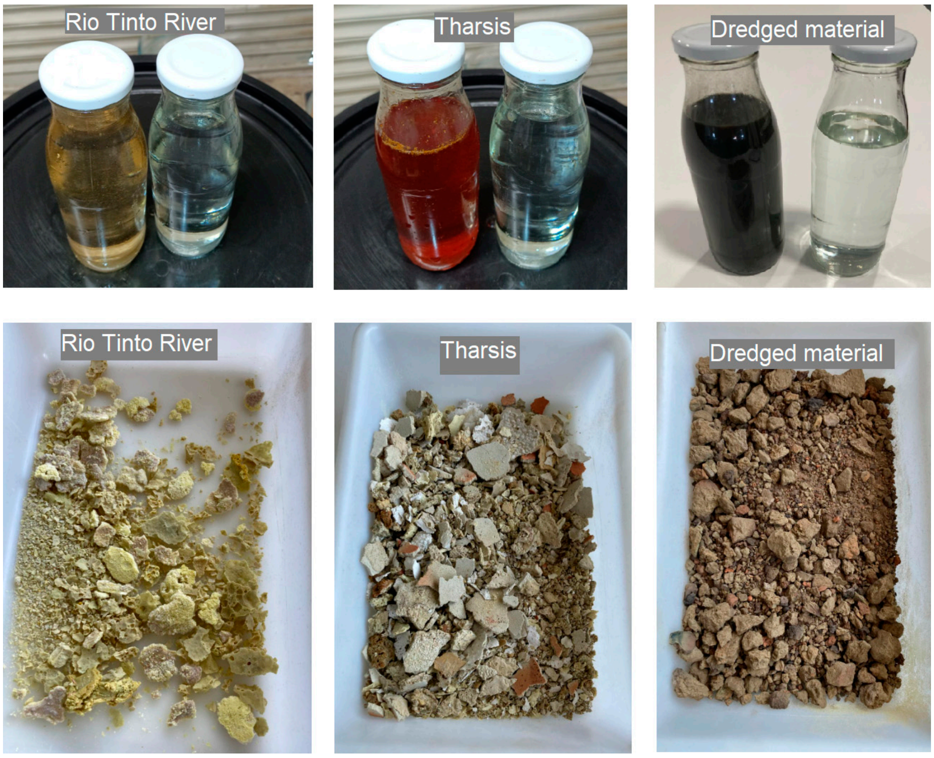

Figure 2. Collected samples (**top left**) and the treated fluid (**top right**) from the Rio Tinto river, Tharsis mine and the dredged material. Below are the results in crystallized solids after the treatment with ASEC technology.

There was a recovery of almost 100% of the injected volume of each sample (50 L) as transparent water, as shown in Figure 2. The depurated obtained water showed properties similar to pure water: pH~6.5–7, EC < 56 µS/cm, TDS< 0.03 mg/L, and the complete removal of the dissolved elements (Table 1). In the case of the mining residues ($I_{RioTinto}$, $I_{Tharsis}$), there was a sharp rise in pH, from 3.70 and 3.15 to pH values of typical pure water 6.69 and 6.79, respectively. The electrical conductivity (EC) decreased in magnitude from mS to µS, from 2.5 mS/cm for $I_{RioTinto}$ and 12 mS/cm for $I_{Tharsis}$ to values of EC below 55 µS/cm. The high concentrations of acidity, sulphate, metals and metalloids (such as Fe, Al, Cu, Zn, As, Co, Cd, Ni), and REE in these mining samples are the resultant product of the intense sulphide oxidation processes.

Table 1. Physicochemical properties of samples (Rio Tinto River, Tharsis mine, and dredged material) before (Input: I) and after (Output: O) treatment with ASEC technology.

			Input			**Output**	
		$I_{RioTinto}$	$I_{Tharsis}$	$I_{Dredged}$	$O_{RioTinto}$	$O_{Tharsis}$	$O_{Dredged}$
Ph		3.70	3.15	7.27	6.69	6.79	7.82
EC	µS/cm	2450	12,140	52,300	55.5	35.8	40.4
TDS	mg/L	1516	7530	32,400	0.034	0.022	0.025
T	°C	10.1	15.0	11.9	9.1	14.2	10.3
Li	ppb	177.1	1356	166.9	n.d.	n.d.	n.d.
B	ppb	255.1	92.9	5367	n.d.	n.d.	n.d.
Na	ppm	41.65	67.44	12.90	n.d	n.d	n.d
Mg	ppm	88.14	1.29	1609	n.d.	n.d.	n.d.
Al	ppb	85,070	806,164	59.6	n.d.	n.d.	n.d.

Table 1. *Cont.*

		Input			**Output**		
		$I_{RioTinto}$	$I_{Tharsis}$	$I_{Dredged}$	$O_{RioTinto}$	$O_{Tharsis}$	$O_{Dredged}$
K	ppm	6.0	4.6	500	n.d.	n.d.	n.d
Ca	ppm	4.3	20.2	50.8	n.d.	n.d.	n.d.
Cr	ppb	912	298	n.d.	n.d.	n.d.	n.d.
Mn	ppm	8.4	134	1.07	n.d.	n.d.	n.d.
Fe	ppm	162	2844	12.6	n.d.	n.d.	n.d.
Co	ppb	547	9348	4.5	n.d.	n.d.	n.d.
Ni	ppb	93.0	4076	1.4	n.d.	n.d.	n.d.
Cu	ppb	116,239	186,798	16,340	n.d	n.d	n.d
Zn	ppb	18,173	367,775	1108	n.d	n.d	n.d
As	ppb	2593	5821	470.7	n.d.	n.d.	n.d.
Se	ppb	2403	1511	1481	n.d.	n.d.	n.d.
Ag	ppb	12.7	11.9	11.8	n.d.	n.d.	n.d.
Cd	ppb	90.3	967.0	n.d.	n.d.	n.d.	n.d.
Ba	ppb	46.5	347.9	104.6	n.d.	n.d.	n.d.
Pb	ppb	199.0	80.1	n.d.	n.d.	n.d.	n.d.
Sr	ppb	179.3	224.5	10,127	n.d.	n.d.	n.d.
La	ppb	27.3	270.4	2.8	n.d.	n.d.	n.d.
Ce	ppb	66.5	795.0	n.d.	n.d.	n.d.	n.d.
Pr	ppb	14.9	112.1	n.d.	n.d.	n.d.	n.d.
Nd	ppb	44.6	495.3	n.d.	n.d.	n.d.	n.d.
Sm	ppb	19.2	149.3	n.d.	n.d.	n.d.	n.d.
Eu	ppb	9.9	39.4	n.d	n.d	n.d	n.d
Gd	ppb	19.7	173.3	n.d.	n.d.	n.d.	n.d.
Tb	ppb	8.4	30.5	n.d.	n.d.	n.d.	n.d.
Dy	ppb	17.1	137.2	n.d.	n.d.	n.d.	n.d.
Ho	ppb	7.2	29.6	n.d.	n.d.	n.d.	n.d.
Er	ppb	11.7	69.6	n.d.	n.d.	n.d.	n.d.
Tm	ppb	6.3	12.9	n.d.	n.d.	n.d.	n.d.
Yb	ppb	10.1	56.8	n.d.	n.d.	n.d.	n.d.
Th	ppb	24.5	732.3	100.8	n.d.	n.d.	n.d.
U	ppb	62.4	320.9	9.7	n.d.	n.d.	n.d.
Y	ppb	86.5	1801	n.d.	n.d.	n.d.	n.d.
Mo	ppb	n.d.	51.3	n.d.	n.d.	n.d.	n.d.
Au	ppb	n.d.	0.9	0.48	n.d.	n.d.	n.d.

n.d.: not detected.

In the case of the dredged material sample ($I_{Dredged}$), the pH was not as low (7.27), but the O sample showed a slightly increased pH of 7.82. Instead, the strong EC of 52.3 mS/cm and the TDS of 32,400 mg/L decreased sharply to 40.4 µS/cm and 0.025 mg/L in the treated water, but also removal of all studied elements was observed in the $O_{Dredged}$.

The mineralogy of the collected solid samples from Rio Tinto, Tharsis and Huelva was analyzed using XRD (Figure 3). The elemental composition of the solid by-product from the Rio Tinto river mining sample after treatment of the water with ASEC technology was 41% S and 35% Fe. These elements appear as hydrated sulphates of Zn, Cu and Al [46] thoughout XRD (Figure 4).

A similar composition was observed for solids collected after the treatment of the Tharsis sample, where the major composition was iron sulphates, and other oxyhydrosulphates, such as Mn, Ca, Zn or Al, but some iron oxides were also detected (Figure 4).

The AMD solid particle sample examination showed that trace elements are included within a matrix of large trace element-rich Fe sulphate particles and Fe-Mn oxides [46] (Figure 4).

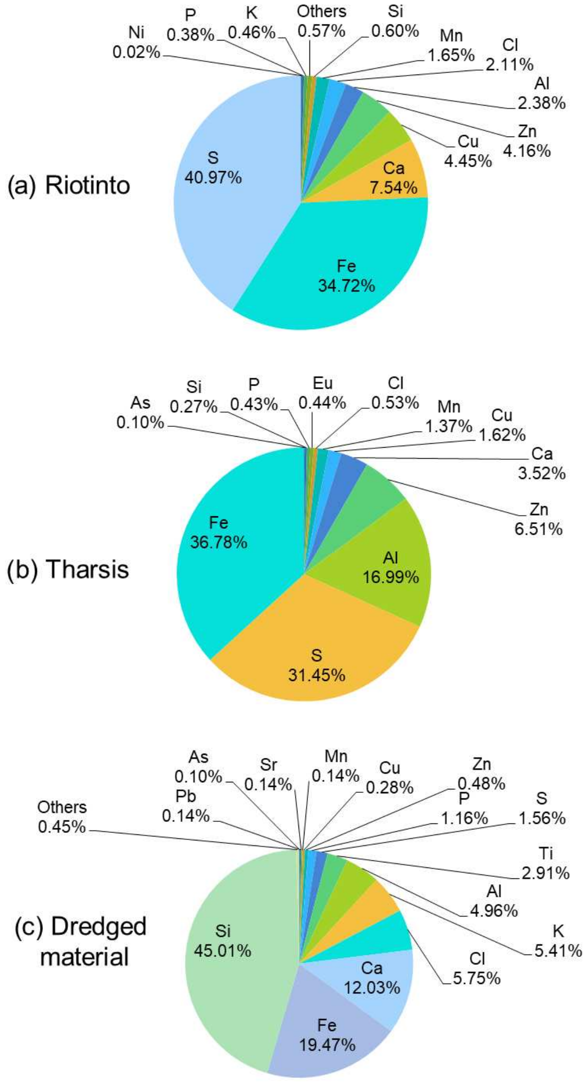

Figure 3. Elemental composition (in %) of the crystallized solid samples from Rio Tinto (**a**), Tharsis (**b**) and dredged material (**c**) after ASEC depuration.

Meanwhile, the dredged material mineralogy is dominated by iron-bearing silicates, where some toxic elements (Cu, Zn, Ni, etc.) (Figures 3 and 4) are preserved by weathering and soil forming processes as less reactive and mobile [47].

Some elements found in elevated concentrations, such as Cu, Cd, Zn or As, are mainly associated with discharges of the Odiel-Rio Tinto watershed due to mining activity [47]. But the binding of these elements also depends on the pH of elements (Table 1), e.g., for low pH, iron and S are associated with As and Pb. In contrast, the estuarine mixing processes promote the precipitation of Fe and Al oxyhydroxysulfates (pH < 6), so contaminants (Cu, Cd, Zn) are associated with S [48]. The main mineral formations found were anhydrous sulphate of Zn, and other sulphates of Cu, Al, and Fe in the Rio Tinto solid sample. In the Tharsis sample, Fe-sulphides prevailed, along with anhydrous sulphate of Fe, oxyhydroxide of manganese (manganite), sulphate of Zn, and Cu sulphides, among others (Figure 4).

SiO_2, Al_2O_3 and CaO are the main chemical compositions of dredged material determined by various authors [11]. In the case of sediments from the Ria of Huelva, apart from the industrial activities (acid mining), there are further salt-induced physical mixing processes and muddy deposits, bioturbation, and urban wastes from the vicinity of Huelva. Contamination input also stems from harbouring activities, the presence of phosphogypsum deposits (from the fertilizer industry), and phosphate carbonates rich in Fe storage [49] that determine a different composition domined by iron-oxide and iron titanate as trace in the obtained dried solid samples (Figure 4).

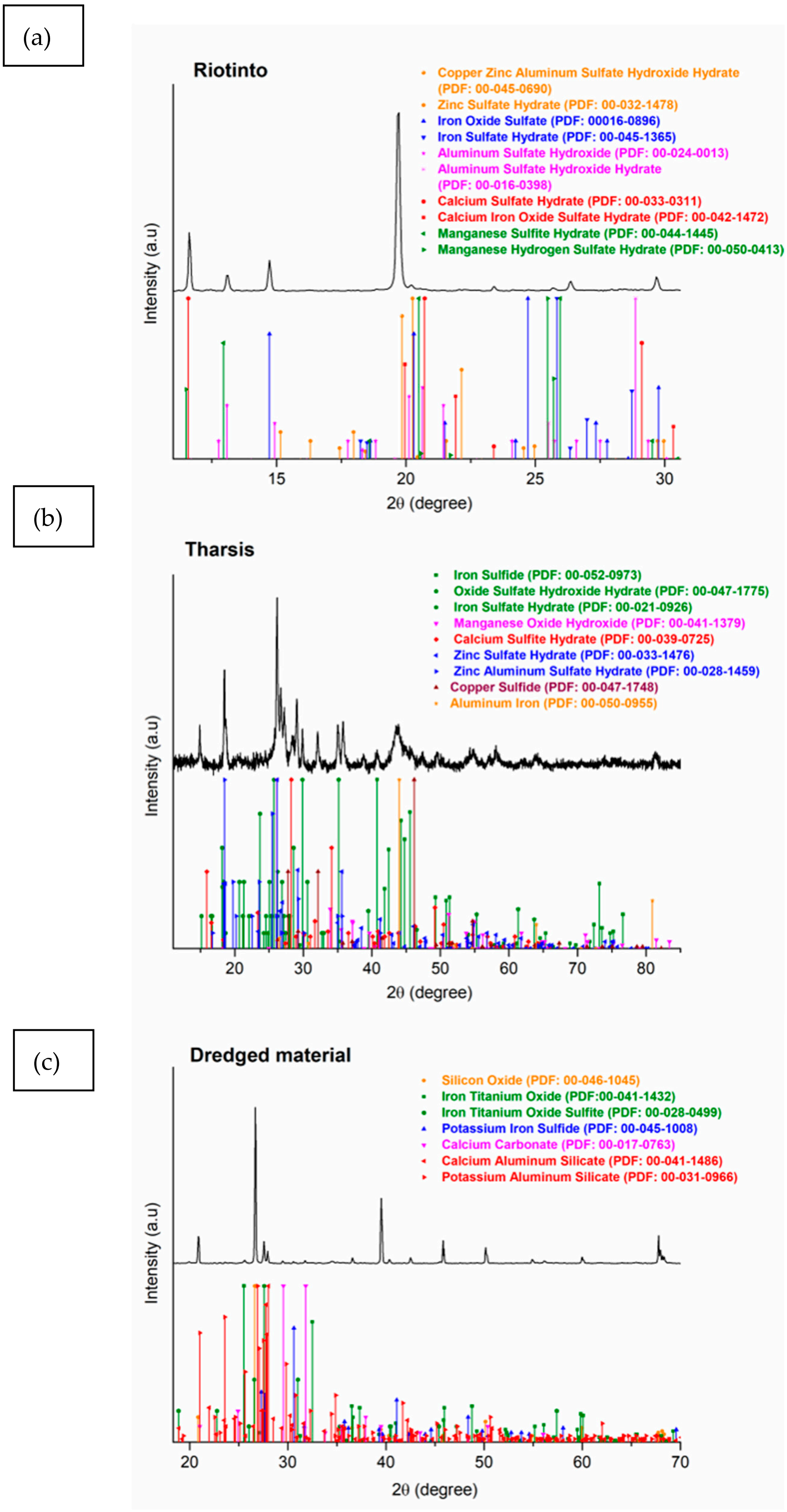

Figure 4. X-ray difraction results of the crystallized solids from Rio Tinto (**a**), Tharsis (**b**) and dredged material (**c**). Correspondence between coloured peaks and composition found in samples.

4. Discussion

The handling of dredged sediments depends on the level of contamination and local regulations, as specific restrictions, costs and treatment requirements may apply [39]. The most common management practices for dredged sediments include landfilling and deep-sea disposal [5], and metal extraction and natural recovery [49]. However, increasingly stricter environmental legislation affects both the availability of landfilling space and the cost of utilizing it. One example is the European Landfill Directive (1999/31/EC), which has caused some landfills to close, resulting in a reduction in the space available for landfill, and higher landfilling costs [5]. Consequently, stakeholders involved in dredged sediment management are increasingly motivated to investigate alternatives to landfilling. Stabilization and solidification methods, which enable the use of dredged materials in construction, is a common strategy internationally [50]; however, its use is limited in different countries, due to geological conditions (soft clays), high salinity and limited knowledge. Stabilization may be more sustainable when combined with a method where metals are extracted from the sediment before it is stabilized. The extraction of metals from sediment can be performed using biological, physical or chemical techniques, which can either be used independently or combined [3,50]. However, ASEC technology obtains solids with humidity below 1%. For the current experiment, the annual production of solids would generate 99.6 tons of dried solids for the treatment of Rio Tinto, 110 tons for Tharsis, and 250 tons for dredged material treatment. The stabilization is instantaneous without the addition of reagents (cost-effective). This is translated as a reduction in storage and transport costs, but it also offers an advantage to concentrate the dissolved elements for secondary mining processing (re-exploitation). In addition, this removal implies the cessation of pollution discharge into aquatic systems, thereby enhancing the ecosystem services provided by these aquatic bodies. Apart from the production of cleaner sediment, another potential benefit of metal extraction is the opportunity to recover valuable metals, thereby reducing the need for mining. Once elements have been extracted from the sediment, the environmental risks of residual sediments are reduced, and management criteria are more likely to be met [51]. Included in these elements are the CRM and other elements of commercial interest, so the extractive process of solid crystallized salts might present a dual economic advantage in terms of re-mining and re-exploitation of resources alongside removal.

As an overview, based on some elemental concentrations found in the solid samples' by-products and the current market price [40,41,52–55], an estimation of the maximum economic potential value was calculated as annual gross production considering an interrupted depuration cycle working for a daily treatment of 10 m^3, with 100% of the extracted element recovered. But also, the estimation accounted for a reduction in extractive expenses and partial recovery (70%), and energy consumption expenses (0.13 USD/kWh for a system of 20 kWh/d) were discounted to estimate the net production obtained (Table 2).

Table 2. Estimation of the quantities of elements extracted as solid crystals (tons per year) and the annual economic revenues (kUSD) obtained after depuration of the three samples along a year for a minimum treatment of 10 m^3 ASEC system (the system could be scaled to thousands of m^3 per day if necessary).

	Annual Production (kg)				**Annual Potential Revenues (kUSD)**		
	Rio Tinto	**Tharsis**	**Dredged Mat.**		**Rio Tinto**	**Tharsis**	**Dredged Mat.**
Al	2422	3005	3074	Al	5.409	6.710	6.864
As	11.62	1.91	29.00	As	2.022	0.332	5.045
B	1.217	3.058	7.614	B_2O_3	3.652	9.173	22.84
Ba	0.244	2.227	27.682	Ba	0.000	0.000	0.005

Table 2. *Cont.*

	Annual Production (kg)			Annual Potential Revenues (kUSD)			
	Rio Tinto	**Tharsis**	**Dredged Mat.**		**Rio Tinto**	**Tharsis**	**Dredged Mat.**
Be	0.009	0.007	0.007	Be	0.009	0.008	0.007
Bi	0.000	0.034	0.637	Bi	0.000	0.001	0.013
Ca	41.87	204	417	$CaCO_3$	13.06	63.54	130
Cd	3.09	3.26	0.15	Cd	0.008	0.008	0.000
Ce	1.87	2.60	7.77	Ce	0.007	0.010	0.029
Co	31.39	23.83	2.46	Co	1.543	1.171	0.121
Cr	1.01	1.44	5.95	Cr	0.008	0.011	0.048
Cu	274	647	106	Cu	2.323	5.481	0.901
Dy	0.382	0.479	0.541	Dy	0.169	0.212	0.240
Er	0.203	0.244	0.271	Er	0.005	0.006	0.006
Eu	0.080	0.102	0.171	Eu	0.023	0.029	0.049
Fe	8444	7062	6656	Fe	887	742	699
Ga	0.21	0.24	1.19	Ga	0.160	0.181	0.898
Gd	0.44	0.57	0.78	Gd	0.011	0.015	0.020
Ho	0.07	0.09	0.10	Ho	0.004	0.005	0.006
In	0.10	0.07	0.05	In	0.054	0.039	0.029
K	-	121	545	KCl	-	60.48	272.30
La	0.719	1.022	4.633	La	0.003	0.004	0.017
Li	4.11	6.70	5.92	Li	0.054	0.088	0.078
Mg	4035	3469	1128	$MgCl_2$	3228	2775	902
Mn	385	353.8	50.83	Mn	12.0	11.06	1.59
Na	123	1121	1434	NaCl	7.64	69.52	88.93
Nd	1.19	1.71	4.12	Nd	0.150	0.215	0.520
Ni	12.87	4.78	2.58	Ni	0.209	0.078	0.042
Pb	-	10.81	44.40	Pb	-	0.023	0.096
Pd	74.6	73.6	174	Pd	0.100	0.098	0.232
Pr	0.250	0.363	0.998	Pr	0.026	0.038	0.104
Se	0.937	1.521	2.024	Se	0.024	0.039	0.052
Sr	-	6.156	28.049	Sr	-	0.083	0.379
Sn	161	1489	17,957	Sn	4.05	37.51	452.28
Sm	0.331	0.473	0.830	Sm	0.004	0.006	0.011
Tb	0.067	0.084	0.100	Tb	0.110	0.138	0.164
Th	0.072	0.242	0.851	Th	0.005	0.018	0.062
Tl	0.006	0.162	0.104	Tl	0.047	1.307	0.840
Tm	0.018	0.023	0.020	Tm	0.925	1.167	1.020
Yb	0.167	0.195	0.219	Yb	0.002	0.003	0.003
Zn	953	550	132	Zn	2.317	1.339	0.322
Total (kg)	16,987	18,170	31,853	Total(kUSD)	4171	3787	2588

Therefore, after a year of depuration of mining residues with ASEC treatment (10 m^3/day), the Rio Tinto River samples depuration would provide 4 million USD in potential revenue and 3.7 million USD in potential revenue from Tharsis samples depuration. In the case of dredged material depuration with the ASEC system, the potential revenue would reach 2.5 million USD for the selected elements. After discounting expenses that decrease the extraction ratio and considering other impurities, the revenue calculations for the selected elements would be between 0.76 and 1.2 million USD (a significant increment might positively affect these numbers). The study of Rosario-Beltré et al. [40] also supports the cost-effective secondary mining of residues from Rio Tinto and Tharsis mine residues to obtain a potential gross profit above one hundred USD based on the recovery of Ag, Zn, Pb, Sb, Cs, Bi and As.

When analyzing the number of elements collected in the dried solids, heterogeneity can be observed in the composition. Significant amounts of Fe (6.6–8.4 ton), Al (2.4–3 ton) and $MgCl_2$ (1.2–4.0 ton) might be obtained as a dried solid conglomerate together with other elements for the three samples. But a significant amount of Sn (18 tons) would be obtained from dredged sediments, compared to mining residues (0.16–1.4 tons). However, according to the market price, the greatest revenues considering the high pre-concentrated amounts would be obtained as $MgCl_2$ (between 0.92 and 3.2 million USD). Iron is also a great contributor with revenues between 700 and 885 kUSD for the three samples. A second group of elements would also be potential economic target materials, such as B_2O_3 (22.84 kUSD), $CaCO_3$ (130 kUSD from dredged materials), KCl (272.30 kUSD), or Sn (452.28 kUSD).

Zinc sulphate is used by agrochemical industries as fertilizer, but also as animal feed and even toothpaste. As significant amounts of Zn sulphate are found in solid samples from Rio Tinto, it might be considered more economically valuable than the individual element.

Some other trace elements catalogued as CRMs and technology metals, used as catalyzers, would be concentrated in lesser amounts, but with higher economic impacts in re-exploitation, such as Tm, Mn, Gd, Dy, Ga, Tb, Be, In, etc. (Table 2).

The primary impact of the ASEC depuration process is the complete purification of water with a quality exceeding that required for direct or indirect consumption (Table 1) and the avoidance of pollution discharges into the environment.

According to Norén et al. [50], the more polluted the site is, the greater the potential new revenue is. Sites such as the Odiel-Tinto River watershed and other highly metal-polluted bays might be considered ore deposits due to their high concentrations of elements with commercial interests. But also, market values will impact the decision to re-mine elements from solids. A secondary advantage of the reuse of these residues as metal resources is the facility of crush due to the very low degree of humidity and the small particle size. The removal of toxic elements from dredged material decreases the harmful charge of the sediments facilitating easier and cheaper the management, either for landfilling, deep-sea disposal or natural recovery [50]. On the other hand, the application of ASEC as a recovery method provides a monetary and environmental contribution to a circular economy treatment of residues from the Huelva and Ria rivers. The study of Crocetti et al. [56] widely reviews the traditional and new operation management of dredged sediments, including the characterization and regulations of European countries and some innovative projects. But it is also aware that management is moving towards a circular economy (sustainable, environmentally friendly, energy and cost-efficient, and low carbon and water footprint) that includes by-products but also impacts the sediment management scheme.

Despite the economic profits that might potentially be obtained from mining and dredged material depuration, there are some other ecosystem services that would strongly benefit, for example, water regulation and water supply by providing important volumes for irrigation or industrial processes; waste treatment by removal of contaminants from aquatic ecosystems; increase of raw materials by revalorizing the by-products of dried solids.

By applying ASEC to different industries, it would impact other global ecosystem services such as climate regulation (a decrease in GHG emissions for a reduction in the productive cycle of minerals and transport) and gas regulation and even food production by supplying products such as fertilizers and pesticides. According to [42], human well-being is affected by the interaction between social, human, built and natural capital. The ecosystem services are a required bridge between natural capital and human well-being, which is reached through a transdisciplinary approach.

5. Conclusions

The Ria of Huelva was selected as a case study to test the industrial fluid depuration efficiency of the ASEC technology due to its strong mining contamination originating from the Odiel-Tinto River basin. Sediment from the port of Huelva derived from dredging activities and two heavily acid/metal polluted sites of the upstream rivers was collected and treated with the active system to obtain pure water and dried solids. The current study has demonstrated the complete efficacy of the removal of dissolved elements and the neutralization of pH from both residues (dredged materials and mining residues) located in the Huelva province: from pH ~3 to pH 6.5–7. The complete recovery of a high-quality water volume represents an environmental milestone in the restoration of aquatic ecosystems. On the other hand, the obtained dried solids represent, based on the characterization results, a new source of critical raw materials and other elements of commercial interest for re-exploitation (especially in terms of mining residue treatment by-products) providing important potential revenues (~1.2 million USD), as well as a reduction in transport and management of dredged material costs. A new alternative has been presented here, which incorporates dredged materials into the circular economy by reutilizing the high-quality water obtained in the process, along with the proper management of the solid phase (moisture < 1%), obtaining potentially high revenues (~700 kUSD, data with 70% discount) and offering traditional construction material alternatives.

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