Journal of Hazardous Materials 466 (2024) 133529

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

A novel heuristic tool for selecting the best upgrading conditions for the removal of potentially toxic elements by soil washing

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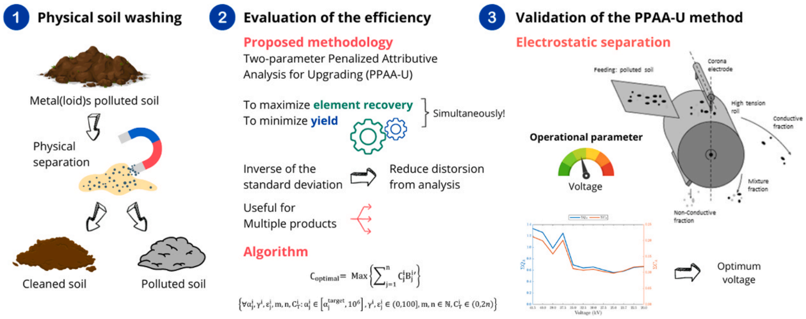
HIGHLIGHTS

- We introduce PPAA-U for soil-washing optimization.
- PPAA-U maximizes recovery and minimizes concentrate yield.
- PAAA-U penalizes experimental contributions with high variability.
- This methodology yields consistently good results.
- PPAA-U has many potential applications in other separation technologies.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Soil pollution
Soil remediation
Optimization
Circular economy

ABSTRACT

Here, we propose two-parameter penalized attributive analysis, PPAA-U, a novel heuristic tool for selecting the best upgrading conditions (BUCs) for soil washing. Given a multi-component feed and a specific set of operating conditions, PPAA-U generates a quality index based on how well recoveries for key components are maximized while minimizing the yield. We demonstrate, through the calculation of families of curves, that this quality index is related linearly to recovery and to the inverse of the yield, meaning that reducing yield values is more important than maximizing recovery. To evaluate our method, electrostatic separation at 12 different voltages was carried out on soil samples from an ex-industrial site in Spain. Values of recovery, yield, and grade were analyzed using basic attributive analysis and PPAA-U with and without target-to-distance correction. Both methods identified the same optimal separation voltage, and the power of PPAA-U to correct for high variation in yields and recoveries was observed as a divergence between results produced by each method at low voltages where variation in these values was greatest. PPAA-U thus offers a convenient tool for soil washing optimization, and we suggest that it could be applied successfully to other industrial processes.

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https://doi.org/10.1016/j.jhazmat.2024.133529
Received 11 October 2023; Received in revised form 11 January 2024; Accepted 12 January 2024

1. Introduction

Many industrial processes lead to the accumulation of potentially toxic elements (PTEs) in soils, and the need to remove them is driving research into soil remediation techniques in several countries [1,2]. Existing soil remediation technologies include various physical, chemical, and biological methods [3,4]. Originally developed in the mining industry to obtain metal concentrates from mineral ores, physical separation technologies have been used for soil remediation in the case of both organic and inorganic pollutants [5-7]. Physical separation can be used to remove potentially toxic elements (PTEs) from soil either directly, where PTEs are present as discrete particles, or, since many PTEs are strongly absorbed by clay, by separating the fraction onto which they are preferentially sorbed [8-10]. Although physical soil washing can be a terminal process, it is usually followed by chemical soil washing [11,12].

Physical soil remediation shares many common processes with mineral beneficiating and recycling. In all cases, the objective is to separate a concentrate of perhaps two or more target components from a multi-component feed. However, whereas in mineral beneficiation (and recycling), optimization may occur for either elements or mineral compounds, in the case of soil washing, we tend to be concerned only with elements.

The principal distinction between soil remediation and mineral processing (beneficiation or recycling) lies in the different economics of these processes. These considerations mean that the concentrate-to-tailings ratio achieved in mineral processing operations is generally closer to one than it is in soil remediation [13]. In mineral beneficiation, for example, the cost of further processing the concentrate (through the pyro- and hydrometallurgical routes) is offset due to the value of the final product [14,15]; however, for soil washing, a high initial concentrate-to-tailing ratio is crucial to avoid compromising the economic viability of the operation [16].

This means that, for soil remediation, in contrast to mineral beneficiation and recycling, concentrate yield minimization is the most important criterion. Moreover, in soil remediation, the washed fraction (tailings) must adhere to environmental standards in terms of grade for it to be declared decontaminated, whereas in mineral recycling and beneficiation, the grade of tailings is dictated more by economic considerations and may remain quite high provided that the process is profitable [17,18].

In any mineral separation process, whether this be a beneficiation or soil remediation process, improving performance implies increasing the concentration of a target element or compound in one of the process flows. Thus, the total mass of the initial material flow, known as the feed (F), is normally separated into two products, the concentrate (C) and the tailings (T), corresponding to the fractions in which the grade of the target element or compound is, respectively, higher or lower than that in the feed. A third fraction is sometimes collected, the middlings (M), which has a grade intermediate between that of the concentrate and the tailings.

The total mass balance is then [19]:

$$F = C + M + T \tag{1}$$

Dividing Eq. (1) by the mass of the feed gives the yield or weight recovery for each mass flow (Eq. (2)) [20]:

$$\frac{F}{F} = 1 = \frac{C}{F} + \frac{M}{F} + \frac{T}{F} \tag{2}$$

Of the three yields, that of most interest is the concentrate yield, $\frac{C}{F} = \gamma$, which contains the highest concentration of the target element or compound.

A parameter known as the grade or assay is used to indicate the proportion of the target element or compound in each of the mass flows. Its value for the concentrate is usually denoted as λ, and this value can be used as an assessment of the quality of the separation process. The grades for the feed, middlings, and tailings are denoted by α, β, and ϑ, respectively. In a successful separation process, the following inequalities should be true: $\lambda > \alpha$; $\vartheta < \alpha$; and $\lambda > \beta > \alpha$ [20]. The recovery, ε, refers to the mass of the target element or compound found in a given mass flow relative to the feed. Thus, for the concentrate fraction, recovery is defined as [20]:

$$\varepsilon = \frac{\lambda}{\alpha}\gamma \tag{3}$$

The same value for the tailings fraction is usually denoted as η, such that [20]:

$$\varepsilon + \eta = 1 \tag{4}$$

Intuitively, it would seem that ε alone could be used as a measure of separation performance due to its relationship to the parameters α, λ, and γ. However, this is not the case and, in fact, assessing separation performance requires consideration of not only ε but at least two of either α, λ, or γ. For example, $\varepsilon = 100\%$ may seem to imply perfect separation, but accompanied by high γ and low λ, this is clearly not so. Thus, if we wish to optimize a separation process, we must maximize ε and λ while simultaneously minimizing γ [20,21,19].

Bearing in mind the above discussion, the aim of this research is to develop a robust method for determining the best upgrading conditions for a given soil washing operation. Specifically, we will:

- Offer an exhaustive analysis of basic attributive analysis in terms of families of curves.
- Discuss the effect of experimental noise on the overall quality of a given experimental set-up.
- Show how attributive analysis can be modified to address this type of distortion.
- Provide a practical example of the use of attributive analysis.

2. Materials and methods

In this section, we discuss the sources of the soil samples and the separation technique used to demonstrate the practical application of attributive analysis. We then explain the principles of basic and penalized attributive analysis.

2.1. Sample preparation and analysis

The site of interest is the Linares mining district (Andalusia, Spain), a center for intense Pb mining, mineral processing, and metallurgical activities for several centuries [22,23]. Ten 2.5 kg samples were collected from the top 35 cm layer of soil at random points across the study site to form a bulk sample of 25 kg. The bulk sample was homogenized and wet sieved (water flow = 0.3 l/min) using sodium carbonate and sodium hexametaphosphate as dispersing agents to produce six granulometric fractions: 63 μm, 63–125 μm, 125–250 μm, 250–500 μm, 500–1000 μm, and 1000–2000 μm (ASTM D-422–63) [24]. Wet sieving continued until 3 kg of the 1000–2000 μm fraction was obtained. This fraction was then divided into thirty-six subsamples for electrostatic separation. Each of these subsamples was subjected to chemical analysis.

Subsequently, representative subsamples weighing 1 g each were extracted. These specimens were digested using "aqua regia" (a mixture of HCl and HNO_3) before analysis via inductively coupled plasma optical emission spectroscopy (ICP—OES) (HP 7700, Agilent Technologies).

Separation of the feed samples was achieved using an EHTP Outotec (Fig. S1) high-tension electrostatic separator. This advanced specification model is equipped with an AC wiper electrode operating at 18 kV and two DC electrodes—one corona and the other static—adjustable to a maximum of 41.5 kV, known as the separation voltage. Additionally, it features a grounded roller brush with interchangeable bristles and infrared roller heating for particle removal. Three fractions are collected: the nonconductive, intermediate, and conductive fractions.

The most important parameters in the separation process include the conductivity of the sample particles, the rotation speed of the roller, the placement of the electrodes, and the corona electrode tension. This enables the separation of materials based on their conductivity properties, making electrostatic separation an invaluable tool for many industrial and research applications.

Samples are loaded onto the roller via the feed hopper and travel towards the corona electrode. The air surrounding the corona electrode is ionized; thus, as the particles on the roller approach the corona electrode, they pick up charge. Conductive particles will lose their charge most rapidly; therefore, the roller's centrifugal force ejects these particles first, and they are collected in the conductors bin. More insulating particles keep their charge and remain on the roller until they are brushed off and fall into either the insulators or the middlings bin.

The apparatus was operated at 12 different separating voltages in a range from 20 kV to 41.5 kV. Separations were repeated three times at each separation voltage, and the results presented here correspond to the average values recorded for the three experiments completed at each voltage. A comprehensive description of the apparatus is provided in the Supplementary material section (SM1).

2.2. Basic attributive analysis

The basic model for attributive analysis was developed and applied to soil washing by Sierra et al. (2010) and Boente et al. [25]. Given the results of a number of soil-washing experiments using a particular separation technique, this method seeks to determine the set of experimental parameters that provides optimal separation. As discussed, this is done by seeking the conditions where the recovery of target elements is maximized while minimizing the yield.

Considering a set of *m* experiments to separate out *n* contaminating elements, the performance of a given experiment, *i*, with respect to target element, *j*, is expressed as a quality factor Q_j^i (Eq. (5)):

$$Q_j^i = \frac{\mathrm{Min}\{\gamma\}}{\gamma^i} + \frac{\varepsilon_j^i}{\mathrm{Max}\{\varepsilon_j\}} \tag{5}$$

where

- $i = 1, \ldots, m$ and refers to the results produced by a specific set of experimental parameters.
- $j = 1, \ldots, n$ and refers to results for a specific target element or contaminant; in this study, $m = 10$ (see Table 1 for all target elements considered).
- Q_j^i: Quality factor of experiment *i* for element *j*.
- γ^i: Yield of experiment *i*.
- ε_j^i: Recovery of element *j* in experiment *i*.

In the present study, the main experimental variable is the separation voltage; thus, $m = 12$, and ten target elements (the values of *j*) were considered (see Table 1). Table 1 presents the yields and recoveries of each element at each separation voltage tested; the values shown are an average of the results from three experimental runs at the same separation voltage.

As in the present study, there are generally numerous contaminants to consider, each of which has a specific target grade, that is, a safe threshold concentration after soil washing. Because some contaminants are significantly more toxic than others, each element to be removed during the soil washing operation is given a weighting coefficient related to its target grade, known as the target-to-distance correction. The sum of these coefficients must add up to 1; thus, we first define A_j^i (*Eq.*6) :

$$A_j^i = \frac{\alpha_j^i}{\alpha_j^{\mathrm{target}}} \tag{6}$$

Table 1

Yield (γ) for each experiment, grade (α), and recovery (ε) for each element in each experiment. Average of three experimental runs at each voltage with a standard error < 5%.

		As		Cd		Cr		Cu		Hg		Mo		Ni		Pb		Sb		Zn	
Voltage (kV)	γ (%)	α (mg/kg)	ε (%)	α (mg/kg)	ε (%)	α (mg/kg)	ε (%)	α (mg/kg)	ε (%)	α (mg/kg)	ε (%)	α (mg/kg)	ε (%)	α (mg/kg)	ε (%)	α (mg/kg)	ε (%)	α (mg/kg)	ε (%)	α (mg/kg)	ε (%)
41.5	8.6	57.8	38.9	11.3	37.8	11.6	35.4	523.5	62.2	1.7	14.4	4.0	81.0	12.3	57.3	2731.75	44.8	15.7	59.6	2375.37	81.5
40	9.0	56.5	41.7	8.7	45.0	10.2	38.8	610.7	74.9	1.5	13.2	3.6	79.7	10.5	53.7	2521.85	48.9	14.5	56.2	2293.01	83.3
39	6.3	45.9	38.7	8.6	29.2	8.4	30.7	555.1	77.7	1.4	13.3	2.3	74.7	6.0	39.0	2311.17	45.7	11.4	57.9	1535.38	82.9
37.5	7.8	55.3	40.9	8.5	34.3	11.4	41.8	802.3	68.7	1.1	25.9	2.4	78.9	12.3	49.4	2691.02	47.6	15.1	58.3	1776.63	82.6
35	5.9	39.2	30.3	6.2	27.4	9.3	28.1	296.0	61.6	0.8	10.9	1.7	64.5	6.4	42.6	2837.74	29.8	11.1	38.8	1106.05	74.2
32.5	4.0	43.5	23.0	6.2	20.0	7.6	24.8	294.8	36.6	0.6	11.1	1.4	54.5	7.4	39.7	3108.21	20.1	16.8	19.1	790.23	65.5
30	3.4	42.7	17.3	6.9	19.1	9.5	16.1	394.5	34.6	0.6	10.6	1.4	49.4	8.0	31.6	2238.23	27.2	10.6	29.6	834.86	55.3
27.5	3.3	50.0	12.3	7.9	11.6	11.5	13.6	314.5	33.5	0.5	12.1	1.5	33.0	9.7	16.1	2522.95	18.7	10.9	17.8	886.28	40.3
25	2.8	49.0	8.4	8.1	15.6	17.4	5.7	245.0	22.5	0.4	6.2	1.3	24.9	10.5	7.8	1797.21	13.3	10.7	12.5	751.50	36.0
23.7	1.7	43.6	5.3	5.8	9.1	10.4	4.7	188.6	9.2	0.4	4.3	0.6	17.3	14.0	3.6	1620.25	7.2	7.5	9.1	376.06	21.6
22.5	1.5	39.6	5.4	7.2	7.8	8.1	7.2	235.1	10.2	0.3	4.9	1.1	11.3	8.3	7.4	1684.47	6.8	8.3	6.6	520.28	17.1
20	1.4	46.2	4.3	5.6	6.4	8.3	5.3	176.1	24.7	0.7	2.0	0.7	16.8	5.6	16.0	2016.94	6.6	9.1	7.3	473.96	17.3
17.5	0.7	66.6	2.0	6.9	2.7	11.4	1.9	286.4	5.1	0.4	2.0	0.9	7.0	8.7	3.0	3455.43	2.9	15.0	3.0	590.56	6.9
Target value (mg/kg)		0.9		0.8		0.35		3.4		0.3		3		0.26		55		3		16	

where

- *i* and *j* are defined as before.
- α_j^i: Feed grade of element *j* in experiment *i*.
- $\propto_j^{target}$: Target grade for element *j*.

Then, to obtain the correct weighting for each element's contribution to overall contamination levels, the following transformation is implemented (Eq. (7)):

$$A_j^{i'} = \frac{A_j^i}{\sum_i^m A_j^i} \tag{7}$$

A global quality index for a given experiment, *i*, for all elements $\left(Q_T^i\right)$ can then be defined as follows (Eq. (8)):

$$Q_T^i = \sum_{j=1}^n Q_j^i A_j^{i\prime} \tag{8}$$

Finally, the best experimental set-up is, then, that for which this value is maximal (Eq. (9)):

$$Q_{optimal} = \mathrm{Max}\left\{\sum_{j=1}^n Q_j^i A_j^{i\prime}\right\} \tag{9}$$

where the following restrictions apply:

$$\left\{\forall \alpha_j^i, \gamma^i, \varepsilon_j^i, m, n, Q_T^i : \alpha_j^i \in \left[\alpha_j^{target}, 10^6\right], \gamma^i, \varepsilon_j^i \in (0, 100), m, n \in \mathbb{N}, Q_T^i \in (0, 2n)\right\}$$

2.3. *Two-parameter penalized attribute analysis (PPAA-U)*

As can be appreciated from Eqs. (5) and (6), experiments for which the yield, recovery, or grade varies greatly compared to the mean values will be given disproportionally more weight than those resulting in less variance. This will clearly bias the final quality assessment; thus, we present a modified method to address and eliminate this problem. Specifically, the inverse of the standard deviation can be used as a weighting factor to penalize large variations in each of the parameters of interest, yield, recovery, and grade, all of which vary for each element and every experiment. In addition, because the range of variation will be of a different order of magnitude for each parameter (for instance, in our case study, ε [%] is in the range {2, 83.3}, while γ [%] is in the range {0.7, 9}, and α [mg/kg] is in the range {0.32, 3108.21}, see Table 1), the weighting factors must be normalized to between 0 and 1.

In this way, we obtain a new value for the quality factor of each experiment and target element, C_j^i (Eqs. (10)–(14)):

$$\Gamma^i = \frac{\mathrm{Min}\{\gamma\}}{\gamma^i}\left(\frac{\sum_{i=1}^m \left|\gamma^i - \bar{\gamma}\right|}{m}\right)^{-1} \tag{10}$$

$$\Gamma^{i'} = \frac{\Gamma^i}{\sum_i^m \Gamma^i} \tag{11}$$

$$E_j^i = \frac{\varepsilon_j^i}{\mathrm{Max}\{\varepsilon_j\}}\left(\frac{\sum_{i=1}^m \left|\varepsilon_j^i - \bar{\varepsilon}_j\right|}{m}\right)^{-1} \tag{12}$$

$$E_j^{i'} = \frac{E_j^i}{\sum_i^m E_j^i} \tag{13}$$

$$C_j^i = \Gamma^{i'} + E_j^{i'} \tag{14}$$

and a new target-to-distance correction coefficient, $B_j^{i'}$ (Eq. (15)–(16)):

$$B_j^i = \frac{\alpha_j^i}{\propto_j^{target}}\left(\frac{\sum_{i=1}^m \left|\propto_j^i - \bar{\alpha}_j\right|}{m}\right)^{-1} \tag{15}$$

$$B_j^{i\prime} = \frac{B_j^i}{\sum_i^m B_j^i} \tag{16}$$

Thus, the corrected global quality index for an experiment *i* for all elements *j* to *n* is:

$$C_T^i = \sum_{j=1}^n C_j^i B_j^{i\prime} \tag{17}$$

Finally, the optimal experimental set up can be found as follows: $C_{optimal} = \mathrm{Max}\left\{\sum_{j=1}^n C_j^i B_j^{i\prime}\right\}$ (18).

where

- *i*, *j*, and *n* are defined as before.
- γ: Yield of experiment *i*.
- $\bar{\gamma}$: Mean yield for element j.
- ε: Recovery of element *j* in experiment i.
- $\bar{\varepsilon}_j$: Mean recovery for element *j*.
- α_j^i: Grade of the feed of *j* element in experiment *i*.
- $\bar{\alpha}_j$: Mean grade for element *j*.
- $\propto_j^{target}$: Target grade for element *j*.

and the following restrictions apply:

$$\left\{\forall \alpha_j^i, \gamma^i, \varepsilon_j^i, m, n, C_T^i : \alpha_j^i \in \left[\alpha_j^{target}, 10^6\right], \gamma^i, \varepsilon_j^i \in (0, 100], m, n \in \mathbb{N}, C_T^i \in (0, 2n)\right\}$$

The Supplementary material section (SM2) contains an example of this methodology used in a scenario involving two experimental set-ups with two elements to be separated.

3. Results and discussion

3.1. *Separation results*

We conducted twelve experimental runs with separation voltages ranging from 41.5 kV to 17.5 kV (Table 1). There was a positive correlation between yield (γ) and voltage with $\gamma = 1.4\%$ at 20 kV and $\gamma = 8.6\%$ at 41.5 kV with a maximum of $\gamma = 9.0\%$ at 40.0 kV (peak-yield voltage). Similarly, the recovery (ε) in the conductive fraction was also positively correlated with the voltage. The peak-yield voltage (40 kV) also produced the highest maximum recoveries for six PTEs of interest: Zn (83.3%), followed by Mo (81.0%), Cu (62.2%), Cu (62.2%), Sb (59.6%) and Ni (57.3%). The same voltage, however, resulted in the lowest recoveries for the other four PTEs studied: Hg (13.2%), Cr (38.8%), Cd (45.0%), and As (41.7%). Considering the variation in the values recorded for the three parameters of interest, yield, recovery, and grade, while the first of these parameters varied in a range from 1.4% to 8.6%, the other two had far wider ranges: 2%–83.3% and 0.3 mg/kg to 3108 mg/kg, for recovery and grade, respectively. All the data are summarized in Table 1.

Attributive analysis generates a family of curves describing the relationships between yield, recovery, and the quality index for a particular experimental set-up. In the following sections, we will consider these curves and compare the performances of basic and penalized attributive analysis in evaluating the quality indices of the twelve separation experiments undertaken here.

3.2. Families of curves

Taking Eq. 5, the attributive analysis equation, and substituting in values for yield (γ) and recovery (Ɛ), it is possible to produce a family of surfaces that share a similar shape and functional relationship. These surfaces represent the ways in which the quality index of an experiment (Q) will vary with changes in either γ or Ɛ. Fig. 1 shows a surface plot of Q for all possible combinations for γ^i and ε_j^i ranging from 0.01 to 0.99.

Sierra et al. (2010) applied the attributive analysis function to process engineering; however, they did not analyze the family of surfaces created. Such analysis enables an exploration of the theoretical consistency of the proposed methodology. In this way, as part of the present study, we will perform a sensitivity analysis and a comparative analysis with known values from our electrostatic separation experiment (see Section 2.1). Concerning the former type of analysis, this considers the relative sensitivity of the quality index to changes in yield and recovery values. Meanwhile, our comparative analysis involves an examination of results derived from the basic version of attributive analysis in comparison to those derived from PPAA-U without target-to-distance correction.

As demonstrated in Fig. 1, the relationship between Q and γ is very different from its relationship with Ɛ; thus, it is useful to examine these two relationships separately. This can be done by keeping either one of the two addends in Eq. 5 ($\frac{\varepsilon_j^i}{\mathrm{Max}\{\varepsilon_j\}}$ or ε_j^i) constant to give two families of curves, one for Q varying with γ and another for Q varying with Ɛ.

Starting with the recovery addend, $\frac{\varepsilon_j^i}{\mathrm{Max}\{\varepsilon_j\}}$, setting $\mathrm{Max}\{\varepsilon_j\}$ to 0.99 and varying ε_j^i between 0.01 and 0.99, we obtain a set of curves corresponding to different values of γ (for values of γ from $\gamma_{\min}$=0.01 to 0.99). These curves correspond to the Q-Ɛ plane (see Fig. 1) at different points along the γ-axis, and as can be seen in Fig. 2, the quality index, Q, and Ɛ are related by a straight line:

$$\mathrm{Q} = \frac{\varepsilon_j^i}{\mathrm{Max}\{\varepsilon_j\}} + K$$

The gradient of the line can be found by taking the derivative of Q with respect to Ɛ:

$$\mathrm{dQ} = \frac{d\varepsilon_j^i}{\mathrm{Max}\{\varepsilon_j\}}$$

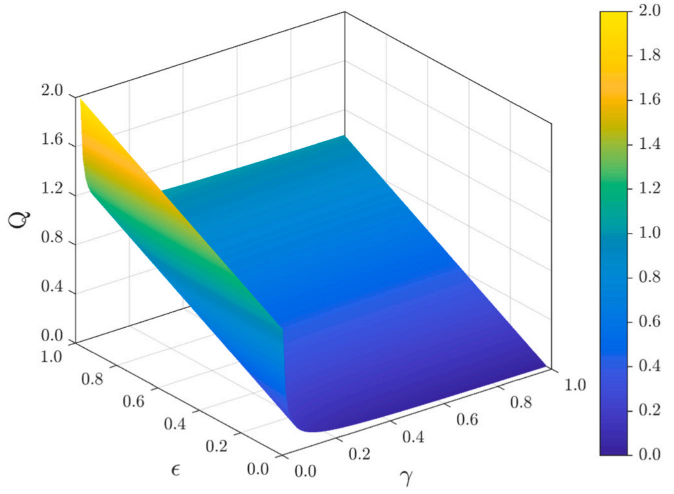


Fig. 1. General shape of a quality index function.

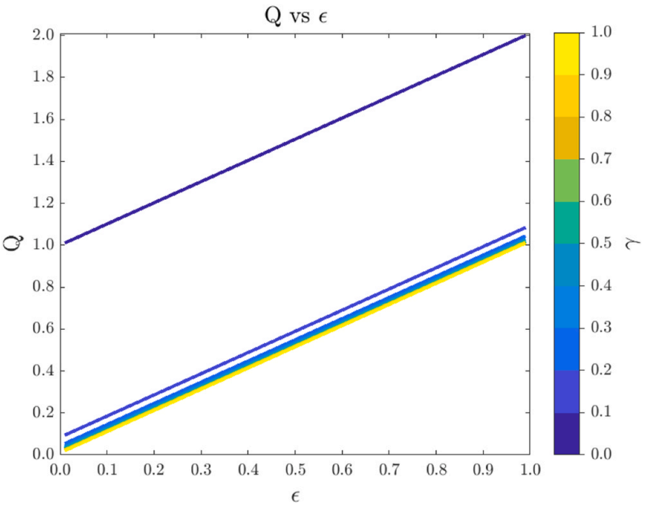


Fig. 2. Quality index function (parallel to the Q-ε plane).

such that (Eq. (19)):

$$\frac{\mathrm{dQ}}{d\varepsilon_j^i} = \frac{1}{\mathrm{Max}\{\varepsilon_j\}} \tag{19}$$

This tells us that as the maximum recovery increases, the slope of the curve generated decreases. Moreover, when $\mathrm{Max}\{\epsilon_j\}$ is large, the variation in Q with recovery will decrease.

Taking the yield addend, $\frac{\mathrm{Min}\{\gamma\}}{\gamma^i}$, setting $\mathrm{Min}\{\gamma\}$= 0.01 and varying Ɛ from 0.01 to Ɛ= $\mathrm{Max}\{\varepsilon_j\}$ = 0.99 gives a second set of curves. These curves are Q-γ planes at different points along the Ɛ axis; see Fig. 3. In contrast to Ɛ, γ has a nonlinear relationship with Q, and calculating the gradient of the curve gives an inverse square function (Eq. (20)):

$$\frac{dQ}{d\gamma} = \frac{-\mathrm{Min}\{\gamma\}}{(\gamma^i)^2} \tag{20}$$

This result shows that the quality index is highly sensitive to yield for values of $\gamma^i < 0.1$; however, for $\gamma^i > 0.1$, the quality index remains almost stable. In this way, reducing yield values is more important than maximizing recovery since, at very low yields, small changes in this parameter have a very large impact on the quality index.

Eq. 20 also demonstrates that the sensitivity of the quality index to changes in yield decreases for higher values of Min{γ}. This fact could be

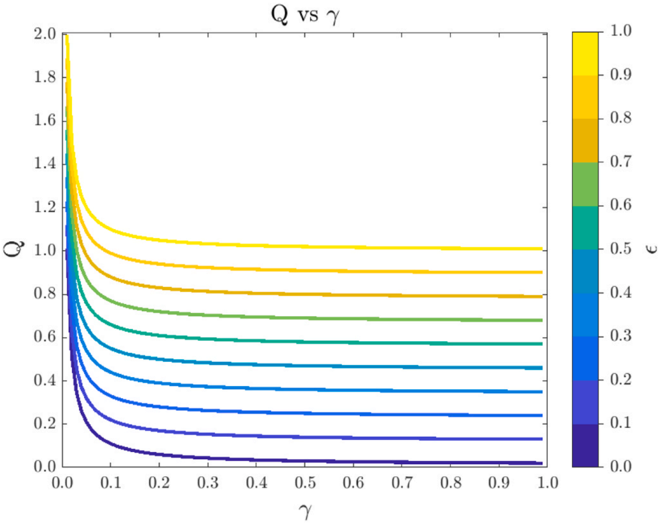


Fig. 3. Quality index parallel to the Q-γ plane.

useful under particularly noisy experimental conditions.

From the above analysis, it becomes apparent that Q should be considered a function of four variables: γ^i and ε_j^i, $\mathrm{Min}\{\gamma\}$ and $\mathrm{Max}\{\varepsilon_j\}$. We will now explore in more detail the effect of variations in $\mathrm{Min}\{\gamma\}$ and $\mathrm{Max}\{\varepsilon_j\}$.

Starting this time with the yield addend, $\frac{\mathrm{Min}\{\gamma\}}{\gamma^i}$, increasing $\mathrm{Min}\{\gamma\}$ reduces the function domain from [0,1] to [$\mathrm{Min}\{\gamma\}$,1]. As was mentioned, while this function is highly sensitive to $\gamma^i < 0.1$, at larger values of γ^i the curve is relatively flat (Fig. 3); thus, increasing $\mathrm{Min}\{\gamma\}$ places the quality function in a largely stable zone. Furthermore, as Fig. 4 demonstrates, higher values of $\mathrm{Min}\{\gamma\}$ lead to flatter curves, meaning that Q becomes increasingly insensitive to variation in γ^i.

Similarly, considering the recovery addend, $\frac{\varepsilon_j^i}{\mathrm{Max}\{\varepsilon_j\}}$, if $\mathrm{Max}\{\varepsilon_j\}$ decreases, this also narrows the function domain from [0,1] to [0, $\mathrm{Max}\{\varepsilon_j\}$]. In addition, since the gradient of the curve (see Eq. 19) is constant and equal to $\frac{1}{\mathrm{Max}\{\varepsilon_j\}}$, increasing $\mathrm{Max}\{\varepsilon_j\}$ will decrease the gradient angle, given by $\tan^{-1}\left(\frac{1}{Max\{\varepsilon_j\}}\right)$. This is demonstrated in Fig. 5.

3.3. Results of Penalized Attributive Analysis (PAA-U)

Bearing in mind the insights of the previous section, we now consider a comparison of basic attributive analysis, AA, and its inverse standard deviation weighted or penalized version, PPAA-U. Fig. 6 presents a comparison between results obtained using AA and PPAA-U (without target-to-distance correction) for the electrostatic soil washing operation described in Section 2.1. We observe significant agreement between AA and PPAA-U, particularly for lower voltages (<25 kV). This is because the variation in recoveries for the elements tested is less at lower voltages (see Table 1) than at higher voltages. In this way, the weighting used in PPAA-U makes little difference at lower voltages; however, at high voltages, the high variances are strongly penalized, lowering the Q values of these experiments and thus causing a divergence in the results obtained via the basic and penalized versions of the method. In addition, both AA and PPAA-U predict the presence of two maxima in the quality index: one at low voltages where, despite low recoveries, the lower yield leads to a peak in the quality index and a second at high voltages where there is high recovery.

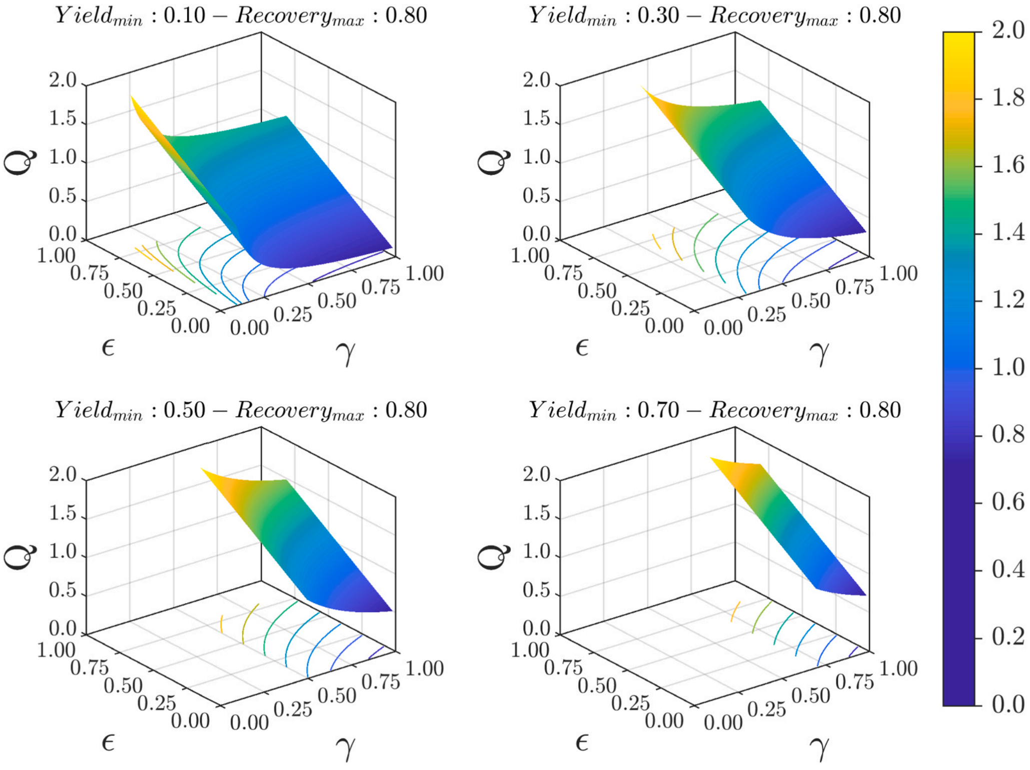
When the target-to-distance correction is introduced, while both methods once again give similar results and, as expected PPA diverges from AA at higher voltages, the overall pattern is very different: specifically, the peak in the quality index at lower voltages disappears (Fig. 7). The target-to-distance correction allows us to compare how different experimental set-ups perform with respect to particularly harmful elements. This, in turn, enables attributive analysis to identify not simply the best overall separation conditions but those that are most environmentally optimal. In the present case, voltages greater than 37.5 kV stand out as providing the best separation conditions, giving priority to the removal of the most harmful PTEs.

The work presented here demonstrates that PPAA-U is a promising tool for identifying the optimal conditions for electrostatic soil washing operations. To further improve the methodology, additional quotients should be incorporated to account for components reporting to the middlings fraction instead of considering it as part of the concentrate, as is done here. In addition, the method could be modified to encompass some of the economic factors involved in the soil washing process, especially those relating to the circular economy, to further optimize conditions.

4. Conclusions

Currently, the available literature offers few means to evaluate the quality of a separation process. Methods that do exist tend to use only two parameters, typically recovery and yield; however, on their own, these two variables do not provide a sufficiently robust way to identify separation conditions that are genuinely optimal.

In its original form, attributive analysis addresses the shortcomings of other methods, providing a tool to assess optimal separation


$Yield_{min}$: 0.10 − $Recovery_{max}$: 0.80

$Yield_{min}$: 0.30 − $Recovery_{max}$: 0.80

$Yield_{min}$: 0.50 − $Recovery_{max}$: 0.80

$Yield_{min}$: 0.70 − $Recovery_{max}$: 0.80

Fig. 4. Quality index function for different minimum yields.

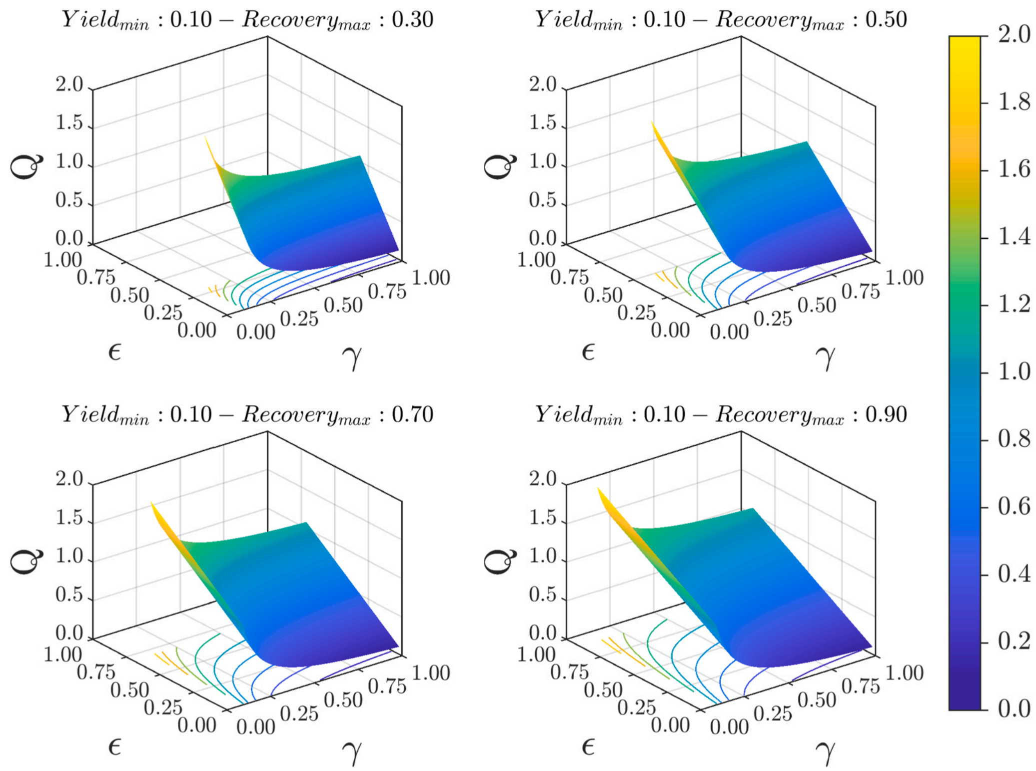


Fig. 5. Quality index function for different maximum recoveries.

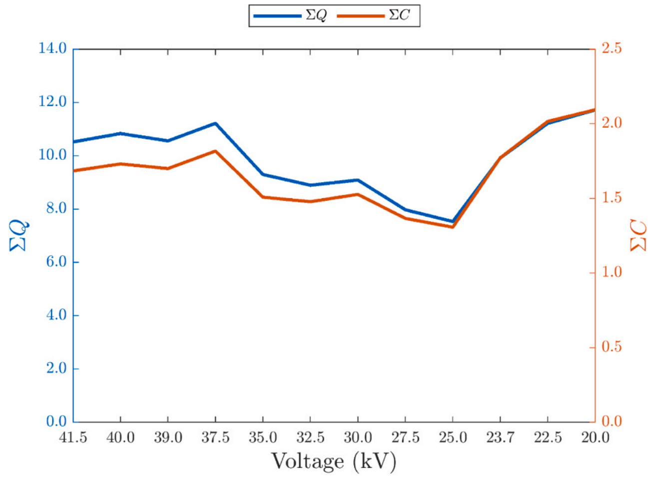


Fig. 6. AA (ΣQ) and PPAA-U (ΣC) quality index results before target-to-distance correction.

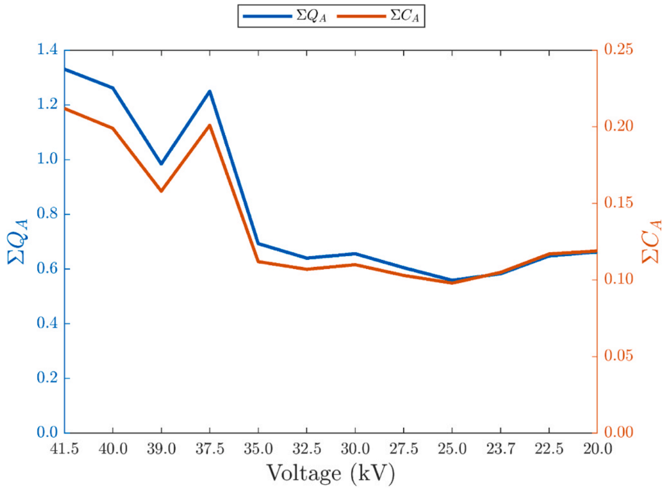


Fig. 7. Quality index as calculated via basic AA (ΣQ_A) and PPAA-U (ΣC_A) with target-to-distance correction.

conditions through a comparison of three variables: yield, recovery, and grade. However, this basic method suffers where there are large variations in the yields and recoveries of the components or elements separated.

Indeed, examination of the attributive analysis function reveals that it exhibits significant sensitivity to dispersion within both the yield dataset and the recovery dataset. However, the sensitivity to variations in yield is greatest, something that can be attributed to the fact that while the quality index is linearly related to recovery (with the slope being dependent on the maximum recovery value), it has an inverse relationship with yield and thus an inverse squared relationship to changes in yield. Penalized attributive analysis, PAA-U, directly addresses the issue of variations in yields and recoveries, most strongly penalizing contributions to the quality index from experimental set-ups for which the largest variations are recorded.

Target-to-distance correction improved the performance of both AA and PPAA-U. For the separation technique used in this work, without this correction, two maxima (one at higher and one at lower voltages) in the quality index were observed, making it difficult to distinguish the true optimum conditions. When the target-to-distance correction was introduced, however, the lower voltage maximum was removed. This shows that while lower voltages might provide effective separation on average, they are poor at removing particularly harmful PTEs; this more targeted separation is achieved only at higher voltages.

Future work should focus not only on the application of PPAA-U to evaluate the soil washing methods used in the remediation of metal (loid)-polluted soil but also as part of feasibility studies for bioremediation or chemical oxidation technologies (where numerous organic contaminants, each with different target concentrations, are addressed simultaneously). Furthermore, this methodology can not only be applied to environmental remediation operations but could also be used to determine optimal operating conditions in a variety of materials processing or manufacturing contexts. In such contexts, the quality factor used in this method might involve variables such as temperature, pressure, or particle size and also the environmental and economic factors such as the costs associated with particular operating conditions, which might depend on energy use or manpower requirements.

Environmental implications

Two-parameter penalized attribute analysis for upgrading (PPAA-U) provides a way to optimize soil remediation operations and is, thus, a valuable tool to improve environmental outcomes. The method assesses how well a given set of operating conditions maximizes recovery while simultaneously minimizing the concentrate yield, and its primary advantage lies in the way it provides a single value, the quality index, to identify optimal separation conditions. Furthermore, PPAA-U could be adapted easily to various processes and thus has numerous potential applications in a range of industries; in particular, its capacity to address multiple variables opens new avenues for sustainable materials processing and manufacturing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

Carlos Sierra would like to thank the EURECA-PRO phase I 2020–2023 co-funded by the European Union's Erasmus + Programme (Ref.: 101004049). Diego Baragaño wants to acknowledge MCIU/AEI/CSIC for his JdC contract under the grant JDC2022-050209-I funded by MCIU/AEI/10.13039/501100011033 and by European Union NextGenerationEU/PRTR.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2024.133529.

References

[1] Wu, Q., Leung, J.Y.S., Geng, X., Chen, S., Huang, X., Li, H., et al., 2015. Heavy metal contamination of soil and water in the vicinity of an abandoned e-waste recycling site: Implications ofr dissemination of heavy metals. Science of the Total Environment 506-507, 217–225. https://doi.org/10.1016/j.scitotenv.2014.10.121.

[2] Sun, L, Guo, D., Liu, K., Meng, H., Zheng, Y., Yuan, F., et al., 2019. Levels, sources, and spatial distribution of heavy metals in soils from a typical coal industrial city of Tangshan, China. Catena 175, 101–109. https://doi.org/10.1016/j.catena.2018.12.014.

[3] Ashraf, S., Ali, Q., Zahir, Z.A., Ashraf, S., Asghar, H.N., 2019. Phytoremediation: Environmentally sustainable way for reclamation of heavy metal polluted soils. Ecotoxicology and Environmental Safety 174, 714–727. https://doi.org/10.1016/j.ecoenv.2019.02.068.

[4] Khalid, S., Shahid, M., Niazi, N.K., Murtaza, B., Bibi, I., Dumat, C., 2017. A comparison of technologies for remediation of heavy metal contaminated soils. Journal of Geochemical Exploration 182, 247–268. https://doi.org/10.1016/j.gexplo.2016.11.021.

[5] Aparicio, J.D., Raimondo, E.E., Saez, J.M., Costa-Gutierrez, S.B., Álvarez, A., Benimeli, C.S., et al., 2022. The current approach to soil remediation: a review of physicochemical and biological technologies, and the potential of their strategic combination. J Environ Chem Eng 10 (2). https://doi.org/10.1016/J.JECE.2022.107141.

[6] Baragaño, D., Berrezueta, E., Komárek, M., Menéndez-Aguado, J.M., 2023. Magnetic separation for arsenic and metal recovery from polluted sediments within a circular economy. J Env Manag 339, 117884. https://doi.org/10.1016/j.jenvman.2023.117884.

[7] Wan, X., Lei, M., Chen, T., 2020. Review on remediation technologies for arsenic-contaminated soil. Front Environ Sci Eng 14 (2), 1–14. https://doi.org/10.1007/S11783-019-1203-7/METRICS.

[8] Baragaño, D., Gallego, J.R., Menéndez-Aguado, J., Marina, M.A., Sierra, C., 2021. As sorption onto Fe-based nanoparticles and recovery from soils by means of wet high intensity magnetic separation. Chem Eng J 408, 127325. https://doi.org/10.1016/J.CEJ.2020.127325.

[9] Gu, F., Zhang, J., Shen, Z., Li, Y., Ji, R., Li, W., et al., 2022. A review for recent advances on soil washing remediation technologies. Bull Environ Contam Toxicol 109 (4), 651–658. https://doi.org/10.1007/S00128-022-03584-6/METRICS.

[10] Liu, J., Zhao, L., Liu, Q., Li, J., Qiao, Z., Sun, P., et al., 2022. A critical review on soil washing during soil remediation for heavy metals and organic pollutants. Int J Environ Sci Technol 19 (1), 601–624. https://doi.org/10.1007/S13762-021-03144-1/METRICS.

[11] Sierra, C., Martínez-Blanco, D., Blanco, J.A., Gallego, J.R., 2014. Optimisation of magnetic separation: a case study for soil washing at a heavy metals polluted site. Chemosphere 107, 290–296. https://doi.org/10.1016/J.CHEMOSPHERE.2013.12.063.

[12] Tran, H.T., Lin, C., Hoang, H.G., Bui, X.T., Le, V.G., Vu, C.T., 2022. Soil washing for the remediation of dioxin-contaminated soil: a review. J Hazard Mater 421. https://doi.org/10.1016/J.JHAZMAT.2021.126767.

[13] Bunge, R., Bachmann, A., Ngo, C.D.1995. Soil-washing: Mineral processing technology in environmental engineering.

[14] Nishiyama, T., Tomoguchi, M., Sasamoto, N.,2000. Soil washing using mineral processing plant in Hanaoka Mine, Dowa Mining Co., Ltd. Second International Conference on Processing Materials for Properties, pp. 601–624. ⟨https://www.researchgate.net/publication/290268173_Soil_washing_using_mineral_processing_plant_in_Hanaoka_Mine_Dowa_Mining_Co_Ltd⟩.

[15] Richardson, W.S., Phillips, C.R., Luttrell, J., Hicks, R., Cox, C., 1999. Application of remedy studies to the development of a soil washing pilot plant that uses mineral processing technology: a practical experience. J Hazard Mater 66 (1–2), 47–65. https://doi.org/10.1016/S0304-3894(98)00211-8.

[16] Schulz, N.F.,1970. "Separation Efficiency." Trans. SME/AIME, 247, pp. 81–87. ⟨https://cir.nii.ac.jp/crid/1570572701370836096⟩.

[17] Gupta, A., Yan, D.2016. Mineral Processing Design and Operations: An Introduction: Second Edition. Mineral Processing Design and Operations: An Introduction: Second Edition, pp. 1–850.

[18] Weiss, N.L.,1985. Metallurgical Accounting and Mill Reports. In SME mineral processing handbook. Society of Mining Engineers of the American Institute of Mining, Metallurgical, and Petroleum Engineers.

[19] Wills, B.A., Finch, J.A., 2015. Wills' mineral processing technology: an introduction to the practical aspects of ore treatment and mineral recovery. Wills' Miner Process Technol. Introd Pract Asp Ore Treat Miner Recovery 1–498.

[20] Drzymala, J., 2006. Atlas of upgrading curves used in separation and mineral science and technology. Physicochem Probl Miner Process 40.

[21] Taggart. A.F.,1947. Handbook of Mineral Dressing. John Wiley & Sons. ⟨https://www.abebooks.com/first-edition/Handbook-Mineral-Dressing-Ores-Industrial-Minerals/4800302279/bd⟩.

[22] Cortada, U., Hidalgo, C., Martínez, J.M., Rey, J., 2018. Impact in soils caused by metal(loid)s in lead metallurgy. the case of La Cruz Smelter (Southern Spain). J Geochem Explor 190 (5a). https://doi.org/10.1016/j.gexplo.2018.04.001.

[23] Rosendo, R., Rey, J., Martínez, J.M., Hidalgo, C., 2022. Geological and mining heritage as a driver of development: the NE sector of the Linares-La Carolina district (Southeastern Spain). Geosciences 12 (2), 76. https://doi.org/10.3390/geosciences12020076.

[24] American Society for Testing and Materials, 1963. ASTM D422–63: Standard Test Method for Particle-Size Analysis of Soils (Reapproved 2007). ASTM International.

[25] Boente, C., Sierra, C., Rodríguez-Valdés, Menéndez-Aguado, J.M., Gallego, J.R., 2017. Soil washing optimization by means of attributive analysis: case study for the removal of potentially toxic elements from soil contaminated with pyrite ash. J Clean Prod 142, 2693–2699. https://doi.org/10.1016/j.jclepro.2016.11.007.