Article

Tackling Arsenic and Mercury Contamination: Implications for Sustainable Mining and Occupational Health Risks

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Abstract: This study investigates the environmental and occupational health risks associated with arsenic (As) and mercury (Hg) contamination in a specific industrial site: a mercury mine site that contains a metallurgical plant within its premises. Utilising a comprehensive sampling and analysis approach, As and Hg concentrations in the soil and air across various zones within the site were assessed. The results revealed elevated levels of both contaminants, particularly in areas proximal to industrial processes such as metal smelting and waste disposal. Risk assessment using the Cancer Risk (*CR*) and Hazard Index (*HI*) indices demonstrated significant health hazards that exceed regulatory thresholds, indicating potential carcinogenic effects from As exposure and risks of non-cancerous occupational diseases. Three distinct risk areas were identified based on the *CR* and *HI* indices, guiding the formulation of tailored risk management strategies. While some zones may permit limited industrial activities under specific conditions, others require stringent safety measures and specialised personal protective equipment (PPE) due to exceptionally high contaminant concentrations. Overall, the findings underscore the critical need for robust safety protocols and regulatory compliance to mitigate the health risks associated with As and Hg exposure in industrial settings, ensuring the protection of worker health, environmental stewardship, and the promotion of sustainable mining practices.

Keywords: sustainability; arsenic contamination; mercury exposure; occupational safety; environmental pollution; risk assessment; sustainable mining

1. Introduction

Estimating the health risks to human populations at sites contaminated with potentially toxic elements (PTEs) is evidently a matter of vital importance. Over the past few decades, various risk estimation models have been proposed; Chartres et al. [1] and Zhang et al. [2] have provided reviews of these models. While Chartres et al. aimed to improve the uniformity and transparency of the assessments carried out by national and international organisations through examining the techniques used for hazard identification and risk assessment regarding environmental hazards, Zhang et al., on the other hand, focused on soil pollution, particularly when examining the risk assessment for human health associated with contaminated locations. The study by Zhang et al. dives deeply into particular aspects of risk assessment related to contaminated sites, such as hazard identification, dose–response assessment, and exposure assessment, whereas that of Chartres et al. provides a broad overview of the methods used across various organisations.

In many cases, health risk analysis follows the US EPA Guidance for Calculating Health Risk at a Specific Location (US EPA site-specific risk assessment methodology) [3–5].

This standardisation allows for risk estimation at a particular site and facilitates comparison of the results with those from other locations (worldwide) where the same methodology has been applied.

A large number of articles focused on assessing human health risks in sites contaminated with PTEs due to various industrial activities has recently been published. Examples of this include the works of Wu et al. [6], Gruszecka-Kosowska et al. [7], Xia et al. [8], Custodio et al. [9], Wcislo [10], Wcislo and Bronder [11], Wcislo et al. [12], and Wu et al. [13]. These publications summarise the research on the dangers that heavy metal contamination poses to human health in a variety of environmental contexts. They use standard techniques and focus on the following important area, albeit in different geographic locations: the variety of heavy metals, such as arsenic, cadmium, chromium, copper, mercury, nickel, lead, and zinc, in soil upon identification of the contaminants. In order to estimate the hazards associated with various exposure pathways, such as oral ingestion and skin contact, health risk assessment uses both deterministic and probabilistic models. There are documented sources of contamination, including both natural and industrial sources. Related evaluations draw attention to possible long-term health consequences, such as cancer and chronic illnesses. Reducing human exposure and improving environmental quality in impacted areas are the goals of policy formulation and environmental remediation recommendations.

Following the same methodology, this study estimates the health risks for humans—particularly, for workers—in the case of La Soterraña, an abandoned mercury mining facility area. The Soterraña Mine is one of the most polluted areas in Europe, mainly due to high levels of arsenic and mercury in the soil [14]. A research project, including some civil works, was conducted recently (2017–2024), and a very extensive restoration project with a budget of EUR 8 million will be carried out during the coming months (2024–2025). In this context, the analysis of the workplace from a health and safety point of view is of the utmost importance. The study focuses only on two contaminants or potentially toxic elements (PTEs)—mercury (Hg) and arsenic (As)—as these are two contaminants that typically reach high concentrations in facilities related to abandoned or already closed mercury mines. In fact, in the case of La Soterraña, the historical production of arsenic was much higher than that of mercury.

The International Agency for Research on Cancer (IARC) has classified arsenic and inorganic arsenic compounds as Group 1 carcinogens, which means they are confirmed as carcinogenic to humans [15]. Arsenic compounds, particularly inorganic ones, are regarded as potent poisons, affecting various systems in the human body by interfering with numerous enzymatic reactions. The most evident effects are seen on the skin, and arsenic is associated with various cancers, such as those of the lungs, skin, and bladder [16]. Exposure to inorganic arsenic can irritate the stomach, lungs, and intestines, cause changes in the skin, and decrease the production of white and red blood cells. Significant exposures have been linked to an increased risk of developing cancer, including lung, skin, liver, lymph, and others. Epidemiological and biological studies have described the metabolic pathways and biological modes of action of arsenic, although further research is needed to confirm the findings. Exposure to arsenic has also been associated with effects on the nervous system and genetic changes, as well as decreased bactericidal activity and alteration to the respiratory tract epithelial barrier. Arsenic increases the risk of vascular diseases and various tumours and is widely absorbed in the body, accumulating especially in the liver and excreting mainly through urination. The toxicokinetics of arsenic depends on various factors, including the duration and route of exposure, as well as the characteristics of the compound and affected biological species [17–19].

Mercury toxicity has been recognised since ancient times, with historical figures such as Pliny, Hippocrates, and others documenting its dangers. Ulrich Ellenberg, in 1473, and Paracelsus, around 1527, were among the early voices discussing occupational risks, while Bernardino Ramazzini coined the term "mercurialism" in his work.

Mercury's toxicity varies depending on its chemical form, notably, in elemental, organic (such as methylmercury), or inorganic compounds. Methylmercury, for instance, accumulates in mammals and fish.

The body can be exposed to mercury through various pathways:

Respiratory Route: Mercury vapours can easily enter the bloodstream through the alveoli, with approximately 80% of inhaled vapours absorbed. Despite some oxidation to divalent mercury, much elemental mercury can still reach the brain due to its quick transit from the lungs [20].

Digestive Route: Ingestion, especially from fish, is another significant route, although absorption rates vary based on compound solubility.

Dermal Route: While less significant, mercury can penetrate the skin, particularly in the case of methylmercury.

The distribution of mercury in the body is widespread, with the kidneys acting as the primary reservoir. Despite high concentrations in the kidneys, the nervous system remains the most sensitive target for elemental mercury exposure [21].

The elimination of mercury mainly occurs through passing urine and faeces, although residual amounts can persist in the body for years after exposure ceases.

The health effects of mercury toxicity are profound, ranging from neurological symptoms, such as tremors, insomnia, and cognitive deficits, to gastrointestinal issues, such as metallic taste, nausea, and gingivitis. Skin contact with inorganic mercury can lead to dermatitis, while psychological disturbances, known as "erethism", may include mood changes, anxiety, and depression [22].

Mercury's impact extends to foetal development, as it readily crosses the placental barrier and transfers through breast milk. Organic mercury compounds, including methylmercury, exhibit high absorption rates through various routes, exacerbating their toxic effects.

At present, the analysis of potential risks to the health of both workers and the public is of the utmost importance. On the other hand, the modern concept of sustainability requires making an effort to use industrial subproducts, instead of natural resources, in the context of the circular economy. In the same way, sustainability requires the restoration and remediation of industrial areas abandoned in the past that remain degraded and contaminated. The use of industry subproducts is an optimal solution for these cases. Simultaneously, in order to reduce the exploitation of natural resources, many projects are now being developed with the aim of extracting strategic and highly valuable metals from abandoned mining waste dumps. In all these cases, workers must work under critical conditions with very high PTE concentrations, which implies a high risk to the health of these workers. Consequently, it is of the utmost importance to utilise any contribution that helps technicians in the planning and implementation of this type of work.

This study delves into a comprehensive assessment of the health risks posed by arsenic (As) and mercury (Hg) contamination within a designated mercury mining area. This investigation builds upon previous research conducted in the study area, ranging from earlier investigations led by the University of Oviedo to recent projects such as SUBproducts4LIFE. These collective efforts have provided a foundational understanding of the contamination landscape, delineating zones of varying risk levels and informing subsequent remediation strategies. However, while prior studies have laid the groundwork, our current work seeks to expand upon these findings through implementing a more nuanced risk assessment framework. This framework considers not only the concentrations of As and Hg in the soil and air, but also their implications for both cancer and non-cancerous occupational diseases. In doing so, we aim to offer a more comprehensive understanding of health risks within the study area and provide actionable insights to mitigate these risks effectively. The research findings can be instrumental in the Health and Safety of upcoming works in the area.

The present research is different from previous works in the following three relevant aspects. In previous papers, the US EPA model was used only with data on the concentra-

tion of PTEs in soils, while, in the present work, we have taken into account the As and Hg concentrations in air both in the dust (As and Hg) and as a gas (only Hg). We have demonstrated that the US EPA model, although it is very extraordinarily useful, can be improved through the use of site data. In the case of small areas with different kinds of wastes, the calculation using parameters from the US EPA yields As and Hg concentrations in the air that are much higher than the recorded ones. The methodology is not used to estimate the risks related to different common activities. Nevertheless, in this study, we use the method to estimate the risk and to classify the different areas according to this risk in the context of the planning of research works that are being carried out currently and the restoration works that will be carried out in the near future (2024–2025).

2. Materials and Methods

2.1. Site Description

The Soterraña Mine is located in the Municipality of Lena, Principality of Asturias, Spain. The mercury mine closed in 1974, and restoration of this area has not yet been carried out. The co-ordinates of the mine are 43.19303, −5.844671. A video showing a general overview of the current situation of the site is presented in [23].

Very thorough prior knowledge about the site exists, allowing us to define six different areas (Figure 1). In general, the first step to improving knowledge is to study the site to understand how and where different mining and metallurgic activities have been carried out in the past. A second step is to take some soil samples to obtain the concentration of PTEs in the soil; in this case, the results from previous studies carried out in the area can be used. From these two steps, different zones with different concentrations of As and Hg (clearly related to the different mining and metallurgical processes in these places) can be defined.

Figure 1. Different zones in the abandoned facilities of La Soterraña.

A. Zone of chimneys and ducts for smoke exhaust;
B. Area of demolition debris from the support structures of the roasting furnace in the metallurgical plant;
C. Upper waste dump, made up of residues from the metallurgical process;
D. Lower waste dump, made up of mining waste;
E. Horizontal platform at the base of this dump, where leachates from it converge;

F.   Other areas, such as transit areas, warehouses, buildings, cleaning facilities, roads, etc., that are different from the aforementioned types.

2.2. Sampling Description

Conducting the study required data on the concentration of As and Hg in the soil and the concentration of gaseous Hg in the air. If there is no prior knowledge of an area, sampling could be conducted by defining 25 × 25 m grids and following the methodology of the US EPA, as carried out by Wcislo et al. [12].

As it stated above, very thorough prior knowledge of the site exists, allowing us to define six different zones in advance. Having this comprehensive understanding before-hand allowed for more targeted and efficient sampling strategies, compared to generic grid-based approaches (Figure 2).
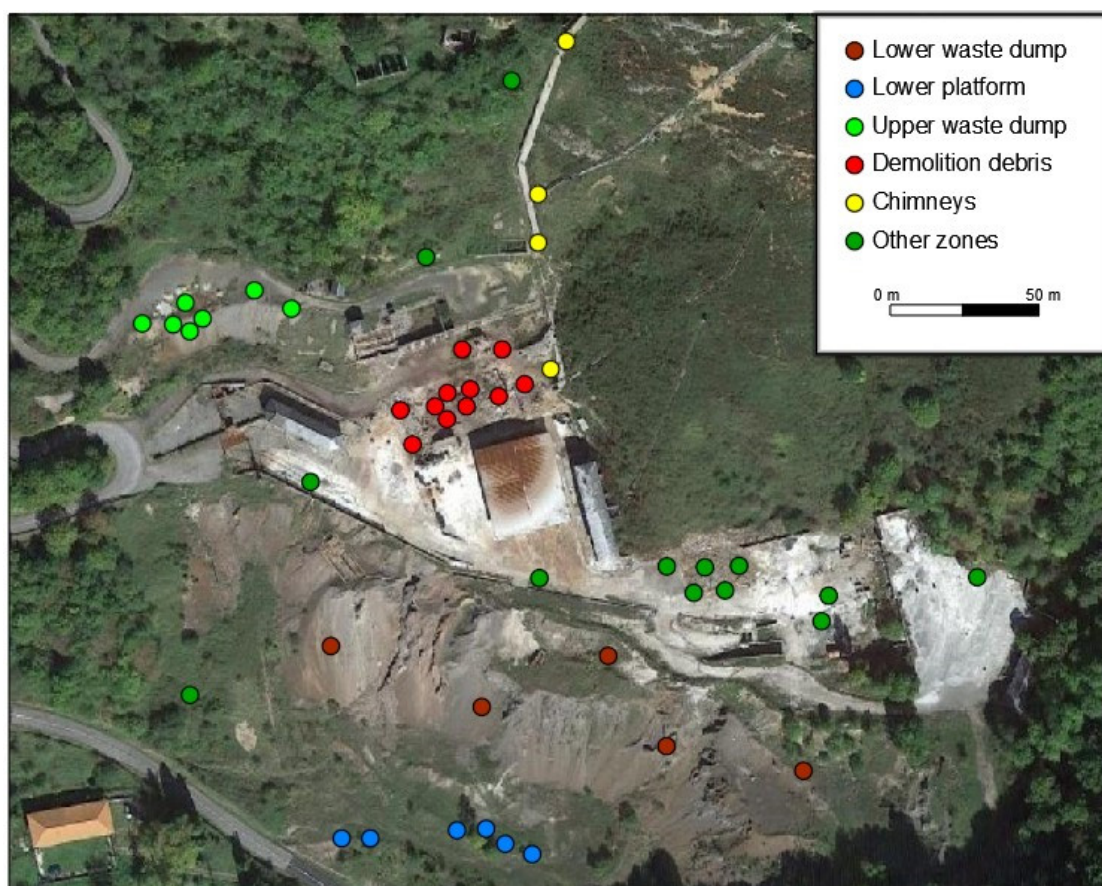

Figure 2. Sampling points in different zones.

The study utilised data from previous investigations, including those by Loredo et al. [24], Soto Yen [25], and Fernández et al. [14]. Additionally, more samples were collected and analysed for verification purposes, some of which have been used in previous works by Ayala and Fernández [26].

Soil samples were collected, allowed to dry at room temperature, ground using a roller, and sieved to remove grains larger than 4 mm. The material was then split (riffled) to obtain 50 g representative subsamples, which were subsequently pulverized into particles smaller than 100 μm. Following this, representative subsamples weighing 0.250 g were leached, diluted, and filtered before quantification using ICP-MS, following the methodology described by Fernández et al. [14,26].

Arsenic and mercury concentrations in airborne dust were collected using a personal pump calibrated to 2 lpm equipped with an IOM sampler and utilizing a Mixed Cellulose Ester (MCE) membrane filter substrate. The collected filters underwent digestion and

analysis using ICP-MS, as outlined by García-González et al. [27]. Airborne mercury was further measured using the continuous monitoring analyser Lumex RA-915 with representative samples from each area, following the procedure described by Rodriguez et al. [28].

2.3. Methodology for Assessing Human Health Risk

Previous studies have shown that, in addition to mercury and arsenic, the soil and metallurgical waste in this area are also contaminated with dioxins and furans [14]. A comprehensive assessment of the health impact for cases involving common industrial activities should include consideration of these additional harmful toxic elements. This is because the risk of contamination from some of these elements in the presence of others may be higher than the sum of their risks.

However, this aspect becomes critical when the individual indices are below the legal limit (separately), yet their combined total exceeds this limit. In our case, the values of the indices for arsenic and mercury exceeded the legal limits by a significant margin, thus reaching the overall limit when considering only these two elements. We have focused solely on arsenic and mercury due to their higher concentrations in the soil and air, as well as their representation of different critical risks—both from soil and air pollution and their carcinogenic and non-carcinogenic effects.

On the other hand, we analysed the risks associated with the necessary works for the restoration and remediation of the area, instead of common industrial activity. In this context, workers will already be using the necessary personal protective equipment (PPE) for arsenic and mercury, which is also effective for other types of toxic elements (with the addition of new filters to the half-masks if necessary). Consequently, the method used to classify the different zones according to their risk was employed when assessing these risks.

The first step was to estimate exposure to contaminants, which is carried out separately according to the route of entry of these contaminants into the body: Oral (direct ingestion), dermal (skin contact), or inhalation regarding either dust or volatile contaminants.

In the first two cases, the aim is to estimate the quantity of contaminant entering the body in mg of contaminant per day per kg of an individual's weight, where *CI* represents contaminant ingestion, and *AD* represents the absorbed dose through the skin.

It is assumed that a quantity of contaminant is accidentally ingested and is then averaged over time, estimating exposure using the following expression:

$$CI = \frac{CS \times EF \times ED \times IR_0 \times RBA \times CF_1}{BW \times AT} \tag{1}$$

The meaning of the parameters involved in the calculation, as well as the characteristic values for the health risk analysis of a common industrial scenario that is commonly used in the cited scientific literature, are given in Table 1.

Table 1. Parameter values used in the calculation [3].

Parameter Code	Parameter Definition	Value
EF	Exposure frequency; EF (days/year)	240
ED	Exposure duration; ED (years)	40
IR_0	Ingestion rate for soil; IR_0 (mg/day)	100
RBA	Relative bioavailability factor; RBA (unitless) As = 0.6, Hg = 1.0	0.6/1.0
CF_1	Conversion factor; CF_1 (10^{-6} kg/mg)	1.0×10^{-6}
SA	Skin surface area—soil contact; SA (cm^2)	3300
AF	Soil-to-skin adherence factor; AF (mg/cm^2/day)	0.20
ABS_d	Dermal absorption fraction; ABS_d (unitless) As = 0.03, Hg = 0.01	0.03/0.01
BW	Body weight; BW (kg)	70
AT	Averaging time for carcinogens; AT (days) = 70 years = 70 × 365 days	70 × 365
AT	Averaging time for no carcinogens; AT (days) = ED × 365 days	ED × 365
ET	Exposure time; ET (h/h) = working shift = 8 h/24 h	0.33

In the case of dermal contact with the contaminant, it is assumed that a quantity of contaminant enters the body of a worker through the skin and is averaged over time:

$$AD = \frac{CS \times EF \times ED \times SA \times AF \times ABS_d \times CF_1}{BW \times AT} \tag{2}$$

In inhalation cases, the goal is to estimate the average concentration of contaminant in the air over time in mg/m^3, separately estimating pulmonary exposure to particulate matter and exposure to volatile substances, PE_p and PE_v, respectively:

$$PE_p = \frac{CS \times EF \times ED \times ET \times \left(\frac{1}{PEF}\right)}{AT} \tag{3}$$

$$PE_v = \frac{CS \times EF \times ED \times ET \times \left(\frac{1}{VF}\right)}{AT} \tag{4}$$

PEF relates contaminant concentration in soil to contaminant concentration in airborne suspended particles. PEF (m^3/kg) is the particulate emission factor, with 1/PEF being the average particulate-matter concentration in the air in kg/m^3. Consequently, the quotient CS/PEF represents the contaminant concentration in air in mg/m^3.

For solids containing substances that sublime, VF (m^3/kg) is used. If the soil itself sublimes, we relate contaminant concentration in the soil to contaminant concentration in the air in gaseous form. Thus, the quotient CS/VF represents the contaminant concentration in air in mg/m^3.

The US EPA provides values for all parameters involved in the estimation or indicates how they can be calculated. Equations can be left in terms of the variables measured on-site: contaminant concentration in solids, CS (mg/kg); contaminant concentration in ambient dust, CS/PEF (mg/m^3; and volatile contaminant concentration in air, CS/VF (mg/m^3):

$$CI = \frac{EF \times ED \times IR_O \times RBA \times CF_1}{BW \times AT} \times CS \tag{5}$$

$$AD = \frac{EF \times ED \times SA \times AF \times ABS_d \times CF_1}{BW \times AT} \times CS \tag{6}$$

$$PE_p = \frac{EF \times ED \times ET}{AT} \times \frac{CS}{PEF} \tag{7}$$

$$PE_v = \frac{EF \times ED \times ET}{AT} \times \frac{CS}{VF} \tag{8}$$

In our case, it is more appropriate to use these equations, as the study area and the facilities are relatively small in size and do not have homogeneous soil. Moreover, the contaminant concentration in air at a specific location is not dependent on the soil concentration at that same location. Indeed, there is a very specific area, the demolition debris zone, where gaseous mercury is being produced and dispersed to other parts of the facilities. Therefore, except for this focal point, there is no relationship between the concentration of mercury in the environment and the soil.

Taking the above into account, the following expressions were used in each zone:

$$\frac{CS}{PEF} = C_{air-p} \tag{9}$$

$$\frac{CS}{VF} = C_{air-v} \tag{10}$$

It should be noted that chimneys may also act as emission sources, but they are further away from the facilities, and their influence is not relevant.

Based on the parameter values, it can be deduced that for oral exposure estimation, it is assumed that a contaminant ingestion of IR_0 = 100 mg/day occurs accidentally.

Similarly, in the case of contact with the contaminant, it is assumed that a quantity of contaminant, $SA \times AF \times ABS_d$, enters the body of a worker through the skin, depending on the skin's ability to absorb that contaminant. That quantity is 19.8 mg/day in the case of arsenic and 6.6 mg/day in the case of mercury.

In order to determine the long-term cancer risk (over a worker's entire life) due to exposure to a carcinogenic contaminant, the value is measured using the cancer risk index, *CR*. This index is dimensionless and is calculated for each of the contaminant entry pathways into the body using the following expressions:

$$CR_O = CSF_O \times CI \tag{11}$$

$$CR_d = CSF_d \times AD = \frac{CSF_O}{ABS_{GI}} \times AD \tag{12}$$

$$CR_{inh-p} = IUR \times PE_p \tag{13}$$

$$CR_{inh-v} = IUR \times PE_v \tag{14}$$

The total cancer risk for a substance is calculated as the sum of all risks:

$$CR = CR_O + CR_d + CR_{inh-p} + CR_{inh-v} \tag{15}$$

The total risk when there are multiple substances acting on the same target organ is equal to the sum of the risks due to all substances:

$$Total\ CR = \sum CR_i \tag{16}$$

In Spain, the limit value for *CR* for a single carcinogenic agent is 1×10^{-5} (Royal Decree 9/2005) [29], whereas according to the US EPA, the limit value of 1×10^{-4} is accepted in the case of multiple carcinogenic contaminants acting on the same target organ.

In the case under study, of the two contaminants analysed, only As is potentially carcinogenic, so the reference value is 1×10^{-5}.

The CR index measures cancer risk. When CR > 10^{-4}, a risk of cancer exists; when $10^{-6} \leq CR \leq 10^{-4}$, the risk of cancer is considered to be within an acceptable range; and when CR < 10^{-6}, the risk is considered negligible and common activities can be carried out. In the case of no cancer occupational diseases, the index used is the hazard index, HI. When HI > 1, there is a risk to people's health [3].

Table 2 provides the parameter values for As, as recommended by the US EPA and commonly accepted in the scientific literature.

Table 2. Parameter values used in the calculation.

Parameter Code	Parameter Definition	Value
CSF_O	Oral cancer slope factor; CSF_O (mg/kg/day)$^{-1}$	1.5
ABS_{GI}	Gastrointestinal absorption factor; ABS_{GI} (unitless)	1
IUR	Inhalation unit risk; IUR (mg/m^3)$^{-1}$	4.3

On the other hand, the risk of developing an occupational disease due to exposure to a non-carcinogenic contaminant during a worker's entire working life is measured by the *HQ* (hazard quotient) indices. These indices are dimensionless and are calculated for each of the entry pathways using the following expressions:

$$HQ_O = \frac{1}{RfD_O} \times CI \tag{17}$$

$$HQ_d = \frac{1}{RfD_d} \times AD = \frac{1}{ABS_{GI} \times RfD_O} \times AD \tag{18}$$

$$HQ_{inh-p} = \frac{1}{RfC} \times PE_p \tag{19}$$

$$HQ_{inh-v} = \frac{1}{RfC} \times PE_v \tag{20}$$

The total *HI* (hazard index) risk for a contaminant is calculated as the sum of the risk indices for each of the pathways:

$$HI = HQ_O + HQ_d + HQ_{inh-p} + HQ_{inh-v} \tag{21}$$

Finally, if there are multiple contaminants, the total risk index is calculated as the sum of all the risk indices:

$$Total\ HI = \sum HI \tag{22}$$

The limit value for the *HQ* and *HI* indices is 1. When one of these *HQ* indices, or the overall *HI* index, exceeds 1, it is expected that adverse toxic effects on the target organ or system (the organ affected by the contaminants) may occur [30].

As mentioned, only As and Hg were analysed in the case study. Table 3 provides the parameter values for As and Hg, which are recommended by the US EPA and are commonly accepted in the scientific literature.

Table 3. Parameter values used in the calculation.

Parameter Code	Parameter Definition	Value
ABS_{GI}	Gastrointestinal absorption factor; ABS_{GI} (unitless) As = 1.0; Hg = 0.07	1.0/0.07
RfD_O	Oral reference dose; RfD_O (mg/kg/day) As = 3×10^{-4}; Hg = 3×10^{-4}	3×10^{-4}
RfC	Inhalation reference concentration; RfC (mg/m^3)—As = 1×10^{-5}; Hg = 3×10^{-4}	$1 \times 10^{-5}/3 \times 10^{-4}$

3. Results and Discussion

3.1. Obtention and Elaboration of the Data

The results of the soil or solid material analysis obtained from 46 sampling points can be seen in Figure 2.

Tables 4–7 contain the values of the specific parameters for the CS site, C_{air-v}, and C_{air-p} from Equations (5)–(10).

Table 4. Arsenic and mercury concentration in soil for CS in mg/kg.

	Lower Waste Dump		Lower Platform		Upper Waste Dump		Chimneys		Demolition Debris		Other Zones	
	As	Hg	As	Hg	As	Hg	As	Hg	As	Hg	As	Hg
N	5	5	6	6	7	7	4	4	11	11	13	13
Min	11,089	454	1074	243	2594	240	12,906	25,114	29,644	530	36	2
Max	26,776	4991	73,374	4991	38,841	7266	445,171	250,842	603,337	41,498	8958	538
Aver	17,159	1708	51,810	2731	16,107	3646	215,223	87,476	136,130	17,199	2386	94
SD	6960	1928	27,206	1840	13,312	2645	177,818	109,054	172,072	13,175	2792	138

Table 5. Mercury concentration in air; C_{air-v} in mg/m^3.

	Lower Waste Dump	Lower Platform	Upper Waste Dump	Chimneys (Estimated)	Demolition Debris	Other Zones
N	40	24	22	42	42	130
Min	3.4×10^{-5}	1.0×10^{-5}	3.0×10^{-5}	1.0×10^{-2}	5.1×10^{-3}	1.0×10^{-5}
Max	7.9×10^{-3}	4.4×10^{-4}	1.3×10^{-3}	1.2×10^{-1}	5.8×10^{-2}	7.4×10^{-3}
Aver	8.0×10^{-4}	7.8×10^{-5}	2.8×10^{-4}	4.6×10^{-2}	2.3×10^{-2}	8.9×10^{-4}
SD	1.3×10^{-3}	9.1×10^{-5}	2.9×10^{-4}	3.3×10^{-2}	1.7×10^{-2}	1.4×10^{-3}

Table 6. Arsenic and mercury concentration in airborne dust; $C_{air\text{-}p}$ in mg/m^3.

	Lower Waste Dump	Lower Platform	Upper Waste Dump	Chimneys	Demolition Debris	Other Zones
As in air (mg/m^3)	5.90×10^{-5}	5.40×10^{-4}	5.90×10^{-5}	5.40×10^{-4}	5.40×10^{-4}	3.30×10^{-5}
Hg in air (mg/m^3)	1.66×10^{-4}	1.66×10^{-4}	1.66×10^{-4}	4.50×10^{-4}	4.50×10^{-4}	8.00×10^{-5}

Table 7. Cancer Risk (CR) index value associated with As.

Cancer risk	Lower Waste Dump	Lower Platform	Upper Waste Dump	Chimneys	Demolition Debris	Other Zones
Oral CR_O	8.3×10^{-3}	2.5×10^{-2}	7.8×10^{-3}	1.0×10^{-1}	6.6×10^{-2}	1.2×10^{-3}
Dermal CR_d	2.7×10^{-3}	8.3×10^{-3}	2.6×10^{-3}	3.4×10^{-2}	2.2×10^{-2}	3.8×10^{-4}
Inhalation CR_{inh}	3.1×10^{-5}	2.9×10^{-4}	3.1×10^{-5}	2.9×10^{-4}	2.9×10^{-4}	1.8×10^{-5}
Total CR	1.1×10^{-2}	3.4×10^{-2}	1.0×10^{-2}	1.4×10^{-1}	8.8×10^{-2}	1.6×10^{-3}

Table 4 provides representative statistical values for the concentration of arsenic and mercury in the soil: sample size, N; minimum, Min; maximum, Max; mean, Aver; and standard deviation, SD.

The data clearly show that the zones with the highest concentration of mercury and arsenic are those of the metallurgical plant, with the roasting furnace and the chimneys. This is because these concrete structures were in contact with the fumes produced and became impregnated with mercury and arsenic.

In the two waste dumps, the levels of As and Hg are very high, although they do not reach the contamination level of the two previous zones. However, beneath the lower waste dump, there is a platform where the concentration of arsenic is significantly higher than in the waste dumps, making it a special zone. This is because it collects all the leachates produced by the waste dump, and during dry periods, arsenic salt precipitates occur due to water evaporation.

Finally, in the other zones (mainly associated with transit areas and buildings), the concentrations of Hg and As are several times lower.

It is worth noting that there was a single point with a high concentration of As far from the most contaminated areas; this was the site of the arsenic loading station.

On the other hand, the concentration of mercury in the air was measured at different points within the facilities (Rodríguez et al. [28]; Rodríguez et al. [31]). Table 5 presents the average concentration of gaseous mercury in the air in the different zones. No measurements were taken in the chimney area, and there is no data on Hg in that area, so an estimate will be used by assuming that the concentration of Hg in the air in the chimney area is about twice that of the demolition debris area. This is a very conservative value because this area is more exposed to the influence of wind than the demolition debris.

It should be emphasised that the concentration of mercury in air is only linked to the mercury present in the soil in the demolition debris zones and the chimneys, which act as emission sources. In the other zones, practically all the mercury in the air comes from these sources.

Regarding the concentration of As and Hg in suspended dust, it is worth noting that very little dust is generated at this site. Measurements conducted in the area yielded an airborne dust concentration ranging from 0.08 to 0.15 mg/m^3 [27].

As a result, the measurements of arsenic and mercury in airborne dust were also low. As described by García et al. [27], three measurement campaigns were carried out: in the demolition debris area, in a nearby area, and in the upper waste dump area. No further control campaigns were deemed necessary, so these measurements are considered for the data in the calculations according to Table 6.

3.2. Analysis of the Results

Following the methodology described, arsenic was first analysed as a potentially carcinogenic element. Table 7 presents the results of the calculation of the total cancer risk index, *CR*. As observed, the values are several orders of magnitude higher than the reference limit of 10^{-5} in Spain. Consequently, it can be inferred that under no circumstances could normal industrial activity be conducted over a full 40-year working life.

However, as will be seen later, in less contaminated areas (e.g., transit zones) and under certain restrictions, the risk decreases to values below the limit.

In order to complete the risk analysis, arsenic and mercury were analysed as pollutants to which exposure poses a risk of occupational diseases (non-cancer). Considering they do not affect the same target organ, the analysis was conducted separately and the corresponding *HI* indices are not summed.

Table 8 shows the health risks associated with the presence of As in the soil for different areas and the health risks associated with the presence of mercury, both in the soil and in the atmosphere, in different zones.

Table 8. Health Risk (HI) index value associated with As and Hg.

Hazard Quotient	Lower Waste Dump		Lower Platform		Upper Waste Dump		Chimneys		Demolition Debris		Other Zones	
	As	Hg	As	Hg	As	Hg	As	Hg	As	Hg	As	Hg
Oral HQ_O	32.2	5.3	97.3	8.6	30.3	11.4	404.3	273.9	255.7	53.9	4.5	0.3
Dermal HQ_d	10.6	5.0	32.1	8.1	10.0	10.8	133.4	258.2	84.4	50.8	1.5	0.3
Inhalation HQ_{inh}	1.3	0.7	11.7	0.2	1.3	0.3	11.7	33.8	11.7	17.1	0.7	0.7
Total HI	44.2	11.1	141.2	16.8	41.5	22.5	549.5	565.9	351.9	121.7	6.7	1.3

The reference value for *HI* is 1.0; however, in practically all zones, the *HI* index is several times higher. Therefore, it is concluded that there is also an unacceptable risk of non-cancerous occupational diseases in all zones.

It should be noted that the greatest health risk to workers is associated with high concentrations of arsenic rather than mercury. Nevertheless, the volatility of mercury and the possibility of encountering high concentrations of Hg in the environment are unique features of these facilities that significantly affect the work.

The analysis of the results allows for the distinction of three main areas based on the risk indices. This division is perfectly coherent with our understanding of the nature and origin of these areas, the working conditions within them, and the tasks undertaken in the SUBproducts4LIFE project.

Figure 3 illustrates the three major zones defined based on CR and HI. As mentioned earlier, it is noteworthy that there is a point with an extremely high health risk within a zone of lower risk. This is because it is a singular point where the former arsenic loading platform was located.

In Table 9, the three ranges of variations in the selected CR and HI indices are defined.

Table 9. Definition of the three areas based on the CR and HI indices.

Area	*CR* Range	*HI* Range
High risk to health (Green) (Area F)	$1.5 \times 10^{-4} < CR < 1.5 \times 10^{-3}$	$0 < HI < 10$
Very high risk to health (Brown) (Areas C and D)	$1.5 \times 10^{-3} < CR < 1.5 \times 10^{-2}$	$10 < HI < 100$
Extremely high risk to health (Red) (Areas A and D)	$1.5 \times 10^{-2} < CR < 1.5 \times 10^{-1}$	$100 < HI < 1000$

There is a less critical zone (in green in Figure 3), although it remains a high-risk area in general, as the indices exceed the *HI* > 1 and *CR* > 10^{-5} limit values. This means that, in this zone, regular work or industrial activity could not be carried out over a 40-year working life. However, conditions could be found where the *CR* and *HI* values decrease below 10^{-5} and 1, respectively.

For instance, in the framework of a research project (SUBprofucts4LIFE), work could be conducted for 6 months (120 days) in areas where the concentration of As and Hg in the soil is below 500 mg/kg and 200 mg/kg, respectively, and the concentration of Hg in the air is below 10^{-3} mg/m^3. Recalculating the former values using these values yields $CR = 4.2 \times 10^{-6}$ for As and $HI = 0.98$ and $HI = 1.0$ for As and Hg, respectively.
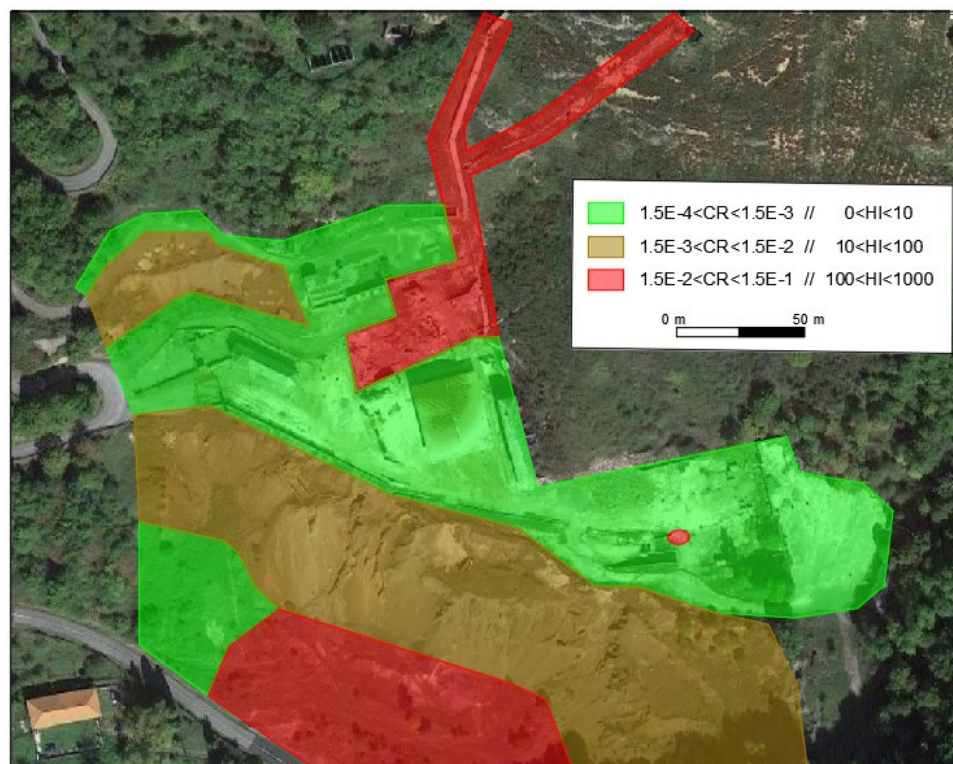

Figure 3. Division of the study area based on risk.

An example of such an area is the entrance zone to the facility (Figure 4A). The average concentrations of As and Hg in the soil in the transit zones are 138 and 77 mg/kg, respectively, and the average concentration of Hg in the air throughout the year is 6.1×10^{-4} mg/m^3. In theory, work could be conducted for up to 6 months without special protection. However, compliance with regulations requires the use of personal protective equipment (PPE), such as dust masks, gloves, boots, helmets, and so on.

This is coherent with the fact that some industrial activity was carried out in the past, with acceptable values for CR and HI, considering that this activity was developed 20 years ago (before establishing the legal limits).

There are other areas (brown colour in Figure 3) where the soil is contaminated with arsenic and mercury but the concentration of mercury in the air is not too high; specifically, less than 10^{-3} mg/m^3. Normal activity cannot be conducted in these areas; they involve special tasks where workers must wear more protection, such as full-body suits to protect the skin from contact with materials, or special gloves. However, work can be carried out in these areas for the entire day (8 h/day) and throughout the year (240 days/year). An example of this is presented in Figure 4B, where a worker is shown collecting samples in the upper waste dump, where the average concentrations of As and Hg in the soil are 16,107 and 3646 mg/kg, respectively, and the average concentration of Hg in the air throughout the year is 5.1×10^{-4} mg/m^3.

Finally, there are two areas (highlighted in red in Figure 3) where, for different reasons, the risk is extremely high. One of them is the platform at the foot of the lower waste dump, where the precipitation of arsenic salts results in much higher concentrations of this element than in other areas.

Another is the demolition debris areas of the metallurgical plant along with the smoke ducts and chimneys which, due to high concentrations of gaseous mercury in the air, form another critical area in terms of health risks to workers. This is due to the extremely high concentrations of Hg in the air, for which the use of the PPE, as mentioned earlier, is not sufficient, and a half-face mask must also be used for protection against Hg gases; here, work must be conducted according to a strict safety protocol that limits working hours based on temperature, as described by Rodríguez et al. [31].

Figure 4. Work in areas with different risk levels.

This situation is reflected in Figure 4C, where work is being carried out in the demolition debris area. As observed, the average concentrations of As and Hg in the soil can reach values as high as 136,160 and 17,199 mg/kg, respectively, and the average concentration of Hg in the air throughout the year is 1.3×10^{-2} mg/m^3. In these areas, the work shift is at most 6 h/day at temperatures below 15 °C, which can be reduced to 3 h/day at a maximum temperature of 25 °C.

As described above, the As and Hg concentrations in the soils in different parts of the site are clearly related to the mining activities developed over the life of the mine. The paragenesis of the ore deposit is composed of cinnabar, orpiment, realgar, pyrite (usually with high concentrations of As), arsenopyrite, marcasite, and pararealgar in a gangue of quartz and calcite [32].

First, the mineral had to be mined out from the rock mass. In this process, two materials were produced: one rich with a high concentration of cinnabar, which was sent to the metallurgical process, and the other poor in Hg, which was stockpiled in a waste dump. This waste dump, in the lower part of the site, is an area with high As and Hg contamination, although it is not the most contaminated area. We should keep in mind that both As and Hg are chemically combined in the minerals (they are sulphates); with this, the mobility and the contaminating potential are lower than in other areas.

The rich minerals were collected at the metallurgical plant, where As and Hg were produced on one side and waste was produced on the other. As the efficiency was not 100%, part of the As and Hg remained in this waste, which was stored in a waste dump in the upper part of the site. The concentration of As and Hg is of the same order of magnitude as in the other waste dump.

The metallurgical plant is divided into two different zones. At first, there were the buildings and structures that supported the roasting furnace. At present, only the debris from the demolition of these structures and buildings is located in this zone. In the other zone, there are the ducts and the chimneys for the smoke. These two zones are the ones

with the highest concentrations of mercury and arsenic. This is because these concrete structures were in contact with the fumes produced in the metallurgical process and became impregnated with mercury and arsenic.

There is a singular area—the platform in the lower part of the site—where the concentration of arsenic is significantly higher than in the waste dumps, making it a special zone. This is because it collects all the leachates produced by the waste dump and, during dry periods, arsenic salt precipitates occur due to water evaporation.

Finally, in the other zones, mainly associated with transit areas and buildings, the concentrations of Hg and As are several times lower than in the previous areas. In these areas, the contamination was produced by the dispersion of PTEs existing in the other areas.

A number of studies have been carried out recently using the US EPA model, demonstrating that it is useful under different conditions. A summary of the research is shown in Table 10, which lists the areas that pose a low or high risk to the health of adults. In general, it analysed the effect of more PTEs than As and Hg; although, in all cases, the weight of As in the total values of the indices for cancer risk (CR) and non-cancer illness (HI) is significant.

Table 10. Summary of recent research that used the US EPA methodology.

Case	Country	Site	As (mg/kg)	Hg (mg/kg)	CR	HI	Reference
1	Poland	Agricultural soils	1.40–16.6	0.00–0.52	1.41×10^{-5}	0.103	[7]
2	Poland	Former chemical plant	2.72–3.26	0.05–0.07	2.8×10^{-7}–4.2×10^{-5}	2.2×10^{-4}–4.6×10^{-2}	[11]
3	China	Industrial area	0.4–70.6	-	1.3×10^{-4}	1.69×10^{-2}	[8]
4	Peru	Agricultural soils	21.1–28.7	-	3.4×10^{-4}	3.6×10^{-1}	[9]
5	China	Automobile industry	88.25–1994.75	-	6.3×10^{-4}	6.4×10^{-5}–1.4×10^{-3}	[33]
6	China	Mercury smelting site	67.42	358.51	1.2×10^{-6}–3.4×10^{-4}	0.37–43.56	[6]
7	Panama	Abandoned gold mine	35.5–5030	0.06–1.37	1.1×10^{-4}–3.5×10^{-3}	1.51–11.54	[34]
8	China	Metalliferous, industrial district	0.54–3895	0.00–3.29	7.1×10^{-7}–3.2×10^{-2}	0.29–211	[13]
9	Spain	Former mercury mine	13–392,238	0.7–4498	8.5×10^{-6}–2.5×10^{-1}	0.1–1000	[12]

The results show that there are two different scenarios. The first is related to agricultural activities or general industry (Cases 1 to 5). In general terms, the contamination is moderate, and the CR and HI indices have acceptable values.

The second is related to mining activities (Cases 6 to 9), gold mines (in which Hg was used in the past), and former mercury mines. Under these conditions, both cancer and non-cancer illness risks are over the legal limits. The worst conditions (Case 9) are present in former abandoned mercury mines and facilities in which the waste from metallurgical processes and debris from the demolition of metallurgical plants are present. The case studied here is similar to this last case.

Only in Case 9 (related to the Terronal Mine in Spain) was there a comparable situation to that of La Soterraña concerning the elevated CR and HI index values. When comparing the CR values associated with arsenic from La Soterraña Mine (Table 7) with Terronal Mine in Mieres (Asturias) [12], notable differences emerge. In Terronal, there are two areas (25 × 25 m) with CR levels of 2.5×10^{-1}, which is significantly higher than the maximum observed at La Soterraña, although it is still within the same order of magnitude as the chimneys, which register levels of 1.4×10^{-1}. In La Soterraña, the second focus of concern lies in the demolition debris, with CR levels reaching 8.8×10^{-2}. Interestingly, in Terronal, there is a grid exhibiting similar levels to those found in La Soterraña, with a CR value of 3.2×10^{-2}.

4. Conclusions

The analysis of results revealed significantly elevated health risks associated with arsenic (As) and mercury (Hg) contamination across different zones within the study area. These risks exceeded the reference limits for both the Cancer Risk (CR) and Hazard

Index (HI) indices, indicating that normal industrial activities cannot be conducted safely within a typical 40-year working lifespan due to the potential carcinogenic effects of As exposure. Furthermore, the HI values for both As and Hg indicate substantially increased risks of non-cancerous occupational diseases, with the majority of zones surpassing the acceptable threshold.

Three major risk areas were identified based on the CR and HI indices. These delineations were consistent with the nature and origin of the areas, the working conditions, and the tasks conducted within the SUBproducts4LIFE project. Zones with lower but still significant risks may permit limited industrial activities under specific conditions, such as reduced exposure duration and enhanced personal protective equipment (PPE) requirements.

Areas characterised by extremely high risks necessitate stringent safety measures and specialised PPE, due to exceptionally high concentrations of As and Hg (either in the soil or air). Work in the most hazardous zones, particularly those with extremely high risks, requires strict adherence to safety protocols, including restricted work hours based on environmental conditions and the use of advanced respiratory protection against Hg gases.

Overall, the reported findings underscore the imperative of implementing robust safety measures to mitigate health risks associated with exposure to As and Hg in various occupational environments, ensuring the well-being of workers and regulatory compliance.

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