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Rethinking soil contamination assessment for urban brownfield management: from total hazard to bioavailability

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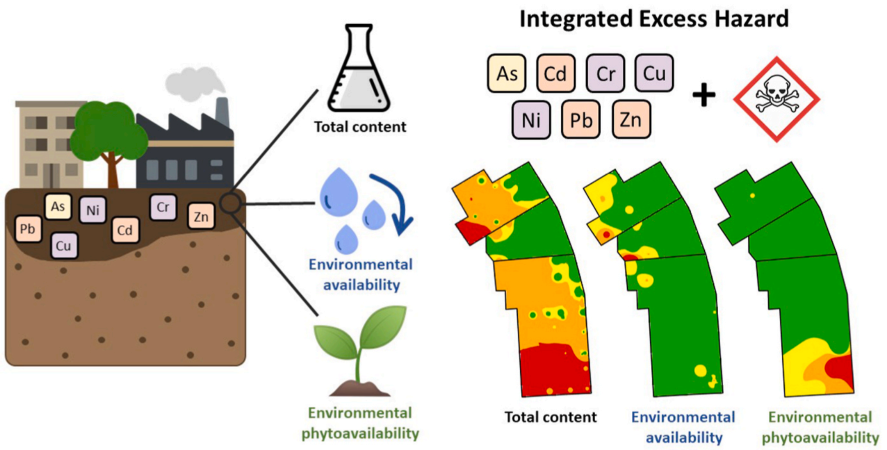
HIGHLIGHTS

- Hazard of metals in soil may be broken down into total, available and bioavailable fractions.
- Defining whether a site is contaminated depends on how the hazard is assessed.
- Accurate hazard identification requires knowledge of local background variation.
- Using hazard indices helps to better anticipate the risks at contaminated sites.

GRAPHICAL ABSTRACT



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ABSTRACT

Hazard identification at contaminated sites usually rely on total concentrations of potentially toxic elements (PTE), but it is largely admitted that only the bioavailable fraction can trigger adverse effects. Assessing total PTE amounts is thus a worst-case scenario, that overestimates environmental risks and often leads to costly remediation actions. This study presents a practical and cost-effective methodology for assessing soil contamination by PTE, taking into account their bioavailability. To address this issue, we performed a three-tiered hazard identification considering: (1) total hazard (total PTE content), (2) environmentally available hazard ($CaCl_2$ leachable fraction), and (3) environmentally bioavailable hazard (accumulation in plant leaves). Additionally, an Integrated Hazard Excess (IHE) index was computed, for aggregating raw contaminant levels into a single integrated hazard value. The methodology was applied to a former industrial site in Saint-Étienne, France. Soil and

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plant samples were analyzed for As, Cd, Cr, Cu, Ni, Pb, and Zn. Mapping of IHE values provided a clear and holistic view of contamination across the site. Results showed that while total PTE levels were very high, the mobile and bioavailable fractions were overall within the range of local background observed in urban parks used for recreational activities. This indicated that actual environmental risk was significantly lower than total concentrations suggested. Thus, distinguishing total, available, and phytoavailable PTE amounts provided a more accurate and ecologically meaningful assessment of soil hazard. This approach could help decision-making and support the adoption of less disruptive and more sustainable remediation strategies, avoiding unnecessary excavation and landfill operations.

1. Introduction

Potentially toxic elements (PTE), such as As, Cd, Cr, Cu, Ni, Pb and Zn, are common by-products of many industrials processes and have long been widely released into the environment. Since they are not biodegradable, PTE accumulated in soils, notably in urban and peri-urban areas, where they now constitute a serious threat for ecosystems and human health (Nieder and Benbi, 2024). Indeed, beside they eco-toxicity (Ding et al., 2022), PTE can cause a number of adverse effects for human health, such as nervous system disorders, cardiovascular and kidney damages, birth defects, and various types of cancer (FAO and UNEP, 2021; Jomova et al., 2025). As a result, soil pollution with PTE is today considered as a major concern by stakeholders and decision makers involved in land rehabilitation and urban governance.

In fact, one of the main challenging issues for decision makers involved in the management of contaminated sites, lies in an accurate characterization of soil contamination (MNGSSP, 2017). Indeed, each polluted site is a special case by nature and is often highly heterogeneous, with respect to the type of pollutants, their concentrations and their spatial distribution. This implies that numerous samples have to be taken and analyzed, both on the surface and at depth, for a precise spatialized hazard identification designed for the site's typology. As a rule, analyses of PTE in soil samples are only carried out to quantify their total contents, *i.e.* to assess total hazard in the soil matrix. The underlying assumption justifying this approach is that, potentially, all of an element present in the soil can move to reach a sensitive target, such as groundwater or living organisms. However, as opposed to air and water, soil is an anisotropic environment (Hartemink, 2016), as it is made of numerous components such as minerals of various sizes and composition (*e.g.* clays, loams and sands, carbonates, Fe/Mn oxide and hydroxide minerals), water, gases, and organic matter. Thus, contaminants are never free to displace in the soil, as they do in air or water, but they are instead more or less strongly bound to the different soil's organo-mineral particles. This refers to the so-called chemical and mineralogical speciation of metals in soil (Manceau et al., 2002; Ure and Davidson, 2008). Consequently, a given total amount in one element can reflect very different situations regarding its mobility, ranging from virtually insoluble species to water soluble and highly mobile forms (Reeder et al., 2006; Li et al., 2022). When considering these two extreme cases, it is obvious that although the estimated hazard (*i.e.* total content) is the same, the risk for a detrimental effect toward living organisms will be quite different. Consequently, simply measuring total contents of PTE in soil is a worst-case scenario, which generally leads to overestimating the actual hazard, thereby increasing the associated risk. The main drawback of this protective approach is that most decisions for site reclamation finally result in "dig and dump" actions aiming at mitigating total hazard (Liu et al., 2018), while alternative strategies that could allow soil preservation are seldom considered. Thus, although very safe for public health, soil management plans solely taken from total amounts of PTE are leading to a significant increase in reclamation's costs, both economically and environmentally (Espinoza-Tofalos et al., 2025).

In this context, there is a need today for developing complementary decision support tools, aiming at better accounting for the chemical and mineralogical speciation and, ultimately, for the fate of PTE in hazard identification. However, PTE speciation relies on multiple parameters (*e.g.* pH, type of organic matter, grain size, cation exchange capacity, Redox potential) that those analyses usually performed for site characterization cannot take into account in routine assessment procedures. By contrast, this issue could be operationally addressed using the concept of bioavailability (Scattolin et al., 2021; Sun et al., 2023; Chen et al., 2023; Li et al., 2024). Although numerous definitions can be found in the literature to explain this concept (Semple et al., 2004; Hodson et al., 2011; Kim et al., 2015), the International Organization for Standardization proposed to consider the following: "*bioavailability is the degree to which chemicals present in the soil can be absorbed or metabolized by human or ecological receptors or are available for interaction with biological systems*" (ISO 17402, 2008; ISO 22190, 2020). Hence, according to this definition, beside total hazard, an accurate evaluation of site contamination should also consider at least two other levels for hazard identification: that of PTE easily transferable into the soil solution (*available for interaction with biological systems*), and that of PTE actually accumulated in living organisms inhabiting the site (*absorbed by ecological receptors*). These two shades of hazard are the so-called environmental availability and environmental bioavailability, respectively (ISO 17402, 2008).

To date, numerous methods using weak chemical extractions have been proposed to specifically assess the most labile fractions of PTE in soils (Kim et al., 2015; ISO 17402, 2008; ISO 22190, 2020). These methods are rapid and easy to use and are indeed efficient tools to address the issue of the potential mobility of PTE. However, though these approaches were long presented, and still are (ISO 22190, 2020), as proxies for bioavailability, extracted PTE generally poorly correlate with those actually accumulated in living organisms in field situations (Remon et al., 2013; Pauget et al., 2015). Thus, chemical weak extractions only help to quantify easily leachable PTE fractions, but can seldom anticipate their contents in biological receptors, which depend on complex eco-physiological parameters. Consequently, chemical weak extractions just highlight one shade of the hazard, that of its environmental availability.

On the other hand, determining PTE accumulated in living organisms highlights the fractions of total contaminants that are actually absorbed by ecological receptors, *i.e.* the environmental bioavailability of PTE, which is another shade of hazard. However, assessing environmental bioavailability is not so obvious, since PTE accumulated in living organisms not only depend on the speciation of the element considered, but also on the target species, its exposure pathways and its trophic level. This means that the environmental bioavailability of PTE must not be seen as an intrinsic soil characteristic, but rather as the result of interactions between soil contaminants and communities of organisms living here, at a given time. Consequently, it makes little sense to consider a single species to assess the bioavailability of PTE on a contaminated site. By contrast, studying communities of organisms is a more relevant approach. In this respect, analysis of composite plant samples is an efficient way to estimate environmental phytoavailability (Remon et al., 2013), which is that part of environmental bioavailability associated with the whole plant compartment. However, the use of plant communities as indicators of PTE mobility (accumulation indicators) still deserves development before standardized protocols can be put forward.

In addition, besides partitioning the hazard into three different shades, data from contaminated sites should be communicated in the most comprehensive but concise form, so that stakeholders and decision makers can understand them as easily as possible. In this aim, data mapping for spatial visualization is of great interest, but it is not sufficient. Indeed, because several PTE are generally present at contaminated sites, and since each PTE has its own toxicity, it is quite difficult to unambiguously interpret contamination maps when expressed in concentration units for numerous elements. Hence, converting raw data from multi-contamination maps into a single spatial integrated hazard index would be of great interest for decision makers, to rapidly and accurately score various plots in the same site, or various sites in the same regional area.

In this context, this study had three main objectives: 1/to investigate the usefulness of layering the hazard into three different shades for site characterization, as obtained from total PTE contents in soil, their environmental availability and their environmental phytoavailability; 2/to define a calculation approach for deriving integrated indices dedicated to each shade of hazard; and 3/to illustrate the usefulness of integrated hazard mapping as a new tool for communication with stakeholders involved in the management of contaminated sites.

2. Material and methods

2.1. Site description

The study site (45°26′33.7″N 4°23′44.9″E, WGS84 coordinate system) was located in Saint-Étienne (Auvergne/Rhône-Alpes region, France). It was an urban area extending over approx. 5700 m^2 (Fig. 1), which was formerly divided into three distinct parcels (EK05, approx. 1400 m^2; EK06, approx. 1100 m^2 and EK25, approx. 3200 m^2) that hosted different industrial facilities from the mid-19th century onward. Although successive activities were not clearly identified, last known occupations were a cycle factory and metal plating facility onto EK05, a waste deposit area at EK06 and a manufacture of metal parts for truck

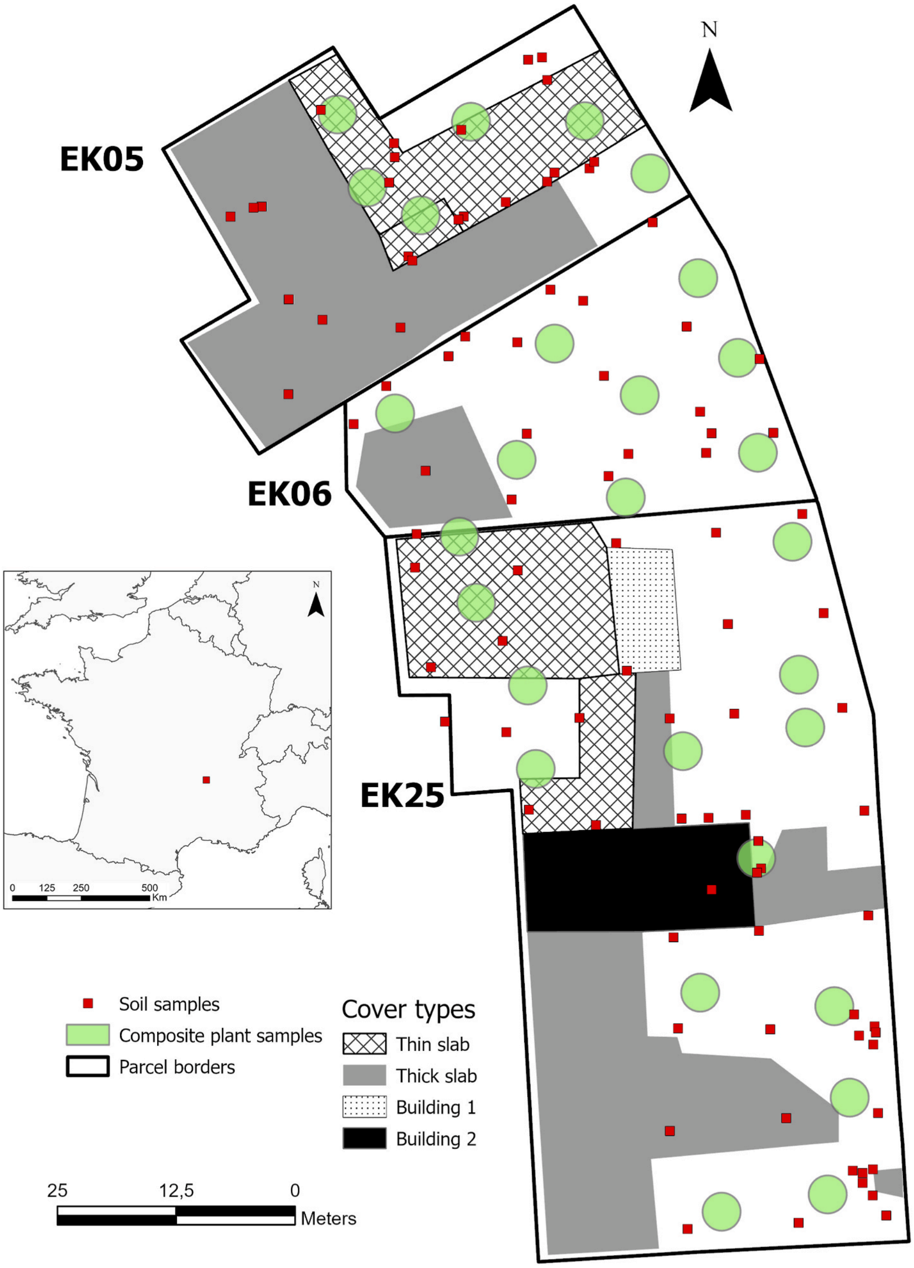

Fig. 1. Map of the study site, showing soil occupation and location of sampling points (red squares: soils samples; green circles: composite plant samples).

industry and steel construction at EK25. All buildings and dividing walls were dismantled by 2020, leaving a large open area showing numerous concrete slabs remaining on the ground. Preliminary soil investigations indicated high levels in several PTE, such as As, Cd, Cr, Cu, Ni, Pb and Zn. However, while soil conditions seemed at first glance very unfavorable for plant development, a rapid floristic survey carried out in spring and summer 2023 showed that a biodiverse plant community was still present, with more than 140 species from 36 different botanical families being identified.

2.2. *Soil sampling design*

Overall, soil samples were taken according to a 10 m × 10 m grid. In sealed areas, samples were collected in broken parts of the slab, when possible. Moreover, some samples were replicated in areas where high levels of PTE were suspected. This resulted in 24 samples at EK05, 21 samples at EK06 and 46 samples at EK25; the location of soil sampling points is given in Fig. 1 (red dots). All samples were taken at 0–20 cm depth. They were oven dried at 60 °C, sieved to pass a 2-mm mesh and stored in close vials at room temperature before further processing.

2.3. *Plant sampling design*

Composite plant samples were taken on areas of approx. 10 m^2. The choice of sampling areas was based on a roughly systematic design, but was constrained by the presence of sealed or highly tamped area, without vegetation. This resulted in 6 composite plant samples at EK05, 8 at EK06 and 14 at EK25; the location of sampling areas is given in Fig. 1 (green circles). At each sampling area, leaves from the 5 most abundant species were individually collected in clean plastic bags. Leaf samples of each species were thoroughly washed with tap water and rinsed with ultra-pure water. They were then oven dried at 40 °C until constant weight, and individually ground (Polymix PX-MFC-90D, Kinematica, Malters, Switzerland) to pass through a 2-mm sieve. The same quantity (routinely 50 mg dry weight) of ground leaves from each of the five species was weighted and mixed into clean 60 mL-capacity PFA digestion vials (Savillex®, Eden Prairie, USA), resulting in a 250 mg composite leaf sample for each sampling area.

2.4. *Extraction of PTE for assessment of total contents in soil*

Aliquots of collected soil samples (approx. 5 g of the < 2-mm soil fraction) were ground to pass a 250-µm mesh. Sub-samples (250 mg) were then digested in aqua regia, according to standardized protocol (NF ISO 11466, 1995). PTE concentrations (As, Cd, Cr, Cu, Ni, Pb and Zn) in the extracts were measured using an inductively coupled plasma mass spectrometry (Agilent 7800 ICP MS, Agilent Technologies, Santa Clara, USA).

2.5. *Extraction of PTE for assessment of environmental availability*

Weakly bound PTE were assessed on the < 2-mm soil fraction according to the NEN 5704 standardized protocol (NEN 5704, 1996). This was a 2 h extraction on a rotary shaker at 40 rpm (Reax 2, Heidolph, Schwabach, Germany), at 20–22 °C, in a 0.01 M calcium chloride solution (Calcium chloride dihydrate 99+% for analysis, Acros Organics, Geel, Belgium), with a solid/liquid ratio of 1:10 (W/V). Extracts were then centrifuged (Sigma 2–16 K centrifuge, Sigma GmbH, Osterode am Harz, Germany) at 8000 rpm for 15 min, and the supernatants were collected for PTE determination by ICP-MS.

2.6. *Extraction of PTE for assessment of environmental phytoavailability*

Composite plant samples (250 mg) were first pre-digested overnight at 60 °C in 2.5 mL sub-boiled distilled concentrated HNO_3 (65 % analytical reagent grade, Fisher Scientific, Illkirch, France) using a hot block digestion system. 500 µL H_2O_2 (Hydrogen peroxide solution >30 % for trace analysis, Merck, Darmstadt, Germany) were then added, and tubes were heated at 90 °C for 1h. The final digestion step was achieved by adding 2.5 mL concentrated HNO_3, then heating at 140 °C for 16 h. Samples were finally evaporated to dryness and solubilized in 37.5 mL 1 % HNO_3 (final solid/liquid ratio of 1:150). Extracts were then analyzed by ICP-MS.

2.7. *Accuracy and quality control of ICP-MS analyses*

ICP-MS analyses were carried out using calibration curves at appropriate concentration ranges, made from certified PlasmaCal standard solutions (SCP Sciences, Baie d'Urfée, Canada). All calibration standards were prepared in the same matrix used for the extracts. For each extraction procedure (soil total, soil $CaCl_2$ and plant leaves), laboratory reagent blanks (LRB) were processed in the same way as samples, through the entire preparation procedure. Method quantitation limits were calculated as mean LRB plus 9-times the standard deviation of at least 10 LRB replicates. The analytical precision was checked by running the same sample every ten analyses; relative standard deviation was routinely less than 5 %.

For quality assurance of soil analysis, the certified reference soil SS-1 (EnviroMAT, SCP Sciences) was used. For quality assurance of plant analysis, the certified reference materials INCT-OBTL-5 (Oriental Basma Tobacco Leaves – Institut of Nuclear Chemistry and Technology, Warsaw, Poland) and ERM-CD281 (Rye grass – European Reference Materials, Geel, Belgium), were used. For each type of matrix (soil total, soil $CaCl_2$-extractable, plant tissues), CRM recoveries were assessed by analyzing these standards alongside samples. Most recoveries ranged between 80 and 120 %, which is in line with internationally accepted quality criteria for trace element analysis (see Supplementary Material Table S1).

2.8. *Calculation of an integrated index for hazard excess*

For each shade of PTE analysis (total content – *Tot* –, extractable content – *Extr* – and plant content – *Phyt* –), and for each element i, raw data of element concentrations were converted into "hazard excess" ratio (HE) as:

$$HE^{i}_{(Tot\ OR\ Extr\ OR\ Phyt)} = T^{i}\left(\frac{C^{i}_{Meas\ (Tot\ OR\ Extr\ OR\ Phyt)}}{C^{i}_{Thr\ (Tot\ OR\ Extr\ OR\ Phyt)}} - 1\right) \tag{1}$$

Where T^i is the toxicity coefficient of element i, $C^{i}_{Meas\ (Tot\ OR\ Extr\ OR\ Phyt)}$ is the measured concentration of element i, and $C^{i}_{Thr\ (Tot\ OR\ Extr\ OR\ Phyt)}$ is the selected threshold value (see *2.10*) for element i, for total content, extractable content and plant content, respectively. Note that in (Eq. (1)) the 1 value was subtracted to the ratio, just to set samples with "no hazard excess" at 0 or below, while samples with "hazard excess" have positive values.

The selected toxicity coefficients where the following (Håkanson, 1980):

$$T^{Zn} = 1;\ T^{Cr} = 2;\ T^{Cu} = T^{Ni} = T^{Pb} = 5;\ T^{As} = 10;\ T^{Cd} = 30.$$

Finally, for each shade of hazard, an integrated hazard excess index (IHE) was calculated for each sampling point, as the sum of individual hazard excess values:

$$\mathrm{IHE}_{(\mathrm{Tot\ OR\ Extr\ OR\ Phyt})} = \Sigma\ \mathrm{HE}^{\mathrm{i}}_{(\mathrm{Tot\ OR\ Extr\ OR\ Phyt})} \tag{2}$$

2.9. *Data mapping*

Interpolation and data mapping were carried out independently for each of the three parcels, using ArcGIS Pro 3.4.3 (Esri, Redlands, USA). We used the inverse distance weighting interpolation method, which is a robust deterministic approach, easy to use with the Geostatistical

Analyst extension of ArcGIS Pro. This method enables the calculation of the expected value for any point in a given space, according to measured neighborhood values. It is worth noting that this method is an exact interpolator, where the measured values are kept at all sampling locations; calculated values are therefore intermediate values.

2.10. Data processing and statistical analyses

Threshold values for total PTE in soil were derived from exploratory data analysis of PTE contents in unpolluted urban parks and gardens (*i.e.* soils allowed for recreational activities) from the conurbation of Saint-Étienne (Seillier et al., 2025). We used the boxplot method (Tukey, 1977; Reimann et al., 2005) to separate "usual" background variations from outliers. Three thresholds were considered: 1) the upper inner fence (*i.e.* 75th percentile plus 1.5-times the interquartile range), which discriminates usual values from anomalous values (“outliers”), 2) the upper outer fence (*i.e.* 75th percentile plus 3-times the interquartile range), which separates moderate anomalies from high anomalies (“far outliers”), and 3) the maximum value of the distribution, to discriminate high anomalies from extreme anomalies (“extreme outliers”).

Threshold values for extractable PTE, phytoavailable PTE as well as for IHE values were computed in the same way, considering upper limits of their background variations in uncontaminated soils. Selected threshold values are given in Table 1.

Comparisons of PTE contents (total, $CaCl_2$ extractable and plant contents) between local background and site’s values were performed using Welch’s *t*-test. Comparisons of PTE contents between the three parcels was done using Kruskal-Wallis rank sum tests. When significant differences were evidenced, post-hoc comparisons were carried out using pairwise Wilcoxon rank sum tests, with Holm’s *p*-adjustment method for multiple comparisons. Differences were stated significant when $p < 0.05$.

All computations and statistical analyses were performed using R software (R Core Team, 2024) under the RStudio environment (Posit team, 2024).

3. Results

3.1. Total contents, environmental availability and environmental phytoavailability of PTE, as compared to local urban control soils

Boxplot representations of total PTE levels, their environmental availability and their environmental phytoavailability are given in Fig. 2; corresponding maps are given in supplementary material (Fig. S1–S7).

For total As (Fig. 2, As, left; Fig. S1), median content in the studied soil was 61 mg/kg. This was not significantly different from the local background (median = 78 mg/kg), and most analyzed samples were in the range of usual content as compared to reference soils. Only small areas at the South-West of EK06 and South-East of EK25 (Fig. S1) showed a few high or extreme anomalies. When the environmental availability of As was considered (Fig. 2, As, center), the median value for the studied soil was slightly, but significantly higher than local background. However, most values were still in the usual range, and only a few points showed high or extreme anomalies. By contrast, the phytoavailable fraction of As (Fig. 2, As, right) was significantly higher than reference value, and ca. 65 % of plant samples exhibited moderate or high anomalies, as compared with control distribution.

For Cd, the site’s median value for total content (Fig. 2, Cd, left; Fig. S2) was 0.68 mg/kg, *i.e.* approx. 2-fold the median level in control soils. However, there was a clear-cut difference between parcels, with EK25 showing both significantly higher total Cd levels and much more high and extreme anomalies than EK05 and EK06. Although no significant difference in Cd content was evidenced between EK05 and EK06, the latter was very close to usual content in uncontaminated soils, and showed only one sampling point with a moderate anomaly. Analyses of the $CaCl_2$ extractable fractions (Fig. 2, Cd, center) and plant contents (Fig. 2, Cd, right) showed that both the environmental availability and the environmental phytoavailability of Cd were globally very low and in the same range as uncontaminated soils; however, a few samples at the South of EK25 showed high or extreme anomalies (Fig. S2).

Table 1

Ranges of variation and thresholds for outlier identification, for total PTE contents, weakly bound PTE contents, and plant contents, in uncontaminated urban soils used for recreational activities in the city of Saint-Étienne.

Element	Threshold	Soil total content[a] (mg/kg)	Soil weakly bound content[b] (mg/kg)	Plant content[c] (mg/kg)
As	Usual background	[10–169]	[0.01–0.26]	[0.02–1.10]
	Moderate anomalies	]169–253]	]0.26–0.39]	]1.10–1.74]
	High anomalies	]253–322]	]0.39–0.44]	]1.74–3.56]
	Extreme anomalies	>322	>0.44	>3.56
Cd	Usual background	[0.09–0.70]	[0.001–0.11]	[0.01–0.90]
	Moderate anomalies	]0.70–0.96]	]0.11–0.17]	]0.90–1.46]
	High anomalies	]0.96–1.85]	]0.17–0.25]	]1.46–2.54]
	Extreme anomalies	>1.85	>0.25	>2.54
Cr	Usual background	[11–61]	[0.002–0.02]	[0.05–1.16]
	Moderate anomalies	]61–81]	]0.02–0.03]	]1.16–1.66]
	High anomalies	]81–189]	]0.03–0.06]	]1.66–4.68]
	Extreme anomalies	>189	>0.06	>4.68
Cu	Usual background	[8–95]	[0.04–0.31]	[1.1–20.5]
	Moderate anomalies	]95–149]	]0.31–0.46]	]20.5–28.3]
	High anomalies	]149–196]	]0.46–0.60]	]28.3–33.4]
	Extreme anomalies	>196	>0.60	>33.4
Ni	Usual background	[9–59]	[0.007–0.77]	[0.1–5.0]
	Moderate anomalies	]59–79]	]0.77–1.23]	]5.0–8.1]
	High anomalies	]79–103]	]1.23–1.66]	]8.1–26.6]
	Extreme anomalies	>103	>1.66	>26.6
Pb	Usual background	[0.1–169]	[0.002–0.41]	[0.05–0.65]
	Moderate anomalies	]169–269]	]0.41–0.64]	]0.65–1.01]
	High anomalies	]269–320]	]0.64–4.77]	]1.01–1.9]
	Extreme anomalies	>320	>4.77	>1.9
Zn	Usual background	[40–235]	[0.02–6.74]	[15.8–112.1]
	Moderate anomalies	]235–345]	]6.74–10.61]	]112.1–158.1]
	High anomalies	]345–530]	]10.61–18.20]	]158.1–188.7]
	Extreme anomalies	>530	>18.20	>188.7

[a] Extracted with aqua regia.
[b] Extracted with 0.01 M $CaCl_2$.
[c] Leaf content of composite samples.

Concerning total Cr in soil (Fig. 2, Cr, left; Fig. S3), the site’s median content was 44 mg/kg and was significantly different from the local background (34.5 mg/kg). Like for Cd, a significant difference between

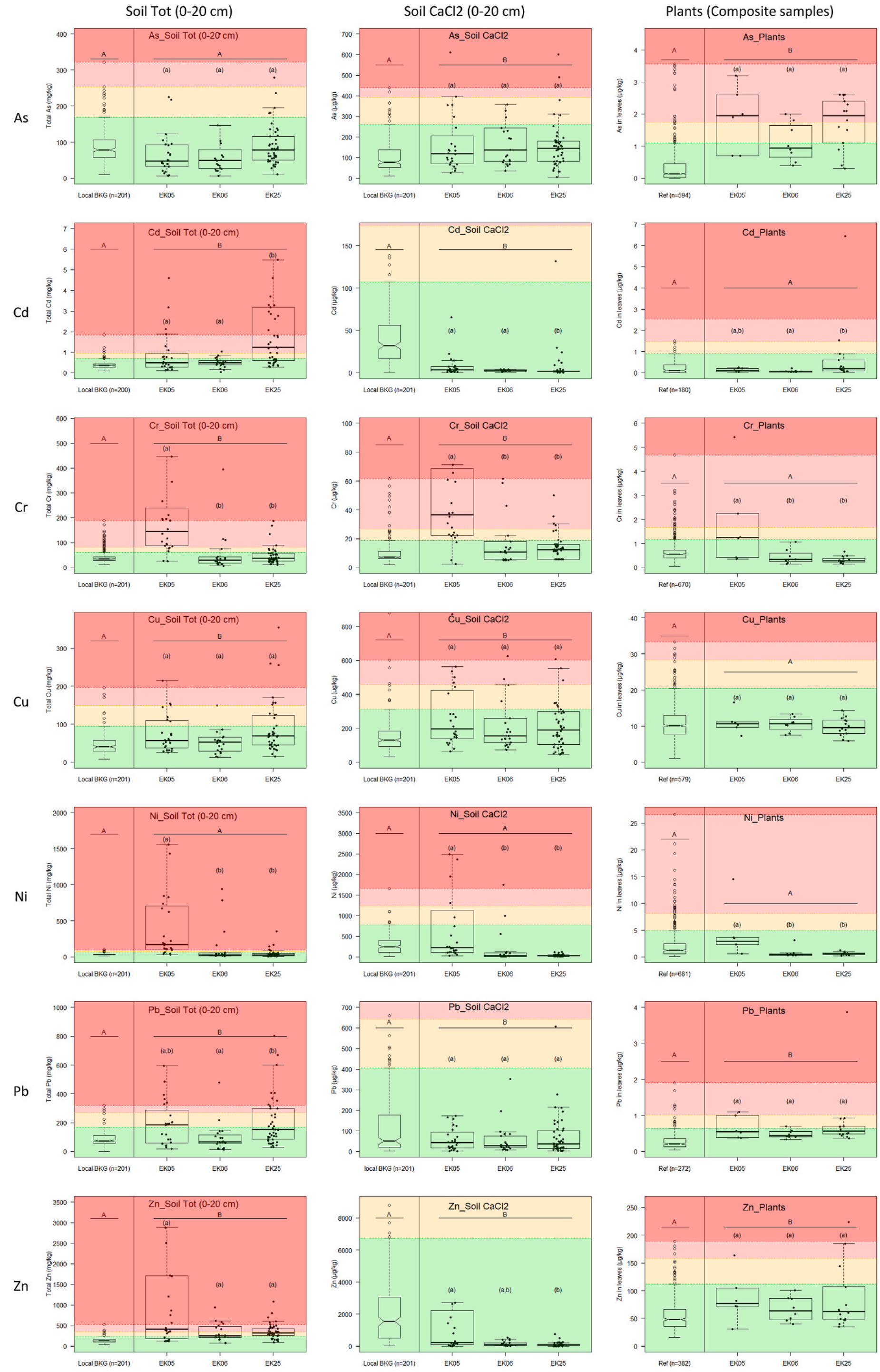

Fig. 2. Distribution of total amounts (left) of As, Cd, Cr, Cu, Ni, Pb and Zn, their environmental availability (center) and their environmental phytoavailability (right) at the study site, as compared with local urban background.

the three parcels was also observed, but it was EK05 which was clearly contaminated, with close to 80 % of high or extreme anomalies and a total level (median value = 145 mg/kg) about 4-fold higher than those of control soils. The EK06 and EK25 parcels also showed approx. 20 % of outlier values, but median Cr levels (29 mg/kg and 37 mg/kg, respectively) were very close to that of control soils. Overall, a similar picture was obtained when considering the environmental availability of Cr (Fig. 2, Cr, center): at the EK05 parcel, $CaCl_2$ extractable Cr was about 5-times the median value of control soils, with about 80 % of samples showing anomalies; while at EK06 and EK25, most values were in the range of usual content, with approx. 20 % of outliers. However, Cr contents in plants growing on the site (Fig. 2, Cr, right) was, overall, not significantly different to those of control plants from uncontaminated sites, and anomalous contents were only evidenced at EK05.

For total Cu (Fig. 2, Cu, left; Fig. S4), the site's median value (59 mg/kg) was slightly but significantly higher than that of control urban soils (41 mg/kg); however, a large number of samples (*i.e.* more than 75 %) were in the range of usual Cu levels and no significant difference between parcels was evidenced. The same trend was observed when considering the $CaCl_2$ extractable fraction (Fig. 2, Cu, center), with almost the same proportion of usual (75 %) *vs* outlier (25 %) values. Nevertheless, Cu contents in plants (Fig. 2, Cu, right) were not significantly different to that of control plants from uncontaminated soils, and all samples were in the range of usual values.

For total Ni (Fig. 2, Ni, left; Fig. S5), the site's median content was 40 mg/kg, and was not significantly different from the local background (32 mg/kg). However, a very significant difference between the three parcels was observed, with EK05 showing more than 5-fold the usual amount in control soils and close to 80 % of high or extreme anomalies. The two other parcels had total Ni contents in the usual range for uncontaminated soils, although a few points showed high anomalies. When the $CaCl_2$ fraction was considered (Fig. 2, Ni, center) more than 70 % of samples from EK05 were in the usual range for uncontaminated soils. Likewise, at EK06 and EK25 most soil samples had usual extractable Ni contents, as compared to control soils. In line with these results, Ni content in plants taken from the site (Fig. 2, Ni, right) was not significantly different from control plants, with only one sample at EK05 showing a high anomaly.

Concerning total Pb (Fig. 2, Pb, left; Fig. S6), the site's median content (124 mg/kg) was significantly higher than that of reference soil (74 mg/kg); however, anomalous Pb contents were mostly found at EK05 and EK25, while EK06 showed overall usual levels. On the other hand, the environmental availability of Pb (Fig. 2, Pb, center) remained within the usual range across the entire site. When foliar Pb levels were considered (Fig. 2, Pb, right), a slightly different picture emerged: EK05 showed moderate to high levels in its central zone; EK06 remained within the usual range and EK25 showed moderate to extreme anomalies in its Southern half, while its Northern half had usual levels (Fig. S6).

Last, for total Zn (Fig. 2, Zn, left; Fig. S7), the site's median content was 327 mg/kg, that is approx. 2.5-fold that of reference soil (129 mg/kg); although no significant difference between parcels was evidence, EK05 exhibited more high and extreme anomalies than EK06 and EK25. This was confirmed considering the $CaCl_2$ extractable fraction (Fig. 2, Zn, center), which showed that anomalies in Zn availability were restricted to a few points at EK05, while more than 90 % of soil samples had usual Zn levels as compared to reference soils. Likewise, Zn accumulation in plants (Fig. 2, Zn, right) was significantly higher than in control plants, but, overall, remained inside the range of usual values, with the exception of a few points at EK05 and in the North-Eastern of EK25 showing moderate to extreme anomalies (Fig. S7).

3.2. *Mapping of the site's hazard using integrated hazard excess (IHE) indices*

Because it is not easy to get a comprehensive picture of a site's hazard when many contaminants are present, raw data for As, Cd, Cr, Cu, Pb, Ni and Zn concentrations were converted into a single Integrated Hazard Excess (IHE) index, to get general maps of total hazard, available hazard and phytoavailable hazard on the study site.

Results showed that, when considering total PTE contents in soil (Fig. 3, left), the Western side of EK05 and a large part of EK25 had high or extreme IHE values. On the other hand, the EK06 parcel had significantly lower IHEs that were mainly in the usual range, as compared to the local background. However, a quite different picture was obtained when the environmental availability was considered (Fig. 3, center); according to this fraction, most of the site had usual values compared to the local background, and only a few small areas, located at the West of EK05, in the South-Western part of EK06 and in the South of EK25,

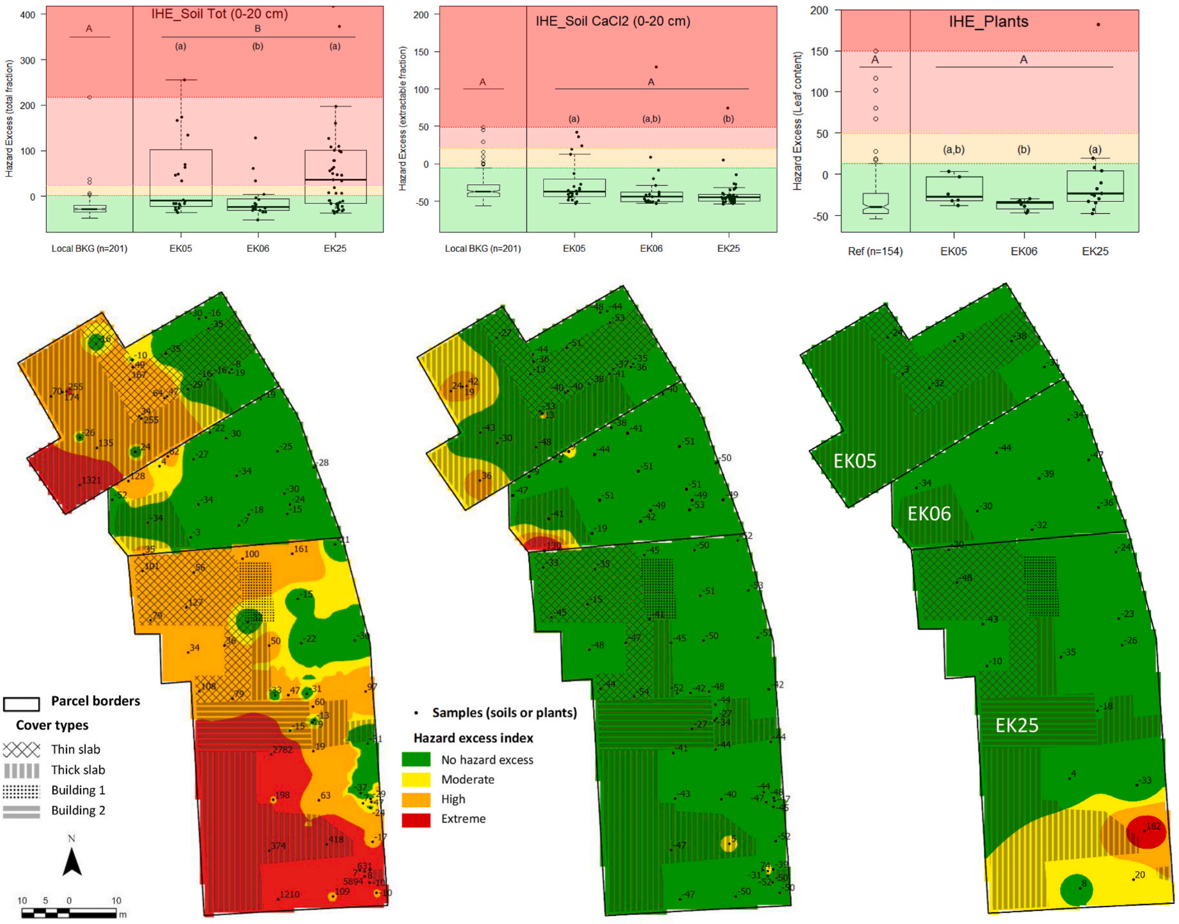

Fig. 3. Distribution and mapping of Integrated Hazard Excess (IHE) values, for total (left), environmentally available (center) and environmentally phytoavailable (right) hazard at the study site, as compared with local urban background.

showed moderate to extreme anomalies. A third vision of the study site was finally obtained by considering the phytoavailable part of the hazard (Fig. 3, right). In that case, results showed plants grown on the site had IHE indices in the range of usual values, except for a few points in the South of EK25, which exhibited extreme anomalies.

4. Discussion

4.1. *Hazard partitioning for assessing PTE contamination helps better anticipating the risk in soil pollution studies*

This case study demonstrated that partitioning PTE into total, extractable and phytoavailable fractions, led to markedly different conclusions. Indeed, while the studied site was classified as highly contaminated by all analyzed elements (As, Cd, Cr, Cu, Ni, Pb and Zn) based on total soil contents, focusing on environmental availability and phytoavailability gave a more nuanced picture. Thus, although labile ($CaCl_2$-extractable) PTE levels were high or very high in some localized areas, most soil samples felt within the usual background range observed in uncontaminated soils. Similarly, though some plant samples showed sharp anomalies in one or several PTE, numerous samples were yet in the range of usual plant content.

Although such a discrepancy between total PTE contents in soils and their (bio)available forms has been previously documented (Black et al., 2011; Farrell et al., 2013; Remon et al., 2013; Pauget et al., 2015) this topic has however seldom been discussed in the context of urban brownfield management. In fact, the only question decision makers and stakeholders want to answer when requesting soil analyses, is whether contaminants may constitute, or not, "*a concern*" for the environment and/or human populations (Chapman, 2007; Vandermoere, 2008). Correctly answering this issue not only implies a precise hazard identification - *i.e.* which harmful substances are present, where are they and what are their amounts in soil? -, but needs also anticipating the likelihood of adverse effects, which depends on their environmental fate and transfer toward biological target (Lofstedt, 2011; Scheer et al., 2014). To deal with this issue, we showed that a tiered hazard identification approach, considering total hazard and its bioavailable fractions, could be of great interest. It is worth to note that in this study we chose a 0.01 M $CaCl_2$ extraction as a proxy for environmental availability, but other weak extraction procedures, such as water extraction or acid rain simulation (Li et al., 2024) could be used as well. Likewise, we considered leaf contents in composite plant samples as a proxy for environmental bioavailability, but other accumulation bioindicators such as earthworms (Suthar et al., 2008; Feng et al., 2025) and/or snails (Pauget et al., 2015; Louzon et al., 2021) could also be considered. In any case, assessing some shades of bioavailability at the early step of hazard identification, would provide decision makers with more precise insights into the fate of PTE in soil, thus enabling to more accurately state for the “concern” issue.

4.2. *The integrated hazard excess indices give a comprehensive view of contaminated sites*

Another important point in soil pollution studies, is to communicate complex analytical results in a form that stakeholders and decision makers should easily understand. For this purpose, a large number of pollution indices have long been proposed (Caeiro et al., 2005; Kowalska et al., 2018; Zhou et al., 2022), with the goal of facilitating a global description of environmental contaminations. One of the most commonly used pollution indices is the “*Potential Ecological Risk*” (RI), first introduced by Håkanson (1980) as a diagnostic tool for lake sediments and water pollution control purposes. A closely similar calculation approach (“*Evaluation of the Risk of the Transferred Metal Elements*”, the ERITME index) was proposed by Pauget and de Vaufleury (2015) for the evaluation of PTE’s bioavailability in polluted soils, using land snails as bioindicators. Accordingly, in this work, we derived the IHE index using the same rationale as for RI or ERITME calculation.

Determination of RI, ERITME or IHE is straightforward. It consists in dividing the concentrations of each investigated PTE (C^i_{Meas} in Eq. (1)) by the corresponding threshold value (C^i_{Thr} in Eq. (1)), then to weight the ratio by the toxicity coefficient of the element considered (T_i in Eq. (1)) and finally to sum the weighted ratio for each element to get the index value (Eq. (2)). Obviously, the result of this calculation, and ultimately the decision that will be taken, is closely dependent on both the thresholds (C^i_{Thr}) and the toxicity coefficient (T_i), selected for each element.

In urban areas, it is highly advisable to refer to the urban pedo-geochemical background (Baize, 2000; MNGSSP, 2017; Belbèze et al., 2023; Binner et al., 2023) rather than the natural background for assessing soil contamination. In this respect, a national database (BDSolU; https://www.bdsolu.fr/fr) is currently under construction in France, with the aim of establishing pedo-geochemical backgrounds for the main French urban areas. However, this database is still incomplete and many cities have not yet been filled in. Consequently, in this work, we used our own database of PTE contents in soils from public parks and gardens (*i.e.* soils used for recreational occupation) in Saint-Étienne city and its urban surroundings (Seillier et al., 2025), to determine the local urban pedo-geochemical background, and derive thresholds for anomalies’ detection. This additionally allowed to establish local background variation and threshold values for the $CaCl_2$-extractable PTE fractions (*i.e.* for the local environmental availability of PTE) as well as for plant content (*i.e.* for the local environmental phytoavailability of PTE).

Another point of attention regarding index calculation is the choice of the toxicity coefficient for each PTE. Indeed, ranking PTE according to their relative toxicity is not trivial, since biological responses of exposed organisms are closely dependent on the species considered. Thus, different toxicity coefficients can be found in the literature (Håkanson, 1980; Pauget and de Vaufleury, 2015; Cao et al., 2023) for a same element. In this study, we retained the values proposed by Håkanson (1980), as they have been the most widely used in environmental studies until now, both to evaluate sediment, water and soil quality.

Anyway, calculating and mapping IHE values provided an integrated overview of the three shades of hazard (total, environmentally available and phytoavailable) on the study site, making it easier to interpret multiple contamination maps. Thus, as compared to the local urban pedo-geochemical background, IHE mapping easily confirmed that total hazard at the study site was overall high or extreme, except at EK06, where values were mostly in the usual range. This was not surprising, owing the different known past activities on the parcels (see Section 2.1). However, when the issue of labile hazard was addressed, both considering the environmental availability and the environmental phytoavailability, IHE values were mostly in the usual range, except at a few localized areas. Consequently, while IHE values for total PTE evidenced high hazard, thereby suggesting high potential risk, the mobile part of the total hazard was overall in the same range as the local background. This indicated that the risk of PTE transfer, either by leaching or accumulation in plants, was not a concern, since it would not be higher than for local urban soils used for recreation in the city.

4.3. *The integrated hazard excess indices are low-cost and efficient decision support tools for soil contamination assessment*

Since the last two decades, methodologies have been developed for performing accurate site-specific risk assessment (Jensen and Pedersen, 2006; Kumar et al., 2019; Qian et al., 2023). In this respect, one of the most promising approach is the Triad procedure (Jensen et al., 2006). This consists in gathering three independent data sets, highlighting the chemical, ecotoxicological and ecological characteristics of the study site, and thus constituting three complementary lines of evidence (LoE) for decision support. Data used to build each LoE are acquired during a tiered process consisting of three successive tiers of investigations, from relatively cheap and rapid analysis (tiers 1: simple screening) to more expensive and precise characterization (tiers 2: refined screening, and tiers 3: detailed evaluation). Although it is a powerful approach, the Triad method is not yet widely used, since the scientific skill needed as well as the costs of analysis rapidly increase when successive tiers are conducted. In fact, to date, studies using successive tiers of the Triad for soil characterization are very scarce (Kim et al., 2015) and most often from academic research groups, while environmental consulting firms rarely propose such complex investigations.

The goal of this study was not to perform a site-specific risk assessment, but rather to propose a complementary approach for a comprehensive hazard identification, which is a prerequisite for an accurate risk assessment. In this sense, partitioning hazard into total, environmentally available and phytoavailable fractions is scientifically sound and technically easy. Computation of IHE indices and mapping of the three shades of hazard is very straightforward, as soon as the local pedogeochemical background of the study site is known and thresholds for outlier detection are established. Consequently, such an approach could be easily integrated in routine procedures for soil characterization, giving stakeholders an efficient decision support tool for anticipating the extent of potential risk and, possibly, the usefulness of carrying out, or not, a more expensive and time-consuming Triad investigation.

5. Conclusion

In this work, we proposed a new methodological approach for a more relevant hazard characterization on PTE contaminated sites. We illustrated that a comprehensive hazard identification should not only consider total PTE contents in soil (*i.e.* the total hazard), as usually performed, but also their most mobile fractions (*i.e.* the environmental availability and environmental bioavailability of PTE). Our results indeed showed that very different conclusions could emerge according the way hazard is considered.

Using a three-tiered hazard assessment for the characterization of urban brownfields is obviously slightly more complicated and expensive, than solely considering total hazard. Indeed, data analysis and interpretation of the results closely depend on the selected threshold values, calculated from the range of usual contents at uncontaminated reference sites. This is not a trivial issue since there is today very few, if any, open public databases gathering available and phytoavailable PTE levels in uncontaminated soils. Consequently, further efforts should be made to get larger and freely accessible regional and national databases. Additionally, standardized protocol (at the national and/or international scale) for assessing PTE content in plant communities should be defined, to allow routine and high output sample processing by private laboratories.

However, considering a three-tiered hazard assessment at the early step of site characterization, could be a relevant strategy for an eco-friendlier management of contaminated soils, while ultimately saving money. As shown in this case study, total PTE contents suggested a high potential risk, thus supporting heavy reclamation strategies such as landfilling and/or covering with clean soil. However, focusing on the available fractions of total hazard revealed that the risk of PTE leaching (environmental availability) and transfer to plant communities (environmental bioavailability) was not higher than for other urban soils accepted for recreational occupation. Consequently, advising for hazard reduction and “dig and dump” actions would be a costly and irrelevant warning. In such a case, more ecologically friendly and less expensive reclamation strategies could be advised, such as gentle remediation options (Drenning et al., 2022), which are more in line with the urgent need for soil preservation, while saving CO_2 emissions due to unnecessary soil excavation and landfilling.

CRediT authorship contribution statement

Rachel Seillier: Writing – original draft, Methodology, Formal analysis, Data curation, Conceptualization. **Frédéric Paran:** Writing – original draft, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Benjamin Pauget:** Visualization, Data curation, Conceptualization. **Cédric Challaye:** Visualization, Data curation, Conceptualization. **Steve Peuble:** Supervision. **Frédéric Gallice:** Formal analysis. **Jérémie Riquier:** Supervision. **Baptiste Bouillot:** Funding acquisition. **Olivier Faure:** Writing – review & editing, Validation, Supervision, Methodology, Investigation, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

The authors declare they have no financial interests or personal relationships which may be considered as potential competing interests.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2025.144769.

Data availability

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