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Keywords: bioavailability, bioaccessibility, metal(loid)s, hazard quotient, carcinogenic risk *Cantareus aspersus*

Abstract

With the new soil uses such as land restoration and to protect wilderness, the human health risk assessment (HHRA) and environmental risk assessment (ERA) should be combined. Based on the relationships demonstrated between an indicator of soil quality, the land snail, and human exposure, the aim of this study is to examine the snail and human risk indicators twenty-nine soils contaminated by metal(loid)s. HHRA was evaluated by both hazard quotient and carcinogenic risk. When the human health indicators were ranked as uncertain, they were weighted by bioaccessibility to refine the risk assessment. The ERA was performed with risk coefficient after *ex situ* snail exposure. The results showed strong and novel relationships between human health and environmental risk indicators that had never been found before. For 62 % of the soils, both indicators revealed either a confirmed risk or an uncertain level of risk. Overall pollutants present greater risk for human than for environment, with 55 vs 28 % of the studied soils classified in the proven risk, respectively. An original integrative risk assessment of polluted soils has been proposed, that shall help setting up relevant strategies to manage contaminated soils considering not only human but also environmental indicators of risk.

1. Introduction

The One Health concept demonstrates that human, animal and ecosystem health are interrelated, notably concerning infectious diseases and chemical contamination (Aguirre et

al., 2016; Lebov et al., 2017; Destoumieux-Garzon et al., 2018; Harrison et al., 2019). This concept is not currently applied to the management of contaminated soils in France, in verifying the compatibility of soil uses (*e.g.*, community gardens, recreational areas), or in assessing the environmental risk (MEEM, 2017). Moreover, the remediation of industrial and urban wastelands requires the assessment of both the human and ecological health risks in the context of increasing alternative uses to both limit urban sprawl and protect the wilderness (such as through land restoration approaches). In most cases, only the total concentration of contaminants in soil is considered (Jia et al., 2018), although according to the bioavailability concept, only a fraction of these contaminants is available and may therefore be assimilated by organisms, leading to adverse effects (Allan, 2002; ISO 17402, 2008; ISO 19204, 2017). To determine an accurate assessment of risk, there is a real need to refine human health and environmental risk assessments (HHRA and ERA, respectively) to account for the bioavailable concentrations of contaminants and to combine these assessments to propose an integrative risk assessment (IRA) of contaminated soils (Alvarenga et al., 2018; Louzon et al., 2020a). This IRA aims associating HHRA and ERA in the frame of the One health concept applied to the management of terrestrial pollutions. Nowadays, HHRA is very often the driving endpoint in the risk assessment of contaminated soils, while the ERA is not systematically carried out in the current methodology for the management of these soils. Yet, usually HHRA and ERA are in most of the cases studied separately. In France, the method to assess the compatibility of the toxicological risks with the current uses of the site, that is called "the interpreting the state of the environment (ISE)" may be used to perform the HHRA (MEEM, 2017). These toxicological risks are assessed by the comparison of hazard quotients (HQ) and carcinogenic risk (CR) to toxicological reference values (TRV). To refine the method of exposure and risk assessment (especially when the values of HQ and CR showed uncertain levels of risk), the bioaccessible concentrations of pollutants measured in soils

according to validated and standardized protocols may be used (Pelfrêne et al., 2013; MEEM, 2017; ISO 17924, 2018). In this case, total soil concentrations are weighted by the bioaccessible fraction determined using the UBM (unified bioaccessibility method) test (MEEM, 2017). Some terrestrial bioassays and bioindicators can be utilized to perform an ERA using for example the risk coefficient (RC) based on the identification of excess transfers and toxicity of contaminants to the land snail (Pauget and de Vaufleury, 2015; Louzon et al., 2020b). As previously determined in soils contaminated by As, Cd and Pb, strong relationships exist between oral bioaccessibility to humans and bioavailability to snails (Louzon et al., 2020a), making these methodologies relevant candidates to perform an IRA for these metal(loid)s. The IRA methodology requires established amplitudes of the responses of indicators used and the convergences and/or divergences between them. The goals of this study were to examine indicators of human health and environmental risks of contaminated soils with using the land snail, *Cantareus aspersus*, as a bioindicator of soil quality (de Vaufleury, 2015; Itziou et al., 2018; Carbone and Faggio, 2019; Baroudi et al., 2020; Radwan et al., 2020), to evaluate their complementarity and propose a way to combine them to perform IRA of contaminated soils. This novel evaluation was conducted in accordance with the French ISE methodology (MEEM, 2017), which is based on comparisons of the state of the environment (*i.e.,* the risk of soil contamination) and the current anthropic uses (HHRA) and the transfers to organisms of the soil fauna (ERA).

2. Experimental

2.1. Soils

Twenty-nine soils contaminated with different mixtures and concentrations of As, Cd and Pb were sampled (0–25 cm deep) in France after humus removal (**Table 1**). Next, these soils were dried (< 40°C) and sieved through 250 µm. The physicochemical parameters of these soils (clay, silt, sand, organic matter (OM), carbonate ($CaCO_3$) contents, pH_{water},

cationic exchange capacity (CEC), and the oxidizable cations of aluminium (Alox) and iron (Feox)) were previously characterized by Louzon et al. (2020a) and were summarized in **table S1**. The minimal and maximal values of the physicochemical parameters were 4.3 and 8.3 for pH water, 4.3 cmol kg^{-1} and 38.8 cmol kg^{-1} for CEC, 0.020 cmol kg^{-1} and 0.054 cmol kg^{-1} for Alox, 0.005 cmol kg^{-1} and 0.008 cmol kg^{-1} for Feox, < 1.00 g kg^{-1} and 169 g kg^{-1} for $CaCO_3$ content, 15.4 g kg^{-1} and 360 g kg^{-1} for OM content, 68.0 g kg^{-1} and 416 g kg^{-1} for clays, 75.0 g kg^{-1} and 734 g kg^{-1} for silts and 66.0 g kg^{-1} and 837 g kg^{-1} for sands. The degree of soil contamination (**Table 1**) were previously characterized in Louzon et al. (2020a), and the min and max concentrations were 3.28 mg kg^{-1} dry weight (dw) and 228 mg kg^{-1} dw for As, 0.10 mg kg^{-1} dw and 257 mg kg^{-1} dw for Cd and 17.8 mg kg^{-1} dw and 8971 mg kg^{-1} dw for Pb.

2.2. Human health risk assessment

HHRA was performed in accordance with the French methodology (MEEM, 2017). On the basis of the concentration of each of the metal(loid)s in the soils, the average daily dose ((ADD) in mg kg^{-1} of body weight day^{-1}) was calculated according to the ingestion of soil particles (Liang, 2017; Jia et al., 2018) with generic input data from the ISE calculation grid (MEDD, 2007a). More precisely, the data considered that 91 mg of soil (Stanek et al., 2001; MEEM, 2017) could consumed daily by a child (15 kg) over 340 days for 6 years of theoretical exposure (**Eq. 1**).

$$ADD = \frac{CS \times IR \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (1)$$

where:

CS: concentration of contaminant in soil (mg kg^{-1})

IR: ingestion rate of soil (mg day^{-1})

EF: exposure frequency (day $year^{-1}$)

ED: exposure duration (years)

BW: body weight (kg)

AT: averaging time (days) for non-carcinogenic contaminants: ED (years) x 365 days year^{-1} (Bevan and Harrison, 2017) or for carcinogenic contaminants: 70 years (*i.e.*, lifetime) x 365 days year^{-1}.

With ADD, both the threshold effects by calculating the hazard quotient (HQ) and the non-threshold effects by calculating the carcinogenic risk (CR) were considered by using the toxicological reference values (TRV) of each contaminant (**Table 2**) recommended by ANSES (2020) and INERIS (2018) (**Eqs. 2 and 3**) (MEDD, 2007b; Liang et al., 2017; Jia et al., 2018), where TRV is expressed in mg kg^{-1} day^{-1} for threshold chemicals and in (mg kg^{-1} day^{-1})$^{-1}$ for carcinogens.

$$HQ = \frac{ADD}{TRV} \quad (2)$$

$$CR = ADD \times TRV \quad (3)$$

The interpretation of HQ and CR for As, Cd and Pb was ranked into three levels: no risk, uncertain risk and potential risk (**Table 3**). When values of HQ and/or CR are uncertain (**Table 3**), ADD can be weighted by the percentage of bioaccessible fraction measured with UBM (ISO 17924, 2018; Louzon et al., 2020a). The bioaccessible fractions (expressed in %) of the metal(loid)s corresponded to the ratio between the measured concentrations and soil concentrations (expressed in mg kg^{-1} dw). To refine the exposure assessment, bioaccessibility can be introduced into HHRA by using a relative bioavailability factor (RB) to quantify the difference in bioavailability of a metal(loid) between the soil matrix and the reference matrix (on which the TRV was built). This factor can be then used in the calculation of ADD (**Eq. 4**).

$$ADD_{adjusted} = ADD \times RB = \frac{ADD \times RB_{soil}}{RB_{TRV}} = ADD \times \frac{BA_{soil} \times fa_{soil}}{BA_{TRV} \times fa_{TRV}} \quad (4)$$

where RB_{soil} and RB_{TRV} are the absolute bioavailability of the metal(loid) in the soil matrix and the reference matrix, respectively (unitless); BA_{soil} and BA_{TRV} is the absolute bioaccessibility of the metal(loid) in the soil matrix and the reference matrix, respectively (unitless); fa_{soil} and fa_{TRV} are the absorbed fraction of the metal(loid) extracted from the soil matrix and the reference matrix, respectively (unitless). Based on INVS (2012), it was found that for As and Cd: (i) fa_{soil} and fa_{TRV} were similar, and (ii) the absolute bioaccessibility in the gastric (G) phase (*i.e.*, stomachal phase) of these elements in drinking water (*i.e.*, fa_{TRV}) was close to 100 %. Thus, for As and Cd, the bioaccessible fraction was directly used to weight the ADD (**Eq. 5**).

$$ADD_{adjusted}\ (As\ or\ Cd) = ADD \times BA_{soil} \quad (5)$$

A similar approach was used for Pb. The TRV established by WHO was calculated considering all sources of Pb (water, air, and food) with the aim of not exceeding a blood level of 50 µg L^{-1} in a child. The calculation was performed on the basis of: (i) a Pb uptake of 20 to 80 % in children's diets (Ziegler et al., 1978; De Michele, 1984) and (ii) a Pb retention of 30 % in the body. According to Oomen et al. (2006), the absolute bioavailability of Pb in the reference matrix can be estimated to average 40 %. Thus, the adjusted ADD for Pb can be determined by considering the maximum value of the absorption (80 %) for all soils (**Eq. 6**) (INVS, 2012).

$$ADD\ adjusted\ (Pb) = ADD \times \frac{BA_{soil} \times 0.8}{0.4} = ADD \times 2 \times BA_{soil} \quad (6)$$

For a conservative estimate, the phase (gastric or gastro-intestinal, *i.e.*, G or GI) in which oral bioaccessibility is the highest was used to weight the ADD (INVS, 2012). Finally, the ISE was performed for each soil to obtain the HHRA by considering the worst interpretation of risk indicators for all contaminants (*e.g.*, if HQ and CR for As and Pb rank a soil as no risk,

but its HQ for Cd rank the soil as proven risk, the soil is then considered to have risk due to the Cd) using the criteria summarized in **table 3**.

2.3. Environmental risk assessment

The RCs for environmental risk assessments were obtained using the snail *ex situ* test over 28 days (Louzon et al., 2020b). Sub-adult snails (*Cantareus aspersus*) were exposed as described in Louzon et al. (2020a). To assess environmental risk by RC calculation based on bioaccumulated concentrations, the following formula was used with the toxicity point (TP) from ATSDR (2015) and *ex situ* threshold guide values (TGV) according to Louzon et al. (2020b) and **table 2** (**Eq. 7**).

$$RC = \left(\left(\frac{Median\ concentration\ of\ contaminant\ in\ snail\ viscera\ at\ 28\ days}{TGV}\right) - 1\right) \times TP \qquad (7)$$

The ERA was performed for each soil by the evaluation of the risk of transferred metallic elements (ERITME) index based on the sum of the RCs (Pauget and de Vaufleury, 2015) (**Eq. 8**). The interpretation of the level of risk as a function of RC and/or ERITME values is possible on the basis of ecotoxic effects on the kinetics of sexual maturity (Louzon et al., *submitted*) (**Table 3**).

$$ERITME = \mathrm{RC}_{As} + \mathrm{RC}_{Cd} + \mathrm{RC}_{Pb} \qquad (8)$$

2.4. Statistical analysis

The statistical analysis was performed with R (version 3.4.2) (R Core Team, 2018). The assessment of the relationships between the indicators of human health and environmental risks were performed using Spearman's correlation. The relationships between the total concentrations of metal(loid)s in the soils and the excess metal(loid) transfers to snails ($C_{28\ days}$ > TGV) were evaluated by comparisons of the identity line (y = x) of the ratio between the total concentration in the soils and the SGV (soil guide values) (Pauget et al., 2013) as a

function of the ratio between the internal concentration in the snails after 28 days of exposure and the TGV.

3. Results and discussion

3.1. Human health risk assessment

HQ and CR were calculated for each soil to estimate human health risk (**Table 4**). By considering the total concentrations of metal(loid)s in the twenty-nine soil samples, the HQ ranged from 0.04 to 2.86 for As, from 0 to 4.04 for Cd and from 0.16 to 80.5 for Pb, respectively. The CR ranged from 2.38×10^{-6} to 1.65×10^{-4} for As and 7.33×10^{-8} to 3.69×10^{-5} for Pb. Overall, for the HHRA, the HQ and CR indicators demonstrated divergence in several cases for the same metal(loid)s. Values of these indicators were uncertain for 16, 9 and 16 soils for the HQ of As, Cd and Pb, respectively, and 27 and 20 soils for the CR of As and Pb, respectively (in **table 3**, the uncertainty area concerns values between 0.2 to 5 for HQ and 10^{-6} to 10^{-4} for CR).

According to INVS (2012), to refine the risk assessment, the HQ adjusted and the CR adjusted were calculated on the basis of the highest bioaccessible fraction obtained between the G and the GI phases (**Table 4**). The adjustment of ADD for Pb can be made by considering the maximum value of the absorption (80 %) and the mean bioavailability (40 %). For As, the bioaccessible fractions were higher in the GI phase than in the G phase for 9 soils (O1, O12, O13, O14, O16, O18, O19, O20 and O22), whereas for Cd and Pb, the bioaccessible fraction was systematically higher in the G phase than in the GI phase (**Table 1**). For these metals, the higher bioaccessibility in the G phase is explained by the acidic pH of the stomach-simulated fluids, which contributes to higher solubilization of metals (Ruby et al., 1996; Wragg et al., 2011). For As, the relatively similar level of bioaccessibility between the G and GI phases could be explained by the reduced adsorption and precipitation reactions

199 caused by the particularly geochemical behaviour of this metalloid in the simulated fluid
200 conditions at a neutral pH (Ruby et al., 1996; Wragg et al., 2011; Rahman et al., 2017).

201 For As, after adjustments of the values in the uncertainty area, the HQ and CR adjusted
202 ranged from 0.02 to 2.86 and 4.28 x 10^{-7} to 1.65 x 10^{-4}, respectively, leading to decreased
203 numbers of soils (7 and 23, respectively) with uncertain risk (**Table 4**). In the current
204 management of As polluted soils CR is shown to be present in very low concentrations in soil,
205 notably at the SGV of As in soils (25 mg kg^{-1}). Indeed, the soil O7 for example, presented a
206 measured concentration of As of 12.8 mg kg^{-1} (**Table 1**), and the unadjusted and adjusted CRs
207 were 9.27 x 10^{-6} and 3.24 x 10^{-6}, respectively (**Table 4**), despite the low bioaccessible
208 fractions (35 % and 25 %, respectively for the G and the GI phases). In this example, the
209 adjustment of the CR by bioaccessibility is not sufficient to reduce the uncertainty of the risk
210 to the level of "no risk" (**Table 4**). The adjustment of the HQ in the area of uncertain risk for
211 Cd by bioaccessibility provides a HQ ranging from 0 to 4.04 with a decrease of 3 % in the
212 number of soils with uncertain risk (*i.e.,* 31 % and 28 % of the soils with uncertain risk for
213 HQ and HQ adj., respectively) (**Table 4**). The decrease in the number of soils with uncertain
214 risk for the HQ of As was higher than for Cd in relation to the lower gastric bioaccessibility of
215 As versus Cd in the soils of this study (G bioaccessible median fraction for As and Cd of 32
216 % and 84 %, respectively) (**Table 1**). For As, and to a lesser extent for Cd, these results
217 validated the inclusion of bioaccessibility to improve the interpretation of human health risks,
218 both in terms of toxicological threshold effects and carcinogenic effects. For Pb, the adjusted
219 HQ and CR ranged from 0.16 to 80.5 and from 7.33 x 10^{-8} to 7.38 x 10^{-5}, respectively (**Table**
220 **4**). However, due to the correction applied to obtain a better ADD consideration for Pb, the
221 use of an orally bioaccessible fraction to refine the HQ or CR of Pb seems inefficient, unless
222 the bioaccessible fraction was smaller than 50 % (see eq. 6 in 2.2. Materials and methods
223 section).

224 The results of the ISE showed that none of the studied soils presented no risk to human health,
225 notably due to the contamination of these soils by Pb. However, after the soils with uncertain
226 risk were ranked based on adjustments of HQ and CR, two soils became no risk (O16 and
227 O22). For an ISE based on results that were risk adjusted or non-risk adjusted with
228 bioaccessibility, the results showed that 11 and 18 soils, respectively, had uncertain risk,
229 leading to a soil status modification of 24 % (**Table 4**). The relevance of adjusting HQ and
230 CR with the bioaccessible fraction to decrease the uncertainty regarding the risks of
231 contaminated soils (carcinogenic or not) to human health was observed, particularly for soil
232 contaminated by As (**Table 4**). However, because Pb and its adjustment with **eq. 6** mainly
233 drive the ISE, the bioaccessibility fraction in this study was not sufficient to decrease the
234 global risk for HHRA for most of the soils. The use of the bioaccessible G fraction is
235 questionable for physiological reasons. Indeed, metal(loid) absorption mainly occurs in the
236 intestine (Mushak, 1991; Diamond et al., 1997). Currently, in the regulations (INVS, 2012),
237 the most penalizing phase (often the G phase) is considered. Consequently, by considering the
238 G phase, which is in the most of the cases, the most penalizing to be more protective and
239 decrease the uncertainty regarding risk, the risk can be overestimated. Due to the
240 consideration of the absorption abilities of Pb in the human digestive tract (**eq. 5**), the oral
241 bioaccessibility fraction improved the HHRA and demonstrated human health risks in 5
242 additional soils (**Table 4**). However, the G phase is the main phase considered to decrease the
243 uncertainty caused by Pb in relation to the stronger bioaccessibility of Pb in the stomach
244 (**Table 1**). For physiological reasons, if the gastro-intestinal phase is considered (*i.e.*, the main
245 site of contaminant assimilation), the results could be different. Indeed, the median (min,
246 max) of the bioaccessible fractions of Pb in the gastric and gastrointestinal phases was 84 %
247 (17 %, 100 %) and 5 % (0 %, 45 %), respectively (**Table 1**).

248 **3.2. Environmental risk assessment**

The snail *ex situ* test was used to assess the environmental risk of these contaminated soils by measuring the bioaccumulated concentrations of metal(loid)s in the visceral mass (**Table 1**). The results showed that the concentrations of metal(loid)s ranged from 0.098 to 21.9 mg kg^{-1} of dw viscera for As, from 2.53 to 333 mg kg^{-1} for Cd and from 1.42 to 856 mg kg^{-1} for Pb. An inter-soil variability of bioaccumulated concentrations in *C. aspersus* was found and this variability reflected the influence of the soil contamination (concentration of metal(loid)s in the soils and mixtures) and the influence of the physicochemical parameters of the soils on metal(loid) mobility and transfer, such as the silt content, the cationic exchange capacity and the organic matter content (Pauget et al., 2012; Louzon et al., 2020a). On the basis of the *ex situ* threshold guide values (TGV), RCs were calculated for each soil and metal(loid) and summed for the assessment of risk by the ERITME index calculation (**Table 5**). RCs ranged from 0 to 35500 for As, 0 to 23400 for Cd and 0 to 37400 for Pb. ERITME for the three metal(loid)s studied varied from 0 to 96300 (**Table 3**). Finally, according to the bioavailability of As, Cd and Pb for the land snail, 11, 10 and 8 soils were classified as without risks, with uncertain risk and with proven environmental risk, respectively (**Table 5**). The relationships between the ratio of the concentrations of metal(loids) in the soils with the soil guide values (SGV) and the ratio of the concentration of metal(loid)s in the viscera of snails after 28 days of exposure to the threshold guide value (when the ratios between the internal concentrations and TGV are higher than 1, a bioavailability of contaminant is characterized) are shown in **figure 1**. Soils below the identity line (y = x) showed a high bioavailability of metal(loids)s in snails based on internal concentrations of the metal(loid)s measured in the snails, while soils above the line showed low bioavailability of excess metal(loids)s in the soils to snails (**Figure 1**). This inter-soil variability according to the total concentrations in soils and in the snail is related to the concept of the bioavailability. More precisely, for As transfer to snails, this variability results from the positive influence of silt

and the negative influence of organic matter content in soils (Louzon et al., 2020a). The values of RCs and the ERITME index constituted a basis to prioritize the management of polluted soils (*e.g.,* remediation, adjustment of use) according to the environmental risk on the basis of the toxicity of metal(loid)s transferred to snails (Pauget and de Vaufleury, 2015; Mariet et al., 2017; Louzon et al., 2020b). For example, by considering the ERITME values for soils O28 and O7 (724 and 12328, respectively), the O7 soil presented a higher environmental risk than O28, and its management should be prioritized to limit environmental risk, although the soil O7 was less contaminated (**Table 5**). Considering the variability of the values of RC that reflect the soil contamination and their physico-chemical properties (**Tables 1 and 5**), the bioavailability assessment is a way to refine environmental impact assessments according to the specificity of each site in terms of physicochemical parameters (Bradham et al., 2006; Pauget et al., 2013).

3.3. Correlations between human health and environmental risk indicators

Spearman's correlation coefficients (ρ) were calculated between human health risk indicators (HQ or CR) either adjusted or not adjusted for bioaccessibility if the values were in the area of uncertain risk and the environmental risk indicator assessed with land snails (RC), and are summarized in **table 6**. Strong correlations were demonstrated between RC and values of human health risk indicators with significant ρ between 0.57 to 0.93. For HQ, the adjustment with bioaccessibility increased the correlations with RC for Pb (+2 %) but not for Cd, and it decreased the correlations with RC for As (-10 %). For CR, the adjustment with bioaccessibility increased the correlations with RC for As (+13 %) and for Pb (+3 %) (**Table 6**). Overall, the correlations between human health and environmental risk indicators were improved by considering oral bioaccessibility for humans. On the basis of the correlations obtained, the relationships discovered between oral bioaccessibility and the bioavailability of metal(loid)s in soils as bioindicators of soil quality by Rahman et al. (2017) and Louzon et al.

(2020a) can be extended between human health (non-carcinogenic and carcinogenic) and environmental health risk indicators.

3.4. Integrative risk assessment

According to the strong correlations observed, the interpretation of the assessment of the human health, whether adjusted or not adjusted with oral bioaccessible fraction and environmental risk was compared for each soil to evaluate similarities and discrepancies (**Figure 2**). In **figure 2**, on the basis of the values used to interpret the indicators of risk (**Table 3**), the worst level of human health risk with HQ and/or CR was used to rank the soils. The comparison between HHRA or HHRA adj. and ERA showed that 52 % and 45 % of the soils, respectively, had the same level of risks (**Figure 2**). These differences can be related to the parameters used to consider the absorption of Pb (see eq. 6 in subsection 2.2. of the Materials and methods) that increases the values of HQ and CR adjusted. For example, without adjustment, the HQ for Pb of soil O1 was 3.86 (uncertain risk), while with adjustment, the HQ was 6.25 (proven risk) (**Table 4**).

These results demonstrated convergences and some differences in terms of interpretation between the conclusions of the ERA and the HHRA of the soils. For example, existing human activities at the site of soil O28 appear inappropriate due to the state and the fate of the soil contamination, while this does not seem to be the case for the soil fauna organisms. Indeed, the RC of Pb in the O28 and O14 soils differed greatly (724 and 11560, respectively) with various environmental risk interpretations (no risk and uncertain risk) (**Table 4**), while their total soil concentrations were higher (467 mg kg^{-1} and 719 mg kg^{-1}) (**Table 1**). Their human health risk was also higher (HQ for Pb of 4.19 and 6.45 for O28 and O14 soils, respectively). The HQ of the Pb in these soils was close to 5, and classifying the soils as having high uncertain and proven risks, respectively (**Table 4**). This example illustrates the different conclusions that can be drawn between HHRA and ERA. For HHRA, these two soils are

classified as high uncertain and proven risk, respectively, which is in agreement with the alert threshold of saturnism risk (300 mg kg^{-1}) and its vigilance threshold (100 mg kg^{-1}) (HCSP, 2014), which implies management of soil pollution (notably by considering the G phase). For soil fauna, soils O14 and O28 were classified as either having uncertain risk and no risk, respectively.

The overall assessment of the environmental risk cannot be performed only on the basis of the response of one bioindicator. However, the use of an accumulation bioindicator that shows the transfer and the bioavailability of contaminants provides essential data regarding risk. This information constitutes the necessary elements of decisions-making to help manage the pollution of the contaminated soils. The IRA was performed for each soil by considering the worst interpretation of the risk to be the most protective for the health of the ecosystem including humans (One Health). Finally, of the twenty-nine investigated soils, often highly contaminated with As, Cd and Pb (in terms of mixture and concentration), fifteen soils presented risks and fourteen were of uncertain risk (**Figure 2**). For these last fourteen soils, supplementary investigations are required to refine the level of risk, potentially by using others bioindicators such as earthworms (Fründ et al., 2011) and specific assays, to assess for example the carcinogenic (Vasseur and Bonnard, 2014; de Souza et al., 2015; Liu et al., 2015) and endocrine disruptor (Höss and Weltje, 2007; Mertl et al., 2014; Druart et al., 2017) potentials of these soils (ISO 19204, 2017). These approaches that would possibly involve integrated biomarker responses (IBR) can be coupled with the ERITME index as demonstrated by Mleiki et al. (2020) for another snail species, *Cantareus apertus*. According to the cost of the decontamination (Lombi and Hamon, 2005), the prioritization of the remediation of the soils could be ranked according to the values of the risk index (HQ adj., CR adj. and ERITME) to better assess the adequacy between the uses of the soils and the risks (MEEM, 2017) and to protect terrestrial ecosystems (Pauget et al., 2013). Currently, the

management of contaminated soils is predominantly based on HHRA. When HHRA concludes that a risk to humans exists, decontamination or other management methodologies should be used. When remediation solutions are too much expensive or when the economic balance of the site redevelopment project does not cover the management overhead, the project can be abandoned and the site remains unclaimed. The assessment of the environmental risk and the pollutant transfers in the environment developed in this study reveal that, although a risk to humans was identified, the contaminants were not necessarily bioavailable (environmental and/or toxicological bioavailability) to an organism of the soil fauna (see yellows stars in **figure 2**). Consequently, the anthropic uses of soil and the risks can be assessed by considering the bioaccessibility to humans and the bioavailability to snails (**Figure 3**). In certain cases, if the IRA shows a higher risk for human health with the current use (ISE > thresholds) but not for the environment (ERITME < threshold), in the future, through developing this methodology to establish a management plan (by using quantitative human health risk assessment), certain sites can be managed by avoiding human activities (*e.g.,* restricting access). In cases such as urban wastelands, the pollutants remain in place to promote land restoration that can increase the biodiversity and/or other alternative uses, such as the production of renewable energy from ecological compensation.

4. Conclusion

This study provides essential knowledge to begin building bridges between human health and environmental risk indicators in the context of the One Health approach and the increasing global environmental crisis. For the first time, methodologies to determine the level of risk for both risk indicators were applied simultaneously to polluted soils. Strong correlations are found between the values of risk indicators for human and environmental health for the three metal(loid)s studied, but in most cases, their interpretation does not indicate the same level of risk. Unexpectedly, for about two-third of the studied soils both

indicators revealed either a confirmed risk or an uncertain level of risk. Another development is that we found that overall polluted soils present greater risk for human than for the environmental bioindicator, with 55 % versus 28 % of the studied soils classified in the proven risk, respectively. The methodology we presented is one way to assess the integrative risk of polluted soils by considering bioaccessibility for humans and bioavailability for bioindicators to better consider the specificity of each site and to protect the health of the ecosystem that encompasses humans and all other life forms.

5. Declaration of competing interest

The authors declare no conflicts of interest.

6. Acknowledgements

The authors would like to thank ADEME for funding the COMBINE study programme (Coupler des indicateurs chiMiques et Biologiques pour une évaluation Intégrative des risques sanitaires et Environnementaux [grant number 1572C0310]) and the ODESSA programme (Optimisation de la gestion Des Sites et Sols pollués par une mesure simple de bioAccessibilité [grant number 1472C0046]). The authors thank Hubert Leprond, Nadia Crini and Francis Douay for their kind advice about our work and Dominique Rieffel and Caroline Amiot for their technical assistance.

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Figure 1: Relationships between the ratio of [metal(loid)s]soils/SGV(metal(loid)s) (mg kg^{-1} dw) and the ratio of [metal(loid)s]snails/TGV(metal(loid)s) (mg kg^{-1} dw) (SGV and TGV were summarized in Louzon et al. (2020b); dotted line represents the identity line y=x).

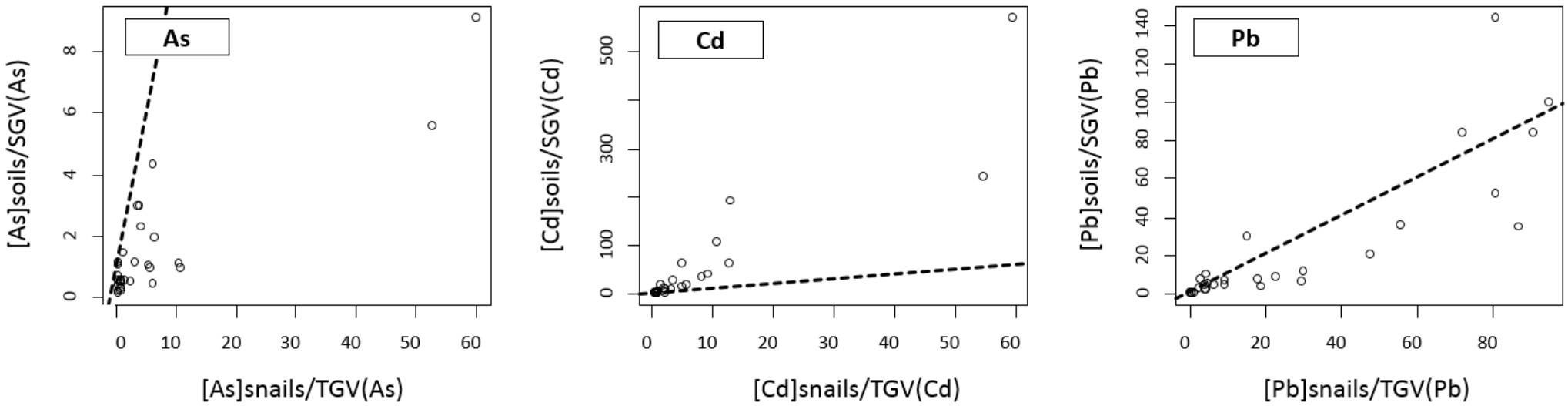

Figure 2: Summary of the HHRA with ISE and ISE adj. and the ERA with ERITME for each of the soils in the study (ISE were adjusted with the oral bioaccessible fraction when HQ or CR were in the area of uncertain risk using the classification in the **table 3**. Yellow stars indicate soils where human health risk is evidenced or is uncertain and the level of environmental risk is no risked. The result of the risk ranking for ISE in this figure is based on the most negative interpretation of the risk for HQ and CR for all of the metal(loid)s studied. ISE, ISE adj. and ERA with ERITME are presented by colored bar plots in blue, orange and green, respectively).

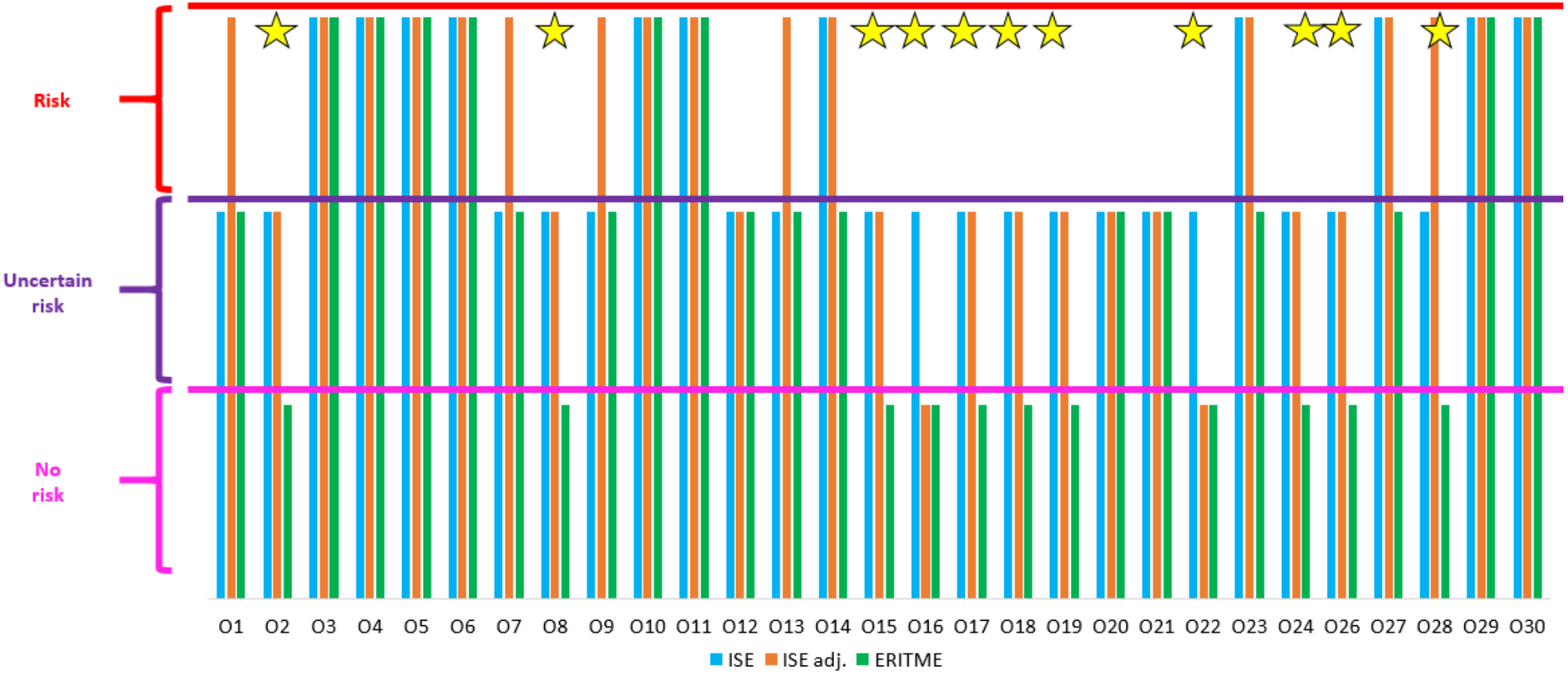

Figure 3: Development of a management plan combining HHRA and ERA (according to results of this study and adapted from the graphical abstract in Louzon et al. (2020a))**.**

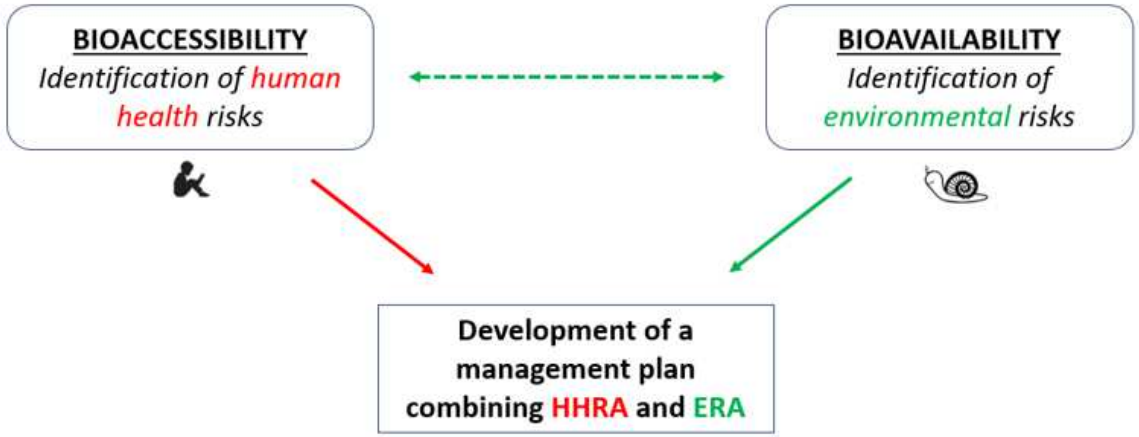

Table 1: Concentrations of As, Cd and Pb in soils and their bioaccessible fractions (gastric and gastro-intestinal) measured using the UBM test and the concentrations bioaccumulated over 28 days in the viscera of snails (mg kg^{-1} dw). Total concentrations of metal(loid)s in soils and their measured bioaccessible fractions are noted as follows for each soil: C_{soil}(G phase|GI phase).

Soil	[As]soil	[As]snail	[Cd]soil	[Cd]snail	[Pb]soil	[Pb]snail
O1	13.4 (38 %|38 %)	0.298	1.32 (95 %|31 %)	5.02	430 (81 %|26 %)	84.6
O2	11.5 (32 %|29 %)	0.298	1.91 (73 %|21 %)	5.63	320 (68 %|2 %)	43.3
O3	228 (100 %|49 %)	21.9	257 (100 %|55 %)	333	6256 (100 %|39 %)	856
O4	74.4 (57 %|38 %)	1.29	48.0 (86 %|25 %)	60.0	5224 (76 %|7 %)	652
O5	108 (60 %|49 %)	2.25	85.7 (89 %|34 %)	73.6	8971 (100 %|20 %)	731
O6	139 (80 %|64 %)	19.2	109 (96 %|45 %)	307	2184 (93 %|5 %)	783
O7	12.8 (35 %|25 %)	0.298	4.20 (92 %|25 %)	18.6	385 (88 %|2 %)	267
O8	5.64 (21 %|21 %)	0.298	0.64 (85 %|39 %)	5.33	30.7 (66 %|10 %)	5.21
O9	23.3 (45 %|42 %)	3.93	7.34 (84 %|37 %)	32.0	534 (96 %|2 %)	208
O10	48.6 (73 %|52 %)	2.31	27.8 (100 %|40 %)	71.9	2233 (100 %|44 %)	503
O11	27.9 (50 %|49 %)	3.78	18.2 (85 %|36 %)	52.5	1260 (88 %|10 %)	431
O12	7.74 (14 %|16 %)	0.298	1.42 (73 %|34 %)	12.5	222 (89 %|45 %)	171
O13	12.6 (27 %|29 %)	0.262	6.50 (74 %|17 %)	28.3	484 (94 %|9 %)	162
O14	23.4 (38 %|65 %)	2.02	15.4 (87 %|57 %)	46.7	719 (85 %|2 %)	271
O15	13.5 (38 %|27 %)	0.482	1.30 (78 %|63 %)	6.04	177 (64 %|0 %)	21.0
O16	5.52 (15 %|16 %)	0.098	0.64 (85 %|32 %)	4.80	17.8 (60 %|0 %)	4.12
O17	17.8 (11 %|8 %)	0.098	0.200 (77 %|60 %)	2.56	33.7 (41 %|2 %)	1.42
O18	74.4 (8 %|8 %)	1.39	0.520 (66 %|26 %)	3.20	46.6 (33 %|1 %)	2.56
O19	10.1 (13 %|14 %)	0.098	4.49 (71 %|28 %)	12.1	117 (72 %|2 %)	37.4
O20	26.0 (23 %|23 %)	1.98	0.370 (71 %|34 %)	4.59	150 (70 %|2 %)	38.9
O21	11.3 (49 %|44 %)	2.23	2.32 (83 %|31 %)	11.5	288 (84 %|1 %)	83.7
O22	3.28 (16 %|18 %)	0.098	0.235 (87 %|57 %)	4.24	19.7 (72 %|18 %)	12.1
O23	57.0 (30 %|25 %)	1.48	0.102 (82 %|35 %)	2.53	660 (17 %|3 %)	39.1
O24	12.0 (34 %|24 %)	0.829	3.57 (96 %|29 %)	12.4	303 (86 %|2 %)	35.6
O26	13.3 (43 %|28 %)	0.098	0.450 (100 %|61 %)	3.20	312 (88 %|3 %)	58.0
O27	28.5 (24 %|16 %)	0.098	7.43 (73 %|38 %)	8.81	1882 (68 %|7 %)	137
O28	25.8 (29 %|23 %)	0.098	2.60 (84 %|28 %)	5.52	467 (63 %|9 %)	25.5
O29	36.1 (24 %|16 %)	0.379	27.2 (49 %|34 %)	28.3	5258 (99 %|32 %)	820
O30	28.3 (27 %|22 %)	1.16	12.8 (81 %|41 %)	20.4	3241 (97 %|24 %)	730
Min.	3.28 (8 %|8 %)	0.098	0.102 (49 %|17 %)	2.53	17.8 (17 %|0 %)	1.42
Med.	23.4 (32 %|25 %)	0.656	3.89 (84 %|34 %)	12.3	426 (84 %|5 %)	110
Max.	228 (100 %|65 %)	21.9	257 (100 %|63 %)	333	8971 (100 %|45 %)	856

Table 2: Toxicological reference values (TRV) for As, Cd and Pb (ingestion) in mg kg^{-1} day^{-1} (ANSES, 2020; INERIS, 2018) for no carcinogenic risks (with threshold) and carcinogenic risk (CR), toxicity point (TP) (ATSDR, 2015) and *ex situ* threshold guide values (TGV) for snails in mg kg^{-1} (Louzon et al., 2020b)

Soil	TRV with threshold	Toxicological target	TRV for CR	*Ex situ* TGV	TP
As	0.00045	Skin	1.5	0.364	600
Cd	0.00036	Kidney	ND	5.60	400
Pb	0.00063	Kidney	0.0085	9.06	400

ND: not determined

Table 3: Interpretation of values of hazard quotient (HQ), cancer risk (CR) (MEEM, 2017) and risk coefficient (RC) (Louzon et al., submitted)

Risk	HQ	CR	RC
No	< 0.2	$< 10^{-6}$	< 2574
Uncertain	0.2-5	10^{-6}-10^{-4}	2574-22720
Proven	> 5	$> 10^{-4}$	> 22720

Table 4: Assessment of human health risk with hazard quotient (HQ), cancer risk (CR) and interpretation the state of the environment (ISE) (when HQ or CR were in the area of uncertain risk, they were weighted (HQ adj. or CR adj.) by bioaccessible fraction. The result of the ISE is the most conservative level of risk for all the metal(loid)s studied according to the ISE grid of interpretation summarized in table 3).

Soil	As				Cd				Pb				ISE	
	HQ	CR	HQ adj.	CR adj.	HQ	CR	HQ adj.	CR adj.	HQ	CR	HQ adj.	CR adj.	risk	risk adj.
O1	0.17	9.76×10^{-6}	no. adj.	3.71×10^{-6}	0.02	no TRV	no adj.	no TRV	3.86	1.77×10^{-6}	6.25	2.87×10^{-6}	uncertainty	proven
O2	0.15	8.36×10^{-6}	no. adj.	2.68×10^{-6}	0.03	no TRV	no adj.	no TRV	2.87	1.32×10^{-6}	3.90	1.80×10^{-6}	uncertainty	uncertainty
O3	2.86	1.65×10^{-4}	2.86	no adj.	4.04	no TRV	4.04	no TRV	56.09	2.57×10^{-5}	no adj.	5.14×10^{-5}	proven	proven
O4	0.93	5.40×10^{-5}	0.53	3.08×10^{-5}	0.75	no TRV	0.65	no TRV	46.82	2.15×10^{-5}	no adj.	3.27×10^{-5}	proven	proven
O5	1.36	7.85×10^{-5}	0.82	4.71×10^{-5}	1.35	no TRV	1.20	no TRV	80.49	3.69×10^{-5}	no adj.	7.38×10^{-5}	proven	proven
O6	1.75	1.01×10^{-4}	1.40	no adj.	1.71	no TRV	1.64	no TRV	19.61	9.02×10^{-6}	no adj.	1.68×10^{-5}	proven	proven
O7	0.16	9.27×10^{-6}	no. adj.	3.24×10^{-6}	0.07	no TRV	no adj.	no TRV	3.45	1.59×10^{-6}	6.07	2.80×10^{-6}	uncertainty	proven
O8	0.07	4.10×10^{-6}	no. adj.	8.61×10^{-7}	0.01	no TRV	no adj.	no TRV	0.27	1.27×10^{-7}	0.36	no adj.	uncertainty	uncertainty
O9	0.29	1.70×10^{-5}	0.13	7.65×10^{-6}	0.12	no TRV	no adj.	no TRV	4.79	2.20×10^{-6}	9.20	4.22×10^{-6}	uncertainty	proven
O10	0.61	3.53×10^{-5}	0.45	2.58×10^{-5}	0.44	no TRV	0.44	no TRV	20.02	9.18×10^{-6}	no adj.	1.84×10^{-5}	proven	proven
O11	0.35	2.03×10^{-5}	0.18	1.02×10^{-5}	0.29	no TRV	0.25	no TRV	11.33	5.19×10^{-6}	no adj.	9.13×10^{-6}	proven	proven
O12	0.10	5.63×10^{-6}	no. adj.	9.01×10^{-7}	0.02	no TRV	no adj.	no TRV	1.99	9.10×10^{-7}	3.54	no adj.	uncertainty	uncertainty
O13	0.16	9.18×10^{-6}	no. adj.	2.66×10^{-6}	0.10	no TRV	no adj.	no TRV	4.34	1.99×10^{-6}	8.16	3.74×10^{-6}	uncertainty	proven
O14	0.30	1.70×10^{-5}	0.20	1.11×10^{-5}	0.24	no TRV	0.21	no TRV	6.45	2.96×10^{-6}	no adj.	5.03×10^{-6}	proven	proven
O15	0.17	9.84×10^{-6}	no. adj.	3.74×10^{-6}	0.02	no TRV	no adj.	no TRV	1.59	7.29×10^{-7}	2.04	no adj.	uncertainty	uncertainty
O16	0.07	4.01×10^{-6}	no. adj.	6.42×10^{-7}	0.01	no TRV	no adj.	no TRV	0.16	7.33×10^{-8}	no adj.	no adj.	uncertainty	no risk
O17	0.22	1.29×10^{-5}	0.02	1.42×10^{-6}	0.00	no TRV	no adj.	no TRV	0.31	1.39×10^{-7}	0.25	no adj.	uncertainty	uncertainty
O18	0.93	5.40×10^{-5}	0.07	4.32×10^{-6}	0.01	no TRV	no adj.	no TRV	0.42	1.92×10^{-7}	0.28	no adj.	uncertainty	uncertainty
O19	0.12	7.34×10^{-6}	no. adj.	1.03×10^{-6}	0.07	no TRV	no adj.	no TRV	1.05	4.81×10^{-7}	1.51	no adj.	uncertainty	uncertainty
O20	0.33	1.89×10^{-5}	0.08	4.35×10^{-6}	0.01	no TRV	no adj.	no TRV	1.35	6.18×10^{-7}	1.89	no adj.	uncertainty	uncertainty
O21	0.14	8.21×10^{-6}	no. adj.	4.02×10^{-6}	0.03	no TRV	no adj.	no TRV	2.58	1.18×10^{-6}	4.33	1.98×10^{-6}	uncertainty	uncertainty
O22	0.04	2.38×10^{-6}	no. adj.	4.28×10^{-7}	0.00	no TRV	no adj.	no TRV	0.17	8.11×10^{-8}	no adj.	no adj.	uncertainty	no risk
O23	0.72	4.14×10^{-5}	0.22	1.24×10^{-5}	0.00	no TRV	no adj.	no TRV	5.92	2.71×10^{-6}	no adj.	9.21×10^{-7}	proven	proven
O24	0.15	8.69×10^{-6}	no. adj.	2.95×10^{-6}	0.06	no TRV	no adj.	no TRV	2.72	1.25×10^{-6}	4.68	2.15×10^{-6}	uncertainty	uncertainty
O26	0.17	9.68×10^{-6}	no. adj.	4.16×10^{-6}	0.01	no TRV	no adj.	no TRV	2.80	1.28×10^{-6}	4.93	2.25×10^{-6}	uncertainty	uncertainty
O27	0.36	2.07×10^{-5}	0.09	4.97×10^{-6}	0.12	no TRV	no adj.	no TRV	16.88	7.75×10^{-6}	no adj.	1.05×10^{-5}	proven	proven
O28	0.32	1.88×10^{-5}	0.09	5.45×10^{-6}	0.04	no TRV	no adj.	no TRV	4.19	1.92×10^{-6}	5.28	2.42×10^{-6}	uncertainty	proven
O29	0.46	2.62×10^{-5}	0.11	6.29×10^{-6}	0.43	no TRV	0.21	no TRV	47.15	2.17×10^{-5}	no adj.	4.30×10^{-5}	proven	proven
O30	0.36	2.06×10^{-5}	0.10	5.56×10^{-6}	0.20	no TRV	0.16	no TRV	29.04	1.33×10^{-5}	no adj.	2.58×10^{-5}	proven	proven

The level of risk is interpreted according to three categories: no risk, uncertain risk and proven risk, labelled as green, orange and red, respectively.

Table 5: Assessment of the environmental risk with risk coefficient (RC) and evaluation of the risk of transferred metallic elements (ERITME) index.

Soil	RC As	RC Cd	RC Pb	ERITME
O1	0	0	3336	3336
O2	0	4	1512	1516
O3	35520	23400	37400	96320
O4	1524	3880	28400	33804
O5	3108	4840	31880	39828
O6	31080	21520	34160	86760
O7	0	928	11400	12328
O8	0	0	0	0
O9	5880	1884	8800	16564
O10	3210	4720	21800	29730
O11	5640	3352	18640	27632
O12	0	492	7160	7652
O13	0	1620	6760	8380
O14	2730	2936	11560	17226
O15	192	32	528	752
O16	0	0	0	0
O17	0	0	0	0
O18	1692	0	0	1692
O19	0	464	1252	1716
O20	2664	0	1316	3980
O21	3078	420	3296	6794
O22	0	0	136	136
O23	1842	0	1328	3170
O24	768	484	1172	2424
O26	0	0	2160	2160
O27	0	228	5640	5868
O28	0	0	724	724
O29	24	1620	35800	37444
O30	1314	1056	31840	34210

The level of risk is interpreted according to three categories: no risk, uncertain risk and proven risk, labelled as green, orange and red, respectively.

Table 6: Spearman correlation coefficients (ρ) for each metal(loids) and each risk index (adjusted or not adjusted with bioaccessible fraction if indicators were in the area of uncertainty risk. Significance of ρ was assessed with p-values and signified with stars: "*", "**" and "***" for < 0.05, < 0.01 and < 0.001, respectively).

	RC As	RC Cd	RC Pb
HQ	0.67***	0.93***	0.90***
HQ adj.	0.57**	0.93***	0.92***
CR	0.67***	ND	0.90***
CR adj.	0.80***	ND	0.93***

HQ: hazard quotient, HQ adj.: hazard quotient adjusted, CR: carcinogenic risk, CR adj.: carcinogenic risk adjusted, RC: risk coefficient, ND = not determined because no TRV for CR of Cd exist.

Graphical abstract