A critical prospective analysis of the potential toxicity of trace element regulation limits in soils worldwide: Are they protective concerning health risk assessment? - A review

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A B S T R A C T

Trace elements (TEs) may have toxic effects to plants and humans; thus, countries and organizations impose maximum allowable regulation limits of their concentrations in soils. Usually such limits are placed in different categories according to soil use, soil properties or based on both attributes. However, some countries have regulation limits irrespective of differentiation in soil properties. In this review, we aimed at collecting TE regulation limits in soils from major countries and organizations around the globe, and critiquing them by assessing potential human health risks in the case of soils attaining the maximum allowable values. We explored the soil-to-human pathway and differentiated among three major exposures from TEs, i.e., residential, industrial and agricultural. We observed the existence of problems concerning TE regulation limits, among which the fact that limits across countries do not regulate the same TEs, not even a minimum number of TEs. This indicates that countries do not seem to agree on which regulation limits of TEs pose a high risk. Also, these regulation limits do not take into account TE mobility to neighbouring environment interphases such as plant, especially edible, and water matrices. Moreover, limits for same TEs are vastly diverse across countries; this indicates that those countries have conflicting information concerning TE-related health risks. Subsequently, we addressed this problem of diversity by quantifying resultant risks; we did that by calculating human health risk indices, taking into consideration the cases in which the highest allowable TE limits are attained in soil. Arsenic limits were found to generate a relatively high hazard quotient (HQ_i, accounting for human intake over the maximum allowable oral reference dose for that same TE), indicating that its risk tends to be underestimated. Other TE limits, such as those of Cd, Cu, Ni, Pb, and Zn typically result in low HQ_i, meaning that limits in their cases are rather overprotective. Our approach reveals the need of reducing diversity in regulation limits by drafting soil legislations of worldwide validity, since risks are common across countries. We suggest that new directions should strategically tend to (a) reduce limits of TEs with underestimated contribution to health risk (such as As), (b) cautiously increase limits of TEs that currently cause minor health risks, (c) quantify TE risks associated with uptake to edible plants and potable water, and (d) consider multi-element contamination cases, where risks are cumulatively enhanced due to TE synergism.

1. Introduction

1.1. *Trace elements: definitions and possible risks in relation to their regulation limits*

The name trace elements (TEs) is given collectively to chemical elements found in relatively small concentrations in soil (with the exception of Fe and Mn, which usually have relatively high total contents); such elements may be potentially harmful to organisms in the environment if present in high concentrations in soil. Some of those elements are also often called "heavy metals" due to their density of higher than 5 g cm^{-3} and their metallic nature, and they typically include Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sn, V, and Zn (Page, 1974; Hodson, 2004; Pourret and Bollinger, 2018; Wu et al., 2018). However, some metalloids (e.g., As, Sb) and non-metals (e.g., Se), which have low atomic density, can also be harmful; thus the necessity of a collective name for all TEs. The occurrence of TEs in the environment can be derived from geogenic processes, as well as from anthropogenic inputs (Liu et al., 2018; Bolan et al., 2014; Rinklebe et al., 2019). However, it is well established that TEs are mostly accumulated in soil due to industrial, mining and agricultural activities, as well as disposal of wastes of communal sources.

Several TEs, such as Mo, Cu, Zn, Ni, Mn, Co, Se, and Cr, have been reported to have essential metabolic functions in living organisms (Brady and Weil, 2002). Nevertheless, these elements and those without any essential biological role (e.g., Zr, Sb, As, Pb, Hg, and Cd) can negatively influence bio-chemo-morphological traits in microbes, plants, animals, and humans beyond their critical exposure level in environmental media. Various research, health and environmental organizations working at national and global scales have defined the deficient, essential, optimal and critical exposure limits of TEs for different organisms under various exposure situations in relation to human and environmental risk assessment. The definition, explanation, objective and values of these various kinds of exposure levels may vary greatly among countries and regulating organizations. Also TE exposure to humans is related to the pathways of ingestion, dermal absorption and inhalation of air-borne soil particles (Fig. 1). While partially understood, it is critically important to understand the question – what critical limit values are fully applicable under certain conditions to certain living organisms?

Food safety and health risk assessment of TEs have received substantial attention worldwide due to their toxic nature and possible transfer in human food chain (Antoniadis et al., 2017a, 2017b, 2019). According to SETAC (2018), "environmental risk assessment determines the nature and likelihood of harmful effects occurring to organisms such as humans, animals, plants, or microbes, due to their exposure to stressors." However, risk assessment calculations based on TE critical exposure limits and other recommended parameters do vary in different literature studies worldwide (as explained later in the discussion). These risk assessment parameters are primarily opted to trace possible route of exposure and associated health risks to living organisms, especially humans. However, the risk assessment predictions based on these critical exposure levels become complicated when different factors are taken into account, such as exposed TE species, bioavailability of TEs, nature of individual, environmental conditions, as well as the source-to-human pathway and the defined regulation. It is understandable that regional and national environmental legislations cannot possibly be identical across countries, as they often derive from many diverse legal and even philosophical principles. However, there is a great necessity of reducing the legislation discrepancies due to the globalization process of the agricultural products, particularly concerning plants, food and fodder. In other words, in case of a product traded across several countries, it may comply with TE permissible limits in some countries but breach the legislations of some other countries. The same applies with other regulated commodities, such as genetically modified organisms and related goods.

Despite considerable progress in these fields, the application of critical exposure concentrations of TEs and associated health risks are still topical and not fully elucidated. In order to better understand the critical exposure concentrations of TEs, it is highly practical to compare these values among different countries and organizations. It is also crucial to address the potential health risks associated with these critical limits of TEs.

1.2. *Trace elements: mobility and adverse health effects*

Generally, the natural concentrations of TEs in soils depend on their concentration in the parent material from which they have been developed. However, anthropogenic activities (responsible for releasing TEs in the ecosystems) often modify natural level, biochemical balance

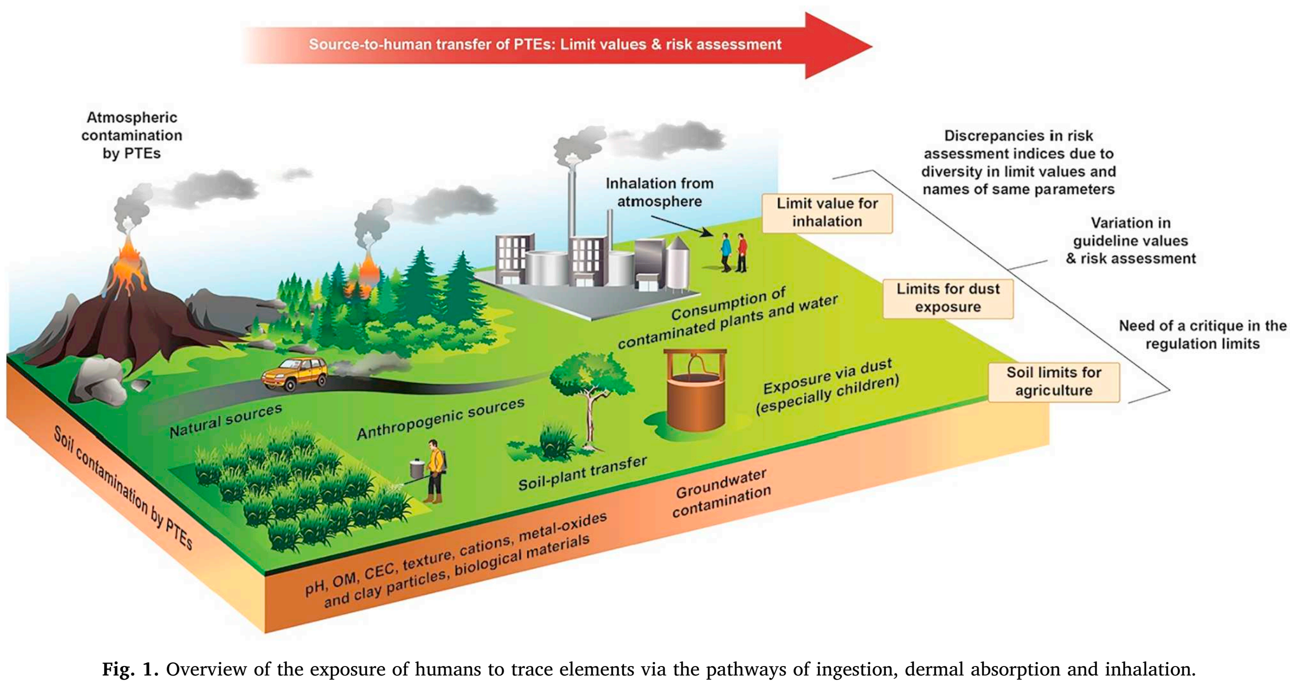

Fig. 1. Overview of the exposure of humans to trace elements via the pathways of ingestion, dermal absorption and inhalation.

and geochemical cycling of TEs in the environment. Soil contamination is defined on the basis of national or regional environmental legislations, which generally do not take into consideration the natural background values. According to an estimation, over 10 million sites are contaminated worldwide, while > 50% of these are contaminated with TEs (Khalid et al., 2017). The majority of these TE-contaminated sites have been reported from high-income and highly developed countries such as the USA, Germany, China, Australia and France, compared to low- and middle-income countries. The difference may be due to the high use of TEs in different industrial processes in high-income countries and the lack of facilities and capacity in the low- and middle-income countries. However, the criteria that have been used to categorize the sites as "contaminated" differ greatly in various countries around the globe. There is a strong need to develop efficient and comparable criteria at international level to categorize these sites as contaminated, as well as to predict the associated environmental and health risks.

Soil is the basic and essential part of ecosystems. Consequently, soil contamination with TEs draws a great interest because of the associated potential risks. It is well established that soil contamination with TEs is the main route of their exposure to humans via food crop consumption. Such exposure is capable of provoking noxious health hazards in humans, if TE exposure contents exceed the safe/critical limits (Bolan et al., 2014; Beckers and Rinklebe, 2017; Rinklebe et al., 2019). Therefore, researchers and regulatory organizations worldwide have given much attention to food safety and risk assessment of TEs in both terrestrial and aquatic environments. Infants and children are at high risk to soil contamination with TEs, and their exposure to these TEs has been numerously documented (e.g., Jiang et al., 2017a; Tang et al., 2017; Kusinm et al., 2018; Yang et al., 2018). It must be added that high levels of TEs in soil and plants are not always evident, and this may lead to relaxed protecting measures. As for soil, it is generally agreed that the soil-to-plant mobility of added TEs (i.e., this generally applies to anthropogenic inputs) decreases with ageing (Antoniadis et al., 2017b). However, McBride (1995), in a notable contribution, argued that added TEs (referring mainly to biosolids-borne TEs) may become highly mobile if certain soil conditions change over time. These could include decreased soil pH, known to induce cationic metal solubility, and thus increased possibility of plant uptake. Another soil property that would enhance TE solubility if changed is soil organic matter (SOM): SOM provides organic retention colloidal phases, increasing TE binding capacity especially in soils with high sand content. In case of fast SOM decomposition (e.g., in a global warming scenario or if land use shifts to agriculture of intense-tillage processes), soil binding capacity will be reduced, and in turn TEs previously bound by it may be solubilized into soil solution and thus easily absorbed by plants. As for plants, they tend to compartmentalize absorbed TEs, shifting them over time to non-vital compounds; by transferring TEs to such niches that do not adversely affect vital physiological functions, plants respond to elevated soil-borne TE-induced stress (Antoniadis et al., 2017a). Thus, plant TE-induced toxicity may become evident when phytotoxic levels are already well established (Chaney, 1980).

Nowadays, it is well established that soil-plant transfer of TEs and their health risks are greatly related to their bioavailable fraction in soil. Generally, TEs exist in soil in various forms: (i) free metal ion, (ii) complexed with organic ligands or inorganic constituents, or (iii) adsorbed onto the surface of soil particles (Pourrut et al., 2011). However, the fraction of TEs that exists in dissolved form in soil porewater is accredited as the most imperative metal fraction, because of its high mobility in soil and availability to plants, and consequently, because of it having high soil-to-plant-to-human transfer potential (Natasha et al., 2018). Therefore, the significance of TEs in soils and their possible soil-to-human transfer depends largely on their fractions that have high solubility and bioavalability (Shahid et al., 2016). However, the limit values are still developed around the globe based on total soil TE contents. The mobility and bioavalability of TEs in soil need to be considered when developing threshold limit values and associated health risks in addition to their total contents.

The solubility and bioavailability (i.e., the ability of TEs to be absorbed by plants, as per Liu et al., 2017) of TEs in soil govern their soil-to-plant-to-human transfer. However, TE bioavailability in soil is highly susceptible to various reactions and processes taking place in soil, such as precipitation/dissolution, adsorption/desorption, and TE–ligand complex formation. Also, TE availability may be affected by plant species and root exudates (Puschenreiter et al., 2003; Antoniadis et al., 2017a). These dynamic soil processes are governed by numerous soil factors such as soil pH, particle size distribution, organic matter, clay content and mineralogy, redox conditions, and competing cations (e.g., Shahid et al., 2016). This implies that TE soil-to-human transfer possibly via food chain depends directly or indirectly on a range of soil processes and properties (Antoniadis et al., 2017a).

The TEs differ greatly with respect to their solubility. Added TEs deriving from anthropogenic inputs tend to be more mobile initially, but their bioavailability decreases over time (Antoniadis et al., 2017a). On the other hand, the majority of TEs of geogenic origin is sparingly soluble, and therefore has very low mobility and bioavailability in soil. This is due to the fact that TEs mainly exist in soils in their residual forms, due to their strong retention by a variety of soil constituents (Shaheen et al., 2017a, 2017b). Indeed, fractionation studies have repeatedly supported the importance of the clay fraction on TE availability and environmental mobility (Rinklebe and Shaheen, 2017; Shaheen et al., 2019a, 2019b). For example, Cu and Pb have been reported to mainly occur in soil in complexed forms (Pourrut et al., 2011), although Cu may also form relatively soluble organometallic complexes. Consequently, these metals have low mobility in soil and bioavailability to plants: soluble soil Pb fractions are generally < 1% of its total soil contents (Natasha et al., 2018). Contrary to Cu and Pb, some metals such as Cd, Ni and Zn have a high soil-plant transfer potential because their soluble soil fractions are typically of higher contribution than that of Cu and Pb (Shahid et al., 2016; Rinklebe and Shaheen, 2014; Shaheen, 2009).

Among TEs, As and Pb are particularly toxic and known to cause cancinogenic and non-carcinogenic health effects to humans (Sun and Chen, 2018). Arsenic (As) is found as anion in soils and aqueous environments; it has two dominant valencies, As(III) and As(V), of which As(III) is more mobile and toxic than As(V) (Niazi et al., 2018). Arsenic causes "dermal lesions, peripheral neuropathy, skin cancer, and peripheral vascular disease" (Pan et al., 2018). Lead, on the other hand, is found in soil as a divalent cation (Pb^{2+}) and is generally considered as readily sorbed onto soil colloids and hence not very bioavailable. Excessive intake of Pb may "damage the nervous, skeletal, circulatory, enzymatic, endocrine, and immune systems of those exposed to it" (Li et al., 2014).

Other well-known elements of high toxicity are Cd and Hg. They are both cationic and without any known beneficial physiological role in living organisms. Cadmium is mostly found in the soil solution as Cd^{2+} and is rather highly mobile, as is often found that its exchangeable and soluble fractions abound in soil (El-Naggar et al., 2018; Rinklebe and Shaheen, 2014); it is also typically poorly sorbed by soil (Shaheen, 2009; Shaheen et al., 2013; Elbana et al., 2018). As for Hg, its characteristics are the tendency to be methylated and thus to increase its mobility compared to its cationic form (Hg^{2+}) (Frohne et al., 2012; Wang et al., 2012; Beckers and Rinklebe, 2017). Additionally, the gaseous emissions from soil to plant and from soil to atmosphere are an important pathway of Hg (e.g., Böhme et al., 2005; During et al., 2009; Rinklebe et al., 2010; Beckers and Rinklebe, 2017). The other two TEs covered in this review, Ni and Zn, do have an essential role in plants; thus, their maximum allowable intake rates (i.e., oral reference dose, RfD_o) are orders of magnitude higher than other TEs such as As, Pb, Cd, and Hg (as further discussed later).

Nowadays, there is a growing trend in predicting the possible health risks deriving from exposure to contaminated soil, atmospheric deposition and water resources. These health risks from soil can be quantified using soil pollution indices of TEs, their soil-to-plant transfer factors, and their levels in edible food crops (Seleiman and Kheir, 2018).

Commonly used health risk assessment indices and parameters include the degree of soil contamination (C_{deg}), uptake/transfer factor, bioaccumulation potential (BAP, also referred as "bioaccessibility"), enrichment factor (EF), translocation factor (TrF), hazard quotient (HQ), estimated daily intake (EDI), and cancer risk (CR). Such indices account for the above mentioned pathways, especially HQ for non-carcinogenic and CR for carcinogenic risks. As for health risk assessment (HRA), values of HQ > 1.00 indicate significant risk, while for CR the risk threshold is 10^{-4} (both indices are unit-less) (Briki et al., 2017).

1.3. Novelty and aims

The issue of national regulations of TEs for soil is rather complicated because countries (a) use similar names for indices which signify different risk levels, (b) categorize limits in different manner, based on land use and soil properties (while some countries on both and some in none of the above), and (c) have limits that vary considerably for each given element. For this reason, it is necessary to collect, critique, and compare regulation limits from various countries and organizations across the globe. Also the possible health risks associated with the scenario of soil TEs reaching the maximum allowable limits should be addressed. The novelty of this approach lies in the fact that such an investigation has never been undertaken before.

In this review, we aimed at (1) presenting under one roof and in a comprehensive manner TE regulation limits concerning soil in major countries and organizations around the globe in order to compare and critique them, and (2) applying health risk assessment caused by human exposure to soil by investigating the scenario that soils have TE concentrations equal to their maximum legislation limits. To this end, we considered different possible exposure scenarios. These included residential exposure to both adults and children — taking into account soil regulation limits regarding residential areas and occupational exposure. Moreover, we examined carcinogenic and non-carcinogenic risks. This approach helps in assessing whether legislation limits are sufficiently protective for the human health or need to be revised and updated in order to reduce possible risks. Such investigations for soil, differentiating by occupation, age groups and associated risk, have never been performed before. We must also note that, although TE-induced risks via water and food consumption are not in any way ignored, they lie beyond the scope of this critical review, and are thus not examined.

2. Theory and equations

2.1. Assessment of non-cancinogenic risk of soil-to-human contamination pathways

Within the soil-related pathways, three routes of TE exposure to the human body were explored: (1) ingestion, (2) dermal absorption, (3) inhalation of air-borne particles.

The human intake via ingestion ($D_{S\text{-}Ing}$), dermal absorption (D_{Derm}), and inhalation (D_{Inh}) (all units in mg TE kg^{-1} BW d^{-1}), is calculated as follows (USEPA (United States Environment Protection Agency), 2002):

$$D_{S-Ing} = C_S \cdot \frac{IngR \cdot EF \cdot ED}{BW \cdot AT} \cdot 10^{-6} \tag{1}$$

$$D_{Derm} = C_S \cdot \frac{SA \cdot SAF \cdot ABS \cdot EF \cdot ED}{BW \cdot AT} \cdot 10^{-6} \tag{2}$$

$$D_{Inh} = C_S \cdot \frac{InhR \cdot EF \cdot ED}{PEF \cdot BW \cdot AT} \tag{3}$$

where C_S is total TE concentration in soil (mg kg^{-1}); *IngR* = Ingestion rate (mg d^{-1}); *EF* = Exposure frequency (d yr^{-1}); *ED* = Exposure duration (yr); *BW* = Body weight (kg); *AT* = Averaging time (d); *SA* = Surface area (cm^2); *SAF* = Skin adherence factor (mg cm^{-2}); *ABS* = Dermal absorption factor (unitless); *InhR* = Inhalation rate (m^3 d^{-1}); *PEF* = Particle emission factor (m^3 kg^{-1}). The factors and their values used in this work for $D_{S\text{-}Ing}$ (ingestion intake), $D_{S\text{-}Derm}$ (dermal absorption intake), and $D_{S\text{-}Inh}$ (inhalation intake) are shown and explained in Table 1, while values used throughout recent similar papers are shown in Table 2 (for $D_{S\text{-}Ing}$) and Table 3 (for $D_{S\text{-}Derm}$ and $D_{S\text{-}Inh}$).

Subsequently, risk was quantified with the use of Hazard Quotient (HQ, unitless), which refers to ingestion (HQ_{Ing}), dermal absorption (HQ_{Derm}), and inhalation (HQ_{Inh}), as follows:

$$HQ_{Ing} = D_{S-Ing}/RfD_o, \tag{4}$$

Table 1
Values of the various factors used in the equations concerning human health risk in this work obtained from USEPA (2002) unless otherwise indicated.

Route to human	Factor (units)	Exposure scenarios		
		Residential-adult	Residential-child	Non-residence (outdoor worker)
Ingestion				
	IngR soil (mg d^{-1}) [water (L d^{-1})]	100 [2]	200 [0.8][a]	100
Dermal				
	SA (cm^2)	5700	2800	3300
	SAF (mg cm^{-2})	0.07	0.2	0.07
	ABS (unitless)	As: 0.03 All else: 10^{-3}	As: 0.03 All else: 10^{-3}	As: 0.03 All else: 10^{-3}
Inhalation				
	InhR (m^3 d^{-1})	20	NA	20
	PEF[b] (m^3 kg^{-1})	1.36×10^9	NA	1.36×10^9
	EF (d yr^{-1})	350	350[c]	225 (agr'ture: 250)
Common factors in all routes				
	ED (yr)	30	6	25 (agr'ture: 50)
	BW (kg)	70	15	
	AT (d)	ED × 365	ED × 365	ED × 365
	LT (yr)	70	–	70

IngR = Ingestion rate; SA = Surface area; SAF = Skin adherence factor; ABS = Dermal absorption factor; InhR = Inhalation rate; PEF = Particle emission factor; EF = Exposure frequency; ED = Exposure duration; BW = Body weight; AT = Averaging time; LT = Lifetime (for cancer risk only).

[a] According to Li et al., 2015a.
[b] According to Li et al., 2017.
[c] According to Singh and Kumar, 2017.

Table 2
Factors for the calculation of D_{Ing} (mg kg^{-1} BW day^{-1}) for children (Ch) and adults (Ad) concerning soil (S) and water (W). IngR = Ingestion rate (mg d^{-1}), EF = exposure frequency (d yr^{-1}), ED = Exposure duration (yr), and BW = Body weight (kg).

	IngR		EF		ED		BW		
	Ad	Ch	Ad	Ch	Ad	Ch	Ad	Ch	References
S	64	104	365		70	6			Chen et al. (2018)
S	450		365		42		60		Briki et al. (2017)
W	2		365		42		60		Briki et al. (2017)
W	3.45		365		42		60		Wongsasuluk et al. (2014)
S	20	50	350		24	6	63	29	Jiang et al. (2017a)
S	50	100	350		30	6	58.8	16.8	Swati et al. (2017)
S	100	200	180		24	6	70	15	Han et al. (2017a)
S	100	200	350		24	6	57.5	15	Giri and Singh (2017)
S	100	200	350		24	6	70	16	Fan and Wang (2017)
S	100	200	350		24	6	56.8	15.9	Xiao et al. (2017)
S	100	200	40[z]		24	6	70	15	Urrutia-Goyes et al. (2017)
S	100	200	250		25	6	70	15	Ihedioha et al. (2017)
S	100	200	180		24	6	61.5	15	Li et al. (2017)
S	X		365		30		60		Fan et al. (2017a)
S	400[a]		40		X		71.6		Pavilonis et al. (2017)
S	100		330		70		65		Lin et al. (2017)
S	200	100	350		30	6	70	15	Ngole-Jeme and Fantke (2017)
W	2.5	0.8	365		70	6	70	15	Singh and Kumar (2017)
S	100	200	X		X	X	55.6	30	Wang et al. (2017a)
S	100	200	250	350	25	6	70	15	Li et al. (2015a)
S	X	X	180		24	6	X	X	Jiang et al. (2017b)
S	100	200	350		30	6	56	15	Wu et al. (2017)
W	2.2		365		70		60		Zhang et al. (2017)
W	2.2	1.8	365		58	10	58.6	22.3	Fei et al. (2017)
S	100	91	243	350	42	10	58.6	22.3	Fei et al. (2017)
S	100	200	350		30	6	70	15	Tepanosyan et al. (2017)
S	100	200	350		60	6	70	15	USEPA (United States Environment Protection Agency) (2002)[b]
W	2	X	350		60	6	70	15	USEPA (United States Environment Protection Agency) (2002)[b]
S	100	X	225	X	25	X	70	X	USEPA (United States Environment Protection Agency) (2002)[c]
W	2	X	225	X	25	X	70	X	USEPA (United States Environment Protection Agency) (2002)[c]

X: Value non-reported.
[a] Concerns gold mining.
[b] Residential use concerning on-site residents.
[c] Non-residential (commercial/industrial) use for outdoor workers.

where RfD_o is the oral reference dose (i.e., via soil ingestion) (mg TE kg^{-1} BW day^{-1}).

Similarly, for the dermal and the inhalation routes:

$$HQ_{Derm} = D_{S-Derm}/RfD_{Derm}, \tag{5}$$

and

$$HQ_{Inh} = D_{S-Inh}/RfD_{Inh}, \tag{6}$$

where RfD_{Derm} and RfD_{Inh} (same units as RfD_o) are reference dose of dermal absorption of soil and reference dose of inhalation of air-borne soil particles, respectively. The RfD_o values used in this work are given for each studied TE in Table 4, while RfD_o values used in other works are reported in Table 5.

The RfD_{Derm} and RfD_{Inh} values were calculated as follows:

$$RfD_{Derm} = RfD_o \times ABS_{GI}, \tag{7}$$

where ABS_{GI} is the unitless gastrointestinal absorption of TEs, given as 1 for As, 0.025 for Cd, and 0.2 for all the rest of TEs (USEPA (United States Environment Protection Agency), 2002).

$$RfD_{Inh}\ [\text{same units as } RfD_o] = RfC\ [\text{mg m}^{-3}] \times 20\ [\text{m}^3\ \text{d}^{-1}]/70\ \text{kg BW} \tag{8}$$

where RfC is the inhalation reference concentration given for each TE (the values we used are reported in Table 4). Values for RfD_o, and RfC commonly used in other similar recent works are given in Table 5.

For each TE, the unitless risk quantified by HQ accounted for individual intake routes can be summed in order to give the overall risk, quantified by Hazard Index of an individual TE, HI_i (unitless), as follows:

$$HI_i = HQ_{Ing} + HQ_{Derm} + HQ_{Inh} \tag{9}$$

Also, hazard index can account for the cumulative risk of all TEs combined, as follows:

$$HI_S = \Sigma(HI_i), \tag{10}$$

where HI_S is the soil-related risk of all studied TEs combined.

It must be noted that HQ_i or $HI_i > 1$ indicates significant risk to human health, while HQ or HI < 1 indicates that TE intake is not harmful.

2.2. *Assessment of carcinogenic risk of the soil-to-human contamination pathways*

Cancer risk is based on the human intake values, D_{Ing}, D_{Derm}, and D_{Inh}, calculated for the soil-to-human pathways as shown earlier.

Soil-related CR for each TE separately, CR_{Si} (unitless) is calculated as follows:

$$CR_{Si} = CR_{Ing} + CR_{Derm} + CR_{Inh} \tag{11}$$

where CR_{Ing}, CR_{Derm}, and CR_{Inh} (all unitless) are cancer risks related to soil ingestion, soil dermal absorption and inhalation of air particles, respectively. These values are calculated as follows:

$$CR_{Ing} = D_{Ing} \times OSF \tag{12}$$

where OSF is oral (i.g., ingestion) slope factor (mg TE kg^{-1} BW day^{-1})$^{-1}$. Values of OSF for the various TEs as used in this study are reported in Table 4, OSF reported in other similar works are reported in Table 6.

$$CR_{Derm} = D_{Derm} \times DSF \tag{13}$$

where DSF is dermal slope factor (mg TE kg^{-1} BW day^{-1})$^{-1}$, calculated as follows:

$$DSF = OSF/ABS_{GI} \tag{14}$$

$$CR_{Inh} = D_{Inh} \times ISF$$

where ISF (mg TE kg^{-1} BW day^{-1})$^{-1}$ is the inhalation slope factor, calculated as follows:

$$ISF = 70\ \text{kg BW}/(UR\ [\mu\text{g m}^{-3}] \times 20\ [\text{m}^3\ \text{d}^{-1}] \times 10^{-3}\ [\text{mg}\ \mu\text{g}^{-1}]) \tag{15}$$

where UR is unit risk (μg TE m^{-3} air), as reported in Table 4, while UR used in various other similar works is shown in Table 5 (UR).

Combined cancer risk of all carcinogenic TEs, CR_S (unitless), was then calculated as follows:

$$CR_S = \Sigma(CR_{Si}) \tag{16}$$

All above equations were obtained from relevant USEPA documents (USEPA (United States Environment Protection Agency), 2002, 2016).

Large differences exist concerning the values utilized across published works, although the concept of risk assessment was developed by the US EPA, and all works cite various US EPA documents as their sole source for obtaining the risk assessment values. This reveals a problem that is related to wrong interpretation of the documents possibly caused by erroneous data gathering and processing or by lack of understanding of the underlying risk assessment concepts. Moreover, it is also observed that the risk assessment parameters (such as HQ, HI, CR) are

Table 3
Factors for the calculation of D_{Derm} and D_{Inh} (both in mg kg^{-1} BW day^{-1}) for children (Ch) and adults (Ad) concerning soil (S) and water (W). InhR = Inhalation rate (m^3 d^{-1}), SAF = Skin adherence factor (mg cm^{-2}), SA = surface area exposure (cm^2), and ABS = Dermal absorption factor (unitless).

	InhR		SAF		SA		ABS	
	Ad	Ch	Ad	Ch	Ad	Ch		References
S	20	10	0.2	X	3300	2800	0.001	Li et al. (2015a)
S	X	X	0.07	0.2	5700	2800	0.001	Jiang et al. (2017b)
S	20	5	10	X	500	1800	0.001, As: 0.03	Wu et al. (2017)
S	X	X	X	X	X	18	0.001, Cr: 0.002	Zhang et al. (2017)
S	16	7.6	X	X	5700	2800	0.001	Jiang et al. (2017a)
S	12.1	7.5	0.07	0.2	2015	1075	0.13	Swati et al. (2017)
S	20	7.6	0.07	0.2	5700	2800	0.001, As: 0.03	Han et al. (2017a)
S	X	X	0.07	0.2	5700	2800	0.001, As: 0.03	Giri and Singh et al. (2017)
S	X	X	0.2	0.07 (sic)	5700	2800	0.01	Fan and Wang (2017)
S	14.5	7.5	0.07	0.2	5709	2176	0.001, As: 0.03	Xiao et al. (2017)
S	20	7.5	0.07	0.2	5700	2800	0.001	Urrutia-Goyes et al. (2017)
S	20	10	0.2	X	3300	2800	0.001	Ihedioha et al. (2017)
S	20	7.6	0.07	0.2	5700	2800	0.001	Li et al. (2017)
S	X	X	X	X	5250	2570	0.01, As: 0.03	Pavilonis et al. (2017)
S		7.63		0.2		1600	a	Lin et al. (2017)
S	20	7.6	0.02	0.04	X	X	0.13	Ngole-Jeme and Fantke (2017)
W	X	X	0.07		5700		0.03	Singh and Kumar (2017)
S	X	X	0.07	0.2	5700	2800	0.001	Wang et al. (2017a)
S	20	X	0.07	0.2	5700	2800	Cd: 0.001, As: 0.03	USEPA (United States Environment Protection Agency) (2002)

X: Value non-reported; a. As: 0.03, Cd: 0.14, Cr: 0.04, Cu: 0.1, Hg: 0.01, Pb: 0.006.

Table 4
Values of oral reference dose (RfD$_o$, μg d^{-1} kg^{-1}), reference concentration (RfC, mg m^{-3}), oral slope factor (OSF, d kg μg^{-1}), unit risk (UR, m^3 μg^{-1}), and tolerable daily intake (TDI, μg d^{-1} kg^{-1}) as used in this study. Values were obtained from USEPA (United States Environment Protection Agency) (2002), unless otherwise indicated.

	RfDo	RfC	OSF	UR	TDI
As	0.3	15×10^{-6}	1.5	4.3×10^{-3}	2.1×10^{-3} [a]
Cd	soil: 1 water: 0.5	10×10^{-6}	NE	NE	10^{-3} [b]
Cr(III)	1500	NE	NE	NE	NA
Cu	40	NE	NE	NE	NA
Hg	0.16	10×10^{-6}	NE	NE	0.7×10^{-3} [a]
Ni	20	10×10^{-6}	NE	NE	NA
Pb	3.5	NE	8.5×10^{-3}	NE	3.6×10^{-3} [a]
Zn	300	NE	NE	NE	NA

NE = non-existent value.
NA = Non-applicable.
[a] Obtained from Antoniadis et al. (2017a).
[b] Obtained from WHO (World Health Organization) (2011).

calculated using various values having different units such as oral reference dose (RfD$_o$, μg d^{-1} kg^{-1}), reference concentration (RfC, mg m^{-3} air), oral slope factor (OSF, d kg μg^{-1}), unit risk (UR). The in-depth review of risk assessment studies also shows some ambiguities in the units used for these risk assessment values.

3. Regulatory limits and health risk assessment

3.1. Trace element limits in regulations

In order to address health risks resulting from exposure to soil TEs, existing national legal regulations have been based on the concept of the protection of human health from being exposed to contamination pathways. That brings the need for the countries to set up maximum limit thresholds. This was a necessity recognised in an early work by Chaney (1973). Often countries' regulations have different limits for different land uses, mostly residential and industrial; however, commercial (often treated as identical to industrial) and agricultural are also cited. Some other countries have regulations for different soil properties (mostly related to particle size distribution, organic matter content and pH) (Poland, Carlon, 2007; Russia, Chernova and Beketskaya, 2011; China, Wang and Shan, 2013). There are also a few countries that base their regulations on factors such as different water table depths or ~~on~~ different uses of groundwater (potable and non-potable) (Poland, Carlon, 2007; Canada-Ontario, Ontario, 2010). Moreover, there is no agreement as to a minimum number of TEs to be regulated; thus various countries regulate different combinations of TEs, some even leaving important toxic elements such as As, Hg, and Cr unregulated.

Among the European countries scrutinised in this study, eleven countries have identified their limits with common terms as follows: precautionary value (PV), trigger value (TV), screening value (SV), action value (AV), intervention value (IV), guideline value (GV), and critical value (CV) (Tables 7 and 8; Table SM-1 in the Supplementary Material). Each of these values represents a certain level of health risk. However, there is not a standard protocol to represent the extent of risk; for some countries, TV represents the lowest (safest) value (Finland, the Netherlands, Latvia, the three Belgium cantons (Wallonia, Flanders and Brussels), Lithuania, and Germany), while for Czech Republic the lowest is PV. Also, AV (for Czech Republic), IV (Lithuania, Belgium, Netherlands, and Austria), GV (Sweden, and Finland), and CV (Latvia) represent the highest permissible values (Table 7 for As, Cd, and Pb; Table 8 for Zn, Cu, Ni, and Hg). Other European countries, including UK, Italy and Poland, use the term SV in a different manner, since they have only one level of risk.

On the other hand, some countries use completely different terms, not in any way common among each other. These include Denmark (Soil Quality Criteria, Ecotoxicology SQC, Indicative Value of Serious Contamination, along with a TV), Slovakia (Maximum Allowable Concentrations, and Value for Decontamination Measures), France (Soil Statement Definition Value, and Impact Statement Value) and Russia (Maximum Permissible Concentration, and Provisional Permissible Concentration) (Table SM-1).

In this regard, different terms signify different levels of potential risk, although the order from the lower to the higher risk is not always identical. Within some of these limits, there is a distinction among different land uses. Residential use and gardens (or allotments for the UK) are usually assigned the lowest value, while industrial and commercial the highest. This, apart from the UK, concerns the Belgium cantons of Flanders, Brussels and Wallonia, as well as Austria, Italy, Czech Republic, Poland, France and Germany for Europe, and Korea, Australia, USA, Canada, and South Africa for countries outside from

Table 5
Chronic ingestion reference dose RfD_o (μg day^{-1} kg^{-1} BW), chronic inhalation reference concentration (RfC, mg m^{-3}) and inhalation unit risk (UR, $(m^3\,\mu g)^{-1}$) as reported in various publications.

	As	Cd	Co	Cr	Cu	Hg	Mo	Ni	Pb	Sb	Se	Zn	References
RfD_o		1		1500	40			20	4			300	Chen et al. (2018)
RfD_o	0.3	0.5		3	40	0.3		20	3.5			300	Wongsasuluk et al. (2014)
RfD_o	0.3	1	20	3	40	0.3		20	3.5	0.4		300	Li et al. (2015b)
RfD_o	0.3	1		3	40	0.3		20	3.5			300	Chen et al. (2015a)
RfD_o	0.3	0.5							3			300	da Silva et al. (2017)
RfD_o	0.3	1	0.00571	0.0286	40.2	0.0857		20.6	3.52	0.4		300	Wu et al. (2017)
RfD_o	0.3	0.5			0.3				1.4				Zhang et al. (2017)
RfD_o	0.3	1		3	40			20	3.5			300	Jiang et al. (2017a)
RfD_o	0.3	1	30	X, 0.3[a]	10	0.04			3.5			300	Okereke and Amadi (2017)
RfD_o	0.3	1	20	3	40	0.3		20	3.5			600	Han et al. (2017a)
RfD_o	0.3	1	0.3	40	3			20	3.5			300	Giri and Singh (2017)
RfD_o				3	40				3.5			300	Fan and Wang (2017)
RfD_o		1				0.3			3.5				Xiao et al. (2017)
RfD_o		1		3	40			20	3.5			300	Li et al. (2017)
RfD_o	50	1		1500	40			4	5			300	Fan et al. (2017a)
RfD_o	0.3		0.3			0.16			0.286			300	Pavilonis et al. (2017)
RfD_o	0.3	1		3	40	0.3			3.5				Lin et al. (2017)
RfD_o	0.3	1	0.3	3	40			20	0.035			300	Ngole-Jeme and Fantke (2017)
RfD_o		0.5			40			20	1.4			300	Singh and Kumar (2017)
RfD_o		1		1500	40				3.5			300	Wang et al. (2017b)
RfD_o				5	4	0.1			38			300	Duan et al. (2017)
RfD_o	0.3	0.5	X	1500, 3[a]	1300	0.3	X	20	X	0.4		300	Bouwes and Hassur (1997)
RfD_o	0.3	1, 0.5[b]	0.3	X, 3[a]	40	0.16	5	20	X	0.4	5	300	RAIS (2018)
RfC	X	X	X	X	X	0.3		X	X	X		X	Bouwes and Hassur (1997)
UR	0.0043	X	X	X	X	X		X	X	X		X	Bouwes and Hassur (1997)
RfC	0.015	0.01	0.006	X, 0.1[a]	X	0.3	0.4	0.09	X	0.3		X	RAIS (2018)
UR	0.0043	0.0018	0.009	X, 0.084[a]	X	X		0.26×10^{-3}	1.2×10^{-6}	X		X	RAIS (2018)

X: Value reported as non-existent.
[a] Refers to Cr(III) and Cr(VI), respectively.
[b] Refers to food and water, respectively.

Table 6
Oral slope factor for carcinogenic effects (μg day^{-1} kg^{-1} $BW)^{-1}$ as reported in various publications.

As	Cd	Cr	Cu	Ni	Pb	Zn	References
1.5							Wu et al. (2017)
1.5					0.0085		Wan et al. (2017)
1.5							da Silva et al. (2017)
1.5							Chen et al. (2015a)
1.5	X	X	X	X	X	X	Wongsasuluk et al. (2014)
1.5	X	0.5	X	X	0.0085	X	Jiang et al. (2017a)
1.5	X	X		X			Han et al. (2017a)
		22					Fan and Wang (2017)
	6.1						Xiao et al. (2017)
					0.0085		Urrutia-Goyes et al. (2017)
					0.0085		Lin et al. (2017)
1.5	X	X	X	0.84	0.0085	X	Ngole-Jeme and Fantke (2017)
1.5		0.5			0.0085		Wang et al. (2017b)
1.5	6.1						Duan et al. (2017)
1.5	X	X	X	X	X	X	Bouwes and Hassur (1997)
1.5	X	0.5	X	X	0.0085	X	RAIS (2018)

X: Reported as non-existent value.

Europe. There are other countries that distinguish within certain limits according to soil properties; such properties usually include pH (China), texture (particle size distribution, Latvia), and both pH and texture (Russia); some countries have a range of values as maximum allowable limits, instead of a single value (Finland and the EU Directive).

As for countries outside Europe, India has only low and high limits (similar to Finland and the EU), while the USA has a SV limit value with two levels assigned to residential and industrial soil use. Apart from that, Korea identifies its limits as "Soil Contamination Warning Limits," and "Soil Contamination Counterplan Limits," and within these there are 3 different limits for various land uses (categorized as Zones 1–3); Australia identifies its limits as "Ecological Investigation Levels" and "Health Investigation Levels" (HIL), and within HIL there are further differentiations for land uses (residential with and without gardens, parks-schools, and industrial-commercial). South Africa does not have any specific name for the limits, but distinguishes limits among various land uses or water protection regimes (water protection, informal and standard residence, commercial-industrial, and a value named "Protection of Ecological Health").

As for Canada, it is interesting that apart from one national regulation, there also exist different regulations applying for the various provinces;, e.g., Alberta categorizes TEs within land uses (natural, agricultural, residential/park, industrial, and commercial), while Ontario has many different limits distinguishing for categories such as uniform/stratified soils, deep/shallow water table, and potable/non-potable groundwater. Also within each category, there are two land uses, residential and industrial. As for the national Canada limits (called "Soil Quality Guidelines," SQG), TEs are put in three categories according to "agricultural," "industrial," and "residential" soil uses. In Table SM-1 we present all these values, including FD ("full-depth generic site condition standard in non-potable ground water conditions") and SS ("stratified site condition standard in a non-potable ground water conditions") of the province of Ontario.

Among the largest differences in terms of threshold values are those concerning the maximum values in the regulations of various countries: giving the example of a well-known toxic element, i.e., As, the regulation values for residential use range from 20 (SV, Italy; Carlon, 2007) to 110 (IV, Flanders, Chernova and Beketskaya, 2011); for non-European countries the diversity is even higher, with regulation limits ranging from as low as 0.68 (SV, USA; USEPA (United States Environment Protection Agency), 2016) to 400 (HIL, Australia; DoEC (Department of Environment and Conservation, Australia), 2010). Likewise, Cd for residential use ranges from 2 (Italy) to 20 in Germany (TV; BBodSchV, 1999) and Czech Republic (AV; Carlon, 2007). Non-European countries have limits that range from 1.2 (SS, Ontario-Canada; Ontario, 2010) to 71 (USA). It is noteworthy that the USA have been criticized for relaxed TEs regulation limits compared to European

Table 7

Limits for As, Cd, and Pb in various countries, corresponding Hazard Quotients (HQ) for adults and children, and Cancer Risk (CR). Significant non-cancer health hazard exists for HQ > 1 and increased cancer risk for CR $> 10^{-4}$. Values in italics indicate such elevated risks. For each country, the highest value for each land use is chosen. Limits are reported in mg kg^{-1}, while HQ and CR are unitless.

Residential

	As				Cd			Pb				
Europe	Limits	HQ-Adult	HQ-Child	CR	Limits	HQ-Adult	HQ-Child	Limits	HQ-Adult	HQ-Child	CR	References for limits
Italy (SV)	20	0.091	0.852	1.76E-05	2	0.003	0.026	100	0.039	0.365	8.00E-06	Carlon (2007)
UK (SV)	32	0.146	*1.363*	2.82E-05	10	0.014	0.128	450	0.176	*1.643*	3.60E-05	Carlon (2007)
Wallonia (IV)	40	0.183	*1.704*	3.52E-05	3	0.004	0.038	200	0.078	0.730	1.60E-05	CO (Coordination Officieuse) (2008)
Germany (TV)	50	0.229	*2.130*	4.40E-05	20	0.027	0.256	400	0.156	*1.460*	3.20E-05	BBodSchV (1999)
Czech R. (AV)	70	0.320	*2.982*	6.16E-05	20	0.027	0.256	300	0.117	*1.095*	2.40E-05	Carlon (2007)
Flanders (IV)	110	0.503	*4.686*	9.68E-05	6	0.008	0.077	700	0.273	*2.555*	5.60E-05	Chernova and Beketskaya (2011)
Brussels	110	0.503	*4.686*	9.68E-05	6	0.008	0.077	700	0.273	*2.555*	5.60E-05	Carlon (2007)
Non-Europe												
USA (SV)	0.68	0.003	0.029	5.98E-07	71	0.097	0.908	NE	–	–	–	Ontario (2011)
Ontario-Ca (SS)	11	0.050	0.469	9.68E-06	1.2	0.002	0.015	120	0.047	0.438	9.60E-06	Ontario (2011)
Alberta-Ca (Res/ Park)	17	0.078	0.724	1.50E-05	10	0.014	0.128	140	0.055	0.511	1.12E-05	Alberta Government (2016)
Canada (SQG)	12	0.0.055	0.511	1.05E-05	10	0.014	0.128	140	0.055	0.511	1.12E-05	CCME (Canadian Council of Ministers of the Environment) (2018)
S. Africa (StandRes)	48	0.219	*2.045*	4.22E-05	32	0.044	0.409	230	0.090	0.840	1.84E-05	MoE (Ministry of Environment, Korea) (2018)
Korea (SCCL, Z. 1)	150	0.686	*6.390*	*1.32E-04*	NE	–	–	600	0.234	*2.190*	4.80E-05	MoE (Ministry of Environment, Korea) (2018)
Australia (HIL)	400	*1.828*	*17.040*	*3.52E-04*	20	0.027	0.256	1200	0.468	*4.380*	9.60E-05	DoEC (Department of Environment and Conservation, Australia) (2010)
HQ1-Ad	219	1			73	1		2564	1			
HQ1-Ch	23		1		78		1	274		1		
$CR10^{-4}$	114			10^{-4}	NA			1250			10^{-4}	

Industrial

	As			Cd		Pb			
Europe	Limits	HQ-Adult	CR	Limits	HQ-Adult	Limits	HQ-Adult	CR	References for limits
Italy (SV)	50	0.147	4.40E-05	15	0.013	1000	0.250	4.00E-05	Carlon (2007)
Poland (SQS, Deep/LHC)	100	0.294	8.80E-05	20	0.018	1000	0.250	4.00E-05	Carlon (2007)
France (ISV)	120	0.353	*1.06E-04*	60	0.053	2000	0.500	8.00E-05	Carlon (2007)
Czech R. (AV)	140	0.412	*1.23E-04*	30	0.026	800	0.200	3.20E-05	Carlon (2007)
Germany (TV)	140	0.412	*1.23E-04*	60	0.053	2000	0.500	8.00E-05	BBodSchV (1999)
Flanders (IV)	300	0.882	2.64E-04	30	0.026	2500	0.625	1.00E-04	Chernova and Beketskaya (2011)
Brussels	300	0.882	*2.64E-04*	30	0.026	2500	0.625	1.00E-04	Carlon (2007)
Wallonia (TV)	300	0.882	*2.64E-04*	50	0.044	1360	0.340	5.44E-05	CO (Coordination Officieuse) (2008)
UK (SV)	500	*1.470*	*4.40E-04*	1400	1.232	750	0.188	3.00E-05	Carlon (2007)
Non-Europe									
USA (SV)	3	0.009	2.64E-06	980	0.862	NE	–	–	USEPA (United States Environment Protection Agency) (2016)
Ontario (SS)	18	0.053	1.58E-05	7.9	0.007	1000	0.250	4.00E-05	Ontario (2011)
Alberta	26	0.076	2.29E-05	22	0.019	260	0.065	1.04E-05	Alberta Government (2016)
Canada (SQG)	12	0.035	1.06E-05	22	0.019	600	0.150	2.4E-05	CCME (Canadian Council of Ministers of the Environment) (2018)
Australia (HIL)	500	*1.470*	*4.40E-04*	100	0.088	1500	0.375	6.00E-05	DoEC (Department of Environment and Conservation, Australia) (2010)
Mexico	260	0.764	*2.29E-04*	450	0.396	800	0.200	3.20E-05	USEPA (United States Environment Protection Agency) (2016)
S. Africa (Comm/Ind)	580	*1.705*	*5.10E-04*	260	0.229	1900	0.475	7.60E-05	GGS (Government Gazette Staatskoerant, South Africa) (2012)
Korea (SCCL, Z.3)	600	*1.764*	*5.28E-04*	NE	–	2100	0.525	8.40E-05	MoE (Ministry of Environment, Korea) (2018)
HQ1-Ad	340	1		1136	1	4000	1		
$CR10^{-4}$	213		10^{-4}	NA		2500		10^{-4}	

Agricultural use (as occupational exposure)

	As			Cd		Pb			
Europe	Limits	HQ-Adult	CR	Limits	HQ-Adult	Limits	HQ-Adult	CR	References for limits
Austria (SV)	20	0.065	1.76E-05	1	0.001	100	0.028	9.00E-06	CF (Common Forum – Working Document), 2009
Wallonia (IV)	30	0.098	2.64E-05	1	0.001	200	0.056	1.80E-05	CO (Coordination Officieuse) (2008)
France (ISV)	37	0.121	3.26E-05	20	0.020	400	0.112	3.60E-05	Carlon (2007)
UK (SGV)	43	0.140	3.78E-05	1.8	0.002	NE	–	–	CLAIRE (2018)
Flanders (IV)	45	0.147	3.96E-05	2	0.002	200	0.056	1.80E-05	Chernova and Beketskaya (2011)

(*continued on next page*)

Table 7 (*continued*)

Agricultural use (as occupational exposure)									
	As			Cd		Pb			
Europe	Limits	HQ-Adult	CR	Limits	HQ-Adult	Limits	HQ-Adult	CR	References for limits
Poland (SQS, Deep/LHC)	55	0.179	4.84E-05	10	0.010	200	0.056	1.80E-05	Carlon (2007)
Czech R. (AV)	55	0.179	4.84E-05	12	0.012	300	0.084	2.70E-05	Carlon (2007)
Germany (AV)	200	0.652	*1.76E-04*	20	0.020	1200	0.336	*1.08E-04*	BBodSchV (1999)
Non-Europe									
Korea (SCCL, Z. 2)	75	0.245	6.60E-05	NE	–	1000	0.280	9.00E-05	MoE (Ministry of Environment, Korea) (2018)
Alberta-Ca	17	0.055	1.50E-05	1.4	0.001	70	0.020	6.30E-06	Alberta Government (2016)
Canada (SQG)	12	0.173	4.66E-05	1.4	0.001	70	0.02	6.30E-06	CCME (Canadian Council of Ministers of the Environment) (2018)
Australia (HIL, Res/Gard)	100	0.326	8.80E-05	20	0.020	300	0.084	2.70E-05	DoEC (Department of Environment and Conservation, Australia) (2010)
Taiwan	NE	–	–	5	0.005	500	0.140	4.50E-05	Lai et al. (2010)
Mexico	25	0.082	2.20E-05	35	0.034	400	0.112	3.60E-05	Perez-Vazquez et al. (2016)
S. Africa (PEH)	150	0.489	*1.32E-04*	37	0.036	100	0.028	9.00E-06	GGS (Government Gazette Staatskoerant, South Africa) (2012)
HQ1-Ad	307	1		1020	1	3571	1		
$CR10^{-1}$	114		10^{-4}	NA		111		10^{-4}	

Use non-specified				
Europe	As	Cd	Pb	References
Russia (PPC, clay/pH > 5.5)	10	2	130	Chernova and Beketskaya (2011)
Denmark (TV)	20	5	400	DEPE (Danish Environmental Protection Agency) (2002)
Spain	25	3	75	Nadal et al. (2016)
Sweden (GV)	25	12	400	INSURE (2017)
Latvia (CV)	40	10	500	INSURE (2017)
Slovakia (VDM)	50	20	600	Carlon (2007)
Austria (IV)	50	10	500	CF (Common Forum – Working Document) (2009)
Netherlands (IV)	55	12	530	MvV (Ministerie von Volkshuissvesting) (2000)
EU Dir (High)	NE	3	300	CEC (Council of the European Communities) (1986)
Lithuania (IV)	80	3	500	CF (Common Forum – Working Document) (2009)
Finland (GV, High)	100	20	750	MotE (Ministry of the Environment, Finland) (2007)
Non-Europe				
China (Grade III, pH > 6.5)	30	1	500	Wang and Shan (2013)
India (High)	NE	6	500	Saha et al. (2015)
Taiwan	60	20	2000	Lai et al. (2010)
Armenia	10	NE	65	Tepanosyan et al. (2017)

SV = Screening Value; IV = Intervention Value; TV = Trigger Value; AV = Action Value; SS = Stratified Site Condition Standards in a Non-potable Ground Water Conditions; Res/Park = Residential/Parks; SQG = Soil Quality Guidelines; StandRes = Value for "Standard Residential Area"; SCCL = Soil Contamination Counterplan Limit; Z.1 = Zone 1 (includes residential area); HIL: Health Investigation Levels; HQ1-Ad = Limit value that would generate an HQ value equal to unity for adults; HQ1-Ch = Limit value that would generate an HQ value equal to unity for children; $CR10^{-4}$ = Limit value that would generate a CR = 10^{-4}; SQS = Soil Quality Standards; Deep/LHC = Agricultural use/deep soil (depth > 15 m/low hydraulic conductivity (< 10^{-7} m sec^{-1}); ISV = Impact Statement Value; Comm/Ind = Commercial/Industrial; Z.3 = Zone 3 (includes industrial area); SGV = Soil Guideline Value; Z.2 = Zone 2 (includes agricultural land use); Res/Gard = Residence with garden; PEH = "Protection Ecological Health." PPC = Provisional Permissible Concentration; VDM = Value for Decontamination Measures; High = High value in a range of values; Grade III = Applying for High Adsorption Capacity Soils Only; NA = Non-applicable; NE = Non-existent.

countries (Harrison and Bouldin, 1999, a work dealing with biosolids-added TEs). The range for Pb is also highly variable, spanning from 100 mg kg^{-1} for residential use in Italy to 700 mg kg^{-1} for residential use in the Flanders. Similar is the case for Pb in the non-European countries (lowest is 120 for Ontario-Canada, while highest is 1200 for Australia) (Table 7; values in mg kg^{-1}). It is noteworthy that As is not even regulated in the EU Directive; because of that, As is non-regulated also in European countries that have complied with the EU Directive (such as Greece); outside Europe, India is also a country not regulating As. Likewise, Pb is not regulated in the UK and the USA. Similar is the case with Hg in residential use (ranging from 1 to 15 in Europe and 0.27 to 60 outside Europe), in industrial use (5 to 600 in Europe; 0.27 to 75 outside Europe), or when soil use is not specified (1.5 to 132 in Europe and 1.5 to 20 outside Europe), while in India Hg is not even regulated. Great is also the diversity concerning limits of Zn, Cu, and Ni whether in specified land uses or when use is not specified (Table 8; values in mg kg^{-1}). All above mentioned evidence shows the great discrepancy in environmental policy across the various countries.

These variations among countries are difficult to be explained with respect to environmental, climatic or soil conditions. Even, there are some countries which share almost similar environmental, climatic or soil conditions, but have high variations in maximum level of values in regulations for TEs. The issue spans at global scale, especially for those countries where the regulations are not well-established for TEs (such as African and South-Asian countries). In these countries, the scientists, researchers and regulatory authorities use regulation guidelines set by other countries or organizations, which makes the findings/recommendations very complicated, because of the difference in environmental, climatic or soil conditions. For example, in Pakistan, scientists and regulators tend to use maximum level of values from USEPA and FAO/WHO.

3.2. Health risk assessment as related to trace elements in regulation limits

3.2.1. Rationale of the use of values for trace element concentrations and critique for the risk assessment values

For the evaluation of carcinogenic and non-cancinogenic human health risk assessment (HRA), soil TE concentrations (C_S) were obtained

Table 8
Limits for Zn, Cu, Ni, and Hg in various countries, corresponding Hazard Quotients (HQ) for adults and children. Significant non-cancer health hazard exists for HQ > 1. Values in italics indicate such elevated risks. For each country, the highest value for each land use is chosen. Limits are reported in mg kg^{-1}, while HQ is unitless.

Residential

	Zn			Cu			Ni			Hg			
Europe	Limits	HQ-Adult	HQ-Child	Limits	HQ-Adult	HQ-Child	Limits	HQ-Adult	HQ-Child	Limits	HQ-Adult	HQ-Child	References for limits
Italy (SV)	150	0.001	0.006	120	0.004	0.038	120	0.008	0.077	1	0.009	0.080	Carlon (2007)
UK (SV)	350	0.002	0.015	NE	–	–	130	0.009	0.083	1	0.009	0.080	Carlon (2007)
Wallonia (IV)	230	0.001	0.010	110	0.004	0.035	150	0.010	0.096	1	0.009	0.080	CO (Coordination Officieuse) (2008)
Germany (TV)	NE	–	–	NE	–	–	140	0.010	0.089	20	0.171	*1.597*	BBodSchV (1999)
Czech R (AV)	2500	0.011	0.106	600	0.020	0.192	250	0.017	0.160	10	0.085	0.798	Carlon (2007)
Flanders (IV)	1000	0.005	0.043	400	0.014	0.128	470	0.032	0.300	15	0.128	*1.198*	Chernova and Beketskaya (2011)
Brussels	1000	0.005	0.043	400	0.014	0.128	470	0.032	0.300	15	0.128	*1.198*	Carlon (2007)
Non-Europe													
USA (SV)	23,000	0.105	0.979	3100	0.105	0.990	NE	–	–	11	0.094	0.878	Ontario (2011)
Ontario-Ca (SS)	340	0.002	0.014	140	0.005	0.045	100	0.007	0.064	0.27	0.002	0.022	Ontario (2011)
Alberta-Ca (Res/Park)	200	0.001	0.009	63	0.002	0.020	45	0.003	0.029	6.6	0.056	0.527	Alberta Government (2016)
Canada (SQG)	250	0.001	0.011	63	0.002	0.020	45	0.003	0.029	6.6	0.056	0.527	CCME (Canadian Council of Ministers of the Environment) (2018)
S. Africa (StandRes)	19,000	0.086	0.809	2300	0.078	0.735	620	0.042	0.396	1	0.009	0.080	MoE (Ministry of Environment, Korea) (2018)
Korea (SCCL, Z.1)	900	0.004	0.038	450	0.015	0.144	300	0.020	0.192	12	0.102	0.958	MoE (Ministry of Environment, Korea) (2018)
Australia (HIL)	28,000	0.127	*1.192*	4000	0.136	*1.278*	2400	0.163	*1.533*	60	0.512	*4.790*	DoEC (Department of Environment and Conservation, Australia) (2010)
HQ1-Ad	219,780	1		29,412	1		14,706	1		117	1		
HQ1-Ch	23,485		1	3131		1	1566		1	13		1	

Industrial

	Zn		Cu		Ni		Hg		
Europe	Limits	HQ-Adult	Limits	HQ-Adult	Limits	HQ-Adult	Limits	HQ-Adult	References for limits
Italy (SV)	1500	0.004	600	0.013	500	0.022	5	0.027	Carlon (2007)
Poland (SQS, Deep/LHC)	3000	0.009	1000	0.022	500	0.022	50	0.273	Carlon (2007)
France (ISV)	NE	–	950	0.021	900	0.039	600	*3.281*	Carlon (2007)
Czech R (AV)	5000	0.015	1500	0.033	500	0.022	20	0.109	Carlon (2007)
Germany (TV)	NE	–	NE	–	900	0.039	80	0.438	BBodSchV (1999)
Flanders (IV)	700	0.002	800	0.018	700	0.031	30	0.164	Chernova and Beketskaya (2011)
Brussels	3000	0.009	800	0.018	700	0.031	30	0.164	Carlon (2007)
Wallonia (TV)	1300	0.004	500	0.011	210	0.009	5	0.027	CO (Coordination Officieuse) (2008)
UK (SV)	NE	–	NE	–	1800	0.079	480	*2.625*	Carlon (2007)
Non-Europe									
USA (SV)	350,000	*1.022*	47,000	*1.028*	NE	–	46	0.252	USEPA (United States Environment Protection Agency) (2016)
Ontario (SS)	15,000	0.044	5600	0.123	510	0.022	0.27	0.001	Ontario (2011)
Alberta	360	0.001	91	0.002	89	0.004	24	0.131	Alberta Government (2016)
Canada (SQG)	410	0.001	91	0.002	89	0.004	50	0.273	CCME (Canadian Council of Ministers of the Environment) (2018)
Australia (HIL)	35,000	0.102	5000	0.109	3000	0.131	75	0.410	DoEC (Department of Environment and Conservation, Australia) (2010)
Mexico	NE	–	NE	–	NE	–	310	1.695	Perez-Vazquez et al. (2016)
S. Africa (Comm/Ind)	150,000	0.438	19,000	0.416	10,000	0.438	6.5	0.036	GGS (Government Gazette Staatskoerant, South Africa) (2012)
Korea (SCCL, Z.3)	5000	0.015	6000	0.131	1500	0.066	60	0.328	MoE (Ministry of Environment, Korea) (2018)
HQ1-Ad	342,466	1	45,714	1	22,857	1	183	1	

Agriculture (as occupational exposure)

	Zn		Cu		Ni		Hg		
Europe	Limits	HQ-Adult	Limits	HQ-Adult	Limits	HQ-Adult	Limits	HQ-Adult	References for limits
Austria (SV)	300	0.001	100	0.002	60	0.003	10	0.061	CF (Common Forum – Working Document) (2009)
Wallonia (IV)	155	0.001	50	0.001	65	0.003	1	0.006	CO (Coordination Officieuse) (2008)
France (ISV)	9000	0.029	190	0.005	140	0.007	7	0.043	Carlon (2007)
UK (SGV)	NE	–	NE	–	230	0.011	26	0.159	CLAIRE (2018)

(*continued on next page*)

Table 8 (*continued*)

Agriculture (as occupational exposure)									
	Zn		Cu		Ni		Hg		
Europe	Limits	HQ-Adult	Limits	HQ-Adult	Limits	HQ-Adult	Limits	HQ-Adult	References for limits
Flanders (IV)	600	0.002	200	0.005	100	0.005	10	0.061	Chernova and Beketskaya (2011)
Poland (SQS, deep/LHC)	720	0.002	100	0.002	210	0.010	10	0.061	Carlon (2007)
Czech R (AV)	720	0.002	190	0.005	210	0.010	10	0.061	Carlon (2007)
Germany (AV)	NE		1300	0.032	1900	0.093	5	0.031	BBodSchV (1999)
Non-Europe									
Alberta-Ca	200	0.001	63	0.002	45	0.002	15	0.092	Alberta Government (2016)
Canada (SQG)	250	0.0008	63	0.002	45	0.002	6.6	0.040	CCME (Canadian Council of Ministers of the Environment) (2018)
Korea (SCCL, Z.2)	1800	0.006	1500	0.037	600	0.029	30	0.184	MoE (Ministry of Environment, Korea) (2018)
Mexico	NE	–	NE	–	NE	–	25	0.153	DoEC (Department of Environment and Conservation, Australia) (2010)
Australia (HIL)	7000	0.023	1000	0.025	600	0.029	15	0.092	Perez-Vazquez et al. (2016)
S. Africa (PEH)	240	0.001	16,000	0.392	1400	0.069	4.1	0.025	GGS (Government Gazette Staatskoerant, South Africa) (2012)
HQ1-Ad	305,810	1	40,816	1	20,408	1	163	1	

Use non-specified					
Europe	Zn	Cu	Ni	Hg	
Russia (PPC, clay/pH > 5.5)	200	250	80	132	Chernova and Beketskaya (2011)
Denmark (TV)	1000	1000	30	3	DEPE (Danish Environmental Protection Agency) (2002)
Spain	80	NE	405	5	Nadal et al. (2016)
Sweden (GV, non-sens)	500	200	120	2.5	INSURE (2017)
Latvia (CV)	700	150	200	10	INSURE (2017)
Slovakia (LVC)	3000	500	500	10	Carlon (2007)
Austria (IV)	NE	600	140	10	CF (Common Forum – Working Document) (2009)
Netherlands (IV)	720	190	210	10	MvV (Ministerie von Volkshuissvesting) (2000)
EU Dir (High)	300	140	75	1.5	CEC (Council of the European Communities) (1986)
Lithuania (IV)	1200	200	300	1.5	CF (Common Forum – Working Document) (2009)
Finland (GV, High)	400	200	150	5	MotE (Ministry of the Environment, Finland) (2007)
Non-Europe					
China (Grade III, pH > 6.5)	400	500	200	1.5	Wang and Shan (2013)
India (High)	270	600	150	NE	Saha et al. (2015)
Taiwan	400	2000	200	20	Lai et al. (2010)
Armenia	132	220	80	2.1	Tepanosyan et al. (2017)

SV = Screening Value; IV = Intervention Value; TV = Trigger Value; AV = Action Value; SS = Stratified Site Condition Standards in a Non-potable Ground Water Conditions; Res/Park = Residential/Parks; SQG = Soil Quality Guidelines; StandRes = Value for "Standard Residential Area"; SCCL = Soil Contamination Counterplan Limit; Z.1 = Zone 1 (includes residential area); HIL: Health Investigation Levels; HQ1-Ad = Limit value that would generate an HQ value equal to unity for adults; HQ1-Ch = Limit value that would generate an HQ value equal to unity for children; SQS = Soil Quality Standards; Deep/LHC = Agricultural use/deep soil (depth > 15 m/low hydraulic conductivity ($< 10^{-7}$ m sec^{-1}); ISV = Impact Statement Value; Comm/Ind = Commercial/Industrial; Z.2 = Zone 2 (includes agricultural land use); PEH = "Protection Ecological Health." PPC = Provisional Permissible Concentration; Non-sens = Non-sensitive soil use; CV = Control Value; VDM = Value for Decontamination Measures; High = High value in a range of values; GV = Guideline Value; Grade III = Applying for High Adsorption Capacity Soils Only; NA = Non-applicable; NE = Non-existent.

directly from the regulation limit values. We considered that this was the most appropriate means of evaluating the potential risks in the case that the upper regulation limit values are indeed achieved. However, this approach creates a two-fold drawback: (a) not all countries have limit values for the same TEs, and, subsequently, (b) absence of limit values for certain TEs simply mean lack of regulation and does not in any way imply lack of risk. Bearing this in mind, for these soil-related risk calculations we decided to use only seven specific TEs which are most commonly reported in regulations: As, Pb, Cd, Zn, Cu, Ni, and Hg. Thus, other TEs, not necessarily of less importance, could not be used for the sake of performing meaningful comparisons among countries. Such non-used TEs observed in regulations often included Ag, Ba, Be, Cr, Mo, Sb, Se, Sn, Tl, and V.

We evaluated the scenario in which the seven studied elements have separately reached the maximum allowable concentrations in residential, industrial, and agricultural soils, as well as in cases where the soil use is not specified. We then evaluated the non-carcinogenic risk to adults and children concerning chronic exposure to the seven studied TEs. For this, only the limit values were obtained that were set for the specific land uses. Moreover, the limit values were distinguished according to soil properties, bearing in mind that in such cases, residential land use is not exempted. In this case (residential, non-carcinogenic for adults and children), we accounted for all three human exposure possibilities, i.e., ingestion, dermal conduct, and inhalation.

Concerning non-residential land uses, we accounted for industrial and agricultural areas. Thus we dealt with occupation-related risks, which are linked by definition to adults only. In that case, we altered the risk assessment parameters so as to accommodate the particular reality of occupational labour workers and similar employees, as suggested by relevant USEPA documents and reported in Table 1.

According to the US EPA, the only carcinogenic TEs are As, Pb, and chromate (Cr(VI)). Because in this study we did not deal with Cr, we focused on As and Pb only concerning cancer risk. The physical meaning of CR is that a CR = 10^{-n} indicates the risk of one additional person out of a group of 10^n persons to develop cancer. For cancer risk (CR), the risk unitless threshold is 10^{-4}; values lower than 10^{-6} indicate non-significant possibility of a risk, while values between 10^{-4} and 10^{-6} are in an intermediate state and still acceptable. In order to present CR in a clear manner, we expressed it as p(CR) = −log(CR) which means that there is a significant carcinogenic risk for p(CR) < 4

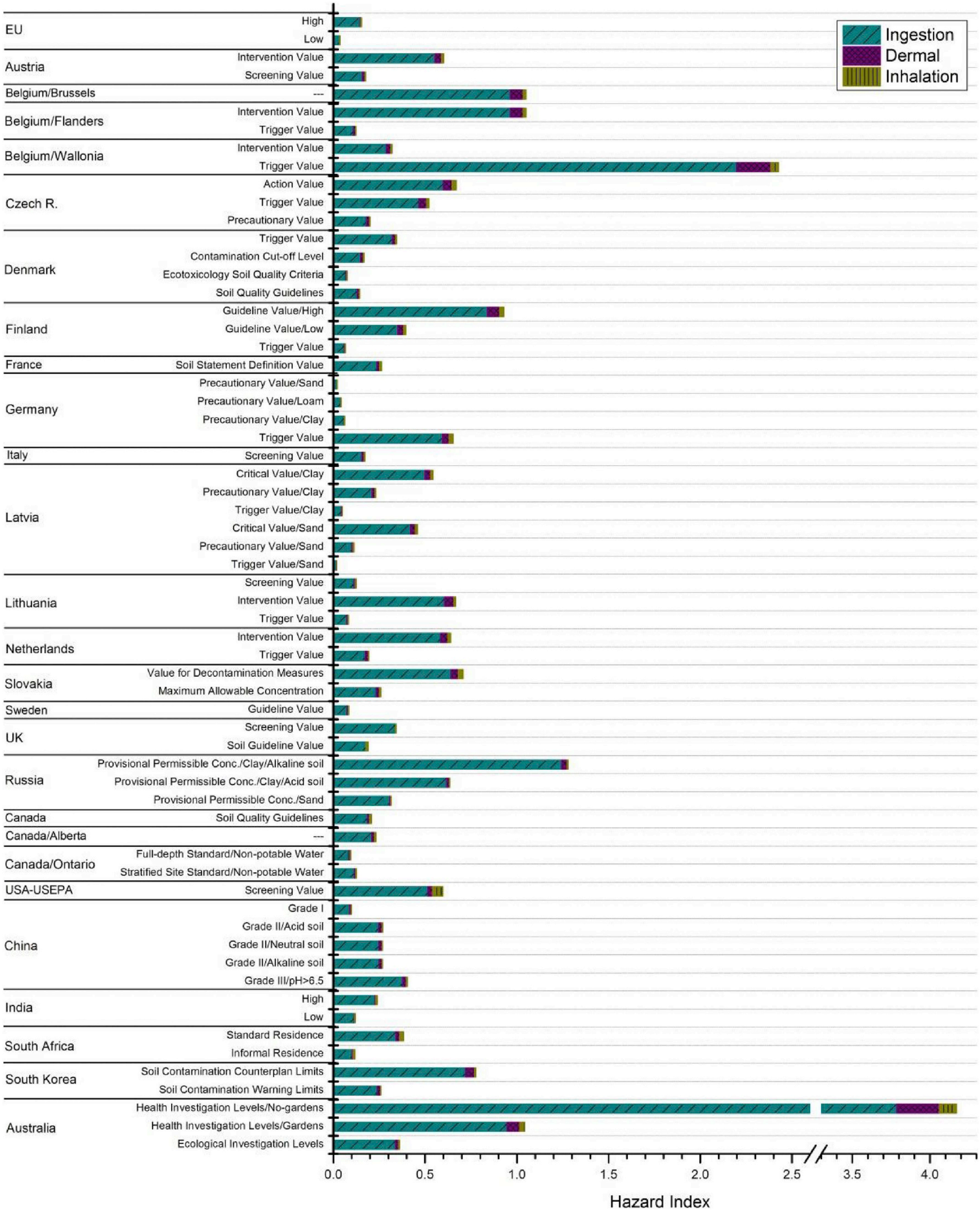

Fig. 2. Hazard Index (HI) for non-cancer health effects to adults concerning soil in residential areas. Values of HI > 1 indicate significant non-carcinogenic health risk.

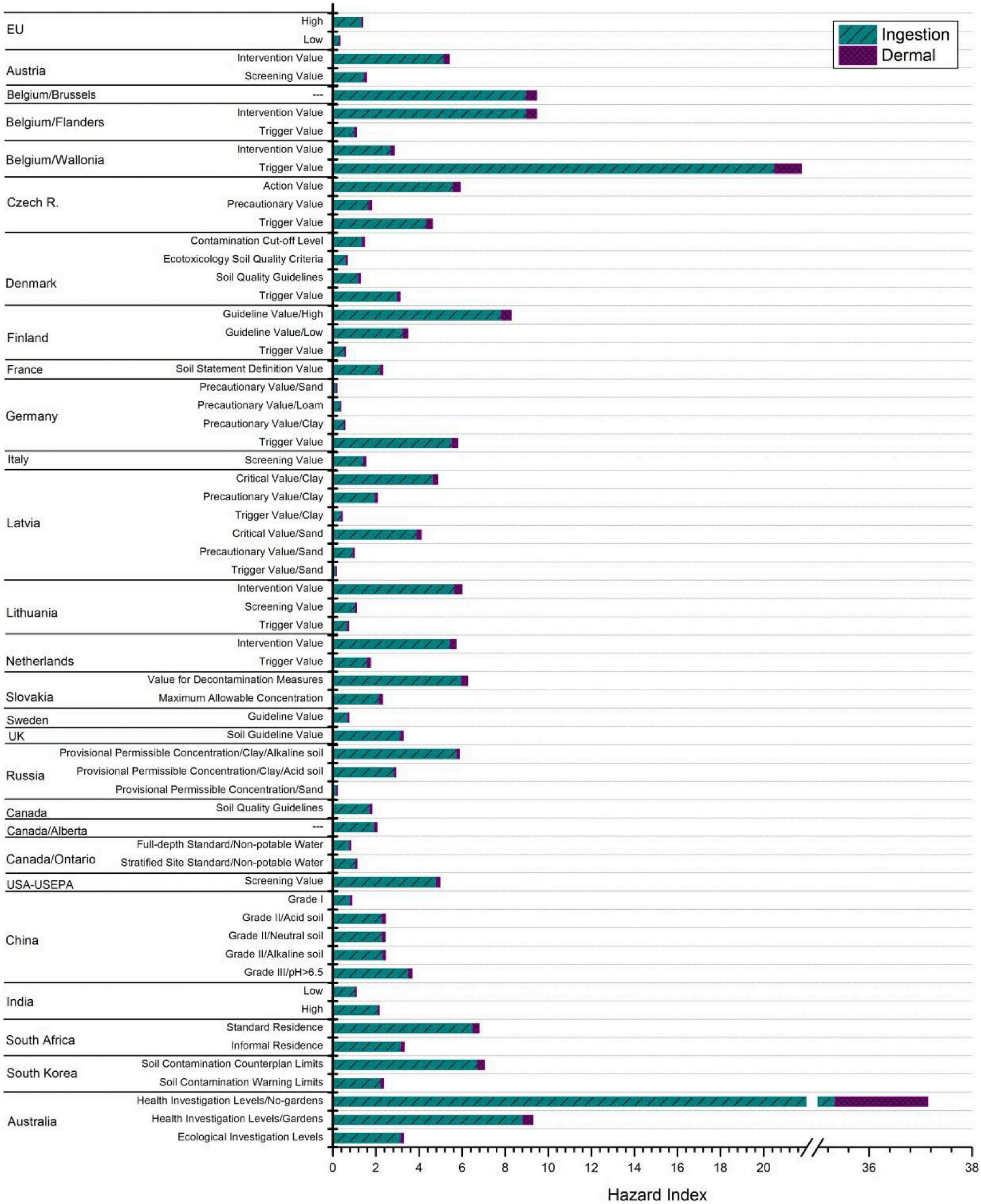

Fig. 3. Hazard Index (HI) for non-cancer health effects to children concerning soil in residential areas. Values of HI > 1 indicate significant non-carcinogenic health risk.

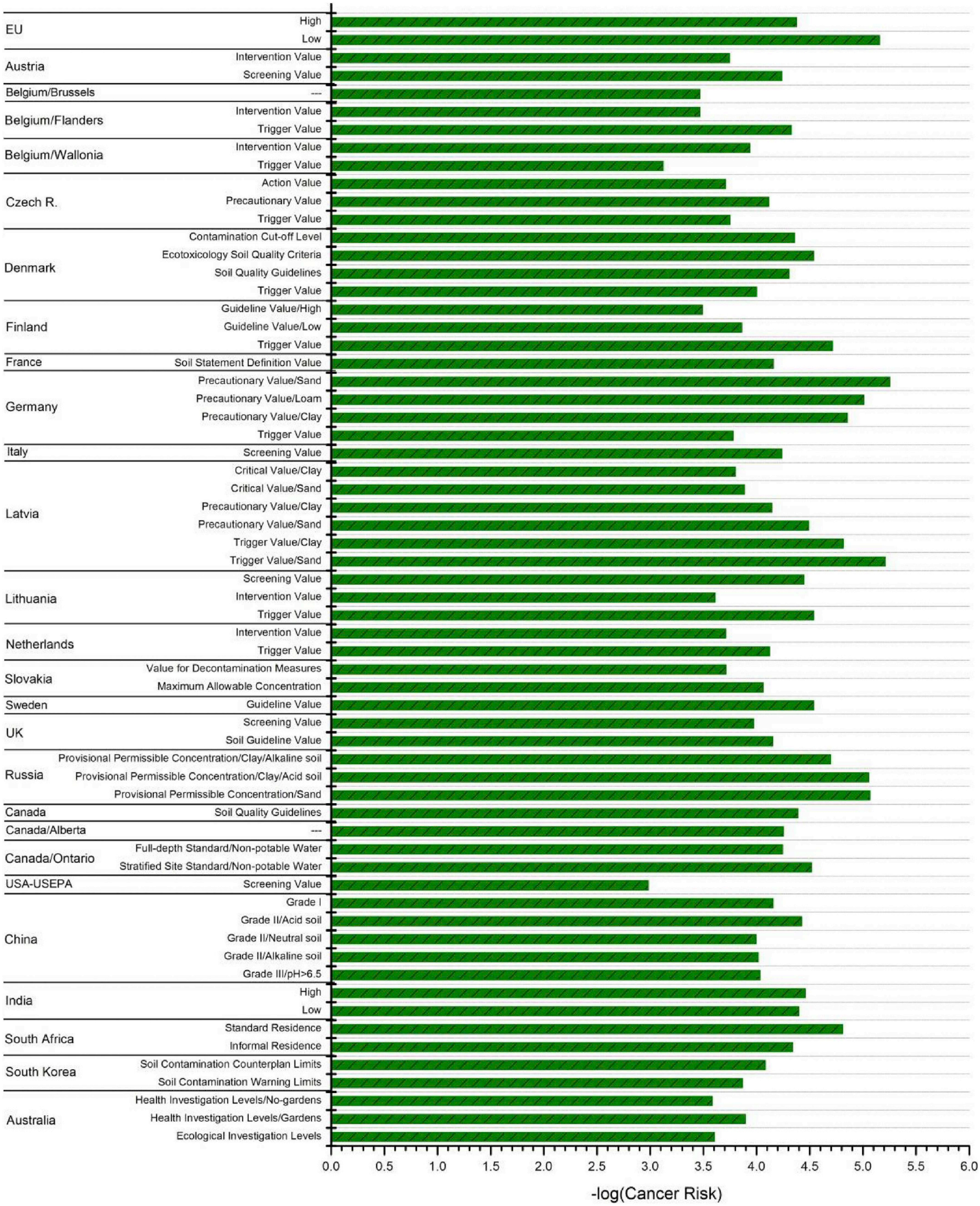

Fig. 4. Cancer health risk (CR) to adults concerning soil in residential areas expressed as –log(CR). Values of –log(CR) < 4 indicate significant carcinogenic risk.

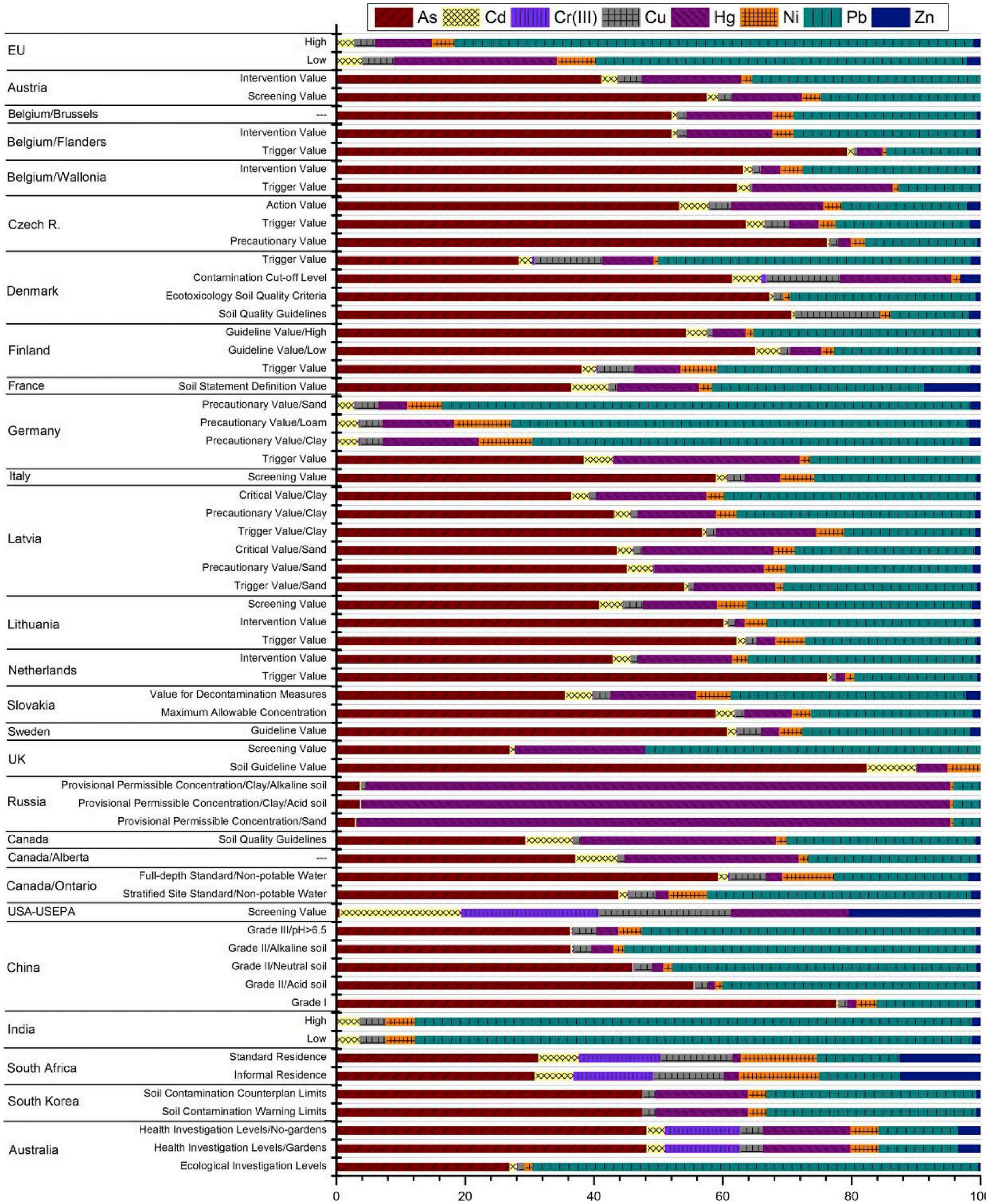

Fig. 5. Percentage of the contribution of potentially toxic element Hazard Quotient values to the total Hazard Index for non-cancer health effects concerning soil in residential areas.

Table 9
Values reported in monitoring papers from various countries across the globe. Unique values represent median values, values in parentheses are the maximum of the reported values, while values presented as range represent minimum and maximum of reported values. All values in mg kg^{-1} soil.

Country	As	Cd	Pb	Zn	Cu	Ni	Hg	Remarks	Area	Ref
2018										
China	7.8	0.17	27.0	131.1	27.9		0.49	Soil	Shanghai industrial area	Bi et al., 2018
	9.4	0.46	105.6	473.5	132.2		0.29	Dust		
	17.6 (538.5)	0.31 (3.42)	51.6 (441)	139 (807.6)	63.4 (3515)			Cu mine soil	Anhui Province, Tongling	Ding et al., 2018
	18.2 (109.5)	69.1 (189)	157.2 (512.3)	90.9 (201.2)	29.8 (79)	36.6 (74.4)	0.43 (2.18)		SouthEast China	Huang et al., 2018
	7.6 (11.6)	0.29 (1.7)	29.7 (160)	76.5 (176)	31.6 (90)	33 (23.8)	1.53 (31.800)		Beijing green space	Lin et al., 2018
		0.9	86.7	178.3	65.6	30.8		Soil 1	Hebei and Jiangxi Provinces	Liu et al., 2018
		0.1	10,332	67.1	23.8	25.4		Soil 2		
		5	847.5	878.1	43	34.6		Soil 3		
		7.6	1354.1	1297.9	46.6	28.2		Soil 4		
	53.1 (352.2)	9.98 (130.6)	77.3 (654.4)	486.4 (5080)	45.7 (352.6)				Xikuangshan, Sb mine area	Long et al., 2018
	5.48 (31)	0.3 (1.39)	27.6 (336)	82.8 (9238.5)	25.9 (106.5)	32.1 (96.4)	0.08 (1.61)		Hechuan County, Upper Yangtze	Ni et al., 2018
	587.3 (1600)	31.9 (630)	6041 (21400)	22,749 (67600)	530.6 (2770)		1.21 (7.99)		Zn smelter area-Guizhou Prov.	Peng et al., 2018
	14.2 (27)	2.31 (8.14)	122 (203)	512 (684)	29.3 (35.9)			Site 1	Hebei Prov.	Sun and Chen, 2018
	80.1 (1695)	10.9 (621)	2725 (26288)	2220 (11461)	209 (1402)			Site 2		
	329 (1464)	106 (164)	26,143 (49973)	23,413 (39353)	429 (956)			Site 3		
	26.3	4.03	115	123.8	72.2	51.9		Commercial	Xiangyang city, Hubei Pr.	Wu et al., 2018
	23.4	3.42	63.0	100.4	59.2	46.3		Roadside		
	19.5	2.54	51.3	82.3	55.9	47.6		Farmland		
	24	3.61	77.8	90.7	57.2	51.3		Residential		
Australia	185	0.4	76	328	59	23	9		Maldon, Victoria, mine area	Desjardins et al., 2018
Canada	3.4	0.9	66.3	230.1	79.3				Montreal botanical gardens	Abraham et al., 2018
Sweden	180	13	800	330	24			Soil 1	Glasswork sites	Hagner et al., 2018
	2200	4.4	12,000	430	62			Soil 2		
	380	6.5	7900	350	140			Soil 3		
	500	5.5	1200	340	42			Soil 4		
	170	19	7000	420	48			Soil 5		
Korea	1540	55	1293						Abandoned mine area	Kim et al., 2018
Russia	3.2	0.7	26.4	106.1	60.6	57.5		Soil 1	Barens Region, Pasvik Valley	Magiera et al., 2018
	1.9	0.6	33.3	379.1	189.9	115.3		Soil 2		
	3.7	0.5	28.7	54.2	170.2	213.4		Soil 3		
	6.8	0.9	30.2	91.8	413.7	651.2		Soil 4		
Spain	4.77	0.16	16.1		20.6	10.8	0.01	Year 2015	Around municipal waste incinerator-Campdora-Girona	Rovira et al., 2018
	4.62	0.20	18.2		37.7	11.4	0.01	Year 2016		
Italy	17.1	0.47	119	156	131	27		Soil 1	Vesuvius National Park, Naples	Memoli et al., 2018
	60.6	0.8	152	149	786	21.9		Soil 2		
	7.42	0.21	48.5	133	107	19.4		Soil 3		
	13.5	0.43	46.9	45.2	120	18.1		Soil 4		
	10.5	0.93	152	166	102	31		Soil 5		
	10.5	0.77	149	162	84.5	36.2		Soil 6		
	6.49	0.1	28.2	40.9	78.1	21.6		Soil 7		
	9.29	0.19	38.2	19.9	84.4	21.1		Soil 8		
	3.1–4.7		1.1–1.6			1.8–2.8			Plain of Gela, Sicily	Salvo et al., 2018
2017										
China	1.1	0.01	73.6	18.1	4.8	5.4			U mine area-Bayan Pr.	Bai et al., 2017
	26.5 (34.1)	0.05 (0.11)	30.3 (41.5)					Soil 1	Mining area-Hunan Pr.	Fan et al., 2017a, 2017b
	142.7 (562.3)	0.86 (1345)	365.2 (756)					Soil 2		
	32.6 (70.2)	0.05 (0.13)	13.7 (33.4)					Soil 3		
			43.7–189.6	131.4–239.5	24.7–37.7				Rb/Zn smelting area-Shaaxi Pr.	Fan and Wang, 2017

(*continued on next page*)

Table 9 (*continued*)

Country	As	Cd	Pb	Zn	Cu	Ni	Hg	Remarks	Area	Ref
	97.7 (106)		728.9 (910)	130.5 (151)				Mine tailing	Midstream area of Yuxi River	Fei et al., 2017
	107.7 (132)		3074 (3590)	971.1 (1123)				Wasterock		
	26.9 (29.3)		70.6 (79.6)	276.1 (294.1)				Farmland		
	72.7 (219.8)	0.18 (0.35)	90.7 (139.8)	125.6 (292.90	40.7 (112.9)	13.6 (26.7)			Tungsten mine area, Lianhuashan Pr.	Guo et al., 2017
	76.3 (93.3)	1.8 (2.47)	61.7 (57.4)	61.7 (106.5)	46.5 (54.1)				Shanghai-waste electrical processing area	Gu et al., 2017
	7.75 (16.7)	0.13 (0.49)	19.8 (31.7)	74.2 (137)	28.3 (137)		0.03 (0.12)		Riparian soils-Miyun Reservoir, $N = 425$	Han et al. (2017b)
		0.3 (0.6)	35.5 (68.9)	45.8 (88.7)	24.7 (53.9)	24.7 (32.4)	51.3 (192)		Industrial city, Baoton, parks	Han et al. (2017a)
	24.5 (104.3)	0.37 95.37)	50 (108)		30.9 (86.8)		0.7 (2.49)	Cropland	Coal-fired power plant-Guizhou Pr.	Huang et al., 2017
	4.46 (31.2)	0.67 (1.03)	36.6 (39)		22.5 (51.3)		0.38 (0.52)	Forest		
	6.38 (19.1)	0.18 (1.83)	42.6 (263)	107 (704)	33.5 (160)	30.4 (293)	0.21 (2.26)		Yangtze River soils, $N = 18{,}212$	Haribala et al., 2016
	7.36 (10.8)	0.11 (0.27)	30.48	57.2 (102.1)	30.8 (55.9)	34.3 (60.1)			Jiangsu Pr, $N = 101$	Jiang et al. (2017a)
		0.81 (3.21)	34.2 (61)	56.6 (109)	77.4 (151)			Street dust	Langhau, Gansu Pr.	Jiang et al. (2017b)
	14.9 (512)	0.59 (8.71)	37.8 (745)	107.2 (1112)	33.3 (719.6)		178.2 (3601)		Linyuan, coal mine city	Liang et al., 2017
	6.78 (14.2)	0.22 (0.75)	24.1 (58.9)	82.2 (153)	30.5 (162)	27.6 (52.4)	0.04 (0.21)	Qingshui R.		Li et al. (2017c)
	5.46 (7.75)	0.23 (0.29)	32.8 (44.8)	93.5 (122)	27.9 (36.4)	24.6 (25.9)	0.09 (0.17)	Yongding R.		
	7.96 (18.5)	0.76 (1.52)	81.5 (265.3)	544.9 (749)	62.9 (111.7)			Utilities	Lanyungang urban soils	Li et al. (2017c)
	10.8 (13.8)	0.56 (0.56)	46.8 (55.1)	564 (580.2)	78.9 (89.7)			Commercial		
	2.82 (11.3)	1.23 (3.71)	155.4 (498.5)	699 (1851)	110.5 (283.1)			Industrial		
	31 (66.6)	0.84 (1.7)	161.4 (389.6)	574.9 (577.7)	57.8 (84.4)			Roadside		
	8.41 (17.7)	0.64 (0.98)	28.2 (66.7)		38.5 (57)		0.90 (1.62)	Jingxi	Guangxi Pr.	Lin et al., 2017
	6.27 (10.8)	0.49 (0.65)	24.8 (51.1)		30.1 (34.1)		0.69 (1.05)	Debao		
	7.27 (17.9)	0.63 (2.54)	25.4 (44.6)		29.4 (56.9)		0.77 (1.54)	Longlin		
	33.6 (68.1)	0.55 (1.94)	14.5 (24.6)		8.48 (25.4)		NA	Leye		
	39.7 (112)	0.97 (4.22)	25.1 (62.7)		21.5 (64.5)		0.03 (0.30)	Lingyun		
		2.07 (6.40)	67.2 (96.4)	344 (549)	149 (252)	35.8 (69.8)		Soil 1	Urban street dust, Chengu city	Li et al. (2017)
		1.95 (3.93)	98.5 (162)	334 (478)	107 (331)	24.7 (33.4)		Soil 2		
		1.14 (2.73)	71.8 (152)	255 (503)	80.1 (121)	19.3 (29.5)		Soil 3		
		1.51 (3.85)	90.2 (164)	272 (607)	97.4 (235)	22.6 (37.2)		Soil 4		
		1.74 (2.49)	78.8 (114)	375 (386)	61 (83.7)	20.1 (25)		Soil 5		
		1.51 (8.52)	102 (760)	216.1 (923.7)	41.3 (108.1)				Pb/Zn mine are, Yunnan	Wang et al. (2017a)
	6.95 (9.58)	0.21 (0.43)	43.7 (52.6)		36.6 (65.5)		0.21 (0.72)	Soil 1	Huainan city, Anhui Pr.	Tang et al., 2017
	6.67 (9.36)	0.27 (0.45)	46.9 (71.5)		38.7 (60.1)		0.29 (3.02)	Soil 2		
	6.95 (9.43)	0.27 (9.43)	44.9 (70.4)		46.7 (87.5)		0.09 (0.15)	Soil 3		
	15.5 1(55.46)	0.33 (4.8)	40.2 (243)				0.52 (1.81)		Suzhou city (industrial)	Wang et al. (2017b)
	28.9 (52.8)		324 (733.9)	322.7 (1194)					Along Dahuanjing R.	Wan et al., 2017
	55 (82)	2.6 (5.6)	423 (708)	1039 (1723)	99 (1212)	39 (68)	0.22 (0.89)	Street dust	Hezhang city, Guizhou Pr.	Wu et al., 2017
Portugal	827 (3568)	5.5 (19)	45 (128)	462 (1262)	346 (766)			Tailing 1	Mine tailings, Panasqueira mine	Avila et al., 2017
	174 (519)	1 (2.3)	17 (23)	157 (267)	135 (243)			Tailing 2		
	513 (2288)	2.5 (10)	46 (153)	320 (707)	269 (879)			Tailing 3		
	614 (1777)	4.8 (15)	15 (25)	448 (1028)	590 (2379)			Tailing 4		
Greece		1.68 (4.95)	180.4 (429)	408.4 (1075)	640.1 (1706)	86.0 (211.5)			Thessaloniki city street dust	Bourliva et al., 2017
Armenia	15.3		54.5	158	39	33	0.88		Yerevan urban areas	Tepanosyan et al., 2017
France	8.32	0.43	99.2	106.9	44.5	14.4		Lawns	Paris urban areas	Foti et al., 2017
	8.30	2.45	188.1	174.8	105.2	14.0		Woods		
India	29.7	0.34	47	210.6	218	94.2			Singhbhum Cu belt	Giri and Singh, 2017
	2.81 (3.16)	2.61 (3.49)	13.8 (14.9)	25 (29)	13.6 (19.8)		0.47 (1.08)		Jharia coal field	Raj et al., 2017
Australia	1.6 (218)	0.04 (5.09)	7.22 (1090)	26.3 (262)	11.2 (150)	9.8 (3871)	0.01 (3.07)		Nationwide data	Reimann and de Caritat, 2017
S. Africa	917–1493	7.3–11.9	39.2–72.5	2204–5340	1029–1350	1076–4041		Tailings 1	Au mine tailings	Ngole-Jeme and Fantke, 2017
	3.09–17.3	1.08–8.42	0.9–28.2	52.7–81.9	4.01–18.9	0.85–26.3		Tailings 2		
	0.4–4	0.03–6	21.1–42.6	46.9–194.4	26.3–53.4	1.84–33.5		Control		
2016										

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Table 9 (*continued*)

Country	As	Cd	Pb	Zn	Cu	Ni	Hg	Remarks	Area	Ref
China	21.6 (60.8)	2.93 (5.90)	37.4 (225.5)	178.7 (830.1)	56.4 (551)	70.2 (395.7)			Tibetan Plateau	Bu et al., 2016
	44.2	0.59	116.1	300.1	108.1	85.4		Soil	Hunan Pr.	Cao et al., 2016
	75.8	2.15	72.1	250.2	33.4	119.3		Dust		
	9.75 (41.6)	0.13 (1.49)	79.6 (26.9)	78.3 (317)	27.3 (107)	25.7 (80.3)	0.12 (9.39)		Beijing urban area	Chen et al., 2016
	0.83 (9.63)0.17 (0.51)	1.73 (3.12)	34.9 (94.3)	30.2 (61.9)	7.74 (15.3)		0.09 (0.14)		U mine area, Tongliao	Haribala et al., 2016
	8.74 (15.21)	0.17 (0.51)	27.3 (1300)		33.3 (82.9)		0.15 (1.51)		Zhangjigang city, Yangtze River	Hu et al., 2016
	220.9 (278.9)	2.72 (3.82)	226.1 (270.3)	391.1 (505.9)	436.1 (505.5)			Paddy soil	Mine-affected soils, Guangdong Pr.	Liao et al., 2016
	177	2.11 (2.95)	193.2 (226.3)	288.5 (453.2)	346.9 (431.2)			Vegetable soil		
	144.1 (205.5)	1.52 (2.44)	156.2 (189.4)	189.6 (266.9)	242.9 (346.4)			Sugarcane soil		
	7.1 (20.3)		61.8 (243)	92 (501)	48.3 (445)	42.8 (628)	0.7 (2.8)		Dongguan, industrial sites	Liu et al. (2016a)
	11.94 (35.5)	0.64 (3.92)	60.32 (194.56)				0.11 (0.35)		Kaifeng city	Liu et al. (2016b)
	57.1 (208.5)	0.35 (0.91)	61.6 (151.1)		35.1 (65.3)		0.27 (0.94)		South China	Ou et al., 2016
	16.1 (77.9)	1.39 (4.32)	76.9 (358)	129 (1359)	67.2 (201)		0.42 (1.33)		Coal-fired power station, N. Guangong	Wang et al., 2016
	10.2	0.13	18.5	48.8	18.4			Soil 1	Tibetan Plateau, roadside soils	Zhang et al., 2016
	45.6	0.34	32.6	132.7	20			Soil 2		
	18.3	0.11	26.8	60.3	18.3			Soil 3		
Sweden		11 (20)	630 (1300)	2300 (8300)	110 (250)	17 (33)		Soil 1	Glass waste-contaminated soils	Augustsson et al., 2016
		830 (39,000	1210 (3100)	1100 (3900)	120 (240)	44 (76)		Soil 2		
		9.6 (17)	1400 (2000)	310 (440)	39 (83)	21 (30)		Soil 3		
		6.9 (25)	290 (540)	130 (240)	32 (56)	16 (21)		Soil 4		
		0.34 (0.46)	130 (200)	20 (31)	7.3 (13)	2.9 (3.4)		Soil 5		
Korea		1.16	14.5	159.4	9.73	8.36		Farmland	Wastewater treatment plant area	Choi et al., 2016
	200.2	6.6	1879.3	782.3	83.0			Soil 1		Kim et al., 2016
	12.4	17.4	981.6	1971.9	331.0			Soil 2		
	357.5	11.4	810.2	12,464	34.6			Soil 3		
Spain	16.6	0.13	9.99		6.16	6.35	0.17	Soil 1	Near landfill (Sesena, Toledo)	Nadal et al., 2016
	4.46	0.04	5.47		5.1	3.52	ND	Soil 2		
	10.51	0.08	10.7		8.18	8.18	ND	Soil 3		
	74.2	1.2	308	1250	40.6			Soil 1	Sierra de Aracena National Park (geogenically-contaminated soils)	Rivera et al., 2016
	74.2	1	301	1330	33.6			Soil 2		
	27.1	4.5	213	1050	4.9			Soil 3		
	346	32.9	1630	7680	26.2			Soil 4		
	94	21.6	947	5540	50.6			Soil 5		
	30.5	7.4	375	2640	40.2			Soil 6		
	82	30.4	3140	14,800	49.2			Soil 7		
	48.2	25.9	5000	8110	37			Soil 8		
	60.4	7.9	2820	14,800	34.6			Soil 9		
	104	116	634	12,000	52.3			Soil 10		
	114	319	774	18,800	55.1			Soil 11		
	70.6	71	4240	18,300	197			Soil 12		
	20.6–7268	0.2–7.8	107.4–15,008	119.5–3488.2	30.8–1894	15.9–109.2	0.6–79.5		Contaminated soil	Wcislo et al., 2016
Mexico	7.06 (10.4)	7.46 (16.9)	227 (247)				0.81 (1.3)	Soil 1		Perez-Vazquez et al., 2016
	5.52 (6.87)	3.72 (8.79)	227 (230)				0.56 (13)	Soil 2		
	51.8 (263)	3.8 (12.9)	108 (432)				0.57 (2.34)	Soil 3		
	8.43 (10.5)	4.15 (4.81)	219 (220)				1.54 (2.02)	Soil 4		
France	48.1	0.31	360					Soil 1	Paleao-pollution mine site, Vosges Mt.	Mariet et al., 2016
	26.6	0.24	100					Soil 2		
	17.2	0.13	74.6					Soil 3		
	28.5	0.33	101					Soil 4		
	201	0.37	1080					Soil 5		
	1280	0.33	14,900					Soil 6		
	230	0.29	760					Soil 7		
	18.9	0.07	64.4					Soil 8		

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Table 9 (*continued*)

Country	As	Cd	Pb	Zn	Cu	Ni	Hg	Remarks	Area	Ref
India	111	2.8	10.3			25.5		Soil 1	West Bengal contaminated sites	Singh et al., 2016
	77.4	1.46	3.7			11.5		Soil 2		
	99.2	6.9	5.06			25.1		Soil 3		
	62.6	163	7.48			36.5		Soil 4		
	117	80.3	79.4			33.5		Soil 5		
	84.2	4.52	9.47			29.3		Soil 6		
	52.4	3.12	4.3			19.5		Soil 7		
	47.5	5.43	7.3			22.3		Soil 8		
	46.2	113	5.2			21.3		Soil 9		
S. Africa	78.4 (115.2)	0.05 (0.05)	4.79 (10.2)	51.3 (82.5)	42.5 (55.8)	112.1 (152.5)	0.09 (0.13)		Witwatersrand Au mine basin	Kamunda et al., 2016
	2.77	3.44	32.8	247	49.5	16		Soil 1	Kwazulu-Natal area	Mahlangeni et al., 2016
	–	1.29	11.2	61.2	14.3	6.44		Soil 2		
	7.86	8.44	–	55	21.1	9.65		Soil 3		
	1.91	2.97	5.14	43.5	16	9.66		Soil 4		
	30.9	13.4	–	50.4	26.8	37.7		Soil 5		
	32.4	13.7	–	97.9	23.6	19.9		Soil 6		
	3.23	3.74	–	35.8	15.3	6.82		Soil 7		
	4.39	5.22	21.8	439	40.6	14.7		Soil 8		
	6.9	6.87	–	38.9	38.5	17.7		Soil 9		
	5.86	6.19	–	548	17.2	11.7		Soil 10		
2015										
China	348.2	0.71	96.5	186.1	62.1	18.9			Pb-acid battery plant	Cao et al., 2015
	22.1 (45.2)	0.25 (0.44)	24.9 (32.3)			36.2 (48.7)	0.45 (0.68)	Soil 1	Hunan Pr.	Zeng et al., 2015
	21.2 (34.8)	3.25 (28)	43.3 (83.6)			36.5 (44.5)	41.1 (56.8)	Soil 2		
	16.8 (45.2)	1.40 (28)	94.5 (262)			33.9 (68.7)	14.8 (192)	Soil 3		
	15.5 (85)	0.47 (12.9)	39.1 (2.3)				0.7 (0.23)		Orange orchards, Xinfeng County	Cheng et al., 2015
	4.05 (17.8)	0.15 (0.51)	19.6 (63.6)	50.8 (142.3)	21.4 (62.3)	26.8 (83.1)			Fuxin city, Liaoning Pr.	Chen et al. (2015a)
	9.7 (25.7)	0.14 (0.51)	25.6 (59.5)	69.8 (141.5)	23.2 (27.1)	27.4 (53.4)	0.05 (0.26)		*N* = 38,393 of China (90th percentile)	Chen et al. (2015b)
	18.7 (117)	1.3 (4.6)	32.8 (459)	89.4 (4150)	45 (1480)	24.7 (54.4)	0.07 (0.48)		Agricultural soils, Daye city	Du et al., 2015
	29.9 (71.7)	5 (75.4)	672 (2485)	1194 (8076)	244 (316)	73.2 (103)	0.33 (2.58)		Pb/Zn smelter, Huize city, Hunan	Li et al. (2015b)
	10.8 (225.7)	0.18 (2.95)	50.1 (284)			24.3 (69.1)			Agricultural soils, Pearl River Delta	Li et al. (2015c)
		63.1 (91)	15 (21.8)		13 (21.4)	9.1 (15)		Soil 1	Waste incinerator site	Li et al. (2015a)
		6.85 (33.9)	5.03 (9.98)		1.5 (5.4)	5.9 (8.1)		Soil 2		
	9.32 (86.8)	0.17 (1.57)	32.1 (92.1)	87.3 (336)	23.6 (112)	27.4 (96.7)	0.09 (0.49)		Pb/Zn mine area, Hunan	Lu et al., 2015
	231 (869)	2.14 (3.82)	204 (436)	244 (472)	36 (99.8)	36 (99.8)	45.7 (68.7)		Agricultural soils affected by mines, Ganxi R.	Ma et al., 2015
	64.5 (341.3)	2.94 (6.47)	179.6 (837.5)		46.6 (148.7)				Suxian County, Hunan Pr.	Song et al., 2015
	11.6 (52.1)	0.24 (1.55)	47 (244)	87.9 (799)	53.5 (629)		0.10 (0.80)		Jiangxi, mine area	Wu et al., 2015
	10.2 (13.7)	0.34 (0.89)	32.6 (51.7)	48.7 (63.4)	20 (34.2)	20.3 (39.9)		Soil 1	Roadside soils along Qinghai-Tibetan motorway	Zhang et al., 2015
	45.6 (69.9)	0.11 (0.44)	26.8 (43.5)	132.7 (364.1)	18.3 (24.2)	20.7 (33.7)		Soil 2		
	18.3 (23.7)	0.14 (0.32)	33.1 (83.4)	60.3 (75.9)	3.13 (29.7)	78 (102.8)		Soil 3		
	17.6 (20.1)	0.12 (0.37)	18.5 (29.7)		18.4 (28.1)	26.9 (42.9)		Soil 4		
Australia	93.4 (26)	2.85 (8.25)	709 (22,000					Soil 1	Mining areas	Dong et al., 2015
	3.2 (3.6)	0.6 (10.0.7)	100 (280)					Soil 2		
	5.1 (16)	1.5 (5.1)	420 (1130)					Soil 3		
	2.7 (3)	0.7 (0.9)	140 (300)					Soil 4		
	11 (15)	8.1 (98)	2360 (2260)					Soil 5		
India		15.9 (18.1)	102.9 (121.8)	67.4 (84.3)	27.8 (47.4)	36.8 (55.4)			Sewage-irrigated soil, Kolkota	Saha et al., 2015
Slovakia	87.4	1.5	102.8	478	196	102.1	6.7	Soil 1	Soils in former mine areas	Sestinova et al., 2015
	71.9	0.9	77.6	360	300	69.9	1.6	Soil 2		
Greece	18.8 (65)	1.54 (3.2)	100.9 (539)	90.6 (207)	33 (77)			Soil 1	Plain in Drama, N. Greece	Sofianska and Michailidis, 2015
	352 (1022)	8.9 (27.5)	1198 (1996)	739.5 (2140)	108.3 (153)			Soil 2		
USA	12. (35.1)	1.13 (3.62	334.1 (1178)	203.6 (634)				Hough	Post-industrial cities	Sharma et al., 2015
	21.5 (38.5)	1.65 (4.36)	123 (1004)	253.2 (545)				Weihland Park		
	7.5 (27.9)	0.34 (2.63)	52.6 (1540)	120 (6010)	39.3 (104.5)				Sacramento, CA	Solt et al., 2015

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Table 9 (*continued*)

Country	As	Cd	Pb	Zn	Cu	Ni	Hg	Remarks	Area	Ref
Denmark	22	2.5	580	1255	450	38				Warming et al., 2015
2014										
China	21.5	0.27	22.1	63.5	71.9	30.8			Near large coking plant, Shanxi Pr.	Cao et al., 2014
	20.8 (105)	0.13 (0.69)	25.6 (140)				0.29 (1.49)		Pearl River Delta	Chang et al., 2014
	10.9 (17.8)	0.21 (0.40)	26.2 (86)	86.1 (121)	20.9 (47.6)	29.8 (50.2)	0.12 (0.39)		Taiyuan city	Liu et al., 2014
	4.89 (10.1)	0.12 (4.79)	29.2 (69.7)	61.3 (156.7)	15.8 (85.7)	19.2 (153.5)	0.03 (3.16)		Yangi basin	Mamat et al., 2014
	9.63 (109)	0.42 (30.5)	85 (625)	190 (1950)	58.8 (180)	52.5 (219)	0.13 (1.1)		Tiexi Sistrict, Shenyang	Ren et al., 2014
	8.63 (12.5)	0.22 (1.26)	33.9 (49.4)	94.9 (188)	32.4 (61.7)	36.4 (51)	0.20 (0.65)		Yangtze Delta-industrial area	Xu et al., 2014
	6.22 (204)	0.4 (3.5)	77 (2764)	122 (1089)	48 (410)		0.03 (0.07)		Auhui Pr.	Zhao et al., 2014
Greece	29 (204)	0.4 (3.5)	77 (2764)	122 (1089)	48 (410)	111 (727)			Athens city areas	Argyraki and Kelepertzis, 2014
Portugal	223.6 (918)	1.3 (3.4)	58.6 (146.1)	323.2 (911)	163.2 (757.6)		0.1 (0.3)	Soil 1	Saint Fransisco de Asis-Panasqueira mine soils	Candeias et al., 2014
	97.8 (350)	0.2 (1.5)	26.7 (78.4)	85.7 (192)	41.7 (146)		0.1 (0.1)	Soil 2		
Spain	4.45 (11.68)	0.10 (0.33)	26.2 (2905)	29.9 (78.7)	8.98 (38.1)	6.54 (7.2)			Alcala urban soils (N = 90–97)	Pena-Fernandez et al., 2014
India		0.6	19.8	33.8	5.4				East coast (max. values)	Satpathy et al., 2014
	21.99	0.43	41.8	114.6	26.8	37.1			Roadside soils, Qinghai-Tibet Plateau	Wang et al., 2014
2013										
China	5.80 (16.5)	0.86 (4.54)	12.2 (762.45)	39.7 (417.1)	11.4 (272.9)	5.41 (302.7)	1.26 (6.56)	Au	Mine soils, Miyun Reservoir, Beijing	Huang et al., 2013
	4.37 (13.9)	0.57 (2.14)	139.2 (384.2)	133.9 (162.7)	65.4 (118.3)	89.3 (156.6)	0.23 (1.59)	Fe		
	8.87 (52.1)	0.38 (2.14)	39.5 (19.3)				0.54 (1.45)		Yangtze River Delta (95th percentile)	Liu et al., 2013
	6.97 (10.1)	0.11 (0.19)	18.8 (32.4)	88.5 (180.2)	25.6 (49.2)	23.4 (30.0)			Beijing city samples	Peng et al., 2013
	22.3	0.27	37.6	108.4	21.8	33.3			Roadside soils, Qinghai-Tibet Plateau	Yan et al., 2013
Sweden	–			40	7600	7		Soil 1	Metal-polluted hotspot areas	Fedje et al., 2013
	34			180	100	960		Soil 2		
Italy	2.5–3.5	ND-0.30	7.9–9	37–41	12–22	17–22	ND		Waste dumping area	Ferrara et al., 2013
France	8.1 (114)	ND (3)	27 (3408)		ND 213)				Playground soils	Glorennec et al., 2013
Spain	239	9	1215	2410	32	16		Soil 1	Abandoned mine soils (1–3: Ag mine; 4–5: Cu mine)	Gutierrez-Gines et al., 2013
	281	5.4	1742	1983	36	14		Soil 2		
	326	25.5	3855	5095	85	36		Soil 3		
	35.5	2.8	126	113	971	17		Soil 4		
	51.5	13.3	152	362	2725	14		Soil 5		
Korea	1.17	0.15	7.09	119	3.68		0.02		Abandoned Nokdang metal mine	Park and Choi, 2013
Italy	419 (793)	731 (1748)	8314 (15605)	38,912 (48451)	1910 (2834)		65.2 (138)	Soil 1	Abandoned mine	Marchiol et al., 2013
	256 (553)	407 (761)	5137 (7371)	92,371 (250080)	1788 (2844)		33.9 (125)	Soil 2		
	111 (210)	710 (1278)	3443 (25890)	11,819 (28664)	657 (1675)		18.3 (93.5)	Soil 3		
	261 (632)	256 (557)	6752 (14766)	27,353 (55947)	1501 (3493)		14.4 (44.1)	Soil 4		
	152 (432)	388 (797)	5359 (13502)	37,722 (133874)	2540 (72334)		24.5 (1.4)	Soil 5		
Greece	209 (634)	82.7 (174.1)	6378 (12567)	9514 (22292)				Site 1	Abandoned mine, Kirki, N. Greece	Nikolaidis et al., 2013
	60.7 (112)	33.5 (95)	2441 (4530)	4029 (9333)				Site 2		
	8.54 (19)	1.93 (7.5)	97.1 (521)	222 (345)				Site 3		
	17.1 (242)	0.5 (0.9)	15.2 (49)	94.6 (190)	94 (430)	165.3 (480)			Apple cultivated area, Central Greece	Skordas et al., 2013
Canada	5.5 (7.8)	0.35 (0.62)	12 (17)		26 (81)	22 (31)			Roadside soils, Toronto	Wiseman et al., 2013
2012										
China	7.32 (137.8)	0.07 (1.12)	41.8 (108.1)	47.4 (248.8)	16.7 (99.8)	11.1 (112)	0.15 (3.39)		Agricultural soils, Huizhou, Guangdong Pr.	Cai et al., 2012
	7.41 (14.9)	0.19 (0.57)	40.2 (70.9)	149.8 (510)	39.6 (196)		0.15 (3.38)			Chen et al., 2012
Canada	14.6	11.4	40.2	149.8	39.6		0.95	Soil 1	Industrial soils, Montreal	Bisone et al., 2012
	13.3	19	70.9	510	196		0.38	Soil 2		
Italy	15.6 (18.88)	1.2 (1.3)	47 (50)	103 (141)	18 (21)	20.5 (29)	0.5 (0.6)	Soil 1	Tuscany soils	Giannaccini et al., 2012
	7.7 (10.4)	0.5 (1.1)	27 (51)	76 (87)	19 (21)	53 (68)	0.3 (0.4)	Soil 2		
	6.8 (11)	0.2 (1)	26 (42)	90 (96)	17 (25)	46 (60)	0.5 (1.1)	Soil 3		
	7.1 (7.6)	0.8 (1)	42 (43)	81 (102)	20 (26)	59 (85)	0.3 (0.3)	Soil 4		
	4.7 (5.4)	1.3 (1.3)	31 (44)	73 (86)	14 (16)	48 (56)	0.0.4 (1.1)	Soil 5		
	711 (2282)	1 (5.7)	5171 (12155)	2031 (40363)	1495 (2746)	40.4 (136)			Fenice Capanne mine soils	Pignattelli et al., 2012

(*continued on next page*)

Table 9 (*continued*)

Country	As	Cd	Pb	Zn	Cu	Ni	Hg	Remarks	Area	Ref
India	210 (711)	ND	MD-114.6	31.9 (136.8)	9.3 (36.4)	17.4 (40.3)	0.4 (82.1)		Yamuna, Delhi	Sehgal et al., 2012
2011										
China	5.02 (60.03)	0.11 (10.02)	18 (980)	69 (3500)	8.4 (12700)	6.5 (1100)	0.05 (0.27)		Guiyu, electronic waste area	Li et al., 2011
	7.7 (20.6)	0.11 (1.79)	14 (301.8)	64.8 (402.3)	24.2 (128.2)		0.06 (1.60)		Beijing and Tianghu cities	Qiao et al., 2011
	23.9 (77.8)	4.4 (8.9)	39.3 (81.8)	220 (463.8)	530.6 (865)				Guixu Pr.	Xiao et al., 2011, 2017
India	ND-6.86	23.2 (38.6)	35.9 (49.2)	62.7 (7120)	51.5 (373)	80.4 (347)	ND (1.54)		Thane Region, Maharashtra	Bhagure and Mirgane, 2011
Sweden	6.3	8.5		225	225				Sulfide ore smelter, Ronnskarsverka	Berglund and Nyholm, 2011
Korea		9.6	1140	5190	105			Tailing 1	Au and Ag mine tailings and soils	Chon et al., 2011
		4.1	276	384	33			Soil 1.2		
		3.4	234	87	22			Soil 1.2		
		4.5	483	533	94			Soil 1.3		
		28.5	1630	5940	267			Tailing 2		
		6.2	128	802	89			Soil 2.1		
		5.1	88	753	62			Soil 2.2		
		5.6	112	312	952			Tailing 3		
		2.9	40	96	142			Soil 3.1		
		2.7	36	197	11,000			Soil 3.2		
		96.6	10,410	14,960	5820			Tailing 4		
		11	742	1620	233			Soil 4.1		
		4.2	336	475	101			Soil 4.2		
France		37.7	3967	4690				Tailing 1	Surface layers only	Escarre et al., 2011
		108	18,770	32,513				Tailing 2		
		745	38,305	129,032				Tailing 3		
Australia	90.6	22.5	12,500	7060	155				Mine-affected soil, Broken Hill	Munksgaard and Lottermoser, 2011
Greece	1.37 (1634)	0.02 (3.40)	3.86 (873.1)	13.4 (5416)	3.25 (184.2)	2.29 (78.7)	0.01 (0.54)		Kavala industrial area	Papastergios et al., 2011
Italy	89.2	5.1	995	2410	2726			Surface	As/metal-polluted soils	Vemerali et al., 2011
	423	2.67	118	860	1719			Subsurface		
2010										
China	8.1	0.22	35.4	92.1	29.7	26.7			Raodside soils, Beijing	Chen et al., 2010
	12.8 (23.2)	0.20 (0.38)	24.3 (32.3)	69.8 (124.3)	24.6 (38.9)		0.03 (0.19)		Yucheng city, Shandong Pr.	Jia et al., 2010
	55 (11)	0.05 (0.05)	9.9 (24)	55 (100)	19 (68)	17 (58)			Miyun Reservoir, Beijing	Luo et al., 2010
	8.01	0.97	236.6	753.3	257.6	66.4	0.14	Urban	Shanghui city areas	Shi et al., 2010
	10.38	0.80	148.4	380.6	127.5	63.2	0.23	Suburban		
		1.7 (7.4)	81.3 (501.9)	177.7 (661.8)	77.9 (227.3)	11.7 (83.8)	0.2 (1)		*E*-waste plant, Wenling city, Taizhou Pr.	Tang et al., 2010
	8.01 (15.28)	0.13 (0.24)	29.4 (41.4)	86.1 (224.4)	27.8 (78.4)		0.15 (0.61)		Paddy soils, Haining city	Wu and Zhang, 2010
India	2.07	0.55	16.5			12			Coal-fired thermal power station, Singraul Region	Agrawal et al., 2010
	10.6 (24.7)		50.1 (81.9)	38.1 (68.1)	18.1 (30.5)	25.4 (54.6)			Siddipet, Medak Distr., Andhra Pradesh	Dantu, 2010
Italy		1.8 (3.5)	100.8 (224.4)	147.2 (542.3)	70.3 (280.8)	40.4 (80.9)			Agricultural soils, Apulia	Buccolieri et al., 2010
Czech R.	1392 (2568)	16.1 (30.6)	50.7 (111)	1514 (2674)		21.9 (26)			Kutna Hora, contaminated soils	Kralova et al., 2010
Taiwan	14.8	25.3	13,100	26,300	9080	12,100	3.34		Contaminated soils, maximum values	Lai et al., 2010
USA	1.2–11.3		93–490						Schoolyard soil, N. Orleans	Presley et al., 2010
Canada	8.59 (38)	0.48 (5)	21.9 (149)	93 (1500)	31 (120)	34.5 (44)			Pb-contaminated soils, Qubec	Saint-Laurent et al., 2010
2009										
China	4.6 (15.5)	0.23 (1.03)	69.4 (182.1)	158.6 (386.1)	38.8 (294.3)		0.61 (2.4)		Near petrochemical complex, Guangzhou	Li et al., 2009
	776 (5520)	29 (184)	582 (5680)	1123 (3140)	120 (448)	41.3 (75.2)			Dachang (mining city)	Zhang et al., 2009
2008										

(*continued on next page*)

Table 9 (*continued*)

Country	As	Cd	Pb	Zn	Cu	Ni	Hg	Remarks	Area	Ref
China	5.45 (14.8)	0.23 (0.64)	46.6 (87.3)	109 (264.9)	34.8 (77.6)	19.5 (36.5)	0.72 (5.78)		Hangzhou, vegetable soils	Chen et al., 2008
	3.05 (5.58)	0.55 (7.86)	51.9 (64.6)		83.8 (236.9)	27.6 (52.7)	0.24 (0.76)		Taizhou, E-waste area	Fu et al., 2008
France	18.9 (65.1)	14.4 (30.4)	905.9 (3710)	995 (5830)	74.8 (189)	34.4 (67.7)	0.52 (1.35)		Polluted by smelter dust	Douay et al., 2008
2007										
China	6.81 (7.64)	0.14 (0.14)	18.3 (21.4)				0.10 (0.14)	Soil 1	Zhengzhou city, agricultural soils	Liu et al., 2007
	6.1 (6.1)	0.12 (0.13)	19.3 (23.9)				0.09 (0.13)	Soil 2		
	6.97 (8.74)	0.11 (0.11)	14.4 (16.5)				0.04 (0.05)	Soil 3		
	6.88 (10.9)	0.68 (1.2)	8.2 (164.9)	54.3 (109.3)	13.5 (64.4)	15.8 (33.3)			Around Guanting Reservoir	Luo et al., 2007, 2010
Germany	0.9	7	136.9	730.7	145.5	55.8	3.4	Soil 1	Surface layers at Central Elbe River	Overesch et al., 2007
	35.8	8.4	142.1	747.7	144.3	46.8	3.7	Soil 2		
	6.9	9	161.6	1214.4	184.4	72	9.5	Soil 3		
	15.1	9.2	164.6	1053.1	197.7	55.1	11.7	Soil 4		
	29.2	1.3	155.4	137.6	110.8	27.3	2.6	Soil 5		
	69.7	14.4	262	1326.2	286.3	72.7	16.7	Soil 6		
India		1.65 (3.80)	13.2 (35)	33.7 (121.8)	19 (52.6)	10.2 (34.4)		Soil 1	Contaminated soils, Varanasi	Sharma et al., 2007
		1.50 (6.95)	15.5 (38.9)	65.8 (387.8)	20.3 (203.5)	115 (32.3)		Soil 2		
		1.45 (8.85)	14.9 (44.5)	65.9 (223.6)	21.3 (107.1)	11.2 (32.2)		Soil 3		
2006										
Canada	101	6	126	28	615	435			Near Ni/Cu smelter, Sudbury, Ontario	Feisthauer et al., 2006
Slovakia	518 (1540)	0.5 (0.8)	5.3 (10.2)	27.2 (40.6)	11.7 (26.5)	11.5 (26.4)			Around coal-burning power station	Keegan et al., 2006
Korea	74		38.7	237 (322)	76.7			Tailing 1	Mine tailings and surrounding soils	Lee et al., 2006
	93.6 (131)		6.2 (11.3)	400 (400)	49.9 (50.3)			Soil 1.2	Tailing 1: Dukeum	
	32.5 (49.8)		300 (418)	100	141 (199)			Soil 1.2	Tailing 2: Dongil	
	55.5		1.2	241 (384)	84.1			Tailing 2	Tailing 3: Dongjung	
	28.1 (32.3)		8.8 (10.6)	109.5 (117)	17.2 (17.7)			Soil 2.1	Tailing 4: Myungbong	
	25.8 (26)		23.4 (27.2)	309 (399)	22 (25.2)			Soil 2.2	Tailing 5: Songchun	
	31.3 (91.2)		317 (590)	134 (231)	135 (183)			Tailing 3		
	14.3 (26.6)		92.3 (184)	140 (144)	82.7 (152)			Soil 3.1		
	30.2 (335.1)		110 (113)	66.9	43.2 (50.5)			Tailing 4		
	30.4		11.8	79.5 (89.5)	16.6			Soil 4.1		
	113,560		16.6 (21.1)	5.33	163.5			Tailing 5		
	196 (368)		19,025	239 (363)	94.7 (174)			Soil 5.1		
Sweden	3.26 (4.62)	0.20 (0.30)	25.9 (36.9)	83.9 (115)	25.4 (35.8)	21.5 (30.9)	0.51 (1.40)		Playground, Upssala	Ljung et al. (2006a)
	3.46 (15)	0.21 (0.71)	25.5 (358)	84 (149)	25.4 (110)	18.5 (39.1)	0.13 (3.66)		Soil in Upssala	Ljung et al. (2006b)

(CR = 10^{-4}).

3.2.2. Non-cancinogenic risk assessment

Fig. 2 indicates that the ingestion is by far the most important exposure pathway to contaminated soil. The HI values that take into account TEs at their maximum concentrations and all three exposures reveal that most European countries are sufficiently safe, since they have an HI < 1. Exception is the Wallonia canton, Belgium (HI = 4.15). If the target age group is children, in Europe only 7 out of 34 countries have an HI < 1, while for non-European countries, it is only 6 out of 18 countries (Fig. 3).

In non-residential areas, where human exposure is less intense due to limited amount of time of exposure compared to the residential exposure, cases with HI > 1 are five (Supplementary Fig. 1).

3.2.3. Cancer-related risk assessment

As for cancer risk, most of the European Union countries have p (CR) < 4 (or CR > 10^{-4}), indicating significant cancer-related risk (CR for residential areas, Fig. 4). This risk would be found mostly in countries with the higher permissible limit values for As (see discussion in Section 3.1). Sufficiently low CR value to indicate no significant cancer risk would be found for Sweden and Slovakia. The same was the case for countries outside the European Union. As will be discussed later, by far the highest contribution to CR was by As, with Pb having only a minor contribution.

4. Problems of regulation limits and associated health risk assessment

The As concentration in soil that generates an HQ value of 1.00 (unity) in residential use for adults is 219 mg kg^{-1}. Any value higher than this threshold indicates significant non-carcinogenic risk for human health (Rinklebe et al., 2019). However, for children in residential use, As generating HQ = 1 will be as low as 23 mg kg^{-1}; thus the As regulations are considered to be non-protective in most of the cases. Table 7 reveals that within Europe only Italy has a limit lower than that (SV = 20), while for countries outside Europe, the countries sufficiently protective for children in residential areas are the USA (SV = 0.68), Canada (SQG = 12), as well as the two studied Canada provinces of Ontario and Alberta (11 and 17, respectively).

As for the carcinogenic risk, our data suggest that the As threshold concentration for induced cancer risk of CR > 10^{-4} is 114 mg kg^{-1}. This threshold is not surpassed by any of the studied European countries, while Korea (SCCL/Z.1 = 150) and Australia (HIL = 400 mg kg^{-1}) regulation limits prove to be insufficiently protective.

The picture for soils of industrial use is none the better: the threshold that would generate an HQ = 1 is 340 mg kg^{-1}; this is surpassed by the UK (SV = 500) in Europe and Australia (HIL = 500), S. Africa (Comm/Ind = 580), and Korea (SCCL/Z.3 = 600) outside Europe. The cancer risk would be elevated if the threshold of 213 mg kg^{-1} was surpassed; this occurs, apart from UK's SV, to the three Belgium cantons (all limits at 300 mg kg^{-1}), as well as to Australia, Mexico, S. Africa and Korea. As for the agricultural use (the HQ = 1 threshold is attained with 307 mg kg^{-1} and CR = 10^{-4} with 114 mg kg^{-1}), regulations seem to be a lot more protective: only Germany (AV = 200 mg kg^{-1}) would generate a CR > 10^{-4} (i.e., 1.76×10^{-4}), and so is the case with S. Africa (PEH = 150 mg kg^{-1}, generating CR = 1.32×10^{-4}).

The fact that As is the most problematic among the regulated elements is shown in Fig. 5, where we present the percentage of contribution of HQ to the total HI among the various elements in residential areas. One can observe that As contributes by far the highest percentage of human health risk in almost all cases; the elements seems to have no contribution only in countries where that have not regulated As (also Supplementary Fig. 2 for industrial areas).

As for Pb, limits seem to be overly protective for adults (HQ = 1 in residential areas would be generated for Pb = 2564 mg kg^{-1}), while the highest among the studied limits is that of Australia (HIL = 1200 mg kg^{-1}). If child protection was taken into consideration, then the threshold for HQ = 1 would be reduced to 274 mg kg^{-1} for Pb. In that case, regulations in most of the countries would be unprotective; these would include UK (SV = 450), Germany (TV = 400), Czech Republic (AV = 300), Flanders and Brussels (700), Korea (SCCL, Zone 1 = 600), and Australia (HIL = 1200) (all values in mg kg^{-1}). For countries which do not specify soil use, the picture concerning Pb is similar: overly protective for adults, but rather unprotective for children, if soils were in residential areas. In industrial soil use, the threshold values of 4000 mg kg^{-1} (sufficient to generate HQ = 1) and 2500 (for CR = 10^{-4}) prove sufficiently protective in all countries. Similar is the picture for the agricultural use for Pb (Table 7).

As for Hg, in the residential use, children would face potential risks to their health if the level of 13 mg kg^{-1} was reached: limits higher than 13 mg kg^{-1} are observed in Flanders (IV = 15), Brussels (15), Germany (TV = 20), and Australia (HIL = 60). For industrial use of soils, the maximum Hg for the risk levels is 183 mg kg^{-1}, and this is surpassed in the case of UK (SGV = 480), France (600), and Mexico (310). In agricultural use, no risks of any kind are possible. In cases where soils with no use assignment were used as residential, the threshold of 13 mg kg^{-1} (generating HQ = 1 for children) and 117 (generating HQ = 1 of non-cancer risk for adults) would be surpassed only by Russia (PPC, clay/pH > 5.5 = 132), while the Taiwan limit of 20 mg kg^{-1} would cause risks to children only.

Concerning Cd (Table 7), Zn, Cu, and Ni (Table 8), the limits set by the countries indicate that they are overly protective, since no limit is anywhere close to the risk level of these elements in any of the studied cases of land use (Table SM-2).

In an attempt to identify how the regulation limits correspond to reported values around the globe, we overviewed in Table 9 all reported values from published papers from 2006 to the present. One can observe that rarely are the Zn, Cu, Ni, and Hg limits exceeded, except in some minor cases of mine tailing samples being examined. It must be noted that such samples do not represent soils, so human exposure does not occur with residential use or with occupational hazard. On the other hand, data seem to suggest that As, Cd and Pb values reported are often high relative to the limits. Especially As limits are very often exceeded in the reported data in all types of soil uses, i.e., agriculture, industrial and residential alike.

5. Conclusions and future directions

Our review reveals that there is diversity in regulation limits. Firstly, limits do not include the same TEs across countries. Secondly, TE limits in various countries are put in categories based on (1) land use, (2) soil properties, (3) both land use and soil properties, and (4) none of the above — in which case, either a single value or a range of values per regulated TE is drafted. Thirdly, regulation limits for any given element vary dramatically from country to country even within same soil use or same soil properties.

This constitutes a major problem: If limits are too low, this means that they are unnecessarily overprotective and result in high law-enforcement and legislation costs; if they are too high, health risks occur. We addressed this problem of diversity by calculating human health risk indices in the "what-if" scenario of having TEs in their highest allowable limit soil concentrations. This approach revealed the fact that As is grossly underestimated in the sense that it would result in HQ_i (accounting for human intake over the maximum allowable oral reference dose for that same TE) values of higher than unity (indicating significant risk), especially in residential use concerning all age groups (but mostly children); this shows the major problem with countries or organizations that fail to even regulate As (as is the case with the European Union, EU, and with European countries that complied with the EU soil Directive). On the other hand, TEs such as Cd, Cu, Ni, Pb, and Zn typically result in low HQ_i, indicating that their limit values are rather overprotective. As for Hg, its limits are problematic for children in residential areas in 4 out of the 14 studied counties, while the risks are non-significant in almost all other cases.

We also identify the lack in legislations concerning the possibility to include methods for estimating the plant available fractions of TEs; we examined the failure of legislations to recognise the possibility (and its associated risks) of adverse effects due to TE synergism in cases where many TEs are concurrently close to their regulation limits without necessarily exceeding them.

We conclude that there is a great necessity of scientific bodies, such as the International Union of Soil Science and national soil societies, as well as regulation bodies and policy makers to strive at dramatically reducing the existing variability of diverse soil legislations by drafting regulation limits of worldwide similarity, so that validity and acceptability may be enhanced. Such attempts should strategically aim at the direction of:

(1) Reducing limits of elements with underestimated risks (based on our analysis, we propose As),
(2) Increasing limits of essential elements currently unnecessarily low (here we suggest Cu, Ni, and Zn, but this could also include Cd, Hg and Pb, provided certain conditions are satisfied),
(3) Taking into consideration common suitable extraction procedure(s) which would be able to act as indicator of the potential mobilization of TEs from soil. A harmonization across continents and countries should be aimed at,
(4) Taking into consideration other environment interfaces, such as plants (i.e., TE mobility to plants, especially when considering edible species), and.
(5) Addressing scenarios of multi-element contamination cases, where cumulative TE human health risks must be considered.

Abbreviations

SV	Screening value
TV	Target value
GV	Guideline value
IV	Intervention value
SGV	Soil Guideline value
PV	Precautionary value
CV	Critical value
SQC	Soil quality criteria
ESQC	Ecotoxicology soil quality criteria
CCOL	Contamination cut-off level
IVSC	Indicative value of serious contamination
MAC	Maximum allowable concentration
VDM	Value calling for decontamination measures
AV	Action value
SSDV	Soil Statement Definition Value
ISV	Impact Statement Value
MPC	Maximum permissible concentration
PPC	Provisional Permissible concentration
Lim Agr	Limited agricultural use
Grade I	Background concentration
Grade II	Upper permissible levels for safe food, healthy plant growth, and no expected effect on water
Grade III	Applying for high adsorption capacity soils only.
SCLW	Soil contamination warning limits
SCCL	Soil contamination counterplan limits
Zone 1	fields, paddies, orchards, pastures, building lots (residential), school sites, ditches, parks, and children's playgrounds, and is subject to the strongest limits.
Zone 2	forests, salterns, building lots (non-residential), warehouses, rivers, historic ruins, pHysical sites, recreational sites, and miscellaneous sites.
Zone 3	factory sites, gas station sites, roads, parking lots, railway sites, and national defense and military facilities.
EIL	Ecological investigation levels
HIL	Health investigation levels
CUL	Clean-up levels
FD	Full-depth generic site Condition Standard in non-Potable Ground Water Conditions
SS	Stratified Site Condition Standard in a non-Potable Ground Water Conditions
SQG	Soil Quality Guidelines

List of equation parameters

Parameter	Name/descriptionUnit
$D_{S\text{-}Ing}$	Human intake via ingestion of soilmg TE kg $^{-1}$ BW day $^{-1}$
$D_{S\text{-}Derm}$	Human intake via dermal absorption of soilmg TE kg $^{-1}$ BW day $^{-1}$
$D_{S\text{-}Inh}$	Human intake via inhalation of air-borne soil particlesmg TE kg $^{-1}$ BW day $^{-1}$
C_S	total TE concentration in soilmg TE kg $^{-1}$ soil
IngR	Ingestion ratemg TE day $^{-1}$
EF	Exposure frequencyday yr $^{-1}$
ED	Exposure durationyr
BW	Body weightkg BW
AT	Averaging timeday
SA	Surface areacm ^{2}
SAF	Skin adherence factormg TE cm $^{-2}$
ABS	Dermal absorption factorUnitless
InhR	Inhalation ratem ^{3} air day $^{-1}$
PEF	Particle emission factorm ^{3} air kg $^{-1}$ soil
HQ_{Ing}	Hazard quotient referring to soil ingestionUnitless
HQ_{Derm}	Hazard quotient referring to dermal absorption of soilUnitless
HQ_{Inh}	Hazard quotient referring to inhalation of air-borne soil particlesUnitless
RfD_o	Reference dose of oral ingestion of soilmg TE kg $^{-1}$ BW day $^{-1}$
RfD_{Derm}	Reference dose of dermal absorption of soilmg TE kg $^{-1}$ BW day $^{-1}$
RfD_{Inh}	Reference dose of inhalation of air-borne soil particlesmg TE kg $^{-1}$ BW day $^{-1}$
ABS_{GI}	Gastrointestinal absorption of TEUnitless
RfC	Reference concentrationmg TE m $^{-3}$ air
HI_i	Hazard Index of TE iUnitless
HI_S	Hazard Index the Hazard Index, HI_i, of various TEsUnitless
CR_{Si}	Cancer risk of soil for TE_iUnitless
CR_{Ing}	Cancer risk related to soil ingestionUnitless
CR_{Derm}	Cancer risk related to dermal absorption of soilUnitless
CR_{Inh}	Cancer risk related to inhalation of air-borne soil particlesUnitless
OSF	Oral slope factor (i.e., related to ingestion) (mg TE kg $^{-1}$ BW day $^{-1}$) $^{-1}$
DSF	Dermal slope factor (i.e., related to dermal absorption of soil) (mg TE kg $^{-1}$ BW day $^{-1}$) $^{-1}$
ISF	Inhalation slope factor (i.e., related to inhalation of air-borne soil particles)(mg TE kg $^{-1}$ BW day $^{-1}$) $^{-1}$
UR	Unit riskμg TE m $^{-3}$ air
CR_S	Cancer risk combined for all carcinogenic TEsUnitless

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Appendix A. Supplementary data

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