Full length article

Future food contaminants: An assessment of the plant uptake of Technology-critical elements versus traditional metal contaminants

Qvarforth A[a,*], Lundgren M[a], Rodushkin I[b,c], Engström E[b,c], Paulukat C[c], Hough R.L[d], Moreno-Jiménez E[e], Beesley L[d,f], Trakal L[f], Augustsson A[a]

[a] Department of Biology and Environmental Science, Linnaeus University, Kalmar, Sweden
[b] Division of Geosciences and Environmental Engineering, Luleå University of Technology, Luleå, Sweden
[c] ALS Laboratory Group, ALS Scandinavia AB, Luleå, Sweden
[d] The James Hutton Institute, Craigiebuckler, Aberdeen, UK
[e] Univ Autonoma Madrid, Fac Sci, Dept Agr & Food Chem, Madrid, Spain; Department of Biology, Chemistry, Pharmacy, Institute of Biology, Freie Universität Berlin, Berlin, Germany; Berlin-Brandenburg Institute of Advanced Biodiversity Research, Berlin, Germany
[f] Department of Environmental Geosciences, Faculty of Environmental Sciences, Czech University of Life Sciences Prague, Czech Republic

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ABSTRACT

Technology-critical elements (TCEs) include most rare earth elements (REEs), the platinum group elements (PGEs), and Ga, Ge, In, Nb, Ta, Te, and Tl. Despite increasing recognition of their prolific release into the environment, their soil to plant transfer remains largely unknown. This paper provides an approximation of the potential for plant uptake by calculating bioconcentration factors (BCFs), defined as the concentration in edible vegetable tissues relative to that in cultivation soil. Here data were obtained from an indoor cultivation experiment growing lettuce, chard, and carrot on 22 different European urban soils. Values of BCFs were determined from concentrations of TCEs in vegetable samples after digestion with concentrated HNO_3, and from concentrations in soil determined after 1) *Aqua Regia* digestion and, 2) diluted (0.1 M) HNO_3 leaching. For comparison, BCFs were also determined for 5 traditional metal contaminants (TMCs; As, Cd, Cu, Pb, and Zn). The main conclusions of the study were that: 1) BCF values for the REEs were consistently low in the studied vegetables; 2) the BCFs for Ga and Nb were low as well; 3) the BCFs for Tl were high relative to the other measured TCEs and the traditional metal contaminants; and 4) mean BCF values for the investigated TCEs were generally highest in chard and lowest in carrot. These findings provide initial evidence that there are likely to be real and present soil–plant transfer of TCEs, especially in the case of Tl. Improvements in analytical methods and detection limits will allow this to be further investigated in a wider variety of edible plants so that a risk profile may be developed.

1. Introduction

For several years now, discussions have intensified around a group of elements collectively referred to as "Technology-critical elements" (TCEs). The "technology" aspect of the expression relates to these elements' key roles in several new and emerging technologies. For example, several TCEs are essential in sustainable technologies: e.g., in electric- and hybrid vehicles, for renewable energy production and energy-efficient lightening (Ali and Katima, 2020a/b; Folens et al., 2017; Savignan et al., 2021). They are also used in modern electronics, communications, digital equipment and in various medical applications (Ali and Katima, 2020b, Brioschi et al., 2013; Rauch and Morrison, 2008; Wahid et al., 2003). A more detailed summary on their application is provided in Table 1. The "critical" aspect of the expression refers to the elements' scarcity relative to the currently increasing demand and use. According to the European COST Action TD1407: Network on Technology-critical elements (NOTICE), 27 metall(oid) elements are classified as TCEs (Cobelo-García et al., 2015). These include: 1) most rare earth elements (REEs) i.e., Yttrium (Y) and almost all lanthanoids (cerium (Ce), dysprosium (Dy), erbium (Er), europium (Eu), gadolinium (Gd), holmium (Ho), lanthanum (La), lutetium (Lu), neodymium (Nd), praseodymium (Pr), samarium (Sm), terbium (Tb), ytterbium (Yb)); 2) the platinum group elements (PGEs) iridium (Ir), osmium (Os), palladium (Pd), platinum (Pt), rhodium (Rh) and ruthenium (Ru); and 3)

* Corresponding author.
 E-mail address: anna.qvarforth@lnu.se (A. Qvarforth).

another seven elements; gallium (Ga), germanium (Ge), indium (In), niobium (Nb), tantalum (Ta), tellurium (Te) and thallium (Tl), sometimes called less studied TCEs (LSTCEs) (Filella and Rodushkin, 2018; Jabłońska-Czapla and Grygoyć, 2021; Romero-Freire et al., 2019).

Since the late 1900's there has been a dramatic increase in the demand for TCEs (Cobelo-García et al., 2015; Graedel and Erdmann, 2012), and consequently also in global extraction rate (Cobelo-García et al., 2015; Kouhail et al., 2020). For example, the extraction of the platinum group elements has increased more than 10-fold in the past decade (Thormann et al., 2017; Zientek et al., 2017), and Brioschi et al. (2013) describe how the demand for REEs has expanded mining operations from approximately 50 kt/year in 1990 to 130 kt/year in 2010. Demands for both the PGEs and REEs are projected to continue to increase, as outlined for example in a recent review article by Hughes et al. (2021), and by figures from a leading company in the market research industry (Research and Markets, 2020). OECD has also assessed the future global demand of critical raw materials and forecasted the use of TCEs to increase from the current 79 billion tonnes/year to 167 billion tonnes/year by 2060 (OECD, 2018).

The consequence, in addition to the environmental impacts directly linked to mining (deforestation etc.), is that concentrations of TCEs will accumulate within environmental compartments – analogous to the global spread of other industrial contaminants (metals, organic chemicals, plastics) that are now ubiquitous in our environment. Indeed, increasing concentrations of several TCEs have already been reported in both urban (Chang et al., 2020a; Kouhail et al., 2020) and rural environments (Barbante et al., 2001 and 2004; Savignan et al., 2021); in soil (Kouhail et al., 2020), groundwater (Kouhail et al., 2020), surface water (Bu-Olayan and Thomas, 2020; Folens and du Laing, 2017; Hatje et al., 2014; Kouhail et al., 2020; Kulaksiz and Bau, 2013), sediments (Folens and du Laing, 2017; Savignan et al., 2021) and biota (Savignan et al., 2021; Tyler and Olsson, 2006). However, little is known about the environmental fate and toxicity of TCEs (Kouhail et al., 2020), with the vast majority of toxicologic studies investigating concentration ranges far in excess of environmental levels. Table 1 gives an overview of the toxic effects according to contemporary literature to date. In addition, previous studies on environmental TCEs have been limited due to the elements' low abundance in nature and associated challenges with their detection, measurement and chemical characterisation (Cobelo-García et al., 2015; Filella and Rodushkin, 2018; Savignan et al., 2021).

Exposure to most traditional metal contaminants occurs through food consumption, and especially via vegetables, fruits and cereals (Cao et al., 2016; EFSA, 2009a; EFSA, 2009b; EFSA, 2010; Glorennec et al., 2016; Parveen et al., 2018). Thus; it is generally motivated to prioritise investigations of different metals' uptake by food crops when seeking to understand the risks they may pose to human health. There are several generic risk assessment frameworks of high international acceptance where bioconcentration factors (BCFs), or the ratio of the concentration measured in edible plant tissue to the concentration in the cultivation soil, are used to estimate the soil to plant transfer of metals and subsequent human exposure (CCME, 2006; Lijzen et al., 2001; US EPA, 1996; Swedish EPA, 2009). Whilst a higher BCF implies a higher potential for plant uptake, risk estimates are affected by both the elemental concentration in growing media and the dose tolerated before toxicologic effects start to appear in an exposed population. Thus, even relatively toxic elements at elevated concentrations in soil can present minimal risk if uptake into edible plants is low or where consumption of the affected plants is low. A key obstacle when assessing TCE health risks is

Table 1
Technology-critical elements; areas of use and known/indicated toxicity to date.

Group	Element	Areas of use, examples		Toxicity/Indications of toxicity
REEs	Cerium Dysprosium Erbium Europium Gadolinium Holmium Lanthanum Lutetium Neodymium Praseodymium Samarium Terbium Ytterbium Yttrium	- Wind power turbines[a,b,c,d,e] - Electric vehicles[a,b,d,e] - Energy-efficient lighting[a,b,e] - Metallurgy[e] - Catalytic converters[a,e] - Medical facilities, e.g. contrast agents, lasers, cancer treatments[a,c,d,e] - Petroleum refining processes[b,c,e] - Military applications[b,c,d,e] - Computer systems[a,b,e] - Mobile phones[a,c]	- Audio equipment[b] - Flat screens[a,e] - High strength magnets[a,b] - Production of (optical) glasses[a,b,e] - Parts of super-capacitors and/or conductors[a,c] - Polishing[a,e] - Ceramics[a,e] - Batteries[a,e] - Fertilisers[b,c]	Animal studies imply that REEs can accumulate in multiple organs and body parts, such as the liver, eyes, bones, spleen, lungs, kidneys, testicles, brain, heart and adipose tissue.[f,g,h,i] Toxic effects stated so far are listed. - Damage to kidneys (Gd)[c] - Neurological impairments such as reduced IQ in children (La)[c] - Pneumoconiosis, i.e. a type of lung disease (Ce)[c] - Anti-testicular effects/male sterility[c] - Bone alteration[c] - Fibrotic tissue injury[c] - Genotoxicity[c] - Oxidative stress (Ce, Gd, La, Nd, Tb, Y, Yb; animal studies)[f,h] - Decreased bodyweight and damage to liver and brain (Ce, La; animal studies)[f,g,j]
PGEs	Iridium Osmium Palladium Platinum Rhodium Ruthenium	- Catalysts (automobile, chemistry, petroleum refining)[a,k,l,m,n,o] - Electric/electronic industry/engineering[a,k,m,p] - Jewelery[k,m] - Medical applications (e.g., cancer therapy, electron microscopy, dental applications, implants such as pacemakers)[k,n,l,m] - High wear applications; instrument pivots, electrical contacts[a] - Magnetic coatings (e.g., in hard discs)[a] - Glass and color ceramics[l,m,o]		The following toxic effects in humans are reported in the literature: - Cytotoxic and allergenic effects (for Pt in particular)[n,q,r] - Damages to lung cells[n,s] - Headaches and dizziness[s] - Toxicity to skin, blood, eyes, liver, gut and brain[n,s]

[a](Ali and Katima, 2020b), [b](Carpenter et al., 2015), [c](Gwenzi et al., 2018), [d](Klinger, 2018), [e](Zhou et al., 2017), [f](Doulgeridou et al., 2020), [g](Huang et al., 2011), [h](Kawagoe et al., 2005), [i](Ramos et al., 2016), [j](Fei et al., 2011), [k](Savignan et al., 2021), [l](Rauch, S. and Morrison, G. 2008), [m](Zientek et al., 2017), [n](Ali et al., 2019), [o](Lenntech, 2022), [p](Ermakov and Naboichenko, 2012), [q](Lustig et al., 1996), [r](Sternbeck and Östlund, 1999), [s](Švorc et al., 2012)

Table 1 (*continued*)

Group	Element	Areas of use, examples		Toxicity/Indications of toxicity
LSTCEs	Gallium	- High-performance computers[a] - Telecommunication[a] - Light-emitting diodes (LEDs) and laser diodes[a,t]	- Photodetectors[a] - Solar cells[a] - Medical equipment[a,u] and drug administration[v] - Transistors[w]	The following have been reported, mostly after animal studies, regarding toxicity: - Antimicrobial and anti-inflammatory activity[t,u] - Immunosuppressive effect[t,u] - Lung toxicity[x,u] - Toxicity to reproductive organs[x] - Kidney impairments[x]
	Germanium	- Wafers for high-efficiency solar cells[a] - Detectors (in airport security)[a] - Fiber- and infrared optics[y]	- Semiconductors[z,y] - Medical uses (cancer treatments)[z] - Catalyst[y]	Low acute toxicity[z,aa] but some observed effects in humans are listed below. - Kidney damage[z] - Neurological toxicity[z] - Pulmonary toxicity[z]
	Indium	- Film coatings on flat-panel displays[a,bb,cc] - Semi-conductor materials (InP, InN)[dd] - Alloys[bb] - Solar panels[bb,cc]	- LEDs[bb] - Laser diodes[bb] - Batteries[bb]	Toxic effects have been reported for listed internal organs in humans. - Heart[cc,ee] - Kidneys[cc,ee] - Liver[ee]
	Niobium	- High-grade structural steel (used for example in vehicle bodies)[a,ff,gg] - Superalloys for rocket nozzles, aircraft engines, terrestrial turbines, chemical industry and turbine blades[a,ff] - Touch screen/sensor technologies[a] - Magnets, used in e.g. particle accelerators, magnetic resonance tomography (MRT), nuclear spin resonance (NSR)[a,ff,gg] - Chemistry, catalyst[gg] - Optics[ff]		Indications of low toxicity[hh,ii]
	Tantalum	- Capacitors (e.g., used in automotive electronics)[jj] - Superalloys (e.g., used in jet engines, nuclear reactors, heat exchangers and in healthcare, see below)[jj] - Personal computers and cell phones, digital photo- and video cameras[a,ff,gg] - Medical applications (pacemakers, prosthetic devices, surgery)[a,ff] - Chemical industry[jj]		Indications of low toxicity[ff]
	Tellurium	- Solar cells (CdTe)[a,f] - Thermal cooling devices[a] - Electronics[a] - Additive in metallurgy[f]	- Telecommunication[f] - Magnetical discs[f] - Optics[f,kk]	Tellurium is distributed to the kidneys, liver, bone, brain and testes.[f] Following experiments on rats, two conclusions have been drawn: - Cytotoxicity (cell damage) to astrocytes (i.e., a kind of braincell).[ll] - A higher toxicity than both arsenite and selenite.[aa]
	Thallium	- Electrical- and electronic industries[f,mm] - Manufacturing of optic lenses and glass[f,mm] - Low temperature thermometers[mm] - Fireworks[mm] - Laser equipment[mm] - Medical applications (e.g., cardiac scintigraphy)[f,mm] - Alloys, that e.g. enhances resistance to corrosion[f,mm]	- Scintillation counters (instruments that detect and measure ionising radiation)[mm] - Crystals for infrared instruments[mm] - Catalyst, organic reactions[mm] - Pigments/dyes[mm] - Protection of wood and leather against fungi[mm]	Thallium can accumulate in several body compartments; in the liver, heart, muscle tissues, lungs, bones, kidneys and nervous system.[f,oo] It is highly[uu] toxic and some effects in humans are listed below. - Stomach- and intestinal ulcers[nn,oo,pp] - Nausea and vomiting[mm] - Tiredness[mm] - Neurological damage[oo,qq,rr,pp] - Blindness[ss] - Hair-loss[ss,oo,rr] - Sleep disorders[mm] - Paralysis[mm,nn,oo] - Headache[mm,pp] - Loss of body mass[oo] - Muscle pain[mm] - Internal bleeding[oo] - Heart failure[oo,ss,tt] - Death[oo,qq,rr]

[t](Chung and Lee, 2013), [u](Chitambar, 2010), [v](Kim et al., 2018), [w](Salminen et al., 2005), [x](Tanaka, 2004), [y](SGU, 2020a), [z](Zheng et al., 2020), [aa](Stjernman Forsberg and Eriksson, 2002), [bb](SGU, 2020b), [cc](Chang et al., 2020b), [dd](Veal et al., 2010), [ee](Moyer et al., 2002), [ff](Nikishina et al., 2013), [gg](Folens and du Laing, 2017), [hh](Ray et al., 2020), [ii](Schulz et al., 2017), [jj](SGU, 2020c), [kk](Kominkova et al., 2017), [ll](Roy and Hardej, 2011), [mm](Kazantzis, 2000), [nn](Duri et al., 2020), [oo](Karbowska, 2016), [pp](Wang et al., 2013), [qq](Liu et al., 2017), [rr](Pavlíčková et al., 2006), [ss](Jiang et al., 2020), [tt](Heim et al., 2002), [uu](Xiao et al., 2004)

that their tolerable intake levels have not yet been established and adopted into standard risk assessment frameworks, meaning that health risk implications cannot be assessed in a regulatory context. However, while the TCE uptake by edible crops is difficult to translate into risks for human consumers, this paper undertakes the initial steps by discussing calculated BCFs of individual TCEs from a risk perspective, i.e., in parallel to what is known about their occurrence in the environment and their toxicity. Urban soils, due to their proximity to multiple sources of contaminants via contrasting pathways (airborne, highway runoff, waste disposal etc.) are a logical media to establish a "baseline" for the TCEs' accessibility for plant uptake. In addition, whilst a relatively small fraction of global food production is currently derived from urban land, vegetables from such environments often contain elevated concentrations of 'traditional' metals currently acknowledged as ubiquitous urban pollutants, e.g. As, Cd, Cu, Pb and Zn (Bretzel and Calderisi, 2006; Clarke et al., 2015; Mitchell et al., 2014; Szolnoki et al., 2013), and need monitoring (Meharg, 2016). Based on current trends, a similar outcome is expected with TCEs in the future.

In this study, cultivation of two leafy vegetables (lettuce and chard) and one root vegetable (carrot) was conducted in 22 urban soils from 6 major cities across Europe. The vegetable types were selected due to a high consumption worldwide. The aims of this study were to; 1) conduct a controlled growth experiment to determine concentrations of TCEs in paired soil and vegetable samples; and 2) undertake an assessment of TCEs potential risk for entering food chains by determining bioconcentration factors (BCFs) for the TCEs, and comparing these to the BCFs of five traditional metal contaminants (TMCs; As, Cd, Cu, Pb and Zn) and two major soil elements, of which one (K) is also a major plant nutrient; thus, actively and easily taken up by plant roots, and the other one (Al) constituting an element with a near-negligible root uptake (Engström et al., 2008; McBride et al., 2013).

2. Method

2.1. Cultivation experiment

To obtain paired data for concentrations of TCEs in soils and plants, a cultivation experiment with selected urban soils was devised. An important motive for using authentic urban soils, rather than spiked soils, is that the elements can be studied in their natural form. Soils were collected primarily from active allotment gardens across Europe, resulting in soils from 22 different areas in 6 cities: Copenhagen, Denmark (N = 7); Widnes, UK (N = 5); Malmö, Sweden (N = 4); Madrid, Spain (N = 3); Pribram, Czech Republic (N = 2); and Berlin, Germany (N = 1). Initial analyses of the soils' basic geochemistry revealed pH values from 5.02 to 7.96, cation exchange capacities between 8.02 and 24.1 cmol+/kg, and clay contents from 0.01 to 2.0 %. The soils' content of organic matter, assessed by loss on ignition, ranged from 4.8 to 14 %.

From each of the 22 sites, a soil composite sample was collected by pooling soil material from different gardeners' plots. After thorough mixing and sieving to < 2 mm, triplicate 1L pots for each soil + crop combination were prepared. Three vegetables were grown: lettuce (*Lactuca sativa*), chard (*Beta vulgaris*) and carrot (*Daucus carota sativus*). The total number of pots was thus 22*3*3 = 198. The cultivation was carried out in an indoor constant room at Linnaeus University in Kalmar. Thereby the deposition of airborne dust particles onto above ground plant tissues was minimised. The temperature was set to 20 °C and the relative humidity to 67 %. Samples were illuminated for 10 h per day with warm fluorescent light (photon flux density of approximately 100 μmol m^{-2} s^{-1}). Watering was made with distilled water every second day and plants were harvested after having reached edible size. Lettuce and chard were cut at about 2 cm above the soil surface. Since not all pots produced enough material for analyses, the cultivation in the end generated 62 samples of lettuce, 62 samples of chard and 51 samples of carrots (N$_{tot}$ = 175). These were prepared and sent for analyses together with paired soil samples.

2.2. Sample preparation

Harvested vegetable samples were carefully rinsed in distilled water and blotted dry with tissue paper prior to drying at 60 °C. After homogenisation, approximately 0.50 g was transferred to 50 mL polypropylene tubes and transported to the accredited laboratory ALS Scandinavia AB in Luleå for digestion/leaching and determination of elemental concentrations. The vegetables were accompanied by paired soil samples, each consisting of approximately 0.50 g of sieved (to < 2 mm) and dried (at 60 °C) material. Every tenth sample of both vegetables and soils were prepared and sent in duplicates.

To minimise contamination of samples by particulate matter from the laboratory environment, all sample preparation and analyses were performed in Class 10 000 Clean Room facilities with HEPA-filtered air by personnel wearing clean room attire. General precautions detailed by Rodushkin et al. (2010) were taken to minimise handling contamination. All utensils had been acid washed, i.e., soaked in 0.7 M HNO$_3$ for 24 h followed by rinsing with high-purity de-ionised Milli-Q water (Millipore, Bedford, MA, USA).

Vegetables were digested using concentrated HNO$_3$ of Suprapur grade, which is a treatment that provides quantitative oxidation of the biologic material but do not provide decomposition of resistant minerogenic fractions potentially attached to the plant surfaces despite rinsing (Rodushkin et al., 1999). To each sample, 10.0 mL of concentrated Suprapur grade HNO$_3$ was added. Samples were left overnight in a fume hood at 20 °C, and thereafter placed on heating blocks (120 °C) for 1 h. The extracts were diluted with Milli-Q water to 20 mL before ICP analyses.

Soil samples were digested to obtain both (pseudo)total concentrations and an indication of the more bioavailable, or geochemically active, concentrations. The (pseudo)total concentrations in soil were determined after *Aqua Regia* (AR) digestion, where approximately 0.50 g of soil was mixed with 7.5 mL HCl and 2.5 mL HNO$_3$, both concentrated and of Suprapur grade. The samples were heated on heating blocks (130 °C) for 2 h, followed by dilution to 50 mL using Milli-Q water. As a proxy for the more directly plant-available concentrations, another 0.50 g aliquot of the soil was leached with 5.0 mL of 0.1 M HNO$_3$ at room temperature. The mixtures were shaken for 16 h on a laboratory shaker and then centrifugated (for 2 min at 4000 rpm) to separate the aqueous phase, which was then diluted with Milli-Q water to 20 mL.

Along with the vegetable and soil samples, each preparation batch also included a set of preparation blanks. Both blanks and calibration standards were prepared in dedicated fume hoods in facilitates that are separated from those where sample preparation is carried out; thereby minimising the risk of contamination. Each preparation batch also included matrix-matched quality control (QC) samples. The one used in the vegetable analyses was an internal quality control sample for plant matrices (IQCSP), consisting of Spirulina powder. Concentrations in this sample have been established through cross-calibration against the certified reference material SRM1547 (peach leaves) from the National Institute of Standards and Technology (NIST, Gaithersburg, MD, USA). Compared to the SRM1547, the IQCSP material contains detectable concentrations of most TCEs. The reference material used in the soil analyses was an internal quality control sample for soil matrices (IQCSS), based on well-homogenised local topsoil. Concentrations in this sample have been established through cross-calibration against the certified reference material GBW 07410 (soil) from the National Research Centre of Geoanalysis (Beijing, China).

2.3. Elemental analyses

All samples were analysed for the elements that BCFs were to be calculated for: 26 TCEs (Ce, Dy, Er, Eu, Ga, Gd, Ge, Ho, Ir, La, Lu, Nb, Nd, Os, Pd, Pr, Pt, Rh, Ru, Sm, Ta, Tb, Te, Tl, Y, Yb), five traditional metal contaminants, TMCs (As, Cd, Cu, Pb, Zn) and two major soil elements (Al and K). Concentrations of indium (In), one of the less-studied TCEs, were

not evaluated since the applied multi-element screening protocol uses In as an internal standard.

As outlined by Rodushkin et al. (2018), the accurate analyses of TCEs and other elements found at ultra-trace levels in environmental samples present significant analytical challenges. Simultaneous determination of multiple elements comes at a price of elevated method limits of detection (LOD) compared to the lower LODs that can be achieved for analytical protocols optimised for a limited group of analytes or a single element. In this study, analyses were performed by double-focusing, sector field inductively coupled plasma mass-spectrometry (ICP-SFMS, ELEMENT XR, Thermo Scientific, Bremen, Germany) equipped with a solution nebulisation sample introduction system. During operation, methane was added to the sample gas to improve sensitivity and reduce spectral interferences, as described by Rodushkin et al., (2005). Matrix effect correction was accomplished by internal standardisation, where indium (In) was added to all measurement solutions at 2.5 µg/l concentration. Quantification was done by external calibration with concentration- and matrix-matched standards. Further details on the multi-element ICP-SFMS analysis can be found in e.g., Engström et al., (2004). Obtained LODs were in the range 0.016–27 µg/kg for the REEs and 0.013–29 µg/kg for the PGEs. The LODs for all separate elements and procedures are listed in the Supplementary Tables (**S1–S5**).

2.4. Quality control and quality assurance (QC/QA)

Throughout the analyses, standard procedures for analytical quality control were applied. The precision and accuracy of the measurements were monitored using reference materials and duplicate samples included in each sample set. The limit of detection (LOD) and limit of quantification (LOQ) for individual elements were calculated as three and ten times the standard deviation of concentrations measured in preparation blanks, respectively. Despite high instrumental sensitivity of the applied analytical technique, 10 out of the 26 analysed TCEs (Eu, Ge, Ir, Os, Pd, Pt, Rh, Ru, Ta and Te) were found below corresponding LOD in most of the vegetable samples (72 %), and in the vast majority (92 %) below LOQ. For the remaining elements, both the remaining 16 TCEs, all TMCs and the major elements, all samples were above the LOD and in more than 95 % of the measurements also above the LOQ. Only these elements, for which the obtained concentrations were assessed to be of high enough accuracy, were included in the calculation and analyses of BCFs.

The reproducibility of the method was assessed based on the results obtained for the duplicate samples. For the soil sample duplicates, average relative standard deviations (RSDs) were below 10 % for most of the elements that remained after the exclusions mentioned above. The exceptions were Cu and La (RSD 11 %), Na (RSD 23 %) and Nb (RSD 37 %). The RSDs were higher for the vegetable samples, which can be explained by the fact that the vegetable concentrations were generally closer to LOD and LOQ than for the soil samples. Moreover, obtaining a representative 0.5 g sub-sample without excessive sample manipulation (and thus without increasing contamination risk) is more difficult for plants than for soils. The vegetable sample RSDs ranged from 5 to 54 %, with a median value of 21 %.

The analytical accuracy was assessed using reference materials. For the soil reference material, target concentrations were available for AR based digestion. Average recoveries were in the range of 96–118 % for all studied analytes. Target concentrations for the vegetable reference material were based on a combination of nitric acid and traces of hydrofluoric acid during the decomposition. These values will therefore be significantly higher for more resistant elements (that can be attached or incorporated in the structure of the plant surfaces) than corresponding values achieved using only nitric acid. With that in mind, the average recovery rates for the 16 remaining TCEs varied from 45 to 100 %, even though recoveries for the majority of elements were found in the 80–100 % range. For the TMCs and major elements, average recovery rates were between 92 and 110 %.

2.5. Calculation of bioconcentration factors (BCFs)

For the elements included in the calculation and analyses of BCFs (see the section about QA/QC), the BCFs for each paired vegetable/soil sample were calculated by dividing the concentration in the vegetable sample (C_v, µg/kg dw) by the corresponding concentration in soil (C_s, µg/kg dw), according to Eq. (1).

$$BCF = \frac{Cv}{Cs} \tag{1}$$

Whilst most BCFs presented in the scientific literature are expressed against the total (or near-total) concentrations in the soil, there are also numerous examples where weaker soil extractions have been used. To provide some information about the significance of the procedure for soil extraction, BCFs in this study were calculated both against soil concentrations after the near-total extraction with AR and after the milder leaching with 0.1 M HNO_3.

2.6. Exposure assessment

While no health-based tolerable daily intake values have been established for any of the TCEs yet, preliminary TDIs have been proposed for the REEs (Rodríguez-Hernández et al., 2019) and for Tl (RIVM, 1998). For these we therefore undertook a simple exposure assessment using Eq. (2) and adopting lettuce consumption by an adult women as the exposure pathway. We choose women since they have a lower average body weight than men (meaning higher exposure per kg) and compared to children (who have an even lower body mass) generally consume more vegetables per kg bodyweight. Eq. (2) calculates the daily exposure via vegetable consumption (EXP in µg/kg/day) from the ΣREE and Tl concentrations in the cultivation soil (Cs), respectively, the elements' bioconcentration factors (BCF), the vegetable dry matter content (TS), daily vegetable consumption (Rig) and the fraction of that consumption which relates to produce from the cultivation soil of interest (fh). BW is the bodyweight.

$$EXP = \frac{Cs \ x \ BCF \ x \ TS \ x \ Rig \ x \ fh}{BW} \tag{2}$$

According to WHO recommendations, the daily vegetable and fruit consumption should reach 400 g. We therefore assumed, very conservatively, that all of these 400 g consist of lettuce that is cultivated on urban soils (fh = 1). Further, we assumed a low bodyweight of the consumer, in this case corresponding to the 5th percentile for adult women (52 kg) as stated by Filipsson et al. (2011), and a lettuce dry matter content of 13.5 % (Augustsson et al., 2015).

3. Results and discussion

3.1. Element concentrations in soil and vegetables

Measured concentrations of the analysed elements, on a dry weight basis for both soil and vegetables, are provided in Table 2, with concentration data specified for each individual soil in **Tables S1 and S2** of the supplementary material, and more detailed concentrations in the examined vegetables in **Tables S3–S5**. The traditional metal contaminants (TMCs) found in the highest concentrations in soil were Zn and Pb, with mean and median AR-extractable concentrations of several hundred mg/kg. None of the TCEs were detected in those concentration ranges. However, several TCEs were present at concentrations comparable to that of Cd, which was the TMC with the lowest concentration in soil; on average about 1 mg/kg. Measured concentrations of REEs were of a similar order of magnitude, as were the two LSTCEs Ga and Nb. The remaining LSTCEs (Ge, Ta, Te, Tl) were less abundant, while the PGEs – when detected at all – were present only at a few µg/kg.

In terms of contamination by TMCs, the 22 soils used in the cultivation experiment displayed significant variability. Using Cd, As and Pb

Table 2

Concentrations of Technology-critical elements, traditional metal contaminants, K and Al in the 22 European cultivation soils and in lettuce, chard and carrot grown thereon. The number of values in soil, lettuce, chard and carrot are 22, 62, 62 and 51, respectively, unless stated otherwise in superscript square brackets. All concentrations refer to dry samples.

	Concentration in soil after AR (0.1 M HNO₃) digestion/leaching (µg/kg)				Concentration in lettuce (µg/kg)				Concentration in chard (µg/kg)				Concentration in carrot (µg/kg)			
	min	mean	median	max	min	mean	median	Max	min	mean	median	max	min	mean	median	max
Light REEs																
Ce	15 000 (110)	30 000 (2 400)	29 000 (1 900)	57 000 (7 400)	25	85	69	360	21	110	57	530	4.9	27	17	160
Eu*	130 (8.8)	450 (61)	440 (47)	850 (170)	1.1[33]	2.9[33]	2.1[33]	8.4[33]	1.2[54]	4[54]	3.3[54]	16[54]	1.1[27]	1.5[27]	1.5[27]	2.2[27]
Gd*	740 (13)	2 100 (230)	2 200 (180)	3 400 (660)	1.4	6.3	5.1	30	1.2	7.9	4.8	38	0.57	2.3	1.6	6.6
La	7 400 (72)	15 000 (1 500)	14 000 (1 200)	26 000 (3 600)	13	43	32	160	14	65	32	250	2.4	13	8.9	37
Nd	5 700 (65)	14 000 (1 100)	13 000 (850)	24 000 (3 100)	11	41	31	170	10	49	26	220	2.8	12	8.6	39
Pr	1 500 (16)	3 500 (270)	3 300 (230)	6 100 (810)	2.5	9.6	7.6	42	2.5	12	6.6	54	0.65	3.0	2.0	9.1
Sm	1 100 (14)	2 700 (210)	2 500 (160)	4 600 (630)	1.7	8.3	6.8	42	1.6	10	6.1	45	0.84	2.8	2.0	7.7
Heavy REEs																
Dy	540 (11)	1 600 (210)	1 600 (170)	2 500 (590)	1.0	5.4	4.2	27	1.4	6.2	3.4	32	0.45	1.8	1.1	5.8
Er	240 (6.3)	750 (110)	800 (90)	1 200 (290)	0.51	2.6	2.0	13	0.50	3.2	1.8	16	0.28	1.0	0.72	3.0
Ho	88 (2.2)	280 (40)	290 (33)	440 (110)	0.21	0.92	0.72	4.3	0.18	1.2	0.70	5.9	0.10	0.36	0.26	1.1
Lu	44 (0.68)	100 (11)	110 (8.9)	160 (29)	0.056[61]	0.30[61]	0.20[61]	1.4[61]	0.23	0.58	0.47	2.5	0.16	0.30	0.28	0.56
Tb	96 (1.8)	290 (34)	300 (27)	460 (99)	0.16	0.90	0.70	5.0	0.39	1.4	0.93	6.4	0.27	0.57	0.47	1.3
Y	2 400 (80)	7 900 (1 600)	8 400 (1 500)	13 000 (3 600)	7.6	36	30	130	13	44	23	180	3.4	11	7.1	32
Yb	230 (4.8)	670 (79)	720 (65)	1 100 (220)	0.43	2.1	1.7	11	0.57	2.6	1.6	13	0.21	0.86	0.64	2.6
LSTCEs																
Ga	1 900 (1.1)	3 700 (63)	3 700 (37)	7 100 (260)	2.5	12	7.5	50	3.1	12	8.5	62	1.8[43]	5.8[43]	4.4[43]	14[43]
Ge	35[18] (0.61[21])	230[18] (22[21])	90[18] (5.7[21])	1 200[18] (170[21])	5.3[51]	14[51]	11[51]	47[51]	5.2[38]	11[38]	7.8[38]	79[38]	5.3[13]	12[13]	11[13]	19[13]
Nb	530 (0.57)	1 300 (3.0)	1 300 (1.9)	2 500 (26)	0.78	5.1	4.4	23	1.3	6.5	4.3	47	0.41	2.3	1.4	8.9
Ta	3.3 (0.046)	6.1 (0.39)	5.0 (0.38)	19 (0.93)	0.20[52]	0.41[52]	0.34[52]	1.2[52]	0.60[54]	0.60[54]	0.35[54]	3.4[54]	0.18[22]	0.53[22]	0.30[22]	1.7[22]
Te	7.6[20] (0.38)	27[20] (6.0)	20[20] (4.0)	86[20] (32)	1.6[5]	1.9[5]	1.7[5]	3.2[5]	1.6[7]	2.3[7]	2.3[7]	2.9[7]	1.5[1]	1.5[1]	1.5[1]	1.5[1]
Tl	55 (0.65)	190 (5.5)	200 (1.7)	320 (23)	0.50	8.6	3.3	40	0.72	79	25	800	0.59	16	5.2	73
PGEs																
Ir	nd (0.040)	nd (0.098)	nd (0.081)	nd (0.25)	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Os	nd (0.057)	nd (0.11)	nd (0.11)	nd (0.18)	0.16[5]	0.19[5]	0.17[5]	0.21[5]	0.16[5]	0.19[5]	0.17[5]	0.27[5]	0.16[11]	0.22[11]	0.20[11]	0.31[11]
Pd	29[17] (0.37)	58[17] (4.6)	60[17] (3.1)	97[17] (25)	6.3[16]	37[16]	14[16]	150[16]	5.3[28]	26[28]	11[28]	120[28]	6.8[3]	7.8[3]	7.8[3]	9.0[3]
Pt	3.6[11] (nd)	5.5[11] (nd)	4.6[11] (nd)	8.7[11] (nd)	0.49[9]	0.56[9]	0.55[9]	0.68[9]	0.73[18]	0.82[18]	0.79[18]	1.1[18]	0.54[1]	0.54[1]	0.54[1]	0.54[1]
Rh	nd (1.1)	nd (3.7)	nd (2.5)	nd (19)	0.81[32]	1.4[32]	1.3[32]	2.3[32]	nd	nd	nd	nd	0.79[1]	0.79[1]	0.79[1]	0.79[1]
Ru	8.8[1] (0.11)	8.8[1] (0.33)	8.8[1] (0.28)	8.8[1] (0.68)	4.6[1]	4.6[1]	4.6[1]	4.6[1]	nd	nd	nd	nd	nd	nd	nd	nd
TMCs																
As	3 900 (130)	21 000 (2 100)	11 000 (910)	130 000 (18 000)	22	390	210	4 100	40	260	110	5 100	15	79	75	180
Cd	92 (21)	1 300 (1 000)	710 (620)	7 400 (5 700)	150	2 900	570	39 000	49	6 200	860	130 000	12	270	130	1 800
Cu	16 000 (130)	140 000 (28 000)	46 000 (11 000)	690 000 (100 000)	5 400	12 000	10 000	28 000	6 200	19 000	17 000	43 000	2 800	4 600	3 900	12 000
Pb	21 000 (16)	330 000 (38 000)	110 000 (23 000)	1 900 000 (210 000)	200	1 700	520	18 000	280	5 600	980	120 000	49	640	220	3 100
Zn	61 000 (3 600)	420 000 (200 000)	190 000 (87 000)	2 000 000 (970 000)	25 000	120 000	62 000	340 000	22 000	390 000	130 000	2 100 000	11 000	39 000	23 000	330 000
Major soil elements																
Al	5 600 000 (30 000)	10 000 000 (760 000)	9 000 000 (760 000)	21 000 000 (1 800 000)	4 400	28 000	21 000	120 000	9 600	37 000	25 000	190 000	3 200	13 000	10 000	45 000
K	390 000 (40 000)	2 400 000 (320 000)	2 200 000 (220 000)	5 800 000 (1 100 000)	42 000 000	92 000 000	98 000 000	120 000 000	35 000 000	88 000 000	91 000 000	130 000 000	5 800 000	21 000 000	21 000 000	37 000 000

*Sometimes classified as heavy REEs (HREEs) (Ramos et al., 2016; Zhou et al., 2016). They can also be classified into light (Ce, La, Nd, Pr), medium (Eu, Gd, Sm) and heavy (Dy, Er, Ho, Lu, Tb, Tm, Y, Yb) REEs (Doulgeridou et al., 2020).

as examples, concentrations measured in soils after AR digestion were in the range 0.092–7.4 mg/kg (Cd), 3.9–130 mg/kg (As) and 21–1900 mg/kg (Pb), with median concentrations of 0.71 mg/kg, 11 mg/kg and 110 mg/kg, respectively. As a comparative example, the Swedish EPA's guidance values for residential areas are 0.5 mg/kg for Cd, 10 mg/kg for As and 50 mg/kg for Pb. The assessment of urban soil relative to background concentrations of TCEs is not feasible at present because data are not available for the TCEs. Therefore, as an alternative, the concentrations of the different elements in each urban study soil was assessed relative to the average concentrations measured in commercially-produced plant soils (N = 6) that had been purchased from a major plant store chain. Such commercially available plant growth media provides the closest possible analogue, at present, to urban soils since it is used in place of/as well as natural soils in urban cultivation.

Mean concentrations in the commercial plant soils were 0.36 (As), 0.081 (Cd), 5.8 (Cu), 2.6 (Pb), 31 (Zn), 44 (∑REEs), 0.55 (Ga), 0.60 (Nb), and 0.031 (Tl) mg/kg, and the resulting elevation values in each urban soil relative to the commercial soils are found in Table 3. The average TMC concentrations are clearly elevated in the urban soils compared to the commercial plant soils; between 7.0 and 250 times. Also the TCE concentrations are somewhat higher in the urban soils, although generally to a much lower degree; 13 times higher at the most.

3.2. Bioconcentration factors (BCFs)

Fig. 1 contains boxplots of the elements more thoroughly assessed and shows how the two major soil constituents included in the analyses, Al and K, provide a distinct framing for the rest of the dataset. The lowest BCF values apply to Al and the highest to K, where the mean AR-based BCFs of these two elements differ approximately 12,000 times. Aluminium - although the most abundant metal in geogenic material - is a good example of an element of known toxicity (Crisponi et al., 2011;

Table 3

Concentration elevations in the 22 urban cultivation soils, i.e., the concentrations in the urban soils relative to concentrations in commercial plant soil, given for the sum of the traditional metal contaminants As, Cd, Cu, Pb, and Zn (∑TMC), the sum of the rare earth elements (∑REE) and Ga, Nb and Tl. Sites 1–4 are from Sweden (Malmö); 5–11 from Denmark (Copenhagen); 12–16 from the UK (Widnes); 17–19 from Spain (Madrid); 20–21 from the Czech Republic (Pribram) and 22 from Germany (Berlin).

	Malmö				Copenhagen							Widnes					Madrid			Pribram		Berlin
Soil no.	1.	2.	3.	4.	5.	6.	7.	8.	9.	10.	11.	12.	13.	14.	15.	16.	17.	18.	19.	20.	21.	22.
∑TMC	7.0	12	8.2	12	10	170	110	14	7.2	130	56	44	33	18	66	29	9.3	13	6.3	28	250	26
∑REE	1.7	2.0	1.9	1.4	1.6	2.4	1.8	1.6	1.4	2.0	1.9	1.3	1.6	0.94	1.1	1.3	2.9	1.7	2.0	3.1	2.5	0.83
Ga	4.9	4.9	4.9	4.4	3.6	13	7.6	4.3	4.3	9.0	7.2	6.7	7.9	4.6	6.8	6.6	9.2	8.7	8.5	11	7.6	3.5
Nb	2.1	2.1	2.1	1.8	2.4	2.9	2.2	2.1	1.8	2.4	2.8	1.4	1.7	0.90	1.4	1.2	3.0	3.8	3.2	2.6	1.7	4.2
Tl	4.6	4.8	5.2	4.0	3.2	9.0	10	4.5	3.6	8.9	7.7	8.7	7.7	4.5	8.5	5.6	6.6	6.4	7.0	6.7	9.1	1.8

EFSA, 2008) where the uptake by plants is very low; thus, generally presents an insignificant health risk following consumption of vegetables. Potassium on the other hand, as a major nutrient, is efficiently taken up and translocated in plants, often using specific transport systems (Nieves-Cordones et al., 2019; Xu et al., 2020). The BCFs for Pb and As are relatively low, which is shown in Fig. 1 as well as in numerous previous publications (Bissen and Frimmel, 2003; de Temmerman et al., 2015; McBride et al., 2013; RIVM, 2007; Sipter et al., 2008). Despite this, a number of exposure assessments have concluded that vegetable consumption alone may render an intake above health-based tolerable intakes for both Pb and As (EFSA, 2010; Uddh-Söderberg et al., 2015). The TMCs Zn and Cd, on the other hand, are well known to be significantly more soluble and plant available (Salminen et al., 2005), and as expected they plot further to the right in Fig. 1. It should be emphasised that contaminant solubility is not the only factor in plant uptake. So are a number of other parameters, for example the presence or absence of active uptake mechanisms and plant physiology (Krämer, 2010; Rascio and Navari-Izzo, 2011; Trakal et al., 2015). The latter should explain why the BCFs were generally higher in the two leafy vegetables compared to the carrots (Fig. 1), which reaffirms previously published results from e.g. Singh et al., 2012. **Figure S1** in the supplementary material shows the intra-soil BCF variability for all elements, based on the triplicate samples that were grown in each soil, and **Table S6** lists the BCFs for all the elements analysed in this study; even the 10 that are not further evaluated due to the high number of samples with concentrations below LOD (Eu, Ge, Ir, Os, Pd, Pt, Rh, Ru, Ta and Te).

3.2.1. Impact of chosen soil extraction on BCFs

Fig. 1 and **Table S6** show that the soil digestion procedure is critical for the calculated bioconcentration factor. While there are indeed a wide range of digestion protocols, with varying capacities of dissolving various mineral phases, the use of AR is a commonly used and widely accepted method (e.g., ISO standard 11466) for the extraction of (pseudo)total *recoverable* metals in soils. It is thus among the most applied methods for assessing the maximum available concentration for plant uptake (Chen and Ma, 2001; Duri et al., 2020; Pavlíčková et al., 2003; Vercoutere et al., 1995). The fraction of elements that resists this digestion are mainly associated with highly stable silicate minerals of negligible solubility; thus, they can be excluded as potentially available for plant uptake (Niskavaara et al., 1997). Characterisation of concentrations more directly available for plant uptake is both uncertain and approximative whatever extraction protocol is applied, with solubilisation rates differing significantly between elements as well as soils (Groenenberg et al., 2017; Kashem et al., 2007; Menzies et al., 2007). Dilute acids, such as 0.1 M HNO_3 or 0.1 M HCl are, however, among the more commonly applied leaching media (Li and Zhang, 2013; Rodrigues et al., 2010; Sutherland, 2002). They have previously been described to provide a fair approximation of the plant available concentrations of traditional metal contaminants such as Cd, Cu, Pb and Zn (Chowdhury et al., 2010; Kashem et al., 2007; Sutherland, 2002), but their suitability for assessing the plant available pool of TCEs is less well known.

Mean BCF values may be between 1.3 (as found for Cd in chard) and 19,000 (Pb in chard) times higher when a weak leaching agent (such as 0.1 M HNO_3) is used to bring the solid phase metals into solution before analyses, as compared to a near-total (AR) digestion. The more resistant an element is to dissolution in the soil matrix, which is the case e.g., for Pb, the more the calculated BCF value is affected. It is therefore important to take methodological differences into account when comparing or using BCF data from other studies, where the majority adopt strong soil digestion techniques which render total or near-total concentrations of most elements (Jiang et al., 2020; LaCoste et al., 2001). There are, however, also examples where weak extractions have been used, motivated by an intention to target concentrations that better reflect the plant-available fraction (Duri et al., 2020). The methodological descriptions of soil (and plant) digestion procedures are quite frequently ambiguous, which makes interpretation difficult and indicates that there is a lack of understanding of the importance of the chosen extraction method. Which approach is most suitable can be
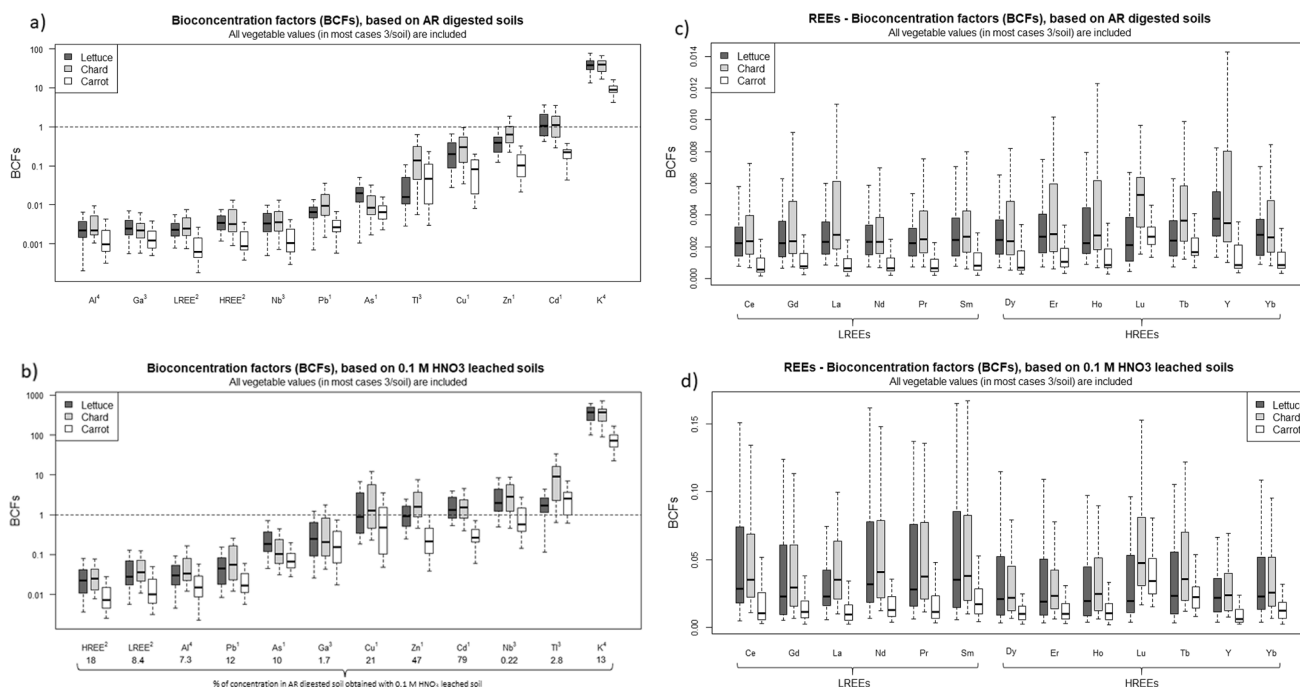

Fig. 1. The subfigures a and b show boxplots with bioconcentration factors for 1) traditional metal contaminants (TMCs), 2) REEs – visualised together as heavy REEs (HREEs) and light REEs (LREEs), 3) LSTCEs (Ga, Nb, Tl) and 4) elements with a well-known high (K) and low (Al) uptake in lettuce, chard and carrot. BCFs are based on soils digested/leached with AR (subfigure a) and 0.1 M HNO_3 (subfigure b). Subfigures c and d show BCFs for the REEs (except for Eu) specifically, based on soils digested/leached with AR (subfigure c) and 0.1 M HNO_3 (subfigure d). Note that the scales are logarithmic in subfigures a–b, while non-logarithmic in c–d. The upper and lower horisontal lines mark the maximum and minimum value of the dataset, respectively. The line within the box shows the median value, the lower edge of the box the 25th percentile and the upper edge the 75th percentile. Outlier values are not shown in these figures, but can be seen in Table S6.

debated, but if one aims to describe *the concentration in plants as a function of the concentration in the soil,* then the most robust measure of the concentration of a certain element in soil is obtained after an analysis of it's total concentration - or the maximum concentration of the element considered available for plant uptake. The most common and practically feasible method of obtaining this concentration is through analyses after digestion with AR. This analysis will render similar concentrations for all soils with the same (pseudo)total concentrations despite geochemical differences, hereby facilitating the comparison of results (e.g., BCF calculations) between studies. When weaker soil extractions are selected, a number of geochemical factors affect how much of the soil's total supply that is dissolved (e.g. soil mineralogy, pH, content of organic matter, clays and Al/Fe/Mn (hydr)oxides etc; Kabata-Pendias and Pendias, 2001). Since such factors are critical for the solubility and phytoavailability of most elements, they are certainly important for the concentrations finally found in plants. However, they add a complexity to the picture that makes the straightforward calculation of BCFs difficult – especially when it comes to comparing and interpreting data from different sites (=different studies). At present, methodological inconsistencies with regards to soil extraction protocols constitute a considerable source of uncertainty within final risk estimates.

3.2.2. Rare earth elements (REEs)

Consistently low BCFs imply a low vegetable uptake of the REEs relative to the other investigated elements (Fig. 1 **a–b** and **Table S6**). Fig. 1 **c–d** further indicate only small variations between the different REEs; not unexpected considering their chemical similarity (Hatje et al., 2014; Ramos et al., 2016; Tyler, 2004a). It thus seems reasonable, at least for this study, to draw generic conclusions on uptake of REEs by plants without necessarily analysing the full range of REEs and soil:plant combinations. Despite the REEs overall analogous geochemical behavior, however, it has previously been suggested that the lighter REEs are slightly more easily taken up by plants than the heavier ones, as a consequence of their lower atomic mass (Tyler, 2004a) or possibly a more efficient passive diffusion (Trakal et al., 2015). The light REEs are also considered more soluble (Ramos et al., 2016; RIVM., 2000), which should further increase their mobility and plant uptake relative to the heavy REEs (Brioschi et al., 2013; Kovaříková et al., 2019). More efficient uptake of the lighter REEs was, however, not confirmed by our data. The BCFs after AR extraction decreased in the order Y > Er > Dy > Ho > Yb > Sm > La > **Tb** > Nd > Ce > Gd > **Lu** > Pr in lettuce, HREEs in bold. In chard and carrot, the order was Lu > Y > Tb > Er > La > Ho > Yb > Sm > Dy > Gd > Pr > Ce > Nd and Lu > Tb > Y > Er > Ho > Yb > Gd > Sm > Dy > Ce > La > Nd > Pr, respectively. And most importantly the observed differences were very small (Fig. 1 **c–d**), with almost similar mean BCFs found for light (0.0025) and heavy (0.0031) REEs when all vegetables were considered together.

The BCFs were similar for the two leafy vegetables, and it was higher than for the studied root vegetable (Fig. 1). For the two leafy vegetables, mean BCFs ranged from 0.0028 (Pr) to 0.0055 (Lu) when looking at the values determined after AR digestion of the soil, and from 0.036 (Y) to 0.089 (Sm) when based on soil concentrations determined following extraction with the weaker 0.1 M HNO₃ (**Table S6**). For carrots, the mean BCFs ranged from 0.00095 (Pr) to 0.0029 (Lu) when concentrations in soils were analysed after AR digestion and from 0.011 (Y) to 0.055 (Lu) after leaching with 0.1 M HNO₃. Previous studies that have set out to compare accumulation of REEs in underground parts of plants and in aboveground tissues have found a higher uptake in underground parts (Carpenter et al., 2015; Hu et al., 2002; Khan et al., 2017b; Tyler, 2004a, Wen et al., 2001), which according to Ramos et al. (2016) could be due to an apoplastic barrier in the roots that blocks translocation into the rest of the plant. Whilst consistent, the results from these studies/ reviews, however, should be used carefully if one wants to draw conclusions about the uptake in leafy vegetables versus root/bulb vegetables. The target crops; radish, tomato, wheat, maize, mungbean, rice and

paprika, are not optimal for such a comparison and concentrations in the edible part of the plant are more relevant to human exposure than the root:shoot distribution.

Since some REEs have been shown to stimulate plant growth, they are often added to fertilisers (Khan et al., 2017a; Kovaříková et al., 2019; Tyler, 2004a/b; Xu et al., 2002). Mechanisms are proposed to include a supported photosynthesis, accumulation of biomass, formation of secondary metabolites and reduced oxidative stress; but knowledge is scarce and higher levels of REEs have shown harmful effects on plants suggesting a threshold of toxicity (Kovaříková et al., 2019). A major part of the literature available on REEs, addresses effects on crop yield, while a much smaller part deals with toxicity or actual REE uptake (Ramos et al., 2016). Among the studies that have focused on plant uptake, however, several point to low BCFs as well (Carpenter et al., 2015; Khan et al., 2017a; Markert and de Li, 1991; Tyler, 2004a, Yoshida and Muramatsu, 1997). Table 4 lists REE BCFs from several other studies, and – when available in the original publication – also presents data for TMCs for comparison. Yoshida and Muramatsu (1997), who studied the uptake of La and Ce in leaves, shrubs and mushrooms (after HNO₃, HF and HClO₄ soil digestion) found high BCFs of several TMCs in mushrooms (Cd = 19.1, Cu = 3.5, Zn = 2.4), but a much lower uptake of the REEs (Ce = 0.012; La = 0.013). In another study, with focus on Beech trees (*Fagus sylvatica L.*), Tyler (2004b) presented BCFs for most REEs in the range 0.10–0.12 after soil digestion with concentrated HNO₃. Europium showed a slightly higher BCF (0.31), proposedly due to its common presence in the divalent cationic form more readily taken up than the trivalent forms, but the TMC BCFs were in general even higher (Cd = 0.79, Cu = 1.00, Pb = 0.20, Zn = 0.80). After soil digestion with Triethanolamine and EDTA, BCFs for REEs varied between 0.04 and 0.09 in forest plants including birch, pine, lingonberry, blueberry, wavy hair grass and hair cap moss according to Markert and de Li, 1991. But still, high BCFs have been reported for some plant groups – subgroups of ferns, grass, and citrus plants for example (Khan et al., 2017b; Ozaki et al., 2000; Turra et al., 2013; Tyler et al., 2004a/b). Furthermore, Wen et al. (2001) recommended not to use fertilisers that contain REEs because of elevated concentrations found in crops (tomatoes, radish, cucumbers, kidney beans, cabbage, Chinese cabbage) exposed to REE fertilisers in comparison with corresponding unexposed control crops. In radish, which was the only root vegetable included, the concentrations were as much as 125 (La), 24 (Ce), 85 (Pr) and 101 (Nd) times higher in the exposed groups than in the controls. The comparison may not be completely relevant though, since the uptake may differ depending on whether the REEs are added to the soil with e.g., fertilisers or whether they originate from other sources. For example, significant differences in time to equilibrate with the soil can have an impact on the result.

With the limited data at hand, one should not rule out the possibility that certain vegetables used for human consumption may accumulate REEs efficiently. High BCFs have been reported on some occasions, but it is important to highlight that this is mainly when soil concentrations have been measured after leaching with weak extractants; thus, increasing the plant:soil concentration ratio (Jiang et al., 2020; Liu et al., 2017). In our study too, there were some examples with relatively high maximum BCF values (>1 for Ce, Dy, Gd, Nd, Pr and Sm), but here too they were found when the weaker soil extraction was used. And even with this finding in mind, we can conclude that our data clearly points to a much lower BCFs for REEs than other TCE metals and traditional metal contaminants.

The ultimate and final question is then would we, based on the BCFs calculated in our study, conclude that vegetable consumption is an exposure pathway of concern either now or in the future? Rodríguez-Hernández et al. (2019) suggest a total oral tolerable daily intake (TDI) of 61 µg per kg bodyweight and day for the sum of Ce, Dy, Er, Eu, Gd, Ho, La, Lu, Nd, Pr, Sc, Sm, Tb, Tm, Y and Yb. The ΣREE concentrations measured in the 22 urban soils of this study result in an exposure to REEs from 0.13 to 0.47 µg/kg/day (Eq. (2)), i.e., <1 % of the suggested TDI of 61 µg/kg/day. However, this calculation is based on the lower BCF

Table 4

Proposed bioconcentration factors (BCFs), in previous studies for REEs in edible and non-edible plants.

Reference	Crop	BCF REEs	BCF TMCs	Soil digestion/leaching
Markert and de Li, 1991	Forest plants including birch, pine, lingonberry, blueberry, wavy hair grass and moss (*Polytrichum*)	0.04–0.09[a]	-	Triethanolamine and EDTA
Yoshida and Muramatsu, 1997	Tree leaves *Pinus thunbergia, Morus bombycis*	La = 0.031[b] Ce = 0.013	Cu = 0.58 Cd = 1.5 Pb = 0.082 Zn = 0.51	HNO$_3$, HF and HClO$_4$
	Shrub *Indigofera pseudo-tinctoria, Vitex rotundifolia, Oenothera lamarckiana, Miscanthus sinensis, Ophiopogon japonicus*	La = 0.055 Ce = 0.018	Cu = 0.84 Cd = 0.7 Pb = 0.069 Zn = 0.46	
	Mushroom *Suillus granulatus, Lactarious hatsudake, Russula mariae, Amanita pantherine, Tricholoma flavovirens*	La = 0.013 Ce = 0.012	Cu = 3.5 Cd = 19.1 Pb = 0.052 Zn = 2.4	
Tyler, 2004b	Beech (*Fagus sylvatica* L.)	0.10–0.12[c] (all investigated REEs except Eu)	Cd = 0.79 Cu = 1.00 Pb = 0.20 Zn = 0.80	HNO$_3$
Ozaki et al., 2000	96 ferns and fern ally species.	La = 0.39[d] Ce = 0.097	Zn = 0.78	Not stated
Turra et al., 2013	Citrus plant (*Citrus sinensis*)	La = 0.62–1.09[e]	-	Not stated
Khan et al., 2017b	**Ferns:** *Dicranopteris dichotoma, mining area* *Dicranopteris linearis (A), mining area* *Dicranopteris linearis (B), natural area*	BCFs for ∑ REEs[f] 53.74 97.19 151.11	 - - -	65% HNO$_3$ and 35% H$_2$O$_2$
	Melastomataceae: *Melastoma malabathricum L., industrial área*	64.26	-	
	Grass: *Cyperus difformis Rottb., road side* *Cyperus kyllingia Rottb., road side* *Cyperus distans L., river side* *Cyperus rotundus L., river side*	 12.39 99.94 151.70 134.13	 - - - -	

[a]Transfer factor (TF) = C$_{plant}$/C$_{soil}$; [b]TF = C$_{plant/mushroom}$/C$_{surface\ soil}$; [c]C$_{root}$/C$_{organic\ soil}$; [d]mean R$_j$ (C$_{j,plant}$/C$_{j,soil}$) value; [e]C$_{leaf}$/C$_{soil}$; [f]BCF=C$_{leaves}$/C$_{soil}$

value, obtained after AR soil digestion. Replacing this value with the higher BCF, calculated based on soils leached with 0.1 M HNO$_3$, gives a corresponding ΣREE exposure from 3 to 11 µg/kg/day, i.e., 5–18 % of the abovementioned TDI.

3.2.3. Less studied TCEs (LSTCEs)

Bioconcentration factors were calculated for three LSTCEs: Tl, Ga and Nb. A common feature of these three elements was low detectable concentrations from the soils studied following leaching with 0.1 M HNO$_3$, indicating low solubilities. The percentage values found under subfigure 1b show the relative fraction of the AR-extracted concentration that was detectable after 0.1 M HNO$_3$ leaching. For Nb, the concentration brought into solution by the milder extraction was only 0.22 % of that following AR treatment, and for Ga and Tl only 1.7 % and 2.8 % were released. The corresponding figures for the remaining elements lie between 7.3 % (Al) and 79 % (Cd). For elements with a low solubility, the plant:soil concentration ratios, or BCFs, will increase more when the soil concentration is determined after a weaker soil extraction, than will elements that are readily soluble. Fig. 1 clearly shows this effect for Ga, Nb and Tl through a shift towards the right, with higher BCFs relative to the other elements, in subfigure 1b compared to subfigure 1a. Other sources as well (Folens and du Laing, 2017; Salminen et al. 2005; Tanaka, 2004) point to these elements' low solubility in pore water.

3.2.3.1. Thallium. Albeit an element that currently attracts attention as a TCE, Tl was discovered as early as 1861 (Xiao et al., 2004). The research that has been undertaken to date already points to the intake of vegetables as a main route of Tl exposure (Karbowska, 2016; Kazantzis,

2000) and various concerns have been raised since high levels have been found in fruits, vegetables and farm animals near contaminated areas, e. g., in China, Germany and North Macedonia (Doulgeridou et al., 2020; Karbowska, 2016; Kazantzis, 2000; Xiao et al., 2004). Thallium concentrations in plants are usually below 0.1 mg/kg (Kazantzis, 2000), or even lower (Karbowska, 2016). On the other hand, Xiao et al. (2004) reported Tl concentrations close to 500 mg/kg in green cabbage from a mining area in China.

Despite the relatively low solubility of the element, a predicted uptake in plants would be significant even if BCFs are calculated from soil concentrations after AR extraction (Fig. 1a). The Tl BCFs in our study were, for example, higher than the BCF values for both Pb and As, for which diet is generally the main route of exposure. When BCFs are calculated based on the 0.1 M HNO$_3$ soil leaching, Tl is the element that would have the highest predicted uptake (if the BCFs calculated in this study were applied to another soil). The only exception is K, which was included for comparison just because of its role as both a major soil constituent and major plant nutrient (Fig. 1b).

Mean BCFs for Tl were found to be highest for chard; 0.31 when calculated against soil AR concentrations and 13 after soil leaching with 0.1 M HNO$_3$. The BCFs in lettuce and carrot were 0.036 (or 1.9) and 0.088 (or 4.9), respectively. Fig. 2 shows a summary of BCFs presented for Tl in previous studies. The results vary by several orders of magnitude (from 0.0027 to 63), probably to a large extent due to the inconsistencies in analytical approaches, see some methodological notes below the graph. However, the observed span is large also for the AR and HNO$_3$ digested samples of our study (no 1 and 2 in Fig. 2), indicating that analytical differences can't be the only explanation. And the overall
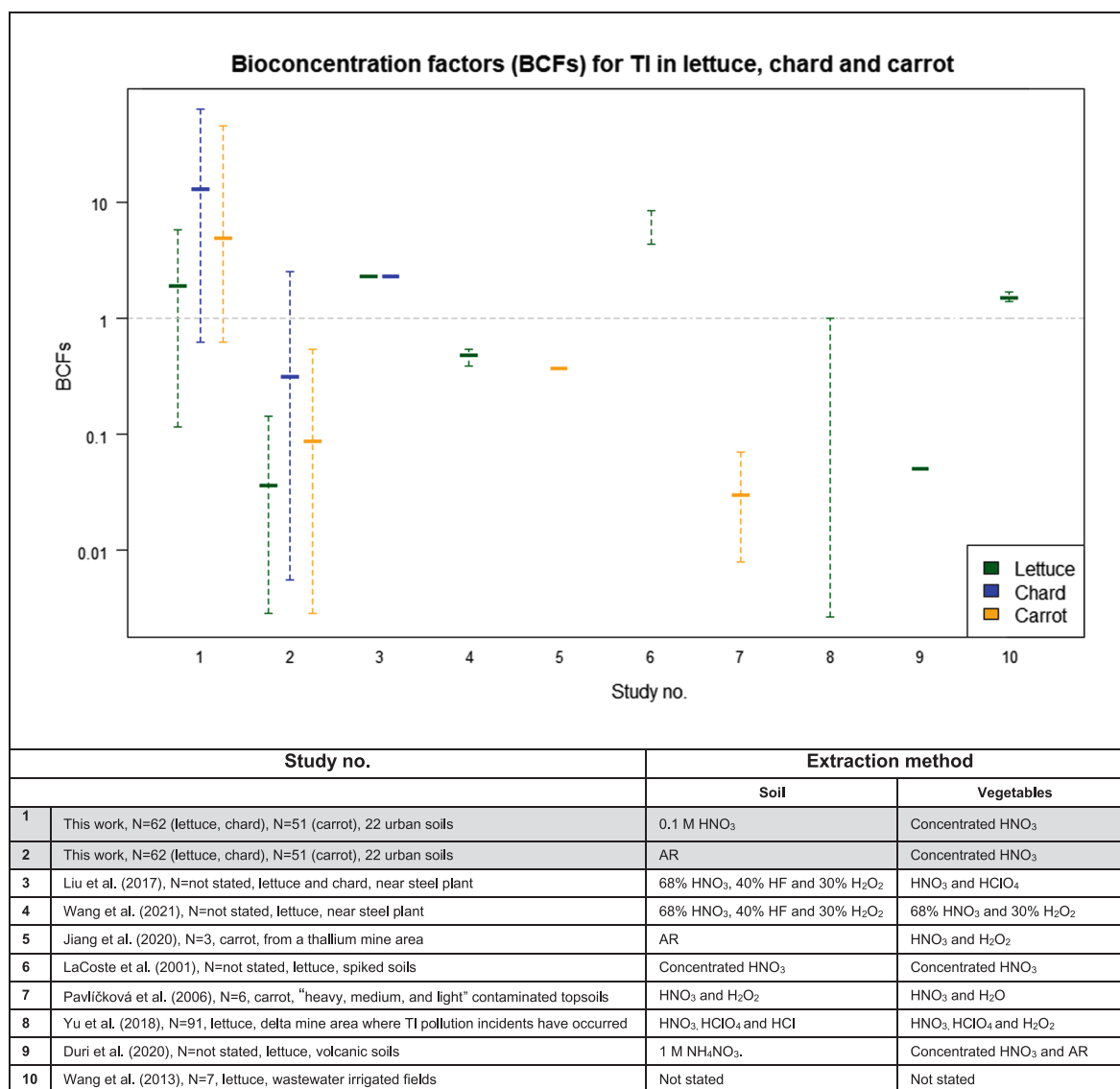
Bioconcentration factors (BCFs) for Tl in lettuce, chard and carrot

	Study no.	Extraction method	
		Soil	**Vegetables**
1	This work, N=62 (lettuce, chard), N=51 (carrot), 22 urban soils	0.1 M HNO$_3$	Concentrated HNO$_3$
2	This work, N=62 (lettuce, chard), N=51 (carrot), 22 urban soils	AR	Concentrated HNO$_3$
3	Liu et al. (2017), N=not stated, lettuce and chard, near steel plant	68% HNO$_3$, 40% HF and 30% H$_2$O$_2$	HNO$_3$ and HClO$_4$
4	Wang et al. (2021), N=not stated, lettuce, near steel plant	68% HNO$_3$, 40% HF and 30% H$_2$O$_2$	68% HNO$_3$ and 30% H$_2$O$_2$
5	Jiang et al. (2020), N=3, carrot, from a thallium mine area	AR	HNO$_3$ and H$_2$O$_2$
6	LaCoste et al. (2001), N=not stated, lettuce, spiked soils	Concentrated HNO$_3$	Concentrated HNO$_3$
7	Pavlíčková et al. (2006), N=6, carrot, "heavy, medium, and light" contaminated topsoils	HNO$_3$ and H$_2$O$_2$	HNO$_3$ and H$_2$O
8	Yu et al. (2018), N=91, lettuce, delta mine area where Tl pollution incidents have occurred	HNO$_3$, HClO$_4$ and HCl	HNO$_3$, HClO$_4$ and H$_2$O$_2$
9	Duri et al. (2020), N=not stated, lettuce, volcanic soils	1 M NH$_4$NO$_3$.	Concentrated HNO$_3$ and AR
10	Wang et al. (2013), N=7, lettuce, wastewater irrigated fields	Not stated	Not stated

Fig. 2. Bioconcentration factors for Tl in lettuce, chard and carrot, defined in this work (study no. 1 and 2) and in previously performed studies. In all cases, concentrations refer to dry weight vegetables. When concentrations and number of samples are not specified, they were not given in the original publication.

picture still supports our results of Tl as an element with relatively high BCF values, considering that average BCFs for TMCs such as Pb and As were 0.0083 (1 0 0) and 0.014 (0.17) respectively. Even though our data suggests a relatively low solubility, in accordance with the results from e.g. Pallavicini et al. (2018) who show that Tl is mainly associated with refractory mineral phases, it should be acknowledged that others, e.g. Karbowska (2016), describe a relatively high solubility of Tl, and many are of the opinion that the Tl uptake is facilitated by its resemblance to K (Duri et al.. 2020; Xiao et al., 2004, Yu et al., 2018). Particularly hyper accumulative edible plants for Tl appear to be *Brassicaceous* plants such as cabbage (Kazantzis, 2000; LaCoste et al., 2001; Ning et al., 2015; Pavlíčková et al., 2006). However, resulting concentrations in vegetables should in most cases still be low relative to the TMCs, as a result of Tl's low concentration in the soils (Table 2).

No recently updated TDI value has been found for Tl in the literature, although this is probably the TCE element with most reported/known toxic effects (Table 1). According to several researchers, the toxicity of Tl is as severe as that of As, Cd, Hg and Pb (Duri et al., 2020; Heim et al., 2002; Wang et al., 2013; Xiao et al., 2004), and the element is found on the US EPA's Priority Pollutant List (US EPA, 2014). If the provisional

oral health- based guidance value of 0.2 µg/kg/bw, proposed in 1998 by the RIVM (RIVM, 1998), is used as a reference point and exposure calculations are made as for the REEs above, the resulting exposure to Tl from lettuce grown on the investigated soils still only equals 0.6–6 % of the tolerable intake. Replacing the lower BCF with the higher, as described for the REEs, gives a Tl exposure of as much as 31–320 % of the specified guidance value. The big difference highlights the importance of using representative BCF values and the need to adopt standards for how BCFs should be determined and used in risk assessments in general.

3.2.3.2. Gallium. As shown in Table 2, the average concentrations of Ga in the cultivation soils of our study were 3–5 times higher than those of Cd, even though a much narrower min–max interval suggests lower spatial variability – so its abundance is not negligible. How much Ga is then taken up in plants, becoming a source of exposure via this route? Our results indicate low Ga BCFs, at least when related to the concentration in the soil measured after AR digestion (Fig. 1a). Mean BCFs for Ga were 0.0030 (lettuce), 0.0035 (chard) and 0.0016 (carrot). The BCF values relative to the other investigated elements were (as expected)

higher after the 0.1 M HNO_3 soil leaching (1.1, 0.84 and 0.45, respectively), but the low solubility of this element means that any uptake from pore water will still result in low concentrations in plants. For example, reviewing Table 2 again, we see that while concentrations of Ga in soil are higher than for Cd, the concentrations in vegetables are much lower.

Putting our results into a wider scientific context is challenging given the lack of previous studies of Ga uptake in consumable vegetables. However, the study by Jensen et al. (2018) shows similar results. They present a low BCF (0.0037) in ryegrass and suggest that exposure via direct soil intake is a more likely route of exposure for this element. They argue that Ga, which is chemically similar to Al, probably remains in the epidermis of the root, i.e., in the outermost cell layer; thus, with limited uptake and translocation within the plant. This may also explain why we found higher Ga concentrations in the leafy vegetables than in carrot (Fig. 1). Although the cultivation was conducted indoors to limit the deposition of airborne particles, fine lithogenic material from the soil may still have adhered to the growing vegetables, and the higher surface:volume ratio of lettuce and chard compared to carrot could then have resulted in more adhering soil material. In a new paper by Chen et al. (2022), Ga is on the contrary pointed out as a possible public health problem due to exposure via rice cultivated in Ga rich paddy fields. No exposure calculation was carried out and evaluated to back this statement up, however, and the authors further show that the vast majority of Ga was accumulated in the roots of the rice plants, with only a smaller part translocated to the grains. There are today no TDI values available to assess the possible severity of Ga exposure via vegetable consumption. While negative health effects have been described (Table 1) and much remains to be revealed about TCE element toxicity, the moderate uptake should also be interpreted against the general view of Ga as an element of low toxicity (Salminen et al., 2005).

3.2.3.3. Niobium. Mean BCFs of Nb were 0.0043, 0.0056 and 0.0016 in lettuce, chard and carrot, respectively, when calculated from the analysis of soil AR digestates; and 3.8, 4.3 and 1.2, when 0.1 M HNO_3-extractable concentration values were used. Similarly to Ga, the predicted uptake into plants, based on our BCFs, is small relative to other elements, in particular when proposed BCFs are related to total elemental concentrations in soils prior to chemical analysis. However, as for Ga, the low solubility probably explains the element's low accessibility for plant uptake. A low solubility and mobility of this element has been described previously too, primarily explained by the formation of stable oxide phases, although association with soluble organic acids may increase the element's mobility (Greger, 2004; Kabata-Pendias and Pendias, 2001; Salminen et al., 2005; Sutliff-Johansson et al., 2021; UK Soil and Herbage Pollutant Survey, 2007). The potential health risks associated with Nb intake should also be assessed as low due to the element's low toxicity (Table 1). Although not too well investigated it has been proposed that the LD50 value of Nb is as high as several thousand mg per kg body weight in rat (Ray et al., 2020; Sigma-Aldrich, 2022), which is probably due to its biological inertness (Schulz et al., 2017).

4. Conclusions

In this study paired urban soil–plant data from 22 soils growing 3 different plants were utilised to obtain bioconcentration factors for Technology-critical elements (TCEs), compared to traditional metal contaminants, in order to evaluate and discuss present and future risks from emerging contaminants. In general, mean BCFs were highest in the leafy vegetables, particularly in chard, and lowest in carrot. The BCF values for the rare earth elements (REEs) were low, both in comparison to the other evaluated TCEs and compared to traditional metal contaminants (TMCs) such as As, Cd, Cu, Pb and Zn. All elements of the REE group, both light and heavy REEs, showed similar BCFs – implying that further assessments of these elements' susceptibility for plant uptake may not need to include the full range of elements, or they can be treated

as a group. The BCF values for Ga and Nb were low as well, and probably due to these elements' low solubility. Thallium on the other hand, was the TCE element with the highest BCF value; in particular after the weaker soil leaching but also after digestion with AR. This may be particularly important considering that available data points to substantial toxicity for this element. The obtained bioconcentration factors were, however, strongly affected, several orders of magnitude, by the soil extraction media (*Aqua Regia* or 0.1 M HNO_3). To calculate adequate BCFs and obtain robust and comparable values, our recommendation is to define soil element concentrations after a total or (pseudo)total extraction, for example after an *Aqua Regia* digestion. Hereby, the influence of geochemical factors other than the concentration of the focus element(s) will be less perceptible than if a weaker leaching method is used.

We can't point to any evident risks from consumption of vegetables grown on urban soils with current concentrations of TCEs. The key phrase here is, however, "current concentrations" since many of the TCEs are considered contaminants of soon-to-be emerging interest. Given that vegetable consumption is a major route of exposure for many other metal contaminants, in many cases with comparable or lower BCFs than several TCEs, we stress the importance of future studies that look into how increasing concentrations of TCEs in soil may affect their future accumulation in plants, and how different soil geochemical variables affect the soil:plant transfer of these elements. In addition, health based toxicological reference values are warranted for assessing the potential risks associated with dietary TCE exposure.

Declaration of Competing Interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2022.107504.

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