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Emerging risks of toxic metal(loid)s in soil-vegetables influenced by steel-making activities and isotopic source apportionment

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A R T I C L E I N F O

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A B S T R A C T

Industrial activities tend to deteriorate adjacent agricultural lands due to accumulation of potentially toxic elements in soils and crops. However, better understanding of their distinctive source partitions and transfer process remains insufficient in steel-making area. The paper focuses on the pollution levels, health risks, and provenance identification of Tl, As, Pb, Cu, Ni, Co, Sb, Cd, Zn, Be, Cr, Fe, Mn, Mo, Sn, and V in common vegetables from different farmlands near a steel-making plant. The results showed that the Tl, As, Pb, Cd, Cr, Cu and Mn were of high-level contamination in soils and generally above the maximum permissible level (MPL). Calculation using hazard quotients (HQ) exhibited that consumption of the studied vegetables may entail significant health risks to residents, especially for children, resulting from the elevated contents of Tl, As and associated toxic elements. Calculation by binary mixing model using Pb isotopic compositions suggested that steel-making activities contributed to 35–80% of the contamination of Pb and As in vegetables. It is necessary to adopt appropriate remediation measures to mitigate the farmland contamination and ensure the food safety of the agricultural products.

1. Introduction

With rapid development of industrialization and urbanization, industrial activities have led to the increased occurrence and levels of toxic elements in the environmental medium (Bi et al., 2018; Dietrich et al., 2018; Joon et al., 2020; Guzinski et al., 2013; Lisak et al., 2013; Ding et al., 2019; Liu and Han, 2020). Toxic metal(loid)s contamination in crop production is receiving widespread public attention in the world due to their stability and resistance to biodegradation (Peng et al., 2020; Muhammad et al., 2020; Wang et al., 2020a; Zeng et al., 2020). They can pose substantial potential risks to the ecological environment and can harm human health through skin contact, inhalation, soil-food chain and other absorption pathways (Harmanescu et al., 2011; Lu et al., 2011; Liu et al., 2013; Rinklebe et al., 2019; Yin et al., 2020). Redox-driven mobilization of some contaminants may facilitate the release process of these toxic metal(loid)s from the soil, which even increase the possibility of transferring to the groundwater, posing unfavorable impacts on human health and water quality (Rinklebe et al., 2020).

Steel-making activity has been regarded as a great contributor to wastewater and dust containing various pollutants (Liu et al., 2017; Petranikova et al., 2020). The pollutants of steel-making plant have been studied extensively. For example, Adamo et al. (2002) found that the soils near a steel-making plant in southern Italy had been considered contaminated with Cu, Cr, Pb, Zn and Ni, whose average contents were

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53.3, 151, 99.2, 253 and 123 mg/kg, respectively. Liu et al. (2013) showed that seriously environmental risk of Cd (0.52–2.55 mg/kg) was derived from steel-making activities near the Jishui River, Jiangxi Province, China. Dietrich et al. (2018) used bulk chemistry and scanning electron microscope data to identify the sediment contamination by Pb, Cd, Zn and Cr from AK steel-making plant, Middletown Ohio, USA, which all exceeded background levels. MacDonald et al. (2011) reported that all birch and larch collected from Sydney steel-making plant exhibited elevated concentrations of Pb ranging from 12 to 52 mg/kg, and 10–30 mg/kg, respectively. Zhou et al. (2019) emphatically detected 17 kinds of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) in surface soils around a steel-making plant in northern China, with PCDD/Fs concentration ranging from 0.16 to 4.5 ng I-TEQ/kg, presenting potential environmental organic pollution risk.

To our best knowledge, the majority of the scientific community efforts concerning steel-making activity focused on traditional contaminants such as Pb, Cd, Cr, Zn, Polychlorinated biphenols (PCBs), and volatile organic compounds (VOCs). Emerging risks of remarkably hazardous metal(loid)s, such as thallium (Tl) and arsenic (As), are rarely concerned (Wang et al., 2020b; Liu et al., 2019d, 2021). In fact, arsenic, as a highly toxic element, is closely associated with the diseases of lung, liver, bladder, skin keratoses and peripheral vascular and kidney cancer (Topal et al., 2020; Bhattacharya et al., 2020; Zhang et al., 2020). In addition, As in the soils is partly mobile and easily gets into the air or water, becoming a secondary pollutant via natural/uncontrollable process typical of leaching and weathering, inducing a wide range of environmental issues (Beiyuan et al., 2017; Cui et al., 2018; Siddiqui et al., 2020). And notably, Tl, it is also a highly toxic element with a pathogenic potential on neurological brain or even death with a lethal dose of only 8–10 mg/kg for an adult, which ranks more dangerous than traditional metals of Pb, Zn, Cd and Cr (Liu et al., 2017; Peter and Viraraghavan, 2005; Wang et al., 2020c; Zhang et al., 2018). Previous studies imply that a great portion of Tl in soils can be transferred to pore waters and plants since humic acid of soils seems to present no strong complexation with Tl (Jacobson et al., 2005; Pavoni et al., 2017). We hypothesize that residents living near steel-making plant may suffer from health threats through food chains due to exposure to these toxic pollutants, especially Tl and As.

Therefore, it is critical to investigate the effects of Tl and As on soil-crop system surrounding a steel-making plant. On the other hand, the original sources of these concerned pollutants in soil-crop system may be diverse. How to clarify metal(loid)s enrichment in the soil-crop system from both natural and anthropogenic emissions, seems to be one of the crucial issues. Pertinent studies have displayed that Pb isotopic composition was very effective to track the source and pathway of metal contaminants in soils, waters, sediments and atmospheric environments in recent years (Peng et al., 2020; Liu et al., 2020; Liu et al., 2018a; Liu et al., 2018b; Zurbrick et al., 2017). Hence, the isotopic “fingerprint” of Pb, which is not affected to a measurable extent by physical or chemical fractionation processes (Peng et al., 2020), can also hypothetically serve as a powerful tool to identify the sources of toxic elements in agricultural soils and crops.

Even though most steel-making related enterprises may be now under strict regulation, potential risks of toxic metal(loid)s in soils may still persist over a long term. Therefore, the present study takes the soil-crop system near a steel-making plant in northern Guangdong province, China as a typical example, the objectives are to (1) evaluate the metal(loid)s of Tl, As, Pb, Cd, Co, Mn, Ni, Sb, Cr, Cu, Zn, Be, Fe, Mo, Sn and V enrichment characteristics in the soils and crops; (2) assess the potential health risks (non-carcinogenic and carcinogenic) of humans via consumption of locally grown vegetables; and (3) estimate the relative contribution of anthropogenic source from the steel-making plant using Pb isotopic fingerprinting technique.

2. Materials and methods

2.1. Study area and sampling

The length and drainage area of Beijiang River are 573 km and 52,068 km^2, respectively, which is the second largest tributary of Pearl River, China (Wang et al., 2017; Wang et al., 2020d). All the 5 sampling sites (S1–S5) were selected in the vicinity of a steel-making plant in Beijiang River Basin. The distance between the upstream and downstream is ~4 km. The steel-making plant was established in 1960s, which intermittently discharged/emitted wastewater, solid wastes and dusts. In particular, the steel-making plant had discharged massive quantities of Tl-containing wastewater over the past decades, leading to serious environmental contamination (Liu et al., 2017). The area is characterized with subtropical monsoon climate, holding an annual average temperature of 22 °C with relative humidity ranging 60–90% (Liu et al., 2017). A map showing the sites is presented in Fig. 1.

In total, 16 crop samples and 16 corresponding rhizosphere soil samples were collected in the study area. Each soil sample was manually collected from a 10 cm deep soil using a stainless-steel shovel and stored in a polyethylene bag. A depth of 10 cm soil was chosen to enclose because the toxic metal(loid)s have relatively high exposure risks to plants and the public within this depth range (Cittadino et al., 2020; Golden et al., 2020). Then the samples were stored at low temperature (4 °C) and immediately transported to our laboratory for pretreatment and analytical determination. The vegetable grown soils are mainly characterized with clay type, and the basic physical and chemical properties of soil samples are provided in Table S1. There are 16 kinds of crops, including *Pachyrhizus erosus (Linn.) Urb.* (S1), *Ipomoea aquatica Forsk* (S1), *Lactuca sativa var longifoliaf. Lam* (S1), *Brassica oleracea L.* (S1), *Amaranthus tricolor L.* (S1), *Beta vulgaris L.* (S1), *Nelumbo nucifera* (S1), *Capsicum annuum L.* (S2), *Lactuca sativa L. var. ramosa Hort.* (S2), *Allium fistulosum L.* (S2), *Lactuca sativa var longifoliaf. Lam* (S2), *Beta vulgaris L.* (S3), *Sonchus lingiaus* (S3), *Cichorium endivia L* (S4), *Allium fistulosum L.* (S4) and *Sonchus lingiaus* (S5) collected to represent the main vegetable varieties near the steel-making plant. The detailed information about growth stage and general conditions of vegetables are shown in Table S2.

The vegetable samples were cleaned by Milli-Q water and then fractured into roots, stems and leaves (Noli and Tsamos, 2016). The vegetables were then oven-dried to a constant weight, followed by grounding to less than 80 μm. Similarly, the rhizospheric soil samples were dried in an oven at 80 °C to a constant weight, then grounded to less than 100 μm.

2.2. Measurement of metal contents

In brief, the finely ground soils (~0.100 g) were digested in a mixture of 8 mL 68% HNO_3, 4 mL 40% HF and 2 mL 30% H_2O_2 by heating on Teflon vessel at 150 °C. Repeated the above steps several times until the solution was relatively clear, then removed additional hydrofluoric acid by heating till dryness. The samples were diluted to 100 mL using double-deionized water (Milli-Q Millipore, 18.25 MΩ/cm). Meanwhile, vegetable samples were digested by 3 mL 68% HNO_3 and 1 mL 30% H_2O_2 at 150 °C. The process was repeated several times till dryness. Then the double-deionized water was used for sample dilution to a final volume of 25 mL. Afterward, all digestions were determined by an Inductively Coupled Plasma Mass Spectrometry (ICP-MS) (PE-Sciex Elan 6100 DRC-II, Perkin Elmer, US) (Liu et al., 2018c). The quality of the analytical measurements was guaranteed with the certified reference materials provided by the National Standard Reference Materials, i.e., GBW07406 (soil) and GBW07605 (tea leaves). The accuracy of the measurements is assured within 5% RSD.

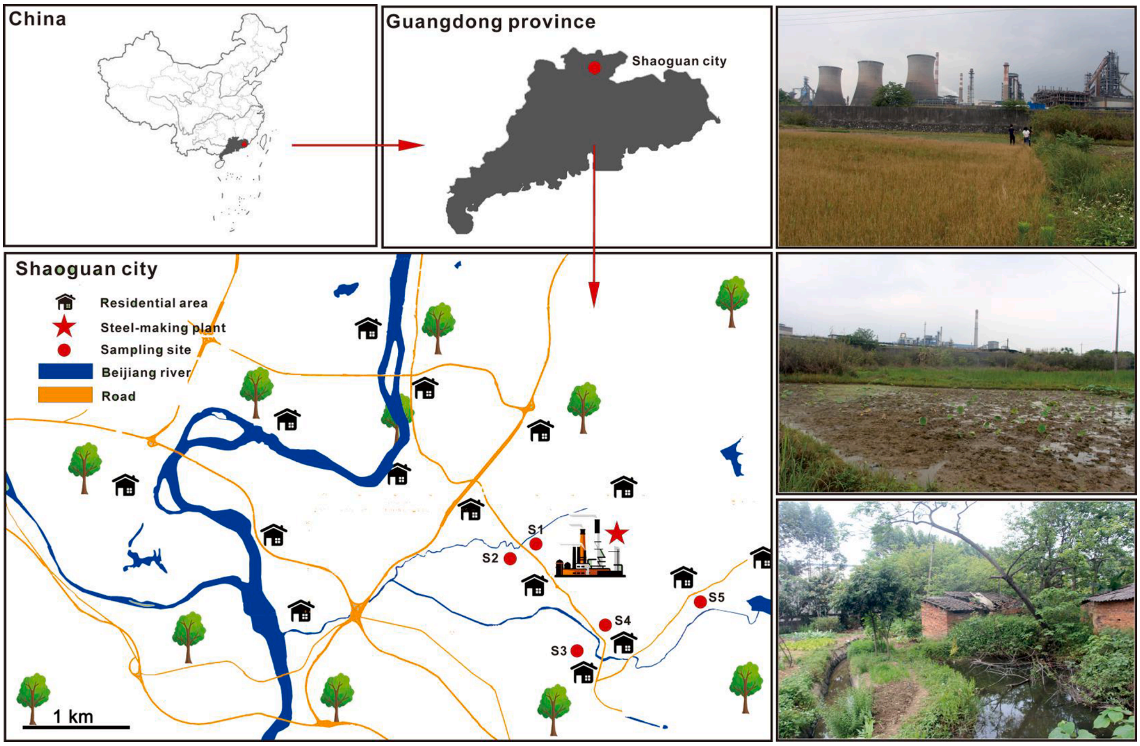

Fig. 1. Schematic display of study area.

2.3. Data process

SPSS Statistics 22 and Origin 2019 were used to analyze and plot the data, respectively. Pearson test was adopted to evaluate the degree of correlation between the contents of Pb and other elements (Tl, Co, Cu, Mn, Ni, As, Sb, Zn, Cd, Be, Fe, Cr, Mo, Sn and V) in the vegetable tissues and rhizospheric soils.

2.4. Evaluation method for pollution risk

2.4.1. Bio-concentration factor (BCF) and transfer factor (TF)

The indices of BCF and TF represented the accumulation capability of toxic metal(loid)s from soil to plant and the transferability of toxic metal (loid)s from root to aerial part, respectively, calculated as follow (Rezapour et al., 2019):

$$TF = \frac{C_{aerial}}{C_{root}} \quad (1)$$

$$BCF = \frac{C_{vegetable}}{C_{soil}} \quad (2)$$

where C_{root} and C_{aerial} are the concentrations of individual elements in the root and aerial part (stalk and leave) of vegetables, and C_{soil} and $C_{vegetable}$ represent the corresponding value in the soil. When the BCF and TF of an element in a vegetable exceed 1, it indicates the enrichment ability of vegetables for the element was high.

2.4.2. Health risk assessment

The hazard quotient (HQ) and daily chronic intake (CDI) were applied for assessing the potential health risks of local vegetable intake to Tl and other metals. The CDI used to determine the total metal concentration in metal exposure assessment was calculated as followed (Opoku et al., 2020):

$$CDI = \frac{C_i \times IR \times ED \times EF}{BW \times AT} \quad (3)$$

where C_i (mg/kg), IR (kg/d), ED (a), EF (d/a), BW (kg) and AT (d) refer to the element concentration in the vegetable, average daily intake of vegetables for residents, exposure duration, exposure frequency, average body weight and average exposure time, respectively. Based on the previous investigation on consumption habits of suburban residents in Guangdong Province, the IR of light and dark green vegetables for children, adults and seniors were 0.355, 0.223 and 0.366 kg/(d·person), respectively (Shi-Cong et al., 2014). The value of EF was 350 d/a. The ED values for children, adults and seniors were 6, 30 and 70 years, respectively (NBSPRC, 2016). The BW for children, adults and seniors were 24.5, 65 and 59.4 kg, respectively (Song et al., 2009). AT is the average exposure time (365 d/a × number of exposure years). The conversion factor was 0.085 used to convert the dry weight to fresh weight of vegetables (Rattan et al., 2005).

The US Environmental Protection Agency points out that elements such as Tl, Pb, Ni, Co, Zn, Mn, Cu and Sb in food have non-carcinogenic effects on human health, while elements such as Cd, As and Cr have carcinogenic effects (US EPA, 1989, 2004, 2009). For non-carcinogenic elements, the hazard coefficient (HQ) was represented by the potential health risks of the daily estimated intake of each element (Karbowska et al., 2014):

$$HQ = \frac{CDI}{R_fD} \quad (4)$$

Herein, the oral reference doses (R_fD) in food are 0.8×10^{-5} for Tl, 3.5×10^{-5} for Pb, 3.0×10^{-1} for Zn, 4.0×10^{-2} for Cu, 2.0×10^{-2} for Ni, 3.0×10^{-4} for Co , 2.4×10^{-2} for Mn, 2×10^{-3} for Be,7×10^{-1} for Fe, 5×10^{-3} for Mo, 6×10^{-1} for Sn and 5×10^{-3} for V (US EPA, 1989, 2009, 2004). Guidelines of HQ are sorted as: HQ $<$ 1, hazard is low; HQ = 1.1–10, hazard is moderate; HQ greater than 10, hazard is high (Ji et al., 2017; USE PA, 1989, 2004, 2009). The carcinogenic elements (As, Cd, and Cr) via vegetable ingestion were calculated by multiplying their CDI values with the corresponding cancer slope factor of 15.1, 6.1, and 42.0 $(mg/kg/d)^{-1}$, respectively (USEPA 1999, 2004). Risk values in the range from 1×10^{-6} to 1×10^{-4} are acceptable.

2.5. Binary mixing model of lead isotope ratios

A binary mixing model was used to quantify the Pb contributions to the vegetable tissues from two sources (Komarek et al., 2008; Notten et al., 2008):

$$X_A = \frac{\left(\frac{^{20X}Pb}{^{20X}Pb}\right)_S - \left(\frac{^{20X}Pb}{^{20X}Pb}\right)_B}{\left(\frac{^{20X}Pb}{^{20X}Pb}\right)_A - \left(\frac{^{20X}Pb}{^{20X}Pb}\right)_B} \times 100 \quad (5)$$

Table 1
Tl and other toxic metal(loid)s (mg/kg dwt) contents in the rhizospheric soils of vegetables near the steel-making plant.

Site	Vegetables	Tl	Pb	Cd	Co	Cr	Cu	Mn	Ni	As	Sb	Zn	Be	Fe	Mo	Sn	V
S1	*Pachyrhizus erosus (Linn.) Urb.*	0.54 ± 0.02	101.0 ± 4.0	2.67 ± 0.11	6.4 ± 0.26	1140 ± 45	57.4 ± 2.30	5510 ± 220	17.9 ± 0.72	34.5 ± 1.38	3.03 ± 0.12	274 ± 10.9	2.84 ± 0.13	8.94 ± 0.35	6.72 ± 0.28	16.9 ± 0.80	189 ± 9.20
	Ipomoea aquatica Forsk	0.57 ± 0.02	118.5 ± 4.7	3.97 ± 0.16	7.1 ± 0.28	1240 ± 49	85.3 ± 3.41	7000 ± 280	24.3 ± 0.97	33.3 ± 1.3	4.02 ± 0.16	343 ± 13.7	4.06 ± 0.18	9.07 ± 0.38	5.99 ± 0.25	19.1 ± 0.85	227 ± 10.9
	Lactuca sativa var longifoliaf. Lam	0.54 ± 0.02	113.0 ± 4.5	2.63 ± 0.11	6.5 ± 0.26	476 ± 19	41.6 ± 1.66	2960 ± 118	21.7 ± 0.87	24.8 ± 0.99	4.77 ± 0.19	295 ± 11.8	3.55 ± 0.16	4.87 ± 0.17	2.56 ± 0.10	11.8 ± 0.42	130 ± 6.10
	Amaranthus tricolor L.	0.72 ± 0.03	183.5 ± 7.3	7.51 ± 0.30	10.0 ± 0.40	180 ± 7.20	54.6 ± 2.18	1140 ± 45	29.4 ± 1.18	48.3 ± 1.93	4.38 ± 0.18	516 ± 20.6	2.89 ± 0.12	3.75 ± 0.15	1.98 ± 0.06	14.7 ± 0.71	83.0 ± 3.90
	Beta vulgaris L.	0.83 ± 0.03	184.0 ± 7.3	5.88 ± 0.24	10.4 ± 0.42	93 ± 3.72	59.7 ± 2.39	689 ± 27.5	29.3 ± 1.17	48.7 ± 1.95	4.68 ± 0.19	476 ± 19.0	2.31 ± 0.11	3.51 ± 0.15	1.93 ± 0.06	52.3 ± 2.12	76.0 ± 3.60
	Brassica oleracea L.	0.44 ± 0.02	81.6 ± 3.26	1.82 ± 0.07	10.2 ± 0.41	77 ± 3.08	28.5 ± 1.14	417 ± 16.6	29.5 ± 1.18	23.1 ± 0.92	2.81 ± 0.11	200 ± 8.00	1.31 ± 0.03	2.74 ± 0.13	1.25 ± 0.05	5.10 ± 0.21	65.0 ± 2.80
	Nelumbo nucifera	0.52 ± 0.02	76.9 ± 3.08	1.71 ± 0.07	9.4 ± 0.38	69 ± 2.76	31.9 ± 1.28	421 ± 16.8	26.4 ± 1.06	24.9 ± 1.00	3.10 ± 0.12	184 ± 7.36	1.37 ± 0.03	2.69 ± 0.12	1.14 ± 0.03	5.60 ± 0.22	66.0 ± 2.80
S2	*Lactuca sativa L. var. ramosa Hort.*	0.40 ± 0.02	68.7 ± 2.75	2.07 ± 0.08	10.2 ± 0.41	200 ± 8.00	28.4 ± 1.14	4240 ± 169	25.1 ± 1.00	30.4 ± 1.22	3.29 ± 0.13	221 ± 8.84	3.10 ± 0.13	5.18 ± 0.21	1.93 ± 0.07	7.90 ± 0.31	218 ± 9.80
	Allium fistulosum L.	0.50 ± 0.02	81.0 ± 3.26	2.10 ± 0.08	12.7 ± 0.51	183 ± 7.32	25.0 ± 1.00	859 ± 34.3	31.4 ± 1.26	38.8 ± 1.55	42.7 ± 0.17	242 ± 9.68	1.72 ± 0.05	4.85 ± 0.22	1.98 ± 0.07	6.10 ± 0.26	114 ± 5.40
	Lactuca sativa var longifoliaf. Lam	0.53 ± 0.02	90.0 ± 3.60	2.59 ± 0.10	14.0 ± 0.56	205 ± 8.20	26.9 ± 1.08	628 ± 25.1	31.8 ± 1.27	43.4 ± 1.74	4.34 ± 0.17	289 ± 11.5	2.98 ± 0.12	5.56 ± 0.24	2.83 ± 0.09	11.4 ± 0.40	239 ± 7.10
	Capsicum annuum L.	0.52 ± 0.02	104.5 ± 5.0	4.30 ± 0.18	7.6 ± 0.28	654 ± 31.6	47.2 ± 1.86	4540 ± 217	19.5 ± 0.66	26.9 ± 1.30	3.09 ± 0.13	332 ± 15.6	1.92 ± 0.05	5.12 ± 0.18	2.33 ± 0.09	6.00 ± 0.26	142 ± 1.90
S3	*Beta vulgaris L.*	1.54 ± 0.06	170.0 ± 6.8	3.29 ± 0.13	12.7 ± 0.51	57 ± 2.28	303 ± 12.1	806 ± 32.2	15.1 ± 0.60	58.9 ± 2.36	10.05 ± 0.4	490 ± 19.6	3.36 ± 0.14	3.43 ± 0.15	7.68 ± 0.28	11.8 ± 0.45	54 ± 1.90
	Sonchus lingiaus	1.42 ± 0.06	198.5 ± 7.9	4.08 ± 0.16	13.4 ± 0.54	61 ± 2.44	372 ± 14.8	864 ± 34.5	16.3 ± 0.65	71.8 ± 2.87	10.90 ± 0.4	592 ± 23.6	3.63 ± 0.15	3.78 ± 0.16	8.67 ± 0.34	11.9 ± 0.22	56 ± 1.80
S4	*Allium fistulosum L.*	1.50 ± 0.06	236 ± 9.44	4.16 ± 0.17	14.2 ± 0.57	52 ± 2.08	386 ± 15.4	823 ± 32.9	18.7 ± 0.75	92.9 ± 3.72	14.10 ± 0.5	605 ± 24.2	4.07 ± 0.18	4.66 ± 0.20	11.3 ± 0.40	14.5 ± 0.66	64.0 ± 3.10
	Cichorium endivia L	1.10 ± 0.04	114.0 ± 4.5	1.39 ± 0.06	6.2 ± 0.23	50 ± 2.00	27.8 ± 1.11	539 ± 21.5	14.2 ± 0.57	48.7 ± 1.95	8.36 ± 0.33	247 ± 9.88	2.82 ± 0.10	3.08 ± 0.11	1.32 ± 0.04	9.70 ± 0.34	54.0 ± 2.50
S5	*Sonchus lingiaus*	0.38 ± 0.02	68.5 ± 2.74	1.64 ± 0.07	9.5 ± 0.38	73 ± 2.92	24.6 ± 0.98	405 ± 16.2	23.3 ± 0.93	21.4 ± 0.86	2.61 ± 0.10	187 ± 7.48	1.21 ± 0.04	2.73 ± 0.10	1.10 ± 0.03	4.50 ± 0.15	64.0 ± 2.80
Minimum		0.38 ± 0.02	68.5 ± 2.74	1.39 ± 0.06	6.2 ± 0.23	50 ± 2.00	24.6 ± 0.98	405 ± 16.2	14.2 ± 0.57	21.4 ± 0.86	2.61 ± 0.10	184 ± 7.36	1.21 ± 0.04	2.69 ± 0.12	1.10 ± 0.03	4.50 ± 0.15	54.0 ± 1.90
Maximum		1.54 ± 0.06	236 ± 9.44	7.51 ± 0.30	14.2 ± 0.57	1240 ± 49	372 ± 14.8	7000 ± 280	31.8 ± 1.27	92.9 ± 3.72	42.7 ± 0.17	605 ± 24.2	4.07 ± 0.18	9.07 ± 0.38	11.3 ± 0.40	52.3 ± 2.12	239 ± 7.10
Mean		0.74 ± 0.03	126 ± 5.03	3.17 ± 0.13	10.2 ± 0.41	277 ± 11.1	104 ± 4.14	1820 ± 73	23.6 ± 0.95	42.9 ± 1.72	5.65 ± 0.23	344 ± 13.8	2.70 ± 0.11	4.62 ± 0.19	3.79 ± 0.14	13.1 ± 0.52	115 ± 4.73
Background soil in China[a]		0.58	26.0	0.10	12.7	61	22.6	583	26.9	11.2	1.21	74.2	1.95	2.94	2.0	2.6	82.4
Background soil in GD Province[b]		0.52	36.0	0.06	7.0	50.5	17.0	279	14.4	8.9	0.54	47.3	1.61	2.42	7.7	5.8	65.3
MPL in soil[c]		1.00	50.0	0.30	40.0	250	50.0	1200	70.0	30.0	10.00	200	4.00	NG[d]	5.00	NG	130

[a] The value of Tl content is taken from Qi et al. (1992); the others are referred to Wei et al. (1991).
[b] GD: Guangdong Province. The value of Tl content is taken from Qi et al. (1992); the others are referred to CEMS (1990).
[c] MPL: Maximum Permissible Level. The values of Tl, Be, Mo, and V content are taken from Canadian Council of Ministers of the Environment (CCME, 1999, 1991, 1997, 2015); and others are referred to Soil Environmental Quality Risk Control Standard for Agricultural Land (GB 15618–2018) (SEP and GAQIQ, 2018; Wei et al., 1991; CCME, 1991, 1997, 2015).
[d] Not given.

where X_A is the contribution of source A, while $(^{20X/20X}Pb)_S$, $(^{20X/20X}Pb)_A$ and $(^{20X/20X}Pb)_B$ are the isotopic ratios (e.g., $^{206}Pb/^{207}Pb$) in the samples, source A ($^{206}Pb/^{207}Pb$ = 1.1690, the average value of steel-making raw materials) and source B ($^{206}Pb/^{207}Pb$ = 1.1952), respectively.

2.6. Pb isotopic analysis

The Pb in the vegetable sample matrix was separated by cation exchange chromatography, utilizing a cation exchange resin (100–150 mesh, Eichrom Sr-spec, US). In brief, Pb was separated from the digested samples in the resin, using HNO_3 and HCl as eluants (Liu et al., 2019c). The Pb isotopic composition was determined by a Multiple-Collector Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS, Nu Plasma HR, Nu Instruments, Wrexham, UK) (Liu et al., 2019c). The mass bias and analytical control of samples were adopted a common Pb isotopic standard (NIST SRM 981) to correct (Barling and Weis, 2008).

3. Results and discussion

3.1. Contents of Tl and associated toxic elements in rhizosphere soils

The total, minimum, maximum and average content of Tl, Zn, Pb, Co, Cr, Cu, Mn, As, Sb, Be, Fe, Mo, Cd, Sn, Ni and V in the rhizospheric soils are displayed in Table 1. The Tl contents in soils grown with distinct vegetables ranged from 0.38 to 1.54 mg/kg. More than half of the soil samples had high Tl contents and exceeded the soil background values of Guangdong Province (0.52 mg/kg) and China (0.58 mg/kg) (CEMS, 1990; Qi et al., 1992). The maximum Tl content was observed in the soil of S3, surpassing the maximum permissible level (MPL) set by the Canadian Council of Ministers of the Environment for agricultural land use (CCME, 1999) (Hitherto, China has no MPL towards Tl). The high Tl contamination generally occurred in the soils around the industrial area of the steel-making plant and other related factories. The higher Tl content in Soil S3 and S4 was mainly found in the rhizospheric soil samples near the Maba River located in the Beijiang River Basin. He (2016) reported that the content of Tl in the sediment of Maba river was in a range of 1.03–3.13 mg/kg (average 1.89 mg/kg), which was 1.78 times of the average Tl content in the background sediment from Guangdong Province (0.682 mg/kg). The minimum Tl content was found in the soil of S5, which was sampled from the remote forest area with little impact from the industrial activities.

The high contents of Pb, Mn, Cu, Zn, Be, Fe, Cd, Cr, Sn and V were also found in the soils, in the range of 68.5–236, 405–7000, 24.6–372, 184–605, 1.21–4.07, 2.69–9.07, 1.39–7.51, 50–1240, 4.50–52.3, 54.0–239 mg/kg, respectively. The concentration levels of Mn, Cr, Cu, Cd, Pb and As in soils were above the MPL of soil environmental quality standard set in China (SEP and GAQIQ, 2018). This suggested a high risk to agricultural crops and human health. It was also detected that the As content (average 42.9 mg/kg) was far beyond its background values of Guangdong Province (8.9 mg/kg) and China (12.1 mg/kg). In addition, the soil As contents in this study were higher than topsoil of some heavily industrialized cities worldwide, such as Idrija in Maribor (20.1 mg/kg) (Bavec et al., 2015), Novi Sad in Serbia (6.3 mg/kg) (Mihailović et al., 2015), Changchun in China (12.5 mg/kg) (Yang et al., 2011), respectively. This indicated that the As pollution in the local soil could not be overlooked. In addition, most of the contents of Co, Ni and Sb approached the risk screening values. Meanwhile, as shown in Fig. S1, there was a significant correlation in soil between Pb and Tl (0.83) As (0.88), Cd (0.75), Cu (0.79), Zn (0.98), Sb (0.79), Be (0.64) and Mo (0.70) (P < 0.01), suggesting that these elements are probably derived from the same source.

3.2. Contents of toxic elements in vegetables

The concentrations of toxic elements in different tissues of vegetables collected are shown in Table 2. The average Tl content (0.29 ± 0.01 mg/kg) of edible parts of all vegetables was higher than that of vegetables from a typical farmland protection areas in the Pearl River Delta region, Guangdong Province, China (0.051–0.13 mg/kg), areas without Tl contamination in Xiangquan city, Gansu Province, China, and areas without Tl contamination in China (0.01–0.06 mg/kg) (Zhou et al., 2007). As Tl threshold is not available in Chinese food guidelines, the German guidelines is adopted in this study. The Tl content of edible parts of a few vegetables exceeded Recommended Standards for Food and Feed of Germany (0.5 mg/kg) (Umweltqualität, 1998). These results indicated that the studied vegetables were moderately contaminated by Tl. Similar Tl contamination has been found in different plants in other countries. Heim et al. (2002) reported that Tl concentrations of the moss *Pleurozium schreberi* ranged from 0.04 to 0.13 μg/g in the Euro Region Neisse (including Germany, Czech Republic and Poland). Even higher Tl contamination was found in fodder of maize and rapea (Germany, up to 50 and 1095 mg/kg) and rape seeds (France, up to 33 mg/kg) (Tremel et al., 1997). These results indicate that Tl can be easily enriched in several agricultural products. Further explanation of the enrichment of Tl in the plants may be that Tl and K have a close geochemical affinity (Xiao et al., 2004). As previously reported, Tl can readily replace potassium in plants due to similar radius between Tl^+ (1.59 Å) and K^+ (1.51 Å) (Krasnodębska-Ostręga et al., 2012; Vaněk et al., 2019; Wei et al., 2020). Therefore, a competitive effect usually occurs between Tl^+ and K^+ during the Tl uptake (Hassler et al., 2007; Renkema et al., 2015; Siegel and Siegel, 1976), and Na^+/K^+–ATPase2 is indispensable in this substitution process (Liu et al., 2019b; Wang et al., 2013).

The contents of Pb, Cd, Cu, Mn, Ni, As, Sb, Zn, Be, Co, Cr, Fe, Mo, Sn and V of edible parts of all vegetables were in ranges of 3.79–25.7, 0.99–13.4, 11.5–79.6, 33.7–437, 4.09–53.3, 0.07–11.4, 0.10–5.74, 102–407, 0.01–0.20, 0.24–1.98, 18.0–887, 703–7211, 0.37–11.2, 0.17–3.96 and 0.90–12.7 mg/kg, respectively. The majority of contents of toxic elements exceeded the Chinese food standards MPL (Liu et al., 2019a; MHPRC, 2012). It should be noted that Pb, Cd, Cr, Zn As, Ni and Cu represented a high level of contamination. The average values of As, Pb, Cd, Cr, Zn even reached 7.4, 38.3, 22.8, 566 and 9.6 times of the MPL, respectively. Lower values of Pb (0.21 mg/kg), Cd (0.09 mg/kg), Cr (0.84 mg/kg), Zn (15.34 mg/kg) were reported in a previous study in kale irrigated by sewage in Machakos municipality in Kenya (Tomno et al., 2020). Moreover, much lower enrichment of As was found in edible *Nasturtium officinale* and *Diplazium esculentum* collected from an As contaminated area in Hawaii (0.57 and 0.075 mg/kg, respectively) (Falinski et al., 2014). The present results showed that there is a significant risk via vegetable ingestion. All these results indicated that high contents of toxic elements have been enriched in vegetables, and the potential risks of the vegetables should not be overlooked. Furthermore, Pb and As (0.66), Cu (0.55), Zn (0.66), Fe (0.65) (P < 0.01) and Co (0.66) (P < 0.05) also showed strong correlations in vegetable tissues (Fig. S2), which may share the common sources and pathways (Liu et al., 2017).

3.3. Contents of toxic elements in different vegetable tissues

The edible portions of vegetables were divided into the aboveground part (stalk and leave) and the underground part (root). The contents of studied elements (Tl, Pb, Cd, Cu, Mn, As, Sb, Zn, Be, Fe, Cr, Mo, Co, Sn, Ni and V) in different vegetable tissues (leaf, stalk and root) are presented in Fig. 2 and S3. The results showed that there were significant differences in toxic metal(loids) concentrations among different tissues of vegetables. Thallium in the majority of vegetables was mostly enriched in the roots. The reason may be that roots are the parts of the plant contacted physically with soil and surface water, where the anomalous high Tl content was retained. In general, Pb, Co, Cr, Cu, Fe,

Table 2
Tl and other toxic metal(loid)s contents(mg/kg dwt, dry weight) in the vegetables near the steel-making plant.

Site	Vegetables	Tl	Pb	Cd	Co	Cr	Cu	Mn	Ni	As	Sb	Zn	Be	Fe	Mo	Sn	V
S1	*Pachyrhizus erosus (Linn.) Urb*[a]	0.14 ± 0.01	21.2 ± 0.85	2.90 ± 0.12	1.98 ± 0.08	612. ± 24	37.8 ± 1.51	171 ± 6.86	53.3 ± 2.13	7.5 ± 0.30	0.56 ± 0.02	240 ± 9.59	0.17 ± 0.01	7211 ± 356	5.12 ± 0.15	2.63 ± 0.11	11.0 ± 0.45
	Ipomoea aquatica Forsk[a]	0.06 ± 0.00	11.4 ± 0.46	2.13 ± 0.09	0.97 ± 0.04	208.0 ± 8.3	11.8 ± 0.47	263 ± 10.5	12.1 ± 0.48	3.67 ± 0.15	5.74 ± 0.23	121 ± 4.82	0.04 ± 0.00	2678 ± 123	7.4 ± 0.23	1.47 ± 0.05	4.76 ± 0.22
	Lactuca sativa var[a] *longifoliaf. Lam*	0.12 ± 0.00	13.3 ± 0.53	3.29 ± 0.13	0.46 ± 0.02	163.8 ± 6.5	11.35 ± 0.45	67.3 ± 2.69	9.85 ± 0.39	2.00 ± 0.08	0.10 ± 0.00	173 ± 6.92	0.05 ± 0.00	1289 ± 56.0	3.85 ± 0.12	1.65 ± 0.05	1.96 ± 0.05
	Amaranthus tricolor L [a].	0.37 ± 0.01	12.0 ± 0.48	6.07 ± 0.24	0.41 ± 0.02	190.8 ± 7.6	21.8 ± 0.87	65.7 ± 2.63	7.18 ± 0.29	1.13 ± 0.05	0.12 ± 0.00	188 ± 7.53	0.1 ± 0.01	1432 ± 67.0	2.67 ± 0.10	3.96 ± 0.12	1.85 ± 0.05
	Beta vulgaris L [a].	0.19 ± 0.01	6.00 ± 0.24	1.70 ± 0.07	0.24 ± 0.01	110.0 ± 4.4	23.5 ± 0.94	33.7 ± 1.35	5.06 ± 0.20	1.03 ± 0.04	0.10 ± 0.00	120 ± 4.82	0.01 ± 0.00	703 ± 28.0	1.53 ± 0.05	1.9 ± 0.08	1.16 ± 0.05
	Brassica oleracea L [a].	0.06 ± 0.00	11.4 ± 0.46	2.13 ± 0.09	0.97 ± 0.04	208.0 ± 8.3	11.8 ± 0.47	263 ± 10.5	12.1 ± 0.48	3.67 ± 0.15	5.74 ± 0.23	120 ± 4.82	0.18 ± 0.01	5630 ± 256	0.86 ± 0.03	1.51 ± 0.05	12.7 ± 0.45
	Nelumbo nucifera[b]	0.09 ± 0.00	13.1 ± 0.52	1.60 ± 0.06	1.26 ± 0.05	121.2 ± 4.8	40.3 ± 1.61	437 ± 17.5	17.8 ± 0.71	11.4 ± 0.46	0.50 ± 0.02	114 ± 4.56	0.10 ± 0.00	5506 ± 256	0.56 ± 0.02	1.38 ± 0.05	4.80 ± 0.23
S2	*Lactuca sativa L. var. ramosa Hort* [a].	0.13 ± 0.01	25.7 ± 1.03	5.46 ± 0.22	0.82 ± 0.03	150 ± 5.98	31.0 ± 1.24	147 ± 5.89	12.6 ± 0.50	4.30 ± 0.17	0.46 ± 0.02	341 ± 13.6	0.04 ± 0.00	3120 ± 134	1.10 ± 0.05	2.43 ± 0.11	4.10 ± 0.14
	Allium fistulosum L [a].	0.14 ± 0.01	6.92 ± 0.28	0.99 ± 0.04	0.34 ± 0.01	18.0 ± 0.72	13.2 ± 0.53	48.6 ± 1.94	4.40 ± 0.18	4.24 ± 0.17	0.18 ± 0.01	102 ± 4.07	0.02 ± 0.00	890 ± 34.0	3.56 ± 0.14	0.68 ± 0.02	1.40 ± 0.04
	Lactuca sativa var longifoliaf. Lam[a]	0.14 ± 0.01	7.18 ± 0.29	1.85 ± 0.07	0.30 ± 0.01	70.9 ± 2.84	16.5 ± 0.66	124 ± 4.96	7.28 ± 0.29	2.64 ± 0.11	0.14 ± 0.01	122 ± 4.88	0.02 ± 0.00	1011 ± 45.0	2.00 ± 0.08	0.51 ± 0.01	1.00 ± 0.03
	Capsicum annuum L[c].	0.28 ± 0.01	5.82 ± 0.19	4.12 ± 0.15	0.81 ± 0.03	42.3 ± 1.5	11.1 ± 0.45	55.9 ± 1.8	3.02 ± 0.12	2.6 ± 0.11	0.19 ± 0.01	99.3 ± 4.2	0.04 ± 0.00	1950 ± 87.0	0.37 ± 0.02	0.63 ± 0.02	2.90 ± 0.12
S3	*Beta vulgaris L* [a].	0.48 ± 0.02	3.79 ± 0.15	5.03 ± 0.20	0.38 ± 0.02	57.5 ± 2.30	59.1 ± 2.36	57.1 ± 2.28	4.09 ± 0.16	1.53 ± 0.06	0.20 ± 0.01	223 ± 8.90	0.04 ± 0.00	916 ± 36.0	4.18 ± 0.13	0.17 ± 0.01	0.90 ± 0.03
	Sonchus lingiaus [a]	0.72 ± 0.03	17.6 ± 0.71	13.4 ± 0.54	1.70 ± 0.07	186 ± 7.44	79.6 ± 3.18	165 ± 6.61	10.4 ± 0.42	8.14 ± 0.33	0.94 ± 0.04	407 ± 16.2	0.20 ± 0.01	4220 ± 187	11.2 ± 0.45	0.92 ± 0.02	3.90 ± 0.13
S4	*Allium fistulosum L* [a].	0.92 ± 0.04	6.92 ± 0.28	2.74 ± 0.11	0.99 ± 0.04	710 ± 28.4	28.4 ± 1.14	97.6 ± 3.90	52.3 ± 2.09	0.07 ± 0.00	ND[d]	231 ± 9.25	0.06 ± 0.00	1211 ± 46.0	6.00 ± 0.20	2.1 ± 0.05	3.52 ± 0.14
	Cichorium endivia L [a]	0.13 ± 0.01	8.31 ± 0.33	10.1 ± 0.40	0.72 ± 0.03	108 ± 4.34	51.3 ± 2.05	77.3 ± 3.09	8.01 ± 0.32	3.60 ± 0.14	0.36 ± 0.01	211 ± 8.44	0.07 ± 0.00	1746 ± 57.0	9.65 ± 0.32	0.5 ± 0.02	1.90 ± 0.05
S5	*Sonchus lingiaus* [a]	0.18 ± 0.01	15.7 ± 0.63	5.02 ± 0.20	1.26 ± 0.05	887 ± 35.5	22.7 ± 0.91	152 ± 6.08	50.3 ± 2.01	1.72 ± 0.07	0.14 ± 0.01	190 ± 7.61	0.09 ± 0.00	2386 ± 112	10.4 ± 0.36	1.37 ± 0.05	4.32 ± 0.18
Minimum	0.06 ± 0.00	3.79 ± 0.15	0.99 ± 0.04	0.24 ± 0.01	18.0 ± 0.72	11.1 ± 0.45	33.7 ± 1.35	4.09 ± 0.16	0.07 ± 0.00	0.10 ± 0.00	99.3 ± 4.2	0.01 ± 0.00	703 ± 28.0	0.37 ± 0.02	0.17 ± 0.01	0.90 ± 0.03	
Maximum	0.92 ± 0.04	25.7 ± 1.03	13.4 ± 0.54	1.98 ± 0.08	887 ± 35.5	79.6 ± 3.18	437 ± 17.5	53.3 ± 2.13	11.4 ± 0.46	5.74 ± 0.23	407 ± 16.2	0.20 ± 0.01	7211 ± 356	11.2 ± 0.45	3.96 ± 0.12	12.7 ± 0.45	
Mean	0.29 ± 0.01	11.5 ± 0.46	4.56 ± 0.18	0.87 ± 0.03	283 ± 11.3	30.3 ± 1.21	124.4 ± 5.3	19.5 ± 0.82	3.7 ± 0.15	0.61 ± 0.03	191 ± 7.64	0.08 ± 0.00	2619 ± 131	4.4 ± 0.22	1.5 ± 0.07	3.90 ± 0.20	
MPL in food[e]	0.5	0.3	0.20	NG	0.5	10.0	NG	1.0	0.5	NG	20	NG[f]	NG	NG	250	NG	

[a] The edible parts are stems and leaves.
[b] The edible parts are stems, leaves and fruits.
[c] The edible parts are leaves.
[d] Not detectable.
[e] MPL: Maximum Permissible Level. Values for Tl and other metal(loid)s are taken from foods and foodstuffs recommended in Germany (Umweltqualität, 1998) and Ministry of Health of People's Republic of China (Umweltqualität, 1998; MHPRC, 2012), respectively.
[f] Not given.

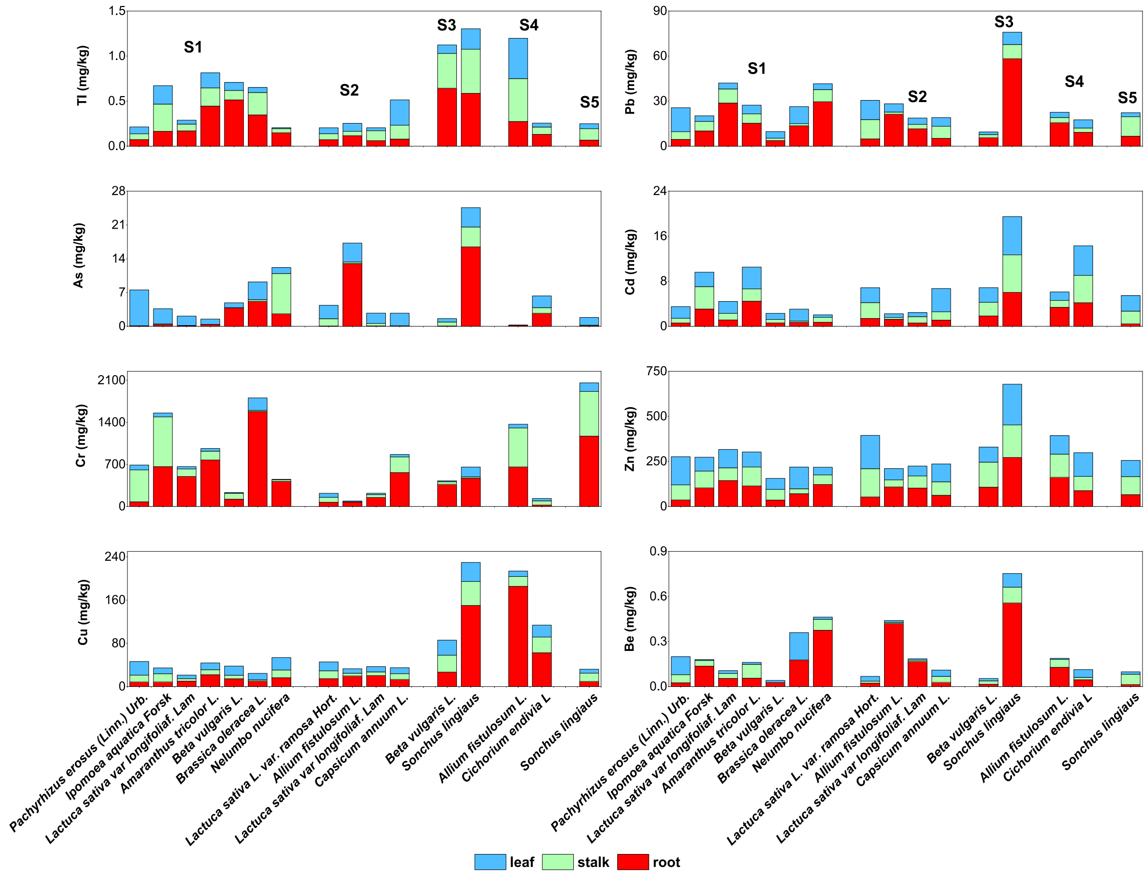

Fig. 2. Contents of thallium and related toxic metal(loid)s in separate organs of the vegetables.

Ni, Be, Sn and V were also enriched in the roots. In particular, the contents of Cr in vegetables' roots were significantly higher than those in above-ground parts. Generally, the excluders of plants and their related rhizospheric microbial composition might restrict the uptake and/or translocation of metals, maintaining low level of pollutants in the above-ground parts (Antoniadis et al., 2017a, 2017b). However, Cd and Zn were distributed relatively higher in aboveground parts (stalks and leaves) of vegetable. It may be related to tolerance of plants to Cd and Zn, which may influence the roots uptake and cell sequestration (Verbruggen et al., 2009). As reported, hyperaccumulators may translocate the toxic elements to the aboveground part to avoid roots being poisoned (Tian et al., 2011). It is worth noting that Be, As and Cu were fairly unevenly distributed in tissues among different vegetables and As concentrations in root of *Allium fistulosum L.* (S2) and *Capsicum annuum L.* (S2) were much higher than those of other vegetables. Overall, the process of uptake and transport is complex, depending on the type of vegetables and specific elements.

3.4. BCF and TF of studied toxic metal(loid)s in vegetables

As shown in Table 3, the BCF values of Tl followed the order: *Brassica oleracea L.* (S1) > *Ipomoea aquatica Forsk* (S1) > *Amaranthus tricolor L.* (S1) > *Sonchus lingiaus* (S3) > *Capsicum annuum L.* (S2) > *Beta vulgaris L.* (S1) > *Allium fistulosum L.* (S4) > *Beta vulgaris L.* (S3) > *Sonchus lingiaus* (S5) > *Lactuca sativa var longifoliaf. Lam* (S1) > *Lactuca sativa L. var. ramosa Hort.* (S2) = *Lactuca sativa L. var. ramosa Hort.* (S2) > *Nelumbo nucifera* (S1) > *Pachyrhizus erosus (Linn.) Urb.* (S1) > *Lactuca sativa var longifoliaf. Lam* (S2) > *Cichorium endivia L* (S4). Multitude BCF values of Tl were lower than 1, but cabbage (1.48), swamp cabbage (1.18) and amaranth (1.13) from S1 were greater than 1, suggesting a high bio-accumulation capacity of Tl in the root. All vegetables BCF values of As were much less than 1, which was lower than untreated sites in Florida (0.20–1.61) and higher than contanminted area in Yangtze River delta (0.002–0.053) (Cao et al., 2004; Mao et al., 2019). This indicated that the BCF was greatly influenced by soil properties and crop species. Most of the analyzed vegetables displayed the BCF magnitude of Cr, Ni, Fe, Cd and Mo exceeding 1, indicating their high enrichment by the studied vegetables. As shown in Table 4, the TF values of most vegetables for Tl, Cd, Cu, Zn, As, Mn, Ni, Be, Mo and Sn were higher than 1, suggesting that such elements in roots can be easily transferred to the aerial parts. Both the values of BCF and TF of Cd and Zn exceeded 1, which indicated that this vegetable had a strong accumulation of the two metals. This revealed that the metal(loid)s in the contaminated soil could be easily transferred from the root to the ground, entailing greater risks to local residents.

3.5. Health risk assessment of vegetables

The potential health effects from carcinogenic elements (As, Cr and Cd) and non-carcinogenic metal(loid)s (Ni, Tl, Co, Mn, Pb, Sb, Zn, Be, Sn, Fe, Cu, Mo and V) of the daily consumption of various vegetables were calculated based on CDI and HQ/cancer risks for local residents. As shown in Table S3 and Table 5, the CDI and HQ values for all the elements were in the following order: children > seniors > adults. It implied that children group were the most susceptible victims to the consumption of the vegetables. It should be noted that HQ value of Tl in children was generally higher than 10, which suggested children in this area were at risk of chronic Tl poisoning, although the concentration of Tl in vegetables was not exceptionally high as those reported elsewhere (Heim et al., 2002; Tremel et al., 1997; Xiao et al., 2004; Antoniadis et al., 2019). Studies have shown that even a low level of Tl can damage the function of liver, kidneys and heart muscle in children and hard to

Table 3
Bioconcentration factor (BCF) of vegetables near the steel-making plant.

Site	Vegetables	Tl	Pb	Cd	Co	Cr	Cu	Mn	Ni	As	Sb	Zn	Be	Fe	Mo	Sn	V
S1	*Pachyrhizus erosus (Linn.) Urb.*	0.40 ± 0.02	0.25 ± 0.01	1.31 ± 0.05	0.37 ± 0.01	0.61 ± 0.02	0.81 ± 0.03	0.03 ± 0.00	3.47 ± 0.14	0.22 ± 0.11	0.19 ± 0.01	1.01 ± 0.04	0.07 ± 0	845 ± 36	0.8 ± 0.04	0.22 ± 0.01	0.06 ± 0
	Ipomoea aquatica Forsk	1.18 ± 0.05	0.17 ± 0.01	2.42 ± 0.10	0.34 ± 0.01	1.26 ± 0.05	0.04 ± 0.02	0.03 ± 0.00	3.28 ± 0.13	0.11 ± 0.00	0.02 ± 0.00	0.80 ± 0.03	0.04 ± 0	715 ± 31	1.37 ± 0.07	0.17 ± 0.01	0.05 ± 0
	Lactuca sativa var longifoliaf. Lam	0.54 ± 0.02	0.37 ± 0.01	1.68 ± 0.07	0.19 ± 0.01	1.39 ± 0.06	0.51 ± 0.02	0.04 ± 0.00	1.43 ± 0.06	0.09 ± 0.00	0.02 ± 0.00	1.07 ± 0.04	0.03 ± 0	630 ± 21.5	2.59 ± 0.13	0.27 ± 0.01	0.06 ± 0
	Amaranthus tricolor L.	1.13 ± 0.05	0.15 ± 0.01	1.40 ± 0.06	0.16 ± 0.01	5.36 ± 0.21	0.80 ± 0.03	0.15 ± 0.01	1.83 ± 0.17	0.03 ± 0.00	0.03 ± 0.00	0.59 ± 0.02	0.06 ± 0	893 ± 39.1	4.65 ± 0.21	0.33 ± 0.02	0.09 ± 0
	Beta vulgaris L.	0.85 ± 0.03	0.05 ± 0.00	0.39 ± 0.02	0.07 ± 0.00	2.48 ± 0.10	0.63 ± 0.03	0.15 ± 0.01	0.64 ± 0.03	0.10 ± 0.00	0.08 ± 0.00	0.33 ± 0.01	0.02 ± 0	608 ± 30.4	1.06 ± 0.05	0.04 ± 0	0.03 ± 0
	Brassica oleracea L.	1.48 ± 0.06	0.32 ± 0.01	1.68 ± 0.07	0.54 ± 0.02	23.5 ± 0.94	0.86 ± 0.03	1.08 ± 0.04	2.34 ± 0.09	0.40 ± 0.02	2.25 ± 0.09	1.09 ± 0.04	0.27 ± 0.01	7324 ± 266	2.75 ± 0.12	0.48 ± 0.02	0.46 ± 0.02
	Nelumbo nucifera	0.47 ± 0.02	0.55 ± 0.02	1.35 ± 0.05	0.33 ± 0.01	7.77 ± 0.31	1.78 ± 0.07	1.47 ± 006	1.64 ± 0.07	0.56 ± 0.02	0.16 ± 0.01	1.28 ± 0.05	0.34 ± 0.02	9454 ± 372	3.37 ± 0.15	0.65 ± 0.03	0.25 ± 0.01
S2	*Lactuca sativa L. var. ramosa Hort.*	0.51 ± 0.02	0.44 ± 0.02	3.31 ± 0.13	0.10 ± 0.00	1.09 ± 0.04	1.60 ± 0.06	0.04 ± 0.00	0.93 ± 0.04	0.14 ± 0.01	0.14 ± 0.01	1.79 ± 0.07	0.02 ± 0	685 ± 31.1	0.75 ± 0.04	0.41 ± 0.02	0.02 ± 0
	Allium fistulosum L.	0.51 ± 0.02	0.35 ± 0.01	1.06 ± 0.04	0.32 ± 0.01	0.50 ± 0.02	1.32 ± 0.05	0.38 ± 0.02	0.37 ± 0.01	0.44 ± 0.02	0.29 ± 0.01	0.87 ± 0.03	0.26 ± 0.01	3276 ± 163	2.47 ± 0.1	0.4 ± 0.02	0.24 ± 0.01
	Lactuca sativa var longifoliaf. Lam	0.39 ± 0.02	0.21 ± 0.01	0.94 ± 0.04	0.09 ± 0.00	1.07 ± 0.04	1.37 ± 0.05	0.27 ± 0.01	0.39 ± 0.02	0.06 ± 0.00	0.03 ± 0.00	0.78 ± 0.03	0.06 ± 0	486 ± 23.4	1.22 ± 0.06	0.49 ± 0.02	0.02 ± 0
	Capsicum annuum L.	0.9 ± 0.04	0.2 ± 0.01	2.7 ± 0.12	0.1 ± 0.01	4.7 ± 0.23	1.3 ± 0.06	0.11 ± 0.01	1.4 ± 0.07	0.0 ± 0.00	0.0 ± 0.00	0.9 ± 0.04	0.06 ± 0	656 ± 32.8	2.75 ± 0.11	0.84 ± 0.03	0.05 ± 0
S3	*Beta vulgaris L.*	0.73 ± 0.03	0.06 ± 0.00	2.09 ± 0.08	0.08 ± 0.00	7.49 ± 0.30	0.28 ± 0.01	0.13 ± 0.01	2.04 ± 0.08	0.03 ± 0.00	0.02 ± 0.00	0.67 ± 0.03	0.01 ± 0	516 ± 25.8	0.96 ± 0.05	0.12 ± 0.01	0.05 ± 0
	Sonchus lingiaus	0.92 ± 0.04	0.38 ± 0.02	4.78 ± 0.19	0.38 ± 0.02	10.8 ± 0.43	0.62 ± 0.02	0.38 ± 0.02	1.89 ± 0.08	0.34 ± 0.01	0.35 ± 0.01	1.15 ± 0.05	0.21 ± 0.01	3947 ± 187	1.9 ± 0.08	0.23 ± 0.01	0.24 ± 0.01
S4	*Allium fistulosum L.*	0.80 ± 0.03	0.10 ± 0.00	1.57 ± 0.06	0.19 ± 0.01	26.3 ± 1.05	0.55 ± 0.02	0.28 ± 0.01	5.10 ± 0.20	0.003 ± 0.0	ND[a]	0.65 ± 0.03	0.05 ± 0	649 ± 32.4	1.05 ± 0.05	0.24 ± 0.01	0.12 ± 0.01
	Cichorium endivia L	0.23 ± 0.01	0.15 ± 0.01	10.3 ± 0.41	0.19 ± 0.01	2.65 ± 0.11	4.09 ± 0.16	0.22 ± 0.01	1.27 ± 0.05	0.13 ± 0.01	0.10 ± 0.00	1.21 ± 0.05	0.04 ± 0	808 ± 40.4	14.21 ± 0.67	0.08 ± 0	0.05 ± 0
S5	*Sonchus lingiaus*	0.66 ± 0.03	0.33 ± 0.01	3.33 ± 0.13	0.32 ± 0.01	28.2 ± 1.13	1.31 ± 0.05	0.53 ± 0.02	4.10 ± 0.16	0.09 ± 0.00	0.05 ± 0.00	1.37 ± 0.06	0.08 ± 0	2633 ± 130	12.81 ± 0.54	0.75 ± 0.04	0.2 ± 0.01

[a] Not detectable.

rehabilitate. Therefore, Tl contamination deserves greater attention and urgent management. Except Tl, the HQ values of non-carcinogenic metals (Pb, Co, Mn and Fe) mostly exceeded 1, particularly for Pb, which were all greater than 10 (even up to 713.57). The results implied that the consumption of locally grown vegetables may have a potential non-carcinogenic risk. Moreover, the carcinogenic risk of As, Cd and Cr was higher than 1×10^{-4}, which exceeded the limit of high risk value (Sun, et al., 2016), indicating that these vegetables have assignable potential carcinogenic risks. The results showed that the majority of collected vegetables could lead to both potential non-carcinogenic and carcinogenic risks for residents, and the consumption of vegetables is more harmful to children. Therefore, it is urgent to formulate practical measurements to safeguard the health of local residents in a better way.

3.6. Source apportionment by Pb isotopic tracing

As displayed in Fig. S2, positive correlations were shown between Pb and As (0.66), Cu (0.55), Zn (0.66), Fe (0.65), Be (0.85), Sb (0.51) and V (0.57) ($P < 0.01$). This revealed that these elements may have the same or similar pollution source. Besides, Zn, Pb, Cu, Fe and Be are typical contaminants from steel-making plant. The vegetable samples were collected from the soil surrounding a steel-making plant near a river, which had been received discharged water for decades. Therefore, the toxic elements of the vegetables may originate from the steel-making activities via fluvial transport. Relatively low correlations were found between in Pb and Tl (0.28), Cr (0.28), Cd (0.28), Sn (0.38), Ni (0.25), respectively, which suggested that these elements may have different sources compared with Pb. According to our previous studies, 6–88% of Tl in Gaofeng River, Yunfu came from the exploitation and utilization of pyrite (Liu et al., 2016) and 80–90% of Tl in the sediment profile near a Pb-Zn smelter in Shaoguan came from Pb-Zn smelting activities (Liu et al., 2019a). In order to further verify the sources of Pb and other elements with high correlation, Pb isotope analysis has been adopted. Fig. 3 exhibited the isotope graph ($^{206}Pb/^{207}Pb$ versus $^{208}Pb/^{206}Pb$) of vegetables in farmland S1, S2, S3, S4 and S5, smelting raw materials (Luo, 2019), and uncontaminated soils in the local area (Zhu et al.,

Table 4
Translocation factor (TF)) of vegetables near steel-making plant.

Site	Vegetables	Tl	Pb	Cd	Co	Cr	Cu	Mn	Ni	As	Sb	Zn	Be	Fe	Mo	Sn	V
S1	*Pachyrhizus erosus (Linn.) Urb.*	1.93 ± 0.08	4.70 ± 0.19	4.89 ± 0.20	5.48 ± 0.22	7.93 ± 0.32	4.42 ± 0.18	9.12 ± 0.36	5.99 ± 0.24	199 ± 7.97	ND[a]	6.60 ± 0.26	6.8 ± 0.3	20.84 ± 1	20.48 ± 1	2.5 ± 0.1	11 ± 0.4
	Ipomoea aquatica Forsk	3.08 ± 0.12	0.99 ± 0.04	2.14 ± 0.09	1.08 ± 0.04	1.35 ± 0.05	2.99 ± 0.12	2.84 ± 0.11	2.18 ± 0.09	10.0 ± 0.40	ND	1.64 ± 0.07	0.32 ± 0.01	0.7 ± 0.03	9.25 ± 0.3	0.84 ± 0.04	0.77 ± 0.04
	Lactuca sativa var longifoliaf. Lam	0.70 ± 0.03	0.46 ± 0.02	2.92 ± 0.12	0.58 ± 0.02	0.33 ± 0.01	1.14 ± 0.05	1.15 ± 0.05	0.46 ± 0.02	16.2 ± 0.65	ND	1.21 ± 0.05	0.93 ± 0.04	0.72 ± 0.04	1.38 ± 0.04	1.05 ± 0.05	0.34 ± 0.02
	Amaranthus tricolor L.	0.83 ± 0.03	0.79 ± 0.03	1.36 ± 0.05	0.33 ± 0.01	0.25 ± 0.01	1.00 ± 0.04	0.60 ± 0.02	0.15 ± 0.01	3.20 ± 0.13	ND	1.65 ± 0.07	1.96 ± 0.09	0.75 ± 0.04	0.41 ± 0.02	4.4 ± 0.22	0.33 ± 0.02
	Beta vulgaris L.	0.38 ± 0.02	1.61 ± 0.06	2.81 ± 0.11	0.54 ± 0.02	0.91 ± 0.04	1.63 ± 0.07	0.48 ± 0.02	0.37 ± 0.01	0.27 ± 0.01	0.37 ± 0.01	3.39 ± 0.14	0.37 ± 0.01	0.49 ± 0.01	3 ± 0.1	11.18 ± 0.4	0.86 ± 0.03
	Brassica oleracea L.	0.16 ± 0.01	0.84 ± 0.03	3.15 ± 0.13	0.22 ± 0.01	0.13 ± 0.01	1.13 ± 0.05	1.47 ± 0.06	0.23 ± 0.01	0.71 ± 0.02	10.4 ± 0.42	1.71 ± 0.07	1.03 ± 0.05	0.41 ± 0.01	1.27 ± 0.06	2.18 ± 0.11	0.75 ± 0.04
	Nelumbo nucifera	0.62 ± 0.02	0.44 ± 0.02	2.23 ± 0.09	0.67 ± 0.03	0.29 ± 0.01	2.47 ± 0.10	2.40 ± 0.10	0.70 ± 0.03	4.41 ± 0.18	ND	0.94 ± 0.04	0.26 ± 0.01	0.28 ± 0.01	0.17 ± 0.01	0.61 ± 0.03	0.4 ± 0.02
S2	*Lactuca sativa L. var. ramosa Hort.*	1.84 ± 0.07	5.27 ± 0.21	3.90 ± 0.16	3.77 ± 0.15	2.15 ± 0.09	2.13 ± 0.09	6.40 ± 0.26	1.18 ± 0.05	72.7 ± 2.91	ND	6.44 ± 0.26	1.96 ± 0.09	7.27 ± 0.3	3.14 ± 0.16	2.93 ± 0.12	3.73 ± 0.12
	Allium fistulosum L.	1.19 ± 0.05	0.32 ± 0.01	0.79 ± 0.03	0.09 ± 0.00	0.25 ± 0.01	0.67 ± 0.03	0.18 ± 0.01	0.60 ± 0.02	0.33 ± 0.01	0.18 ± 0.01	0.94 ± 0.04	0.05 ± 0	0.06 ± 0	2.68 ± 0.1	0.38 ± 0.02	0.05 ± 0
	Lactuca sativa var longifoliaf. Lam	2.35 ± 0.09	0.62 ± 0.02	3.11 ± 0.12	0.30 ± 0.01	0.48 ± 0.02	0.81 ± 0.03	2.79 ± 0.11	1.44 ± 0.06	28.4 ± 1.14	ND	1.19 ± 0.05	0.12 ± 0	0.6 ± 0.03	1.37 ± 0.05	0.1 ± 0.01	0.23 ± 0.01
	Capsicum annuum L.	0.9 ± 0.04	0.2 ± 0.01	2.7 ± 0.12	0.1 ± 0.01	4.7 ± 0.23	1.3 ± 0.06	0.11 ± 0.01	1.4 ± 0.07	0.0 ± 0.00	0.0 ± 0.00	0.9 ± 0.04	2.86 ± 0.12	2.73 ± 0.1	0.57 ± 0.03	1.04 ± 0.05	1.68 ± 0.07
S3	*Beta vulgaris L.*	0.75 ± 0.03	0.67 ± 0.03	2.74 ± 0.11	0.60 ± 0.02	0.16 ± 0.01	2.20 ± 0.09	1.13 ± 0.05	0.15 ± 0.01	23.2 ± 0.93	ND	2.08 ± 0.08	2.67 ± 0.13	1.07 ± 0.04	1.3 ± 0.05	0.14 ± 0.01	0.47 ± 0.02
	Sonchus lingiaus	1.22 ± 0.05	0.30 ± 0.01	2.23 ± 0.09	0.50 ± 0.02	0.40 ± 0.02	0.53 ± 0.02	0.99 ± 0.04	0.51 ± 0.02	0.49 ± 0.02	0.32 ± 0.01	1.49 ± 0.06	0.36 ± 0.01	0.39 ± 0.02	2.14 ± 0.1	0.5 ± 0.03	0.4 ± 0.02
S4	*Allium fistulosum L.*	3.37 ± 0.13	0.44 ± 0.02	0.81 ± 0.03	0.57 ± 0.02	1.08 ± 0.04	0.15 ± 0.01	0.72 ± 0.03	1.21 ± 0.05	0.40 ± 0.02	ND	1.43 ± 0.06	0.47 ± 0.02	0.67 ± 0.02	1.03 ± 0.05	1.45 ± 0.04	0.9 ± 0.05
	Cichorium endivia L	0.95 ± 0.04	0.90 ± 0.04	2.42 ± 0.10	1.53 ± 0.06	4.48 ± 0.18	0.82 ± 0.03	2.00 ± 0.08	0.08 ± 0.03	1.33 ± 0.05	0.83 ± 0.03	2.41 ± 0.10	1.15 ± 0.05	1.46 ± 0.05	2.99 ± 0.12	1.03 ± 0.02	1.33 ± 0.06
S5											ND						

(*continued on next page*)

2001). The graph of Pb isotopic compositions ($^{206}Pb/^{207}Pb$ versus $^{208}Pb/^{206}Pb$) indicated that the Pb isotopic signatures in the vegetable tissues showed a significant linear correlation ($r^2 = 0.81$) along with the signatures of steel-making raw materials and uncontaminated soils within the Pearl River Basin (Fig. 3). The ratios of lead isotopes from the five sampling sites were between the ratios of uncontaminated soil and the steel-making raw materials, indicating that the sources of lead pollution in the vegetable samples were mainly from the steel-making raw materials and geogenic-Pb. The values of isotopic ratios of $^{206}Pb/^{207}Pb$, $^{208}Pb/^{206}Pb$, $^{208}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$ were listed in Table S4. Absolute uncertainty errors (2-sigma) for $^{206}Pb/^{207}Pb$, $^{208}Pb/^{206}Pb$, $^{208}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$ were ±0.00005, ±0.0001, ±0.004, ±0.002 and ±0.003, respectively. The $^{206}Pb/^{207}Pb$ ratios of vegetable tissues ranged from 1.1695 to 1.1882, with an average of 1.1758 ± 0.000012, which were significantly lower than the $^{206}Pb/^{207}Pb$ values (1.1952) of the unpolluted soil in the Pearl River Basin (Zhu et al., 2001). It showed that vegetables were remarkably affected by anthropogenic activities. The $^{206}Pb/^{207}Pb$ ratios of most vegetable tissues were close to the $^{206}Pb/^{207}Pb$ ratio (1.1715) in the steel-making raw materials (Luo, 2019). This indicated that Pb in the vegetable tissues might be closely related to solid wastes generated by the steel-making activities.

To quantify the contribution of two end-members, as shown in Fig. 4, the results of the binary mixing model showed that the steel-making raw materials contributed 34.8–80.2% to the Pb contamination in vegetables from S1–S5. The results showed that the Pb and As of steel-making plant were important contaminant sources with high risks to vegetables because of the positive correlation between Pb and As (0.66). Notably, the vegetables of S3 (~2.9 km) and S4 (~3.0 km) of the downstream area revealed similar sources of Pb contamination of those of S1 (~0.5 km) and S2 (~0.5 km) where are closer to the steel-making plant. This indicated that steel-making activities have affected not only the vegetables adjacent the steel-making plant, but also farther downstream. The dispersion of contamination has increased the health risks to residents of the area. In addition, S5 was located ~3.6 km upstream, which lies within the natural protected water source area. Therefore, it was much less contaminated.

Table 5
Hazard quotient (HQ) of vegetables near the steel-making plant.

Site	Vegetables	Consumer	Tl	As	Cd	Co	Cr	Cu	Mn	Ni	Pb	Sb	Zn	Be	Fe	Mo	Sn	V
S1	*Pachyrhizus erosus (Linn.) Urb.*	Children	16.65 ± 0.83	713.57 ± 35.68	0.02 ± 0.00	7.80 ± 0.39	30.39 ± 1.52	1.12 ± 0.05	8.43 ± 0.42	3.15 ± 0.15	0.13 ± 0.00	2.90E-04 ± 0.00	0.94 ± 0.04	0.10 ± 0.00	12.17 ± 0.61	1.21 ± 0.06	5.18E-03 ± 0.00	2.60 ± 0.13
		Adults	3.94 ± 0.20	168.95 ± 8.45	0.00 ± 0.00	1.85 ± 0.09	7.20 ± 0.36	0.26 ± 0.01	2.00 ± 0.01	0.74 ± 0.04	0.03 ± 0.00	6.86E-05 ± 0.00	0.22 ± 0.01	0.02 ± 0.00	2.88 ± 0.14	0.29 ± 0.01	1.23E-03 ± 0.00	0.62 ± 0.03
		Seniors	7.08 ± 0.35	303.44 ± 15.17	0.01 ± 0.00	3.32 ± 0.16	12.92 ± 0.64	0.47 ± 0.03	3.59 ± 0.02	0.00 ± 0.00	0.06 ± 0.00	1.23E-04 ± 0.00	0.40 ± 0.02	0.04 ± 0.00	5.17 ± 0.25	0.51 ± 0.02	2.20E-03 ± 0.00	1.10 ± 0.05
	Ipomoea aquatica Forsk	Children	59.88 ± 2.99	339.76 ± 16.99	0.05 ± 0.00	4.87 ± 0.24	44.34 ± 2.21	0.76 ± 0.04	8.50 ± 0.43	3.23 ± 0.16	0.06 ± 0.00	4.31E-05 ± 0.00	0.67 ± 0.03	0.03 ± 0.00	4.52 ± 0.23	1.75 ± 0.09	2.89E-03 ± 0.00	1.13 ± 0.05
		Adults	14.18 ± 0.71	80.45 ± 4.02	0.01 ± 0.00	1.15 ± 0.06	10.50 ± 0.52	0.18 ± 0.00	2.01 ± 0.01	0.76 ± 0.04	0.01 ± 0.00	1.02E-05 ± 0.00	0.16 ± 0.00	0.01 ± 0.00	1.07 ± 0.05	0.41 ± 0.02	6.85E-04 ± 0.00	0.27 ± 0.01
		Seniors	25.46 ± 1.27	144.48 ± 7.22	0.02 ± 0.00	2.07 ± 0.1	18.85 ± 0.94	0.32 ± 0.02	3.62 ± 0.02	0.01 ± 0.00	0.03 ± 0.00	1.83E-05 ± 0.00	0.28 ± 0.01	0.01 ± 0.00	1.92 ± 0.09	0.74 ± 0.03	1.23E-03 ± 0.00	0.48 ± 0.02
	Lactuca sativa var longifoliaf. Lam	Children	14.05 ± 0.70	449.66 ± 22.48	0.02 ± 0.00	1.83 ± 0.09	8.13 ± 0.40	0.34 ± 0.02	3.31 ± 0.02	0.58 ± 0.03	0.04 ± 0.00	4.98E-05 ± 0.00	0.68 ± 0.03	0.03 ± 0.00	2.17 ± 0.11	0.91 ± 0.04	3.25E-03 ± 0.00	0.47 ± 0.02
		Adults	3.33 ± 0.17	106.47 ± 5.32	0.01 ± 0.00	0.43 ± 0.02	1.92 ± 0.09	0.08 ± 0.00	0.78 ± 0.04	0.14 ± 0.00	0.01 ± 0.00	1.18E-05 ± 0.00	0.16 ± 0.00	0.01 ± 0.00	0.51 ± 0.03	0.22 ± 0.01	7.69E-04 ± 0.00	0.11 ± 0.00
		Seniors	5.98 ± 0.30	191.21 ± 9.56	0.01 ± 0.00	0.78 ± 0.04	3.46 ± 0.18	0.14 ± 0.00	1.41 ± 0.07	0.00 ± 0.00	0.02 ± 0.00	2.12E-05 ± 0.00	0.29 ± 0.01	0.01 ± 0.00	0.92 ± 0.04	0.39 ± 0.02	1.38E-03 ± 0.00	0.20 ± 0.01
	Amaranthus tricolor L.	Children	43.70 ± 2.18	406.10 ± 20.31	0.04 ± 0.00	1.60 ± 0.08	9.47 ± 0.48	0.64 ± 0.03	3.23 ± 0.02	0.42 ± 0.02	0.02 ± 0.00	6.26E-05 ± 0.00	0.74 ± 0.40	0.06 ± 0.00	2.42 ± 0.12	0.63 ± 0.03	7.79E-03 ± 0.00	0.45 ± 0.02
		Adults	10.35 ± 0.52	96.15 ± 4.81	0.01 ± 0.00	0.38 ± 0.02	2.24 ± 0.11	0.15 ± 0.00	0.76 ± 0.04	0.10 ± 0.00	0.00 ± 0.00	1.48E-05 ± 0.00	0.18 ± 0.00	0.01 ± 0.00	0.57 ± 0.03	0.15 ± 0.00	1.85E-03 ± 0.00	0.11 ± 0.00
		Seniors	18.58 ± 0.92	172.69 ± 8.63	0.02 ± 0.00	0.68 ± 0.03	4.03 ± 0.20	0.27 ± 0.01	1.37 ± 0.07	0.01 ± 0.00	0.01 ± 0.00	2.66E-05 ± 0.00	0.32 ± 0.02	0.03 ± 0.00	1.03 ± 0.05	0.27 ± 0.01	3.31E-03 ± 0.00	0.19 ± 0.00
	Beta vulgaris L.	Children	22.91 ± 1.15	202.56 ± 10.13	0.01 ± 0.00	0.94 ± 0.04	5.46 ± 0.27	0.69 ± 0.03	1.66 ± 0.08	0.30 ± 0.01	0.02 ± 0.00	5.03E-05 ± 0.00	0.47 ± 0.02	0.01 ± 0.00	1.19 ± 0.06	0.36 ± 0.02	3.74E-03 ± 0.00	0.28 ± 0.01
		Adults	5.42 ± 0.27	47.96 ± 2.40	0.00 ± 0.00	0.22 ± 0.01	1.29 ± 0.05	0.16 ± 0.00	0.39 ± 0.02	0.07 ± 0.00	0.00 ± 0.00	1.19E-05 ± 0.00	0.11 ± 0.00	0.00 ± 0.00	0.28 ± 0.01	0.09 ± 0.00	8.85E-04 ± 0.00	0.07 ± 0.00
		Seniors	9.74 ± 0.48	86.14 ± 4.31	0.01 ± 0.00	0.40 ± 0.02	2.32 ± 0.11	0.29 ± 0.01	0.71 ± 0.04	0.00 ± 0.00	0.01 ± 0.00	2.14E-05 ± 0.00	0.20 ± 0.01	0.00 ± 0.00	0.50 ± 0.47	0.15 ± 0.00	1.59E-03 ± 0.00	0.12 ± 0.00
	Brassica oleracea L.	Children	6.61 ± 0.33	384.67 ± 19.21	0.02 ± 0.00	3.81 ± 0.15	10.32 ± 0.05	0.35 ± 0.02	12.94 ± 0.65	0.71 ± 0.03	0.07 ± 0.00	2.95E-03 ± 0.00	0.47 ± 0.02	0.11 ± 0.00	9.50 ± 0.47	0.20 ± 0.01	2.97E-03 ± 0.00	3.00 ± 0.15
		Adults	1.57 ± 0.08	91.08 ± 4.55	0.00 ± 0.00	0.90 ± 0.04	2.44 ± 0.12	0.08 ± 0.00	3.06 ± 0.15	0.17 ± 0.00	0.02 ± 0.00	6.98E-04 ± 0.00	0.11 ± 0.00	0.03 ± 0.00	2.25 ± 0.11	0.05 ± 0.00	7.04E-04 ± 0.00	0.71 ± 0.03
		Seniors	2.81 ± 0.14	163.58 ± 8.18	0.01 ± 0.00	1.62 ± 0.08	4.39 ± 0.22	0.15 ± 0.00	5.50 ± 0.28	0.00 ± 0.00	0.03 ± 0.00	1.25E-03 ± 0.00	0.20 ± 0.01	0.05 ± 0.00	4.04 ± 0.20	0.09 ± 0.00	1.26E-03 ± 0.00	1.28 ± 0.06
	Nelumbo nucifera	Children	10.98 ± 0.55	440.52 ± 22.03	0.01 ± 0.00	4.94 ± 0.25	6.01 ± 0.30	1.19 ± 0.05	21.54 ± 1.01	1.05 ± 0.05	0.20 ± 0.00	2.57E-04 ± 0.00	0.45 ± 0.02	0.06 ± 0.00	9.29 ± 0.46	0.13 ± 0.00	2.72E-03 ± 0.00	1.13 ± 0.05
		Adults	2.60 ± 0.13	104.30 ± 5.22	0.00 ± 0.00	1.17 ± 0.05	1.42 ± 0.07	0.28 ± 0.02	5.10 ± 0.25	0.25 ± 0.01	0.05 ± 0.00	6.09E-05 ± 0.00	0.11 ± 0.00	0.01 ± 0.00	2.20 ± 0.10	0.03 ± 0.00	6.43E-04 ± 0.00	0.27 ± 0.01
		Seniors	4.67 ± 0.23	187.33 ± 9.37	0.00 ± 0.00	2.10 ± 0.15	2.56 ± 0.13	0.51 ± 0.03	9.16 ± 0.45	0.00 ± 0.00	0.09 ± 0.00	1.09E-04 ± 0.00	0.19 ± 0.00	0.02 ± 0.00	3.95 ± 0.20	0.06 ± 0.00	1.16E-03 ± 0.00	0.48 ± 0.02
S2	*Lactuca sativa L. var. ramosa Hort.*	Children	15.59 ± 0.78	865.52 ± 43.28	0.04 ± 0.00	3.24 ± 0.16	7.41 ± 0.37	0.92 ± 0.05	7.24 ± 0.36	0.75 ± 0.04	0.08 ± 0.00	2.38E-04 ± 0.00	1.34 ± 0.07	0.03 ± 0.00	5.26 ± 0.26	0.26 ± 0.01	4.78E-03 ± 0.00	0.97 ± 0.05
		Adults	3.69 ± 0.18	204.93 ± 10.25	0.01 ± 0.00	0.77 ± 0.03	1.75 ± 0.09	0.22 ± 0.01	1.72 ± 0.01	0.18 ± 0.00	0.02 ± 0.00	5.63E-05 ± 0.00	0.32 ± 0.02	0.01 ± 0.00	1.25 ± 0.06	0.06 ± 0.00	1.13E-03 ± 0.00	0.23 ± 0.01
		Seniors	6.63 ± 0.33	368.05 ± 18.40	0.02 ± 0.00	1.38 ± 0.07	3.15 ± 0.16	0.39 ± 0.02	3.08 ± 0.15	0.00 ± 0.00	0.03 ± 0.00	1.01E-04 ± 0.00	0.57 ± 0.03	0.01 ± 0.00	2.24 ± 0.11	0.11 ± 0.00	2.03E-03 ± 0.00	0.41 ± 0.02
	Allium fistulosum L.	Children	16.30 ± 0.81	233.50 ± 11.68	0.01 ± 0.00	1.35 ± 0.07	0.89 ± 0.04	0.39 ± 0.02	2.39 ± 0.12	0.26 ± 0.01	0.08 ± 0.00	9.45E-05 ± 0.00	0.40 ± 0.02	0.01 ± 0.00	1.50 ± 0.07	0.84 ± 0.04	1.34E-03 ± 0.00	0.33 ± 0.02
		Adults	3.86 ± 0.19	55.29 ± 2.76	0.00 ± 0.00	0.32 ± 0.02	0.21 ± 0.01	0.09 ± 0.00	0.57 ± 0.03	0.06 ± 0.00	0.02 ± 0.00	2.24E-05 ± 0.00	0.09 ± 0.00	0.00 ± 0.00	0.36 ± 0.02	0.20 ± 0.01	3.17E-04 ± 0.00	0.08 ± 0.00
		Seniors	6.93 ± 0.34	99.29 ± 4.96	0.00 ± 0.00	0.57 ± 0.03	0.38 ± 0.02	0.17 ± 0.00	1.02 ± 0.05	0.00 ± 0.00	0.03 ± 0.00	4.02E-05 ± 0.00	0.17 ± 0.01	0.01 ± 0.00	0.64 ± 0.03	0.36 ± 0.02	5.69E-04 ± 0.00	0.14 ± 0.00

Site	Vegetables	Consumer	Tl	Pb	Cd	Co	Cr	Cu	Mn	Ni	As	Sb	Zn	Be	Fe	Mo	Sn	V
	Lactuca sativa var longifoliaf. Lam	Children	17.01±0. 85	242.28±12.11	0.01±0. 00	1.19±0. 06	3.52 ±0.18	0.49±0. 03	6.10 ±0.35	0.43±0. 02	0.05 ±0.00	7.19E-05±0.00	0.48±0. 02	0.01 ±0.00	1.71±0.09	0.47 ±0.02	1.00E-03±0.00	0.24 ±0.01
		Adults	4.03±0. 20	57.36±2.92	0.00±0. 00	0.28±0. 01	0.83 ±0.41	0.12 ±0.00	1.44 ±0.07	0.10 ±0.00	0.01 ±0.00	1.70E-05±0.00	0.11 ±0.00	0.00.±0.00	0.40 ±0.02	0.11 ±0.00	2.38E-04±0.00	0.06 ±0.00
		Seniors	7.23±0. 36	103.03±5.15	0.01±0. 00	0.51 ±0.03	1.50±0. 07	0.21±0.01	2.59 ±0.13	0.00 ±0.00	0.02 ±0.00	3.06E-05±0.00	0.20±0. 01	0.00 ±0.00	0.73 ±0.04	0.20 ±0.01	4.27E-04±0.00	0.10 ±0.00
	Capsicum annuum L.	Children	33.19±0.17	196.39±9.82	0.03±0. 00	3.18±0. 16	2.10±0. 10	0.33±0.02	2.75 ±0.14	0.18 ±0.00	0.05 ±0.00	9.50E-05±0.00	0.39±0. 02	0.02 ±0.00	3.29 ±0.16	0.09 ±0.00	1.24E-03±0.00	0.68 ±0.03
		Adults	7.86±0. 39	46.50±0. 23	0.01±0.00	0.75±0. 04	0.50±0. 03	0.08±0. 00	0.65 ±0.03	0.04 ±0.00	0.01 ±0.00	2.25E-05±0.00	0.09±0.00	0.01 ±0.00	0.78±0.04	0.02 ±0.01	2.94E-04±0.00	0.16 ±0.01
		Seniors	14.11±0.71	83.51±0. 42	0.01±0. 00	1.35 ±0.07	0.89±0. 05	0.14 ±0.07	1.17 ±0.06	0.01 ±0.00	0.02 ±0.00	4.04E-05±0.00	0.17 ±0.00	0.01 ±0.00	1.40 ±0.07	0.04 ±0.02	5.27E-04±0.00	0.29 ±0.01
S3	*Beta vulgaris L.*	Children	56.69 ±2.83	127.89±6.39	0.04±0.00	1.49±0.07	2.85±0.14	1.74±0.09	2.81±0.14	0.24±0.01	0.03±0.00	1.08E-04±0.00	0.88±0.04	0.02±0.00	1.55±0.08	0.99±0.05	3.35E-04±0.00	0.21 ±0.01
		Adults	13.42±0.67	30.28±1.51	0.01 ±0.00	0.35±0.02	0.68±0.03	0.41±0.02	0.67±0.03	0.06±0.00	0.01±0.00	2.55E-05±0.00	0.21±0.01	0.01±0.00	0.37±0.02	0.23±0.01	7.92E-05±0.00	0.05 ±0.00
		Seniors	24.11±1.21	54.38±2.72	0.02±0.00	0.63±0.03	1.21±0.06	0.74±0.04	1.19±0.06	0.01±0.00	0.01±0.00	4.59E-05±0.00	0.37±0.02	0.01±0.00	0.66±0.03	0.42 ±0.02	1.42E-04±0.00	0.09 ±0.00
	Sonchus lingiaus[a] *Sonchus lingiaus*[a]	Children	84.56±2.43	594.90±29.75	0.10 ±0.00	6.70±0.33	9.23±0.46	2.35±0.12	8.13±0.41	0.62±0.03	0.15±0.00	4.81E-04±0.00	1.60±0.08	0.12±0.00	7.12±0.36	2.65±0.13	1.81E-03±0.00	0.92 ±0.05
		Adults	20.02±1.00	140.85±7.04	0.02±0.00	1.59±0.08	2.19±0.11	0.56±0.03	1.93±0.09	0.15±0.00	0.03±0.00	1.14E-04±0.00	0.38±0.02	0.03±0.00	1.69±0.08	0.63±0.03	4.29E-04±0.00	0.22 ±0.01
		Seniors	35.96±1.80	252.97±12.65	0.04±0.00	2.85±0.14	3.93±0.19	1.00±0.05	3.46±0.17	0.02±0.00	0.06±0.00	2.04E-04±0.00	0.68±0.03	0.05±0.00	3.03±0.15	1.13±0.05	7.70E-04±0.00	0.39 ±0.02
S4	*Allium fistulosum L.*	Children	109.13±5.46	233.64±11.68	0.02±0.00	3.89 ±0.19	35.24±1.71	0.84±0.04	4.80±0.24	3.09±0.15	0.00±0.00	0.00E+00±0.00	0.91±0.04	0.04±0.00	2.04±0.10	1.41±0.07	4.13E-03±0.00	0.83 ±0.04
		Adults	25.84 ±1.44	55.32±2.77	0.00±0.00	0.92±0.05	8.34±0.42	0.20±0.01	1.14±0.05	0.73±0.03	0.00±0.00	0.00E+00±0.00	0.22±0.01	0.01±0.00	0.48±0.02	0.33±0.02	9.79E-04±0.00	0.20 ±0.01
		Seniors	46.40±2.32	99.35±4.97	0.01±0.00	1.65±0.08	14.98±0.75	0.36±0.02	2.04±0.01	0.02±0.00	0.00±0.00	0.00E+00±0.00	0.39±0.02	0.02±0.00	0.87±0.04	0.60± 0.03	1.76E-03±0.00	0.35 ±0.02
	Cichorium endivia L	Children	14.76±0.74	280.41±14.02	0.07 ±0.00	2.83±0.14	5.38±0.27	1.51± 0.08	3.80±0.19	0.47±0.02	0.06±0.00	1.86E-04±0.00	0.83±0.04	0.04±0.00	2.95±0.15	2.28±0.12	9.84E-04±0.00	0.45 ±0.02
		Adults	3.50±0.17	66.39±3.32	0.02 ±0.00	0.67±0.03	1.27±0.06	0.36±0.02	0.90±0.04	0.11±0.00	0.02±0.00	4.40E-05±0.00	0.20±0.01	0.01±0.00	0.70±0.04	0.54±0.03	2.33E-04±0.00	0.11 ±0.00
		Seniors	6.28±0.31	119.24 ±5.96	0.03±0.00	1.21±0.06	2.29±0.11	0.64±0.03	1.62±0.08	0.00±0.00	0.03±0.00	7.90E-05±0.00	0.35±0.02	0.02±0.00	1.25±0.07	0.97 ±0.05	4.19E-04±0.00	0.19 ±0.00
S5	*Sonchus lingiaus*[a]	Children	21.61±1.08	529.53±26.48	0.04 ±0.00	4.98±0.25	44.03±2.20	0.67±0.03	7.48±0.37	2.97±0.15	0.03 ±0.00	7.34E-05±0.00	0.75±0.04	0.05±0.00	4.03±0.20	2.45±0.12	2.70E-03±0.00	1.02 ±0.05
		Adults	5.12±0.25	125.38±6.27	0.01±0.00	1.18±0.06	10.42±0.52	0.16±0.00	1.77±0.09	0.70±0.03	0.01±0.00	1.74E-05±0.00	0.18±0.00	0.01±0.00	0.95±0.05	0.58±0.03	6.38E-04±0.00	0.24 ±0.01
		Seniors	9.19 ±0.46	225.18±11.26	0.02±0.00	2.12±0.10	18.72±0.94	0.28±0.01	3.18±0.16	0.00±0.00	0.01±0.00	3.12E-05±0.00	0.32±0.02	0.02±0.00	1.71±0.08	1.04±0.05	1.15E-03±0.00	0.43 ±0.02

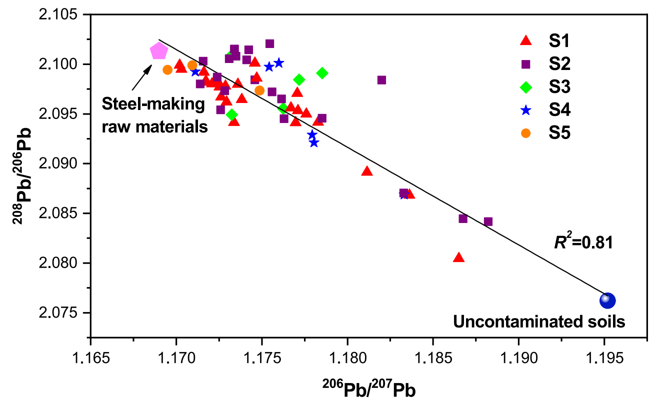

Fig. 3. Three isotope graph ($^{206}Pb/^{207}Pb$ versus $^{208}Pb/^{206}Pb$) displaying the distribution of the isotopic compositions of studied vegetables, uncontaminated soils from the studied area and the steel-making raw materials.

4. Conclusions

Besides the vegetable grown soils surrounding a steel-making plant were extremely enriched with traditional contaminants such as Pb, Cd, Cr and Cu, notably, highly toxic metal(loid)s of Tl and As were also observed to be remarkably accumulated. The results of TF and BCF indicated that high enrichment capacity of Tl was found in vegetables like *Brassica oleracea L., Ipomoea aquatica Forsk and Amaranthus tricolor L.* The CDI and HQ values calculated for residents at different ages indicated that vegetable consumption posed a potential health risk under Tl exposure, which cannot be ignored, especially for children. Carcinogenic risk was higher than the threshold for both As, Cd and Cr. Results of Pb isotopic fingerprinting with a simple binary model suggested that the average contribution of steel-making activities to contamination of most metals in vegetables from the studied sites (S1, S2, S3, S4 and S5) were 75.7%, 80.0%, 70.0%, 80.2%, and 34.8%, respectively. This further quantitatively confirms that the toxic elements contamination was predominantly ascribed to the steel-making activities. This work highlights that the areas near the steel-making plant may be enriched with non-expected toxic pollutants (i.e. Tl and As). Also, our results reveal that the steel-making activities contributed 35–80% of Pb and As in vegetables via a binary mixing model. The findings indicate that (1) the steel-making plant should be received with special attention due to emerging risks of the toxic metal(loid)s in the nearby soil-crops system, and (2) the present study site may serve as a model region to monitor and compare the pollution features surrounding the steel-making or related activities in other nation.

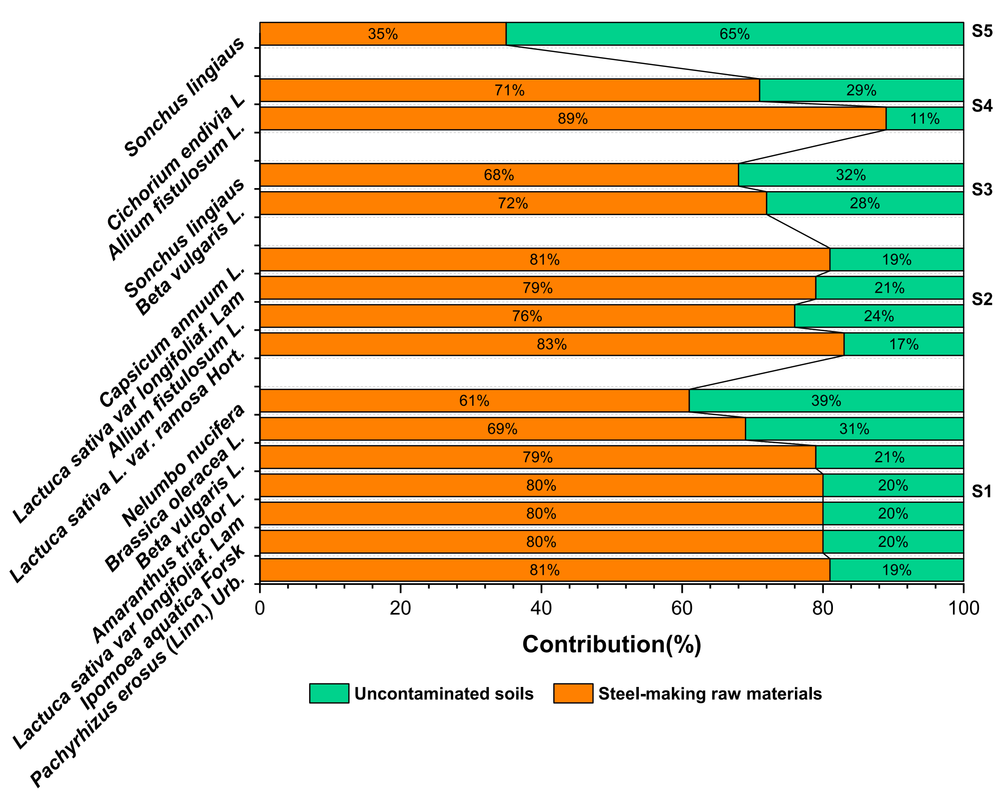

Fig. 4. Source apportionment of toxic metal(loid)s contamination in different vegetables by Pb isotopic tracer analysis according to the binary mixing model.

CRediT authorship contribution statement

Jin Wang: Writing - original draft, Writing - review & editing, Project administration. **Lulu Wang:** Formal analysis, Investigation, Writing - original draft. **Yuxuan Wang:** Methodology, Writing - review & editing. **Daniel C.W. Tsang:** Writing - review & editing. **Xiao Yang:** . **Jingzi Beiyuan:** Writing - review & editing. **Meiling Yin:** Writing - review & editing. **Tangfu Xiao:** Writing - review & editing. **Yanjun Jiang:** . **Wenli Lin:** . **Yuchen Zhou:** . **Juan Liu:** Conceptualization, Writing - review & editing, Funding acquisition. **Liang Wang:** Funding acquisition. **Min Zhao:** .

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.

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Appendix A. Supplementary material

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