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Low presence of potentially toxic elements in Singapore urban garden soils

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Abstract

Background: Urban agriculture is potentially an important piece of the food security puzzle for a rapidly growing urban world population. Community gardening is also promoted as a safe and viable form of exercise for aging populations in crowded settings where opportunities to participate in other action activities may be limited. Knowledge of potential site-specific health risks to environmental contaminants is important in dialogues promoting urban farming.

Methods: We assess the pseudo-total concentrations of selected potentially toxic elements (PTEs) in the soils of community gardens, public parks, and woodlands in the tropical urban island nation of Singapore. We compare concentrations of cadmium, copper, lead, and zinc with amalgamated risk guidelines to form a baseline understanding of the level of contamination in these spaces. We also perform provenance tracking with lead isotopes to identify potential sources of contaminants.

Results: All pseudo-total concentrations of Cd, Cu, Pb, and Zn in the soil were below threshold concentrations considered to represent substantial risk. Further, PTE concentrations in gardens were largely equivalent to those found in community parks and woodlands, but the geographical distribution varied. Provenance tracking with Pb isotopes indicated Pb in gardens was both anthropogenic and natural, but spatially variable. The lack of strong spatial clustering of areas with the highest PTE concentrations was inconsistent with a common point source of contamination. However, the correlation between Cu and Zn suggest a common source for these elements, such as road/trafficking or atmospheric deposition.

Conclusion: We find limited risk of urban gardeners to exposure to Cd, Cu, Pb, and Zn—elements that are commonly abundant in urban settings with dense transportation networks and substantial industrial activities. The low levels of PTEs are encouraging for the promotion of urban farming for food production and leisure in this dense urban setting. However, as concentrations were low, we did not assess bioavailability and bioaccessibility of the PTEs. These assessments would need to be determined in cases of with higher levels of contamination to provide a more thorough consideration of actual human risk.

Keywords: Urban agriculture, Green spaces, Parks, Woodlands, Environmental risk assessment, Heavy metals

Background

Urban gardening risks

Urban agriculture activities are increasingly being

incorporated into the planning of cities worldwide (Taguchi and Santini 2019; Edmondson et al. 2020; Langemeyer et al. 2021), with urban hubs such as Hanoi, Havana, New York, and Singapore being examples of productivism (Hou 2017; Martin-Moreau and Ménascé 2020). Embedded within UN Sustainable Development Goal 11.3 is the importance of agriculture as part of sustainable cities

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(Mansfield and Mendes 2013; McClintock et al. 2018): “countries should aim to work to enhance inclusive and sustainable urbanization for participatory, integrated, and sustainable human settlement planning and management”. Nearly a decade ago, Orsini et al. (2013) recognized that the food security benefit of urban agriculture was evidenced by 100–200 million people providing fresh horticultural goods to city markets. Arguably urban agriculture in developing countries plays a critical role in food availability and affordability, but there is need for improved understanding potential risks and limitations (De Bon et al. 2010; Zezza and Tasciotti 2010; Hamilton et al. 2014). While urban agriculture may not achieve food self-sufficiency per se, it is an alternative way of “feeding citizens differently” (Martin-Moreau and Ménascé 2020). Urban gardening is also a practical means of exercise, leisure, therapy, and perhaps supplementary income, in crowded urban spaces for all citizens (Orsini et al. 2013; Kalantari et al. 2018; Rogge et al. 2018; Winkler et al. 2019). Regardless of economic setting, it is increasingly important to preform foundational work addressing the potential realm of risks that await the growing wave of urban farmers, many of which may be advanced in age, and therefore in some cases, susceptible to the adverse effects of contaminated urban soils (Beckie and Bogdan 2010; Hamilton et al. 2014; Yang and Na 2017).

Pollution research worldwide has revealed the potential for people engaged in outdoor urban activities to be exposed to various PTEs that originate from past and present anthropogenic sources such as transportation networks, energy production systems, mining and industrial activities, and improper handling of waste (McLaughlin et al. 2000; Mitchell et al. 2014; Jean-Soro et al. 2015; Spliethoff et al. 2016; Bechet et al. 2018; López et al. 2019). Elevated PTE concentrations in urban soils are often legacies of past activities conducted before a garden was established (Kessler 2013; Wortman and Lovell 2013). While some elements such as copper and zinc are essential in very small amounts for normal health, they may be toxic when high bioavailable fractions are encountered (De Vries et al. 2007).

Exposure to high concentrations of PTEs can cause a variety of health problems including dizziness, nausea, cramping, coughing, diarrhea, and vomiting (Jaishankar et al. 2014; Gautam et al. 2016). Long-term and repeated ingestion may induce element-specific maladies such as hypertension and weakening of bone density (Cd), renal dysfunction (Pb, Cd), anemia (Zn), and other afflictions (ATSDR 2015; Satarug and Moore 2004; Swaddiwudhipong et al. 2011; Cabral et al. 2015). The potential for adverse health risks stemming from a seemingly healthy and sustainable activity is one aspect

of the “dark side of urban gardening” (Kessler 2011; Mancebo 2016)—however, others conclude the risk is sometimes more perceived than real (Brown et al. 2016; Lupolt et al. 2021). Nevertheless, assessing the potential risk is beneficial to promoting healthy outdoor activities.

Human urban gardening exposure chain

Community risk to exposure to potentially toxic elements (PTEs) while working in urban gardens has been a topic of concern for more than half a century (Chaney et al. 1984). Lead in the USA during the 1960s and 1970s, for example, was found in very high concentrations owing to contamination from a variety of sources including automotive exhaust, lead-based paint degradation, smelter emissions, sewage sludge, pesticides, and element-enriched manure fertilizers (Preer and Rosen 1977; Spittler and Feder 1979). Similarly, Purves (1966) found urban garden soils in Edinburgh and Dundee to be elevated in copper and boron, probably as a result of atmospheric deposition of soot, which was also applied as a garden soil amendment. Warren et al. (1971) later reported elevated concentrations of Cd, Cu, Pb, and Zn to be a growing concern for vegetable gardening in urban and industrial areas of Canada. Over the years, dozens of studies have examined the levels of these and other potentially harmful elements in urban and peri-urban soils worldwide (Table 1). The high element concentrations in the soils of many of the latter studies was often linked to legacy activities (e.g., Pb from paint; Cu, Pb, Zn from smelters and mines), trafficking phenomena (e.g., vehicle emissions, vehicle and tire wear), deposition following the release from a variety of urban and industrial sources, irrigation with wastewater, inter alia (Table 1; Additional file 1: Table S1).

These prior works have demonstrated that the “human urban exposure chain” to PTEs in the soil is a complex issue involving behavioral, biochemical, developmental, geophysical, physiological, and societal variables (cf. Clark et al. 2006; Schram-Bijkerk et al. 2018). To frame our research, which is a baseline assessment of Cd, Cu, Pb, and Zn in the soils of urban gardens in Singapore, we portray this exposure chain as a complex dynamic system. In the following narrative, the italicized terms are key variables in a simplified causal loop diagram shown in Fig. 1.

Element concentrations in the soil are influenced by several factors: mineralogy and geochemical composition of the parent rock, organic element content, particle size distribution, soil horizonation, drainage, vegetation, management, and anthropogenic inputs (Kabata-Pendias 1993; Wuana and Okieiman 2011). The elevated presence of a PTE in an urban soil could be naturally part of the background *geogenic signature* or associated with

Table 1 Ranges of Cd, Cu, Pb, and Zn concentrations reported for surface soils in (peri)urban garden soils worldwide (mg/kg; total/pseudo-total concentrations)

Location; garden type	Cd	Cu	Pb	Zn	Issues mentioned	References
Belo Horizonte, MG, Brazil; urban gardens	0.11–0.20 ^a	16–42 ^a	11–30 ^a	ND	Proximity to potential sources	Dala-Paula et al. (2018)
Jos, Nigeria; cultivated soils	0.08–0.19	1–4	3–5	8–13	Urban ash (deposition, soil amendment)	Pasquini and Alexander (2004)
Nanjing, China; peri-urban ag soils	0.08–0.15 ^a	21–30 ^a	31–33 ^a	53–71 ^a	Deposition; industrial effluent	Huang et al. (2006)
Salamanca, Spain; urban gardens	0.07–0.33	ND	10–26	ND	Fertilizers (Cd); trafficking (Pb)	Sanchez-Camazano et al. (1994)
Visakhapatnam, India; semi-urban	ND	11–31 ^a	15–48 ^a	43–50 ^a	Proximity to industry	Srinivas et al. (2009)
Alice, S Africa; home garden soils	0.10–0.80 ^a	5–8 ^a	5–14 ^a	18–53 ^a	Minor contamination	Bvenura and Afolayan (2012)
Toronto; community gardens	0.27–0.40	22–33	19–96	ND	Trafficking processes	Wiseman et al. (2013)
Guangzhou, China; urban veg gardens	0.02–1.9	6–25	25–55	57–195	Atmospheric deposition	Xiong et al. (2016)
Namyangju, Korea; urban field near road	bdl	15–30	14–18	61–77	Inconclusive; some road effects	Kim et al. (2017)
Thessaloniki, Greece; Peri-urban farm plots	0.16–0.26	18–40	16–37	3–124	Proximity to industry was NOT an issue	Vousta et al. (1996)
Wuxi, China; peri-urban ag soils	0.14–0.17 ^a	35–37 ^a	38–42 ^a	104–112 ^a	Deposition; industrial effluent	Huang et al. (2006)
Rostock, Germany; urban garden soils	0.05–0.60	1–59	3–57	3–95	Diffuse; highest in city center	Kahle (2000)
Sakai, Osaka; (peri)urban veg farms	~0.25–0.90 ^a	~10–30 ^a	~10–35 ^a	~30–125 ^a	Redistribution of legacy metal; trafficking	Komai and Yamamoto (1982)
Skarzysko-Kamienna, Poland; allot. gardens	0.30–5.8	3–15	7–33	2–99	Inconclusive; probably industrial	Świercz and Zajęcka (2018)
Kabul, Afghanistan; urban gardens	0.15–0.20	50–66	25–33	113–184	Traffic, war, contaminated irrigation water	Safi and Buerkert (2011)
New Brunswick, CA; residential gardens	~0.02–0.05	~18–34	~27–115	~77–145	Industrial legacy; long-term deposition	Pilgrim and Schroeder (1997)
Baoding, China; sewage-irrigated soils	0.17–0.40	26–44	27–58	113–250	Sewage irrigation water	Xue et al. (2012)
Szeged (Bakto), Hungary; urban gardens	0.27–2.86	19–580	5–61	33–199	Traffic; pesticides	Szolnoki et al. (2013)
Jiangsu cities (China); peri-urban ag soils	ND	38–45 ^a	88–120 ^a	45–47 ^a	Industrial, urban processes	Hao et al. (2009)
Manitoba province, CA; urban ag soils	0.8–1.7	16–37	24–43	62–110	Road phenomena most likely	Mills & Zwarich (1975)
Beijing, China; gardens and green spaces	0.14–0.24 ^a	28–50 ^a	30–74 ^a	78–118 ^a	Age, traffic processes, urban sources	Beavington (1975)
Torun, Poland; botanical garden topsoil	bdl	bdl–30	bdl–148	6–142	Natural soil genesis	Charzyński et al. (2018)
Cowra (Au); commercial/residential gardens	0.01–0.80	1–96	7–12	9–336	No main pollution sources	Kachenko and Singh (2006)
Beijing (Tongzhou); irrigated soils	0.41–1.71	22–49	48–53	136–176	Waste-water irrigation	Khan et al. (2008)
Varnasi city, India; suburban garden soils	0.90–5.65	3–30	9–25	22–208	Waste-water irrigation	Sharma et al. (2006)
Koszalin, Poland; allotment soils	0.04–0.53 ^a	8–34 ^a	8–57 ^a	78–220 ^a	Natural signature; garden management	Bielicka-Giełdoń et al. (2013)
Singapore; urban gardens	bdl–3.2	10–77	bdl–37	71–164	Proximity to industry/roads & industry; diffuse urban signal	This study
Mumbai, India; railway gardens	0.1–1.1	60–231	3–7	15–111	Irrigation water; pesticides; railway signal	Vazhacharickala et al. (2013)

Table 1 (continued)

Location; garden type	Cd	Cu	Pb	Zn	Issues mentioned	References
Ho Chi Minh City; urban ag lands	ND	8–70	bdl–27	bdl–272	Amendments (fert/pest); urban waste	Tran et al. (2020)
Vancouver, Ca; community garden	ND	36–63	22–82	51–133	Legacy deposition, roads, industry	Oka et al. (2014)
Kano City, Nigeria; urban veg gardens	2.3–4.0	5–14	2–17	17–233	Irrigation wastewater	Abdu et al. (2011)
Oregon (Corvallis, Portland); food gardens	bdl–0.57	25–109	0.2–347	ND	Not determined; compost concerns	Nelson (2018)
Seoul, Korea; rooftop gardens	0.9–1.9	6–118	6–21	51–268	Deposition (local and long-distance)	Kim et al. (2015a, b)
Shangdong towns (China); garden soils	0.03–0.99	1–160	9–27	25–223	Trafficking; industry; waste irrigation	Liu et al. (2011)
Zurich; urban gardens	ND	16–34	19–1076	ND	Legacy (esp. Pb); deposition	Tresch et al. (2018)
Nanjing, China; peri-urban ag soils	0.19–0.58	22–58	21–150	76–181	Fertilizers; deposition; industry emissions	Hu et al. (2018)
Guelph, Ontario, CA; Community gardens	0.38–1.84	ND	16–151	97–503	Low-income area; legacy	Montaño-López and Biswas (2021)
Kaduna, Nigeria; river bank gardens	0.07–0.95	11–36	26–154	53–218	Deposition; vehicular emissions	Agbenin et al. (2009)
Lisbon area; urban allotment soils	ND	10–62	1–110	33–146	Uncertain; roads/airport likely	Leitão et al. (2018)
Nanjing; urban/suburban gardens	ND	23–72	35–84	100–283	Decreases from urban to rural	Fang et al. (2011)
Kunming, China; peri-urban garden	0.53–0.80	87–208	42–62	77–148	Geogenic; manure inputs; uncertain	Zu et al. (2014)
Bragança, Portugal; peri-urban gardens	0.9 ± 0.2 ^a	128 ± 28 ^a	56 ± 78 ^a	89 ± 28 ^a	Organic farming; new city sources	Arrobas et al. (2017)
Dar es Salaam, Tanzania; urban ag	0.15–0.26 ^a	4–8 ^a	9–33 ^a	13–57 ^a	Uncertain; contaminated irrigation water	Kibassa et al. (2013)
Delhi, India; peri-urban soils	2.28–2.51 ^a	24–77 ^a	21–59 ^a	73–175 ^a	Wastewater irrigation, fertilizer, deposition	Bhatia et al. (2015)
Kayseri, Turkey; urban veg gardens	1–48–2.11 ^a	44–68 ^a	67–108 ^a	128–179 ^a	Not determined	Tokalioglu et al. (2006)
Danang, Vietnam; urban ag/ garden soils	0.1–0.7	30–495	1–5	53–562	Air-borne pollutants; ag amendments	Thuy et al. (2000)
Johannesburg, SA; school veg gardens	ND	bdl–80	27–100	91–427	Proximity to pollution sources	Kootbodien et al. (2012)
Hanoi, Vietnam; urban farm soils	3.9–4.5	195–230	125–145	187–263	Contaminated irrigation water	Nguyen et al. (2010)
Harare, Zimbabwe, metropolitan farms	1.0–3.4	21–94	17–59	42–228	Irrigation with wastewater	Mapanda et al. (2005)
Hyderabad, Pakistan; unclear	1.6–4.3 ^b	11–32	21–67	105–209	Wastewater irrigation	Jamali et al. (2007)
Kano City, Nigeria; garden soil	1.4–9.6 ^b	ND	16–91 ^a	77–246	Inconclusive; irrigation water suspected	Egwu and Agbenin (2013)
Tacoma, Washington, USA; urban gardens	0.30–0.80 ^a	32–118 ^a	38–110 ^a	100–219 ^a	Uncertain; past management effects	McIvor et al. (2012)
Melbourne; urban garden soil	0.12–1.04	ND	13–773	ND	House age; distance to roads	Kandic et al. (2019)
Lisbon, Portugal; urban allotment gardens	bdl	5–87	23–245	35–208	Industry; traffic phenomena	Bechet et al. (2018)
Hue City, Vietnam; peri-urban farms	0.03–0.44	3–39	4–25	12–212	Irrigation water; flood disposition	Pham et al. (2021)
Ottawa, CA; garden soils	0.11–0.75	6–42	16–547	50–380	Age; limited industrial activity	Rasmussen et al. (2001)
Hangzhou; urban–rural vegetable soils	0.10–0.64	14–78	20–87	59–265	Agrichemicals; vehicles exhaust; industry	Chen et al. (2008)
Xianyang (China); suburban garden soil	0.2–2.45	6–50	24–132	40–526	Plastic, fertilizers, traffic, industry	Wang et al. (2018)

Table 1 (continued)

Location; garden type	Cd	Cu	Pb	Zn	Issues mentioned	References
Hong Kong area; market garden soils	0.25–2.17	5–67	24–113	20–232	Urbanization and traffic volume	Wong (1996)
Plock city, Poland; allotment gardens	0.41 [2.12]+	9 [38]+	9 [24]+	313 [2947]+	Dust fall (Industry, transportation)	Mikula and Indeka (1997)
Perth, Australia; former market gardens	0.02–0.66	2–68	3–174	6–304	Construction; roads; waste disposal	Rate (2021)
Wuxi, China; peri-urban ag soils	bdl–0.14	27–152	17–162	30–445	Trafficking, industry, manufacturing	Zhao et al. (2007)
Connecticut; community gardens	bdl	20–75 ^a	22–450 ^a	60–238 ^a	Not identified specifically	Stilwell et al. (2008a)
Wollongong, Australia; home gardens	1.99±0.3	847±114	13.3±3.1	229±17	Proximity to Cu smelter	Beavington (1975)
Taiwan; urban park soils++	bdl–3.33	9–117	8–57	49–379	Industry sources	Jien et al. (2011)
Guangdong, China; vegetable gardens	0.26–1.17	210–450	73–134	92–142	E-waste processing	Luo et al. (2011)
Kaduna, Nigeria; urban field gardens	ND	18–85	160–200	156–433	Wastewater irrigation; waste disposal	Akpa and Agbenin (2012)
Naples, Italy; urban gardens	ND	~20–85	~75–325	~50–450	Traffic, Industry, ag amendments	Imperato et al. (2003)
Vienna, Austria; buried pots in gardens	~0.10–0.75	ND	~5–165	74–2600	Great variability reported	Ziss et al. (2021)
Southampton (UK); urban horticulture soils	0.14–0.80	19–86	34–488	60–485	Geology; mixed (indust, petrol, burning)	Crispo et al. (2021)
Adelaide, Australia; urban ag sites	0.01–0.38	25–183	30–268	<1–662	Legacy (industry)	Salomon et al. (2020)
Vancouver, CA; urban gardens/farms	bdl	35–127 ^a	69–208 ^a	77–887 ^a	Not explored	Thomas (2013)
Nantes, France; urban allotment gardens	0.1–17.7	13–117	19–229	26–272	Industry; traffic phenomena	Bechet et al. (2018)
Bottrop, Germany; variety of urban gardens	~1.8–2.7	~15–145	~50–155	~125–295	Inconclusive	Burghardt and Schneider (2018)
Zagreb, Croatia; urban/suburban ag	0.25–3.85	4–183	1–139	15–277	Multiple anthropogenic processes	Romic and Romic (2003)
Harrisburg, PA (USA); potential gardens	ND	11–46 ^a	26–402 ^a	30–216 ^a	Legacy Pb; road phenomena	Kessler et al. (2013)
Madrid, Spain; community gardens	ND	11–59	15–598	72–309	Landuse history; proximity to urban center	Izquierdo et al. (2015)
Île-de-France, France; market gardens	0.55–0.99	81–156	229–436	231–367	Legacy (industry, sludge disposal)	Barbillon et al. (2019)
Cotorro, Havana; peri-urban gardens	ND	50–945	21–731	91–7739	Wind direction (smelter)	Díaz Rizo et al. (2015)
Torino, Italy; gardens and parks	ND	40–293	17–565	75–550	Diffuse pollution	Hursthouse et al. (2004)
Baton Rouge, LA, USA; peri-urban	ND	6–189	11–967	32–1031	Industry, construction, traffic	Weindorf et al. (2012)
Sheffield, UK; gardens/allotments	ND	~45–180	~70–600	~185–750	Varies by land use; e legacy metals	Weber et al. (2019)
Gyongyosoroszim, Hungary; veg gardens	0.22–13.60	ND	21–694	97–2050	Flood water from old Zn/Pb mine	Sipter et al. (2008)
Hangzhou, China; suburb veg gardens	ND	4–144	14–98	76–302	Organic manuring	Wuzhong et al. (2004)
Porto, Portugal; urban ag/farm soils	0.1–1.1	10–248	27–415	92–465	Airport; oil refinery; roads	Cruz et al. (2014)
Madrid; parks and gardens; other	ND	18–194	31–463	82–514	Waste sludge; fertilizers; roads; other	De Miguel et al. (1998)
Nates, France; community garden	ND	16–151 ^a	28–5785 ^a	32–193 ^a	Anthropogenic and geogenic	Jean-Soro et al. (2015)

Table 1 (continued)

Location; garden type	Cd	Cu	Pb	Zn	Issues mentioned	References
Nova Scotia; community/yard gardens	ND	11–108	10–767	42–350	Various potential anthropogenic sources	Heidary-Monfared (2011)
Washington DC; urban farm/garden	ND	13–159	16–869	32–607	Legacy and urban/trafficking sources	Long (2010)
Liverpool (UK); urban horticulture soils	0.19–2.2	27–166	76–514	94–497	Geology; mixed (industry, petrol, burning)	Crispo et al. (2021)
Leicester (UK); urban horticulture soils	0.25–4.8	27–110	76–454	127–614	Geology; mixed (industry, petrol, burning)	Crispo et al. (2021)
Columbus and Cleveland; established gardens	0.93–4.15	23–101	31–553	97–645	Legacy (Pb paint); prior activities	Kaiser et al. (2015)
Ghent, Belgium; urban gardens	0.16–4.91	11–107	55–385	39–1325	Road phenomena; general industry signal	Folens et al. (2017)
Hong Kong; urban parks	0.02–5.89	5–190	5–404	39–435	Age; amendments (fertilizers), road dust	Li et al. (2001)
Sevilla, Spain; parks/ag/gardens	0.18–4.85	14–198	23–725	39–326	Trafficking; organic amendments	Ruiz-Cortes et al. (2005)
Newcastle (UK); urban horticulture soils	0.43–1.1	25–246	82–501	185–629	Geology; mixed (industry, petrol, burning)	Crispo et al. (2021)
Montreal, CA; urban gardens	0.4–1.8 ^a	15–400 ^a	150–400 ^a	210–300 ^a	Compost; highly variable	Murray et al. (2011)
Buffalo & NYC, NY; urban farms/gardens	BDL–3.6	40–83	18–3620	136–1060	Deposition; non-uniform contamination	McBride et al. (2014), Cai et al. (2016)
Wales & England; urban soils only	0.4–1.9	4–75	14–680	6–744	Trafficking, industry, mining, other	Davies (1978)
Dublin area; garden soils	> 3–5	11–145	39–540	117–940	Wind direction from urban sources	Fleming and Parle (1977)
Sevilla, Spain; allotments and parks	ND	14–698	14–1080	38–611	Vehicular sources	Hursthouse et al. (2004)
Leeds (UK); urban horticulture soils	0.24–2.5	33–372	104–1682	139–503	Geology; mixed (industry, petrol, burning)	Crispo et al. (2021)
Milton Keynes (UK); urban horticulture soils	0.21–6.5	19–228	29–3943	89–413	Geology; mixed (industry, petrol, burning)	Crispo et al. (2021)
Bangkok, Thailand; general urban soils++	0.05–2.53	5–283	12–269	3–814	Soil type; urban processes; roads	Wilcke et al. (1998)
Brescia, Italy; home gardens	bdl–4.00	28–410	36–207	58–845	Proximity to industry	Ferri et al. (2015)
Connecticut; comm and res gardens	bdl–3.5	bdl–900	bdl–3490	15–520	Not identified specifically	Stilwell et al. (2008b)
Los Angeles; urban gardens	0.11–4.27	ND	18–1720	ND	Proximity to roads; age of neighborhood	Clarke et al. (2015)
Edinburgh (UK); urban horticulture soils	0.30–2.4	33–103	84–1229	138–996	Geology; mixed (industry, petrol, burning)	Crispo et al. (2021)
Nottingham (UK); urban horticulture soils	0.54–4.6	42–127	124–1019	235–862	Geology; mixed (industry, petrol, burning)	Crispo et al. (2021)
Quezon City, Philippines; cropped soils	ND	84–784	300–6550	bdl–1817	Urban processes; landfill legacy	Navarrete et al. (2017)
Melbourne (AU); residential veg gardens	ND	bdl–323	bdl–3341	bdl–3003	Age (e.g., for lead); road/rail proximity	Laidlaw et al. (2018)
Mexico City; garden, park & other soils	ND	15–398	5–452	36–1641	Trafficking/roads; Industrial sources	Morton-Bermea et al. (2009)
Sydney (AU); urban veg gardens	ND	33–717	14–3080	50–2020	Inner city has high lead legacy	Rouillon et al. (2017)
Wroclaw, Poland; allotment gardens	ND	13–595	13–659	38–2103	Proximity to pollution sources	Kabala et al. (2009)
Chicago; various urban gardens/farms	ND	9–19 ^a	34–449	38–69 ^a	Not substantiated	Witzling et al. (2010)
Christchurch; urban gardens	0.33–10.7	12–118	23–2615	68–799	Age (pre-1950 neighborhoods)	Ashrafzadeh et al. (2018)

Table 1 (continued)

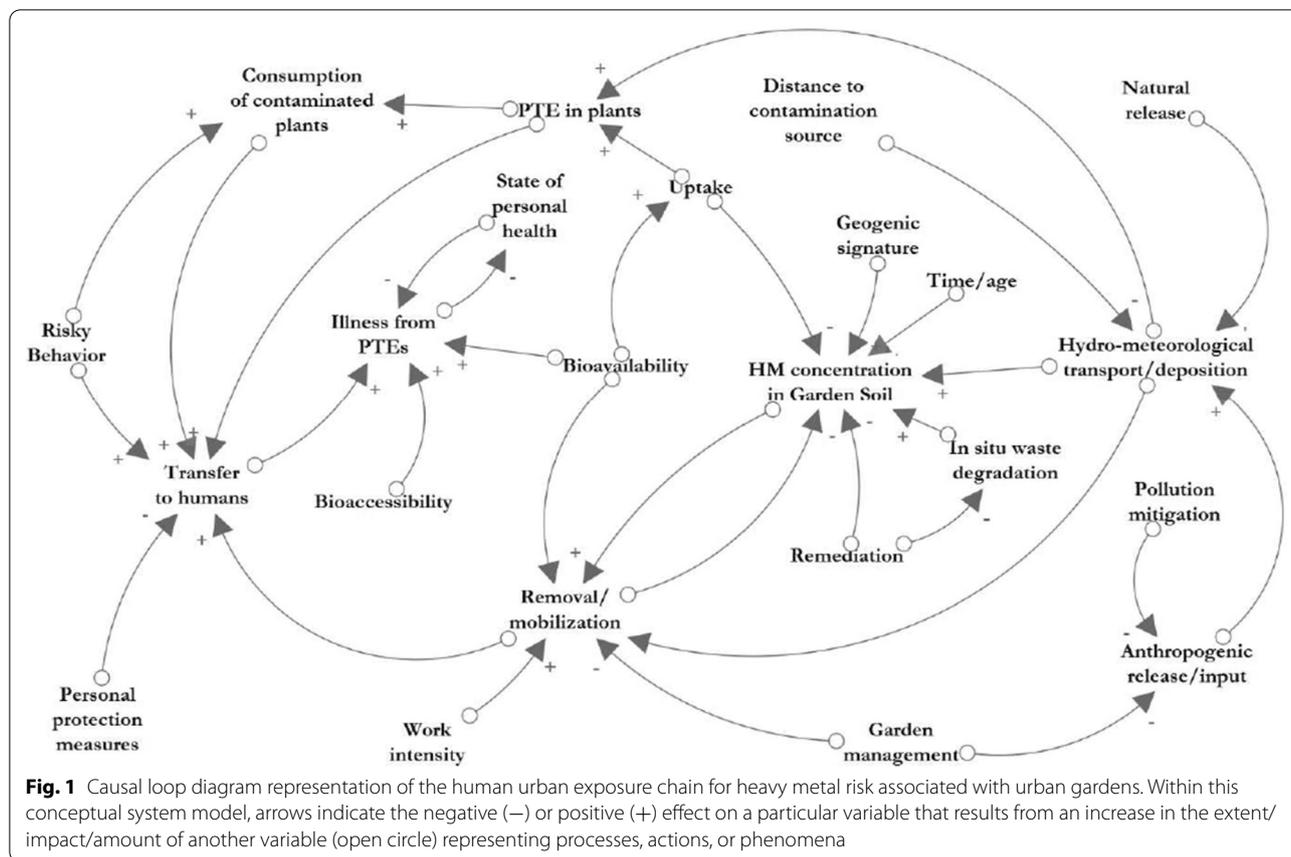
Location; garden type	Cd	Cu	Pb	Zn	Issues mentioned	References
Cardiff (UK); urban horticulture soils	0.15–4.3	10–216	30–2149	46–1213	Geology; mixed (industry, petrol, burning)	Crispo et al. (2021)
Bristol (UK); urban horticulture soils	0.46–3.3	14–752	88–1211	115–960	Geology; mixed (industry, petrol, burning)	Crispo et al. (2021)
Copenhagen, Denmark; urban gardens	~0.3–3.7 ^a	~10–450 ^a	~10–600 ^a	~40–1260 ^a	Inconclusive; urban signal	Warming et al. (2015)
Gijón (Asturias, Spain); peri-urban soils	0.4–2.3	6–244	33–585	64–1047	Coal combustion; unidentified sources	Boente et al. (2017)
Marseille, France; veg gardens	0.16–8.86	14–473	20–566	43–1020	Chemical fertilizers, fungicides	Zhong et al. (2022)
New York (USA); community gardens	bdl–3.1	6–598	11–2455	21–2317	Legacy Pb; long-term urban processes	Mitchell et al. (2014)
Baltimore, MA, (USA); urban gardens	0.02–13.65	1–97	1–10,900	<1–4880	Legacy (inner city); urban activities	Mielke et al. (1983)
Paris area (France); Ag + urban gardens	0.90–15.0	ND	42–854	100–1292	Deposition legacy (mining, smelting)	Pelfrène et al. (2012)
Baltimore (USA); urban farms/gardens	ND	~20–300	~7–1000	~50–600	Not determined; produce was safe	Lupolt et al. (2021)
Brooklyn, NY; urban gardens	ND	53–328 ^a	66–1025 ^a	150–1474 ^a	Legacy urban pollutants	Egendorf et al. (2018)
San Francisco (USA); garden soils	bdl–8.14	2–2630	7–2040	33–707	Home age	Gorospe (2012)
Washington DC; urban gardens	0.2–6.6	8–240	44–5300	74–2600	Proximity to homes (esp. Pb); variable	Preer et al. (1984)
Noyelles-Godault (France); kitchen gardens	2.8–21.1	ND	190–1425	199–2429	Former mining/smelter area	Douay et al. (2013)
Paris area (France); kitchen garden soils	17.2–23.9 ^a	ND	387–1708 ^a	2350–2871 ^a	Garden amendments; ag practices	Waterlot et al. (2011)
Kampala City, Uganda; peri-urban ag	0.12–21.0	26–1690	10–653	49–3270	Inappropriate waste disposal	Nabulo et al. (2012)
Boolaroo (Australia); residential veg garden	0.01–27.49	3–223	12–1626	45–4014	Former Pb–Zn smelter area	Kachenko and Singh (2006)
Port Kemba (Australia); residential veg garden	0.01–7.01	65–1032	14–430	87–1947	Former Cu smelter area	Kachenko and Singh (2006)
Palmerton, PA (USA); garden soils	18–113	41–595	90–370 ^a	1420–13,100	Proximity to Zn smelter	Chaney et al. (1988)
Glasgow, Scotland; allotments and parks	ND	52–484	250–7051	164–1004	Diffuse pollution	Hursthouse et al. (2004)
Hong Kong; roadside gardens/parks	ND	13–553	42–1034	173–11,316	Road pollution	Tam et al. (1987)
New York (USA); urban gardens	0.1–11.0	5–1286	3–8912	35–2352	Legacy (lead; incineration); pesticides	Cheng et al. (2015)
New Orleans, LA (USA); urban ag	0.25–8.80	2–200	1–9540	18–7330	Legacy (Pb); current road/industry	Moller et al. (2018)
Newcastle, Australia; private spaces	3.0–9.0	44–1040	42–11,600	140–8570	Legacy Cu; smelting (slag waste)	Harvey et al. (2017)
Baia Mare, Romania, (peri) urban gardens	1.2–13.8	171–2130	797–10,200	376–2130	Legacy and deposition (mining, smelter)	Mihali et al. (2012)
Dalnégorsk, Russia; garden/other soils	7–26	29–805	213–5621	593–5438	Mining, smelter, industry	Von Braun et al. (2002)
London Boroughs (UK); garden-veg plot soils	bdl–68	8–2320	28–13,700	34–2780	Home age	Culbard et al. (1988)

^a Pertains to a range of means or a mean ± standard deviation; otherwise, entries are ranges (min–max)

~Values are approximated from a graph

+Signals that values are means and maximums

++Not an agriculture study, but in close proximity to Singapore



enhancements caused by *natural processes/sources* such as volcanic eruptions, groundwater seepage, and fluvial transport (Briffa et al. 2020; Müller et al. 2020). Often, however, enrichment occurs when contaminants are *transported* to and *deposited* at a site from *anthropogenic sources* by atmospheric (related to wind and dry deposition) and hydrologic (e.g., rainfall, surface runoff, waste water irrigation) dispersal pathways, as well as the degradation of discarded waste or applied chemicals, including compost, fertilizers and disease/pest control agents that are applied in *garden management* (Kelly et al. 1996; Holt 2000; Wuana and Okiemen 2011; Su et al. 2022).

While contamination may be acute, being associated with only a few pollution events (e.g., accidental spill/release; flooding), it is often chronic, stemming from an accumulation over longer periods time (He et al. 2013; Ashraf et al. 2014). Thus, *time* or *age* are critical factors. For example, the soils associated with old homes/neighborhoods often have elevated concentrations of some elements (inner city legacy), for example Pb from paint used to protect wooden and metal construction materials (Culbard et al. 1988; Li et al. 2001; Mitchell et al. 2014; Clarke et al. 2015; Rouillon et al. 2017; Kandic et al. 2019; Montaña-López and Biswas 2021). Gardens

built on the former sites of long-running smelters, mines, factories, and waste dumps often are enriched in selected elements, representing a deposition legacy long after disuse (Kachenko and Singh 2006; Nabulo et al. 2012; Pelfrène et al. 2012; Douay et al. 2013). Garden soils may become enriched over time from chronic atmospheric deposition (Kargar et al. 2013; Laidlaw et al. 2018). Thus, a new fill soil may not reflect the long-term pattern of contamination. Long-term application of metal-based amendments can also elevate PTEs in garden soils.

Gardens that are close in *distance* to mines, power generation plants, incineration facilities, factories, and manufacturing centers tend to receive higher pollution loads, but long-range dispersal is possible (Chaney et al. 1988; Marx and McGowan 2010; Hall et al. 2021). Gardens located in low-income neighborhoods are often in close *distance* to polluting activities (Mielke et al. 1983; Kaiser et al. 2015; Oguntade et al. 2020; Montaña-López and Biswas 2021). Roads and various phenomena related to trafficking (density/usage; road, vehicle and tire wear; emissions, etc.) are documented contributors to the PTEs found in roadside gardens (Tam et al. 1987; Hursthouse et al. 2004; Morton-Bermea et al. 2009; Folens et al. 2017). Airports, trains, and shipping are also potential

sources of PTEs in urban gardens (Vazhacharickal et al. 2013; Cruz et al. 2014; Laidlaw et al. 2018; Leitão et al. 2018). As shown in the Table 1, other common sources of elevated PTE presence in garden soils are the application of contaminated sludge or irrigation water (Jamali et al. 2007; Abdu et al. 2011; Xue et al. 2012; Vazhacharickal et al. 2013) and element-based garden agrochemicals (Waterlot et al. 2011; Tran et al. 2020; Zhong et al. 2022). Other types of interesting sources of PTEs reported in Table 1 include plastics (Wang et al. 2018), E-waste processes (Luo et al. 2011), and war activities (Safi and Buerkert 2011).

Children are generally considered of greatest risk from exposure to particular PTEs because of two issues: (a) their *state of health*, which is generally characterized as having an undeveloped immune system; and (b) their *risky behavior*, which involves hand-to-mouth and object-to-mouth actions (Zeng et al. 2016; Rózański et al. 2021). Elderly gardeners may also be at risk if their *state of health* has been compromised or they have experienced long-term exposure to particular elements (Sun and Gu 2008; Alghamdi et al. 2022). Nevertheless, the risk can apply to anyone working at contaminated sites who experiences prolonged dermal exposure (increased by lack of personal protection equipment), *ingests* high concentrations of elements through inhalation of dust during *intense work* or through hand-to-mouth transfer, or by *consuming contaminated plants* (Zahran et al. 2013; Antisari et al. 2015; Hough 2007; Tong et al. 2020).

Important is that the degree of toxicity experienced from exposure to PTEs in the contaminated media is not simply related to the total concentration of the element involved, but importantly, the *bioavailability* and *bioaccessibility* of the element (Chapman et al. 2003; Janssen et al. 2003). Bioavailability refers to the fraction of PTEs that is released from a mineral matrix into a liquid form that can be taken up by plants or released into the gastrointestinal tract upon digestion (Chen et al. 2020; Petruzzelli et al. 2020); and bioaccessibility is the proportion that is available to induce a potentially toxic effect within the body. Bioavailability is determined by the characteristics of the element, soil, and biological organisms (Smith and Huyck 1999; Kim et al. 2015a, b; Bidar et al. 2020). Important soil properties include pH, organic matter, texture, cation exchange capacity, redox potential, oxide/hydroxide presence, and time (Petruzzelli et al. 2020). In performing risk assessments of element *toxicity*, bioavailability and bioaccessibility must be determined to avoid over-estimating the safety by using pseudo-total element concentrations alone (Izquierdo et al. 2015).

Pseudo-total concentrations are however useful for initial risk assessments that compare concentrations

with national guidelines—although these guidelines vary greatly worldwide (Additional file 1: Table S2). If a garden soil, and/or the produce grown in it, contain elevated levels of a particular element, *remediation* actions may reduce concentrations to safe levels, for example by replacing the soil or constructing raised beds with safe media (Clark et al. 2008; Rai et al. 2019; Paltseva et al. 2020). Further, risk can be reduced through *protection measures* that reduce the longevity/intensity of exposure, limits extensive disruption of the soil, and/or restrict access to those at high risk (Briffa et al. 2020; Lupolt et al. 2021). Once concern advances to the stage where remediation is considered, for example if the concentration exceeds an intervention threshold, bioavailability and bioaccessibility studies would be highly appropriate to assess risk in greater detail. However, a viable risk may be present to some individuals in cases of repeated exposure (for example consumption of particular crops) for concentrations lower than established thresholds.

The indicated interactions in the simplified model shown in Fig. 1 affect the final outcome of the complex system, which is toxicity or illness associated with exposure to particular PTEs in gardens. Again, potential illness largely stems from ingestion/inhalation/contact with high levels of various elements in the soil and the *consumption* of plants with high levels of PTEs, but in consideration with their bioavailability/bioaccessibility (Rai et al. 2019; Briffa et al. 2020). One relevant feedback in the system is associated with the state of *health*, in the sense that a variety of types of element toxicity reduces one's health, yet various aspects of an individual's health may provide some degree of resilience (Monachese et al. 2012; Mitra et al. 2022). Nevertheless, most avenues for reducing the negative health effect are *behavior* risk reduction, site *remediation*, and appropriate garden *management* and *mitigation* actions (Mitchell et al. 2014).

Fundamentally, understanding the underlying *soil and elemental properties* is an important process in governing many of the processes of the human exposure chain depicted in Fig. 1 (Selim and Amacher 1996; Violante et al. 2010; Li et al. 2022). Thus, the baseline information required in investigating this issue is knowledge of the PTE concentrations in garden soils in context with the behavior of the at-risk group—this type of assessment aligns with the scope of our work and preliminary investigation (Goh 2018). Even in areas where health risks have not been reported, new and site-specific knowledge is needed to make informed decisions regarding potential risks to exposure (Chaney et al. 1984; Kim et al. 2014; Cooper et al. 2020). Once obtained, this information helps guide additional follow-on work to fully assess

health risks to PTE exposure in contaminated urban garden soils (cf. EPA 2007).

Objectives

In this study we were concerned with assessing the presence of PTEs in the soils of community gardens in highly developed Singapore where urban gardening in housing estates is being promoted to encourage a growing, aging community to be active and self-sufficient (Tan and Neo 2009). Community gardens are also anticipated to supplement and diversify local food sources in the city (Chandran 2019). To that end, the nation aims to double the number of community gardens and allotments from 1500 to 3000 plots by 2030 (Begum 2020). The growing prevalence of gardens within public housing estates in Singapore that are visited frequently by onsite residents potentially represent an uncertain health risk because the gardens are inherently located in a dense matrix of industrial and residential land-uses with high road densities (Ng et al. 2006; Joshi and Balasubramanian 2010; Yuen et al. 2012). Further, other green landscapes such as public parks and accessible woodlands are also frequently visited locations of potential exposure to PTEs (Han et al. 2017).

We focus on the presence of four PTEs (Cd, Cu, Pb, Zn) that are often associated with contamination in urban garden soils worldwide (Xia et al. 2011; Kaiser et al. 2015; Table 1). These elements are also among the most studied in past assessments of urban contamination set in Singapore (discussed below). Lead has a legacy presence in Singapore soils despite most leaded petrol being phased out in Southeast Asia in the 1990s (Lee et al. 2014), as well as the regulation of Pb in other important sources such as paint (Kessler 2014). Our focus is on gardens constructed on soils; soil-less, raised bed, rooftop, and hydroponic systems are not considered. We also examine key soil properties (pH, Total Organic Carbon (TOC), texture) that may influence the mobility of elements in the soil (Zwolak et al. 2019). Absent in our assessment with respect to the exposure model (Fig. 1, “Human urban gardening exposure chain” section) are the determination of bioavailability, bioaccessibility, and PTE presence in vegetables. In foreshadowing our results, we did not perform these advanced experiments because pseudo-total element concentrations in soil were not alarmingly high. Finally, we use Pb to track the potential anthropogenic pollution sources of element contaminants in the Singapore urban soils (Lee et al. 2021).

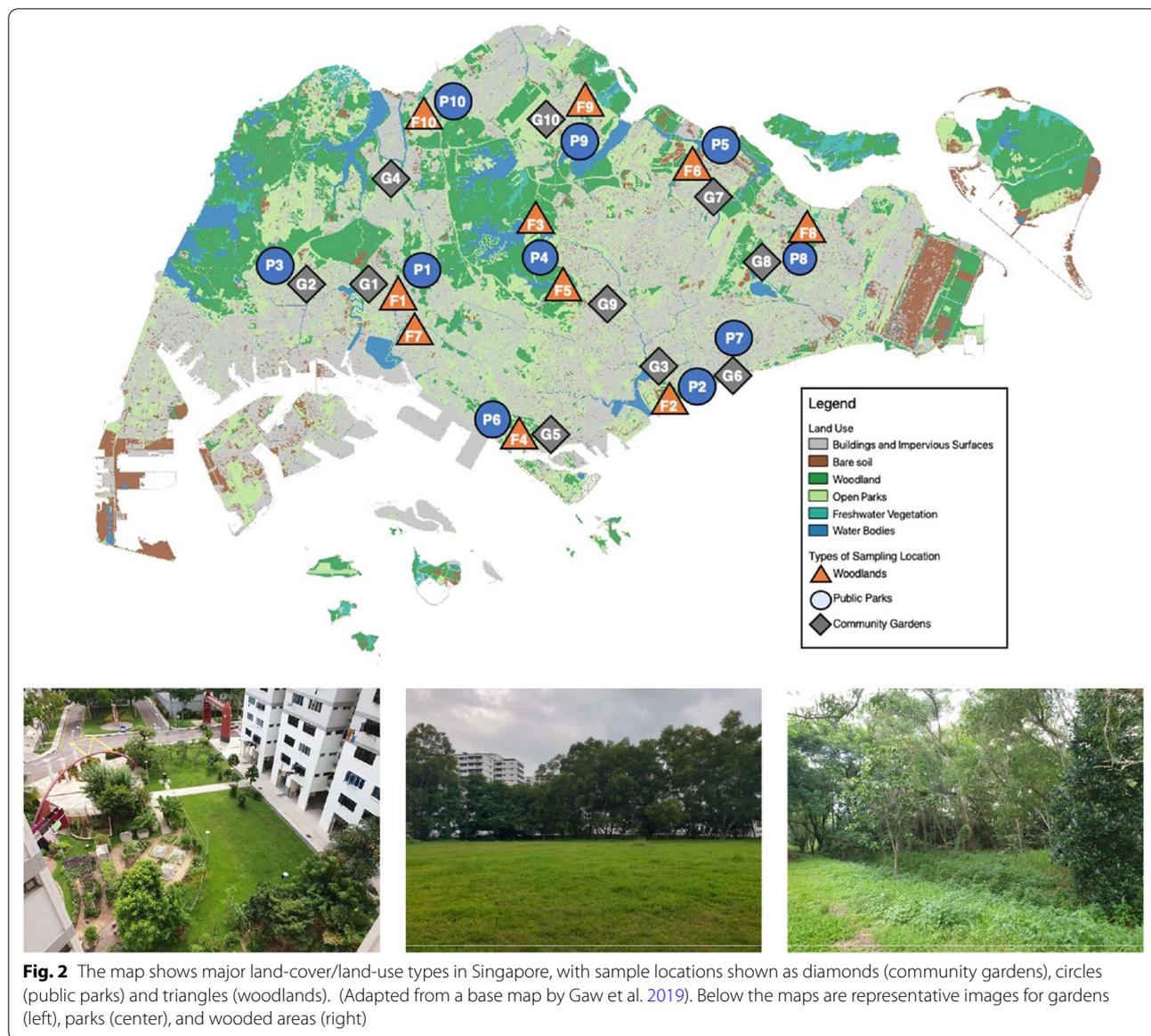
Study area

Singapore geography

Singapore has a tropical equatorial climate (mean annual temperature = 27 °C; total annual rainfall = 2300 mm) with two monsoon seasons and short dry periods. The geology of Singapore is somewhat diverse, with major formations including igneous rocks such as granite and norite, various sedimentary rocks (sandstone, mudstone), isolated metamorphic rocks, Quaternary sediments, as well as marine clay deposits (Sharma et al. 1999). Soils are generally acidic (pH 3.7–6.2), have low cation exchange capacities (maximum mean value: 100 mmolc/kg), and have low P concentrations (mean values ≤ 0.28 g/kg) irrespective of geology (Leitgeb et al. 2019). Soils developed on sedimentary rocks and sediments tend to have higher concentrations of soil organic carbon, total P, base forming elements, exchangeable base cations and moderate levels of some PTEs, for example Cr, Cu, Ni, Pb, and Zn (Leitgeb et al. 2019). Nguyen et al. (2019, 2020) determined that relatively high concentrations of PTEs may be associated with particular types of geological material (e.g., norite) and native soils.

Singapore's land-use is characterized by a dense matrix of urban areas with some lands dedicated to green spaces (Fig. 2). These areas differ somewhat in the degree of human disturbance. Woodland areas are made of (replanted) secondary forests located in either nature reserves or urban parks (Gaw and Richards 2021; Ramchunder and Ziegler 2021). Some have been disturbed by old village settlements, military activity, road building, and other activities requiring forest clearance followed by subsequent reforestation. However, these woodlands are not currently being disturbed due to strict land-use regulation. Open parks, in contrast, are grassland fields with few trees, but may be surrounded by substantial forested areas in some locations. Although they are considered open green spaces, soils tend to be compacted, both from construction and subsequent recreational activities where trampling occurs (Henderson 2013).

While some of the woodland soils may be in situ, the surfaces on all parks and urban gardens contain substantial fill material that was likely either imported from several quarries that were once operational in Singapore, or from other reclamation and sand mining projects (cf. Kog 2015). Thus, soils in gardens likely do not correspond with the underlying geology. Gardens are often built on marginal pockets of land sandwiched between housing blocks. Most of the construction of modern Singapore, including the spaces we investigated, took place in the last 50 years (Guo 2016). One garden we studied was built by top-filling over an old asphalt parking lot; another was built over a concrete basketball



court. These fill soils tend to be deficient in important plant nutrients, which necessitates the addition of fertilizers or construction of raised beds using garden soils purchased from local manufacturers. Store-bought soils and inorganic fertilizers may originate from Singapore or be imported from overseas. Gardeners also apply mulches and other organic materials (eggshells, coffee grounds, compost) to enrich soil nutrients. These additions may modify the amount of PTEs present in the garden. Garden soil disturbances are mainly related to reworking of the soil with hand tools.

Singapore urban pollution

Several studies have examined selected PTEs in a variety of Singaporean soils/sediments, including forests (Nguyen et al. 2018, 2020; Leitgeb et al. 2019; Wang et al. 2022), landfills (Patra et al. 2017), mangroves (Cuong et al. 2005), water bodies (Sin et al. 1991; Chen et al. 1996, 2016; Nguyen et al. 2018), marine sediments (Rahman et al. 1979; Goh and Chou 1997; Wood et al. 1997; Cuong et al. 2008), military lands (Nguyen et al. 2019), road dust (Joshi and Balasubramanian 2010; Joshi et al. 2009; Yuen et al. 2012); fly ash (Tan et al. 1997; Wu and Ting 2006); and various types of urban lands (Zhou et al. 1997; Ng et al. 2006). The studies of Chen (1999), Zhou et al. (1997), and Wang et al. (2022)

Table 2 Pseudo-total concentrations of Cd, Cu, Pb, and Zn found in a variety of soils (if not specified) and other media (e.g., sediment, ash) in Singapore

Ref.	Site	Cd	Cu	Pb	Zn
d	Forest; Nee Soon, upper Catchment [medians]	bdl	5 ± 5	13 ± 6	23 ± 6
d	Forest; Nee Soon, lower Catchment [medians]	bdl	12 ± 4	18 ± 7	33 ± 11
b	Golf course (3 locations)	NA	12–14	20–47	52–73
d	Disturbed lands, Nee Soon Catchment [medians]	bdl	16 ± 6	24 ± 9	43 ± 15
e	Forest soils (variable geology)	bdl–0.03	7–20	9–17	13–61
f	Park soils (4 locations) ^a	0.05–0.08	5–11	14–21	11–24
p	Park soils (3 locations) ^a	0.46–1.66	6–14	14–37	45–71
f	Airport/industrial area soils (2 locations) ^a	0.06–0.08	7–16	20–26	13–31
i	Reservoir susp. sediment (Kranji & MacRitchie)	NA	15–16	20–43	49–73
g	Residential soils (7 sites island-wide)	0.45–1.68	3–90	6–70	9–93
g	Reservoir sediments (3 sites)	0.79–1.73	8–61	15–65	1–68
d	Military lands, Nee Soon Catchment [medians]	bdl	44 ± 34	56 ± 37	77 ± 49
b	Forest; Nature Reserve (MacRitchie)	NA	6–19	9–38	29–123
b	Residential area (7 sites in Bishan)	NA	13–57	16–46	70–151
i	Reservoir sediments (MacRitchie)	NA	13–21	19–64	75–133
q	Mangrove sediment (S. Buloh & S. Khatib Bonus)	0.18–0.27	7–32	12–32	51–120
g	Field soils (7 sites island-wide)	0.45–1.42	1–105	5–89	1–150
a	Parks (Island wide; 10 locations)	bdl	9–70	bdl–61	33–190
h	MacRitchie Reservoir sediments (9-m core)	~ bdl–0.40	~ 1–370	~ 1–55	~ 1–135
a	Gardens (10 locations island-wide)	bdl–3.2	10–76	bdl–37	71–164
a	Woodlands (10 locations island-wide)	bdl–6.4	11–50	bdl–33	19–146
c	Forest; Bukit Gombak [medians]	5.76 ± 5.16	36 ± 24	14 ± 6	100 ± 27
o	Johore Strait marine sediments	0.11–0.36	11–93	27–70	69–231
i	Reservoir sediment (Kranji)	NA	21–29	35–82	128–367
r	Estuary sediment (Punggol)	bdl–1.37	4–439	1–157	NA
s	Coastal sediments (island wide)	bdl–1.6	1–1781	1–82	95–281
t	River sediment (Singapore river)	~ 0.1–1.0	~ 15–95	~ 40–240	~ 100–600
b	Industrial (Jurong West; 7 sites)	NA	12–194	29–167	111–837
b	Road side soils (heavy traffic area)	NA	30–68	27–1981	124–938
k	Road sediment; residential [means]	0.71 ± 0.30	247 ± 54	69 ± 26	275 ± 55
k	Road sediment; commercial [means]	0.30 ± 0.10	98 ± 76	111 ± 15	620 ± 186
p	Industrial zone soils (8 locations) ^a	0.66–1.63	16–258	10–242	40–708
l	Suspended sediment; residential road runoff	0.89–3.03	11–74	5–206	16–941
l	Suspended sediment; industrial road runoff	2.13–8.59	30–671	25–182	112–2006
g	Industrial soils (3 sites)	1.11–3.21	1–485	51–235	15–3594
j	Residential road sediment	NA	203–1294	135–395	704–2105
k	Road sediment; industrial [means]	2.41 ± 1.0	9069 ± 3742	338 ± 56	1696 ± 446
j	Road sediment (industrial areas)	NA	251–4271	144–744	836–11,960
n	Fly ash from municipal incinerator	95	570	2000	6288
m	Fly ash, Tuas solid waste incinerator [means]	130	987	2120	8072

Listed informally in order of increasing level of enrichment

Most values are ranges, means ± standard deviation, or medians ± MAD; ^aindicates a range of means/medians; ~ signifies the values were estimated from a graph. ND is not determined; bdl is below detection limit

References:

(a) This study; see “Methods and materials” section for details on the analysis methods

(b) Zhou et al. (1997); HCL–HNO₃–HF digestion and a flame atomic absorption spectrophotometer for high concentrations and a graphite furnace atomic absorption spectrophotometer for low concentrations

(c) Nguyen et al. (2020); microwave-assisted digestion methods EPA 3051a (USEPA 2017) followed by ICP-OES (Perkin Elmer 8300)

(d) Nguyen et al. (2018); four-acid digestion protocol (MA300 method by Bureau Veritas Labs (Vancouver, Canada) and ICP-MS (Perkin Elmer ELAN 9000)

Table 2 (continued)

- (e) Leitgeb et al. (2019); pseudo-total element concentrations were determined by digesting soil samples with aqua regia using a microwave oven (MARS 6, CEM, Kamp-Lintfort, Germany) and by analysing the solution using ICP-OES (Optima 8300, Perkin Elmer, Waltham, MA)
- (f) Ng et al. (2006), samples digested using microwave oven (Milestone, Ethos D, Monroe, C.T., USA) with a mixture of 9 ml HNO₃ and 3 ml of HF in a closed vent medium pressure vessel with a ramp to 180 °C at 600 W for 10 min and held at 180 °C, 600 W for 10 min
- (g) Chen (1999), mixed acid method (1 ml HF (49%) added to 14 ml 3:1 HCL (36%): HNO₃ (68%) added by 20-min microwave digestion; analysis on ICP-AES
- (h) Chen et al. (2016); leached with ultrapure grade 1.75 mol/l HNO₃-3 mol/l HCl in an ultrasonic bath, followed by analysis using a quadrupole inductively-coupled plasma mass spectrometry (Q-ICP-MS, VG PlasmaQuad 2+)
- (i) Chen et al. (1970); HNO₃ digestion in a microwave followed by analysis with ICP Perkin-Elmer Plasma 400 Emission Spectrometer
- (j) Yuen et al. (2012); digestion with strong acids HNO₃-H₂O₂-HF; analysis on a Thermo Scientific X Series2 Quadrupole ICP-MS
- (k) Joshi et al. (2009); digestion with a combination of strong acids (HNO₃-H₂O₂-HF) in a microwave digester, and analyzed using inductively coupled plasma-mass spectrometry (ICP-MS, Perkin Elmer Elan 6100)
- (l) Joshi and Balasubramanian (2010); digestion of the filter paper containing suspended solids in a microwave digester with a combination of strong acids HNO₃-H₂O₂-HF, followed by analysis on ICP-MS
- (m) Tan et al. (1997); sequential extraction; analysis on a Perkin-Elmer ICP Plasma Emission Spectrophotometer
- (n) Wu and Ting (2006); total digestion according to US EPA SW 846 Method 3050B using ICP-OES (Perkin-Elmer Optima 3000V)
- (o) Wood et al. (1997); digestion with a mixture of HNO₃, HClO₄, and HF; analysis by atomic absorption spectrometry (AAS)
- (p) Wang et al. (2022); means (3 reps) for three parks and eight industrial areas. ICP-OES; microwave-assisted digestion (details uncertain)
- (q) Cuong et al. (2005); micro-wave assisted HNO₃-HF digestion; Perkin-Elmer AAnalyst 600 GFAAS
- (r) Nayar et al. (2004); digestion with suprapure HNO₃; Perkin-Elmer AAnalyst 600 GFAAS
- (s) Goh and Chou (1997); digestion with suprapure HNO₃; analysis on Hitachi Polarised AAS
- (t) Sin et al. (1991); digestion not specified; AAS/FASS

are particularly informative to our study because they show loading of Cu, Pb, and Zn in soils are higher in the proximity to industrial areas compared with natural or residential areas (Table 2). Chen (1999) found very high maximum values of Cu (485 mg/kg) and Zn (3594 mg/kg) in industrial soils. In the most recent study, Wang et al. (2022) found maximum concentrations of Cu and Zn were an order of magnitude higher in soils adjacent to industrial activities, compared with parks.

The main sources of high PTE concentrations in Singapore soils are likely vehicular phenomena and industrial activities (Zhou et al. 1997; Wood et al. 1997). For example, Yuen et al. (2012) reported roads in or near industrial areas had higher levels of PTEs in surficial dust compared with residential areas (Table 2). Joshi et al. (2009) reported very high concentrations of Cu (9069 ± 3742 mg/kg) and Zn (1696 ± 446 mg/kg) for road sediment in industrial areas. Wood et al. (1997) reported that elevated Pb and Zn levels in sediments of the Johor Straits along the Malaysia-Singapore causeway likely resulted from gasoline and tire wear, respectively. Fly ash from incinerators in Singapore have very high concentrations of PTSs (Table 2): Cu (570–087 mg/kg), Pb (2000–2150 mg/kg), and Zn (6288–8072 mg/kg). Potential contamination sources in Singapore are indicated in the map in Additional file 3: Fig. S2.

A prior study of airborne element-bearing aerosols in Singapore showed an enrichment of a variety of PTEs (Pb, Zn, Cd, V, Ni, Cr, and Cu) that originate from several sources including oil-fired power plants, element

processing industry, land reclamation, construction activities, municipal solid waste incinerators, and traffic emissions (Balasubramanian and Qian 2004). Ng et al. (2006) determined that element accumulations in lichens across Singapore were the result of atmospheric deposition, with peak concentrations of Cu (45 mg/kg), Pb (17 mg/kg), and Zn (84 mg/kg) found in locations associated with heavy petroleum refinement, shipping industries, and road traffic, respectively. Rivellini et al. (2020) found evidence that trace elements in aerosols may originate from shipping emissions and the oil refinery industry. Once airborne, these contaminants can be deposited in the environment during rain events and via dry dust deposition (López et al. 2019).

Hu and Balasubramanian (2003) determined that the removal of the trace elements from the atmosphere by precipitation was influenced by the rainfall volume as well as pH. Further, the magnitude of mean annual wet deposition fluxes of combustion-generated elements (V, Ni, and Cu) were high relative to other locations because of abundant rainfall. The southwest monsoonal winds may cause slight increases in PTE concentrations of airborne particles due to the presence of industrial estates and petrochemical plants in the southwestern region of Singapore (Balasubramanian et al. 2003; Rivellini et al. 2020). Another potential source for the occasionally elevated concentrations of Cu and Zn found in residential areas of Singapore is the degradation of buildings materials.

Methods and materials

Sampling and analyses

We adopt an exploratory survey of PTEs in soils across the breadth of the island of Singapore to eliminate spatial bias that may be related to proximity to probable sources of element pollution. In May–June 2018 we collected 300-g samples from 10 sites in the following three land uses (Fig. 2): woodland; parks; and community gardens (raw data reported in Additional file 1: Table S2).

Soil pH was measured in a soil suspension with 1:5 soil:water ratio using a calibrated 9157BNUMD RossUltra pH/ATC Triode with AgCl–Hg electrolytes (Thermo Fisher Scientific, Waltham, USA). Soil particle size was determined using the Mastersizer 2000 (Malvern Instruments, Malvern, UK) via the laser obstruction method. Total organic carbon was determined by combusting 0.03 g of ground soil that was first pre-wetted, fumigated for 6 h to remove inorganic carbonate, and then oven-dried. The samples were combusted at 950 °C in the Vario TOC Cube (Elementar Analysensysteme GmbH, Langensfeld, Germany).

Soil element pseudo-total concentrations were determined using the PerkinElmer Optima 8300 Inductively-Coupled Plasma Optical Emission Spectrometer (ICP-OES). Prior to analysis we used a microwave-assisted acid approach for digestion (US EPA Method 3051A17). First, 0.5 g of the finely ground material was placed in digestion vessels before being reacted with 9 ml of Nitric acid (65%) and 3 ml of Hydrochloric acid (37%) in a fume hood for 15 min. The samples were then heated in a microwave digester, ramping up to 175 °C within 10 min, then maintaining that temperature for another 15 min. Multi-elemental standard solutions of 0.5 mg/kg, 1 mg/kg, 5 mg/kg and 10 mg/kg in 2% HNO₃ were used to calibrate the ICP-OES instrument.

Table 3 Quality assurance/quality control information for Al, Cd, Cu, Pb, and Zn determinations for one standard reference material (USGS 2711a)

	Cd	Cu	Pb	Zn
Certified value (mg/kg)	54.1 ± 0.5	140 ± 2	1400 ± 10	414 ± 11
Analyzed value (mg/kg)	58.6 ± 0.8	190.6 ± 8.6	1462 ± 13	418.4 ± 4.7
Recovery (%)	108	136	104	101
Bias (%)	8.3	36.1	4.4	1.1
Precision	0.01	0.05	0.01	0.01

Certified value is the concentration reported by the SRM manufacturer; analyzed is the mean ± Stdev determined by ICP-OES (this study); recovery = analyzed value/certified value × 100%; bias = (analyzed value – certified value)/certified value × 100; precision = Stdev/mean (i.e., coefficient of variation of analyzed values)

We used the USGS 2711a Certified Reference Material to perform quality control and quality assurance of both the accuracy and repeatability of PTE measurements (Table 3). The percentage recovery of PTEs in this research (% Lab recovery) is generally higher than the recovery rates from the USGS Contract Laboratory Study Programme (CLSP). We found higher recoveries compared to those obtained in the USGS CLSP, which mainly used the weaker EPA Method 200.7 (US EPA 2001) for soil digestion via hotplate reflux (Mackey et al. 2010). Microwave-assisted digestion results in higher recoveries because the high temperatures and pressures achieved in microwave vessels allows for a more complete digestion of the soil's PTEs (Chen and Ma 1998, 2001). Closed microwave vessels also reduce the potential loss of trace elements through volatilization, which is a feature in open hotplate digestion (Link et al. 1998).

The accuracy and bias of Cd, Pb, and Zn are acceptable; however, the over estimation of Cu (34%) is noted and considered in our interpretation of the results. We deemed that ICP-OES and di-acid digestion approach was acceptable for the expected range of total concentrations that would indicate potential risk (Olesik 2020). Over the years, a variety of methods have been used to determine pseudo concentrations of PTEs in garden soils (summarized in Additional file 1: Table S1).

Finally, we determined lead isotope ratios in five samples to investigate the origin of Pb at selected sites (Komárek et al. 2008). In doing so we assume that Pb is a representative indicator of general pollutant dispersal pathways (Wiederhold 2015; Harvey et al. 2017; Carrasco et al. 2018; Crispo et al. 2021). We analyzed the soil from five sites, one woodland (#4) and two each for parks (#1 and 7) and gardens (#2 and 10), to track likely sources of Pb in the three community spaces (see locations on Fig. 2). The Pb isotope values were determined by the Environmental Chemistry and Microbiology Unit at the Charles Darwin University in Darwin, Australia. The samples were selected because they were of relatively high concentrations. The finely-ground samples underwent acid digestion with HNO₃ and HClO₄. Isotope values were determined using an Inductively-Coupled Plasma Mass Spectrometer.

Calculations and statistical handling

As the number of soil samples collected at each site were few (n = 10), we used nonparametric statistics for data inference, including the Spearman rank correlation test, the Kruskal Wallis (KW) for multiple treatment comparisons, and the Mann–Whitney U (MW-U) as a post-hoc test. Differences were considered significant at $\alpha = 0.05$ (P-values < 0.05). We used an Enrichment

Factor (EF) to assess the anthropogenic enrichment of PTEs in the soils of different land covers (Chester and Stoner 1973). Although the validity of EF values is debated (Reimann and Garrett 2005), we find them useful for indicating sites of potentially high contamination (Szolnoki et al. 2013). In the calculation of EF, we used Al as a normalizing element:

$$EF_M = \frac{\left(\frac{X_M}{X_{Al}}\right)}{\left(\frac{BG_M}{BG_{Al}}\right)}, \quad (1)$$

where X_M and X_{Al} are the concentrations of an element M and Al in the sample, respectively; and BG_M and BG_{Al} are their calculated background values. For background values we used those from a prior study conducted in the forested Nee Soon Forest Catchment (Nguyen et al. 2019): Al (72,900 mg/kg); Cd (0.2 mg/kg); Cu (5 mg/kg); Pb (13 mg/kg); and Zn (23 mg/kg).

Soil element concentration risk guidelines

Finally, as Singapore does not have published guidelines pertaining to health risks associated with concentrations of PTEs in soils, we determined three thresholds based on a review of guidelines for nearly 50 countries/regions worldwide (Table 4): (1) Threshold risk concentration (TRC), probable risk concentration (PRC), and elevated risk concentrations (ERC). The values for these

three thresholds for Cd, Cu, Pb and Zn (Table 4) are determined as the 20th, 50th (median), and 80th percentile of the values summarized globally (Additional file 1: Table S3). The ERC values are of approximately the same magnitude as those employed by Wang et al. (2022) as “intervention” values. We caution that while these guidelines are useful for examining potential levels of risk in our assessment, they may not be appropriate for policy/management applications without a more critical review.

In Tables 1 and 2 we list studies based on a first-order ranking of level of contamination. For each study we calculate a composite score that is determined by awarding 1 point if the maximum value of an element exceeds the TRC; 2 points if the PRC is exceeded; and 3 points if the ERC is exceeded. Four points are assigned if the maximum is tenfold the ERC (twofold for Cd). For studies, not reporting maximum values we use the mean (median) + stdev (MAD) as the upper value. If an element was not determined (ND) we adjust the ranking to align with level of contamination relative to other studies. These rankings are made to facilitate discussion in this paper, not to compare cities/locations directly (see below).

Results

Pseudo-total concentrations

The main difference between PTE presence in soils in gardens, parks, and woodlands was in concentrations of

Table 4 Number of samples exceeding threshold risk concentrations (TRC), potential risk concentration (PRC), and elevated risk concentrations (ERC) for four metals and three treatments

TRC, PRC, ERC concentrations ^a					
Level ^a	Unit	Cd	Cu	Pb	Zn
Background	mg/kg	0.2	5	13	23
TRC	mg/kg	0.5	46	57	100
PRC	mg/kg	2	100	140	200
ERC	mg/kg	10	200	400	700
Number of samples exceeding TRC, PRC, ERC thresholds					
Threshold	Land cover	Cd	Cu	Pb	Zn
TRC	Garden	3	5	0	7
PRC	Garden	2	0	0	0
ERC	Garden	0	0	0	0
TRC	Park	0	2	1	1
PRC	Park	0	0	0	0
ERC	Park	0	0	0	0
TRC	Woodland	1	2	0	1
PRC	Woodland	1	0	0	0
ERC	Woodland	0	0	0	0

^a The pseudo-total concentration indicators of level of risk are determined as the 20th (TRC), 50th (PRC), and 80th (ERC) percentile of the ranked concentrations from values reported for numerous countries worldwide (summarized in Additional file 1: Table S2). In the bottom panel, n = 10 for each land cover

Table 5 Summary statistics for PTE pseudo-total concentrations and soil properties for the three land covers

Element	Units	Gardens	Parks	Woodlands
Al	mg/kg	99,070 ± 10,130 [61412–123700]	118,850 ± 18,070 [96980–143440]	110,590 ± 32,970 [40832–156400]
Cd	mg/kg	bdl [bdl–3.2]	bdl [bdl–bdl]	bdl [bdl–6.4]
Cu*	mg/kg	42.5 ± 11.5 (b) [10.0–76.6]	26.4 ± 6.2 (a) [9.2–69.6]	17.6 ± 5.5 (a) [10.6–49.6]
Pb	mg/kg	bdl [bdl–36.6]	bdl [bdl–60.8]	bdl [bdl–32.6]
Zn*	mg/kg	124.8 ± 33.8 (b) [70.8–164.2]	49.8 ± 11.2 (a) [32.6–189.6]	51.5 ± 17.4 (a) [19.2–146]
TOC	%	3.4 ± 1.1 [1.3–5.9]	3.7 ± 1.1 [1.1–5.5]	4.5 ± 0.8 [1.7–9.0]
pH		6.9 ± 0.6 (c) [5.8–7.8]	6.0 ± 0.5 (b) [4.8–8.0]	5.0 ± 0.7 (a) [4.0–8.6]
Clay	%	7.8 ± 0.5 [5.8–9.7]	6.9 ± 0.5 [5.8–8.0]	7.8 ± 0.5 [5.4–9.6]
Silt	%	55.1 ± 2.1 (a) [51.5–62.2]	67.0 ± 4.1 (c) [52.9–74.9]	62.2 ± 6.5 (b) [41.9–78.5]
Sand	%	35.5 ± 1.1 (b) [27.1–39.7]	24.4 ± 1.7 (a) [17.0–38.8]	29.6 ± 5.5 (b) [12.0–47.8]

Values reported are medians ± standard absolute deviations (n = 10 samples). Ranges are reported in brackets. bdl refers to below detection limit, which are 1.62 (Cd), 5.82 (Cu), 12.6 (Pb), and 3.54 (Zn). Values in rows with different letters in "()" are significantly different, based on post-hoc testing (KW, followed by MW-U; $\alpha = 0.05$)

*The differences among landcovers is significant at $\alpha = 0.5$ (KW Test), with garden values being significantly higher than the other treatments (KW; $\alpha = 0.05$). Values in rows with different letters in "()" are significantly different, based on post-hoc testing (MW-U; $\alpha = 0.05$)

Cu and Zn (Table 5). Firstly, detectable concentrations of Cu and Zn were found in all 30 samples. Garden soils had statistically higher concentrations of Cu (42.5 ± 11.5 ; [10.0–76.6] mg/kg Cu) than soils in parks (26.4 ± 6.2 ; [9.2–69.6] mg/kg Cu) and woodlands (17.6 ± 5.5 ; [10.6–49.6] mg/kg Cu). The values reported are medians ± median absolute deviations; the range is in brackets; and n = 10 for all three treatments. Zinc concentrations in gardens (124.8 ± 33.8 [70.8–164.2] mg/kg Zn) were also statistically ($\alpha = 0.05$; KW) higher than in park soils (49.8 ± 11.2 ; [32.6–189.6] mg/kg Zn) and woodland soils (51.5 ± 17.4 ; [19.2–146] mg/kg Zn).

Unlike Cu and Zn, Cd was rarely detectable in any sample (detection limit = 1.6 mg/kg Cd): three garden soils had Cd concentrations ranging from 1.7 to 3.2 mg/kg Cd; one woodland soil sample had a relatively high value of 6.4 mg/kg Cd (Table 5). No detectable Cd was found in park soils. Similarly, Pb was also rarely present in concentrations above our (high) detection limit of 12.6 mg/kg (n = 5, 5, and 4 in gardens, parks, and woodlands, respectively). Detectable Pb concentrations ranged from 13 to 61 mg/kg Pb across the three treatments (Table 5). The maximum concentrations found in gardens, parks, and woodlands were about 36, 61, and 33 mg/kg Pb, respectively. The generally low PTE concentrations we found are generally comparable to those reported by Chen (1999), Zhou et al. (1997), Ng et al. (2006), Nguyen et al. (2018, 2020), and Wang et al. (2022) for a variety of urban soils in Singapore.

PTE enrichment

In comparison with elemental background concentrations from soils in the forested Nee Soon Catchment, the Cu and Zn values found in soils of the

three land uses investigated are considered “enriched” to some degree. For example, the median enrichment factor (EF_M ; Eq. 1) for Cu in gardens was 7; median EF_M values were 3 and 4 in parks and woodlands, respectively. Simply, these values indicate the medians of the samples measured in gardens, parks, and woodlands are all three- to sevenfold higher than the background concentration of 5 mg/kg Cu. Enrichment factors for Zn ranged from 5 in garden soils to 2 for both park and woodland soils (background = 22 mg/kg Zn). Here, enriched does not necessarily indicate high risk. It may simply indicate an anthropogenic contribution; alternatively, it may simply the particular mineralogical composition of the soil—which again may be imported or natural for that particular location.

Correlations

The garden soils were slightly sandier (median of 36%) than those in parks or woodlands (24 to 30%; Additional file 3: Table S4). Copper and Zn concentrations were negatively associated with clay content in garden soils ($\rho = -0.663$ and -0.725 , respectively; $\alpha < 0.01$; Additional file 3: Table S4). In the woodlands, Cu concentrations were positively correlated with silt percentage (0.674), and negatively correlated with the sand fraction (-0.609 ; $\alpha < 0.01$). This same textural association was present to a lesser degree for Zn in woodland soils: silt ($\rho = 0.371$; $\alpha < 0.05$); and sand (-0.255 ; $\alpha < 0.05$). The pH of garden soils ranged from 5.8 to 7.8, with a neutral median value (6.9 ± 0.6 ; Additional file 3: Table S4). In comparison, the soils of the other two land uses were more acidic (median pH was 5 in the woodland soils; 6 in park soils) and displayed larger ranges: 4.8–8.0 (parks); 4.0–8.6

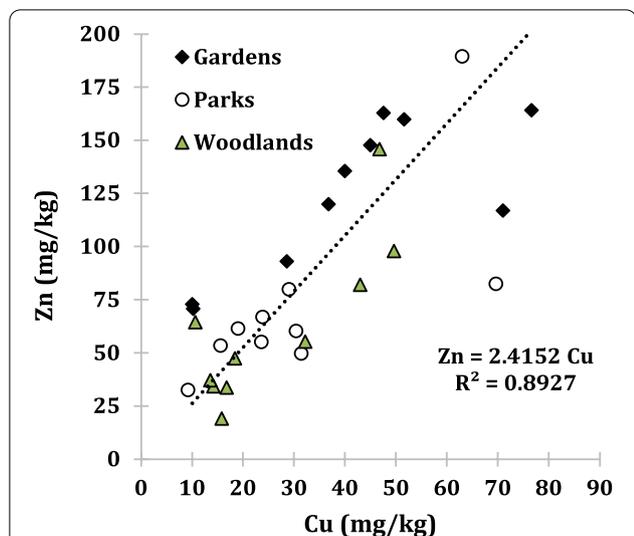


Fig. 3 Relationship between copper (Cu) and zinc (Zn) in garden, park, and woodland soils in Singapore. The Spearman rank correlation coefficient is $\rho = 0.78$ (p -value < 0.001)

(woodlands). The only significant correlation involving pH was a weak correlation with Cu concentration in garden soils (0.387 ; $\alpha < 0.01$; Additional file 3: Table S4).

None of the soils had particularly high TOC, with median percentages ranging from 3.4 to 4.5 (Table 5). The highest value was found at a woodland site (9%); low values $< 2\%$ were found among all three treatments. TOC concentrations were positively correlated with Cu and Zn in garden soils ($\rho = 0.761$ and 0.667 ; $\alpha < 0.01$; respectively), as well as park soils (0.447 and 0.667 , but with different levels of significance). Collectively, these significant correlations between element concentrations and soil properties provide some indication of elements being bound to the organic matter or particular texture fractions in soils, but without a consistent pattern across land uses. Unexpected is the negative association with clay content (Additional file 3: Table S4).

With regard to the PTEs, the correlation between Cu and Zn was significant ($\rho = 0.78$; $\alpha < 0.001$; Fig. 3). Correlation analysis involving Cd and Pb was hindered by so many samples having concentrations below detection limits.

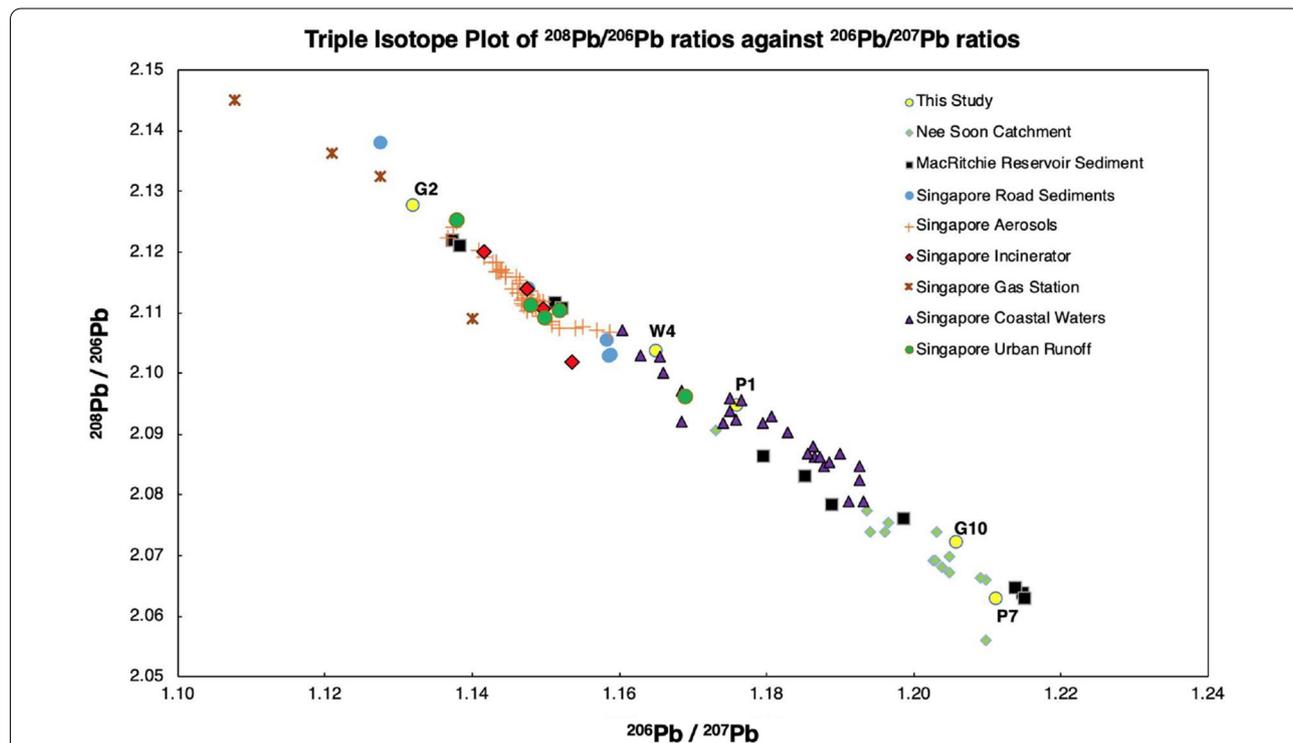


Fig. 4 Triple Pb isotope plot of Pb sources in Singapore, which includes roadside sediments in blue circles (Yuen et al. 2012), aerosols in orange crosses (Lee et al. 2014; Chen et al. 2016), incinerator ash in red diamonds (Chen et al. 2016), gas station soil residuals in brown asterisks, Singapore coastal water in purple triangles and urban runoff in green circles (Carrasco et al. 2018), MacRitchie reservoir sediments (Chen et al. 2016) and Nee Soon Catchment soil samples (unpublished data)

Lead isotope ratios

Chen et al. (2016) suggested that due to the close proximity of industrial estates with incinerator plants, Pb pollution in Singapore usually comes from multiple sources including industrial activities, incineration, the past use of leaded petrol, and the weathering of parent rock material. The Pb isotopes for garden site G2 were similar to those found in urban runoff from petrol stations and shipyards (Fig. 4). This result suggests that even though Pb was banned from petrol in 1998 (Chen et al. 2016) the legacy of Pb contamination has persisted, probably because of the low solubility (Sutherland and Tolosa 2001; Yuen et al. 2012). Abrasive blasting of element surfaces in shipyards during repair and construction works are another potential source of Pb aerosols. Considering the down-wind location of G2 from the Tuas shipyards, the aerial transport of Pb particles to the site is plausible. Wang et al. (2022) reported high concentrations of selected elements in the plant foliage in this area—although lead was not elevated. Again, potential sources of pollution are shown in the map in Additional file 3: Fig. S2.

Sample W4 is similar to the roadside sediments collected from Tuas and Senoko Industrial Estates and the incinerator from Keppel Seghers (Fig. 3). Again, incinerator ash from Singapore contains high concentrations of Pb (≥ 2000 mg/kg; Table 2). The presence of Pb at W4 could therefore be due to the upwind (S and SW winds) location of the Keppel Seghers

incinerator and the airborne transport of Pb in fly ash from industrial activity in Tuas and West Coast, followed by deposition (Hu and Balasubramanian 2003). Industrial emissions of Pb also include plastics production and element processing plants (Ragaini et al. 1977; Siddiqui et al. 2009), which are found at the Tuas (southwest) and Senoko industrial estate in the north.

The Pb isotope ratio at park site P1 is similar to that of coastal waters of the Central Business District, Kranji and Sembawang (Fig. 3). Rahman et al. (1979) state that element accumulation in Singapore waters and marine sediments must come from natural sources or be introduced from land- and marine-based activities, including leakage from landfills, oil/fuel spills, and/or runoff from shipyards, wood mills, motor works shops, and small industries. Lead concentrations in marine sediments at seven locations ranged from 20 to 28 mg/kg Pb (Rahman et al. 1979). The Pb ratios for P1 likely reflect the mixing of natural Pb and that associated with land-based runoff; however, this inference has uncertainty.

Two of the soil samples tested did not contain an anthropogenic Pb signature (Fig. 3). Isotopic ratios for sites P7 and G10 are close to those found at the base of the MacRitchie Reservoir sediment core, which is believed to reflect naturally occurring Pb in Singapore that was transported with eroding material from the forested headwaters. The G10 signature is most similar to those from soils in the relatively undisturbed Nee Soon Catchment on the island.

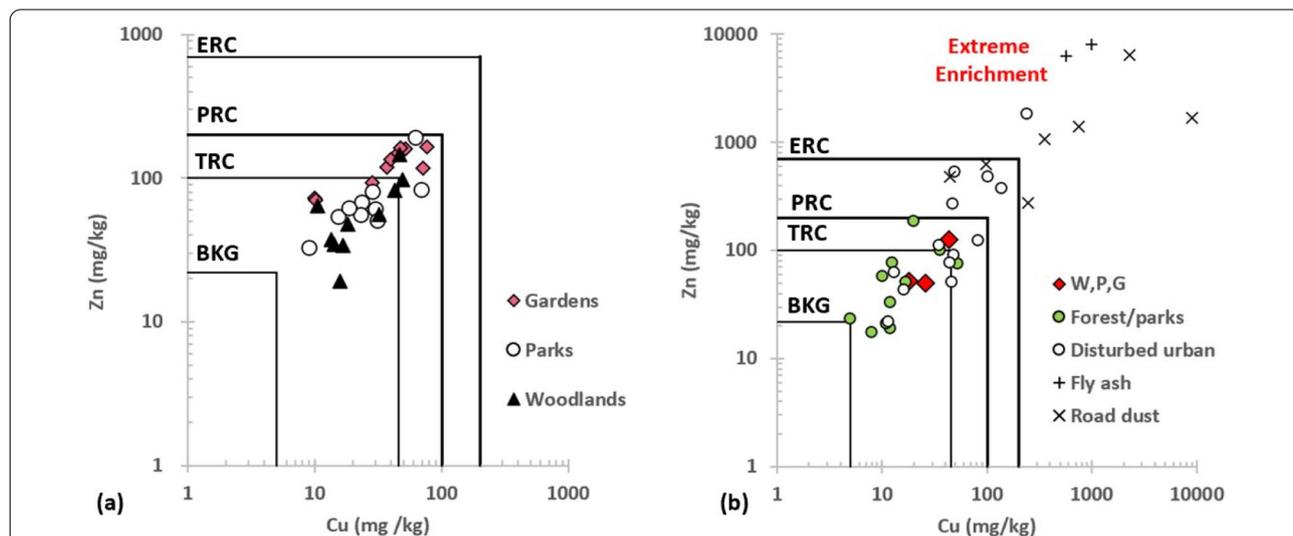


Fig. 5 Plots of copper (Cu) versus zinc (Zn) concentrations for: (a) garden (G), park (P) and woodland (W) sites investigated in this study (n = 10 samples each); and (b) various soils, road dust, and fly ash in Singapore (from Table 1), along with median values from this study. Background (BKG), Threshold Risk Concentrations (TRC), Potential Risk Concentrations (PRC), and Elevated Risk Concentrations (ERC) are listed in Table 4 (top panel). In panel a, all thirty values for each land use type are shown. In panel b, element concentrations beyond the ERC thresholds are considered to have a substantial anthropogenic enrichment (as in the case of fly ash and most road dust samples)

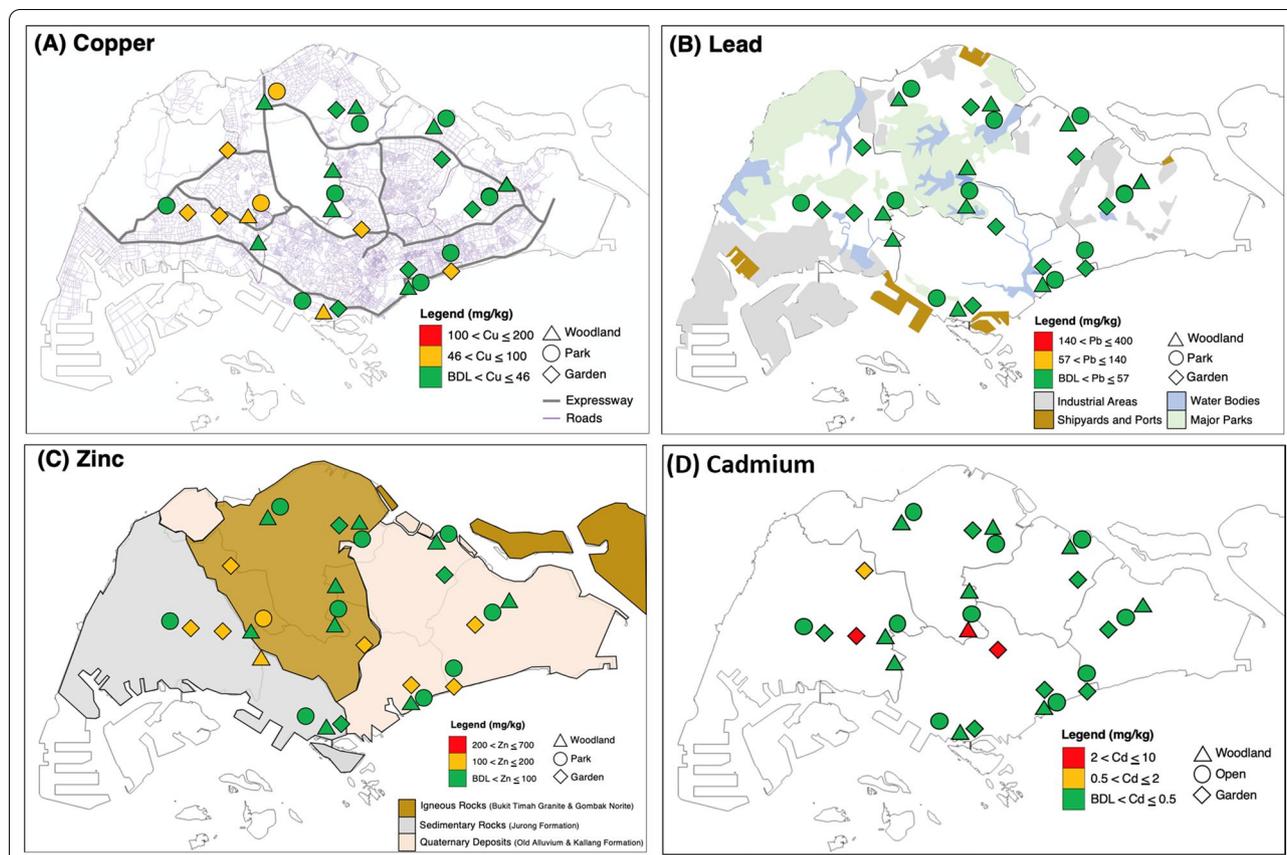


Fig. 6 Concentrations of Cu (A), Pb (B), Zn (C) and Cd (D) at garden (diamond), park (circle), and woodland (triangle) sample sites in Singapore. Concentrations are presented relative to levels of risk (top of Table 4). The background in each map varies to allow comparison of the study sites with the road network (A), major parks, industrial areas, water bodies and shipyards (B) and large geological formations (C). The base maps in A–C do not necessarily reflect an impact on the element that is mapped

Discussion

Spatial patterns

The observed median values of Cu (18–30 mg/kg) and Zn (50–52) for the three treatments are at the high end of the range of central tendency values (reported means, median, midpoints of ranges) associated with soils in forests and natural spaces determined by others for Singapore (Table 2; Fig. 5). The median concentrations for garden soils were slightly higher (43 and 125 mg/kg Cu and Zn, respectively), and therefore, resembled the lower values associated with disturbed soils in Singapore (Table 2). Comparison with reservoir sediment values supports the idea of some degree of anthropogenic enrichment above the natural geochemical signal on the island. With respect to geography, the highest Cu concentrations had no obvious pattern, while Zn concentrations tended to be higher in the western and south-eastern region of Singapore (Fig. 6). Higher Pb concentrations were found in the northern part of Singapore, although a more obvious relation was the occurrence of detectable concentrations in park soils (n = 3) versus garden (1) and

woodland soils (1). Again, most Cd concentrations were low and had no strong geographic pattern.

Elevated concentrations of elements in samples from the western and northern area of Singapore were somewhat expected because of the close proximity to industrial element processing, wood combustion, and waste incineration facilities, which are shown in gray shading in Fig. 6b (Nriagu 1979; Wang et al. 2022). Prior analysis of emissions from two incineration plants in the southwest of Singapore showed that high levels of Cu and Zn attached onto particulate matter (Tan et al. 1997), which may lead to elevated concentrations via deposition. We also expected some enrichment in this area because of the role of the southwest monsoonal winds blowing over industrial estates and petrochemical plants (Balasubramanian et al. 2003). Of note, five of the western-most sites had concentrations of Cu and Zn that were higher than their respective TRCs (Fig. 6d; Table 3).

The great variability in Pb isotopic ratios (Fig. 4) indicates different potential sources, both natural and anthropogenic (Fig. 4). The absence of common sources

for lead aligns with the general lack of spatial patterning in the concentrations (Fig. 6). Relevant is that many of the samples enriched in Cu and Zn found in prior studies were often associated with industrial activities in similar locations (Zhou et al. 1997; Chen 1999; Wang et al. 2022). Both our results and those of Wang et al. (2022) suggest that the footprint of many potential pollution sources is relatively small—and therefore contribute to the “spotty” nature of enrichment across the island.

Singapore urban garden soil contamination context

The apparent combination of several activities/processes that contribute to the variable enrichment of particular PTEs makes Singapore different from several studies conducted in very contaminated urban environments (Table 1). Briefly, the sites in Table 1 are ordered informally according to general contamination level of four elements. The studies at the top of the table have concentrations that rarely exceed the conservative risk metrics we used herein (Table 4); the locations at the bottom of the table have concentrations for multiple PTE that exceed the ERC concentration indicating elevated risk. Singapore sits at position 27 on this list of 142 entries (Table 1). Again, one should be careful in making literal distinctions among sites/studies owing to differences in methods, timing, and rationales for each study. For example, many early studies were conducted in areas where contamination from known activities was.

Generally, soil PTE concentrations in gardens worldwide tend to be affected by legacy contamination from the following (Table 1): leaded paint and various construction materials used in old neighborhoods (e.g., Baltimore, Christchurch, London, New York, New Orleans, San Francisco, Washington DC), smelting/mining (Baia Mare, Dalnegorsk, Gijon, Palmerton, Paris area) and unsafe waste disposal/usage in gardens as irrigation or fertilizer (Kampala, Paris, Marseille, New York, New Castle). The proximity to heavily used roads may also be an important source of Cd, Cu, Pb, and Zn in gardens, for example, as is the case in Bristol, Cardiff, Hong Kong and New Orleans (Table 1). Older roads may have high levels of Pb stemming from past leaded petrol use.

Noticeable in Singapore is the generally low PTE concentrations in garden, park, and woodlands soils, compared with road-associated sediments and fly-ash (Table 2). As concentrations of Cu and Zn decrease dramatically with increasing distance from roads (Chen et al. 2010; Kim et al. 2017), the moderate PTE concentrations we found are plausible because many of these spaces not often located directly adjacent to roads. Gardens are often ensconced within housing complexes and occasionally sheltered by buildings and trees.

Enriched values in garden soils may also be result from degradation of building materials associated with the residential units (Chaney et al. 1984; Lim et al. 2021).

Many authors in the reviewed studies refer to general urban atmospheric deposition of elements in dense cities with much industrial activity and traffic congestion. This type of “new urban deposition” phenomena is likely an apropos description explaining the variations we found in Singapore. In support, Balasubramanian and Qian (2004) reported that the enrichment of aerosols in Singapore was more than 1000 times for Cd, Pb, and Zn; 100 times for Cu. Deposition is almost certainly superimposed on the signatures of the garden soil and may have a legacy signature associated with the age of the building complex. Management activities might be partly responsible for our finding of enriched Cu and Zn concentrations despite the coarse nature of the garden soils, as reported worldwide (Table 1).

For example, the high TOC percentages we found in gardens, which are maintained by application of composted dead organic matter containing element-fixing humic acid, potentially contribute to the enrichment of Cu and Zn in surface soils (Veeken et al. 2000; Amir et al. 2006; Sánchez-Monedero et al. 2002). In addition, NPK fertilizers may facilitate the leaching of Cu and Zn from the topsoil (Young 2013; Bolan and Duraisamy 2003; Loneragen and Webb 1993; Carbonell et al. 2011; Rajneesh et al. 2017; Warman and Cooper 2000). Further, Cu- and Zn-based fertilizers often contain insoluble compounds (e.g., CuSO_4 , ZnO, and ZnSO_4) that potentially elevate Cu and Zn concentrations (Tsang et al. 2014; Jones and Belling 1967). Garden patrons often remedy acidity by applying Ca-rich fertilizers including eggshells to raise the soil pH (Ok et al. 2011), which may in turn facilitate the precipitation of Cu and Zn compounds in the topsoil (Degryse et al. 2006; Kabata-Pendias 2011; McBride 1994). Fertilization may also elevate concentrations in park soils. Focused research would be needed to understand these processes and relationships more thoroughly.

Environmental risk of gardening

In general, comparison of the pseudo-total concentrations of PTEs with risk thresholds do not indicate substantial risk. In the Singaporean context where gardening is an activity that is promoted as beneficial for the growing, aging community to be active and self-sufficient, our analysis indicates limited risk of exposure to contaminated soil. Most Singapore gardeners only spend a few hours per day in their gardens (Goh 2018); and therefore, inhalation and exposure rates are very low (Additional file 3: Table S5). For completeness, however, those concerned about health risks would

also need to consider all exposure routes, including the consumption of produce grown in gardens (visualized in Fig. 1). Others report that consuming the products of community gardens comprise about 60% of overall PTE exposure because some garden produce can bioaccumulate elements via root intake or foliar uptake (Spliethoff et al. 2016; Finster et al. 2004). Plants can potentially accumulate up to 10 times the concentration of Cd and a third of the concentrations of Cu, Zn and Pb in the soil (Kabata-Pendias 2011; Byrne et al. 1976). Dalenberg and van Driel (1990) state that atmospheric deposition is a significant source of Pb for field crops, accounting for 73–95% of total Pb found in plants.

Gardener activities such as applying fertilizers, pesticides, mulching, or soil amendment may influence the presence of elements in the soil (Mortvedt 1996; Atafar et al. 2010), but our analyses did not reveal a consistent association between element concentrations and soil variables in Singapore urban gardens. Nor did we identify sources of substantial contamination. Even for such cases of low levels of contamination, the presence of pollution sources (e.g., industry, road networks) necessitates that urban garden activities should be monitored from time to time to safeguard and inform the public of the level of risk, even if none (cf. Chaney et al. 1984; Kim et al. 2014). Patrons of parks and gardens should be aware that the presence of elements in the soil are a function of background residual signatures and potential enrichment via atmospheric deposition, surface runoff deposition, and application of soil amendments.

Finally, with respect to adverse health risk from consuming elements, bioavailability/inaccessibility should be considered to assess the potential for a toxicity effect if concentrations in soils and in farmed produce are high (Intawongse and Dean 2008). Again, we did not perform this assessment because of low pseudo-total PTE concentrations. We assume that the Singapore context is analogous to that of reported by Izquierdo et al. (2015) in Madrid with slightly higher levels of PTE contamination. They concluded that only in the worst-case scenario of children playing in urban gardens and eating produce grown from them, would risk exceed limits of acceptability. This assessment is in agreement with those of Sialelli et al. (2011) for urban soils in Torino (Italy), and Sipter et al. (2008) studying contaminated vegetables in Gyongyosoroszi (Hungary), but not of Bielicka-Giełdoń et al. (2013) in Koszalin (Poland) for conservative guidelines.

Limitations

We recognize that our elemental recovery methods were not sensitive enough to determine many low-level

concentrations associated with Cd and Pb samples. While this issue did not affect our ability to assess environmental risk to these elements, it did hinder our attempt to identify associations between all elements and soil physical factors. Further the recovery of Cu from the certified reference material was high, indicating our Cu determinations are potentially elevated. However, we did not determine the observed pseudo-total concentrations of Cu to be unrealistically high. With regard to the methods employed, others working in Singapore have had better luck deriving total concentrations using the techniques employing strong or mixed acids and analysis with an ICP-MS, particularly for low concentration elements such as cadmium (Yuen et al. 2012; Nguyen et al. 2018).

We are also somewhat uncertain of how the observed associations in soil pH, TOC, and texture play a role in element retention in garden soils of Singapore. For example, the negative correlation between elements (Cu and Zn) and clay content is inconsistent with most reports of higher element concentrations in clayey soils due to the greater presence of negatively-charged sites for element cation adsorption (Scokart et al. 1983; Quenea et al. 2009; Huang et al. 2014; Thomas and Lavkulich 2015). Nevertheless, some studies have reported greater PTE adsorption on coarse sediments in roadside dust and soils (Varrica et al. 2003; Wang et al. 2006; Acosta et al. 2011).

Finally, we were also limited by the inability to source the location of the soil material used in the construction of the gardens and parks. Doing so would have allowed us to fully understand element enrichment over the background concentrations that have taken place since construction of the gardens. Related, we also do not know the age of the gardens, with respect to element accumulation time.

Conclusions

While provenance tracking using lead isotopes indicated a range of potential sources for PTEs entering urban gardens, measured pseudo-total concentrations of Cd, Cu, Pb, or Zn did not indicate substantial health risk to those practicing urban gardening in Singapore. Further, estimated intake rates determined in a preliminary study (Goh 2018; Additional file 2: Table S5) were much below guideline specifications of maximum allowable doses, although the method to estimate these rates was coarse. Given the low concentrations of PTEs in the garden soils, we did not expand the analysis to determine element concentrations in produce grown in the gardens or the bioavailability/bioaccessability of the elements if ingested or inhaled. However, when combined with findings

from similar studies performing these assessments worldwide, the observed low pseudo-total concentrations observed leads us to conclude that urban gardening activities represent a viable and safe form of leisure, with the potential to produce food for consumption in Singapore. Risk would increase if vegetables in very contaminated soils were consumed, particularly by small children. These positive results add to the discussions on the viability and sustainability of urban agriculture in most urban settings—in agreement with Brown et al. (2016) and Lupolt et al. (2021) that the benefits often outweigh the risks. With respect to urban contamination in general, the growing work in Singapore, summarized in Table 5, supports the notion of a highly heterogenic signal of PTEs, whereby high concentrations associated with particular point sources, especially roads and industrial activities, are mapped onto a variable natural signal that has probably been also been enriched diffusely by the deposition of PTEs over time.

Abbreviations

Al: Aluminum; Cd: Cadmium; Cu: Copper; EF: Enrichment factor; Efm: Median enrichment factor; ERC: Elevated risk concentration; KW: Kruskal–Wallis test; MAD: Median absolute deviation; MW-U: Mann–Whitney U test; Pb: Lead; PTE: Potentially toxic element; PRC: Potential risk concentration; ρ : Spearman rank correlation coefficient; TRC: Threshold risk concentration; TOC: Total organic carbon; Zn: Zinc.

Supplementary Information

The online version contains supplementary material available at <https://doi.org/10.1186/s43170-022-00126-2>.

Additional file 1: Table S1. Ranges of Cd, Cu, Pb, and Zn concentrations reported for surface soils in (peri)urban garden soils worldwide (mg/kg; total/pseudo-total concentrations). **Table S2.** Raw data. **Table S3.** Summary of various health guidelines and threshold risk criteria for Cd, Cu, Pb, and Zn used worldwide.

Additional file 2: Table S4. Spearman rank correlation coefficients (ρ values) indicating associations between Cu, and Zn and soil clay fraction, silt fraction, sand fraction, total organic carbon (TOC), and pH. **Table S5.** Estimated exposure concentrations of copper, zinc, lead and cadmium to community gardeners compared to maximum allowable contaminants guidelines set out by Singapore AVA, WHO, and USA ATSDR. **Figure S1.** Relationship between Cu and Zn. The Spearman correlation coefficient is $\rho = 0.78$ ($\alpha < 0.001$). There are no meaningful correlations between other elements pairs. Shown in the figure is the linear regression equation and coefficient of determination (R^2).

Additional file 3: Figure S2. Map of Singapore showing the locations of many of the places mentioned in the discussions, including potential sources of contamination.

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Author contributions

GTA: as part of thesis work, conducted all phases of sampling, analysis, interpretation, and writing; ADZ: supervised the research, contributed to data analysis, and writing of the final manuscript; SJR: contributed to interpretation

of the findings and writing of final draft. All authors read and approved the final manuscript.

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Availability of data and materials

All new data are available in the supplement and at the following site: www.adziegler.com/data/Singapore_gardens_2020.

Declarations

Ethics approval and consent to participate

Not applicable. We did not do research on human subjects.

Consent for publication

Not applicable.

Competing interests

The authors declare that they have no competing interests.

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