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Heavy metals in suburban gardens and the implications of land-use change following a major earthquake

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ABSTRACT

Numerous studies have shown that urban soils can contain elevated concentrations of heavy metals (HMs). Christchurch, New Zealand, is a relatively young city (150 years old) with a population of 390,000. Most soils in Christchurch are sub-urban, with food production in residential gardens a popular activity. Earthquakes in 2010 and 2011 have resulted in the re-zoning of 630 ha of Christchurch, with suggestions that some of this land could be used for community gardens. We aimed to determine the HM concentrations in a selection of suburban gardens in Christchurch as well as in soils identified as being at risk of HM contamination due to hazardous former land uses or nearby activities. Heavy metal concentrations in suburban Christchurch garden soils were higher than normal background soil concentrations. Some 46% of the urban garden samples had Pb concentrations higher than the residential land use national standard of 210 mg kg^{-1}, with the most contaminated soil containing 2615 mg kg^{-1} Pb. Concentrations of As and Zn exceeded the residential land use national standards (20 mg kg^{-1} As and 400 mg kg^{-1} Zn) in 20% of the soils. Older neighbourhoods had significantly higher soil HM concentrations than younger neighbourhoods. Neighbourhoods developed pre-1950s had a mean Pb concentration of 282 mg kg^{-1} in their garden soils. Soil HM concentrations should be key criteria when determining the future land use of former residential areas that have been demolished because of the earthquakes in 2010 and 2011. Redeveloping these areas as parklands or forests would result in less human HM exposure than agriculture or community gardens where food is produced and bare soil is exposed.

1. Introduction

Heavy Metals (HMs), comprising metal(loids) with a density >5, can be toxic to organisms at relatively low concentrations (Robinson et al., 2009). The total HM concentration in soil is a function of the background concentration (Guagliardi et al., 2012) plus anthropogenic contributions, which include past and current application of soil conditioners (composts, manures and fertilisers), metal-containing agrichemicals, paints, vehicle emissions, local industries, and coal and fuel combustion (Paramashivam et al., 2016; Simmler et al., 2013; Szolnoki and Farsang, 2013).

Even small increases in concentrations of HMs such as cadmium (Cd) and lead (Pb) in soils may endanger the environment and human health (Ajmone-Marsan and Biasioli, 2010). Humans may ingest soil-borne HMs by direct consumption of soil or consumption of plants that either take up HMs into the edible portions or have HMs attached to the surfaces of the edible portions (Robinson et al., 2009). Exposure can also occur through inhalation of suspended soil particulates, and dermal contact (De Miguel et al., 1999; Kachenko and Singh, 2006). Children are particularly vulnerable because they have greater hand-to-mouth activities and gastrointestinal absorption (Calabrese et al., 1997). Childhood Pb poisoning remains a major environmental health concern in cities with Pb-contaminated soils (Ikem et al., 2008).

Urban and suburban soils are more likely than rural soils to become contaminated with HMs because they are more affected by human activities (Hough et al., 2004). Elevated HM concentrations occur in urban soils worldwide (Ajmone-Marsan and Biasioli,

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2010). Table 1 shows common heavy metal(loid)s that may be found at elevated concentrations in urban and suburban soils, along with their anthropogenic source.

El Khalil et al. (2013) showed that concentrations of copper (Cu), Pb and zinc (Zn) in soils from Marrakesh city, Morocco could be used as indicators of industrialisation. Similarly, areas of Annabeh city centre (Algeria) contained Pb, up to 823 mg kg^{-1} (Maas et al., 2010). Garden soils in Nantes, France, also revealed high arsenic (As) and Pb concentrations, and suggested human activities as the main origin (Jean-Soro et al., 2015). Analysis of soil samples from Madrid showed concentrations of Cd, Cu, Pb and Zn were inversely correlated with distance from the city centre (Vázquez de la Cueva et al., 2014).

Christchurch is New Zealand's third largest city with a population of some 390,000 (Statistics New Zealand, 2014). It was founded some 150 years ago and has vast tracts of suburban land (Wilson, 2005) where humans may be exposed to metals through home grown fruits and vegetables as well as direct ingestion of soil by children playing in backyard gardens. Major earthquakes in 2010 and 2011 have resulted in some 630 ha of the city being demolished with redevelopment of the affected land prohibited due to land stability issues (Scott and Carville, 2016). Potentially, this affected land may be used for agriculture, recreation, or community gardens, which may exacerbate human exposure to soil-borne HMs. A single study on a Christchurch residential red zone, Avon-Otakaro, showed that Pb was the only element, among As, Cd, Cu and Pb, exceeding the residential land use standard (260 mg kg^{-1}) (Gilmour, 2013).

This study aimed to determine the nature and extent of soil HM contamination in suburban and rural garden soils in an around the city of Christchurch as a function of the age of the district, with a view to identifying potential risks associated with home or community gardens. We sought to compare HM concentrations in Christchurch suburban soils with background concentrations in Canterbury soils (Tonkin and Taylor, 2007), as well as the New Zealand Soil Contaminant Standards for health (SCSs) for inorganic substances (Ministry for the Environment, 2012) and Dutch Standards.

2. Materials and methods

2.1. Soil sampling: suburban gardens

Crowd sourcing was used to obtain soils from 31 vegetable gardens in Christchurch city and surrounding areas in 2009–2010. Following advertisements in local newspapers, participants were sent labelled and sealable plastic bags along with instructions on soil collection. After removal of any surface litter or vegetation, ca. 250 g samples were collected from the top 10 cm of soil, representing the ‘A’ horizon in most soils, and certainly within the root-zones of most vegetable plants. For each garden, up to six samples were collected from different locations and individually analysed. Fig. 1 shows the sampling locations of the vegetable garden soils.

2.2. Soil sampling: suspected contaminated sites

The Canterbury Regional Council (CRC) is responsible for investigating land for the purposes of identifying and monitoring contamination. In conducting its regulatory duties, CRC created the Listed Land Use Register (LLUR) of sites it considers are potentially contaminated or known to be contaminated. A site will be classed as potentially contaminated if it previously or currently had an activity or land use occurring at the site which is present on the Ministry for the Environment (MfE) Hazardous Activities and Industries List (HAIL), which are activities it considers to be hazardous (MfE, 2016). HAIL sites include fuel storage sites, orchards, timber treatment yards, landfills, sheep dips and any other activities where hazardous substances could cause land and water contamination.

Soils from 99 sites where the residential buildings had suffered earthquake damage were sampled between 2012 and 2015. The sites were assessed as they were listed on the LLUR as potentially contaminated due to the land having been used previously for horticulture. The site investigation focused on the house rebuild footprint (within 1 m of the house) on ‘easily accessed areas’ (in other words, exposed or vegetated soils – not paved or otherwise covered). For each site, four separate soil samples (250 g) were collected from 0 to 0.3 m below ground level within or around the house footprint. The samples were collected using a stainless steel spade or hand auger, and placed directly into sample jars supplied by Hills Laboratories. The sampling equipment was decontaminated between samples. Fig. 1 shows the locations of the HAIL sites.

2.3. Soil analysis

Garden soils were dried at 105 °C and sieved to <2 mm using a Nylon sieve. Pseudo-total elemental analysis was carried out using microwave digestion in 8 mL of Aristar nitric acid (±69%), filtered using Whatman 52 filter paper (pore site 7 μm), and diluted with milliQ water to a volume of 25 mL and stored for chemical analyses. Concentrations of aluminium (Al), As, boron (B), Cd, Cr, Cu, iron (Fe), potassium (K), manganese (Mn), nickel (Ni), phosphorus (P), Pb, and Zn were determined using inductively coupled plasma optical emission spectrometry (ICP-OES Varian 720 ES). Mercury (Hg) was analysed using hydride generation coupled with the aforementioned ICP-OES. Wageningen reference soil (ISE 921) material was analysed for quality assurance. Recoverable concentrations were 91%–108% of the certified values. The HAIL soil samples were analysed by Hill Laboratories. Hill laboratories are an accredited laboratory (IANZ, 2017) and therefore have rigorous quality assurance procedures. Briefly, soils were air dried at 35 °C and sieved to <2 mm. These dried and sieved samples were then composited and

Table 1
Common heavy metal contaminants that may occur in urban or suburban soils (LaCoste et al., 2001; Mills et al., 2005; Robinson et al., 2006, 2009).

Heavy Metal(loid)	Source
Arsenic (As)	As-based insecticides used in horticulture, treated timber, or former sheep-dip sites
Cadmium (Cd)	Cd-rich phosphate fertilisers, sludges, industrial emissions, runoff from roads
Chromium (Cr)	Fixative in treated timber, industrial emissions
Copper (Cu)	Cu-based fungicide, Cu used for roofing
Mercury (Hg)	Industrial emissions, crematoria
Nickel (Ni)	Industrial emissions
Lead (Pb)	Historic use of leaded petrol, Pb-based paints, historic use of Pb-based pesticides. Pb flashings on roofs
Thallium (Tl)	Coal combustion
Zinc (Zn)	Galvanised metal

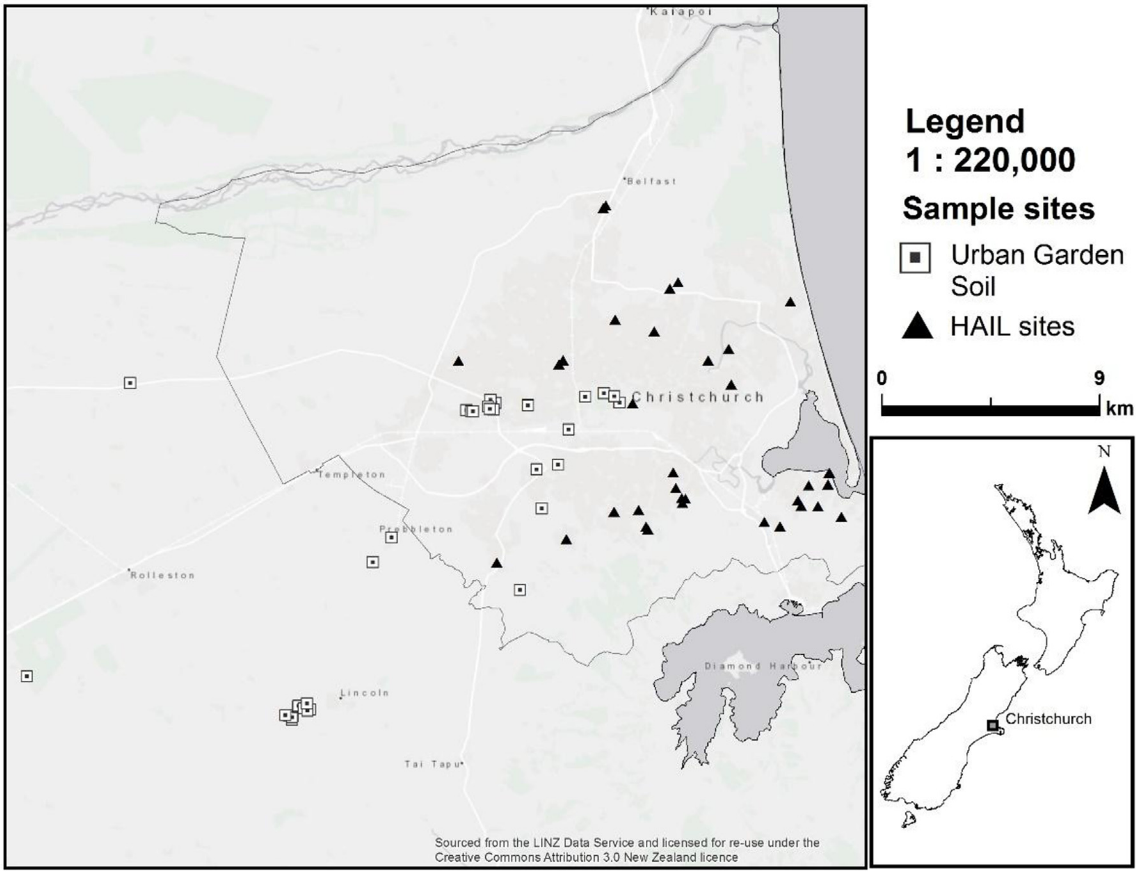

Fig. 1. Map of suburban and rural gardens as well as HAIL sites, where soil samples were taken.

Table 2
The concentrations of selected heavy metals in the Christchurch urban and rural gardens, and HAIL sites.

Element	Land Use	Number of Samples	Concentration (mg/kg)					
			Median	Mean	Range	Background[a]	National Standard[b]	Dutch Standard[d]
As	Rural	15	4.05	4.54	2.46–12.9	0.9–36.9	17	
	Urban	50	16	12.1	4.34–72.2	3.3–13.2	20	55
	HAIL	99	6	6.24	2–31			
Cd	Rural	15	0.58	0.86	0.27–9.69	0.01–0.34	0.8	
	Urban	50	1.54	1.20	0.33–10.7	0.01–0.15	3	12
	HAIL	99	0.11	0.11	<0.1–4.3			
Cu	Rural	15	9.25	9.50	4.32–22.9	2.1–27.3	>10,000	
	Urban	50	36.3	34.6	11.7–118	3.1–13.3	>10,000	190
	HAIL	99	14	15.8	3–2000			
Pb	Rural	15	15.7	18.8	7.96–77.6	3.63–44.4	160	
	Urban	50	123	137	22.6–2615	9.48–57.3	210	530
	HAIL	99	36	40.4	8.6–380			
Zn	Rural	15	68.4	67.3	42.1–120	12.1–116	–	
	Urban	50	216	189	67.5–799	26.6–91.7	400[c]	720
	HAIL	99	87	97.8	32–3700			
Hg	Rural	15	0.03	0.04	0.006–0.50	0.01–0.6	200	
	Urban	35	0.22	0.29	0.03–308	0.02–0.18	310	10
	HAIL	11	0.05	0.05	<0.1–0.25			
Cr	Rural	15	16.4	17.1	11.4–24.2	4.6–26.4	290	
	Urban	50	20.5	22.7	14.1–133	8.2–22.1	460	380
	HAIL	99	16	16.5	9–132			
Ni	Rural	15	8.6	9.69	6.64–18.1	2.9–20.7	–	
	Urban	50	11	11.4	8.77–18.2	6.3–15.6	105[c]	210
	HAIL	99	11	11.3	6–21			

[a] Background concentrations (level 1) of selected heavy metals in Canterbury soils reported by Environment Canterbury in 2007.
[b] New Zealand Soil Contaminant Standards for health (SCSs) for inorganic substances reported by Ministry for the Environment in 2012.
[c] Soil contaminant standards reported by Auckland Regional Council in 2010 (used where national standards are not available).
[d] Dutch soil pollutant standards (intervention values).

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analysed for 'total' heavy metals using a conc. HNO_3/HCl digestion and analysed using ICP-MS. While the detection limits for individual elements in dried soil were <0.05 mg/kg, we used a quantifiable limit of 0.1 mg kg^{-1} for all elements because concentrations below this value are unlikely to be biologically relevant.

2.4. Statistical treatment and analysis

Data that were log-normally distributed were log-transformed for statistical analyses. Data were considered lognormal when the geometric mean was closer to the median than the arithmetic mean. Statistical analyses were conducted using IBM SPSS 23 software. The significance of differences among the parameters was tested by one-way analysis of variance (ANOVA) followed by Tukey's test, at the 95% confidence level ($p < 0.05$). Correlations between concentrations of selected HMs were determined using Pearson correlation coefficient.

3. Results and discussion

3.1. HM concentrations in suburban and rural soils

Table 2 shows that suburban garden soils from Christchurch have significantly higher HM concentrations than rural soils, which is consistent with reports (Wong et al., 2006) that elevated HM concentrations are generally associated with urbanization. Specifically, concentrations of As, Cu, Pb, Zn and Hg were significantly higher in the urban garden soils than in soils of the rural gardens (Table 2). Mercury and Pb mean concentrations were >7 times greater, followed by Cu, As and Zn mean values some threefold greater. Wei and Yang (2010) also reported the largest ratios of urban-to-rural concentrations for Pb, Cu, Zn as well as Cd in soils of different areas of China. Here, mean concentrations of selected HMs in the urban garden soils were also higher than those in the HAIL sites. Mean Cd, Hg, and Pb concentrations were respectively 10, 5 and 3 times higher in the urban garden soils than in the HAIL sites (Table 2). This may be due to the fact that activities listed in the HAIL do not include common practices that contaminate soils, such as the widespread use of lead-based paint, the use of galvanised steer, or Cu-based pesticides (Ministry for the Environment, 2013).

Concentrations of Cu, Zn, Hg, Cr and Ni in all samples taken from the rural gardens were within the natural background ranges, whereas the Cd concentrations in 20% of these samples exceeded the rural national Cd standard (0.8 mg kg^{-1}) with the maximum value of 9.7 mg kg^{-1} (Table 2). Cadmium concentration in eight urban garden soil samples also exceeded the urban national Cd standard (3 mg kg^{-1}). While we did not measure mobility in this study, Li et al. (2017) reported that Cd was the most mobile element in urban soils from Lianyungang, China and that this element posed the greatest ecological risk.

The maximum concentrations of all other HMs were significantly higher than their respective background values in all the three land categories (rural garden, urban garden and HAIL). One urban garden contained a Hg concentration of 308 mg kg^{-1}, compared to its respective background concentration of 0.18 mg kg^{-1}. This concentration is thirtyfold higher than the Dutch Standard for Hg (10 mg kg^{-1}). Similarly, there were two HAIL sites containing high concentrations of Cu (2000 mg kg^{-1}) or Cd (4.3 mg kg^{-1}) compared to the Dutch and New Zealand Standards, respectively (Table 2).

Some 46% of the urban garden samples had Pb concentrations higher than the national standard of 210 mg kg^{-1}. The maximum Pb concentration (2615 mg kg^{-1}) found was 40 times higher than its background concentration, and more than 12 and 5 times greater than the national Pb standard (210 mg kg^{-1}) and Dutch Standard for Pb (530 mg kg^{-1}), respectively. Seven HAIL sites also exceeded the national standard for Pb. Moreover, 20% of the urban garden samples and 5 percent of the HAIL site samples exceeded the national standard for both As and Zn. The highest As concentration (72 mg kg^{-1}) in the urban garden samples was higher than both the national and Dutch Standard concentrations (20 and 55 mg kg^{-1} respectively). Similarly, the highest Zn concentrations among the HAIL and urban garden soil samples exceeded the Zn standard by 9 and 2 times, respectively. Most concentrations of HMs in rural gardens were within their respective background concentration ranges. However, the rural gardens contained significantly elevated Cd concentrations, with some samples in the range of 10 mg kg^{-1}, which is well above the New Zealand Cd standard. This is most likely due to extensive application of Cd-rich phosphate fertilisers in rural areas.

3.2. Correlation between HM concentrations

Correlation between HM concentrations across all samples showed that Zn was positively correlated with Cu and Cd concentrations in both the garden soils and HAIL sites (Table 3). This may be associated with the application of fertilisers, which can contain these three HMs (Romić et al., 2004) or more generally, represent the influence of human activity on soil contamination. Likewise, significant positive correlations between Ni-As, Ni-Cd and Cr-Pb occurred at the HAIL sites. In the garden soils, only Cr and Hg as well as Cd and Cr, Hg or As contents were not significantly correlated (Table 3) Gulan et al. (2017) also reported significant positive correlations between Zn and Pb for urban soils in Pristina, Kosovo. However, unlike our findings, this study also reported positive correlations between As and Cd. This difference is likely due to contrasting provenances of HMs in Christchurch and Pristina.

3.3. HM concentration as a function of neighbourhood age

HMs of anthropogenic origin would be expected to accumulate in soil over time (Hough et al., 2004), due to the activities listed in Table 1. Our data are consistent with this hypothesis because in general, soils from older neighbourhoods had significantly higher concentrations of HMs (specifically As, Cu, Pb and Zn) than those in younger neighbourhoods (Fig. 2). Of particular note is the significantly higher Pb concentrations in the pre-50s gardens, where the mean concentration (282 mg kg^{-1}) was well above the New Zealand Pb guideline (210 mg kg^{-1}). These soil Pb concentrations are similar to those found in Baltimore, Boston, and Chicago in the USA, and Naples, Palermo, and Rome in Italy (Ajmone-Marsan and Biasioli, 2010). Leaded gasoline and Pb-based paint were reported as the two main historical sources of the soil Pb contamination in the Boston gardens (Clark et al., 2006).

Table 3
The correlation between concentrations of selected heavy metals in the HAIL and garden soil samples.

Garden	HAIL							
	As	Cd	Cu	Pb	Zn	Hg	Cr	Ni
As		0.21*	0.02	0.02	0.15	−0.09	0.03	0.62**
Cd	0.11		−0.02	−0.08	0.94**	−0.12	−0.01	0.23*
Cu	0.61**	0.16*		−0.04	0.18*	−0.05	−0.02	0.04
Pb	0.51**	0.32**	0.55*		−0.08	−0.41	0.81**	−0.03
Zn	0.50**	0.34**	0.67**	0.88**		−0.14	−0.03	0.14
Hg	0.22**	0.07	0.17*	0.22**	0.28**		−0.20	−0.19
Cr	0.82**	0.04	0.59**	0.65**	0.54**	0.12		−0.02
Ni	0.20**	0.31**	0.33**	0.30**	0.36**	0.18*	0.26**	

**,*: Correlations are significant at 0.01 and 0.05 levels, respectively.

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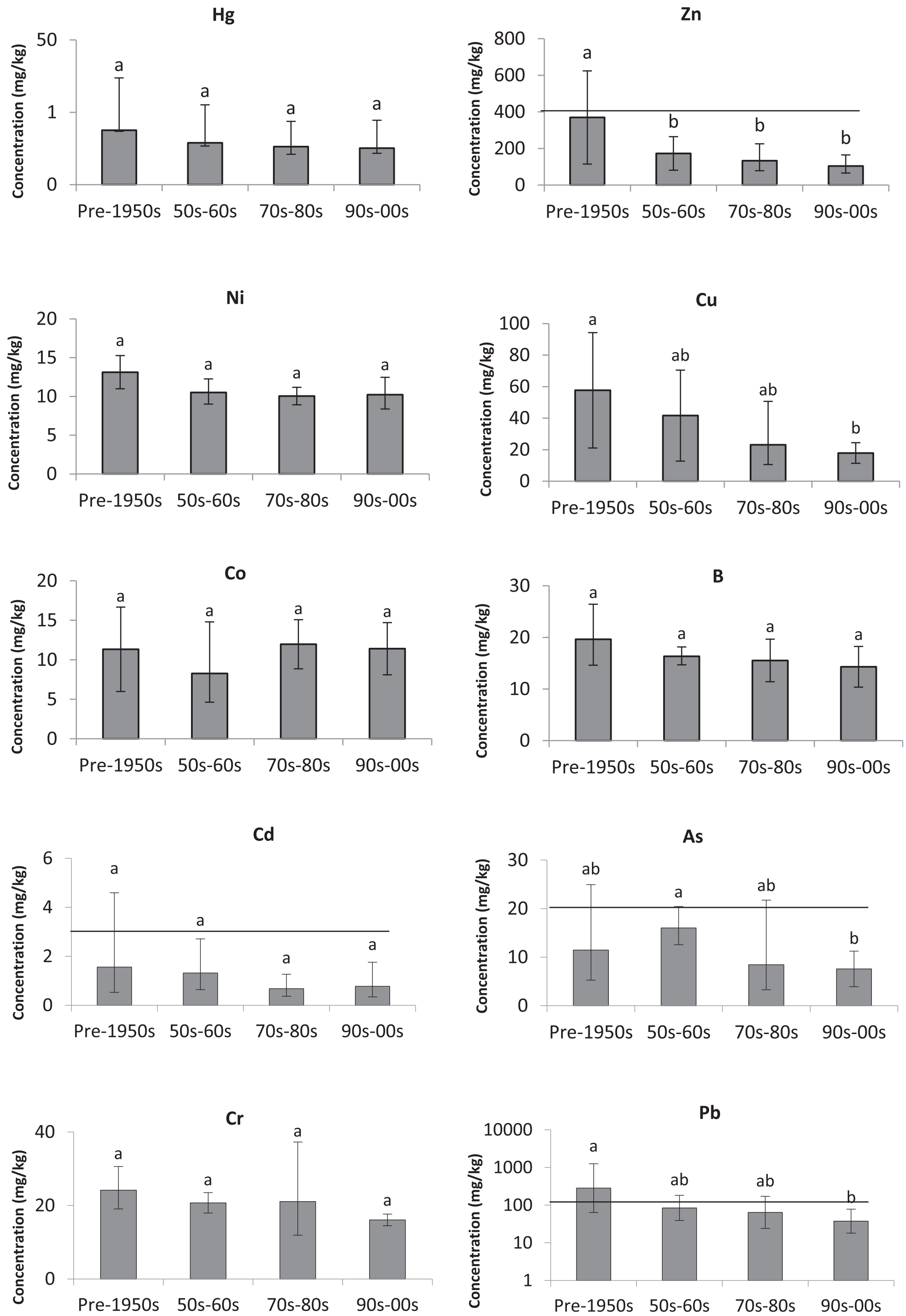

Fig. 2. The concentrations of selected heavy metals in soils of urban and peri-urban gardens made in Pre-50s (n = 21), 50s-60s (n = 16), 70s-80s (n = 11) and 90s-00s (n = 16). Bars labelled with the same letter are not significantly different ($P > 0.05$, Tukey's Test) and the horizontal lines represent the New Zealand contaminant standards.

3.4. *Implications of soil HM concentrations in Christchurch suburban soils*

As with other cities, a significant number of suburban soils from Christchurch gardens were above the national standards for HM concentrations, particularly soils from older neighbourhoods. Unlike some other cities, many suburban soils are used for food production. While we did not test food crops from suburban gardens in this study, there is clearly an increased risk that such crops may contain HM concentrations above food safety standards, which may result from plant uptake or attached soil particles. Vegetable HM contamination was found in vegetables grown in a Pb-contaminated community garden in Omaha, Nebraska (USA) (Sangster et al., 2012). Lead concentrations in samples of leafy greens, eggplant, okra, and tomato grown in soils containing more than 100 mg Pb kg^{-1} exceeded a limit of 0.5 mg kg^{-1} established by the U.S. Food and Drug Administration (FDA) for Pb. While the most concerning contaminants, As, Hg and Pb are not readily taken up by plants into their aerial tissues (Robinson et al., 2009), contaminated dust may be attached to edible portions, which is difficult to remove, even by repeated washing (Hinton et al., 1995). Root vegetables may accumulate significant concentrations of HMs (Alexander et al., 2006).

High soil HM concentrations may present a risk to local populations through the entry of dust indoors and its subsequent inhalation and ingestion (Laidlaw et al., 2005). Tong (1998) found elevated concentrations of Pb and Cu in both indoor and outdoor dust among residential properties in Cincinnati (Ohio, U.S.A.). She did not find a significant relationship between indoor and outdoor dust concentrations across the 121 sites analysed; however, these concentrations were higher than the background soil concentrations and were related, among other things, to the age of the building and the neighbourhood. Subsequent review by Laidlaw and Filippelli (2008) suggested that elevated blood Pb levels in children may be linked to soil transported into residences as dust; they also highlighted the importance of variables that influence the likelihood of wind erosion (e.g. soil moisture and ground cover) contributing to indoor dust levels.

Christchurch is regularly subject to strong seasonal winds and low levels of precipitation, which promote the formation of airborne soil particles. Moreover, there has been considerable demolition and reconstruction work in the Christchurch area subsequent to the earthquakes, which may have contributed to additional dust generation in some neighbourhoods. We did not analyse indoor dust samples, so we cannot comment on whether this may have increased the risk posed by high HM concentrations in household dust; however, this should be investigated further.

Human exposure to soil borne HMs can be limited by using soil conditioners that reduce HM bioavailability (Al Mamun et al., 2016; Clarke et al., 2015; Mitchell et al., 2014) or by selecting plants where the edible portions are unlikely to contain significant concentrations of HMs. Tree borne fruits are unlikely to have significant amounts of attached soil, as are fruits or vegetables that are peeled or contained within a capsule or pod. The highest risks would be associated with low-growing leafy vegetables or unpeeled root vegetables.

4. Conclusions

Soil HM concentrations should be key criteria when determining the future land use of former residential areas that have been demolished because of the earthquakes in 2010 and 2011. Redeveloping these areas as parklands or forests would result in less human HM exposure than agriculture or community gardens where food is produced and bare soil is exposed. The likely risk of any soil being contaminated is higher for older neighbourhoods. Future research could investigate HM concentrations in fruits and vegetables grown in suburban gardens in Christchurch. Communication of the results of this and subsequent studies to the public should be done in a way to avoid unwarranted hysteria: there is no evidence that Christchurch residents are suffering more ill effects of HMs than the populations of other cities.

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