BENCHMARKING ANTHROPOGENIC HEAVY METALS EMISSIONS: AUSTRALIAN AND GLOBAL URBAN ENVIRONMENTAL HEALTH RISK BASED INDICATORS OF SUSTAINABILITY

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ABSTRACT

In Australia, the impacts of urbanisation and human activity are evident in increased waste generation and the emissions of metals into the air, land or water. Metals that have accumulated in urban soils almost exclusively anthropogenically can persist for long periods in the environment. Anthropogenic waste emissions containing heavy metals are a significant exposure pathway for urban soils. The purpose of this paper is to present indicators of sustainability for assessing the environmental health risk from exposure of urban soils to anthropogenic waste emissions containing lead, copper, zinc and chromium. By benchmarking urban surface soil concentrations of these four metals against Australian and international Soil Standards, a data set of indicators of sustainability can be construed for evaluating the potential long-term environmental health risks posed by continued exposure of urban soils to heavy metals.

KEYWORDS

Heavy metals, benchmarking, urban soils, sustainable development (SD), environmental health indicators, anthropogenic emissions

1. INTRODUCTION

The present study focuses on the chemical dimension of soil quality. Soil contamination is a significant threat to sustainable soil management (European Commission, 2002a, 2006) and impacts the social and economic well-being of societies (Jónsson et al, 2016). A 'healthy' environment is defined on the basis of environmental risk factors by the World Health Organisation (WHO): 'Environmental risk factors, such as air, water and soil pollution, chemical exposures, climate change, and ultraviolet radiation, contribute to more than 100 diseases and injuries' (WHO, 2016). Human and environmental health are coupled; human health and well-being cannot be considered in isolation of the environment: 'An estimated 12.6 million people died as a result of living or working in an *unhealthy* environment in 2012 – nearly 1 in 4 of total global deaths' (WHO, 2016). The aphorism that human and environmental well-being are 'coupled' applies to resource consumption and economic growth; societal activities (creating 'economic gains') reliant on resource consumption (with attendant waste emissions) to sustain economic growth are inextricably coupled (Daly, 1995; El Serafy, 2006).

In 1987 the members of the World Commission on Environment and Development (WCED) perceived humanity's predicament at the time to be sufficiently dire as to issue the warning that 'the same processes that have produced these gains have given rise to trends that the planet and its people cannot long bear' (WECD, 1987, p.12). Since the issue of this 'warning' almost 30 years ago, background (natural) levels of metals including lead (Pb), copper (Cu), zinc (Zn) and chromium (Cr) in air, water and soil have increased by anthropogenic flows of metals into the ecosphere by waste disposal and other societal activities (Hamon et al, 2004). Establishing background concentration for heavy metals has been difficult owing to many decades (and centuries) of anthropogenic release of metals (Hamon et al, 2004). Regional and urban background metal concentrations in Australia are essential data for gauging pollution levels and signaling need for intervention when environmental and human health thresholds are reached (NEPM, 2013).

Major anthropogenic airborne sources of lead Pb, Cu, Zn and Cr pollution occur via direct emission or loss into the ecosphere (European Commission, 2002b). The hexavalent (Cr-VI) form of chromium, which is toxic by inhalation and has been classified as a Class A inhalation carcinogen (IARC, 1990) and, environmentally speaking, Cr(VI) compounds are generally considered the most toxic (Shanker et al, 2005; Zayed and Terry, 2003). Major anthropogenic sources of atmospheric Cr(VI) are presented in Table 1. Windblown dusts contaminated with heavy metals (including the Cr-VI species of chromium) emanating from urban soils are a significant source of emissions to air in urban environments.

Table 1. Top 20 sources of chromium emissions to air, land and water in Australia (1999-2014)

Rank	Source
1.	Aeroplanes
2.	Basic Ferrous Metal Manufacturing
3.	Basic Non-Ferrous Metal Manufacturing
4.	Burning (fuel red., regen., agric.)/ Wildfires
5.	Cement, Lime, Plaster and Concrete Product Manufacturing
6.	Ceramic Product Manufacturing
7.	Coal Mining
8.	Commercial Shipping/Boating
9.	Electricity Generation
10.	Fuel Combustion - sub reporting threshold facilities
11.	Gaseous fuel burning (domestic)
12.	Lawn Mowing
13.	Metal Ore Mining
14.	Motor Vehicles
15.	Other Transport Equipment Manufacturing
16.	Paved/ Unpaved Roads
17.	Pulp, Paper and Paperboard Manufacturing
18.	Recreational Boating
19.	Water Transport Support Services
20.	Windblown Dust

Source: (NPI, 2016).

Notes: -For National Pollutant Inventory (NPI) reporting purposes, emissions are defined as the release of an NPI substance to the environment whether in pure form or contained in other matter and/or in solid, liquid or gaseous form. It includes the release of substances to the environment from landfill, sewage treatment plants and tailings dams (National Pollutant Inventory, DEH, 2006-07 Report, p.18).

-Approximately $75x10^3$ tonnes of chromium is emitted globally into the atmosphere annually by these sources with approximately one-third occurring as the Cr(VI) species (Kieber et al, 2002; Pacyna and Nriagu, 1988).

1.1 Geochemical Indicators: Rationale for the Study

Global urban and rural soil investigations have shown that anthropogenic waste outputs contaminate soil in cities including: Minneapolis, USA (Mielke et al, 1984); Berlin, Germany (Birke and Rauch, 1997; Mekiffer et al, 2000); Aberdeen, Scotland (Paterson et al, 1996); Wolverhampton, England (Hooker et al, 1996; Bridge et al, 1997); Birmingham, England (Wang et al, 1997); Tallinn, Estonia (Bityukova et al, 2000); Trondheim, Norway (Ottesen and Langedal, 2001); Karlsruhe, Germany (Norra et al, 2001; Norra and Stuben, 2003); Gainesville and Miami, Florida, USA (Chirenje et al, 2003); Gibraltar (Mesilio et al, 2003); Newcastle upon Tyne, England (Pless-Mulloli et al, 2004); the Totley suburb of Sheffield, England (Knight, 2004); and Seville, Spain (Madrid et al, 2004).

These geochemical studies have focused on soil contamination of large cities with particular emphasis on metals such as Pb, mercury (Hg), Cu and Zn. There is a paucity of research on the environmental health risks of atmospheric deposition of Cr(VI) and long-term accumulation in the ecosphere, particularly urban soil studies of Cr(VI) contamination with accompanying human health risk based indicators of sustainability. This study aims to address this research gap by a systematic evaluation of extant data on anthropogenic accumulation of Pb, Cu, Zn, Cr and Cr(VI) in urban soils and positing a data set of environmental health risk indicators (vis-a-vis urban soil studies) by benchmarking geochemical data (Pb, Cu. Zn and Cr soil concentrations) in 13 cities against Australian (NEPM, 2013) and international (CCME, 1999; CEPA, 2005) Soil Standards.

2. METHOD FOR DEVELOPING INDICATORS OF SUSTAINABILITY

Table 2 summarises the geochemical studies used for calculating environmental health risk indicator scores for Pb, Cu, Zn and Cr.

Table 2. Australian and global geochemical studies used for calculating health risk indicator scores for Pb, Cu, Zn and Cr

Study		Author(s)
Australia	Sydney (estuary catchment)	[a]Birch & Vanderhayden, 2011
	Iron Cove sub-catchment	Snowdon and Birch 2004
	Homebush Bay sub-catchment	[b]Hodge, 2002
	Parramatta sub-catchment	[c]Olmos, 2004
	Wollongong City area	Beavington, 1973
Global	Seoul, Vietnam	Chon et al, 1995
	Danang–Hoian Area, Vietnam	Thuy et al, 2000
	Berlin metropolitan area	Birke and Rauch, 1997
	Great Britain	Culbard et al,1988
	Oslo, Norway	Tijhuis et al, 2002
	Xuzhou, China	Wang and Qin 2007
	Madrid, Spain	De Miguel et al. 1998
	Glasgow, Scotland	Gibson and Farmer 1986

[a] Birch & Vanderhayden, 2011; soil metal concentrations shown for 50th percentile, normalised

[b] Unpublished study by Hodge (2002) data cited in Birch and Vanderhayden (2011)

[c] Unpublished study by Olmos (2004) data cited in Birch and Vanderhayden (2011)

2.1 Principles and Precepts of Sustainable Development

Sustainable development (SD) translates into a series of ‘precepts’ that should be followed to prevent both the decline in the quantity and quality of ‘natural stocks’ including soils (Lawn, 2006). Holmberg et al (1996) formulated indicators underpinned by four principles ‘that should be fulfilled by a sustainable society’. The principles are:

Precept 3: The rate of high entropy waste generation should not exceed the ecosphere’s waste assimilative capacity (Lawn, 2006)

Principles 1-2: Substances extracted from the lithosphere must not systematically accumulate in the ecosphere**;** and society-produced substances must not systematically accumulate in the ecosphere (Holmberg & Eriksson, 1996**;** Azar et al, 1996).

These principles and precepts alone do not directly make provision for ‘measuring’ the ‘sustainable development’ of a nation; they provide parameters for indicators and measures to be formulated. For the purpose of facilitating the development of environmental risk indicators as measures of SD in Australia and globally, a conflation of precept 3 (Lawn, 2006) and principles 1 and 2 will be the reference for formulating indicator scores for the four elements. A conflation of the sustainability principles and precept leads to the following formulation:

Formulation 1

The sum of the anthropogenic emissions and the natural flows from the lithosphere to the ecosphere should not exceed the ecosphere’s waste assimilative capacity.

2.2 Method for Establishing Background (Natural) Levels of Cu, Pb, Zn, Cr

Table 3 shows: the background ranges used for calculating indicators; presents Australian and international Soil Standards Health Investigation Levels (HILs) data; and provides three sets of background ranges posited by different authors (Berkman, 1989; Hamon et al, 2004; Shanker et al, 2005; Zayed and Terry, 2003).

Table 3. Benchmarking 13 geochemical studies of soil metal concentrations against soil standards

Metal	Soil Standard			[d]Background (Natural) ranges (N_n)			[h]Geochemical Study (anthropogenic flows) (A_{1-13})												
	[a]NEPM	[b]CCME	[c]CEPA	[e](N_1)	[f](N_2)	[g](N_3)	1	2	3	4	5	6	7	8	9	10	11	12	13
Cu	1000	63	3000	2-100	<70	-	60	170	81	74	343	84	90	43	54	32	38	72	97
Pb	300	140	80	2-200	<40	-	150	1069	227	217	21	240	84	78	240	56	43	161	216
Zn	7000	200	23000	10-300	<130	-	259	927	281	341	82	271	153	159	260	160	144	210	207
Cr	-	-	-	5-1000	-	65	[i]50.0	-	-	-	-	-	[i]92.2	-	-	[i]32.5	[i]78.4	[i]74.7	-
Cr(VI)	100	0.4	17	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-

Notes: -All metal soil concentrations are mean concentrations shown as mg/kg.

[a]Australian Soil Investigation Standard (Residential HIL A) (NEPM, 2013)

[b]Canadian Standard for residential soil (CCME, 1999)

[c]Californian Standard for residential soil (CEPA, 2010)

[d]Background ranges are natural (N_n) environmental flows. Values shown account for natural flows of the element from weathering and volcanic eruptions

[e] (Berkman, 1989)

[f](Hamon et al, 2004). Background ranges posited by Hamon (et al, 2004) vary depending on iron (Fe) levels in soil; N_2 ranges provided are for soils containing 10% Fe

[g](Shanker et al, 2005; Zayed and Terry, 2003). The mean background total chromium concentration of 65mg/kg is derived from US, Canadian, Japanese and Swedish soils

[h]Metal concentrations are anthropogenic (A_1) environmental flows determined from the 13 soil studies in Table 2.

[i]Total chromium (non-speciated) concentration shown.

Geochemical studies: 1 (Birch & Vanderhayden, 2011); 2 (Snowdon & Birch 2004); 3 (Hodge, 2002); 4 (Olmos, 2004); 5 (Beavington, 1973); 6 (Chon et al, 1995); 7 (Thuy et al, 2000); 8 (Birke & Rauch, 1997); 9 (Culbard et al, 1988); 10 (Tijhuis et al, 2002); 11 (Wang & Qin, 2007); 12 (De Miguel et al, 1998); 13 (Gibson & Farmer 1986)

2.3 Method for Creating Environmental Health Risk Indicators

Environmental health risk indicator scores I_1, I_2 and I_3 are calculated as anthropogenic metal flows from the lithosphere to the ecosphere relative to the corresponding background ranges N_1, N_2 and N_3. The relationship between the flow of elements from the lithosphere into the technosphere, and from the technosphere into the ecosphere can be shown as:

$$I_{(1,2,3)} = A_{(1\ldots13)} / N_{(1,2,3)} \qquad \text{(Equation 1)}$$

Where ***I***= Indicator score; ***A***=Anthropogenic flows (based on geochemical studies measuring soil metal concentrations in mg/kg-see Table 2); and ***N***=Natural flows (based on data sets positing naturally occurring background soil metal concentrations in mg/kg- see Table 3) (Construed from Azar et al, 1996).

If the indicator scores calculated from geochemical data for the cities in Table 2 are greater than unity, the ‘conditions for sustainability’ embedded in formulation 1 are not met; the inference being:

If $I_{N1(1\ldots13)}$, $I_{N2(1\ldots13)}$ ***and*** $I_{N3(1\ldots13)}$ *scores are >1,* ***then*** *the current rate of anthropogenic outputs and deposition in the ecosphere of Pb, Cu, Zn and Cr is not sustainable in those cities (and the risk to human health vis-à-vis contaminated soils is increased through continued exposure to Pb, Cu, Zn and Cr through anthropogenic release into the ecosphere in those cities).*

3. FINDINGS AND DISCUSSION

Table 4 shows environmental health indicator scores $I_{N1\ (1\ldots13)}$, $I_{N2\ (1\ldots13)}$ and $I_{N3\ (1\ldots13)}$ derived from urban surface soil levels of Pb, Cu, Zn and Cr and Cr(VI) relative to background levels. The indicators in Table 4 lie in the range 0.32-26.72; the wide range is due to the Iron Cove sub-catchment determination for the metal Pb (26.72 mg/kg) being an outlier (Snowdon and Birch, 2004). Where the indicators in Table 4 are equal to unity, then the present soil metal concentrations are sustainable (the conditions for SD embedded in formulation 1 are met) posing less risk to soil contamination in urban environments. Indicator scores of greater than unity indicate metal accumulation in the environment at a rate that is greater than the attendant assimilative capacity of the environment; the inference drawn here is continued contamination of urban soils with Pb, Cu, Zn and Cr and Cr(VI) is not sustainable (the conditions for SD embedded in formulation 1are not been met). Environmental risk is proportional to the indicator score; indicator scores orders of magnitude greater than unity are evident for several studies: the Iron Cove sub-catchment study indicating levels of Zn>1($I_{2(2)}$=7.13) and Pb>1($I_{1(2)}$=5.35) (Snowdon and Birch 2004); the Wollongong study indicating levels of Cu >1 ($I_{1(e)}$=3.43) (Beavington,1973); the Madrid Study indicating levels of Pb>1 ($I_{2(12)}$=4.03) (De Miguel et al,1998); and the Glasgow study indicating levels of Pb>1 ($I_{2(13)}$=5.40) (Gibson and Farmer,1986).

The indicator scores in Table 4 reveal: the conditions for SD embedded in formulation 1 have not been met for 90% of the cities on the basis of N_2 (Hamon et al, 2004) background levels for Pb and Zn; and environmental health indicator scores of greater than unity are observed for 12 of the 13 cities on the basis of N_2 background concentrations. On the basis of N_1 background concentrations, 6 cities (Sydney, estuary catchment); Danang–Hoian Area, Vietnam; Berlin metropolitan area; Oslo, Norway; Xuzhou, China; and Madrid, Spain) met the conditions for SD in formulation 1 when assessed for Pb, Cu and Zn.

Table 4. Environmental Health Risk Indicator Scores

Study	**Cu**			**Pb**			**Zn**			**Cr**		
	$I_1=A_1/N_1$	$I_2=A_1/N_2$	$I_3=A_1/N_3$	$I_1=A_1/N_1$	$I_2=A_1/N_2$	$I_3=A_1/N_3$	$I_1=A_1/N_1$	$I_2=A_1/N_2$	$I_3=A_1/N_3$	$I_1=A_1/N_1$	$I_2=A_1/N_2$	$I_3=A_1/N_3$
1	0.60	0.86	-	0.75	3.75	-	0.86	1.99	-	-	-	0.77
2	1.70	2.43	-	5.35	[a]26.72	-	3.09	7.13	-	-	-	-
3	0.81	1.16	-	1.14	5.68	-	0.94	2.16	-	-	-	-
4	0.74	1.06	-	1.09	5.43	-	1.14	2.62	-	-	-	-
5	3.43	4.90	-	0.11	0.53	-	0.27	0.63	-	-	-	-
6	0.84	1.20	-	1.20	6.00	-	0.90	2.08	-	-	-	-
7	0.90	1.29	-	0.42	2.10	-	0.51	1.18	-	-	-	1.42
8	0.43	0.61	-	0.39	1.95	-	0.53	1.22	-	-	-	-
9	0.54	0.77	-	1.20	6.00	-	0.87	2.00	-	-	-	-
10	0.32	0.46	-	0.28	1.40	-	0.53	2.00	-	-	-	0.50
11	0.38	0.54	-	0.22	1.08	-	0.48	1.11	-	-	-	1.21
12	0.72	1.03	-	0.81	4.03	-	0.70	1.62	-	-	-	1.15
13	0.97	1.39	-	1.08	5.40	-	0.69	1.59	-	-	-	-

Notes: -Sustainability indicators ('scores') determined by benchmarking geochemical investigations of soil metal concentrations against NEPM, CCME and CEPA Soil Standards.

-Indicator scores greater than unity are shaded. An Indicator scores of >1 specifies that the conditions for sustainable development embedded in Formulations 1 and 2 have not been met.

[a]An outlier (Snowdon and Birch, 2004).

Table 5 summarises the percentage of geochemical studies showing metal concentrations that are greater than unity relative to the posited background ranges; 15.4%, 46.2% and 15.4% of geochemical studies showed that anthropogenic flows for Pb, Cu and Zn were greater than natural flows when compared to N_1 background data (Berkman, 1989). Environmental health indicator scores for 61.5%, 92.3% and 92.3% of geochemical studies showed that anthropogenic flows for Pb, Cu and Zn respectively were greater than natural flows when compared against N_2 background data (Hamon et al, 2004).

Table 5. Percentage of geochemical studies within standard EIL limits and background range

Metal	**% of geochemical studies showing metal concentrations > NEPM standard**	**% of geochemical studies showing metal concentrations > CCME standard**	**% of geochemical studies showing metal concentrations > CEPA standard**	**% of geochemical studies showing metal concentrations > N_1**	**% of geochemical studies showing metal concentrations > N_2**	**% of geochemical studies showing metal concentrations > N_3**
Cu	0	61.5	0	15.4	61.5	
Pb	7.7	61.5	61.5	46.2	92.3	
Zn	0	61.5	0	15.4	92.3	
Cr	-	-	-	-	-	60
[a]Cr(VI)	0	100	100	-	-	

[a]Environmental health risk indicator scores ($I_3=A_1/N_3$ in Table 4) for Cr are used as a basis for benchmarking Cr(VI) against the Soil Standards. The resulting percentages are based on the probability that approximately one-third of the atmospheric releases of chromium are believed to be in the hexavalent form (Kieber et al, 2002; Pacyna and Nriagu, 1988).

Construing the indicator scores (Table 4) and the percentage of geochemical studies outside of EIL limits (Table 5) together, the inferences drawn from these data are: Pb, Cu and Zn from the lithosphere have spread at a rate which has given rise to a systematic increase in the ecosphere thereby implicating the soils in all of the cities forming part of this study; and on the basis of N_3 background levels, Cr(VI) has spread in cities located in Australia, Vitenam, Norway, China and Spain at a rate that potentially increases risk to human health through long-term exposure and inhalation of Cr(VI) contaminated dust.

4. CONCLUSION AND RECOMMENDATIONS

In Australia there is a significant challenge with the quality of voluntarily reported data from emitters of Cr and the other metals. Cr(VI) emissions and total chromium data have large associated errors due to the reliance on emitters measuring and reporting quantitative data accurately. According to the Australian National Pollutant Inventory (NPI, 2016), the total emissions levels are lower than actual due to: the suspected low capture rate (possibly up to 50%) of potential industry emitters voluntarily reporting data; and out of date diffuse source estimates and lack of uniform reporting of diffuse sources by jurisdictions (DEH, 2005). It is therefore highly probable that the environmental health indicator scores posited are lower than actual.

Under reporting of emissions has implications for human health as it is plausible that soil contamination with Cr(VI) and Pb, when benchmarked against Soil Standards (CCME, 1999; CEPA, 2010) have exceeded human toxicity thresholds in cities including Sydney (Birch & Vanderhayden, 2011), Hoi An in Vietnam (Thuy et al, 2000), Xuzhou in China (Wang and Qin, 2007) and Madrid in Spain (De Miguel et al, 1998).

The findings of this study recommend a deeper analysis and review of the current approach to the Australian Human Health Risk Assessment framework (NEPM, 2013).The current definition of ‘potential impact’ is too broad; a narrow, specific definition accompanied by provisions in the NEPM framework for the long-term environmental monitoring of air and soil Cr(VI) levels would benefit urban populations given that continued anthropogenic release of such metals is unlikely to decrease significantly soon. Goodland and Daly (1996) conflate equity and sustainability; ‘sustainability indeed has an element of not harming the future’ (Goodland and Daly, 1996). Between 1996–97 and 2006–07, the volume of waste produced per person in Australia grew at an average annual rate of 5.4%. Australians generated approximately 1,200kg of waste per person in 1996–97 and this increased to 2,100kg per person in 2006–07 (Productivity Commission, 2006).

On the basis of the data indicating a trend of continually increasing consumption and waste generation, the impacts of continued anthropogenic release of Pb, Cu, Zn and Cr, and particularly Pb and Cr(VI), into the ecosphere are likely to result in long-term harm, especially to the health of future generation dwelling in urban areas. Further research is recommended to assess the long-term environmental and human health impacts of urban soil contamination with heavy metals.

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