

Identifying Sources, Determining Exposure Risk and Assessing Management Strategies of Environmental Contamination in Australia

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BEnv. (Hons)

Thesis Presented for the Degree of
Doctor of Philosophy

Department of Earth and
Planetary Sciences

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Acknowledgements

First and foremost, thank you to Associate Professor Heather Handley and Professor Mark Taylor. Without your guidance, passion and dedication for the topic, this thesis would not have been possible.

Secondly, thank you to my family, Wendy, Peter, Nathan, Angie and Cindy. Your continued and unwavering support has been a huge source of strength in completing this thesis.

Those who have been a mentor throughout my PhD: Associate Professor Norman Pearson, Dr Richard (Dick) Flood, Associate Professor Kelsie Dadd, Professor Damian Gore, Professor Brian Gulson, Dr Anthony (Tony) Morrison and Dr Mark Lackie, your smiling faces and big hearts go a long way - thank you.

Thank you to the team: Chenyin Dong, Dr Louise Kristensen, Marek Rouillon, and Liqin Wu. What adventures we have had!

To everybody who has been a support, big or small along the way, a big thank you: Ron Aggs, Dr Olivier Alard, Sandy Aureli, Dr Danielle Camenzuli, Bruno Colas, Sophia Bratenkov, Beñat Oliveira Bravo, Russell Field, Sarah Gain, Louise Goode, Dr Yoann Gréau, Hadrien Henry, Anthony Lanati, Nóra Liptai, Dr Nicole McGowan, Stuart Mead, Dr Rosanna Murphy, Tetsuya Okada, Dr Montgarri Castillo Oliver, Phoebe Peterson, Dr Ed Saunders, Emma Singh, Catherine Stuart, Romain Tilhac, Aditi Verma, Yu Wang, Peter Wieland, Nigel Wilson and anybody else I have forgotten.

To the wonderful people (past and present) at the National Measurement Institute: Dr Luke Baker, Dr Andrew Evans, Dr Jasminka Jaksic, Dr Danny Slee, Dr Michael Wu and all the team, thank you for sharing your labs and your knowledge to facilitate the research in this thesis.

Finally, thank you to all the welcoming, friendly and endlessly hospitable people that I have met travelling Australia for this research.

Funding is acknowledged from a Macquarie University Research Excellence Scholarship (MQRES2012195) awarded to P.J. Harvey in association with an Australian Research Council Future Fellowship (FT120100440) awarded to H.K Handley.

Statement of Originality

This is to certify that the work in this thesis has not been submitted for a higher degree to any other university or institution. This thesis contains original work of myself and does not contain work published or written by another person, except where acknowledged.

Paul James Harvey

Abstract

Environmental contamination is a globally ubiquitous problem and is one which generates huge ongoing economic and health burdens for impacted communities. This thesis investigates the breadth of the environmental contamination problem in Australia through a number of case studies. Research was conducted across three themes: at the household, in agriculture and city-wide, to illustrate how environmental contamination impacts a wide population. The range of inter-related case studies include contamination of drinking water and associated environments by metal(loid)s in plumbing infrastructure in rural Tasmania and New South Wales. Followed by examination of the risks of metal(loid) exposure presented to household gardeners in urban gardens and the pollution of watersheds from poorly regulated intensive farming operations. The three final studies examine contamination at the city scale: surrounding the former Pasminco Cockle Creek Smelter and assessment of its soil remediation strategy, soil dust contamination surrounding a former antimony processing facility and the city-wide metal(loid) and polycyclic aromatic hydrocarbon contamination of soils in the city of Newcastle. Each of the case studies develop and expand on a multiple lines of evidence approach for identifying the source(s) of environmental contamination when ambiguity in source(s) prevents adequate management response and ultimately clean-up. In addition, the case studies presented in this thesis adopt the concept of citizen science and so commentary on the use of citizen science and engaging the community in scientific investigation to enhance the outcomes and impact of research is presented.

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1 Introduction

1.1 Defining Contamination

Environmental contamination and pollution are the central focus of this thesis. While contamination and pollution are often used interchangeably, there are critical differences between them. Contamination is defined as the **presence** of a substance in the environment when it should not be there, or, when it is in concentrations above what would be considered background for a given environment (Chapman, 2007). Subsequently, contamination is most simply a physical characteristic of an environment. In contrast, pollution describes substances in the environment that should not be there (contamination) and that have an (adverse) **impact** on the biological and health functions of an organism (Chapman, 2007). Pollution is therefore, according to this definition, multi-dimensional and captures the interaction of a range of organisms, particularly humans, with the environment. To complicate the meaning of pollution, Australian legal definitions remove the health-exposure related aspect, as shown here by the *Protection of the Environment Operation Act 1997* (NSW) which defines pollution as:

“an incident or set of circumstances during or as a consequence of which there is or is likely to be a leak, spill or other escape or deposit of a substance, as a result of which pollution has occurred, is occurring or is likely to occur. It includes an incident or set of circumstances in which a substance has been placed or disposed of on premises, but it does not include an incident or set of circumstances involving only the emission of any noise.”

This thesis will focus primarily on literature defined environmental contamination – those substances that are foreign to an environment. Much of the work in this thesis will demonstrate that, typically, environmental contamination is in-fact pollution, and that it has substantial human health impacts.

1.2 A Brief History of Environmental Contamination

The life-long exposure of living organisms to contaminants in the environment, in the form of industrial chemicals, was poignantly described by Rachel Carson in her 1962 text *Silent Spring* (Carson, 1962):

“For the first time in the history of the world, every human being is now subjected to contact with dangerous chemicals, from the moment of conception until death”

Although Carson (1962) focused on the manner in which pesticides were applied haphazardly to the environment in an endless pursuit to improve crop yields in agriculture, the sentiment of this work which highlights the link between the foreign substances, contaminants, and the health impacts that they may have is applicable across the spectrum of environmental contaminants. The extensive observations of Carson (1962) documenting the health impacts of pesticides revealed that in addition to their role as environmental contaminants, these substances also acted as environmental pollutants. Well ahead of her time, Carson’s speculation about the inter-generational impacts of environmental pollutants thrust the topic into the scientific vernacular, leading to the contemporary environmental movement (Lytle, 2007). Despite the warnings from Carson (1962), we still see today, in the form of emission of metal(loid)s and chemicals into the environment, the results of human disregard for both the environment and health throughout the world.

Ultimately, policy and decision makers control the emission and fate of substances that become environmental contaminants. This occurs through policy, regulation and legislation. This is best demonstrated in the United States of America (USA), where the governmental support for the lead industry during the 1900’s led to the creation of one of the world’s longest running, pervasive and damaging incidence of environmental contamination in history (Markowitz and Rosner, 2000, Needleman, 1997, 1998, 2000). What has been described as the ‘aggressive industry and government fuelled cover-up’ of the impacts of lead exposure and the simultaneous marketing of everyday commodities containing lead (Figure 1.1) has resulted in generations of ongoing social welfare issues, racial inequality and detrimental health outcomes (Leech et al., 2016, Needleman, 2005). The World Health Organisation (WHO) global statistics predict that on an annual basis 143,000 lead poisoning related deaths occur (based on 2004 data) and 600,000 children develop a lead related intellectual disability (WHO, 2010). Bellinger (2016) commented on the institutional decision to allow the widespread use of lead in products including leaded fuel and paint saying:

“One would be challenged to design a better strategy for maximizing population exposure to a poison than to have it emitted by a ubiquitous mobile source and to line the surface of dwellings with it”



Figure 1.1 Advertising for lead products available in Australia during the early 1900's.

Although the lessons learned from the prolific global usage of lead have changed the way lead is presently managed in industry and the environment, there persists a worldwide problem whereby a range of agents of concern to human health, including metal(loid)s (e.g. arsenic), and more complex compounds (e.g. perfluorinated chemicals) are released into the environment with limited or no regulation and control (Cousins et al., 2016, Tweedale, 2017). Poor or absent urban planning often exacerbates this problem as unregulated industrial activities generating large volumes of waste and emissions are situated within major population hubs (Caravanos et al., 2013). The WHO estimates that approximately 8.9 million people die each year, many in low- and middle-income countries, as a result of exposure to environmental contaminants (Landrigan and Fuller, 2015).

1.3 Global Burden of Disease from Environmental Contamination

Extensive environmental research, particularly in the USA, has shown strong correlations between negative human health outcomes and heavy metals, pesticides, herbicides, various fire

retardants and industrial chemicals among others (e.g. Lanphear, 2015). The international literature provides an insight into the detrimental health impacts of environmental contamination and pollution and to demonstrate this, this section will briefly outline two examples of major global environmental contamination and pollution problems which are presently challenging policy and health decision makers.

1.3.1 Electronic Waste

Informal electronic waste (e-waste) recycling has become a global environmental contamination challenge. As mobile devices, televisions, computers and other electronic goods are continually developed and upgraded, there is an increasing volume of waste generated from 'out of date' devices (Chi et al., 2011). These devices are typically shipped to facilities in Asia, Africa and South America where the metal(loid)s are recovered for re-use (Wittsiepe et al., 2016). The processes used in this recovery involve heat and acid treatment, with the low value plastics often burned to allow access to the metal(loid)s (Wittsiepe et al., 2016). These practices are commonly carried out by children, adolescents and women who work in artisanal processing facilities with very limited personal protective equipment (Schmidt, 2006). E-waste recycling has resulted in cities throughout the world bearing a significant environmental contamination burden with many cities becoming polluted by metal(loid)s and organic compounds known to cause significant health impacts ranging from respiratory illness to severe carcinogenic outcomes (Chen et al., 2011, Luo et al., 2016). Of particular concern is the compounding effect that multiple-chemical exposure may have on the developing body (Grant et al., 2013). Management of e-waste is an international challenge and the Basel Convention aims to prevent the use of developing nations as dump sites for hazardous waste, however many developed nations navigate this by shipping old goods in 'working order' to recycling facilities (Schmidt, 2006, Sthiannopkao and Wong, 2013). E-waste remains one of the greatest environmental challenges resulting in diminished child and adolescent health outcomes (Zeng et al., 2016).

1.3.2 Artisanal Mining and Smelting

Artisanal mining and smelting is the process of extracting and purifying ore by amateur miners, on a small scale and often in the home living environment. Artisanal smelting occurs throughout the world, predominantly in the developing world (Bose-O'Reilly et al., 2010, Cheyns et al., 2014, Ping et al., 2008, Steckling et al., 2011). One of the most recently significant artisanal smelting incidents was in Zamfara State in Nigeria where processing for gold resulted in the widespread distribution of lead dust (a co-associated metal in the host ore) (Plumlee et al.,

2013). Processing of ore in home-made mills, with limited protective equipment or dust mitigation measures generated soil lead concentrations up to 185,000 mg/kg (Plumlee et al., 2013). Subsequently, a health emergency ensued from the exposure of the community to the dust, with an estimated 400 children dying from lead poisoning and many thousands having severe permanent health impacts (Dooyema et al., 2012, Lo et al., 2012, Plumlee et al., 2013). The remediation and soil recovery efforts in Zamfara State where soils were stripped out and replaced with clean-fill material at a substantial cost provided the opportunity for international collaboration on development of best-practice environmental contamination mitigation strategies (Tirima et al., 2016).

1.4 Australian Perspectives on Environmental Contaminants

The following chapters of this thesis identify and examine in-depth a number of Australian examples where environmental contamination poses environmental management conundrums. It is important to note at this point that management of environmental contaminants often poses a challenge for regulators as there is a balance between social, political, environmental and ethical factors that must be maintained (Forum, 2009, Kuppusamy et al., 2016, Rizzo et al., 2015). Subsequently, management approaches may not always be satisfying to all stakeholder groups and it is often perceived that regulators are not doing enough to effectively manage environmental contamination events (Döberl et al., 2013).

In order to determine if a site is contaminated in Australia, environmental media (e.g. soil or water) are compared to a range of environmental and human health investigation threshold guidelines (Table 1.1). Exceedance of the concentrations specific to the sampling environment will result in the site being deemed contaminated. One of the shortfalls of reliance on guideline concentrations for determining a site's contamination status is that globally, there is great variation in investigation guideline concentrations (Table 1.1) (Jennings, 2013). This leads to questions about which is the most correct guideline to follow. Over history, guidelines have been routinely reduced to reflect new understandings in health-exposure and tolerance thresholds. More recently, the international literature demonstrates that health tolerance thresholds for a range of environmental contaminants, particularly for young children is extremely low (Joe et al., 2006, Lanphear et al., 2005, Rauh and Margolis, 2016). Therefore, dependence on the lowest conservative investigation thresholds is most appropriate.

At various levels of government, contaminated sites are required to be notified and managed. In addition, and to complicate matters, each Australian jurisdiction also has its own set of

regulatory requirements that must be met. In New South Wales (NSW) alone, there was 1,617 known contaminated sites reported to the NSW EPA at the 21 June 2016 (Figure 1.2).

Table 1.1 Comparison of a range of global drinking water and soil health investigation guideline values for residential environments.

Jurisdiction	Metal(loid)	Drinking Water ($\mu\text{g/L}$)	Soil (mg/kg)
Australia	As	10	100
	Cu	2,000	6,000
	Pb	10	300
United States of America	As	10	0.61
	Cu	1,300	310
	Pb	15	400
Canada	As	10	12
	Cu	1,000*	63
	Pb	10	140
Hong Kong	As	10**	22.1
	Cu	2,000**	2,950
	Pb	10**	258

*Aesthetic investigation guideline

**Based on WHO (2011) Guideline

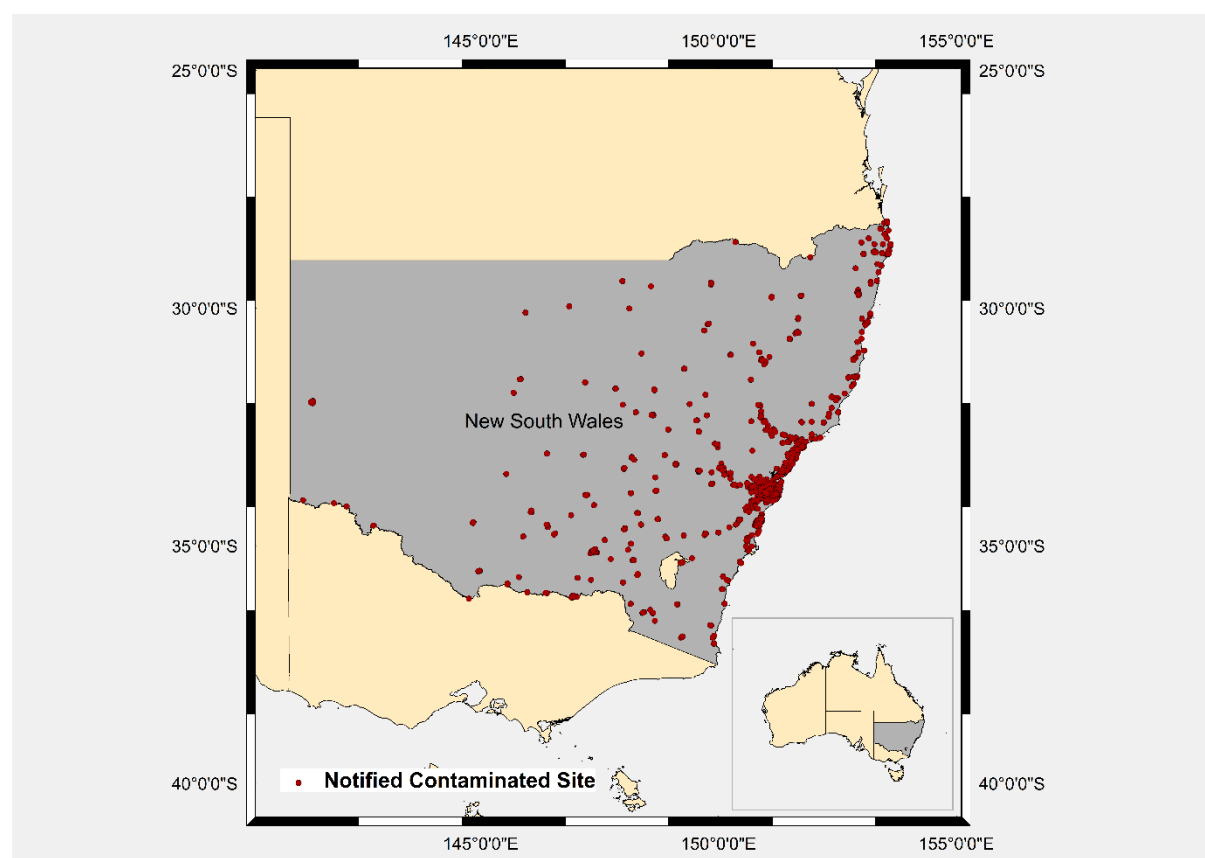


Figure 1.2 Contaminated sites notified to the New South Wales Environmental Protection Authority as at 21 June 2016.

To consider the challenges of environmental contamination in more detail, we return to one of the longest studied and most ubiquitous environmental contaminants, lead. The large body of work on this contaminant has provided many opportunities to study its impacts. The lead mining town of Broken Hill in NSW, Australia exemplifies the multiple-stakeholder interest in environmental contamination and management. Broken Hill has a very long history of environmental lead contamination contributing to the burden of disease in that town (Boreland et al., 2002, Dong et al., 2015, Gulson et al., 1994a, Gulson et al., 1994b, Taylor et al., 2014a). As early as 1893, only 8 years after the commencement of large scale mining operations at the Line of Lode ore body which was surrounded by the urban township, there was speculation that the mines were contributing to the environmental contamination problems (NSW Legislative Assembly, 1893). Almost 100 years later, in 1991, the issue of lead dust deposition over the urban area surrounding the mine was again discussed, suggesting that ‘fugitive losses’ from the mine were a significant contributor to environmental lead contamination (van Alphen, 1991).

After more than 130 years of mining operation there still persists a significant public health burden associated with environmental lead contamination in Broken Hill. More than 50 of the children under 5 years of age have blood lead levels $> 5 \mu\text{g/dL}$, the national level of concern (Taylor et al., 2014b). While on its face, this would appear to be a marked environmental and health management failure, there has been vigorous debate about the relative impacts of mining on the environment and human health (Dong et al., 2015, Earl et al., 2016, Kristensen and Taylor, 2016, Taylor et al., 2014a). Recent evidence-based research has resulted in considerable progress in understanding the risks, educating the Broken Hill community (Figure 1.3) and reducing the burden of disease (Dong et al., 2015, Taylor et al., 2014a). In early 2015, the NSW government provided AUD\$13 million over five years to reduce environmental lead exposure in Broken Hill via a range of environmental, community health and education programs (Kristensen and Taylor, 2016, NSW EPA, 2015).



Figure 1.3 An environmental lead dust management strategy in Broken Hill, NSW, aimed at encouraging children to wash their hands following playing on playground equipment that gathers atmospherically derived dust.

One of the key themes in this thesis is to examine the management and regulator responses to known environmental contamination sources. These range from the deliberate obfuscation of the source of lead in drinking water infrastructure in Tasmania, to the identification of lead additives in plumbing products in the home and even more broadly on the city-scale management of atmospheric and slag bound metal(loid)s in the urban environments surrounding former ferrous and non-ferrous metal smelters.

1.5 Aims and Structure of this Thesis

The aim of this work is to demonstrate that there persists a range of significant environmental contamination problems in Australia, that have the potential to pose substantial impacts to human health, and which are largely unknown to the general public. This thesis is structured to consider contaminants at multiple scales and studies have been undertaken which examine contaminants at the home scale, the home being the place where the majority of living and ultimately exposure time is spent for young children. Studies have also been included at the agricultural scale as these contamination scenarios are often overlooked and are poorly

regulated. Finally, studies are also conducted on the city-wide scale. Cities are examined for the exposure risk they represent for their inhabitants.

By engaging in this research one of the key questions surrounds the response of regulators in managing the environmental contamination events once they have been identified. Considerations of planned management strategies, contemporary best-practice strategies and global management failures for these environmental contamination scenarios are offered to provide a solution to the problems identified and to remedy the site specific contamination scenarios.

The following case studies are included in this thesis, each of which captures a niche, previously unstudied contamination scenario, with further details provided in their relevant chapters:

1.5.1 Contaminants in the Home Environment

P.J. Harvey, H.K. Handley, M.P. Taylor. Identification of the Sources of Metal (lead) Contamination in Drinking Waters in North-eastern Tasmania using Lead Isotopic Compositions. Environmental Science and Pollution Research 2015, 22(16), 12276-12288.

This study investigates a long term and pervasive drinking water metal contamination problem in the complex Ringarooma River catchment using a multiple lines of evidence approach for delineating the source(s) of contamination. A range of environmental tracing techniques are applied to differentiate the multitude of potential contributing source(s) including agricultural runoff, degraded infrastructure and household plumbing. Crucial to this study was the need to provide unequivocal evidence of the source(s) of lead contamination to debunk any suggestion by the water utility and environmental regulator that the contamination was naturally occurring.

P.J. Harvey, H.K. Handley, M.P. Taylor. Widespread Copper and Lead Contamination of Household Drinking Water, New South Wales, Australia. Environmental Research 2016 151, 275-285.

This study expanded on the problem of drinking water contamination in the Ringarooma River catchment by asking the question: how widespread is the problem in Australia? Through extensive community engagement and outreach, a sufficient sample size was obtained to comment on that problem. We present the case in this study that there is a failure in health management as products for use in potable water are still manufactured, almost 20 years after the international and Australian literature identified lead-bearing products as an exposure risk.

M. Rouillon, P.J. Harvey, L. K. Kristensen, S. G. George, M. P. Taylor. VegeSafe: A Community Science Program Measuring Soil-metal Contamination, Evaluating Risk and Providing Advice for Safe Gardening. Environmental Pollution 2016

The VegeSafe program takes a step away from drinking water contaminants however it is no less important as an assessment of health exposure risk in the household setting. VegeSafe considers metal(loid)s in household soils across Australia. This study highlights that the home environment has a number of ‘hidden dangers’ that a homeowner should be alert to, but not alarmed about. This study also marks the transition from contaminants in the home to contaminants in agriculture as many of the community participants in the VegeSafe program were from community gardens and small-hold farms.

1.5.2 Contaminants in Agriculture

P.J. Harvey, M.P. Taylor, H.K. Handley. Widespread Environmental Contamination Hazards in Agricultural Soils in Above Ground Large-scale Water Supply Pipelines. Water, Air & Soil Pollution 2015, 226 (6), 1-9.

This study is the first of those considering contaminants in the agricultural environment. Notably, this study considers one of the problems demonstrated in the first study of this thesis: degrading end-of-life drinking water infrastructure. Through examining soils surrounding multiple large lead-jointed pipelines in NSW and WA, and, following a strategic sampling regime, it was clear that the soils were the only plausible source of soil lead surrounding these pipelines. Elevated blood lead concentrations detected in cattle grazing alongside the pipeline corridor in the Hunter Valley could therefore be attributed to the pipeline itself. Once again, this study demonstrates a failure of environmental managers and regulators as the issues of lead contamination of soils along the Hunter Valley pipeline corridor had been known for many years. Additionally, the assessment of other pipeline soils in Australia demonstrates that this is not an isolated problem, however this remains an ongoing environmental contamination issue as no work has been conducted to reduce the exposure risk associated with these other pipeline corridors.

P.J. Harvey, M.P. Taylor, H.K. Handley, S. Foster, M.R. Gillings, A.J. Asher. Tracking Chicken Slaughterhouse Waste in a Peri-urban Waterway Using Chemical, Biological, and DNA Markers. Environmental Research. In Review.

As one of the more unique studies in this thesis, this investigation draws on a number of the themes already presented. Water contamination is examined in the light of effluent discharge

from a chicken slaughterhouse facility. The slaughterhouse resides in the headwaters of a catchment flowing into an ecologically sensitive national park used for recreational swimming and fishing. The source of effluent water was traced to the facility using a range of techniques including basic water chemistry, analysis of arsenic and organo-arsenic compounds via speciation and the detection of DNA and genetic markers in the water. This study demonstrates another ongoing and pervasive contamination incident, overlooked by the environmental regulator. The study also adds to multiple lines of evidence assessment toolkit.

1.5.3 Contaminants at the City-Scale

P.J. Harvey, M.P. Taylor, L.J. Kristensen, S. Grant-Vest, M. Rouillon, L. Wu, H.K. Handley. Evaluation and Assessment of the Efficacy of an Abatement Strategy in a Former Lead Smelter Community, Boolaroo, Australia. Environmental Geochemistry and Health 2016, 38(4), 941-954.

Contaminants in both the home and the agricultural scale present considerable management challenges. This study examines soil environmental contamination across a range of home environments, or, at the city scale. This study demonstrates the failure of yet another management strategy, the Lead Abatement Strategy (LAS), and the health exposure risk associated with that failure. This paper revisits the multiple lines of evidence approach for determining source(s) of pollution and demonstrates beyond doubt that the source of soil contamination is a result of the failed LAS initiated following the closure of the former lead smelter.

P.J. Harvey, H.K. Handley, M.P. Taylor. Determining the Source and Health Risk Arising from Legacy Environmental Contamination Surrounding a Former Antimony Processing Plant. Submission Ready.

The small regional city of Urunga was once the location of an antimony processing plant. The processing plant, which was rudimentary in design, leading to the generation of significant dust from the crushing plant, was situated on the boundary of the urban/bushland area. This study set out to determine if there was an exposure risk associated with one of the host ore (stibnite) co-associated elements, arsenic. By applying a spatially resolved sampling strategy combined with determination of arsenic species in the soil, it is apparent that the forms of arsenic (As^{V}) expected to be associated with the processing plant, were not substantially present in the soils of the urban area. This study is a nice contrast to the other studies in this thesis because it details a site that does not present a health exposure risk. This is important from an environmental management perspective because it provides environmental managers with an opportunity to

understand why no environmental contamination was detected in the urban environment and this is discussed in the study.

P.J. Harvey, M. Rouillon, C. Dong, V. Ettler, H.K. Handley, M.P. Taylor, E. Tyson, P. Tennant, V. Telfer, and R. Trinh, Geochemical Sources, Forms and Phases of Soil Contamination in an Industrial City. Science of the Total Environment. In Press.

Newcastle is a regional city in NSW that has a rich industrial heritage. For many years, it has been the home of very large scale non-ferrous and ferrous metal smelters. This study was designed to draw in a range of analytical and investigatory approaches including the VegeSafe program used previously in this thesis (study 3) to demonstrate how an environmental assessment can be carried out on a large scale. This study documents for the first time in the international literature, the extensive problem of environmental contamination in this city and offers suggestions for ways to mitigate this contamination. This study draws the thesis to a close by demonstrating the multiple scales (home, agriculture and city) of environmental contamination and the significance that each of these plays in health-risk exposure.

1.6 Methods and Approach

This thesis adopts a range of methods to develop an understanding of the understudied pathways of environmental contamination in Australia. This thesis does not specifically focus on one particular contaminant, but rather encompasses contaminants ranging from metal(loid)s in soil and water to biological contaminants in effluent water.

This body of work focuses on applying a range of geochemical and environmental assessment techniques to identifying environmental contamination events in Australia. Part of this work considers the community-academic nexus and so projects were designed to incorporate, as far as possible, an engagement with the community in one form or another. Critical to many of the projects that are incorporated into this thesis was the need to report findings back to members of the community. Examples of how this is achieved through community meetings, media output and facilitated discussions is presented with the relevant publications.

One of the best approaches to changing the way environmental contaminants are managed is to engage and inform the community. Citizen science involves public engagement with science and is a strong tool that can help to raise public awareness of environmental contamination problems and subsequently trigger a management response. The Office of the Chief Scientist has identified citizen science as an important part of research and learning in Australia. The

Office identifies a number of perceived benefits from community interaction with scientists and scientific projects across three main platforms (Office of the Chief Scientist 2015):

- *“For the research community: increased scale of data collection, new or greater access to resources, access to private lands and information;*
- *For citizen scientists: education (either formal or informal) leading to new knowledge and skills, empowerment, friendships and more active lifestyles;*
- *For society: new information for government decision-making, greater interest in science and understanding of scientific principles, greater environmental stewardship and more engaging teaching methods for science and mathematics subjects, whilst helping scientists to understand people’s concerns and aspirations.”*

This thesis utilises citizen science to engage the community of Australia across a number of projects. The aim of this is to enhance the community understanding of the potential contaminant exposure pathways in the home and local community.

In order to identify contamination sources and to understand the impact and potential risk of harm of environmental contaminants, multiple analytical techniques and approaches have been applied in this thesis. In this sense, the thesis applies a mixed methods approach to delineating sources, types and concentrations of a range of organic and inorganic contaminants. Throughout this thesis, collaborations have been developed with experts in the fields of research relevant to each case study. In the introductory text for each publication, the percentage contribution of each author is presented next to their name.

Analytical procedures used in this thesis include, among others, solution inductively coupled plasma mass spectrometry (ICP-MS) and in-situ and lab based X-ray fluorescence spectrometry to establish total element concentrations, lead isotopic ratio quantification using single collector ICP-MS to establish potential contaminant sources, high performance liquid chromatography and ICP-MS (HPLC-ICP-MS) to determine sources and the toxicity of arsenic in the both the soil and water environment, gas chromatography and ICP-MS to determine the presence of polycyclic aromatic hydrocarbons in the soil environment, other wet chemistry techniques such as in-vitro bio-accessibility extractions to establish the health risk associated with lead and arsenic in the soil environment and a range of microscopy techniques including scanning electron microscopy to establish sources of soil bound contaminants.

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2 **Contaminants in the Home Environment**

This first chapter of case studies considers contamination sources in the home environment. This section is weighted towards drinking water contaminants as to date, little research has been conducted in Australia on this potential exposure pathway. In addition, a broad assessment of home soil contamination has been conducted through the VegeSafe program. This chapter introduces the idea of a multiple lines of evidence approach to identifying the sources of environmental contaminants and also the engagement of citizen scientists in academic research.

Identification of the Sources of Metal (lead) Contamination in Drinking Waters in North-eastern Tasmania using Lead Isotopic Compositions.

P.J. Harvey (90%), H.K. Handley (5%), M.P. Taylor (5%)

Environmental Science and Pollution Research 2015, 22(16), 12276-12288.

This project was conceived by Harvey after hearing a radio broadcast about a community in north-eastern Tasmania experiencing lead in their drinking water supply. The source of the drinking water lead contamination at the site was unknown but it was suggested that it may be naturally occurring, or resulting from the historic tin mines in the area. It became clear from initial research that this was unlikely to be the case and that an anthropogenic source was likely to blame. Two sampling trips were conducted, the first to gather information about the natural environment (geology, soils and fluvial sediments) as well as some household water testing. The second trip was far more spatially constrained in an attempt to pinpoint the exact location of the lead in the supply network, once the natural environment had been discounted. As this study details, the main contributing factors were lead-weld pipelines, PVC piping and the household plumbing. Following the publication of this work the water supply company tried to discredit the findings. The response of TasWater to our research is included in Supplementary Data 7.1.2 for reference, and our reply to that commentary is included here. This project attracted significant public attention, with engagement with community members occurring throughout the project in the form of community meetings and media campaigns. This project was an important catalyst for the subsequent installation of a pipeline to many parts of the Ringarooma River catchment for safe, filtered drinking water. It also drew attention to lead contamination of drinking water in another town within the catchment and the trial of a portable filtration plant. Furthermore, it resulted in the installation of rainwater tanks and contractual continual supply from the water utility to Pioneer and nearby towns and the shift in water quality management by the government such that water quality data will be updated and published live online for all community members to view.

Pages 23-35 of this thesis have been removed as they contain published material under copyright. Removed contents published as:

Harvey, P.J., Handley, H.K. & Taylor, M.P. (2015) Identification of the sources of metal (lead) contamination in drinking waters in north-eastern Tasmania using lead isotopic compositions. *Environmental Science and Pollution Research*, 22, 12276–12288. <https://doi.org/10.1007/s11356-015-4349-2>

Reply to “Review of Harvey et al. (2015) Project 1094-15 by TasWater and Water Research Australia.

P.J. Harvey (80%), H.K. Handley (10%), M.P. Taylor (10%)

Reply to “Review of Harvey et al. (2015) Project 1094-15” by TasWater and Water Research Australia

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Summary

This reply responds to and outlines a number of important failures, oversights and shortcomings of the recent TasWater commissioned review of “*Identification of the sources of metal (lead) contamination in drinking waters in north-eastern Tasmania using lead isotopic compositions*” by Harvey, P.J, Handley, H.K. and Taylor, M.P. (2015) published in *Environmental Science and Pollution Research*. The TasWater review was authored by Priestley, T., Gaston, T. and Mosse, P. from Water Research Australia.

Critically, this reply responds to the following aspects raised in the TasWater review:

- 1. Connection and relevance of the Moorina Power Station to the Pioneer Drinking Water Supply**
- 2. Quantity of data**
- 3. Questions and comments relating to site descriptions, sampling and analytical methods**
- 4. Application of lead isotope compositions for determining the source of contamination**
- 5. Professionalism**
- 6. Sediment**
- 7. True colour**

The authors conclude that based upon the evidence presented in this reply that TasWater’s review has not added anything new or material to Harvey et al. (2015).

Reply to Review of Harvey et al. (2015) Project 1094-15

1. Connection and relevance of the Moorina Power Station to the Pioneer Drinking Water Supply

One of the largest points of contention raised in the TasWater review is the connection of the Moorina Power Station to the drinking water supply of Pioneer. As a result of the erroneous conclusions drawn by the review described herein, Harvey et al. dismiss any statements that the Moorina Power Station does not supply *any* water to the Pioneer Dam and subsequent drinking water supply.

Page 7 of the TasWater review outlines the reticulation network and provides a schematic diagram to illustrate the connection (Figure 1 of the review), dated **2010**. The accompanying text says the following (page 7):

*“The diagram shows that at that time water **was** [our emphasis] supplied to the Pioneer Dam from the **Frome Dam** [our emphasis] via an open raceway and a series of 100 mm diameter polyvinyl chloride (PVC) pipes”.*

The text then informs the reader (page 7):

*“Information from TasWater states that, since around **2009** [our emphasis], the water supply to Pioneer is **no longer** [our emphasis] sourced from Frome Dam or the raceway infrastructure immediately below it”.*

Later in the TasWater review a citation is provided for a communication from Wright (2015) that states the following, without providing a specific date for disconnection of the Frome Dam (page 13):

*“The Pioneer Dam is fed by the open Pioneer water race which originally extended from Frome Dam to Pioneer and supplied water to the Moorina Power Station on the way. Water from the Frome Dam has not now supplied Pioneer for **several years** [our emphasis]. Water that is transferred from the race to the Pioneer Dam is water trapped by the open race from surface water runoff and small watercourse intercepted by the race between the site of the old Moorina PS and Pioneer”.*

The statements made by and relied upon within the TasWater review are inherently contradictory and as a result provide no concrete or useful information.

In an earlier report (2013) commissioned by Ben Lomond Water (now TasWater), the following statement was made:

*“The **Pioneer water scheme sources water from the 3600 ML Frome Dam** [our emphasis] owned by Moorina Hydro Pty Ltd and located to the south of the town. The water is transferred via an 8 km water race and 480 m PVC pipe to PE pipe suspension bridge crossing the Ringarooma River” (GHD 2013a).*

To complicate the issue further, the schematic provided as Figure 1 of the review, which has no clear scale, depicts only one disconnection between the Frome Dam and the section labelled Pioneer Race. Assuming that this is correct and referring to the text again, this disconnection only prevents transfer of water from the Frome Dam through the first 900 m of the 8 km water race. This contention is supported by another Ben Lomond Water (now TasWater) commissioned report from 2013 that states (GHD 2013b):

*“Ben Lomond Water holds a licence to extract water from the 3600 ML Frome Dam. However the water race to supply the town from this source is not operational for the first 900 m of its 8 km length due to the recent installation of an irrigation pipe in **2010** [our emphasis]”.*

Applying some deductive reasoning to clarify these important arguments about the source of lead we note the following:

Given that Moorina Power Station is, according to the TasWater review and confirmed on Google Earth, located a distance of approximately 5 km from the town of Pioneer, a disconnection of 900 m of the water race downstream of the Frome Dam results in 2.1 km of race upstream of the Moorina Power Station from which surface water can be derived from the surrounding catchment.

This distance of open-channel network is further confirmed by Mr Mike Cooke (former Water

Warden for Pioneer, former part owner/operator of the Moorina Hydro-Electric Scheme and overseer for the Winnaleah Irrigation Scheme). Mr Cooke reported in a communication to M.P. Taylor (04/05/2015) that the connection between the Moorina Power Station and the Frome Dam was/is as follows:

“No further pipework was incorporated in the channel work from the weir [at Frome dam] below the dam to the head of the penstock supplying the power station, a distance of around 2.7 kms.”

Clarification of this issue is quintessential to the crux of the sources of lead because the TasWater review hinges, in part, on the complete rejection of Harvey et al.'s (2015) data collected from the Moorina Power Station. It is important because the data from the Moorina Power Station delineates a significant source of contamination and one that is likely to have been ongoing for decades, representing chronic exposures via the potable water supply.

The TasWater review comments further (page 10):

“It appears that the paper’s authors did not fully understand or appreciate the exact scope of the current water supply system”.

Careful analysis of the review reverses this argument back to the authors of the TasWater review by virtue of their contradictory statements on page 7, Figure 1 (TasWater review), the additional evidence provided in the GHD reports (GHD 2013a; b) and commentary by Mr Cooke. Thus, on the basis of the information supplied in the TasWater report it is impossible to **not** consider the Moorina Power Station and its infrastructure as a source of contamination as was proposed in Harvey et al. (2015). Consequently, all comments regarding the disconnection of the Moorina Power Station from the Pioneer drinking water supply in the TasWater review should now be disregarded.

2. Quantity of data

Comments are made throughout the TasWater review about the quantity of data collected. Harvey et al. (2015) collected 179 samples, generating 716 data points. Given that the TasWater review collected zero new data, it is difficult to comprehend how the study could be described as being deficient in data.

3. Questions and comments relating to site descriptions, sampling and analytical methods

The quality and reliability of the data was ensured through a rigorous QA/QC program, as detailed in the Supplementary Data section of the paper (Supplementary Data 1 – see attached). The quality assurance program for the water samples was 17 (field blanks and trip blanks)/96 samples in total (18 % of samples). The reviewers would know that this exceeds the minimum 10 % for QA/QC for analytical purposes. Laboratory analysis was carried out using the National Measurement Institute's NATA accredited method, which includes all relevant QA/QC protocols.

The TasWater review makes a number of references to sample locations. It is unclear if the TasWater reviewers have viewed the supplementary material available with the online publication but all details of sample locations are included as GPS co-ordinates, which can be plotted easily using Google Earth. Samples collected from private properties cannot be accurately identified as this poses significant issues with privacy. Homeowners were protected by the addition of a suitable margin of error to the georeference location data.

The reviewers also suggest the methods and protocols used in the study were unclear (page 12):

“A lack of duplicate samples and repeat analyses, combined with an unclear sampling protocol, is a major weakness of the paper.”

However, judicious review of the article's section on Material and Methods, the Supplementary Data and the references provided at the end of the paper makes it abundantly clear that the study used a robust approach that relied on Australian Standards or world's best practice. The TasWater review identifies an omission of the Supplementary Data (temperature). This is available from the authors on request. Concern is raised about the flush periods for samples collected from within the properties. Australian Standard AS/NZS 5667.5:1998 s.4.1.5 Sampling procedure - Consumer's Taps says *“if the effects of materials on water quality are being investigated then the initial draw-off should be sampled”*. The TasWater review raises concern about residents collecting a water sample from their own taps. This procedure has been applied in multiple domestic and international sampling programs and is *de facto* approach (see examples: Fertmann et al. 2004, Meyer et al. 1998, Pieper et al. in press).

Another concern raised was that samples were collected from stagnated water (page 20):

“Much of the data in Harvey et al. (2015) appears to be based on first flush sampling

of water after extended periods of stagnation in the pipes and local plumbing. These samples do not accurately represent the quality of the main body of water within the distribution system”.

Not only were first draw samples collected following overnight stagnation (8 hr equivalent) for investigation of property plumbing, a 30 second and three minute (where possible) draw was also collected to determine the contribution of lead from off the property. The TasWater review’s own calculations about stagnation in the reticulated supply note the following (page 11-12):

“Based on the system drawing provided by TasWater, and only including pipes in the direct line from the primary 100 mm feed main, the calculated volume for the western part of the town (including the 100 mm feed main) is approximately 5.2 kL and for the eastern part of town (including the 100 mm

feed main) is approximately 3.8 kL. Therefore it seems likely that the water in the mains is fairly well turned over on a daily basis”.

Harvey et al. therefore fail to see how the water collection protocol in the private properties is erroneous and that water was collected from anything but normal usage conditions.

Of the 29 sites sampled as filtered and un-filtered on the first field visit, only six of these sites exhibited a difference in lead concentration between the fractions. Based on that dataset, the collection of further filtered and un-filtered samples was deemed to not be a worthwhile avenue of further investigation. Consequently, only un-filtered samples were collected on the second field visit. Regardless, the filtered and un-filtered fractions for the first field visit were reported in the Supplementary Data. In hindsight, the filtered and un-filtered samples may have been useful from the second sample collection period to determine the particulate lead (>0.45 µm) fraction of those samples with detectable lead concentrations. Nonetheless, given the weight of evidence regarding the source of lead contamination in the drinking water supply presented by Harvey et al. (2015) it would have had little impact on the conclusions reached.

4. Application of lead isotope compositions for determining the source of a contamination event

It is evident that the authors of the TasWater review do not fully comprehend the application of lead isotopic compositions for determining the source of lead. This is evidenced by their apparent poor understanding of Soto- Jiménez and Flegal’s (2011) Torreón lead smelter study from Mexico, upon which they rely. The TasWater review stated the following in regard to this study’s contamination source discrimination using lead isotopic compositions (page 8):

“For example, Soto-Jiménez and Flegal’s (2011) found that the lead isotope ratios of dust, soil and aerosols from a smelting area of Mexico were indistinguishable from each other making it impossible to apportion the dominant source of lead.”

The review erroneously goes on to state (page 8):

“Therefore, investigations require that the different sources of lead be isotopically distinct...”

Careful reading of Soto-Jiménez and Flegal’s (2011) study should have resolved that conundrum for the reviewers, because the dominant and prevailing source of environmental lead in Torreón is a single dominant source of ore, producing a uniform environmental isotopic composition largely indistinguishable from the source materials. In the case of Torreón the environmental lead impacting air, soil, dust and blood lead is the ore processed by the local smelter. Indeed in the conclusions of the Soto-Jiménez and Flegal’s (2011) study, the authors state clearly:

“The data show that lead concentrations and stable isotopic composition in environmental samples revealed that pollutant lead is mostly from contemporary emissions from the smelter”

Lead isotopic analysis can be used to supplement environmental investigations in two ways: (1) To discriminate between sources where there are multiple possible contaminant origins; (2) To assess the effect of a single pollutant source on the environment, where analysis of samples can confirm that the source investigated is responsible for the dominant contamination.

We note that M.P. Taylor had suggested to Mike Brewster, the TasWater CEO, that it might be prudent to source an expert in lead isotopic composition analysis, and Dr Ashley Townsend (University of Tasmania) was suggested. Alternatively, Professor Brian Gulson, who is one of Australia’s most published experts in the use and application of lead isotopes for environmental analysis would have been an excellent choice.

The TasWater review attempts to further discount the lead isotope compositions determined

in the Harvey et al. (2015) study by suggesting that repeat sample collection may result in different lead isotopic ratios. Given the consistency of the drinking water lead isotope compositions across different sample sites, including the sample collected 35 km away in Ringarooma, it is clear that repeat sampling would not have provided any further insight, which is that the lead sources are not natural.

Concern was also raised about the reproducibility of the laboratory results. Each sample analysed for lead isotopic analysis is bracketed with a standard reference material and therefore the standard reference material is analysed repeatedly, generating robust indicative standard deviation data output. Further, the isotope compositions of the water presented in Harvey et al. (2015) were significantly different from the local bedrock isotope compositions. This fact is undisputable and allows us to draw the conclusion that the lead contamination is **not** natural.

Application of source apportionment calculations was rejected by the TasWater review. Rather than discount these calculations, our study shows clearly that large quantities of the lead in the drinking water can be apportioned to the same lead used in the large water supply pipelines. This shows clearly that the lead in the drinking water supply is not naturally occurring and has been contributed to the water supply by the infrastructure. Aside from these things, two points remain abundantly clear:

- (1) The occurrence of measurable and significant lead only occurs once the water passes through the infrastructure network. This conclusion is also reflected in additional (unpublished) data gathered by the Tasmanian Department of Health and Human Services;
- (2) Isotopic data from relevant published sources and this study make it clear that the source is not naturally occurring.

5. Professionalism

In section 4.1 it reads (page 13),

“the use of the term Cesspit to describe part of a water supply system lacks a professional and objective approach to the study”.

This is the name given to the inflow area above the Pioneer Dam, which the community uses to identify the site and is meaningful to them.

We were disappointed that after conversations with TasWater about our willingness to ensure the reviewers understood the approach and purpose of the study, these offers were not taken up. If they had been, then some of the aforementioned issues would not have arisen. Misunderstandings around the geo-location of data could have been rectified in one of two ways (1) plotting up of the GPS locations of the data (Supplementary Data 2) using Google Earth; (2) Contacting the authors for clarification.

6. Sediment

The TasWater review dedicates a large proportion of text to the water and sediment in the Pioneer dam. Specifically, attention is drawn to the elevated lead concentrations in the sediment of Table 1 (416 µg/L and < 7 µg/L). A section of text ascribed to TasWater reveals the following in regard to that concentration (page 15):

“Attached are the two analytical reports covering these sampling events. Previously unless otherwise specified as a sediment investigation, the laboratory’s normal process for “sediment” samples with very low solids was that they are tested as water and hence reported with “water” units (AST#59735)”.

and

“Unfortunately this only adds to the problem of interpreting the data. If the samples only had “very low solids” then it is difficult to see how they are in fact sediment samples”.

This is an important point as TasWater have attributed the lead in the water supply to the sediment. The analysis that was performed on these samples (as a water method) would not extract the lead fraction bound to the limited sediment. TasWater have relied upon this invalid data to form the basis of their sediment bound lead argument. Harvey et al. (2015) has shown that there is no naturally occurring high concentrations of lead anywhere in the Ringarooma River Catchment, either in soils, bedrock or water. Moving forward, this argument is redundant as there is no evidence of sediment bound lead in significant concentrations to generate lead in water above the Australian Drinking Water Guideline.

The TasWater review fails to address the remainder of the data presented in Harvey et al. (2015), which highlights elevated lead concentrations detected in tap water samples collected in Gladstone and Ringarooma. Had these samples been considered by the TasWater review it would have been clear that any notion of sediment bound lead, or naturally occurring lead contaminating the water supply is misleading and incorrect.

7. True colour

A section on true colour of the Pioneer Dam is included on page 15 and 16, under section 4.2. Figure 2 (erroneously referred to and labelled in the TasWater review as Figure 1) relied upon by the authors does not have a y-axis or a clear x-axis, rendering the bar charts meaningless. In any case, these new data are irrelevant to the Harvey et al. (2015) study as colour does not explain source apportionment and was not relied upon in the study.

Conclusion

Based upon the evidence presented here, it is clear that TasWater's review has not added anything new or material to the Harvey et al. (2015) study. Indeed, it has merely confused the matter. The review contains several inaccurate and contradictory statements with regard to the Pioneer water scheme; contains poor understanding of the methods and data quality used in the study; and displays a weak grasp of the utility of lead isotopes, which are critical to the source apportionment aspect in the Harvey et al. (2015) study.

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Widespread Copper and Lead Contamination of Household Drinking Water, New South Wales, Australia.

P.J. Harvey (90%), H.K. Handley (5%), M.P. Taylor (5%)

Environmental Research 2016 151, 275-285.

Drinking water is one of those environmental exposure pathways that is often overlooked in countries like Australia where the majority of the community can turn on their taps, get a clear and tasteless glass of water and not have to worry about the potential for arising illnesses. While this is vastly true for organic contaminants such as bacteria, there is a hidden danger in the inorganic contaminants (metals) that can enter the drinking water through the household plumbing or reticulated supply. The work in Pioneer Tasmania (Paper 1), in conjunction with work conducted previously by other authors demonstrates that there is a real and appreciable risk associated with lead and other elements entering the drinking water. In 1993, the National Health and Medical Research Council (NHMRC) flagged lead in drinking water as a major public health concern. Over time, this concern dwindled and the public focus was taken away from this potential source of environmental exposure. This study set out to establish the risk of potential exposure to inorganic contaminants (arsenic, copper, lead and manganese) in Australian drinking water. A community science outreach program was conducted to engage the community of New South Wales, and in turn, provide access to a sample of their first draw tap water. Over 200 samples were collected from private homes and the data from these samples indicates that there is a problem around lead is leaching from many plumbing fittings within the home. This study highlighted the shortcomings in drinking water management in Australia, particularly surrounding manufacturing standards for products in contact with potable water. The study received significant community attention, feeding back into the citizen science aspect of the project, and was covered widely by a range of media including television, radio and print (Supplementary Data 7.2.2). This work was presented in-part at the Goldschmidt 2015 conference in Prague, Czech Republic.



Widespread copper and lead contamination of household drinking water, New South Wales, Australia



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ARTICLE INFO

Article history:

Received 3 May 2016

Received in revised form

27 July 2016

Accepted 29 July 2016

Available online 8 August 2016

Keywords:

Contamination

Brass

Leaching

Exposure

ABSTRACT

This study examines arsenic, copper, lead and manganese drinking water contamination at the domestic consumer's kitchen tap in homes of New South Wales, Australia. Analysis of 212 first draw drinking water samples shows that almost 100% and 56% of samples contain detectable concentrations of copper and lead, respectively. Of these detectable concentrations, copper exceeds Australian Drinking Water Guidelines (ADWG) in 5% of samples and lead in 8%. By contrast, no samples contained arsenic and manganese water concentrations in excess of the ADWG. Analysis of household plumbing fittings (taps and connecting pipework) show that these are a significant source of drinking water lead contamination. Water lead concentrations derived for plumbing components range from 108 µg/L to 1440 µg/L ($n=28$, mean = 328 µg/L, median = 225 µg/L). Analysis of kitchen tap fittings demonstrates these are a primary source of drinking water lead contamination ($n=9$, mean = 63.4 µg/L, median = 59.0 µg/L). The results of this study demonstrate that along with other potential sources of contamination in households, plumbing products that contain detectable lead up to 2.84% are contributing to contamination of household drinking water. Given that both copper and lead are known to cause significant health detriments, products for use in contact with drinking water should be manufactured free from copper and lead.

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1. Introduction

Metal contamination of drinking water and its potential health effects has impacted human populations for centuries (Bellinger, 2016). Perhaps most famously, albeit controversially, ancient Rome's use of lead in water supply infrastructure has been argued to have caused lead poisoning that contributed to the fall of the empire (Delile et al., 2014; Evans, 1997; Scarborough 1984; Waldron, 1973). Better understanding of the health impacts and consequent corrosion control measures followed the identification of elevated lead and copper concentrations in drinking water resulting from the use of lead service lines in Boston, United States of America (USA), Washington DC USA and also Edinburgh and Glasgow, Scotland (Addis and Moore, 1974; Edwards et al., 2009; Karalekas et al., 1983; Macintyre et al., 1998; Troesken 2008). Further to these cases of drinking water copper and lead contamination, there has been a multitude of similar examples throughout the world (e.g. Fertmann et al., 2004; Gulson et al., 2014; Renner, 2010; ul-Haq et al., 2009). More recently, in the city

of Flint, in Michigan USA, a public health catastrophe has unfolded after 100,000 residents received drinking water via the reticulated town supply contaminated with lead (example of water testing: $n=271$ samples, mean = 10.0 µg/L, max = 1050 µg/L, Flint Water Study, 2015) due to the absence of corrosion control measures (US EPA 2016).

Global research of contaminated drinking water supplies has revealed that some contaminants derived from the local environment, such as arsenic and manganese from bedrock, are widespread and pose a persistent problem (Das et al., 1995; Khan et al., 2011, 2012; Lu et al., 2014; Oulhote et al., 2014). The World Health Organisation described the contamination of Bangladesh ground water supplies by the regional bedrock as a 'public health emergency' (Smith et al., 2000, p. 1093). Its discovery prompted more than a decade of public health research attempting to remedy contamination and prevent further arsenic poisoning (Gardner et al., 2011; Kippler et al., 2016; Rahman et al., 2013; Wasserman et al., 2004).

Previous studies over the last two decades have also revealed potable water contamination in Australia, mainly from lead used in the roof catchments of tank supplies and corrosion of plumbing fittings in small study cohorts (Alam et al., 2008; Cohen, 22nd June, 2000; Gulson et al., 1994, 1997a, 1997b; Harvey et al., 2015;

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Huston et al., 2009, 2012; Magyar et al., 2014; NSW DET, 2011; Rajaratnam et al., 2002; Rodrigo et al., 2012; van der Sterren et al., 2013; Yau and Thorne, 2012). However, no single study has previously conducted large-scale regional sampling to determine the extent of water contamination in the household at the kitchen tap. A comprehensive study examining source bulk water supplies from New South Wales has shown that 98% (copper) and 91% (lead) of systems providing drinking water to consumers on a reticulated scheme are free of those contaminants (Li et al., 2009). Nevertheless, source water analysis may be inadequate for the delineation of population exposures because many contaminants, including copper and lead, commonly arise from corrosion of household plumbing infrastructure within the property boundary (Clark et al., 2015; Gulson et al., 1994; Harvey et al., 2015; Pieper et al., 2015).

In regard to reducing lead exposure, the National Health and Medical Research Council (NHMRC) (1993), the peak medical research body in Australia, recommended the following:

'The use of products containing lead could be prohibited from use in drinking water or plumbing systems. Even if high lead levels in drinking water are found to be relatively uncommon in Australia, elimination of unnecessary sources of lead would be of benefit in reducing the exposure of children to lead' (Greene et al., 1993) (p. 116).

The NHMRC (1993) made the additional recommendation that Australia's governments and policy makers:

'Implement [a] pilot program for testing lead in first flush drinking water. Conduct [an] education program to inform home handypersons of the dangers in using lead solder in plumbing. Review drinking water guidelines for lead in context of an overall lead reduction strategy' (Greene et al., 1993) (p. 182).

Internationally, regulations pertaining to the use of lead in plumbing products in contact with potable water have undergone reform in recent years. In 2014, USA federal legislation commenced that was designed to reduce substantially the permissible lead content of plumbing fixtures and fittings in contact with potable water. The new legislation prescribed a maximum limit of 0.25% weighted average lead content on newly manufactured or installed products, substantially reducing the previously permissible content of lead of 8% (NSF, 2016).

Despite the two-decade-old warning from the Australian NHMRC, focused case studies, international research and amendments to international legislation demonstrating that there is a persistent and widespread issue associated with drinking water and metal contamination, there has been no study comprehensively examining these risks at the population scale in Australian home environments. Thus, this study addresses this knowledge gap by examining household exposure risk to arsenic, copper, lead and manganese contamination via drinking water. In addition, the study also investigates and evaluates sources of copper and lead contamination from plumbing and drinking water infrastructure within properties in households across New South Wales, Australia.

2. Methods

A citizen science approach was adopted in this study whereby members of the community, following media awareness campaigns, submitted household drinking water samples for analysis. The project adopted three phases of investigation, which are summarized in Fig. 1. For Phase 1, first draw samples ($n=212$)

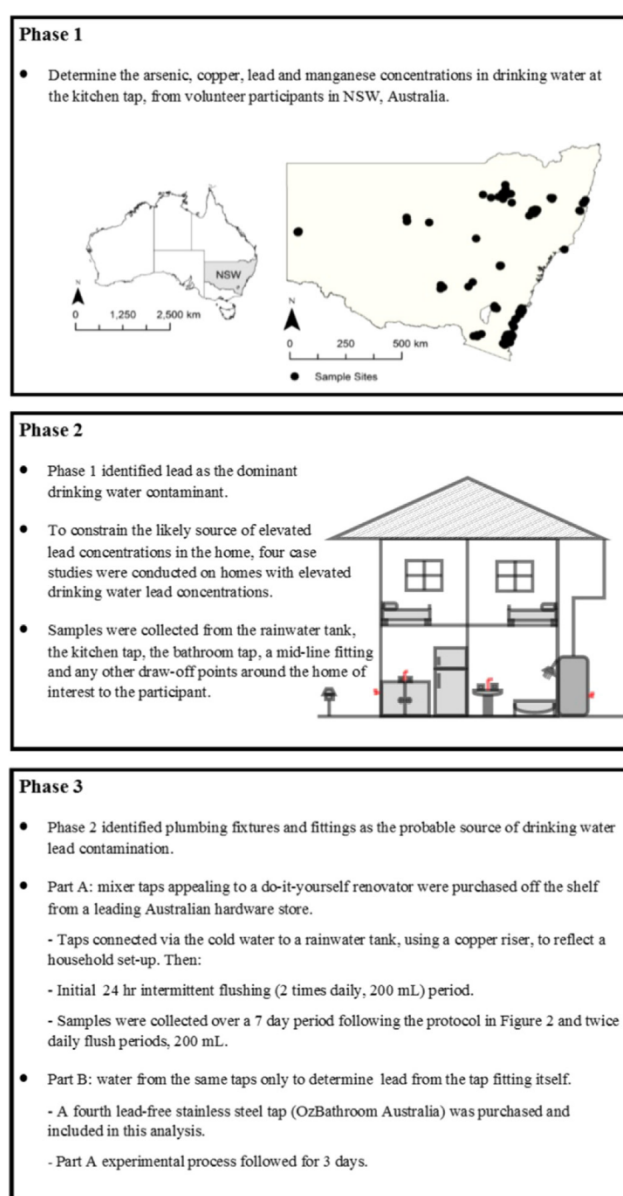


Fig. 1. Structure of the investigation, dividing the project into three key phases of interest.

were collected from each participant's kitchen tap after a 9 h stagnation period (Fig. 2). Sampling instructions, summarized in Fig. 2, were provided to participants for Phase 1. Participants were instructed to collect water samples from a tap that did not have an in-line filtration device. The sampling method was based on the Australian Standard (AS/NZS, 5667.5:1998). Each participant was provided with an acid washed (reagent grade concentrate MERCK HNO_3 diluted to 10% with Milli-Q water) 1 L Nalgene HDPE bottle, and instructed to sample the first draw water from their tap (e.g. to simulate filling a kettle first thing in a morning). Phase 2 (Fig. 1) used 150 mL bottles to capture the water present within the fittings themselves. More detailed site-specific sampling instructions, including the primary sample instructions as per Fig. 2, were provided to each household depending on their plumbing setup. Phase 3 also used 150 mL bottles and applied a sampling regime designed to mimic the typical domestic potable water use accessed via standard plumbing fixtures and fittings that meet

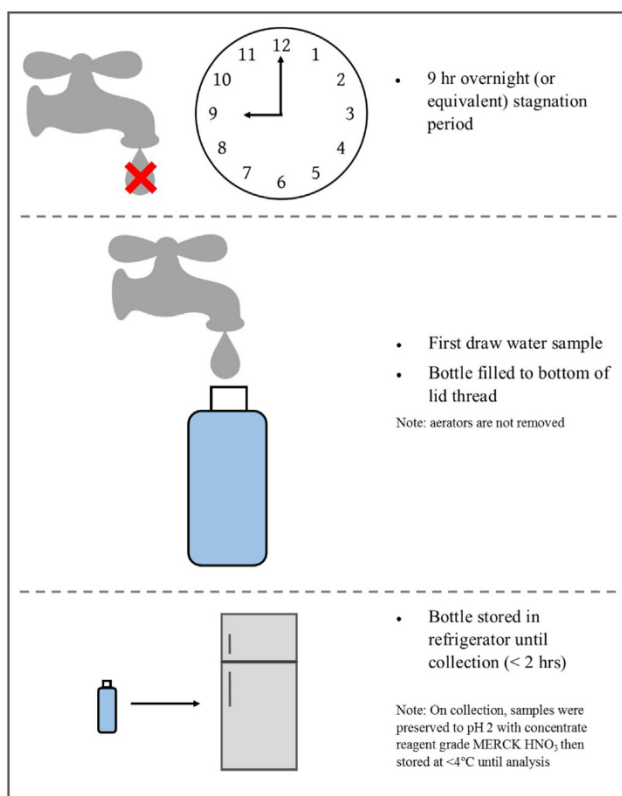


Fig. 2. Sample collection protocol.

Australian Standards for products for use in contact with drinking water (AS/NZS, 4020:2005).

Ten additional samples were taken as part of the Phase 1 sampling process (Fig. 1) that involved flushing stagnant water from the internal plumbing for two minutes prior to collection, reflecting the practice recommended by NSW Health (NSW HEALTH, 2014). Samples were collected at random from any participant who could sample consecutively over a 2-day period. In addition, for quality control purposes, additional samples were collected from Phase 1 sampling (Fig. 1) to determine fluctuations in element concentration due to sampling method or environmental changes at an individual site over time (i.e. a replicate sample separated by a minimum of 24 h) ($n=6$). Sample blanks were also collected ($n=25$).

Samples from Phase 1 were analysed for four elements (arsenic, copper, lead and manganese) and compared to the Australian Drinking Water Guidelines (ADWG, arsenic – 10 $\mu\text{g/L}$, copper – 2000 $\mu\text{g/L}$, lead – 10 $\mu\text{g/L}$, manganese – 500 $\mu\text{g/L}$) (ADWG, NHMRC, 2011a). Samples were analysed by direct injection into an Agilent 7900 octopole Inductively Coupled Plasma Mass Spectrometer (ICP-MS) at the National Measurement Institute (NMI), North Ryde, Australia. A total of 253 samples were collected, generating 1012 data points from Phase 1.

Low copper concentrations identified in Phase 1 resulted in examination of lead only in Phase 2 of the sampling program (Fig. 1) of targeted case study properties. Sampling in Phase 3 Part A (Fig. 1), which evaluated the leaching of copper and lead from taps and fittings, returned low concentrations comparative to the ADWG for copper. Subsequently, only lead was examined in the Phase 3 Part B (Fig. 1) tap leaching experiment.

An aliquot of 30 mL of each sample was digested with 2 mL concentrate reagent grade MERCK HNO₃ for 1.5 h at 95–100 °C (NMI Internal Method NT2_47). An 8 mL aliquot was spiked with

400 μL rhodium/iridium internal standard and diluted with 2% HNO₃ (reagent grade, MERCK) to a 10 mL final volume. An NMI in-house standard reference material (Cal-24, 1 ppm 24 element mixed standard) was added to blank and matrix spike samples (200 μL). Concentrations below the instrument limit of reporting (LOR, 1 $\mu\text{g/L}$) were treated as 0.5 $\mu\text{g/L}$ for analysis. Laboratory duplicate sample relative percent difference for all elements was < 1%. Laboratory control spikes and matrix spikes recoveries were 95–102% and 91–101%, respectively, for all elements. All laboratory blanks returned concentrations < 1 $\mu\text{g/L}$. Sample bottle blanks contained < 1 $\mu\text{g/L}$ except a small number ($n=10$) that contained < 2 $\mu\text{g/L}$ copper.

Participants were provided with a results report (Supplementary Data 1) and advice on approaches to manage any exceedance of the ADWG. Additional water chemistry analysis (i.e. pH, hardness, dissolved oxygen) was not undertaken in this study as gathering this data reliably, in situ, when participants were collecting samples was not plausible.

2.1. Field portable X-ray fluorescence analysis of taps and plumbing components

A field portable X-ray fluorescence spectrometer (pXRF, Olympus Delta Innovex 50 kV) operating in the Alloy Plus mode (a calibration designed specifically for analysis of metal and alloys) was used to assess taps, fittings and components prior to the laboratory water leaching trial (Phase 3). Correct operation of the Alloy Plus mode was determined by analysis of a Brammer Standards CDA 360 Yellow Brass reference material. Taps and fittings were dissected using a grinder fitted with a hardened metal working disk. Once dissected, surfaces were washed with Milli-Q water and metal components in contact with drinking water were analysed.

2.2. Determination of potential exposure risk to drinking water lead contamination

Barton et al. (2002) developed a model to assess the potential health impacts of exposure through ingestion of drinking water contaminants. This modelling approach was applied to the average and maximum copper (average – 364 $\mu\text{g/L}$, max – 3800 $\mu\text{g/L}$) and lead (average – 3.9 $\mu\text{g/L}$, max – 89 $\mu\text{g/L}$) concentrations for first draw samples in this study. The model was applied to a 2 year old, 12 kg infant who consumes 1.5 L of water a day.

3. Results

3.1. Phase 1 first draw samples

First draw samples ($n=212$) contained predominantly low concentrations of arsenic and manganese (arsenic: mean – < 1 $\mu\text{g/L}$, median – < 1 $\mu\text{g/L}$; manganese: mean – 11.0 $\mu\text{g/L}$, median – 3.9 $\mu\text{g/L}$; Supplementary Data 2). Except for the presence of arsenic in a small number of samples ($n=15$) collected mainly from the Far Western region of NSW (Fig. 3, Table 1), there was no significant health concern with respect to this element. For this reason, these elements are not examined here in the same detail as the more abundant water contaminants of copper and lead. Copper was detected in almost 100% of samples ($n=211$) with varying concentrations (mean – 364 $\mu\text{g/L}$, median – 123 $\mu\text{g/L}$). Lead was detected in 56% ($n=118$) of samples (mean – 3.7 $\mu\text{g/L}$, median – 1.3 $\mu\text{g/L}$) (Table 2). Copper exceeded the ADWG in 5% ($n=10$) of the samples and lead in 8% ($n=17$) of the samples (Table 2). The sample replicates collected at least 24 h apart showed good reproducibility between sampling events (Supplementary Data 3).

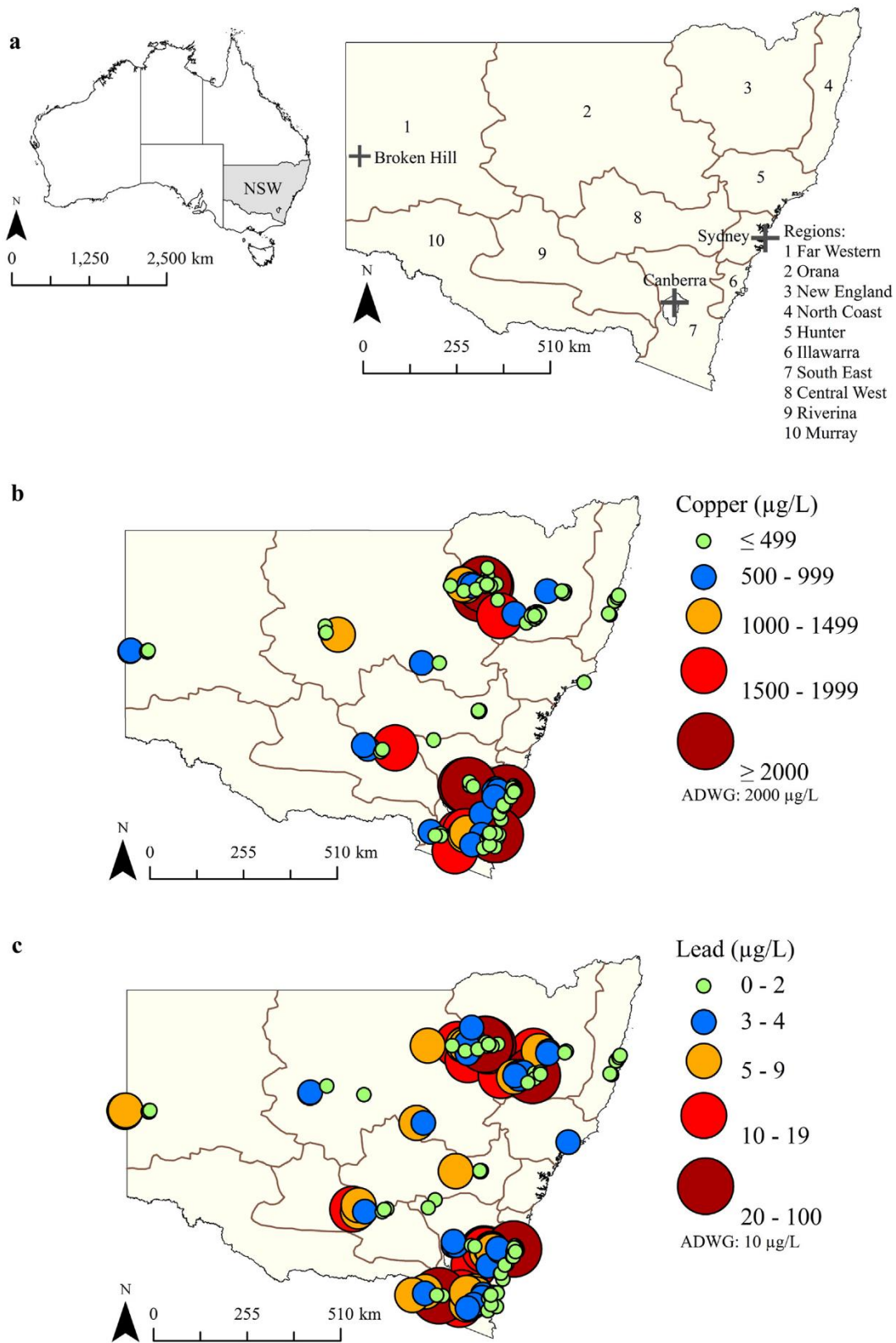


Fig. 3. Map of Australia and New South Wales (NSW) depicting the regions of NSW (a). Copper (b) and lead (c) concentrations in first draw drinking water samples from across NSW, Australia.

Table 1
First draw water concentrations divided by NSW regions.

Region	Element	Mean (µg/L)	Median (µg/L)	Max (µg/L)	Min (µg/L)
Far Western n=18	Arsenic	1	1.1	1.3	< 1
	Copper	146	38.0	1200	2.3
	Lead	1.4	< 1	5.5	< 1
	Manganese	4.7	1.6	23.0	< 1
Orana n=38	Arsenic	< 1	< 1	5.8	< 1
	Copper	617	155	3800	< 1
	Lead	8.2	2.2	89.0	< 1
	Manganese	14.0	8.1	82.0	< 1
New England n=26	Arsenic	< 1	< 1	1.5	< 1
	Copper	316	163	2330	1.5
	Lead	4.4	2.5	28.0	< 1
	Manganese	9.6	2.8	50.0	< 1
North Coast n=10	Arsenic	< 1	< 1	< 1	< 1
	Copper	90.0	64.0	188	15.0
	Lead	0.6	0.5	1.6	< 1
	Manganese	1.0	< 1	2.5	< 1
Hunter n=1	Arsenic	***	***	< 1	< 1
	Copper	***	***	200	200
	Lead	***	***	3.4	3.4
	Manganese	***	***	4.9	4.9
Illawarra n=0	Arsenic	***	***	***	***
	Copper	***	***	***	***
	Lead	***	***	***	***
	Manganese	***	***	***	***
South East n=93	Arsenic	< 1	< 1	< 1	< 1
	Copper	376	140	2640	2.4
	Lead	3.2	1.3	21.0	< 1
	Manganese	9.0	2.9	140	< 1
Central West n=15	Arsenic	< 1	< 1	< 1	< 1
	Copper	246	180	844	6.6
	Lead	2.0	1.1	6.3	< 1
	Manganese	28.0	16.0	80.0	3.1
Riverina n=11	Arsenic	< 1	< 1	1.1	< 1
	Copper	278	29.0	1510	5.4
	Lead	2.8	1.4	10.0	< 1
	Manganese	11.0	8.7	28.0	< 1
Murray n=0	Arsenic	***	***	***	***
	Copper	***	***	***	***
	Lead	***	***	***	***
	Manganese	***	***	***	***
Total n=212	Arsenic	< 1	< 1	5.8	< 1
	Copper	364	123	3800	< 1
	Lead	3.9	1.3	89.0	< 1
	Manganese	10.7	3.9	140	< 1

*** Cannot be calculated.

Table 2
Copper and lead concentrations divided by percentage of ADWG. Code n^t=total sample population, n^d= number of detections (> 1 µg/L), n^f=population within the ADWG bracket. Rows in red highlight samples > ADWG.

		Copper	Lead
n ^t		212	212
n ^d		211	119
LOD – 50% ADWG	n ^f	190	78
	% of n ^d	90	37
50–100% ADWG	n ^f	12	23
	% of n ^d	6	11
> ADWG	n ^f	10	17
	% of n ^d	5	8

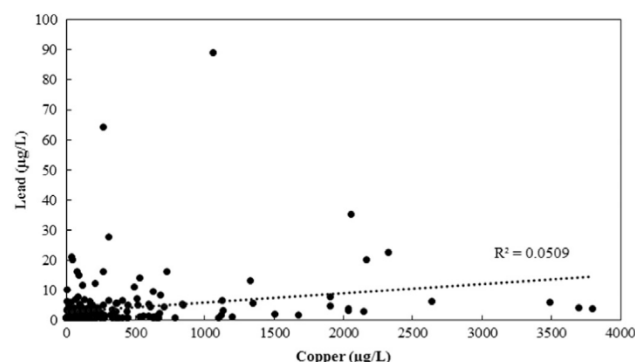


Fig. 4. Copper and lead concentrations in first draw water samples (n=212). Dashed line represents the line of best fit for data correlation.

Elevated copper and lead concentrations were detected throughout NSW (Fig. 3, Table 1). Copper and lead are often co-associated contaminants in drinking water due to their presence in plumbing, fittings and taps (Masters et al., 2016). However, there was no clear correlation between copper and lead concentrations in the first draw water samples (Fig. 4, $R^2=0.05$).

3.2. Flushed samples

Samples collected following a 2-min flush period returned inconsistent contaminant concentrations (Fig. 5, Supplementary Data 4). Arsenic was consistently < 1 µg/L in all samples. Manganese concentrations, although only low, increased in 9 of the 10 samples collected (Fig. 5, Supplementary Data 4). Copper concentrations generally decreased following the 2-min period, however lead concentrations were more variable (e.g. Sample 5 reduced from 10.0 µg/L to 1.1 µg/L whereas Sample 9 increased from 28.0 µg/L to 150 µg/L, Fig. 5, Supplementary Data 4).

3.3. Phase 2 case study properties

Four case study properties (Phase 2, Fig. 1) were examined following identification of elevated lead concentrations. Three of those case study properties had elevated lead concentrations arising only at the draw off point associated with a brass-containing tap (Fig. 6, Supplementary Data 5). Initial assessment of these properties from Phase 1 sampling showed that copper is not an element of concern (mean – 290 µg/L, median – 93.0 µg/L). Copper was therefore not examined further. A comparison of tank and tap water lead concentrations at properties where both existed revealed that tank supplies were significantly lower than concentrations detected at the tap (Fig. 6, samples 1, 2 and 3). All kitchen tap water samples exceeded the ADWG (10 µg/L) for lead concentrations. The fittings analysed in these case studies were all < 1 year of age. The fourth case study property had a lead concentration of 14.0 µg/L from a rainwater tank that collected run-off from a shed roof (Fig. 6, sample d). In this instance, the source of the lead appeared to be the screws used on the roofing material of the shed that contained 6320 mg/kg lead (Supplementary Data 4).

3.4. Phase 3 Laboratory leaching experiment

Aliquots collected during the 7-day sampling period contained very high concentrations of lead (mean – 328 µg/L, median – 225 µg/L, n=28, Fig. 7, Supplementary Data 6). By contrast, water entering the system and monitored at the purge valve or tank contained a negligible lead concentration (mean – 1.6 µg/L, median – 1.6 µg/L, n=3). The lead concentration of the fitting with the copper riser only (fitting 1) decreased markedly over the 7-day

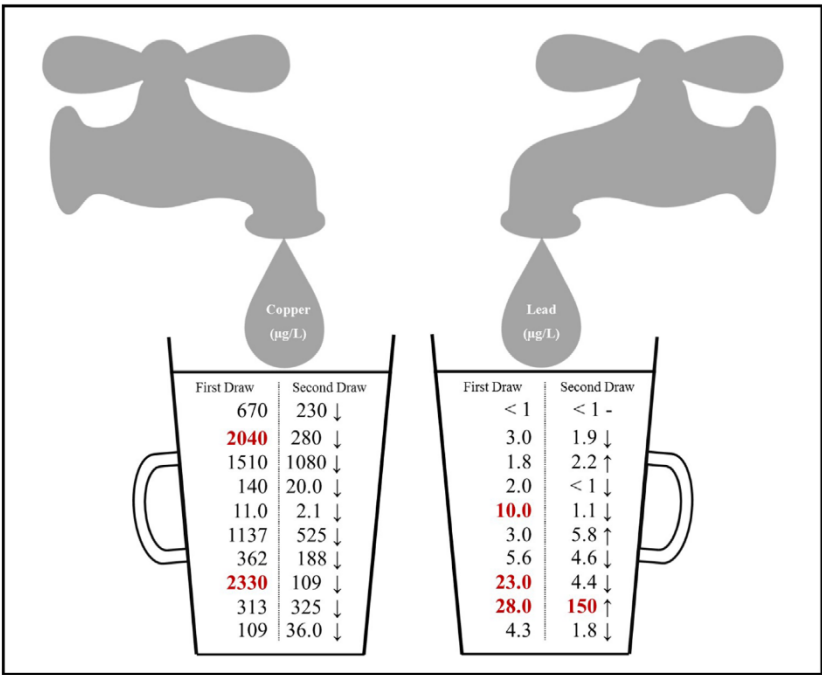


Fig. 5. Copper and lead concentrations in first draw and flushed (2 min) drinking water samples from NSW, Australia. Bold number indicate exceedance of the ADWG (copper=2000 µg/L, lead=10 µg/L). Directional arrows ↑= increase in concentration in second draw, ↓= decrease in concentration in second draw, – = no change in concentration in second draw sample.

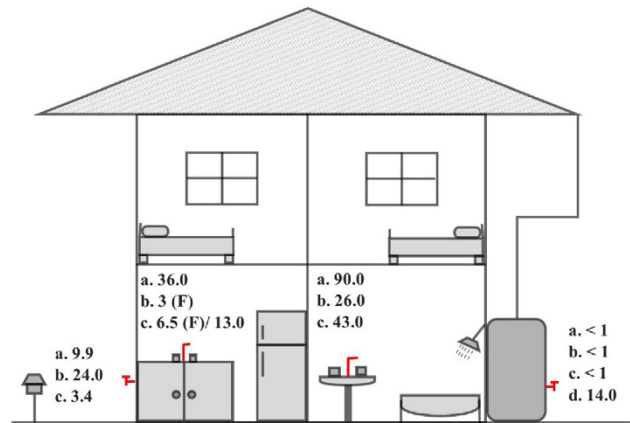


Fig. 6. Lead concentrations in water samples collected from four case study properties (a – CS1, b – CS2, c – CS3, d – CS4; refer to Supplementary Data 4) in rural NSW. Samples are from four household locations (left to right): outdoor tap, kitchen tap (sample b (F) and c (F) have been collected from a filter), bathroom tap and tank. Concentrations are in µg/L.

sampling period (1440 µg/L to 108 µg/L). Samples collected from the riser and tap (fittings 2, 3 and 4) had less water in direct contact with the brass fitting of the riser resulting in lower lead concentrations compared to the riser only samples. Lead concentrations from the riser and tap set-up decreased over the sampling period, but remained elevated above the ADWG over the 7-day sample period (n=28, mean – 327 µg/L, median – 225 µg/L). By contrast, copper concentrations were not elevated (n=28, mean – 428 µg/L, median – 389 µg/L) compared to the ADWG (2000 µg/L, Supplementary Data 6).

Further analysis of water from the tap fittings (Part B Fig. 1 and Panel b Fig. 7) showed very high lead concentrations (n=9, mean – 63.4 µg/L, median – 59.0 µg/L). Lead concentrations remained

constant throughout the 3-day sampling period. Analysis of the lead-free tap fitting showed that < 1 µg/L was leached to water samples over the sampling period. Due to the copper water values being below the ADWG in the riser set-up, water samples from tap fittings were not subsequently analysed for this element. Analysis of the brass components in contact with water within the taps showed a very high concentration of lead (2.84%).

3.5. Determination of potential exposure risk to drinking water lead contamination

Barton et al.'s (2002) model for potential health impacts of exposure used in this study showed that the child exposed to the average copper and lead concentrations detected in the study (copper – 364 µg/L, lead – 3.9 µg/L) would be exposed through ingestion to 318 µg/L copper and 3.4 µg/L lead per kg body weight in a seven day period. By comparison, for the maximum concentrations in this study, copper at 3800 µg/L and lead at 89 µg/L, the same infant would be exposed to 3330 µg/L copper and 77.0 µg/L lead per kg body weight in a seven day period. The modelled exposures in this study are markedly greater than those identified in Barton et al. (2002).

4. Discussion

Of the four potential toxic contaminants examined in this study (arsenic, copper, lead and manganese), two contaminants, copper and lead, present a concern for public health due to their frequent occurrence and high concentrations (Fig. 3, Supplementary Data 2). In contrast to lead and copper water concentrations, arsenic and manganese were detected only rarely in the drinking water and when present were at low concentration (Supplementary Data 2). Therefore, lead and copper form the focus of the following discussion.

Given that sampling of household water took place across NSW,

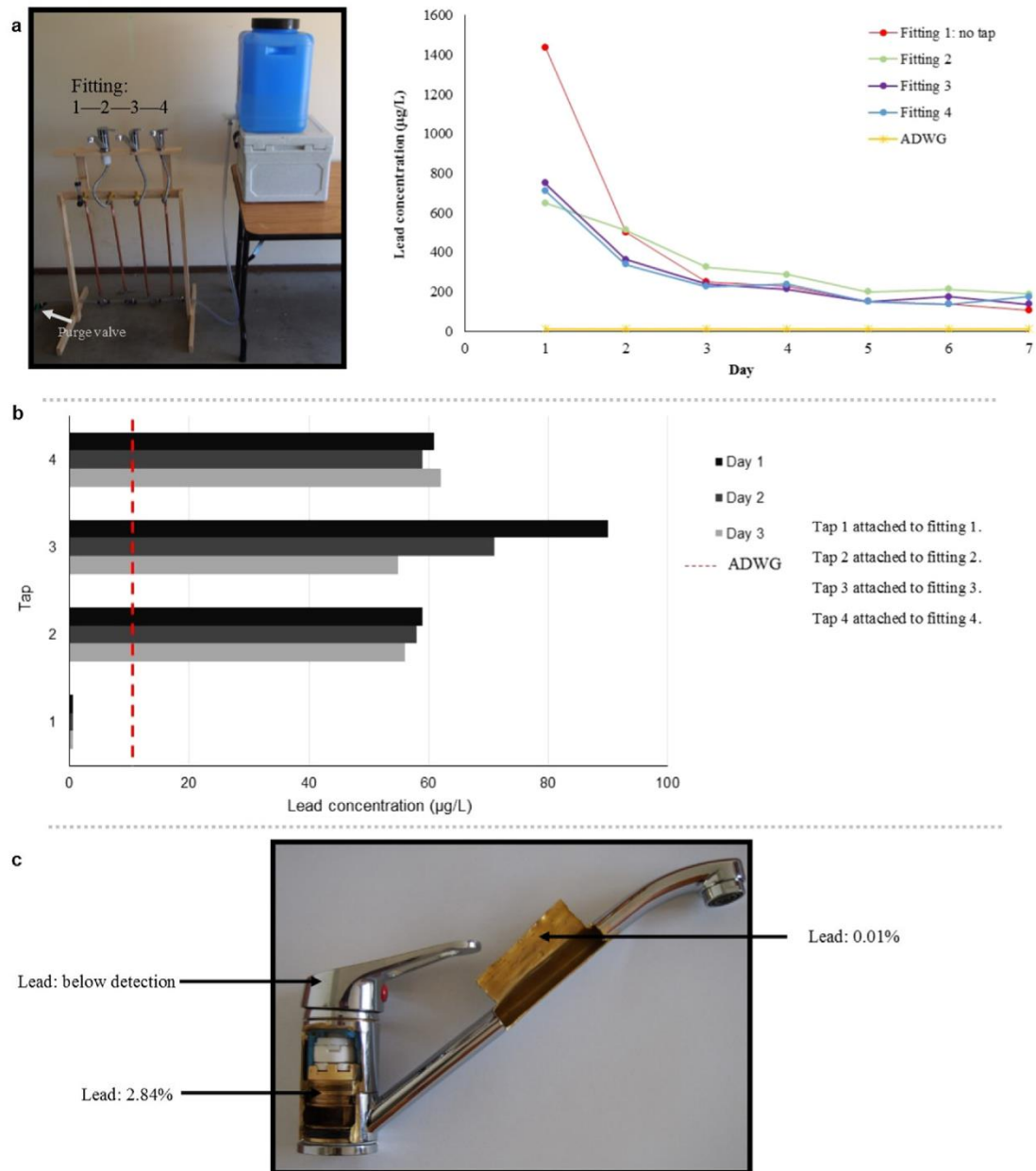


Fig. 7. Domestic plumbing component lead testing. Panel a is the first test using both rise and tap, panel b is the tap only and panel c is the metal composition. Panel a 1 uses all fittings that contain lead. Panel b 2 uses lead containing fittings for taps 2, 3 and 4. Tap 1 is a lead free fitting.

the spatially diverse distribution of the samples containing concentrations above the ADWG for copper and lead illustrates the unpredictable occurrence of drinking water contamination in NSW (Fig. 3). Drinking water contamination is not regionally specific (Fig. 3) and there is no clear regional correlation between the two elements. Nevertheless, there is a significant health risk associated with consumers, particularly infants consuming formula (Edwards et al., 2009; Gulson et al., 1997a; Shannon and Graef, 1989; Troesken, 2008), being potentially exposed to elevated concentrations of copper and lead in tap water, with maximum recorded values of 3800 µg/L for copper and 89 µg/L for lead. Consistency in replicates (Supplementary Data 3) shows that concentrations detected in this study are not one-off events and are prolonged over time, increasing the exposure risk.

The individual case studies reveal that new properties (< 5

years) are also at risk of lead entering the drinking water, demonstrating this is not necessarily an issue associated with lead water supply lines of older properties. Interestingly, copper, although present, was not noted to be of particular concern in these properties (Supplementary Data 5).

4.1. Lead in plumbing components

Brass taps and fittings have, for many years, been a known source of drinking water contamination (Dudi et al., 2005; Elfland et al., 2010; Gardels and Sorg, 1989; Gulson et al., 1994, 1997a; Lytle and Schock, 1993; Schock and Neff, 1988). Despite this knowledge, plumbing components containing large concentrations of lead continue to remain available to consumers (Fig. 7). Plumbing components referred to as 'end-of-line fittings' in the

Australian/New Zealand Standard, are 'any product, or part of a product, installed within 250 mL draw-off of a drinking water delivery point' (AS/NZS, 4020:2005). Samples collected in this study are 1 L of the first draw water, meaning that a large proportion (25%) of the sample was drawn from the end-of-line fitting after the water had been in contact with those fittings for a prolonged period of time. The case studies (Phase 2) show that the likely source of drinking water lead contamination is the components within the internal property plumbing (maximum detected 90 µg/L). The lead concentrations from the source water tank are considerably lower than the water reaching the consumer (Fig. 6). The highest lead concentration was detected in the bathroom, reflecting the quality of the fittings, their manufacturing process, the size of the fittings or the flow conditions due to location within the property (Patch et al., 1998). Examining the lead concentrations of Phase 3 samples, it is evident that end-of-line fittings are contributing to the elevated lead concentration in household drinking water (Fig. 7, Supplementary Data 6). These results, after almost two decades, mirror those of Gulson et al. (1994, 1997a) who showed that lead brass was a contributor to drinking water lead contamination in suburban Sydney homes. Lead-brass taps, purchased over the counter from a national Australian retail hardware store, are clearly contributing very high concentrations of lead to drinking water (Fig. 5). Importantly, in terms of providing end users with relevant knowledge, it is worth noting that the products did not contain any labelling or warnings of the potential health risks associated with use of the product. These tests of a random sample of commercially available taps and fittings show lead leaching from lead-brass taps and fittings is a persistent problem and contradicts the ADWG (2011a) statement that 'the issue of plumbosolvency is rare in Australia' (p. 136). It is evident that the current Australian method of regulation and testing of products containing lead pursuant to the goal that the 'elimination of unnecessary sources of lead would be of benefit in reducing the exposure of children' (Greene et al., 1993) (p. 116) has not been reached.

4.2. The association between copper and Lead Contamination

Copper and lead concentrations were not co-associated in the first draw samples (Fig. 4, $R^2=0.05$). Galvanic reactions occurring at the boundary between copper pipes and brass fittings in household plumbing while the water is stagnant is known to cause both contaminants to leach from the metals, but not always concurrently (Clark et al., 2015; Dudi et al., 2005; Nguyen et al., 2010; Schock and Neff, 1988). The decreased concentrations of copper in the samples following a 2-minute flush period suggest that once the stagnated water has passed and water is flowing, there is reduced dissolution of the copper pipe, limiting exposure to the consumer. In contrast, lead concentrations did not decrease following flushing in all instances (Fig. 5). The reasons for this have been examined extensively in the literature and include, among other things, lead particulate re-mobilisation under high pressure flow, which occurs when the lead precipitate scale lining on the inside of the pipes is degraded during intermittent fluctuations in water chemistry and flow (Triantafyllidou et al., 2007).

4.3. Recommendations for the consumer

4.3.1. Flushing samples to reduce contaminant concentrations

The common advice provided to consumers is to flush the taps prior to consumption for 2–3 min (NSW HEALTH, 2014). Despite following this established protocol, some of the flushed water samples still contained considerable concentrations of both copper and lead (Fig. 5). Moreover, it is not always practical for water-scarce rural districts of Australia to purge the water for extended

periods each day. For example, if consumers were to flush the taps for 2-min each morning, using approximately 3.7 L/min, approximately 2000 L would be wasted annually, per household. Further, this information may be misleading and ineffective for consumers who rely on rainwater tanks as the captured run-off may have already become contaminated by lead flashing, solder or paint prior to water reaching the internal plumbing, which may only exacerbate the problem.

4.3.2. Tank supplies

Elevated copper and lead concentrations detected from water supplies sourced from water tanks indicate that there is an apparent complacency among the community when collecting water for consumption. Tank water supplies are a well-documented source of drinking water contamination either through leaching of roofing components including lead paint or uPVC pipes, or from external sources such as contaminated atmospheric fallout onto the roof catchment (Al-Malack, 2001; Huston et al., 2009, 2012; Magyar et al., 2014; Rodrigo et al., 2012; van der Sterren et al., 2013). Many participants indicated on their consent form that they had noted lead flashing on their roof catchment only when collecting for this study. Conversations with members of the community indicated a lack of knowledge surrounding the risks associated with drinking water contamination, particularly exposure in high-risk populations (pers. comm. Sandy Aureli, Milton CWA Secretary 2015). While common sense often discourages homeowners from drawing from a roof catchment visibly impacted by bird droppings or leaf litter, the dangers of metal contamination is typically overlooked.

4.3.3. Lead free taps

Testing in the study shows that using a tap made from stainless steel, which does not contain lead, prevents lead entering the water at the tap stage of the plumbing infrastructure. However, the evidence indicates there remains an unresolved contribution to drinking water supplies from the in-line brass fittings within the household plumbing system. In the USA, the lead content of new products in contact with potable water is strictly regulated and cannot exceed 0.25% weighted average (NSF, 2016). This lower lead content has been shown to present a substantially reduced risk of lead release into drinking water even when aggressive (i.e. acidic) water is present (Pieper et al., 2016). Lead free taps are available for purchase from some suppliers, however these are often far more expensive (~3 times the price) than the lead-brass taps. Further, we noted that these lead-free taps were not readily available at the same hardware stores and it would require specific consumer knowledge to request such a product.

4.3.4. Managing corrosion control

Various corrosion control measures are used in urban reticulated water supplies to reduce the movement of copper and lead into the drinking water scheme. Many reticulated water schemes in Australia have corrosion control measures in place (NHMRC, 2011b). Water used in the home environment on rural properties is often sourced from a private rooftop catchment and rainwater tank. These supplies are unlikely to be treated to prevent corrosion of internal plumbing from existing lead containing fittings. Installation of concrete tanks can release calcium into the water, reducing the pH and subsequently the corrosion of pipes and fittings (Abbasi and Abbasi, 2011). Adding a buffering agent such as lime to the tank water may also assist in corrosion control (Moore et al., 1985; NSW DPI, 2014). The most ideal solution to this problem is to remove all lead from fittings (tap 1, Fig. 7).

4.3.5. Limitations of this study

Pieper et al. (2015) highlight the potential for data to be skewed

due to volunteer participants only responding when they suspect an issue with their drinking water, resulting in the most contaminated samples being analysed. Volunteer participants are also untrained in sampling processes, which can potentially increase sampling errors and contamination. While this is a risk, the large spatial coverage of sampling and the detailed instruction provided to participants likely negates these influences.

While there is a clear contribution of lead to the drinking water from tap fittings (Fig. 4), there are potential additional contributions from lead paint (on the roof), lead flashing, uPVC piping and degraded infrastructure in the supply network that can influence water contamination measures (Harvey et al., 2015; Magyar et al., 2014). These additional contributions are further impacted in tank and bore water supplies by regional rainfall patterns, atmospheric pollutant loading and aquifer contamination which can accelerate roof and pipe corrosion (Magyar et al., 2014).

This study did not examine the source waters supplying the individual properties and in doing so makes the assumption based on the limited literature that these supplies are contributing negligible concentrations of contaminants to the property (Li et al., 2009). Regardless, the purpose of this study is to examine the risk of drinking water contamination at the consumer's tap under everyday conditions.

4.3.6. Childhood exposure to lead and copper in drinking water

Infant exposure to environmental contaminants places those infants at increased risk of developing a range of detrimental health outcomes, including among many others, diminished cognitive function and physical development delay (Edwards et al., 2009; Gulson et al., 1997a; Troesken, 2008). The greatest health risks related to copper and lead exposure occur in the developing baby, both pre- and post-natal (Hanna-Attisha et al., 2015). Infants fed formula reconstituted by lead and copper contaminated water has been extensively documented as an important exposure pathway (Edwards et al., 2009; Gulson et al., 1997a; Shannon and Graef, 1989; Troesken, 2008). Further, the cooking of absorbent food (e.g. potatoes and pasta) in contaminated water poses a significant exposure pathway (Baxter et al., 1992; Little et al., 1981). Both copper and lead are presented in elevated concentrations in this study. The modelling in this study, based on Barton et al. (2002), shows that the children under the worst case scenario seen in this study have a significant risk of exposure to concentrations of contaminants that can pose detrimental health impacts.

4.3.7. Implications of this research in the Australian and international context

While this study examines only one state of Australia, in terms of population it represents an area occupied by approximately 32% of Australians. When the results of this study are extrapolated out to Australia more broadly, the findings imply there is a significant and widespread risk of preventable copper and lead contamination of drinking water. This conclusion is supported by the literature, where more spatially isolated studies across Australia have identified major drinking water contamination events (Alam et al., 2008; Cohen, 22nd June, 2000; Gulson et al., 1994, 1997a, 1997b; Harvey et al., 2015; Huston et al., 2009, 2012; Magyar et al., 2014; NSW DET, 2011; Rajaratnam et al., 2002; Rodrigo et al., 2012; van der Sterren et al., 2013; Yau and Thorne, 2012).

For the first time, this study shows clearly that drinking water contamination in Australia is comparable to that identified in the Americas, Asia and Europe (Del Toral et al., 2013; Edwards et al., 2009; Fertmann et al., 2004; Jane Wyatt et al., 1998). Although this study only specifically examines four elements of interest, it highlights the potential for a more widespread problem of elemental drinking water contamination. As lead contaminated

drinking water supplies are rapidly becoming identified across the United States of America (Bellinger, 2016; Hanna-Attisha et al., 2015; Pieper et al., 2015; Wang 2016; Young and Nichols 2016), this study serves as a reminder that drinking water contamination is a global problem.

In terms of mitigating preventable exposures, the study acknowledges that education and public awareness is the first step in resolving this widespread problem (Pfadenhauer et al., 2016). Homeowners should be made aware of the risk present in the products that they purchase for use in contact with their potable water supply. Products containing these contaminants should be either removed from sale or have suitable product labelling and consumer information attached. There also needs to be an increase in awareness for those who draw water from roof catchments of the risks associated with the potential contamination sources used within their catchment.

5. Conclusions

The results of this study show a long term, ongoing exposure pathway to environmental copper and lead through drinking water. Copper and lead detected in samples from across New South Wales are an ongoing concern as they pose a risk of exposure for consumers, especially infants. The widespread distribution of samples with elevated copper and lead concentrations demonstrates that this is not a spatially isolated problem, and that domestic supplies across Australia are likely to be subject to similar issues. The laboratory leaching study of new taps demonstrates that commercially available plumbing products pose an appreciable source of exposure to known toxic contaminants. Overall, this study highlights a dearth of knowledge in the community about the potential risk associated with exposure to copper and lead via drinking water, a problem exacerbated by the availability of predominantly lead containing products for household plumbing.

Acknowledgements

Without the community of New South Wales, particularly of the Country Women's Association of New South Wales (CWA) this study could not have occurred. Donna Wilkins of the CWA is thanked for her assistance with initial project development. Chenyin Dong, Nathan Harvey, Wendy Harvey, Nóra Liptai, Beñat Oliveira Bravo, Marek Rouillon and Yu Wang are thanked for their work assisting in sample collection. Ron Aggs, Sandy Aureli, Anne Bayliss, Kathryn Rogan, Carol Whiting and many others are thanked for their tireless effort organising sample collection and their hospitality. Dr Ping Di, Dr Andrew Evans and Dr Michael Wu of the National Measurement Institute, Inorganics Laboratory are thanked for analytical assistance. A/Prof Norman Pearson and Peter Weiland of the Geochemical Analysis Unit, Macquarie University are thanked for laboratory assistance. P.J. Harvey would like to thank Prof Damian Gore and Prof Brian Gulson who assisted with some aspects of project design. Dr Louise Kristensen and Catherine Stuart are thanked for early manuscript reviews. Some participants are thanked for contributing to the laboratory cost of sample analysis. P. Harvey is funded by a Macquarie University Research Excellence Scholarship (MQRES) (2012195) associated with an Australian Research Council Future Fellowship awarded to H. Handley (FT120100440).

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at <http://dx.doi.org/10.1016/j.envres.2016.07.041>.

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VegeSafe: a Community Science Program Measuring Soil-metal Contamination, Evaluating Risk and Providing Advice for Safe Gardening.

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Growing vegetables is a fun and rewarding hobby however many home growers are unfamiliar with the potential environmental contamination risks in their gardens. This study examines the potential for metal(loid) contamination of garden soils in urban Sydney. This study employs a citizen science approach to sample collection whereby members of the community were invited to submit samples to the Macquarie University VegeSafe program for free analysis, complete with sample metadata such as sample collection location and details about the age of the property. In return, participants were provided with a results report and information on ways to mitigate any potential health risks associated with metal(loid) contamination. This study identifies the older inner-city areas of Sydney to contain the greatest soil metal(loid) concentrations, with lead the most commonly detected metal. Soil metal(loid) contaminants were identified in greatest concentrations adjacent to property driplines and where heavy industry, older roadways or un-clean fill was present. This study adds nicely to the contaminants in the home section of this thesis because it highlights yet another overlooked source of environmental exposure.



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Invited paper

VegeSafe: A community science program measuring soil-metal contamination, evaluating risk and providing advice for safe gardening[☆]

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ARTICLE INFO

Article history:

Received 15 September 2016

Received in revised form

3 November 2016

Accepted 5 November 2016

Available online xxx

Keywords:

Lead

Soil contamination

Sydney

Urban gardens

X-ray fluorescence spectrometry

ABSTRACT

The extent of metal contamination in Sydney residential garden soils was evaluated using data collected during a three-year Macquarie University community science program called VegeSafe. Despite knowledge of industrial and urban contamination amongst scientists, the general public remains under-informed about the potential risks of exposure from legacy contaminants in their home garden environment. The community was offered free soil metal screening, allowing access to soil samples for research purposes. Participants followed specific soil sampling instructions and posted samples to the University for analysis with a field portable X-ray Fluorescence (pXRF) spectrometer. Over the three-year study period, >5200 soil samples, primarily from vegetable gardens, were collected from >1200 Australian homes.

As anticipated, the primary soil metal of concern was lead; mean concentrations were 413 mg/kg (front yard), 707 mg/kg (drip line), 226 mg/kg (back yard) and 301 mg/kg (vegetable garden). The Australian soil lead guideline of 300 mg/kg for residential gardens was exceeded at 40% of Sydney homes, while concentrations >1000 mg/kg were identified at 15% of homes. The incidence of highest soil lead contamination was greatest in the inner city area with concentrations declining towards background values of 20–30 mg/kg at 30–40 km distance from the city. Community engagement with VegeSafe participants has resulted in useful outcomes: dissemination of knowledge related to contamination legacies and health risks; owners building raised beds containing uncontaminated soil and in numerous cases, owners replacing all of their contaminated soil.

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1. Introduction

Urban agriculture in the form of city farms, community gardens and verge gardens, are rapidly growing in popularity in many cities across the world (Alaimo et al., 2008; Keisling and Manning, 2010; Bugdalski et al., 2013; Mitchell et al., 2014; City of Sydney, 2016; City of West Hollywood, 2016). While the motives for urban agriculture may be diverse, the social and mental health benefits of urban green spaces and community involvement are well

established (Alaimo et al., 2008; Wise, 2014). In Australia, 89% of the population live in cities (World Bank, 2013), with almost half (48%) of all households in metropolitan areas growing some form of edible produce (Wise, 2014). A recent Australian survey by Wise (2014) found most food gardens are located in either front or back yards (74%), 13% of participants grow food on street verges, 12% on balconies or in container gardens and only 1% from community gardens. In Sydney, food gardens are often located in older, heavily urbanized suburbs and are harvested from both above ground, and in ground vegetable plots.

Urban soils are notorious sinks for lead (Pb) and other metal and metalloid (hereafter referred to as metal) contaminants as a result of industrial and historic traffic emissions, waste incineration and application of paint containing Pb (hereafter referred to as Pb paint)

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<http://dx.doi.org/10.1016/j.envpol.2016.11.024>

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Please cite this article in press as: Rouillon, M., et al., VegeSafe: A community science program measuring soil-metal contamination, evaluating risk and providing advice for safe gardening, Environmental Pollution (2016), <http://dx.doi.org/10.1016/j.envpol.2016.11.024>

(Birmili et al., 2006; Szolnoki et al., 2013; Laidlaw et al., 2014; Kristensen, 2015). In Sydney alone, approximately 34,000–41,000 tonnes of Pb were emitted from the tailpipes of motor vehicles in the metropolitan area during 70 years of leaded fuel use (Kristensen, 2015). Widespread application of Pb paint (1–50 wt% Pb) on houses built pre-1970 was common with the majority of houses built during this period being painted with Pb paint at some point (NSW EPA, 2016a).

Metal contamination in residential garden soils is well documented. Contaminated garden soils are a global problem: Canada (Hendershot and Turmel, 2007), Denmark (Warming et al., 2015), France (Douay et al., 2013), Hungary (Szolnoki et al., 2013), Italy (Ferri et al., 2015) and the United States (Finster et al., 2004; Clark et al., 2008; Cheng et al., 2011; Mitchell et al., 2014; Filippelli et al., 2015). Despite a range of industrial metals being present in garden soils (Birmili et al., 2006; Szolnoki et al., 2013), Pb is typically the metal of most concern due to its toxicity, persistence and abundance in the urban environment (Lanphear et al., 1998; Mielke et al., 1999). To address this pervasive issue, screening programs, such as the New York State Department of Health soil screening and outreach program (New York State Department of Health, 2016), have been set up to raise awareness of soil Pb contamination within communities.

Widespread environmental Pb contamination has been previously documented in Sydney and its suburbs (Laidlaw and Taylor, 2011). A study by Fett et al. (1992) measured garden soils at 18 inner Sydney residential properties (hereafter referred to as homes) and found soil Pb concentrations up to 5400 mg/kg, with a median of 1944 mg/kg. A further study by Olszowy et al. (1995) showed that soils with the greatest mean (379 mg/kg) and median Pb (225 mg/kg) concentrations were from older Sydney suburbs with higher traffic density. The lowest soil Pb concentrations were from newer Sydney suburbs with lower traffic density. In the inner-Sydney suburb of Glebe, Markus and McBratney (1996) analyzed 219 soil samples from home gardens, parks, and nature strips and found 50% of soil samples exceeded the Australian residential Pb guideline of 300 mg/kg. Recent studies of urban soils by Snowden and Birch (2004) and Birch et al. (2011) identified median soil Pb of 203 mg/kg ($n = 374$) in the Sydney Iron Cove catchment and 150 mg/kg ($n = 491$) in Port Jackson (Sydney harbour) estuary catchment, respectively. In contrast, studies that specifically investigate vegetable garden soil contamination in Australian homes are scarce. Kachenko and Singh (2006) measured metals in food-growing soils and vegetables from 24 sites within the Sydney basin, and despite low soil metal concentrations (Pb 2.8–198 mg/kg), 32% of vegetables exceeded the allowable Pb levels set by the Australian and New Zealand Food Authority, respectively.

Previous studies in Sydney have often focused in publicly accessible areas (e.g. street verges, open spaces and parks), mainly due to the difficulties of accessing a large number of private properties for a suitable sample size. Metals found in public open spaces may not be representative of contamination at homes where different sources and soil practices exist (Mielke et al., 1983; Schwarz et al., 2012). Deterioration of Pb paint from old buildings (Gulson et al., 1995; NSW EPA, 2016a) along with the domestic use of pesticides (Kessler, 2013; Szolnoki et al., 2013) are sources of copper (Cu), zinc (Zn), and arsenic (As) contamination in domestic gardens.

1.1. Public access to soil metal screening via the VegeSafe program

To assist urban gardeners in Sydney and Australia more broadly to understand their soil environment, we initiated a free soil metal screening program called VegeSafe in 2013 (VegeSafe, 2015) to delineate, evaluate, and advise about potential risks arising from

metal contamination (ABC, 2014, 2015). This community focused program provides free soil metal screening to participants using field portable X-ray Fluorescence (pXRF). The program provides participants with a formal report on their soil metal results and advice about 'what to do next' in the event of soils containing elevated concentrations of metals (VegeSafe, 2015). Participants provide a signed consent form with their samples permitting the use of their de-identified data in research. Similar programs, such as the Safe Urban Gardening Initiative in Indianapolis, USA, have been successful in engaging urban gardeners to better understand metal contamination in an urban setting (Filippelli et al., 2015). As of September 2016, VegeSafe has provided >5200 free soil metal tests to >1200 homes and community gardens across Australia, representing unprecedented access to the otherwise largely inaccessible private soil environment of Australia. The program aims to encourage participants to grow their own food in the knowledge that their soils contain metals below what are considered to be acceptable thresholds for gardening. Hence, this study investigates soil metal concentrations at Sydney homes using data obtained through the VegeSafe program. Discussion of Pb sources in the urban environment is also presented.

2. Methods

2.1. Sample collection and preparation

Samples were collected by VegeSafe participants from up to four areas around their homes including: front yards, drip lines (adjacent to houses), back yards (non-food growing soil) and vegetable gardens (food growing soil). Participants followed a sampling protocol that involved sampling soil at 0–2 cm depth and placing into labelled zip-lock plastic bags (Supplementary Fig. 1). Participants completed consent forms (Supplementary Fig. 2) and also provided metadata including sample location, age of house and presence of exterior painted surfaces. Samples were received via mail parcels to the University or at one of various VegeSafe events across Sydney (VegeSafe, 2015). For this study, 410 soil samples were selected from 203 homes within a pre-defined area consisting of 22 local government areas (LGAs) of Sydney (Fig. 1). The study area was determined using soil metal results from previous studies in public areas (e.g. Olszowy et al., 1995; Birch et al., 2011) as well as from the VegeSafe program to focus on homes most likely impacted by metal contamination. Twenty six reference samples were collected from eight semi-rural properties outside of this study area to represent soil concentrations in what were estimated to be areas largely unaffected by metal contamination (Fig. 1).

Samples were air dried at 40 °C for 72 h, crushed using a mortar and pestle and sieved through a 2 mm stainless steel sieve to remove coarse debris before passing through a 500 µm sieve. Composite sample preparation was carried out when multiple samples of the same type (i.e. 3 vegetable garden samples) were collected from the same home. Equipment was cleaned between samples with ethanol and KimWipes. Samples were thoroughly mixed by turning the sample bags over for 180 s each prior to packing in XRF cups. Approximately 10 g of <500 µm soil sample were packed in 35 mm open ended PANalytical XRF cups using 3.6 µm Chemplex Mylar Thin-Film for analysis.

2.2. Field portable X-ray Fluorescence (pXRF) analysis

An Olympus Premium Innov-X XRF Analyser fitted with a 50 kV, 4 W Ta anode X-ray tube and a silicon drift detector was used for the measurement of Mn (manganese), Cu, Zn, As and Pb concentrations in soils. Operational procedures followed throughout this study included: daily measurements of an energy calibration check,

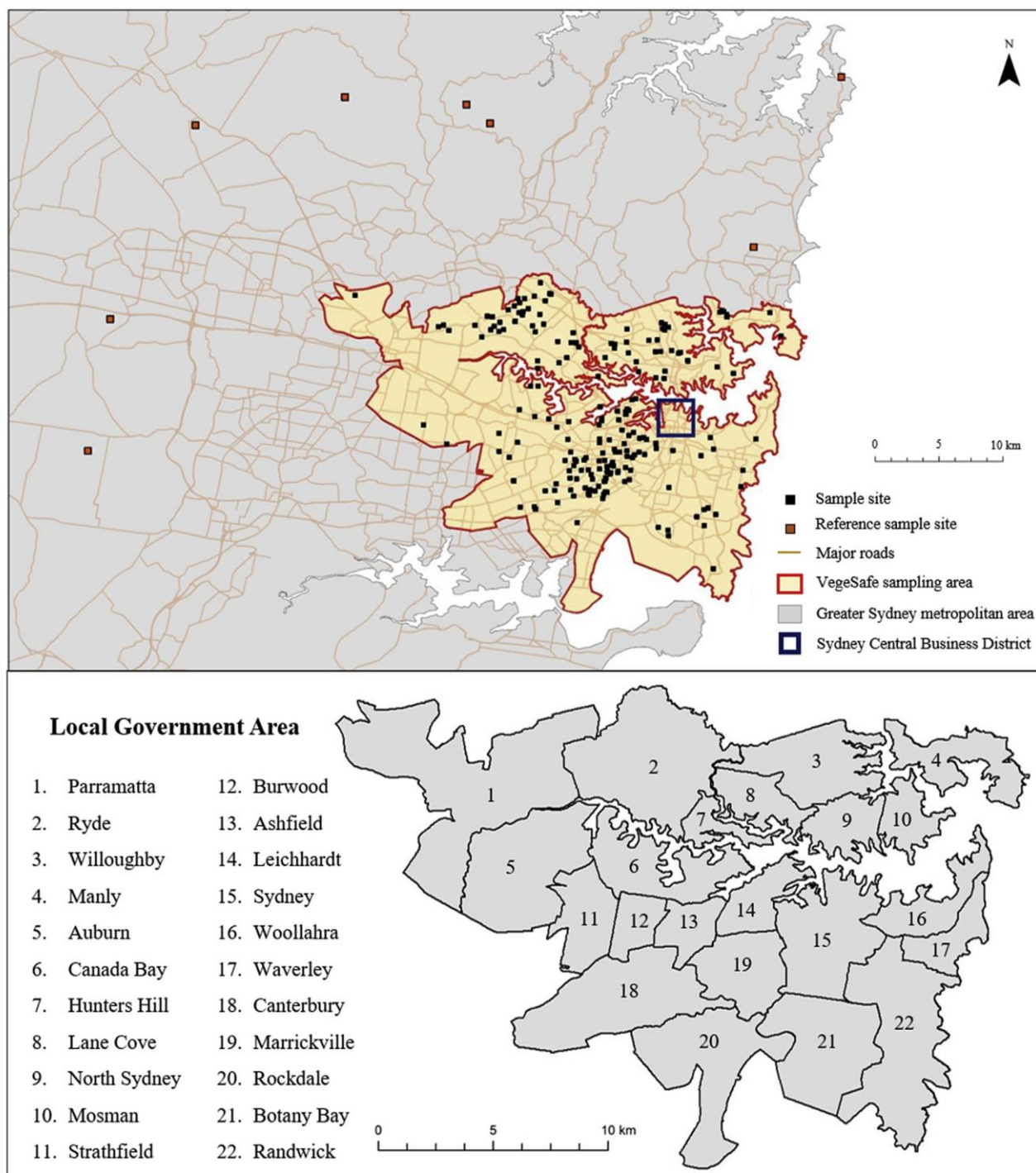


Fig. 1. Sample site locations of 203 homes within the sampling area of 22 Local Government Areas. Eight reference homes are also displayed towards the outer Sydney metropolitan area. Note: this image was generated prior to New South Wales council amalgamation in 2016 ([Local Government New South Wales, 2016](http://www.localgovernment.nsw.gov.au/)).

measurements of a silicate (SiO_2) blank at the beginning and end of daily sample analysis to ensure no instrument contamination had occurred, and measurement of National Institute of Standards and Technology certified reference materials (CRMs: NIST 2586, 2587, 2709a, 2710a, 2711a) every 20–25 samples to monitor instrument

performance. Soil samples and CRMs were measured at 60 s per X-ray beam (180 s total measurement time) using the proprietary soil mode. A matrix matched calibration was applied to the pXRF using the method defined in [Rouillon and Taylor \(2016\)](#) to optimize analytical performance for metal-contaminated silicate-based soils.

Table 1

Summary of detection limits, precision and accuracy achieved by pXRF. CRM values and instrument detection limits are displayed as mg/kg. Relative standard deviation of ten CRMs^a and ten <500 µm soil samples^b. Recovery was calculated as (pXRF value/reference value × 100), relative proximity as (absolute (100 – recovery value)) and the relative standard deviation as (standard deviation/mean × 100).

	Mn	Cu	Zn	As	Pb
Minimum CRM value	229	19	103	9	17
Maximum CRM value	2140	3420	4180	1540	5520
Instrument detection limits	9	4	5	3	3
Analytical precision ^a	0.8%	2.4%	0.8%	7.3%	1.8%
Analytical precision ^b	1.0%	2.8%	0.7%	9.4%	0.8%
Sample homogeneity	5%	4%	4%	18%	4%
Recovery	102%	108%	99%	106%	99%
Relative proximity	3%	8%	5%	12%	3%

The pXRF instrument was operated in a shielded test stand to eliminate radiation exposure associated with extensive pXRF use (Rouillon et al., 2015).

2.3. Quality assurance and quality control

Ten CRMs of similar compositional matrix and trace element concentration were selected to assess the quality of pXRF measurements (cf. Rouillon and Taylor, 2016). The CRMs were mixed thoroughly in their bottles by shaking prior to packing in 35 mm open ended PANalytical XRF cups using 3.6 µm Chemplex Mylar Thin-Film. Instrument detection limits, analytical precision, sample homogeneity, mean recoveries and relative proximity (RP) of each element are provided in Table 1. Analytical precision was determined from the relative standard deviation (RSD) calculated from the five measurements that used for determining each measurement mean. Sample homogeneity was measured through triplicate preparation and analysis of ten soil samples. An additional validation step for the pXRF analytical data was obtained by processing 49 soil samples in a four acid sample decomposition (HClO₄, HNO₃, HF and HCl acid) and analysed using a Varian 725-ES inductively coupled plasma atomic emission spectrometer (ICP-AES) for their 'near total' metal concentrations (ALS, 2009). Three procedural laboratory blanks returned mean concentrations of <2 mg/kg for Cu, Zn and Pb and <5 mg/kg for Mn and As. Excellent agreement between pXRF data with CRM (r^2 0.999) and ICP-AES data (r^2 0.999) demonstrates pXRF was a robust alternative to ICP-AES for the measurement of metals in soils (Supplementary Fig. 3; Rouillon and Taylor, 2016).

3. Results and discussion

3.1. Soil metal concentrations

Soil metal data for Sydney soils are presented in Table 2. Soil concentrations were benchmarked against reference values determined from control sites at outer Sydney homes (Fig. 1) and the Australian health investigation level for residential soils (HIL-A) (NEPM, 2013). The mean Mn concentration within the sampling area (306 mg/kg) was not significantly different ($p > 0.05$) to the reference mean (360 mg/kg) despite the brief use of methylcyclopentadienyl manganese tricarbonyl (MMT) as an additive in Pb replacement petrol (2001–2004) during the phasing out of leaded petrol (Australian Government, 2005; Cohen et al., 2005). These findings are consistent with work by Bhuie and Roy (2001) who found no significant increase in soil Mn away from highways, despite 25 years of MMT use in Canada. Similarly, Gulson et al. (2014) found no significant relationships between traffic

proximity and Mn concentrations in soil. The maximum Mn concentration of 1040 mg/kg was well below the Australian HIL-A guideline of 3000 mg/kg (Table 2) and subsequently was not investigated further.

Concentrations of Cu, Zn and As were significantly greater ($p < 0.001$) than reference values. This is not unexpected given the widespread use of Cu, Zn and As in a range of industrial products including pesticides (Szolnoki et al., 2013), paints (Gulson et al., 1995; NSW EPA, 2016a), and motor-vehicles (Wuana and Okiemen, 2011). The maximum concentration of Cu (717 mg/kg) and Zn (2880 mg/kg) did not approach the Australian HIL-A guideline of 7000 and 8000 mg/kg, respectively. By contrast, several homes had soil As concentrations near the Australian guideline value of 100 mg/kg. While no samples exceeded the HIL-A guideline for As, the data demonstrates that almost half (46%) would exceed the more conservative Canadian soil guideline of 12 mg/kg (CCME, 2013). Even though none of the metal concentrations other than Pb exceeded the Australian HIL-A guideline, this does not preclude potential adverse health effects from exposure to these contaminants at lower levels (Aelion et al., 2009).

The mean Pb concentration within the study area was 415 mg/kg, considerably greater than the reference mean of 33 mg/kg measured in outer Sydney soils, and the mean geogenic Pb concentration (<30 mg/kg) established in the literature (Olszowy et al., 1995; Kachenko and Singh, 2006; Wu et al., 2016). Maximum soil Pb concentrations were 2460 mg/kg (front yard), 6490 mg/kg (drip line), 1810 mg/kg (back yard), and 3080 mg/kg (vegetable garden) (Table 2). Forty percent of homes within the study area contain soil in excess of the Australian HIL-A guideline of 300 mg/kg for Pb in one or more garden areas; 21% exceeded this value in two or more garden areas. Soil Pb concentrations >1000 mg/kg were identified at 30 of 203 (15%) Sydney homes.

3.2. Spatial distribution of Pb (home scale)

Soil metal concentrations varied significantly across homes. Median soil Cu, Zn, As and Pb concentrations were greatest at drip line locations and typically decreased towards the other three garden locations sampled (Table 2). Median soil Pb concentrations were 174 mg/kg in front yards, 345 mg/kg in drip lines, 126 mg/kg in back yards and 135 mg/kg in vegetable gardens. This trend of increasing soil Pb concentrations towards building structures is consistent with previous studies. Mielke et al. (1983) found soil Pb concentrations greatest adjacent to buildings in inner city Baltimore, USA, attributing it to airborne leaded petrol emissions. Similarly, Schwarz et al. (2012) demonstrated a near exponential increase in soil Pb concentrations towards buildings in their study of 61 homes in Baltimore. These studies suggested two primary contributors to the high soil Pb concentrations found adjacent to buildings: 1) buildings provide a surface for airborne Pb particles to adhere to and ultimately deposit on to adjacent soil and 2) deteriorating Pb paint. The data from this study demonstrates a similar trend for soil Zn concentrations, and to a lesser extent Cu and As, suggesting these metals have been deposited in a similar manner around homes. Surprisingly, vegetable gardens had slightly higher Zn and Pb concentrations than back yard soils, despite typically being located furthest from houses in Australia. Industrial sources of Zn include traffic emissions, paint, and galvanized coated roofing, gutters and stormwater pipes. The presence of existing vertical structures (e.g. fences and trees) along the boundaries of homes can enhance the deposition of atmospherically sourced metals (Mielke et al., 1983; Schwarz et al., 2012). This may be more relevant for older homes where long-standing fences or trees have retained undisturbed soil for many years. Urban gardeners who grow vegetables in plots beneath the drip lines of their house, are likely to be

Table 2

Soil Mn, Cu, Zn, As and Pb concentrations (mg/kg) from front yard, drip line, back yard and vegetable garden areas. Values are displayed up to 3 significant figures.

	Mn	Cu	Zn	As	Pb		Mn	Cu	Zn	As	Pb
Front yard						Drip line					
Number detected	92	92	92	92	92	Number detected	97	97	97	91	97
Minimum	85	36	72	3	32	Minimum	83	32	50	2	15
10th percentile	154	43	113	6	47	10th percentile	155	46	154	6	58
25th percentile	209	51	174	7	69	25th percentile	183	56	242	9	163
Median (50%)	291	67	262	10	174	Median (50%)	266	77	472	15	345
75th percentile	403	90	439	24	542	75th percentile	340	120	739	26	752
90th percentile	482	121	793	31	1190	90th percentile	425	149	1080	41	1480
Maximum	1040	413	1240	76	2460	Maximum	743	233	2880	97	6490
Aus soil guideline ^a	3000	7000	8000	100	300	Aus soil guideline ^a	3000	7000	8000	100	300
% > guideline ^a	—	—	—	—	32%	% > guideline ^a	—	—	—	—	57%
Mean	317	80	349	16	413	Mean	282	90	574	21	707
Reference mean	301	46	142	6	31	Reference mean	397	54	178	10	35
Back yard						Vegetable garden					
Number detected	80	80	80	79	80	Number detected	141	141	141	140	141
Minimum	81	29	64	3	22	Minimum	91	33	50	3	14
10th percentile	151	42	110	5	36	10th percentile	174	48	129	5	34
25th percentile	203	49	152	6	48	25th percentile	224	59	196	7	60
Median (50%)	274	68	238	11	126	Median (50%)	304	73	284	10	135
75th percentile	369	98	418	17	339	75th percentile	383	96	469	18	334
90th percentile	452	129	777	24	705	90th percentile	480	132	736	32	745
Maximum	686	646	1790	55	1810	Maximum	813	717	2020	92	3080
Aus soil guideline ^a	3000	7000	8000	100	300	Aus soil guideline ^a	3000	7000	8000	100	300
% > guideline ^a	—	—	—	—	24%	% > guideline ^a	—	—	—	—	26%
Mean	299	89	367	14	266	Mean	319	88	387	15	301
Reference mean	297	41	101	7	29	Reference mean	412	55	163	7	36

^a Australian health investigation level for soils under category residential A – garden/accessible soil (home grown produce <10% fruit and vegetable intake (no poultry), also includes childcare centres, preschools and primary schools) (NEPM, 2013).

doing so in contaminated soils; 57% of drip line samples exceeded the Australian HIL-A guideline for soil Pb of 300 mg/kg.

3.3. Spatial distribution of Pb (city scale)

Spatial distribution of soil Pb data are displayed in Fig. 2, while soil Mn, Cu, Zn and As data are provided in Supplementary Figures 4–7 respectively. The greatest soil Pb concentrations were located to the west and south-west of the Sydney central business district in an area known as the Inner West (Fig. 2). This area is dominated by old medium-high density housing and high traffic roads, which has been contaminated by the widespread historic use of leaded petrol and paints (Markus and McBratney, 1996; Laidlaw and Taylor, 2011). Low soil Pb (<99 mg/kg) concentrations were found predominately towards the outer parts of the study area, demonstrating a trend of decreasing soil Pb concentrations away from the older, inner areas of Sydney (Fig. 2). However, low soil Pb concentrations were also observed throughout inner Sydney, most likely a result of clean soil introduction, particularly for the planting of vegetable gardens.

The majority of high soil Pb concentrations were located within three LGAs: City of Sydney, Leichhardt Municipal Council and Marrickville Council (Figs. 1 and 2). Mean soil Pb concentrations in these council areas were 883 mg/kg, 960 mg/kg and 689 mg/kg respectively. A significant proportion (74%) of homes within these LGAs contain soil Pb in excess of the Australian guideline; one third of these homes recorded >1000 mg/kg Pb in one or more garden samples. There are >150,000 houses within the boundaries of the City of Sydney, Leichhardt and Marrickville LGAs (ABS, 2011), and although this study examines 161 samples from 74 homes in these areas, the consistency of the results reveals a concerning pattern of environmental Pb contamination. Homes within these LGAs are some of the first built during the central Sydney expansion

between 1788 and 1917 (Kelly, 1987). As a result they have a higher proportion of Pb paint surfaces, which are known to correlate strongly with high soil Pb concentrations (Mielke, 1999; Schwarz et al., 2012).

3.4. House age, and contribution of leaded petrol and paint to soil Pb

Leaded paint and petrol are the two most dominant sources of Pb in the residential environment of Sydney over the last century (Markus and McBratney, 1996; Laidlaw and Taylor, 2011), with minor contributions from former and present industries. In Australia, Pb paint contained up to 50 wt% Pb during its early use and was incrementally reduced to 1.0 wt% and 0.1 wt% Pb by 1970 and 1997, respectively (Australian Government, 2014). Following the reduction of Pb in paint, houses were painted with titanium- or zinc-based paints. The median age of homes within the study area is 80 years, while the median age of reference homes is 30 years. The increase of soil Pb concentrations due to the age of house and prevalence of Pb paint have been documented previously (e.g. Sutton et al., 1995; Jacobs et al., 2002; Schwarz et al., 2012). There is an absence of research, however, examining the relationship between age of house, soil Pb concentrations and the presence or absence of exterior painted surfaces, especially in Australia.

Painted homes have greater median soil Pb concentrations (366 mg/kg) than non-painted homes (130 mg/kg). Consequently, painted homes have a higher proportion of soil exceeding the HIL-A guideline of 300 mg/kg (60% for painted versus 19% for non-painted). Soil Pb concentrations increase with age of house for both painted and non-painted homes, but at different rates (Fig. 3). Soil Pb concentrations at non-painted homes gradually increase at an average rate of 22 mg/kg per decade until median concentrations plateau at 226 mg/kg for homes built pre-1914. In contrast,

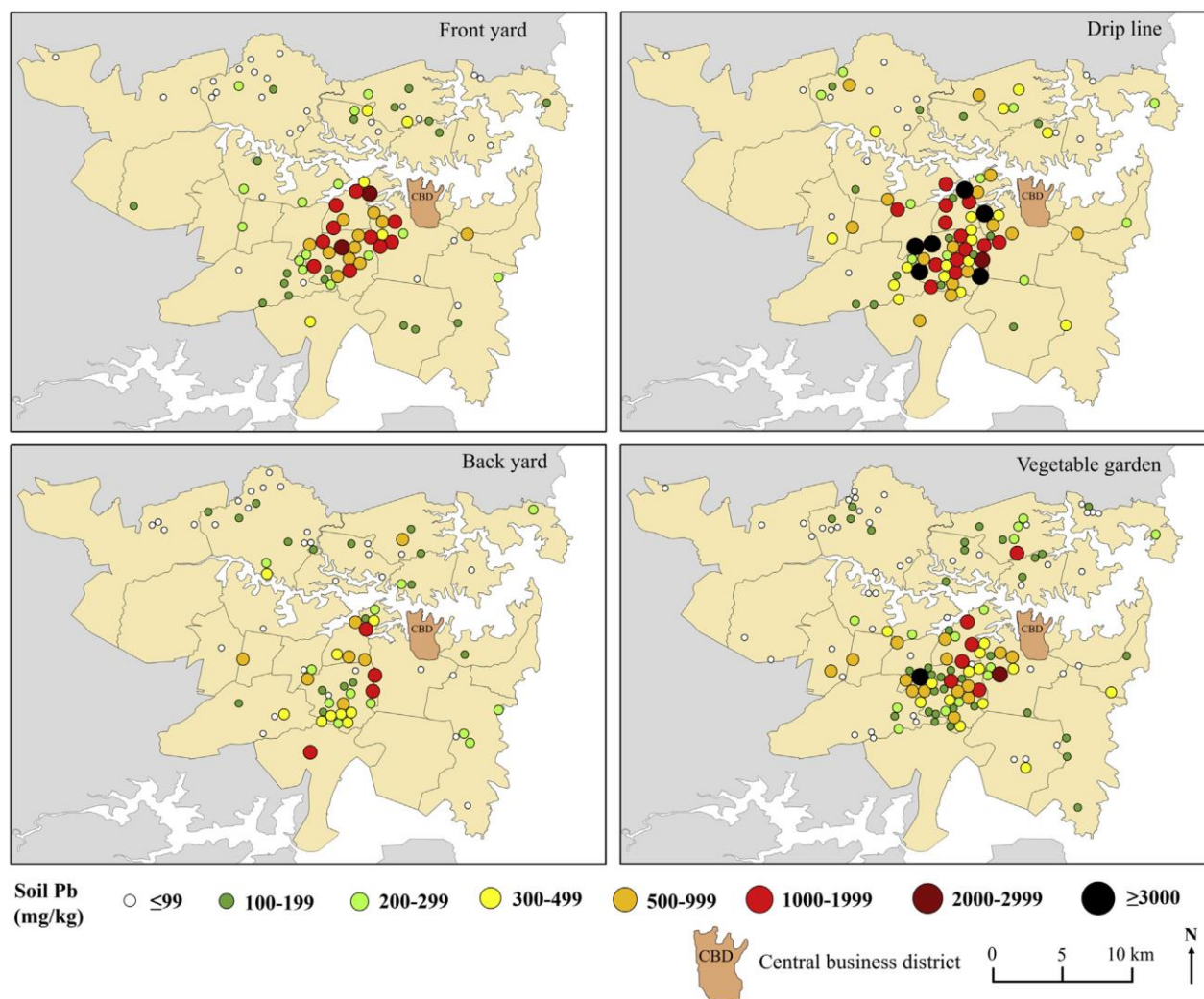


Fig. 2. Soil Pb concentrations of front yard ($n = 92$), drip line ($n = 97$), back yard ($n = 80$) and vegetable gardens ($n = 141$) areas in Sydney homes, Australia. Distributions of soil Mn, Cu, Zn and As data are provided in [Supplementary Figures 4-7](#), respectively.

median soil Pb concentrations at painted homes increase by an average of 76 mg/kg per decade from 45 mg/kg for post-1975 homes to 806 mg/kg for homes built pre-1895 (Fig. 3). Soil from older painted homes are more likely to exceed the 300 mg/kg soil guideline than younger painted homes; 85% of painted homes built between 1895 and 1914 had one or more soil samples in excess of this guideline compared to 43% of painted homes built between 1955 and 1975. The withdrawal of Pb from paints can be seen in Fig. 3. Comparable median soil Pb concentrations at painted (45 mg/kg) and non-painted (50 mg/kg) homes built after 1970 suggests the additional Pb loading from Pb paints has been eradicated in modern homes.

Source apportionment in environmental investigations is typically estimated using elemental isotopic ratios (e.g. [Gulson et al., 1995](#); [Kristensen et al., 2015](#)). However for this study, we estimate the contributions of three main Pb sources using metadata collected as part of the VegeSafe program. Source contributions to soil Pb are estimated using median soil Pb data from non-painted and painted homes applying the assumption that soil from non-painted homes is comprised of geogenic Pb (conservative

estimate of 30 mg/kg for the Sydney Basin, see [Wu et al., 2016](#)) and leaded petrol emissions. Soil from painted homes are assumed to have similar contributions with an additional input from Pb paint. The temporal shifts in Pb contribution from these three Pb sources for soils at painted homes over the last 120 years are estimated in Fig. 3. According to the data, the dominant source of soil Pb for older painted homes is Pb paint, most likely a function of its early use, combined with a longer time period for deterioration of painted surfaces and deposition into adjacent soil. However, improper removal of these paints by blasting, sanding and scraping can expedite generation of Pb rich dust particles that have been demonstrated to contaminate nearby homes ([Gulson et al., 1995](#); [Jacobs et al., 2003](#)). This could explain why a few non-painted homes had abnormally high soil Pb concentrations; for example a 70 year old double brick (unpainted exterior) Marrickville home had soil Pb up to 3220 mg/kg. Marrickville is a suburb of Sydney where 'do it yourself' renovations of old homes are increasingly common, with approximately 320 development applications for alterations and additions each year ([New South Wales Department of Environment and Planning, 2015](#)).

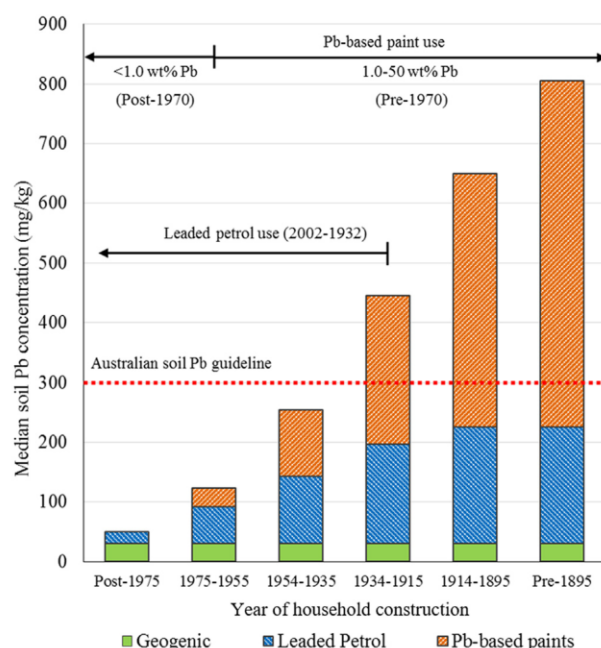


Fig. 3. Estimated Pb contributions from geogenic, leaded petrol emissions and Pb paint sources to median soil Pb concentrations of painted Sydney homes based on household construction era. The Australian health investigation level (HIL-A) for soil Pb is 300 mg/kg and is displayed as the red dashed line (NEPM, 2013). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The lowering of Pb concentration in both petrol emissions and Pb paint, and the eventual removal of both products is reflected by estimated decreased anthropogenic contributions to soil Pb at newer homes (Fig. 3). These findings are further supported by Wu et al. (2016), who showed that anthropogenic Pb isotopic signatures shifted to more geogenic signatures in lichens (used as environmental proxies) during the phasing out of leaded petrol in the Sydney basin.

There are estimated over 3.5 million homes in Australia built pre-1971 (Berry et al., 1994), the majority of which were likely coated with Pb paint at some stage. The impact of Pb paint is further demonstrated by the difference in median soil Pb concentrations between painted and non-painted homes across the four garden areas in Fig. 4. The distribution of soil Pb around homes (Fig. 4) is broadly consistent with cross sectional schematics presented in Olszowy et al. (1995) and Mielke (1999). However, this study demonstrates a marked shift of median soil Pb concentrations from non-painted to painted homes built pre-1970 (Fig. 4), supporting the estimated Pb loading from paints in Fig. 3.

3.5. Public health implications

Soil, including that used for edible produce, forms part of the exposure pathway for Pb absorption (Paustenbach et al., 1997; Hunt et al., 2006; Spliethoff et al., 2016). In Australia, exposure risks are assessed using the formal environmental health risk assessment system (Enhealth, 2012). In addressing the risk, strategies to eliminate hazardous exposures before they occur (i.e. primary prevention) could be supported with this and other relevant datasets (e.g. Birch et al., 2011; Laidlaw and Taylor, 2011; Gulson et al., 2014). Lanphear et al. (2005) and Lanphear (2015) identified two relevant salient facts about primary prevention: (1) the

effects of Pb toxicity has its greatest relative impacts at $<10 \mu\text{g/dL}$ of blood Pb and (2) the majority of population IQ points are lost from Pb exposure below the current United States and Australian acceptable maximum blood Pb level of $5 \mu\text{g/dL}$. The data show that soil from painted pre-1970 homes have higher Pb concentrations than non-painted pre-1970 homes, likely due to the additional loading from Pb paints (Figs. 3 and 4). Many pre-1970 Sydney homes still contain paint with up to 50 wt% Pb on exterior walls, fences, eaves, doors and window frames. As a result, these homes still have the potential to further contaminate the surrounding environment through the deterioration or improper removal of these paints. Over time, deterioration or deliberate removal of Pb paint increases remobilization into soils and household dusts.

This risk is evidenced by Australian blood Pb notification data which identify the primary causes of non-occupational Pb exposures are associated with Pb paints. For instance, in 2003, 78% of Queensland notifiable blood Pb cases ($>15 \mu\text{g/dL}$) were attributed to Pb paint. Childrens (0–4 years of age) blood Pb concentrations ranged $15\text{--}49 \mu\text{g/dL}$ (median $26 \mu\text{g/dL}$) (Queensland Health, 2003). Indeed, more than half (55%) of all non-occupational Pb exposures in Queensland between 2000 and 2011 were related or directly attributed to Pb paints (Queensland Health, 2011). The equivalent Pb exposure data for New South Wales (NSW) was obtained from NSW Health. The dataset could not be used because it contained incomplete entries on blood Pb exposures cases. Blood Pb assessment of Sydney residents primarily occurs as a result of individuals requesting a blood Pb test (unlike Australian mining and smelting communities where there is ongoing testing of children under 5 years of age – e.g. Taylor et al., 2011, 2014). As a result, any such data from cities is likely to represent only a fraction of actual Pb exposure cases in Australia (Taylor et al., 2012).

To demonstrate the importance of lead-safe paint removal, Jacobs et al. (2003) presented a case study from New Orleans, USA, where the improper, uncontained sanding of Pb paint from the exterior walls of a house led to the death of a Labrador retriever (blood Pb $177 \mu\text{g/dL}$), lead poisoning of three children who were immediately hospitalized and over US\$195,000 in Pb contamination cleanup costs. The professional painting contractors did not determine the Pb content of the painted walls prior to sanding and did not use lead-safe practices during the 6 week renovation (Jacobs et al., 2003). As a result of such cases, the United States Environmental Protection Agency (US EPA) introduced the 'Renovation, Repair and Painting' rule where contractors involved in the renovation, repair or painting projects, that disturb Pb paint from homes built pre-1978, must be certified and follow explicit work

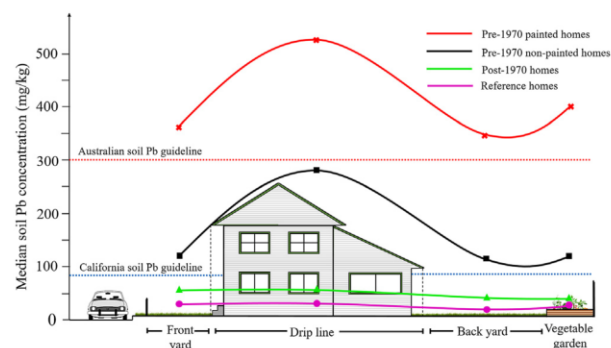


Fig. 4. Cross section schematic of a typical inner Sydney residential home with median soil Pb concentrations for painted pre-1970 homes, non-painted pre-1970 homes, post-1970 homes and reference homes. The vegetable garden is displayed at the rear of the back yard, as this was the case for the majority of homes.

practices to prevent further Pb contamination and exposure (HUD, 2016a). The rule also states that all contractors must provide Pb hazard information pamphlets (see US EPA, 2013) to all pre-1978 homes where children (0–6 years) live. In addition, the United States introduced the 'Lead Disclosure' rule in 1996 where the seller or lessor of a house built pre-1978 must disclose any known information regarding Pb paint or Pb hazards before the sale or lease of the property (HUD, 2016b). Non-compliance of either rule can result in fines issued by the US EPA (e.g. Harrington, 2012). These legislative instruments were implemented to prevent further Pb contamination and exposures associated with high-risk housing coated with Pb paint. Unfortunately in Australia, there are no legislations in place remotely similar to the 'Lead Disclosure' or 'Renovation, Repair and Painting' rules implemented in the United States. This is despite knowledge of the widespread use of up to 50 wt% Pb in paints until 1970 and the high proportion of Pb painted-related blood Pb cases in children (Queensland Health, 2011).

This study identified 40% of Sydney homes and >70% of inner city homes contain soil in excess of the Australian HIL-A guideline for Pb. However, amendments to relevant policies are unlikely to occur without further evidence of existing sources (e.g. Pb paint) directly causing numerous elevated blood Pb cases in urban communities. The true number of elevated blood Pb cases in Sydney and other major cities are likely to be much greater than the available data indicates. Applying Pb exposure rates (>5 µg/dL) in children in the United States, Taylor et al. (2012) estimated approximately 100,000 Australian children aged 0–4 years may have blood Pb concentrations in excess of the current national intervention guideline of 5 µg/dL. Such an estimation is certainly plausible given the high soil Pb concentrations found at Sydney homes. A systematic blood Pb survey of children living in high-risk areas, such as inner city suburbs with high soil Pb concentrations, would be a more accurate representation of current Pb exposures. The only national Australian blood Pb survey was in 1995 (Donovan, 1996). Available resources should be focused on informing the community on Pb paint hazards, as was recommended by Gulson et al. (2014).

3.6. How can residents become further informed about possible lead contamination at home?

As part of the VegeSafe program, each participant receives a formal report with their soil metal results, a table of relevant Australian soil metal guidelines to benchmark results and further information on the best ways to mitigate soil metal contamination (Supplementary Fig. 8). Typical advice varies between participants depending on the severity of soil Pb contamination in their yards, however some common guidance is given in most cases. For example, if Pb concentrations exceed the 300 mg/kg guideline in non-food growing soil, such as the front yard or drip line locations, VegeSafe recommends maintaining year-round coverage of lawn or mulch to minimize potential dust generation. However, if the 300 mg/kg guideline is exceeded in food growing soil, such as a vegetable garden, we recommend replacing the existing soil with new, uncontaminated soil or simply relocating the food growing to an above ground vegetable plot.

The information and advice VegeSafe provides to the community supports the NSW EPA fact sheets on Pb safety (NSW EPA, 2016b), which have been distributed to major hardware stores, childcare centers, and home and renovation shows across NSW. However, further outreach is required to distribute such information directly to residents living in areas with high soil Pb contamination. In this regard, it is recommended that local councils should become more involved in circulating such information. VegeSafe participants also have recommended the program to neighbours, family and friends, particularly those living in high-risk areas such

as the inner city. Such endorsements promote discussion of environmental health throughout communities, resulting in the public becoming better informed about how to mitigate sources of metal contamination in gardens. This assists the public in utilizing their urban space more safely, hence VegeSafe's motto 'carry on gardening'.

3.7. Limitations

While we are confident our sampling instructions were detailed and repeatable, we acknowledge our sample collection method could not guarantee consistent sampling between participants. The VegeSafe program collected soil from participating homes across Sydney and subsequently did not produce a systematic coverage of the entire city. Additionally, VegeSafe participants were not required to submit soil samples from every garden area, rather we suggested that participants sample from the following four locations: front yards, drip lines, back yards and vegetable gardens. We acknowledge that obtaining samples from every garden area per home would have improved the consistency of the sample collection method.

Typically, Pb source apportionment is carried out using Pb isotopic composition ratios (e.g. Chiaradia et al., 1997). However, we estimated source contributions to soil Pb loading using metadata provided by each VegeSafe participant. We acknowledge there are limitations of this approach, such as the generalization of painted houses to contain Pb paint if constructed prior to 1970. However, the overall results from this study correspond to the timing of the withdrawal of Pb from paint and petrol providing support for, and confidence in our conclusions.

4. Conclusions and recommendations

This study found soil Pb concentrations in excess of the Australian health guideline of 300 mg/kg at 40% of Sydney homes. Fifteen percent of sampled homes contain soil Pb >1000 mg/kg. Soil metal concentrations were typically greatest at drip line locations around homes in the inner city, with concentrations decreasing with distance away from the city center. Median soil Pb concentrations at painted homes were significantly greater than non-painted homes of a similar age (Fig. 3), demonstrating a significant contribution to soil Pb loading from Pb paint. The removal of leaded petrol and the reduction in Pb concentration in exterior paints (to <0.1 wt%) are also reflected within our dataset, demonstrating the efficacy of eliminating such products from use. Unfortunately, Pb paint is still present on countless homes in Australia and has the potential to further contaminate the home environment through deterioration and its improper removal.

The VegeSafe program has provided >5200 free soil metal tests to Australian residents and subsequently has re-focused the community's attitude towards environmental contamination, largely because their properties have been directly impacted. Our data demonstrates Pb paint is a major source of Pb contamination around Sydney homes, particularly in the inner city. Young families are increasingly moving into older homes that often require renovation, and frequently are not informed on Pb paint hazards. We recommend further effort is required by local councils and regulatory bodies to adequately inform families living in Pb prone areas of the dangers of soil contamination and Pb paint hazards in the home environment. Additionally, this study demonstrated the effectiveness of crowd sourced sample collection of private residential soils through a community science program and should be replicated in other major cities around the world. Adaptation of this citizen science approach could also be applied to other environmental investigations of traditional and emerging contaminants.

Acknowledgements

We thank Olympus Australia and the National Measurement Institute for contributing funding and practical support towards this research. VegeSafe gratefully acknowledges the donations it has received from members of the public that help to sustain the program. For technical support, discussions, sample collection and analysis we thank Sam Habib and Andrew Taylor (Olympus Australia), Andrew Evans and Shiva Prasad (National Measurement Institute), Danielle Camenzuli, Peter Davies, and Damian Gore (Macquarie University) and Paul Rouillon. Marek Rouillon is funded by Australian Postgraduate Award. Paul Harvey is funded by a Macquarie University Research Excellence Scholarship (MQRES) (2012195) associated with an Australian Research Council Future Fellowship awarded to Heather Handley (FT120100440).

Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.envpol.2016.11.024>.

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3

Contaminants in Agriculture

The agricultural environment is often not thought of as a point of exposure to environmental contaminants. The typically higher tolerance thresholds for livestock and lack of physical expression of disease from environmental exposures results in challenges for environmental managers to identify problems in these settings. This chapter examines recent examples of where environmental contaminants on the agricultural scale have caused considerable degradation of the environment, and in some instances, generated significant livestock and human health risks. This chapter serves to highlight some of the challenges around confidently identifying polluters in the agricultural setting and also the complexities of monitoring and regulating environmental contaminants, on what is often, a very large spatial scale.

Widespread Environmental Contamination Hazards in Agricultural Soils in Above Ground Large-Scale Water Supply Pipelines.

P.J. Harvey (85%), M.P. Taylor (10%), H.K. Handley (5%)

Water, Air & Soil Pollution 2015, 226 (6), 1-9.

Research relating to agriculture is usually focused on organic contaminants. This novel study, which marks a transition in this thesis from the home environment to the agricultural environment, stemmed from concerns raised by landowners whose cattle were unexplainably dying on their property. Investigations of the cattle showed substantial concentrations of blood lead. It became apparent that work to reseal the lead weld joints of the Chichester Trunk Gravity Main, which passes through hundreds of properties, could be the cause. This study demonstrated that the pipeline was a significant source of environmental lead contamination in the soils surrounding the pipeline where cattle grazed. Investigation of other similar pipelines in New South Wales and Western Australia shows that these pipelines are common and that their lead weld joints are a significant source of environmental lead contamination. The results of this study contributed to the decision by Hunter Water Corporation, the utility that owns the pipeline and related infrastructure corridor, to fence along the corridor to restrict livestock access to the contaminated soils.

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Harvey, P.J., Taylor, M.P. & Handley, H.K. (2015) Widespread Environmental Contamination Hazards in Agricultural Soils from the Use of Lead Joints in Above Ground Large-Scale Water Supply Pipelines. *Water, Air, & Soil Pollution*, vol. 226, Article No. 178.

<https://doi.org/10.1007/s11270-015-2397-3>

Tracking Chicken Slaughterhouse Waste in a Peri-urban Waterway Using Chemical, Biological, and DNA Markers

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Environmental Research, In Review.

Continuing with the theme of contaminants in agricultural environments, this study examines the pollution of a peri-urban catchment as a result of discharge waters from a chicken slaughterhouse. The study focuses on the method applied to constrain the potential source(s) of pollution in this catchment. Critically, the wording ‘pollution’ is used in this study to reflect that of the Protection of the Environment Operations Act 1997 (POEO Act). The method utilised a number of unique techniques including arsenic speciation and analysis of DNA markers, in addition to more conventional techniques such as faecal coliform counts. Through this weight of evidence approach to apportioning a source, it became apparent that the chicken slaughterhouse was the primary contributor to the effluent pollution in the catchment, resulting from illegal discharging of waste water. This project originated with M.P. Taylor and has been contributed to by a number of students but the bulk of the work contributing to this manuscript was conducted by P.J. Harvey. In addition, research collaborations were sought by Harvey and Taylor with S. Foster and M.R. Gillings in order to facilitate the more unique aspects of the laboratory analysis. This work was presented orally at the American Geophysical Union (AGU) Fall Meeting in San Francisco in December 2016.

Chemical, biological, and DNA markers for tracing slaughterhouse effluent

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Keywords: arsenic; water quality; urban catchment; mitochondrial DNA; integron-integrase gene; environmental regulation

Abstract

Agricultural practices, if not managed correctly, can have a negative impact on receiving environments via waste disposal and discharge. In this study, a chicken slaughter facility on the rural outskirts of Sydney, Australia, has been identified as a possible source of persistent effluent discharge into a peri-urban catchment. Questions surrounding the facility's environmental management practices go back more than four decades. Despite there having never been a definitive determination of the facility's impact on local stream water quality, the New South Wales Environment Protection Authority (NSW EPA) has implemented pollution requirements to manage noise and water pollution at the facility. However, assessment of compliance remains complicated by potential additional sources of pollution in the catchment. To unravel this long-standing conundrum related to water pollution we apply a forensic, multiple lines of evidence approach to delineate the origin of likely pollution source(s). Water samples collected between 2014-2016 from a watercourse exiting the facility had concentrations of faecal coliforms (max: >2,419 forming units/ 100 mL), ammonia (max: 63,000 $\mu\text{g/L}$), nitrogen (max: 67,000 $\mu\text{g/L}$) and phosphorous (max: 39,000 $\mu\text{g/L}$) significantly higher than samples from adjacent streams that did not receive direct runoff from the facility. Arsenic, utilised as a growth promoting compound, was detected in water discharging from the facility up to ~ 4 times (max 3.84 $\mu\text{g/L}$) local background values (<0.5 $\mu\text{g/L}$), with inorganic $\text{As}^{(\Sigma\text{V}+\text{III})}$ dominating. The spatial association of elevated water pollution to the facility could not demonstrate causation and consequently DNA analysis of a suspected pollution discharge event was undertaken. Analysis of catchment runoff from several local streams showed that only water sampled at the downstream boundary of the facility tested positive for chicken DNA and not duck DNA, which was a potential confounder given that wild ducks were present in the area. Further, PCR analysis showed that only the discharge water emanating from the slaughter facility tested positive for a generalized marker of anthropogenic pollution, the clinical class 1 integron-integrase gene. The environmental data collected over a three-year period

demonstrates that the facility is indisputably the primary source of water-borne pollution in the catchment. Moreover, application of DNA and PCR for confirming pollution sources creates potential for application by regulators in fingerprinting pollution and polluters as presently none of the Australian EPA's use these powerful techniques.

Introduction

When agriculture and industry are situated alongside residential land, conflicts arise from the different needs and expectations of land users (Fowler and Shi, 2016, James and O'Neill, 2016, Pribadi and Pauleit, 2016, Wei et al., 2016). The New South Wales Environment Protection Authority (NSW EPA) is an independent statutory authority whose role, inter alia, is to 'work with the community, business, industry and government to maintain a balance between protecting the environment, managing competing demands on the environment and supporting sustainable growth' (NSW EPA, 2016a). Community feedback about environmental problems, often through the NSW EPA's Environment Line, is used to inform it in executing compliance with respect to environmental protection licence requirements (NSW EPA, 2013; 2015a).

One of the key roles of the NSW EPA, as set out by the *Protection of the Environment Operations Act 1997* (NSW) (POEO Act), is to manage and regulate pollution via environment protection licences (NSW EPA, 2016a). Licences set conditions that aim to achieve 'pollution prevention and monitoring, and cleaner production through recycling and reuse and the implementation of best practice'. Licences are applied to specific activities associated with a premise as well non-premise-based activities and scheduled development works (Schedule 1, POEO Act). Activities that are not listed in Schedule 1, POEO Act, may also be subject to licencing to regulate pollution from activities. Environmental protection licences can protect the polluter from prosecution (e.g. s 120 of the POEO Act (water pollution)) where activities are compliant with licence conditions (NSW EPA, 2014).

In challenging polluters and identifying sources, the primary hurdle is the demonstration of a causal link. There is considerable difficulty in obtaining unbiased and definitive evidence, particularly when major changes to the polluter's operations are required to bring it in line with licence arrangements or public expectations. This hurdle can be overcome in two main ways: (a) using the weight of evidence derived from a suite of biological, chemical and physical environmental measures and their spatial-temporal associations with respect to a possible

polluter; (b) the production of evidentiary material to show an unequivocal causal link between indices of pollution and known activities at a site.

In addressing the causation challenge, this current study sets out a forensic, multiple lines of evidence approach to identify the source of environmental pollution from a livestock (chicken) slaughtering and processing activity (the facility) in Sydney, New South Wales, Australia. The facility and its operations are licenced under a NSW EPA environment protection licence (NSW EPA, 2016b). The facility adjoins semi-rural residential allotments (~ 5 hectares) and light agricultural activities. The facility is situated at the top of a small headwater catchment with runoff passing through a number of adjoining properties before entering an ecologically sensitive National Park (Figure 1). The NSW EPA licence permits irrigation of the facility's effluent water at a set number of adjoining allotments (Figure 1) but at a rate such that the irrigation does not result in off-site discharge.

Animal husbandry and slaughter facilities are frequently the subject of environmental investigation by regulatory authorities, with claims of pollution often vigorously rebutted (e.g. Bienkowski 2015 and Heaney et al., 2015). The environmental impact and management practices of the facility examined here has been the subject of debate including controversial discussion in the NSW Parliament and local media (e.g. Cordina, 2000; Gallacher, 2000; Hornsby Advocate, 1998, 2000a,b; Howard, 2000; Inshaw, 2000, 2001; Rhiannon, 2000, Ward, 2000a,b). The facility has received numerous non-compliance notifications over the period of its operating licence (NSW EPA, 2015).

Poultry processing facilities produce significant volumes of contaminated waste water that can present a risk of harm to local environments (Gan and Hu, 2016). In assessing the efficacy of environmental management practices it is imperative that regulatory authorities are equipped with precise and effective tools to ensure (a) maximum environmental protection is achieved; and (b) that licence arrangements are adhered to. Typically, regulatory authorities rely on adhoc sampling of soils and waters using routine analyses e.g. total concentrations of target pollutants.

While these are useful, they are not always definitive in delineating either the source or cause of pollution. As a result, these relatively 'blunt' analytical tools can limit regulatory authorities from exercising their full power where they are unable to confidently fingerprint the pollution to a particular facility. In this regard, this study details a forensic approach and assessment of water-borne contaminants from multiple waterways that either drain or are adjacent to a chicken slaughter facility to delineate the source, cause and extent of pollution.

In completing this forensic assessment of water pollution a range of geochemical and biological techniques were applied to identify the source(s) of the environmental pollution. One of the more unique techniques considers the clinical class 1 integron-integrase gene, a known indicator of anthropogenic pollution (Gillings et al. 2015). The clinical class 1 integron-integrase gene is often associated with antibiotic resistance genes and has been proposed previously as a marker of pollution from human activities (Gillings et al. 2015). This study examines the utility of using the clinical class 1 integron-integrase gene as a novel environmental pollution tracer to supplement more standard biological and chemical measures of water quality and its sources.

Methods and Approach

The setting for this study is complex as there is a number of separated drainage pathways and an above-ground pipe network that conveys discharge from the facility to domestic lots. The facility is licenced to irrigate adjoining non-domestic lots in an effluent utilisation zone (Figure 1). A targeted sampling strategy has been used to capture any run-off from the facility, understand the quality of water piped to domestic lots and the effluent utilisation zone, as well as to capture any flows in adjacent drainage pathways not receiving run-off from the facility (Figure 1).

Sampling

This study was conducted over a three year period and incorporated multiple sampling times and locations. Sampling was opportunistic and all samples were collected only from flowing (not standing) water at the time of collection. Preliminary *in situ* water quality sampling and assessment for total nitrates and phosphates was undertaken in 2013, which indicated anomalous values associated with discharges relating to the facility. Subsequent field observations of unusual discharges (i.e. when there was no prevailing rainfall) were followed up with sampling of watercourses draining and, adjacent to, but not directly impacted by the facility on the 7th and 10th October 2014 (Figure 1). The water samples were analysed for total ammonia, nitrogen, phosphorous and faecal coliforms. Following presentation of this data to the NSW EPA, a pollution reduction program was added to the facility's licence conditions (3rd February 2015) to investigate ways of improving environmental performance from its effluent management and irrigation practices.

Following the addition of the pollution reduction program additional samples were collected from watercourses draining and abutting the facility on 22nd July 2015 (Figure 1). Samples were also collected from an irrigation pipeline on a private property adjoining the facility (Figure 1, samples F and G) to assess the quality of water being discharged via spray irrigation from the facility. Total ammonia, nitrogen, phosphorous and faecal coliforms were analysed in these samples. Samples were also collected on 29th July 2016 and analysed for total ammonia, nitrogen and phosphorous to assess the nature on possible ongoing effluent discharges to the local waterway.

In all instances, samples were collected in sterilised HDPE or polystyrene sample containers (depending on laboratory requirements). Samples for phosphorous were preserved at pH 2 with analytical grade HNO₃ (MERCK), and those for ammonia and nitrogen with analytical grade H₂SO₄ (MERCK). All samples were stored at < 4 °C prior to analysis.

Tests for ammonia, nitrogen, phosphorous and faecal coliforms were conducted at commercial NATA (National Association of Testing Authorities) laboratories in Sydney, NSW. Total

phosphorous was determined by solution Inductively Coupled Plasma Mass Spectrometry (ICP-MS, Agilent, 7900). Analysis of phosphorous blank samples returned <Limit of Reporting (LOR, 50 µg/L), analysis duplicates had a relative percent difference (RPD) of 0% and concentration recovery based on 10 ppb matrix spike solutions of 96%. Field duplicates had <2% RPD and field blank <LOR. Nitrogen was analysed using a potassium persulphate (Sigma-Aldrich, ReagentPlus 99% purity, K₂S₂O₈) digestion method and Lachat Flow Injection System (QuickChem 8000). Field duplicate samples had <2% RPD and field blank <LOR. Nitrogen analysis blank samples returned <LOR (50 µg/L), analysis duplicates had an RPD of 0% concentration recovery based on 10 ppb matrix spike solutions of 108%. Ammonia was analysed using a hypochlorite and alkaline phenol reaction with sodium nitroprusside persulphate (Sigma-Aldrich, ReagentPlus 99% purity, Na₂[Fe(CN)₅NO]·2H₂O) and an AquaKem 250 discrete analyser. Field duplicate samples had <1% RPD and field blank <LOR (5 µg/L). Ammonia analysis blank samples returned <LOR µg/L, had a duplicate RPD of 8.7% and concentration recovery based on 10 ppb matrix spike solutions of 87%. Faecal (thermotolerant) coliforms were analysed using the Colilert-18 (IDEXX Laboratories 2015) method. Field duplicate samples had <1% RPD and field blank <LOR (1 cfu/100 mL (cfu/100 mL)). Analysis was performed with both positive (spiked with 100 viable cells of the target *Klebsiella pneumoniae*) and negative (spiked with 100 viable cells of the non-target *Pseudomonas aeruginosa*) culture control spiking. Negative control spike cultures act as analysis blanks. All control spike cultures returned <10% RPD. Analysis duplicates are not performed in this method, but field duplicate analysis demonstrated the reproducibility of the data.

Arsenic analysis

Arsenic is commonly used as an additive in chicken feed. The most common compound, Roxarsone (3-nitro-4-hydroxyphenylarsonic acid, HOC₆H₃(NO₂)AsO₃H₂) is commonly detected in environmental discharges from poultry facilities (Anderson and Chamblee, 2001,

Bednar et al., 2003, Brown et al., 2005, Cortinas et al., 2006, Fisher et al., 2015, Garbarino et al., 2003, Jackson et al., 2003, Jackson et al., 2006). While this compound has been banned in many countries (Hileman, 2007 and Yao et al., 2016), it is still available *for* use in Australia and may contribute to elevated arsenic levels in soils water surrounding poultry facilities. To this end, samples collected in 2015 were also analysed for total arsenic and their speciated forms using High Performance Liquid Chromatography Inductively Coupled Mass Spectrometry (HPLC-ICP-MS). Two arsenic-related sampling events took place, one on the 22nd July 2015 and a second on the 30th July 2015, to capture temporal variations in run-off arsenic concentrations.

Water samples for arsenic analysis (n = 8, Sites: A–C, E, G–I, Figure 2) were collected in HDPE bottles and preserved by acidifying to pH 2 using analytical grade HNO₃ and stored at < 4 °C. Two sample duplicates were collected. Samples were analysed for total arsenic and arsenic species at the University of Canberra Ecochemistry Laboratory. Samples were filtered using a 0.45 µm syringe filter and preserved using analytical grade HNO₃. Speciation was achieved using the method for detection of Roxarsone in environmental samples set out by Nachman et al. (2013). Inorganic As(III) was oxidised to As(V) using 10 µL hydrogen peroxide (Sigma-Aldrich) solution. Samples were injected onto a Hamilton PRPX-100 anion exchange column (10 µm, 250 x 4.1 mm) fitted to a Perkin Elmer Flexar 10 HPLC. A gradient mobile phase (5.6 pH adjusted with ammonium hydroxide, Sigma-Aldrich) method was used requiring 4 min of 20 mM malonic acid (Sigma-Aldrich, ReagentPlus 99% purity) followed by 4 - 8 min of 100 mM malonic acid and finally 8 - 10 min of 20 mM malonic acid. A flow rate of 1.0 mL min⁻¹, column temperature of 30 °C and injection volume of 20 µL was used. Arsenic was detected on a Perkin Elmer NexION 300D ICP-MS monitoring *m/z* 75 and ArCl interference masses. Field duplicate samples had <10% RPD and field blank <0.05 µg/L. Laboratory analysis blank samples returned <0.16 mg/L. Standard curves were developed for each of the species measured and their identity determined through retention time-matching and spiking with authentic

standards. Spiked recoveries of Roxasone at 10 ppb was performed on one sample in triplicate ($10.4 \pm 1.5 \mu\text{g L}^{-1}$ As ($104 \pm 14 \%$ recovery)).

DNA extractions

In order to establish causal links between contaminants in the effluent discharge water, analysis was conducted to detect any chicken DNA in the catchment runoff associated with the facility, as well as that in adjoining, but separate streams. Water samples collected on 29th July 2016 were analysed for the presence of chicken and duck DNA in the Department of Biological Sciences, Macquarie University. Waters were tested for the presence of duck DNA because they are common in the local area and could be a potential confounder. Duplicate 100 ml water samples were collected into sterile containers from sites C, E and H (Figure 1). Samples were transported on ice to the laboratory where they were frozen prior to analysis to prevent degradation of genetic material. Two 50 ml subsamples were taken from each of the six samples and centrifuged at 4°C for 10 min at 4,000 rpm. The pelleted material was resuspended in 780 μL of 100 mM NaPO_4 buffer (pH 8.0), transferred to a FastPrepTM lysing tube E (MP Biomedicals) and processed by bead beating using MT buffer as previously described (Gillings, 2014b). Reference DNA was prepared from retail chicken and duck meat using a proteinase K/salting out method (Sunnucks and Hales, 1996). DNA extractions and PCR assays were analysed by electrophoresis on 2% agarose gels, poured and run in TBE buffer (Sambrook et al., 1989). DNA was visualized by post-staining with GelRedTM (Biotium) and UV transillumination.

Polymerase chain reaction (PCR) assays

To test for the presence of chicken tissue in the water samples, an assay specific for chicken mitochondrial DNA was performed. Mitochondrial DNA primers chicken-f (5' GGGACACCCTCCCCCTTAATGACA) and chicken-r (5' GGAGGGCTGGAAGAAGGAGTG) (Lahiff et al., 2001) were used in a 50 μL reaction using

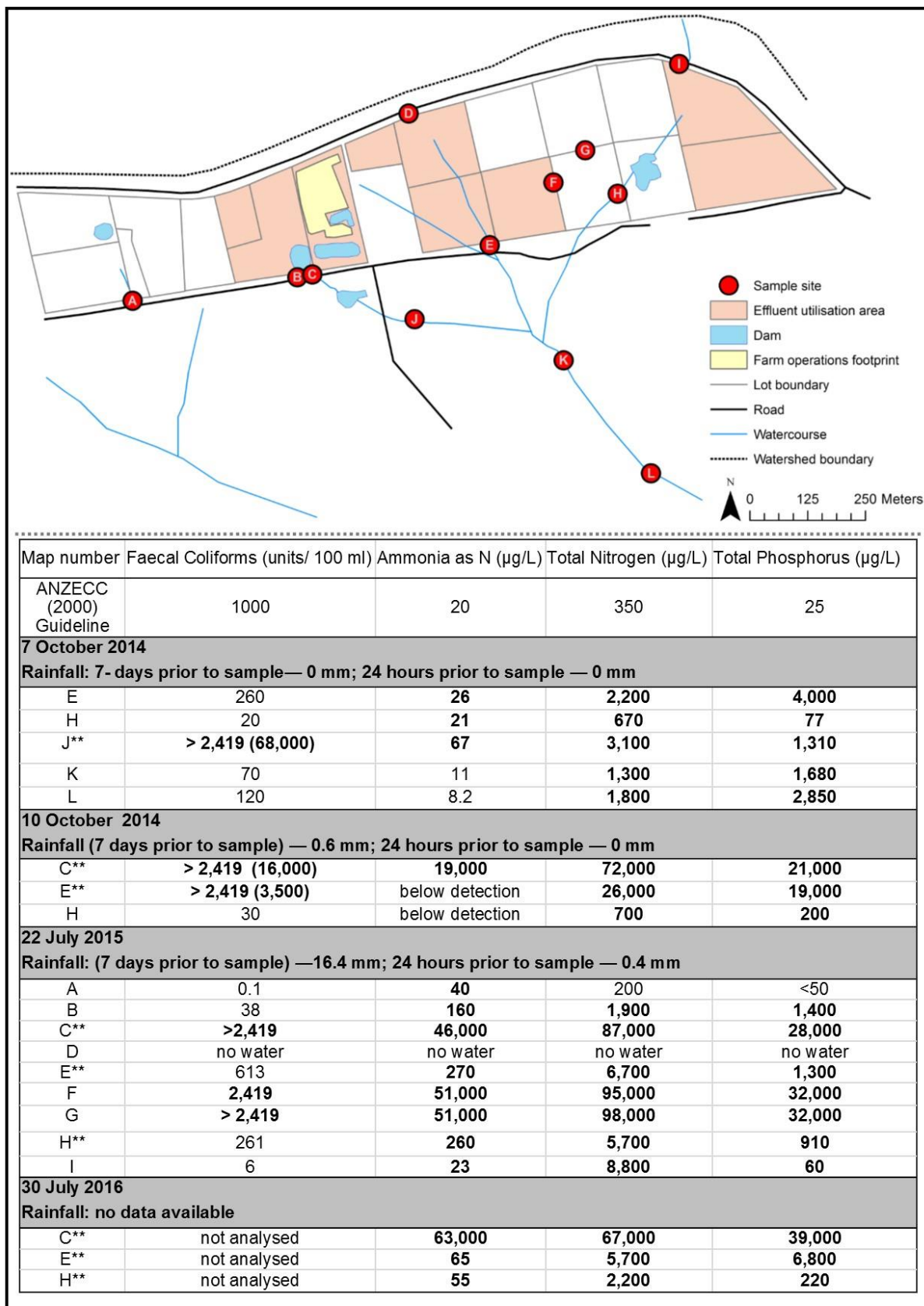
GoTaq, as per the manufacturer's instructions (Promega), at an annealing temperature of 66 °C. Chicken DNA was used as a positive control, duck DNA and water only negative controls were also included in every reaction set. Amplicons were purified using ExoSapTM (Thermo Fisher Scientific) as per the manufacturer's instructions, and DNA sequenced using the amplification primers supplied by Macrogen, Korea. Sequences were analysed using the BLASTn suite on the NCBI website (NCBI 2016). The PCR competency of the extracted DNA was confirmed by successful amplification of the bacterial 16S gene.

Samples were also tested for a known marker of anthropogenic pollution. A PCR was performed to detect the clinical variant of the integron-integrase gene, *intI 1*. This gene is found in antibiotic resistant bacteria that are common inhabitants of the digestive tracts of humans and their domestic animals, and has been proposed as a proxy for human impact on a variety of environmental systems including water (Gillings et al., 2015). Specific primers intI1F165 (5'CGAACGAGTGGCGGAGGGTG) and intI1R476 (5' TACCCGAGAGCTTGGCACCCA) were used in 50 µL PCRs as above, but with an annealing temperature of 60 °C.

Results and Discussion

Given that samples were only collected from flowing water to demark the quality of runoff, some sites are absent of data at particular times due to lack of flow (Figure 1)

Samples ($n = 4$) collected on 7th October 2014 reflect the baseline flow conditions through the upper catchment watercourses (mean values: faecal coliforms 118 cfu/100 mL; ammonia 17 $\mu\text{g/L}$; nitrogen 3,100 $\mu\text{g/L}$; phosphorus 1,310 $\mu\text{g/L}$; Figure 1). Site J, which is downstream of a small dam on an adjoining property that receives runoff directly from the facility, exhibited higher values for most measures (faecal coliforms $> 2,419$ cfu/100 mL; ammonia 67 $\mu\text{g/L}$; nitrogen 1,493 $\mu\text{g/L}$; phosphorus 2,059 $\mu\text{g/L}$).



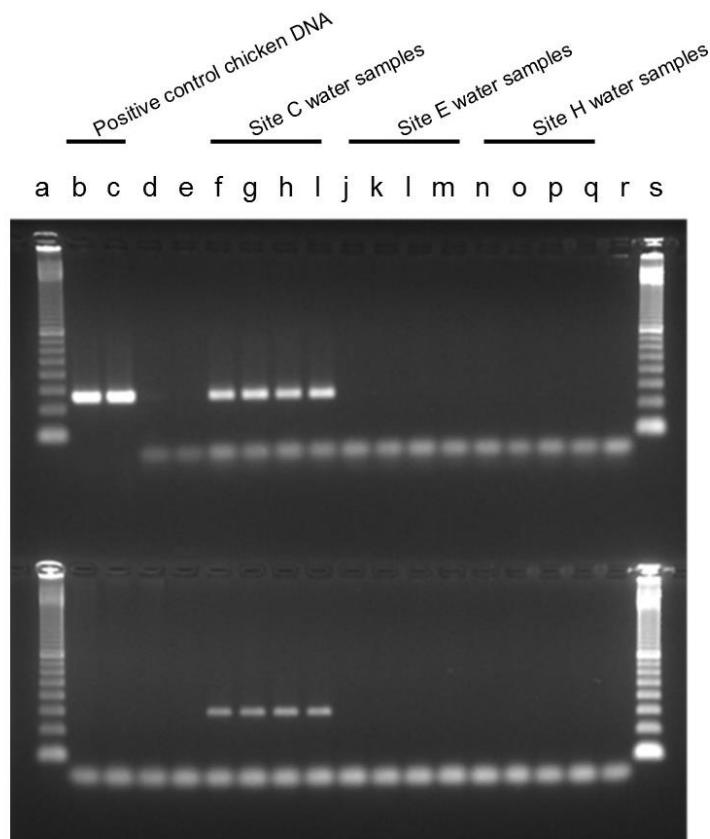


Figure 3 Detection of DNA originating from chickens in urban water samples. PCR was used to directly detect chicken DNA (upper panel) (Lahiff et al., 2001), and to detect a DNA proxy for anthropogenic pollution, the clinical class 1 integron-integrase gene (lower panel) (Gillings et al., 2015). Samples are as follows: (a),(s) 100 base pair size standard; (b-c) positive control chicken DNA; (d-e) negative control duck DNA; (f-i) water samples site C; (j-m) water samples site E; (n-q) water samples, site H, (r) distilled water control.

Samples collected on 10th October 2014 were taken when discharge from the facility was thought to be occurring. Water sampled at Site C, immediately downstream of one of the facility's waste water dams and property boundary, contained grossly elevated concentrations of all contaminants (faecal coliforms >2,419 cfu/100 mL; ammonia 19,000 µg/L; nitrogen 72,000 µg/L; phosphorus 21,000 µg/L; Figure 1). Similarly, Site E, which drains an adjoining paddock within the effluent utilisation area contained elevated concentrations of contaminants (faecal coliforms >2,419 cfu/100 mL; ammonia <5 µg/L; nitrogen 26,000 µg/L; phosphorus

19,000 µg/L; Figure 1). It is relevant to note that contrary to the topographic map, the channel confluence is immediately upstream of the southern property boundary at this site, not downstream of the road as depicted (Figure 1). Therefore, site E also receives water from lot A, which is outside the effluent utilisation area. Relevantly, there was no precipitation on the day of sampling and only 0.6 mm was recorded during the preceding 7 days at the nearest weather station, approximately 2.6 km away, thus eliminating other 'natural' sources of catchment run-off.

Sampling on 22nd July 2015 took place on a day the facility appeared to be discharging. The data contained concentrations of contaminants that varied according to the location of sampling (Figure 1). No precipitation was recorded on that day at the nearest weather station with the preceding 24 hours returning 0.4 mm of rainfall, and 16.4 mm in the 7 days prior to sampling (BoM 2016). Site D, at the top of the catchment and upstream of the facility, could not be sampled due the absence of any flow. Sites C, the watercourse discharging the facility and sites F and G, samples collected from irrigation pipes provided water to adjacent domestic properties, contained elevated concentrations of faecal coliforms (> 2,419, 2,419 and >2419 cfu/100 mL, respectively); ammonia (46,000 µg/L, 51,000 µg/L and 51,000 µg/L, respectively); phosphorous (28,000 µg/L, 32,000 µg/L and 32,000 µg/L respectively). Sites F and G were collected from irrigation pipeline outflow points on a property outside of the effluent utilisation area. This water is provided to residents for domestic use on their gardens. Samples collected distal to the facility, on neighbouring, but not adjoining watercourses, contained markedly lower concentrations of all contaminants (Figure 1, sites A, E and L). Site E contained slightly elevated faecal coliforms (613 cfu/100 mL).

Water samples collected from sites (C and E) draining the facility throughout the study were predominantly elevated well above the ANZECC (2000) guidelines for lowland rivers (Figure 1). Site C, which directly drains the facility, consistently exceeded the ANZECC (2000) guideline values with markedly higher concentrations than adjoining sampled sites (Figure 1). The presence of faecal coliforms (regularly >2419 cfu/100 ml) in waters draining the facility,

compared values in adjacent but not directly connected watercourses, demonstrates a biological contribution to the discharge. By contrast, neighbouring streams have substantially lower concentrations of analytes (Figure 1).

The environmental water pollution markers (faecal coliforms, ammonia, nitrogen and phosphorous) used in this study indicate that the facility was a likely source of environmental pollution. The elevated levels of water contaminants can be contrasted to lower and non-detectable concentrations upstream from, or adjacent to, the facility (Figure 1).

Arsenic as a Source Apportionment Tool

Arsenic was detected in all samples. Samples collected from sites B, C and G (all connected to discharge waters from the facility) had higher total arsenic concentrations than all other samples (2.83 µg/L, 3.70 µg/L and 2.05 µg/L respectively; Figure 2). Sample sites B and C, were collected from drainage channels exiting the facility (Figure 2). Water from Site G was sampled from an irrigation pipeline on a residential property that receives facility wastewater for use on the property lot. Based on faecal coliforms, ammonia, nitrogen and phosphorous concentrations, this water appears almost identical to that analysed at site C.

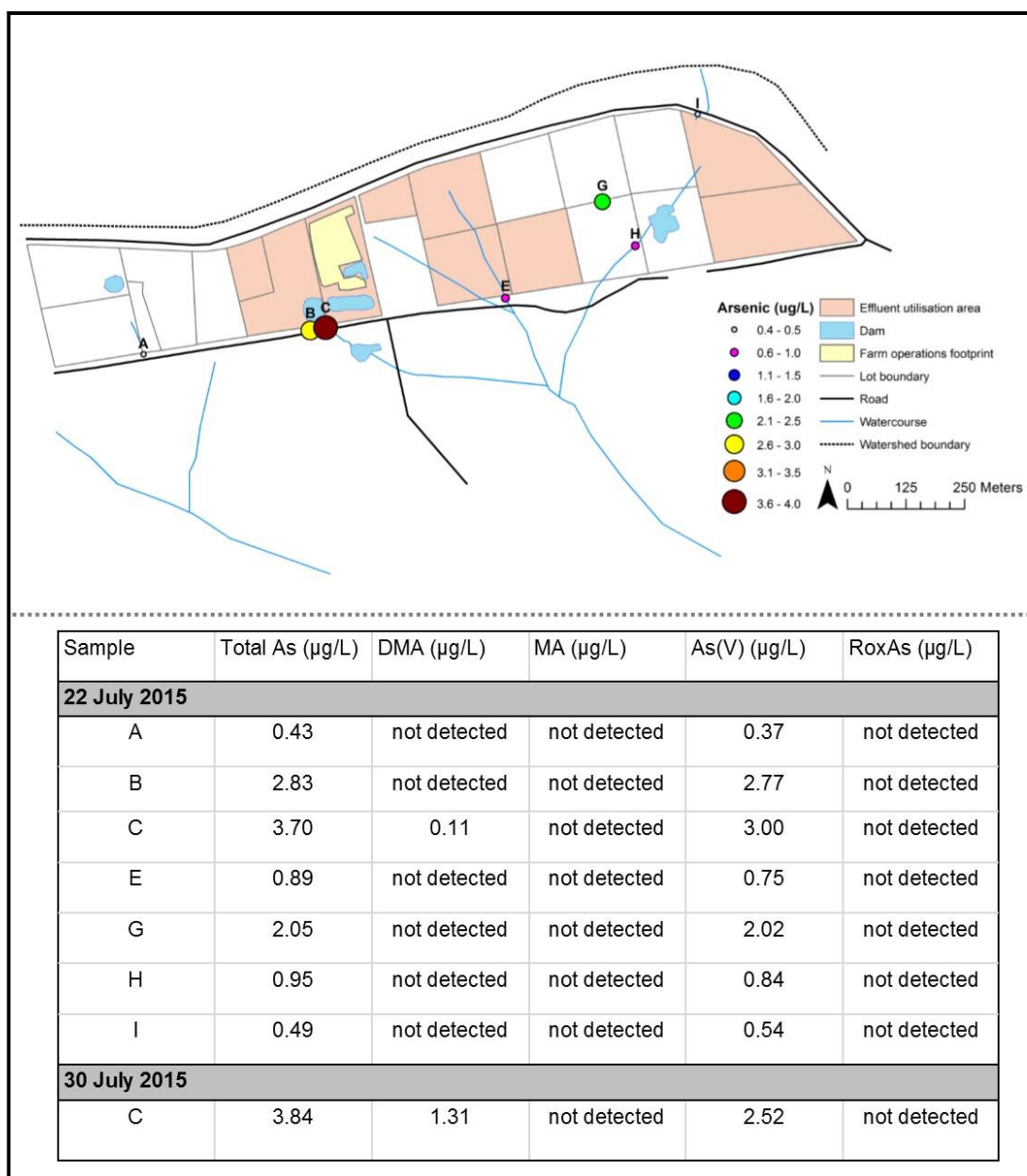


Figure 2. Arsenic concentrations in the water samples collected the study area. Total As in water samples was analysed using ICP-MS as outlined in Maher et al. (2003). Detection limits for total As are calculated at 0.16 µg/L and for HPLC-ICPMS detection limits are calculated at 0.11 µg/L for DMA, 0.25 µg/L for MA, 0.15 µg/L for As(V) and 0.25 µg/L for roxarsone.

Arsenic speciation revealed that the dominant form in all waters was inorganic arsenic $\text{As}^{(\Sigma\text{V}+\text{III})}$ reported as As(V) (Figure 2). Roxarsone was not detected in the samples. Roxarsone is a rapidly degrading compound that when exposed to heat, oxygen and UV light degrades to

inorganic forms (Garbarino et al., 2003, Bednar et al., 2003, Jackson et al., 2006, Brown et al., 2005). Roxarsone is available for purchase in Australia, as one of a wide range of organoarsenic containing products used in poultry agriculture. However, no industry, commercial or governmental information is publicly available to determine its actual usage. A second possible source of inorganic arsenic in environmental waters associated with the facility is the grains and foodstuffs that comprise commercial chicken diets (Yao et al., 2016). Regardless of the primary arsenic source, repeat water sampling (22nd July and 30th July) at Site C confirmed the presence of arsenic (total arsenic - 3.84 µg/L) at concentrations above natural local values. Five reference waterways sampled by the Hornsby Shire Council during 2014–15 show natural waterways across the local region contain ≤ 0.5 µg/L total arsenic (n = 17 samples). The geology of the region is the same as the study catchment—Triassic age Hawkesbury sandstone overlaid by the Wianamatta shale. Therefore, maximum arsenic concentrations in waters associated with facility discharges are up to approximately 8 times higher than local regional samples (Figure 2). However, arsenic(V) concentrations did not exceed the Australian 95% species protection trigger value for freshwater (24 µg/L).

DNA extractions and PCR Assays

Polymerase chain reaction (PCR) amplifications were performed on DNA extracted from water samples collected at Sites C, E and F on 29th July 2016 to detect the presence of chicken tissue. A positive result was obtained *only* from water samples collected at Site C. Each sample from this one location generated a strong DNA product of exactly the same size (266 base pairs)(Lahiff et al., 2001) as the product generated from positive control chicken DNA (Figure 3). No DNA product was generated from water samples collected at Sites E and H, or from the negative controls (duck DNA and sterile distilled water, respectively). PCR amplicons from Site C were submitted for DNA sequencing, and these sequences exhibited 100% homology to chicken mitochondrial DNA sequences lodged in GenBank. In addition, the waters were further analysed for a known proxy of anthropogenic pollution using PCR to detect the clinical class 1

integron-integrase gene, which is known to be resident in gut microbiota of chickens. As per the DNA analysis, only water samples collected from Site C were positive in this assay, generating the expected product (~310 base pairs) (Figure 3).

The detection of chicken DNA and the clinical class 1 integron-integrase gene in water discharged from the facility at Site C confirms unequivocally that the source of environmental pollution is water exiting the facility at site C. In addition to the DNA markers, these waters are also contaminated with elevated levels of ammonia, arsenic, faecal coliforms, nitrogen and phosphorous, providing supporting evidence that the source of the pollution cannot be attributed to any other source but the facility. The absence of any chicken (or duck) DNA indicators in water samples collected from sampling sites not draining the facility indicates that there is no additional source of this biological material in the catchment. This finding corresponds with the conclusion that slightly elevated water quality values identified in watercourses not connected to the facility are probably caused by other sources, such as horse grazing and/or the application of fertilizers (Figure 1).

Source of discharges surrounding the facility

The POEO Act defines water pollution as introducing into water any matter ‘so that the physical, chemical or biological condition of the waters is changed’. Pursuant to the facility’s environment protection licence, offsite discharge of effluent into adjacent waterways is not permitted. Consequently, any such discharges may constitute breach of the licence, and pollution pursuant to the POEO Act.

Following the initial sampling on the 7th and 10th October 2014, the issue of offsite discharges was reported to the NSW EPA (Taylor and Mould 2014). The NSW EPA investigated the matter and subsequently required that the facility undertake a pollution reduction program (NSW EPA 2015b). Specifically, the pollution reduction program required that:

The licensee must investigate options for addressing any limitations of the current wastewater management system; including treatment methods, pond storage capacity (wet and dry weather); irrigation areas; and effluent management practices at the premises.

Despite this recent licence variation, the data presented here demonstrates that regular discharges have continued from the facility throughout 2014–2016. Moreover, the discharges sampled on 22nd and 30th July 2015 and again in July 2016 demonstrate two key issues: (a) the persistent and prolonged nature of effluent discharge into the adjoining catchment; (b) the difficulty facing regulators in being able to unequivocally source polluting activities to enforce compliance.

Discharge of slaughter effluent into natural systems: A public and environmental health risk?

The facility waste water is also piped to adjoining domestic property lots for irrigation. These properties are not included in the facility's licenced effluent irrigation area, for use by local residents. This usage occurs through an informal arrangement between the facility owner and the residential recipients. Analysis of this irrigation water at Sites G and H showed that it was the same grade as that discharged into the watercourse at Site C. This provided additional confirmation that the discharges at Site C were sourced from the facility's waste water and not some other supply, including inter-alia, natural catchment runoff. The data show the irrigation supply was highly polluted with faecal coliforms (>2419 cfu/100ml), a level that is inappropriate for application in a domestic setting where there is a reasonable risk of human exposure. Furthermore, the impacted water course drains into a National Park, where recreational water use is common. This places Park users at risk through exposure to faecal coliforms and other bacteria, and importantly, to the antibiotic resistance genes that are known to be carried by these bacteria and the class 1 integrons that we detected during this study (Gillings 2014a).

However, gathering unequivocal evidence delineating the source and cause of pollution can be challenging, especially where there are a range of potential sources. Even where source

apportionment has been achieved, implementing new licence conditions and assessing compliance remains the ‘Achilles’ heel’ of regulation.

The data from this study illustrate that significant unchecked pollution can enter the environment as a result of livestock (chicken) slaughtering and processing activities. The long record of environmental management issues including non-compliance events associated with waste water discharge from the study facility highlights the need for tighter and better policed regulation to ensure maximum protection of the environment.

Improving the toolkit for environmental regulators

This study adds to emerging research evidence involving the application of genetic markers for tracing sources of pollution. For example, DNA markers were used recently to successfully track effluent pollution from intensive factory pig farming in rural watersheds in North Carolina (Heaney et al. 2015). Identification of antibiotics and their resistance genes is another developing but infrequently applied approach for tracing sources of pollution in the environment. Antibiotics can be sourced from direct anthropogenic sources such as a sewerage discharge, or they can be derived from the administering antibiotics in agricultural settings (Krishnasamy et al. 2015).

Antibiotics in the environment are becoming an increasing concern with a number of key studies predicting an environmental and human health catastrophe arising from the overuse of antibiotics, their prevalence in the environment and subsequent antibiotic resistant diseases (Davis et al. 2011, Hatcher et al. 2016). Consequently, not only is the issue of global concern, the presence of antibiotics, or their resistance genes provides a unique opportunity to trace pollution in the environment as per this study.

While researchers have begun to adopt these approaches, this has not been paralleled by regulators. In Australia for instance, no environmental regulatory body uses genetic markers for pollution source apportionment investigations, relying instead on standard biological and water quality analyses. Consequently, prosecution may not be achievable due to deficiencies in

data with respect to precisely fingerprinting the polluter. This study demonstrates that, along with other evidence, genetic markers provide a solid, robust and investigatory tool that can be adopted by regulators to better inform and direct pollution investigations

Conclusions

Regulators and researchers attempting to source apportion pollution face multiple hurdles with respect to compiling unequivocal evidence. This study's application of a multiple lines of evidence approach using chemical, biological, DNA and PCR markers for tracing slaughterhouse effluent offers a reliable and robust framework for delineating pollution sources and for understanding and investigating the efficacy and appropriateness of environmental management practices. Specifically in this example, the application of analytical tools not used currently by Australian regulatory authorities including arsenic speciation, DNA and PCR provide unambiguous data fingerprinting grossly elevated and persistent pollution of natural watercourses to a chicken processing facility. Although assessing and pinpointing sources of environmental pollution remains a key challenges for regulators wishing to uphold the best and highest standards of environmental management, contemporary and effective environmental analytical techniques and approaches are available to ensure sustainable land use practices, and to prevent degradation of environments surrounding agricultural facilities.

Acknowledgements

P.J. Harvey is funded by a Macquarie University Research Excellence Scholarship (MQRES) (2012195) associated with an Australian Research Council Future Fellowship awarded to H. K. Handley (FT120100440). M.R. Gillings is supported by the Australian Research Council. At the time of writing, M.P. Taylor was undertaking a Review of the New South Wales Environment Protection Authority's (NSW EPA) Management of Contaminated Sites, which was commissioned by the NSW Minister for the Environment. One of the authors is a member

of the local community in which the study was conducted. The authors thank to Rhianon Cunningham for Technical assistance and Simon Mould and Macquarie University students for fieldwork assistance. The local community are thanked for providing access to field sampling locations.

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4 **Contaminants at the City-Scale**

This chapter is a culmination of both the contaminants in the home and agriculture chapters. Studies in this chapter examine environmental contamination at a much larger scale, but the inherent problems are still impacting the community at a more personal household scale. This chapter examines some of the more recent attempts to manage environmental contaminants at the city-wide scale, such as the Lead Abatement Strategy applied to soils surrounding a former lead smelter. Additionally, this chapter builds on the multiple lines of evidence approach to quantifying and identifying the source(s) of environmental contaminants at the much larger city scale. This chapter draws on the investigatory approaches applied throughout this thesis (e.g. VegeSafe) to provide useful and usable information to land managers about environmental contaminants in Australia.

Evaluation and Assessment of the Efficacy of an Abatement Strategy in a Former Lead Smelter Community, Boolaroo, Australia.

P.J. Harvey (70%), M.P. Taylor (10%), L.J. Kristensen (5%), S. Grant-Vest, M (5%).
Rouillon (5%), L. Wu (2%), H.K. Handley (3%)

Environmental Geochemistry and Health 2016, 38(4), 941-954.

Management of contaminated sites can be challenging to get right. This study, which reflects a change in scale to the city level, is an example of an environmental lead contamination technique that was not fit for purpose. In this study, the remediation strategy (Lead Abatement Strategy) for soils in the urban area surrounding the former Pasminco Cockle Creek Smelter (PCCS), a lead-zinc smelter at Boolaroo in New South Wales, is examined. The data show that the strategy adopted was not the most effective approach for this environmental contamination event. The data shows that the everyday living environment, i.e. front and backyard soils, were elevated significantly above the Australian soil guideline concentrations. The study considers the remediation strategy as a management tool designed to limit the exposure risk of human, particularly children, in this environment and compares the approach to other best-practice management techniques globally. Once again, the community was invited to participate in this study by providing access to private property soils and contributing their knowledge of site contamination, including 149 contaminated site assessment certificates to the study. This study was conducted in partnership with the Fairfax Media Ltd Newcastle Herald and subsequently generated extensive media coverage, bringing the issue of environmental lead exposure and contaminated site management back into the public spotlight in Boolaroo and NSW more generally. The outcomes of this study contributed to the formation of the NSW EPA The Lead Expert Working Group, which aimed to determine the effectiveness of the Lead Abatement Strategy at Boolaroo, including a renewed blood lead testing regime.

Pages 121-133 of this thesis have been removed as they contain published material under copyright. Removed contents published as:

Harvey, P.J., Taylor, M.P., Kristensen, L.J. et al. (2016) Evaluation and assessment of the efficacy of an abatement strategy in a former lead smelter community, Boolaroo, Australia. *Environmental Geochemistry and Health*, 38, 941–954 (2016). <https://doi.org/10.1007/s10653-015-9779-8>

Determining the Source and Health Risk Arising from Legacy Environmental Contamination Surrounding a Former Antimony Processing Plant.

P.J. Harvey (90%), H.K. Handley (5%), Taylor, M.P. (5%).

Submission ready.

Non-ferrous ore processing is well documented globally to cause significant environmental contamination burdens, resulting in population health concerns. Antimony (stibnite) ore processing has received considerably less attention than other more popular and traditional ores such as lead. This study was developed by Harvey and Handley in collaboration with the National Measurement Institute (NMI). The study seeks to consider the potential off-site impact of atmospherically transmitted dust surrounding an antimony processing plant in a regional city in Australia. This study uses arsenic, an element co-associated with the stibnite ore deposit, in soils of the urban area surrounding the processing plant as a proxy for environment contamination arising from the processing plant. In addition to using portable X-ray fluorescence spectrometry to calculate total metal(loid) concentrations, this study applies high performance liquid chromatography inductively coupled plasma mass spectrometry (HPLC-ICP-MS) to glean more information about distribution and health risk associated with any potential dust deposition in the urban area surrounding the processing plant. The application of HPLC-ICP-MS as a source tracing tool is underutilised in Australia. In order to utilise this analytical method for this study, the HPLC-ICP-MS technique was required to be set-up and calibrated at the NMI laboratory, which was carried out by Harvey. The data presented here demonstrate that there is no substantial environmental or health risk associated with the soils sampled however there is some minor offsite migration of soil metal(loid)s on the boundary of the former processing plant. This is important for environmental managers as there is an opportunity to further examine the soil and atmospheric environment surrounding the processing plant to determine why no environmental contamination is present. This study is presented here in a publication ready format, however, subsequent to discussion with the NSW EPA, additional sampling and analysis of soils and sediment in the downstream wetlands and overbank deposits will be included in an updated version prior to final publication.

Determining the source and health risk arising from legacy environmental contamination surrounding a former antimony processing plant.

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Keywords

Urban geochemistry, Mining, Health, Speciation

Abstract

Low level naturally occurring arsenic concentrations are ubiquitous in the global environment, however, as a result of anthropogenic activities there has been an increase in environmental As to the extent that in certain situations, it presents a significant health concern. Often, As is co-associated with other environmental contaminants including Sb and Pb, compounding health impacts. One such example is the urban environment surrounding the former Sb processing plant at Urunga, New South Wales, Australia. The co-associated elements Sb and As are examined in soil samples from the adjacent urban area (n = 47) to determine the impact of this former processing plant on the safe living environment of the town. Surface soil total Sb (mean - 3.1 mg/kg, median - < 1 mg/kg, max - 39 mg/kg) was elevated above the regional background (Sb: <1 mg/kg), while As (mean - 10.3 mg/kg, median - 9.3 mg/kg, max - 39 mg/kg) was not enriched above regional background values (As: depth, 30-40 cm, mean - 15.1 mg/kg, median - 17.3 mg/kg, max - 22.1 mg/kg). The data show that soil Sb concentrations close to the boundary of the former processing plant (max - 39 mg/kg) were higher than more distal samples (< 1 mg/kg). Speciation analysis of As by HPLC-ICP-MS (n = 16) indicates a greater proportion of inorganic As in samples collected from the boundary of the processing plant compared to

soil samples from wider locations. Although Australia does not have a soil Sb guideline and no samples exceed the Australia Health Investigation Level for As in soils (100 mg/kg), comparison to international guideline values may indicate a potential health risk associated with those soils immediately adjacent to the processing plant.

Introduction

Australia has a long and prosperous connection to mining, with many mines and their processing facilities situated within urban environments (Harvey et al., 2015b). Often, due to the proximity of industry activities to the urban environment, there are management challenges arising from legacy and ongoing environmental contamination by a range of pollutants including heavy metals (Harvey et al., 2015b, Kristensen et al., 2015, Taylor et al., 2013, Taylor et al., 2014). The town of Urunga, a small town located on Australia's east coast, approximately 450 km north of Sydney (Figure 1), was, until late 2015, the former site of a Sb ore processing plant. The Urunga local environment itself is comprised predominantly of quartz rich sands from the overbank deposits of the Kalang River with intrusions of coastal marine sediment overlying the Nambucca Slate Belt, a highly metamorphosed unit of the marine sediment derived Nambucca Beds (Leitch, 1975, Osborne et al., 1998). The Sb ore processing plant, which operated between 1969 and 1974 and originally owned and operated by Broken Hill Antimony NL then later Australian Antimony Corporation NL, crushed and separated the ore from the nearby stibnite deposits of the Wild Cattle Creek prospect (DPI, 2015, Rabone, 2010, Figure 1). In the five year period, the plant was responsible for crushing 16,500 tonnes of ore and generating 300 tonnes of high purity Sb, also generating by-products As, Cu, Pb, Zn, Cr and Mg (DPI, 2015, Rabone, 2010). Due to a falling market price for Sb and subsequent failure of the company, the plant was closed in 1974 (Rabone, 2010). Remediation of the processing site and adjoining wetlands commenced in July 2015.

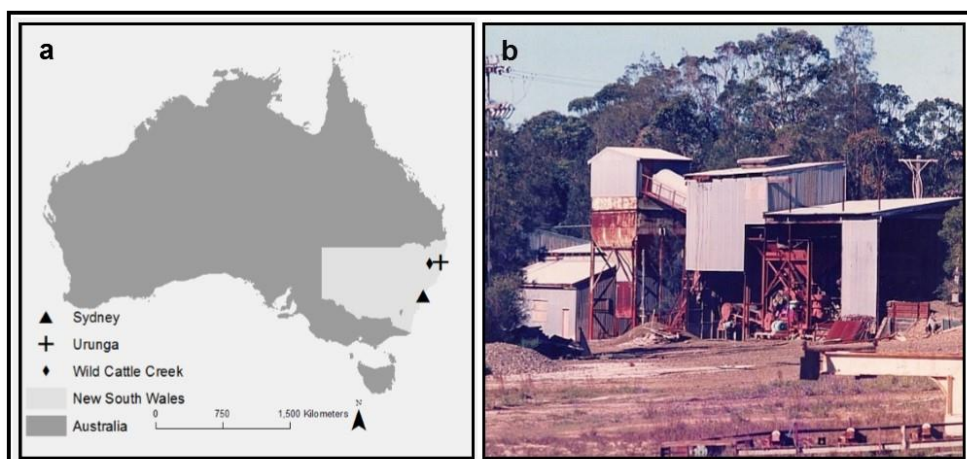


Figure 1 Inset a: Location map of the former Sb processing plant, Urunga NSW in relation to Sydney and the major ore body, Wild Cattle Creek. Inset b: the former Sb processing plant infrastructure, note the artisanal style of the facility and the absence of dust control measures around the chutes – image provided by Bellingen Museum, NSW.

One of the primary by-products of the former processing plant is As, derived from the arsenopyrite associated with the stibnite mineralisation zone in the source ore deposits (Rabone, 2010). Arsenic is becoming a widespread environmental contaminant and exposure to it at elevated levels is responsible for enormous rates of disease, with conditions ranging from skin lesions to terminal carcinoma (Henn et al., 2016, Rahman et al., 2009, Ramirez-Andreotta et al., 2013, Tchounwou et al., 2004). Epidemiological studies show that relatively low concentrations of As in the environment, combined with other low concentration exposures through food and water pose the greatest concern for human health (Kordas et al., 2016). More broadly, metal mining is often also associated with dispersed As contamination (Sultan, 2007), due to mines with insufficient pollution control measures releasing high As concentration tailings into the fluvial environment (Bruce et al., 2003, Ashley and Lottermoser, 1999, Telford et al., 2009, Telford et al., 2008). According to Smith et al. (2002), Australia has >10,000 sites that are affected by As and are considered contaminated. Gaining considerable attention is contamination of drinking water sourced from groundwater on the Australian coastal fringes caused by the dissolution of localised As-bearing deposits following intrusion of seawater into aquifers (Mukherjee et al., 2009, O'Shea et al., 2007, Appleyard et al., 2006, Smith et al., 2003).

Arsenic has historically been used in weed control chemicals, for example, Smith et al. (2006) documented extensive As contamination of disused railway corridors in South Australia earmarked for land use change, resulting from herbicide application during former track maintenance. Similarly, fruit orchards and livestock dip sites are a significant sink of anthropogenically derived As from the application of industrial chemicals (Edvantoro et al., 2003, Juhasz et al., 2007, McLaren et al., 1998, Merry et al., 1983). More globally, in Bangladesh, local bedrock derived As contaminated groundwater has potentially impacted more than 77 million people, resulting in severe chronic preventable morbidities and mortality including renal disease, cardiovascular disease, various cancers and death (Smith et al., 2000). There is also an intergenerational component to As exposure via its adverse impacts on unborn children, which result in a range of unfavourable pregnancy outcomes (Kun et al., 2014). While drinking water has recently become the most documented exposure pathway to As in the environment, ingestion of dust and soil also presents an appreciable risk to human health, particularly for young children (Hwang et al., 1997, Martin et al., 2014).

Globally, there is a huge disparity between health investigation guideline concentrations for As in the soil environment (Table 1). Arsenic has a number of oxidation states, or species, with

differing levels of toxicity. The differences in soil investigation guideline concentrations are mainly due to debate surrounding the toxicity of those species (Teaf et al., 2010). The oxidation or reduction of species is dependent on a range of geochemical properties of the environment including soil elemental chemistry, pH and redox potential (Smith et al., 2002). The inorganic species, arsenite (As^{III}) and arsenate (As^{V}) are commonly believed to be the most detrimental to human health (Smith et al. 2006). In addition to the inorganic species, a number of organic species are often determined, the most common of which include methylated dimethylarsinic acid (DMA) and monomethylarsonic acid (MMA), trimethylarsine oxide (TMAsO) and arsenobetaine (AB) (Maher et al., 2015, Wilson et al., 2010).

Table 1. A selection of global soil total As guideline concentrations, after Teaf et al. (2010).

Location	Total concentration (mg/kg)
Finland	5
Canada	12
United Kingdom	32
Netherlands	76
Australia	100
Japan	150

High Performance Liquid Chromatography coupled with Inductively Coupled Plasma Mass Spectrometry (HPLC-ICP-MS) is a robust technique used to determine the species of As present within a range of matrices (Leybourne et al., 2014). Due to its efficiency, and relatively fast and high volume sample throughput, the HPLC-ICP-MS technique has been used on a broad range of applications for As detection, including ecotoxicology, health studies, environmental forensics and mine site impact assessment (Foster et al., 2006, Marcinkowska et al., 2015, Rehman et al., 2016, Telford et al., 2008, Telford et al., 2009, Zheng et al., 2003,).

In this study we use HPLC-ICP-MS to assess As speciation and broader environmental contamination associated with ore processing at Urunga. Aside of the on-site reports undertaken to assess the plant's impact (DPI, 2015, GHD 2012, 2014, Leahy, 2013) there has been no publically available study examining off-site impacts on the urban environment. Moreover, the absence of any As speciation assessment means that the toxicological risk off-site contamination remains unconstrained. This study: 1) determines soil As and Sb concentrations in the suburban living environment potentially derived from atmospheric dust deposition from the former processing plant via pXRF and ICP-MS; 2) forensically investigates the source of the contamination by HPLC-ICP-MS analysis; and 3) Evaluates the toxicological risk associated with any off-site contamination of soils in the suburban living environment. To the best of the author's knowledge, this study represents the first application of HPLC-ICP-MS in the Australia research literature for differentiating a contamination source and understanding health exposure risk associated with a potential pollution source, at the broad town scale.

Methods

Field sampling

Soil samples were collected in October 2014 from the urban area surrounding the former Urunga Sb processing plant (Figure 2). Soils (n = 47) were collected from the surface (0 – 2 cm) on public land. Sampling of the uppermost surface soils is consistent with the findings of

the Lead in Soil and Dust Working Group (1994) who determined that the upper 2 cm is the most toxicologically relevant sampling medium for health risk assessment purposes. Depth samples ($n = 15$; 4 profiles, maximum depth 40 cm) were also collected to determine the local background soil element concentrations. Soil sampling was conducted according to published methods (Harvey et al. 2015a; Taylor et al. 2010). A small number of sites ($n = 14$) were also analysed in-situ using a portable X-ray fluorescence spectrometer (pXRF) (Olympus® InnovX Delta series, 4 W 50 kV pXRF operating in soil mode with factory calibration) for As. Instrument analytical precision was determined by a routine calibration check, continuous monitoring of a blank (SiO_2) sample and a NIST 2710a CRM with relative standard deviation (RSD) of 80-120%.

Laboratory Analysis

Soil samples were oven dried at 40°C for 24 hrs then sieved using a 180 μm mesh. The < 180 μm fraction was then packed into sampling cups for analysis using a pXRF (Olympus® InnovX Delta series, 4 W 50 kV pXRF operating in soil mode with factory calibration) in a desktop test stand. Analysis by pXRF in this manner has been shown to produce equivalent data quality to wet chemistry methods (Kenna et al., 2011, Rouillon and Taylor, 2016). Analysis was performed using three beams of 30 second duration in order to detect Sb and As. Due to the possibility of interference from the known peak overlap on the pXRF from Pb with respect to As, this too was quantified but it does not form part of the main analysis of the study. Samples were bracketed with a low trace element Certified Reference Material (CRM) (NIST 2711a, Montana II Soil) which contains low concentration Sb and As returning Sb 11% and As 6% RSD and recoveries of Sb 112% and As 91%.

A sub-set of soils ($n = 16$) was selected for analysis by HPLC-ICP-MS to determine the As species present within those soils. Analysis was performed at the Inorganics Laboratory, National Measurement Institute (NMI), Sydney. To determine the acid-extractable concentration of As, approximately 1 g of sample was digested in 5 mL analytical grade HNO_3 (MERCK) on a boiling water digestion block for 1 hr at 95 - 100°C followed by dilution in milli-Q water to 9 mL. Due to similar elution times of arsenobetaine (AB) and As^{III} , 1 mL of hydrogen peroxide solution (Sigma-Aldrich, 30% w/w in H_2O , H_2O_2) was added to the solution to oxidise all As^{III} to As^{V} for reporting total inorganic As (Tziaras et al., 2015). An Agilent 7500cc octopole ICP-MS was coupled with an Agilent Infinity 1200 HPLC system. Samples were introduced via a Mira Mist nebuliser connected via PEEK tubing to an anion exchange Hamilton PRPX-100 column (10 μm , 250 x 4.1 mm). The method utilised an isocratic mobile phase of 10 mM malonic acid (Sigma-Aldrich, ReagentPlus 99% purity, $\text{C}_3\text{H}_4\text{O}_4$) diluted in

1000 mL milli-Q water and adjusted to pH 5.6 with ammonia solution (Sigma-Aldrich, ACS reagent 30% w/w in H₂O, NH₄OH) (Nachman et al., 2012). A 20 µL sample injection volume was used with a 1 mL/minute flow rate. The heat block was maintained at room temperature (20°C) and a column backpressure of 40 bar. An 11 minute stop time was applied to the method. The Agilent ChemStation software in time resolved analysis mode was used to detect *m/z* 75 and monitor isotopic interference masses of Cl (Maher et al., 2015). Blanks, sample spikes, replicates and standards were analysed throughout the HPLC analysis. To validate the concentrations of Sb and As in the soils and to calculate HPLC column recovery, a 2 mL aliquot of the digested sample was further diluted using 400 µL internal standard and milli-Q water to a final volume of 10 mL and analysed for total concentrations on an Agilent 7900 octopole ICP-MS. All blank samples were below the level of instrument detection (Agilent 7900: 1 µg/L, Agilent 7500 2 µg/L). The ICP-MS total As determination duplicate and matrix spike samples had a RPD of 0.75% and recovery of 109% respectively. The HPLC-ICP-MS analysis had digestion duplicate RPD, analysis duplicate RPD and recovery of 16.6%, 2.2% and 71 – 121% respectively. The 7500 ICP-MS for HPLC-ICP-MS was tuned daily with a 10 ppb in-house SRM solution. Both ICP-MS instruments were calibrated daily using a 10 ppb in-house SRM mixed element solution and had recoveries of between 90 and 110%. Samples analysed for total concentration on the Agilent 7900 ICP-MS were spiked with a 20 ppb rhodium/iridium standard and returned recoveries between 90 and 110%.

Results and Discussion

Laboratory pXRF analysis of the soils from the 47 sites in the urban environment surrounding the former Urunga processing plant show relatively low concentrations of both As and Sb. The mean concentration for soil As was 10.3 mg/kg (median – 9.3 mg/kg, max – 39 mg/kg, Table 2) and the Sb 3.1 (median – < 1 mg/kg, max – 39 mg/kg, Table 2). The maximum soil total As pXRF concentration (39 mg/kg) is detected in the soil from the channelised area downstream of the processing plant (Figure 2, Table 2). A number of samples close to the processing plant and in the adjoining estuary have the highest total As concentrations. Aside of these samples, most surface soil concentrations are comparable to concentrations in soils at depth indicating no evidence of surface enrichment by anthropogenic processes (Figure 3). This is in contrast to other mining environments where extensive atmospheric dust deposition creates a distinctly enriched surface layer compared to depth (Taylor et al. 2010). Total soil As concentrations close to the M1 motorway which passes through the town (max – 19.2 mg/kg) are greater than soils in more isolated suburban pockets of the town (Figure 2). The Australian Soil Guidelines (NEPM, 2013) Health Investigation Level (HIL) for As is 100 mg/kg. Overall, the As pXRF concentrations in the urban environment surrounding the former Urunga processing plant are

low compared to this guideline. Likewise, the concentration of Sb is most elevated in soil immediately surrounding the processing plant and in the downstream reach of the surrounding estuary (Figure 2, Table 2). The maximum soil Sb concentration (39 mg/kg) is detected on the boundary of the former plant indicating some minor off-site migration of Sb (Figure 2, Table 2). Although Australia does not have a soil Sb guideline value for comparison, the detected values at the surface, benchmarked against natural values found at in soil depth profile (~40 cm) show that there is surface enrichment at sites closest to the processing plant.

Overall, based on the distribution and concentration of total As and Sb in the soils throughout Urunga and contrary to other urban environments adjacent to mine and processing facilities in Australia (e.g Taylor et al. 2014), the data indicates no substantial surface enrichment of As or Sb in soils beyond the boundary of the processing plant. Although the concentrations of elements at sites adjacent to the processing plant are low compared to NEPM (2013) guidelines, the evidence demonstrates that the only probable source of these elements in the soil environment at those sites is the former processing plant.

It has been previously noted that off-site surface migration of contaminants into the adjacent wetland at Urunga has occurred (DPI, 2015). Organic matter and the free colloid fraction, pH and Fe content of the soil profile play an important role in the retention of many metals and metalloids. In the absence of one of these natural controls, metals and metalloids can readily pass through the profile (Drahota et al. 2016, Cutler et al., 2013, Moreno-Jiménez et al., 2010, Weng et al., 2001). Acid sulphate soils derived from processing the leachate of the sulphate bearing Sb ore has historically mobilised the on-site surface contaminants into the groundwater, causing off-site sub-surface contamination (GHD, 2012). Although there may be no clear off-site dust deposition in the urban environment, the channel sediment deposit analysed in this study demonstrates that there is a transfer of contaminants through the waterway. This poses a potential risk for users of this environment, particularly the species dependant on the wetlands for food and that bio-accumulate metals. Regional research into the accumulation of contaminants by pearl oysters shows they are efficient filters of water and retain contaminants in their flesh and shell (Gifford et al., 2005). As these are first and second order feeders, bio-accumulation in the food chain is therefore likely. Urunga lagoon, Kalang and adjoining Bellingen Rivers are popular commercial and recreational fishing locations so this is a notable exposure risk.

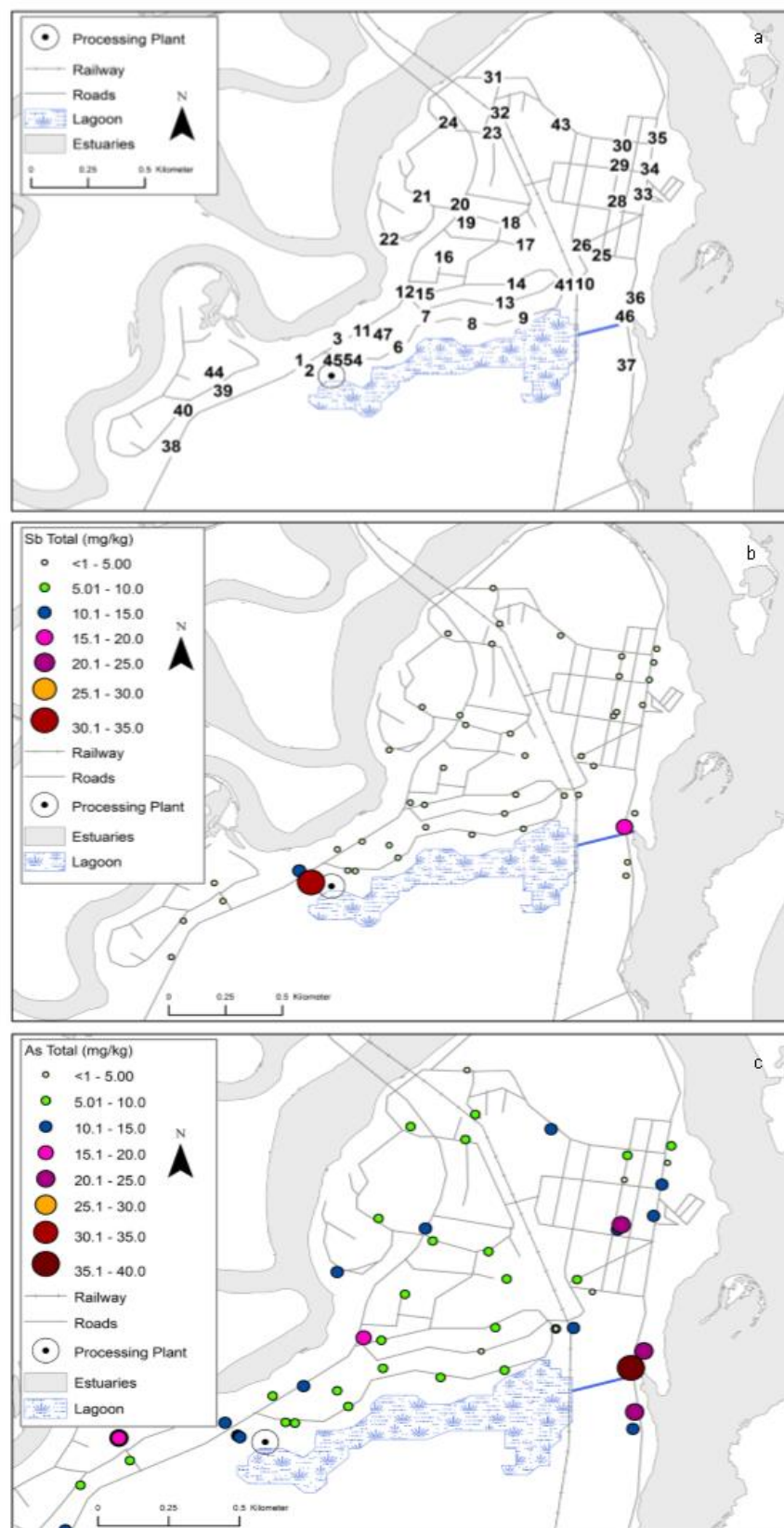


Figure 2 a) Sampling site locations around the town of Urunga. Soil total Sb (b) and As (c) concentrations by laboratory based pXRF in soil surrounding the former Sb processing plant at Urunga, New South Wales.

Table 2 Total As, Sb and Pb laboratory pXRF soil concentrations in the Urunga sampling area. All concentrations are mg/kg.

Sample Site	X co-ordinate	Y co-ordinate	As	Sb	Pb
1	153.008300	-30.504130	14.0	13.0	94.0
2	153.008700	-30.504580	10.0	21.0	39.3
2 DUP 1	153.008700	-30.504580	11.4	25.0	38.0
2 DUP 2	153.008700	-30.504580	8.3	28.0	40.8
2 REP 1	153.008700	-30.504580	9.8	24.0	39.9
2 REP 2	153.008700	-30.504580	10.0	27.0	42.0
3	153.009810	-30.503200	9.4	ND	30.3
4	153.010517	-30.504139	5.9	ND	12.4
5	153.010222	-30.504117	6.8	ND	22.2
6	153.012210	-30.503565	7.8	ND	15.0
7	153.013310	-30.502240	9.3	ND	81.0
8	153.015140	-30.502560	9.7	ND	17.4
9	153.017170	-30.502310	6.7	ND	16.8
10	153.019360	-30.500840	11.0	ND	32.3
11	153.010790	-30.502860	10.3	ND	28.7
12	153.012700	-30.501180	19.2	ND	36.8
13	153.016430	-30.501650	ND	ND	271
14	153.016880	-30.500830	5.6	ND	18.9
15	153.013260	-30.501270	7.4	ND	39.6
16	153.014010	-30.499670	5.2	ND	24.3
17	153.017240	-30.499140	7.5	ND	86.0
18	153.016660	-30.498190	6.2	ND	24.2
19	153.014890	-30.497820	10.0	ND	106
20	153.014660	-30.497390	14.0	ND	220
21	153.013170	-30.497040	5.2	ND	11.8
22	153.011860	-30.498900	13.6	ND	12.0
23	153.015930	-30.494300	9.8	ND	34.1
24	153.014190	-30.493850	7.9	ND	27.0
25	153.019960	-30.499590	ND	ND	319
26	153.019470	-30.499160	7.3	ND	18.4
27	153.020750	30.497430	13.5	ND	81.0
28	153.020870	-30.497260	20.3	ND	17.6
29	153.020970	-30.495700	ND	ND	294
30	153.021070	-30.494850	9.8	ND	32.5
31	153.015980	-30.491890	ND	ND	73.0
32	153.016240	-30.493430	6.9	ND	29.2
33	153.021900	-30.496950	10.9	ND	44.4
34	153.022164	-30.495861	13.6	ND	92.0

Sample Site	X co-ordinate	Y co-ordinate	As	Sb	Pb
34 DUP 1	153.022164	-30.495861	4.5	ND	39.4
35	153.022460	-30.494520	8.8	ND	22.2
36	153.021590	-30.501640	21.7	ND	16
37 0-2	153.021244	-30.504344	13.8	ND	30.7
37 2-10	153.021244	-30.504344	15.8	ND	7.5
37 10-20	153.021244	-30.504344	17.8	ND	ND
37 20-30	153.021244	-30.504344	14.6	ND	4.5
37 30-40	153.021244	-30.504344	21.2	ND	ND
38	153.003230	-30.507860	11.9	ND	46.6
39	153.005280	-30.505440	7.9	ND	18
40	153.003710	-30.506300	6.7	ND	14.7
41 0-2	153.018800	-30.500870	4.4	ND	25.9
41 2-10	153.018800	-30.500870	6.7	ND	31.7
41 10-20	153.018800	-30.500870	7.1	ND	27.5
41 20-30	153.018800	-30.500870	5.3	ND	20.4
41 30-40	153.018800	-30.500870	3.6	ND	15.2
42	153.008770	-30.504630	12.1	34.0	22.7
43 0-2	153.018650	-30.493940	8.1	ND	23.6
43 2-10	153.018650	-30.493940	6.9	ND	24.3
43 10-20	153.018650	-30.493940	10.2	ND	27.5
43 20-30	153.018650	-30.493940	8.2	ND	22.8
43 30-40	153.018650	-30.493940	13.3	ND	18.6
44 0-2	153.004930	-30.504660	16.8	ND	22
44 0-2 DUP 1	153.004930	-30.504660	20.1	ND	20.7
44 0-2 REP 1	153.004930	-30.504660	17.6	ND	23.7
44 0-2 REP 2	153.004930	-30.504660	18.0	ND	19.5
44 2-10	153.004930	-30.504660	18.6	ND	20.8
44 10-20	153.004930	-30.504660	10.9	ND	20.1
44 20-30	153.004930	-30.504660	22.1	ND	15.8
45	153.009796	-30.504136	29.9	39.0	23.7
46	153.021180	-30.502230	39.0	16.0	91
47	153.011860	-30.503020	5.9	ND	22.3

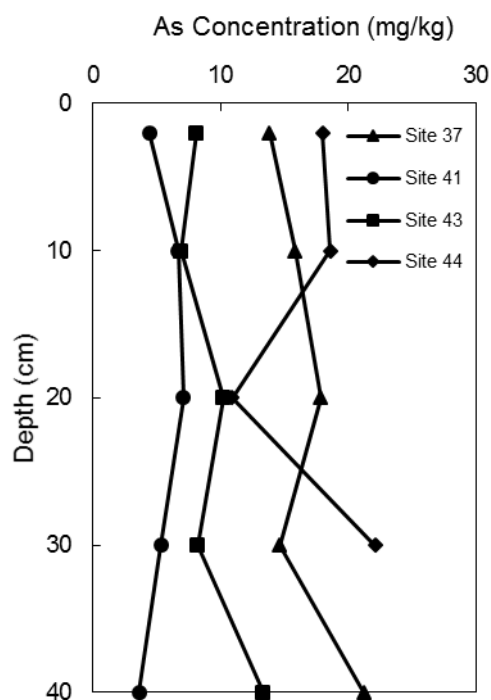


Figure 3 Soil depth profiles from the Urunga study area. Site 44 reached an impenetrable soil horizon at 30 cm.

Arsenic Speciation of Soils and Sediment

To further constrain the source and toxicological risk of the near boundary off-site As concentrations, HPLC-ICP-MS was used. Arsenic speciation of samples throughout the study area reveals that the samples collected close to the former processing plant, which have some of the greatest As concentrations in the data set, have a higher proportion of total inorganic As species (as either As^{III} or As^V in the sample) compared to those sites further from the processing plant which have a greater organic fraction (Table 2, Figure 2). Sites 29 and 33 in the central business area of Urunga also have a high proportion of inorganic As and these locations are situated next to commercial premises with heavy vehicle access (Table 2). The remainder of the samples have a very low proportion of total inorganic As (Table 2). Inorganic As has been associated with industrial processes such as mining and metal production (Huang et al., 2014) and is commonly found in elevated concentrations surrounding mining and processing locations (e.g. Casiot et al., 2007). Organic As species (AB, DMA and MMA) are detected as the dominant species in the remainder of the samples, suggesting that these samples are more significantly influenced by the background (organic) concentrations of As in the environment than inorganic contamination that has emanated from the former processing plant. Comparison of the speciation fingerprint, particularly noting the inorganic fraction, from the soil at depth at Site 37 compared to surface soil at Site 37 illustrates a surface enrichment with inorganic As from an introduced source. The As speciation data suggests that a proportion of the total As in

the broader urban environment has originated from the migration of contamination from the processing plant.

Table 3 HPLC-ICP-MS As speciation of samples in the Urunga sampling area. Inorganic As^V is inclusive of the oxidised As^{III} fraction. All concentrations in mg/kg.

Sample Site	ICP-MS	AB	DMA	MMA	As ^V	Σ Species	Column Recovery (%)
1	3.9	2.1	< 0.05	0.72	< 0.05	2.8	71.7
2	5.3	1.7	< 0.05	1.4	2	5.1	96.2
5	2.3	1.8	< 0.05	< 0.05	0.78	2.6	113
6	2.5	1.5	< 0.05	0.38	< 0.05	1.9	75.9
11	4.5	1.8	1.5	2.1	< 0.05	5.4	121
22	7	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05
24	5.3	3.1	1	1.8	< 0.05	5.9	112
29	3	0.24	0.91	0.8	1.3	3.3	108
33	7.2	3.9	< 0.05	< 0.05	2.2	6.1	84.8
34	2.3	1.5	< 0.05	0.5	0.5	2.5	107
37	5.2	1.9	1.6	< 0.05	2.3	5.8	112
37 30 – 40	8.5	2.6	2.7	2.5	< 0.05	7.8	92.2
39	3.3	1.7	< 0.05	0.72	< 0.05	2.4	74.4
45	21.3	11.5	< 0.05	< 0.05	5.4	16.9	79.5
46	43.8	21.4	7.9	< 0.05	8.9	38.2	87.1
47	2.3	1.5	0.44	< 0.05	0.73	2.7	116

The method used does not quantify the compound TMA₂AsO, however, TMA₂AsO is often associated with atmospheric emissions so it is likely that it is present in the samples and was undetected, or formed part of the detected inorganic or AB fractions (Huang et al., 2006, Huang et al., 2010, Huang et al., 2014). The column recoveries (< 100%, Table 3) compared to total As via ICP-MS could indicate that additional compounds are present in the sample and not detected by the HPLC-IPC-MS analysis method used. Conversely, the column recoveries >100% are likely to be a function of sample in-homogeneity between the two separate aliquots of the bulk sample analysed independently by ICP-MS for total As and HPLC-ICP-MS.

Soil Guidelines in Environmental Investigations and the Need for Universal Clarity

There is significant global diversity among soil As health guidelines (Table 1). Guidelines are developed and implemented following consideration of a number of factors including natural soil loading and exposure pathways (Jennings, 2013). Most importantly, the soil guidelines are significantly influenced by the toxicological research surrounding each contaminant (NEPM, 2013). With the onset of exposome research, it has been shown that increasingly lower exposure concentrations are linked to subtle health impacts (Hornung and Lanphear, 2014, Wild et al.,

2013). Australia has a total soil As guideline value of 100 mg/kg in residential soils and up to 3000 mg/kg in commercial soils (NEPM, 2013). Compared to other international guidelines (Table 1) these are highly conservative numbers for environmental protection. One of the notable limitations of this study is the deficiency of the Australian NEPM (2013) soil guideline for Sb. For example, Canada has a soil interim remediation Sb guideline of 20 mg/kg in residential environments and 40 mg/kg for industrial sites (CCME, 2007). International research is beginning to demonstrate that Sb exposure can have detrimental impacts on the developing human body and include, among a range of other outcomes, respiratory disease and cancer (Amarasiriwardena and Wu, 2011, Li et al., 2014, Schnorr et al., 1995, Wu et al., 2011,). Although Sb exposure occurs predominantly in the workplace, the growth of manufacturing waste recycling facilities in close proximity to residential areas, Sb in road dusts, the inclusion of Sb in fire retardants and in plastics, is increasing the opportunity for exposure to occur more generally (He et al., 2012, Månsson et al., 2009). According to the Canadian guideline (CCME, 2007), the data in this study shows that a number of the sample sites bordering the residential area of Urunga are contaminated by Sb.

Due to on-site soil exceedance of the NEPM (2013) values, the former processing plant is currently undergoing a clean-up implemented by Crown Lands to remove the on-site soil contamination risk and to convert the land use to a public parkland (DPI, 2015). The project is estimated at a cost of \$4 million, covering an area of 4 hectares and includes the construction of a soil containment cell and the importation of clean soil (DPI, 2015). The clean-up does not target off-site soils in the urban environment.

Arsenic and Antimony in the Global Environment.

Arsenic and particularly Sb are often considered emerging contaminants, with one area of concern being electronic waste recycling areas where, for example, highly contaminated dust is generated that can eventually contaminate drinking water and food products, causing significant health impacts (Luo et al., 2011, Leung et al., 2008). In one electronic waste study area of the Guangdong Province, China, Sb concentrations were 147-fold greater than those of control sites. Liu et al. (2002) also described the health and environmental impact of arsenic release from coal combustion in China. In this study, 3,000 patients with arsenic poisoning presenting with severe skin lesions, lung dysfunction and other diseases were identified. The population exposure was exacerbated by indoor combustion of coal (Liu et al. 2002).

This study has examined the potential off-site contamination of soils from the Urunga processing plant following previous assessments indicating on-site contamination. One by-product of the processing, arsenic, was used as an environmental tracer to assess off-site deposition in the urban area. In terms of public health risk, it is fortunate that the total and

speciated arsenic concentrations do not indicate significant deposition of ore processing dusts. This conclusion is supported by the very low Sb concentrations in soils throughout the urban environment. In contrast, the impact of the Xikuangshan Mine, China, which is the world's largest Sb mine resulted in significant As and Sb environmental contamination and degradation of rice, drinking water and vegetables (Fu et al., 2011, Okkenhaug et al., 2011, Wu et al., 2011). Analysis of the Sb by HPLC-ICP-MS speciation in the environmental waters surrounding this mine indicate waters were significantly contaminated by Sb^V, one of the most toxic Sb species (Liu et al., 2010, Wu et al., 2011). While the Urunga former processing plant does not appear to be significantly contributing Sb to the aquatic environment, the risk of exposure through seafood consumption in the lagoon and estuary warrants further investigation to assess the toxicity of low concentration Sb in that environment.

This study sets out an approach for the fast and efficient identification and assessment of potential environmental As and Sb contamination. The application of As speciation analysis to understanding environmental sources and risks is relevant to developing a proper evidentiary basis for the protection of human health in the urban environment. This approach has significant potential to be applied to other similar locations in Australia and globally.

Conclusion

Soil Sb and As concentrations surrounding the former Sb processing plant at Urunga suggest minor off-site migration at the site boundary and into the adjoining wetland and lagoon area. However, the data indicate that off-site emissions have not resulted in significant deposition of contaminated processing dust across the urban area. Soil Sb and As values are elevated at the ore processing boundary with some urban soil As being slightly above local background concentrations. However, no soil concentration exceeds the Australian soil HILs. As speciation analysis of soils shows samples closer to the processing plant have a greater inorganic proportion compared to more distal samples, indicating a minor contribution from the processing plant. This study sets out a novel forensic approach, using HPLC-ICP-MS, to identifying the source of environmental contamination in an urban-industrial environment. The source apportionment method presented here provides scope for future contaminated site assessments where more conventional approaches such as lead isotope analysis may not be suitable. Additionally, the insight provided by the health exposure risk assessment component of the element speciation technique is valuable for clean-up and future site management strategies.

Acknowledgements

P.J. Harvey is funded by a Macquarie University Research Excellence Scholarship (MQRES) (2012195) associated with an Australian Research Council Future Fellowship awarded to H.K. Handley (FT120100440). This project is the result of a collaborative research agreement between Macquarie University and the National Measurement Institute. Michael Wu, Andrew Evans, Ping Di, and Jasminka Jaksic from the Inorganics Laboratory, National Measurement Institute are thanked for laboratory assistance and comments on the draft manuscript. Dr Simon Foster from the University of Canberra and Giovanni Agosti from EnviroLab Group, Fred Fryer and Michael Gleeson from Agilent Technologies are thanked for assistance with method application. A/Prof Norman Pearson of the Macquarie University Geochemical Analysis Unit is thanked for his support throughout the project. Yu Wang is thanked for fieldwork assistance.

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Geochemical Sources, Forms and Phases of Soil Contamination in an Industrial City

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Science of the Total Environment. In Press.

Newcastle is an Australian industrial city. For many years the city was synonymous with the hard-working middle class of Australia's industrial era. The city was dominated by steel manufacturing and shipping ports. The city, built on the inter-tidal flats of the Hunter River, sprung up around the industry, creating a sprawling urban area. Despite over 100 years of industrial activities in the city of Newcastle, there had been no examination of the potential soil environmental contamination there. This study, conceived by Harvey and Rouillon, originally set out to determine if there was an environmental contamination problem in Newcastle. A student group interested in urban geochemistry was invited to assist with sample collection and metal(loid) analysis. It became evident from this first batch of soil samples that the Newcastle urban area was substantially contaminated by a range of metal(loid)s. Additional investigation of the private space soils was conducted which revealed an even worse contamination problem in those soils. In addition, we were interested in understanding the source(s) of contaminants in the soil. Soil lead isotope compositions and mineralogy were determined and these indicated an industrial, likely ferrous and non-ferrous smelting, source. This study employed a multiple lines of evidence approach to build the case that the environmental contamination of the soils was a result of the historic industrial activities of the city. The historic source of contaminants means that this is a legacy contamination issue. The study considers ways that legacy contamination issues can be identified and managed in a contemporary setting. This work was presented at the Goldschmidt 2016 conference in Yokohama, Japan.



Geochemical sources, forms and phases of soil contamination in an industrial city

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ARTICLE INFO

Article history:

Received 5 December 2016

Received in revised form 9 January 2017

Accepted 9 January 2017

Available online xxx

Editor: F.M. Tack

Keywords:

Soil

Metal(loids)

Lead isotopes

Bio-accessibility

Mineralogy of metal-bearing particles

Newcastle

ABSTRACT

This study examines current soil contamination in an Australian industrial city, Newcastle. Public (roadside verges and parks) and private (homes) surface soils ($n = 170$) contained metal(loids) elevated above their respective Australian Health Investigation Levels (HIL). Lead (Pb), the most common contaminant in the city, exceeds the HIL for residential soils (HIL-A, 300 mg/kg) in 88% of private soils (median: 1140 mg/kg). In-vitro Pb bio-accessibility analysis of selected soils ($n = 11$) using simulated gastric fluid showed a high affinity for Pb solubilisation (maximum Pb concentration: 5190 mg/kg, equating to 45% Pb bio-accessibility). Highly soluble Pb-laden Fe- and Mn-oxides likely contribute to the bio-accessibility of the Pb. Public and private space surface soils contain substantially less radiogenic Pb (range: $^{208}\text{Pb}/^{207}\text{Pb}$: 2.345–2.411, $^{206}\text{Pb}/^{207}\text{Pb}$: 1.068–1.312) than local background soil ($^{208}\text{Pb}/^{207}\text{Pb}$: 2.489, $^{206}\text{Pb}/^{207}\text{Pb}$: 1.198), indicating anthropogenic contamination from the less radiogenic Broken Hill type Pb ores ($^{208}\text{Pb}/^{207}\text{Pb}$: 2.319, $^{206}\text{Pb}/^{207}\text{Pb}$: 1.044). Source apportionment using Pb isotopic ratio quantification and soil mineralogy indicate the city's historic copper and steel industries contributed the majority of the soil contaminants through atmospheric deposition and use of slag waste as fill material. High-temperature silicates and oxides combined with rounded particles in the soil are characteristic of smelter dust emissions. Additionally, a preliminary investigation of polycyclic aromatic hydrocarbons in soils, sometimes associated with ferrous metal smelting, coal processing or burning of fossil fuels, shows that these too pose a health exposure risk (calculated in comparison to benzo(a)pyrene: $n = 12$, max: 13.5 mg/kg, HIL: 3 mg/kg).

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1. Introduction

Urban environmental contamination is an issue common to industrial cities throughout the world (Filippelli et al., 2015). Historically, employees would live close to their work, resulting in dense urban development around industrial operations. Due to the proximity of industrial operations to urban environments there is the potential for adverse impacts on human health arising from industrial emissions (Csavina et al., 2012; Dong et al., 2015; Ettler et al., 2009; Gulson et al., 1994; Gulson et al., 2004; Gulson et al., 2009; Morrison, 2003; Taylor et al., 2010; Taylor et al., 2013; Taylor et al., 2014a; Taylor et al., 2014b).

By 2050, the majority of the global population is expected to transition from rural to urban-metropolitan living, increasing the risk associated with contaminated urban lands (United Nations (UN), 2015). The resulting environmental contamination burden from industrial cities is demonstrated in Detroit, USA, which has been considered one of the most contaminated cities in the USA (Lougheed, 2014). Sulfur dioxide, heavy metals, hydrocarbons and other organic contaminants that were emitted to the atmosphere during the city's automobile manufacturing era, accumulated in the urban soil environment and have been subsequently linked to a range of health impacts in-

cluding chronic respiratory diseases and mental disabilities, particularly in infants and young adults (Dong et al., 2015; Lougheed, 2014; Canfield et al., 2003; Taylor et al., 2016). Although Detroit battles legacy environmental contamination issues, modern industrial processes can also contribute to the environmental contamination burden. More recently, the rapid and spontaneous expansion of China's manufacturing and industrial economy has resulted in an overlap of land-use with urban-residential areas. The emissions generated from industrial processes have been linked to substantial carcinogenic health effects in the city of Beijing (Chen et al., 2015). In order to address this problem, a better understanding of environmental contamination and the impacts on population health in urban-industrial environments is required.

This study investigates an Australian industrial city, Newcastle, and the risk of exposure associated with environmental contaminants in the soils of the city. Newcastle, approximately 200 km north of Sydney (Fig. 1), is historically a manufacturing-based city. However, the city has recently become a regional population center, with an economy moving towards services, arts and tourism (Newcastle City Council (NCC), 2013; Newcastle City Council (NCC), 2015). Large-scale industrial operations began in the mid 1800's with the Wallaroo Copper Company smelter (1868 to 1892) processing annually ~ 1300 tonnes of low grade Wallaroo Mine (South Australia) copper ore (Newcastle City Council (NCC), 1997; SMH, 1871a; SMH, 1871b; The Mercury, 1867). During that time, a number of other operations were also established, including the Waratah Coal Com-

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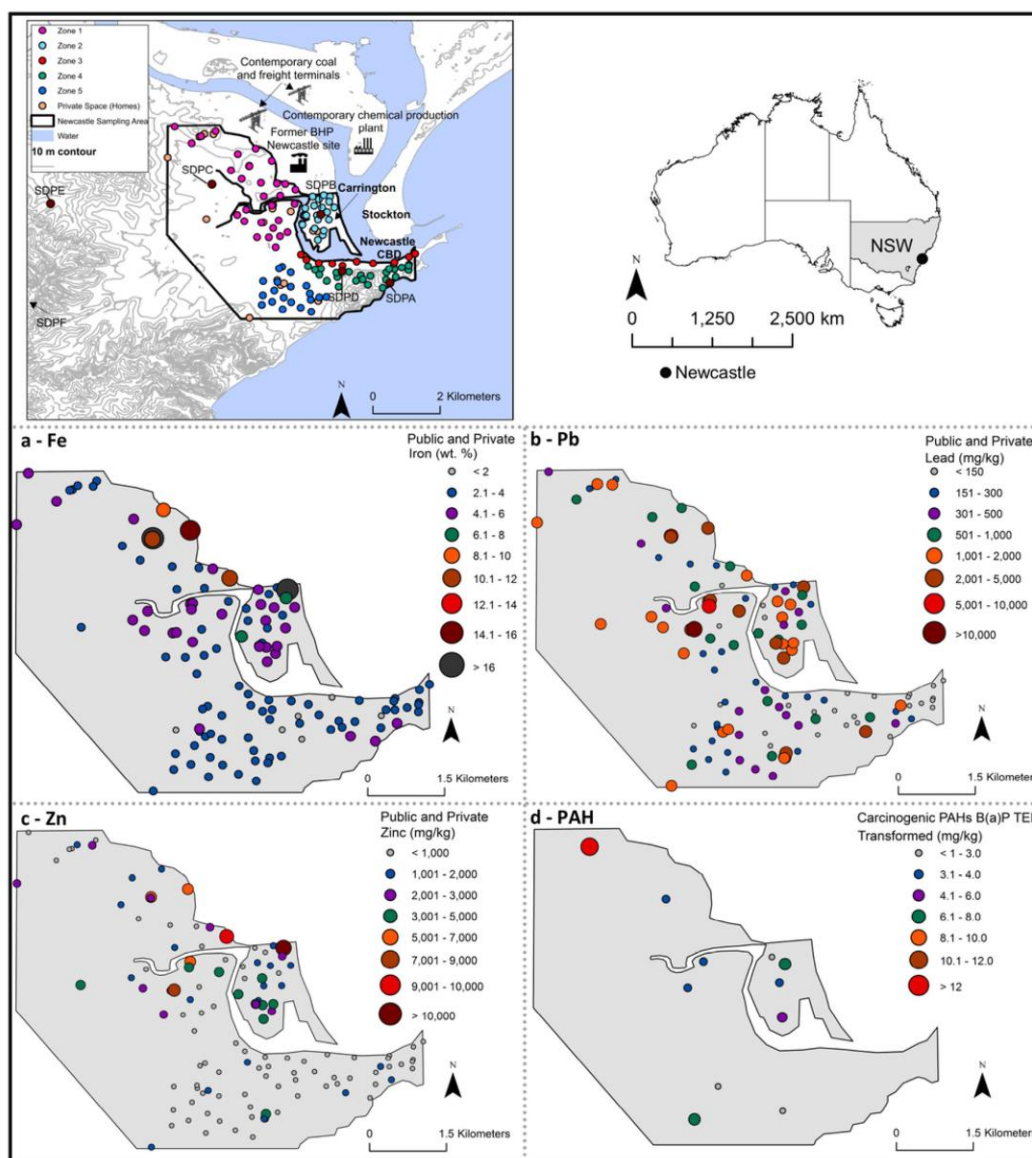


Fig. 1. Soil analyte concentrations within Newcastle city (Australia) (panel a - iron, b - lead, c - zinc, d - TEF transformed PAHs). Site marked SDP- are soil depth profiles.

pany (SMH, 1871b), the English and Australian Copper Company (EACC) smelter in 1867 (SM, 1872) and the Sulphide Corporation Pb smelter at nearby Boolaroo in 1897 (Dames and Moore, 1994). The port and shipping facilities of Newcastle quickly became strategic industrial infrastructure as Pb sulfide ore was shipped through the Port of Newcastle to Boolaroo (Quorn Mercury, 1900a; NMH, 1902). In 1907 the Melbourne and Great Northern Smelter also joined the copper smelting operations at Newcastle (MDM, 1907; SCT&WA, 1907). The Broken Hill Proprietary (BHP) Newcastle Steel Mill and subsidiaries were the industrial focus of Newcastle from 1915 until their closure in 1999 (Parliament of New South Wales, 1999). At peak operation, the facilities employed ~ 12,000 people (Parliament of New South Wales, 1999; BHP Co. LTD, 1949). The BHP operation became an industrial giant with the addition of, and partnership with, two other large scale manufacturers of steel products (Parliament of New South Wales, 1999; BHP Co. LTD, 1949;

NS, 1925). Today, coal and freight shipping terminals and an ammonium nitrate facility dominate the industrial activities of the city.

Despite the industrial heritage of Newcastle, there has been no publicly accessible wide-scale geochemical assessment of soils in the city area. This study investigates soil metal(loid)s and polycyclic aromatic hydrocarbons to help close this knowledge gap and highlight potential environmental contamination risks associated with living in an industrial city.

2. Methods

2.1. Soil sampling

2.1.1. Soil metal(loid) concentrations

Newcastle city was divided into five sampling areas (Fig. 1) to reflect the dominant land use types covering open spaces, retail and in-

dustrial (Zone 1), residential (Zone 2), rehabilitated industrial land along the foreshore now used for high density residential (Zone 3), Central Business District (CBD) (Zone 4) and low to medium density residential (Zone 5). Soils collected in public spaces ($n = 103$) were sampled from roadside verges, parklands and other accessible locations. Soils collected from private spaces ($n = 67$ from 23 homes) were obtained through the Macquarie University VegeSafe soil metal testing program (Rouillon et al., 2016), where Newcastle community members submitted samples for analysis. Samples were also received from outside of the specified target area but were included here to provide additional detail to the investigation. Public space samples were collected at the surface (0–2 cm) in accordance with the standard methods (Taylor et al., 2010). A number of control sites ($n = 6$) with pits dug to a defined depth of 40 cm irrespective of soil horizons were sampled throughout the city to determine local background soil concentrations.

2.1.2. Slag deposits

A slag waste deposit was identified outcropping on a beach adjacent to a children's playground in Stockton (Fig. 1). Large chunks of a black glass-like slag and a ferrous slag material were collected using a rock hammer to separate them from the consolidated mass. This deposit has been documented in the local media as one of many civil works sites in the city where BHP-derived material was used as stabilization fill (NMH, 1948a; NMH, 1948b; NMH, 1949a; NMH, 1949b; NS, 1926; NS, 1948).

2.1.3. Soil polycyclic aromatic hydrocarbon concentrations

Coal, ferrous metal smelting and fossil fuels present significant sources of environmental PAH contamination (Albuquerque et al., 2016; Yang et al., 2016; Odabasi et al., 2010). To further compliment the metal(loid) analysis, a preliminary investigation of soil PAH concentrations was conducted. Soil samples for polycyclic aromatic hydrocarbon (PAH) analysis were collected from 12 locations around the city (Fig. 1d). Soils were collected in clean glass bottles with a Teflon lid and stored at $< 4^\circ\text{C}$ until analysis.

2.2. Laboratory analysis

2.2.1. Soil metal(loid) concentrations

Soil samples ($n = 170$) were oven dried at 40°C for 72 h, then sieved to $< 180\ \mu\text{m}$. This fraction was selected for analysis because the research evidence shows that finer soil fractions are susceptible to re-suspension and subsequent ingestion or inhalation (Horowitz, 1991; Youn et al., 2016). Approximately 10 g of the $< 180\ \mu\text{m}$ sample was packed in 35 mm open ended PANalytical XRF cups using 3.6 μm Chemplex Mylar X-ray film for analysis. An Olympus Premium Innov-X XRF analyzer (pXRF) fitted with a 50 kV, 4 W Ta anode X-ray tube and a silicon drift detector was used for the measurement of Ti, Cr, Mn, Fe, Cu, Zn, As, Cd and Pb concentrations in surface and subsurface soils. Analysis was conducted at Macquarie University. Recommended operational procedures were followed, including: daily measurements of an energy calibration check, measurements of a silicate (SiO_2) blank at the beginning and end of analysis to ensure no instrument contamination had occurred, and measurements of NIST certified reference materials (CRMs) conducted every 25 samples to monitor instrument performance (Table 1). Soil samples and CRMs were placed on the measurement window and analyzed using the proprietary soil mode at 60 s per measurement condition (180 s total measurement). A matrix-matched calibration was applied to optimize the pXRF in the measurement of metal-contaminated silicate-based soils (Rouillon and Taylor, 2016).

Table 1

Summary of the limits of detection, analytical precision, sample homogeneity and recoveries for pXRF.

	Ti	Cr	Mn	Fe	Cu	Zn	As	Cd	Pb
pXRF									
Limit of detection	^c	8	9	^c	4	5	3	2	3
Analytical precision ^a	< 1	3	< 1	< 1	2	< 1	7	9	2
(%)									
Analytical precision ^b	< 1	1	< 1	< 1	1	< 1	7	12	< 1
(%)									
Sample homogeneity	2	5	1	< 1	2	< 1	7	10	1
(%)									
Mean CRM recovery	101	102	102	100	108	99	106	103	99
(%)									

^a Relative standard deviation of reference material suite.

^b Relative standard deviation of soil samples.

^c Limits of reporting were not calculated for Ti and Fe as no CRM values between 3 and 10 times the expected manufacturer detection limit were present in the CRM suite.

Instrument detection limits, analytical precision, sample homogeneity and recoveries of each element are presented in Table 1. Analytical precision was determined by the relative standard deviation (RSD) of samples ($n = 18$) analyzed in triplicate (Table 1), while soil ($n = 7$) homogeneity (RSD) was measured by triplicate preparation and analysis.

2.2.2. In-vitro Pb bio-accessibility

Spatial diverse soil samples with a range of total Pb concentrations ($n = 12$) were digested using the US EPA (2012) method for in-vitro Pb bio-accessibility on the $< 180\ \mu\text{m}$ soil fraction (Horowitz, 1991; Youn et al., 2016). Soil ($\sim 1\ \text{g}$) was digested in 100 ml 0.4 M glycine in de-ionized water adjusted to pH 1.5 using hydrochloric acid (HCl, Reagent Grade, Sigma Aldrich) and analyzed for total Pb on the National Measurement Institute's (North Ryde, Sydney) Agilent 7900 ICP-MS, with an LOR of 0.5 mg/kg. All blanks were $< 0.5\ \text{mg/kg}$ and laboratory control spikes were between 90 and 130%. Relative percent difference for replicate analysis ($n = 2$) was $< 20\%$. Sample duplicate analysis ($n = 1$) had an RSD $< 20\%$.

2.2.3. Soil Pb isotopic compositions

To understand potential sources of soil metal contaminants, Pb isotopic compositions were determined for soils with a range of concentrations and locations, slag samples and reference ore specimens (Broken Hill and Wallaroo mine galena) ($n = 18$). Broken Hill galena was used as an end-member due to its prolific use in commercial and industrial commodities (Gulson et al., 2004). Wallaroo galena was used as a reference specimen as the copper ore processed at the Wallaroo Copper Company smelter was often co-associated with galena deposits as impurities in the host orebody. Samples were extracted using an aqua regia (HNO_3/HCl) digestion to mobilize the labile Pb bound to the soil matrix (Ayuso et al., 2013; Civitillo et al., 2016; Das et al., 2016; Gallego et al., 2016). Following digestion, the samples were centrifuged and the supernatant filtered using a 0.45 μm Minisart syringe filter. Soils were analyzed on an Agilent 7900 ICP-MS at the National Measurement Institute following the method and instrument conditions detailed previously in Kristensen and Taylor (2016). Measurement values were collected for ^{202}Hg , ^{204}Pb , ^{206}Pb , ^{207}Pb and ^{208}Pb and then corrected for variation based on blank values and the effect of ^{202}Hg on ^{204}Pb . Samples were bracketed during analysis with NIST 981 SRM (certified values: $^{206}\text{Pb}/^{204}\text{Pb}$: 16.94, $^{206}\text{Pb}/^{207}\text{Pb}$: 1.09, $^{208}\text{Pb}/^{207}\text{Pb}$: 2.37). This method for Pb isotope ratio quantification reports $^{204}\text{Pb}/^{206}\text{Pb}$: 0.0590 ± 0.0005 , $^{207}\text{Pb}/^{206}\text{Pb}$: 0.915 ± 0.004 and $^{208}\text{Pb}/^{206}\text{Pb}$: 2.168 ± 0.009 as a maximum uncertainty for NIST 981 (Kristensen et al., 2016).

2.2.4. Soil mineralogy

To explore the mineralogy of contaminants in public and private soils ($n = 9$), heavy mineral separation was conducted on the $< 180 \mu\text{m}$ sieved fraction for soils from a range of metal(loid) concentrations and locations around the city, following established methods (Ettler et al., 2016).

The phase composition of the samples ($n = 9$) was assessed by X-ray powder diffraction analysis (XRPD) using a PANalytical X'Pert Pro diffractometer with an X'Celerator detector (PANalytical, the Netherlands) (analytical conditions: $\text{CuK}\alpha$ radiation at 40 kV and 30 mA, 2 θ range 2–80°, step 0.02°, counting time 150 s per step). The XRPD pattern was analyzed using X'Pert HighScore Plus 3.0 software coupled to the Crystallography Open Database (COD) (Grazulis et al., 2012). Relative abundances of phases were estimated from XRPD patterns using the relative intensity ratio (RIR) method.

Heavy mineral fractions, prepared as polished sections, were examined under a Leica DM LP polarizing microscope (Leica, Germany) followed by an automated scanning electron microscope (TIMA 3LM, TESCAN Integrated Mineral Analyzer, Czech Republic) operating at 25 keV using a 'dot-mapping' mode method (dot spacing of 2 μm). The integrated images were then used to identify metal(loid)-bearing particles and their distribution.

Particles, from two representative samples from private land with the highest metal(loid) content, were examined using a scanning electron microscope (SEM; TESCAN VEGA3 XM, Czech Republic operating at 20 kV) equipped with an energy dispersion spectrometer (EDS; Quantax 200 X-Flash 5010, Bruker, Germany).

2.2.5. Soil polycyclic aromatic hydrocarbon concentrations

Approximately 1 g of the $< 180 \mu\text{m}$ fraction was mixed with anhydrous sodium sulfate ($\text{Na}_2\text{O}_4\text{S}$, Reagent Grade, Sigma Aldrich). The sample was then extracted in 1:1 dichloromethane (CH_2Cl_2 , Reagent Grade, Sigma Aldrich) and acetone ($\text{C}_3\text{H}_6\text{O}$, Reagent Grade, Sigma Aldrich) and then concentrated prior to analysis on an Agilent 5975 gas chromatography mass selective detector (GC-MSD) at the National Measurement Institute, North Ryde. The instrument was operated in selected ion monitoring mode for 15 PAHs (naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)&(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-cd)pyrene, dibenzo(a,h)anthracene and benzo(g,h,i)perylene). All PAHs have a LOR of 0.5 mg/kg except for benzo(b)&(k)fluoranthene which has an LOR of 1 mg/kg. Laboratory blanks contained $< \text{LOR}$ for each analyte and laboratory control spikes were 94–130% for all analytes.

2.3. Data analysis

Inorganic elemental concentrations were benchmarked against Australian Health Investigation Levels for residential soils (HIL-A) ((National Environment Protection Measure (NEPM), 2013), Table 2). The NEPM (2013) identifies eight carcinogenic PAHs categories (reported here as seven PAHs categories as benzo(b)&(k)fluoranthene were not separated in the analysis method): benzo(a)anthracene, chrysene, benzo(b)&(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-cd)pyrene, dibenzo(a,h)anthracene and benzo(g,h,i)perylene. The toxicity of these eight PAHs are weighted using a toxicity equivalence factor (TEF) relative to the most toxic PAH benzo(a)pyrene, the HIL for the sum of these weighted PAHs is 3 mg/kg (Table 3).

Table 2

Newcastle city (Australia) soil element concentrations (mg/kg) (pXRF derived) for public spaces in addition to median soil element concentrations for each sampling zone. Complete dataset is presented in Supplementary data 2.

	Ti	Cr	Mn	Fe ^a	Cu	Zn	As	Cd
Private spaces								
n detected	67	67	67	67	67	67	67	11
Minimum	2520	54	120	1.31	44	140	8	3
25th percentile	4090	96	600	2.38	130	740	21	4
Median	5180	128	790	3.36	180	1190	35	5
75th percentile	6210	179	1240	4.50	300	2070	63	6
Maximum	12,200	2400	3600	16.0	1040	8570	283	9
Mean	5160	190	1020	4.00	252	1690	56	5
NEPM (2013)	No	100	3000	No	7000	8000	100	20
HIL-A	value	(VI)		value				
n > guideline	—	^b	2	—	0	1	8	0
% > guideline	—	^b	4%	—	0	2%	12%	0
Public spaces								
n detected	103	100	100	103	103	103	103	13
Minimum	1670	38	180	1.36	34	96	5	3
25th percentile	3870	76	600	2.43	70	270	12	4
Median	4660	92	920	3.00	110	550	22	6
75th percentile	5390	140	1540	4.11	240	1230	44	11
Maximum	14,760	710	4490	17.1	1170	1.2 ^a	380	16
Mean	4850	120	1230	3.67	180	1120	37	7
NEPM (2013)	No	100	3000	No	7000	8000	100	20
HIL-A	value	(VI)		value				
n > guideline	—	^b	8	—	0	2	5	0
% > guideline	—	^b	8%	—	0%	2%	5%	0%
Median per zone (public spaces)								
Zone 1 (n = 29)	4650	140	1450	3.99%	170	980	34	6
Zone 2 (n = 19)	5130	140	1810	4.34%	330	1860	44	6
Zone 3 (n = 10)	4490	85	750	2.75%	100	340	12	< LOD
Zone 4 (n = 25)	5120	92	670	2.65%	74	270	15	3
Zone 5 (n = 20)	4240	77	730	2.56%	78	410	16	< LOD

— Cannot be calculated due to the absence of a NEPM (2013) HIL.

^a Concentration presented as wt%.

^b Indicates that the soil NEPM (NEPM) HIL is based on element species rather than total concentration, this analysis was not conducted in this study and cannot be calculated.

3. Results and discussion

3.1. Soil metal(loid) concentrations

Newcastle was divided into five soil sampling zones to characterize different land uses across the city (Fig. 1). Within these zones, public and private space surface soils were analyzed. Summary surface soil metal(loid) concentrations are presented in Fig. 1 and Table 2, with the complete data set provided in Supplementary data.

Public space surface soils – The greatest concentrations of Fe, Pb and Zn were detected in the Zone 1 and Zone 2 sampling areas (Fig. 1, Table 2). Soil Fe concentrations were detected $> 17 \text{ wt}\%$ in soils of the Zone 2 sampling area (Fig. 1, Table 2). Soil Pb concentrations were detected at a maximum of 4650 mg/kg (median: 280 mg/kg, $n = 103$) with the greatest median Pb concentrations in Zone 2 (median: 900 mg/kg, $n = 19$) followed by Zone 1 (median: 500 mg/kg, $n = 29$). Zinc concentrations were detected at a maximum of 1.2 wt% in Zone 1 (Fig. 1). The Zone 2 sampling area had the greatest median

Table 3

Soil toxicity equivalence factor (TEF) transformed PAH concentrations for Newcastle city (Australia) surface soil samples.

Sample	x co-ordinate	y co-ordinate	TEF PAH 1	TEF PAH 2	TEF PAH 3	TEF PAH 4	TEF PAH 5	TEF PAH 6	TEF PAH 7	Σ TEF PAH
Zone 5 S1	151.765942	-32.935172	0.08	0.0068	0.14	1.0	0.0061	***	0.0076	1.2
Zone 5 S4	151.751481	-32.936583	0.35	0.035	0.69	5.2	0.029	0.57	0.034	6.9
Zone 5 S5	151.755347	-32.931136	0.15	0.015	0.31	2.2	0.014	***	0.016	2.7
Zone 2 S1	151.765858	-32.91974	0.24	0.021	0.45	3.3	0.02	***	0.024	4.1
Zone 2 S2	151.765405	-32.914048	0.20	0.020	0.42	2.7	0.019	***	0.022	3.4
Zone 2 S2**	151.765405	-32.914048	0.23	0.022	0.45	3.2	0.020	***	0.024	3.9
Zone 2 S3	151.764169	-32.909803	0.061	0.006	0.15	0.96	0.0072	***	0.0089	1.2
Zone 2 S4	151.766314	-32.910977	0.40	0.038	0.73	5.4	0.031	0.70	0.035	7.3
Zone 1 S5	151.746816	-32.900268	0.20	0.020	0.44	2.9	0.020	***	0.025	3.6
Zone 1 S2	151.750378	-32.914897	0.18	0.020	0.43	2.8	0.018	***	0.020	3.5
Zone 1 S4	151.752958	-32.910561	0.17	0.018	0.37	2.7	0.016	***	0.020	3.3
Zone 1 S7	151.734307	-32.891691	0.73	0.075	1.4	9.8	0.061	1.4	0.076	13.5

TEF PAH: 1 - Benz(a)anthracene, 2 - Chrysene, 3 - Benzo(b)&(k)fluoranthene, 4 - Benzo(a)pyrene, 5 - Indeno(1,2,3-cd)pyrene, 6 - Dibenzo(a,h)anthracene, 7 - Benzo(g,h,i)perylene. All concentrations in mg/kg.

Concentrations in red exceed the NEPM (2013) HIL for TEF transformed PAH (3 mg/kg).

Cells denoted with *** cannot be calculated as the non-transformed data was < LOR.

Sample marked with ** is a field duplicate.

concentration of soil Zn (1860 mg/kg) (Fig. 1, Table 2). All elements of interest (Table 2), except Ti, had the highest median concentration in Zones 1 and 2. Titanium (n = 170; median: 4830 mg/kg; max: 14,760 mg/kg) is predominantly a geogenic element and had no clear spatial concentration pattern, however, concentration spikes were detected across the city.

Private (residential) space surface soils – Surface soil samples collected from private gardens were markedly more contaminated than those in public spaces (Table 2). Soil Fe was detected with a maximum of 16 wt% in Zone 1 with a median across all sample zones of 3.36 wt% (n = 67). Soil Pb was detected up to a maximum concentration of 1.16 wt% (Zone 1, Fig. 1) (median: 1140 mg/kg, n = 67). The maximum Zn concentration detected (8570 mg/kg) was in Zone 1 (Fig. 1, Table 2). Private soils contained a median Zn concentration of 1190 mg/kg. Over 90% of homes exceeded the 300 mg/kg Australian soil Pb HIL-A (National Environment Protection Measure (NEPM), 2013), with As, Mn and Zn exceeding their respective HIL-A values at 30%, 9% and 4% of homes (Table 2). Soil samples were not collected from private spaces in Zone 3 and 4 where high-density housing and ground surface cover limited access to soil.

Soil metal(loid) concentrations from depth profiles in Newcastle are presented in Fig. 2 and in Supplementary data 1. Subsurface soils collected from depth profiles within the urban areas (profiles a–d, Fig. 1) showed that metal(loid) concentrations did not necessarily decrease with depth (Fig. 2, panels a–d). Soil metal(loid) concentrations remain at ~ 100 mg/kg for Mn, Pb and Zn at 30 cm in depth profile a, while all other depth profiles (b–c, Fig. 2) have concentrations > 100 mg/kg for all metal(loid)s. Soil samples from 2 to 4 cm at depth profiles b and d (Fig. 2) increase in all metal(loid) concentrations from the surface (0–2 cm). Soil depth profile b had Pb concentrations > HIL-A of 300 mg/kg (National Environment Protection Measure (NEPM), 2013); until > 10 cm depth. Soil depth profile c showed an increase in soil metal(loid) concentration to depth, with Mn concentrations (1430 mg/kg) at 30 cm depth double surface values (700 mg/kg) (Fig. 2). Soil Pb concentrations in profile 3 also remained above the HIL-A (300 mg/kg (National Environment Protection Measure (NEPM), 2013)) at 30 cm depth. Other metal(loid)s not presented in Fig. 2, but reported in Supplementary data 1, show that concentrations also increase with depth (e.g. Fe: 2.29 wt% at surface; 3.79 wt% at 30 cm depth) implying that profile c likely occupies a brownfield site now repurposed as a parkland. By contrast two additional soil depth profiles from remnant bushland

(Figs. 1 and 2, e–f) showed a clear decrease in metal(loid) concentrations with depth. These data are comparable to uncontaminated soil concentrations at depth from similar studies of industrial cities (e.g. Rouillon et al., 2013, Mn: ~ 100 mg/kg; Pb: < 5 mg/kg; Zn: ~ 30 mg/kg). The data show that surface soil metal(loid)s from Newcastle have been enriched significantly by anthropogenic source(s).

3.2. Potential source(s) of inorganic soil contaminants

It is well documented that copper smelter waste, and more recently, steel smelter waste was used prolifically around the Newcastle city area as landfill and stabilization material (NMH, 1948a; NMH, 1948b; NMH, 1949b; NS, 1926; NMH, 1925; The Australasian, 1905; SA, 1951; SMH, 1948; TNC, 1950). Much of this material may have contained Broken Hill Pb ore due to the movement of this ore through the Newcastle Ports from BHP to the Sulphide Company smelter (later the Pasmenco Cockle Creek Smelter) at Boolaroo (Quorn Mercury, 1900a; Petersburg Times, 1901; PPR, 1904). The Sulphide Company also contributed large volumes of smelter waste for capital works (Gulson et al., 2004; Morrison, 2003; Morrison and Gulson, 2007; Morrison et al., 2016).

This study's bulk mineralogical investigation of soils showed that high-temperature silicates (clinopyroxenes, olivines and glass) typically originating from smelting industry, as well as smelter-derived oxides (spinel) were present in the majority of the samples (Supplementary data 4). All the samples contained geogenic/pedogenic heavy minerals such as zircon, Ti-, Fe- and Al-bearing oxides in addition to traces of quartz and other geogenic minerals. No metal(loid)-bearing phases were detected by XRPD indicating that contaminants are bound in trace phases in the soil materials (Supplementary data 4).

Fe and Mn oxides were the most common metal-bearing phases in soils, with Fe oxides containing high Pb levels (up to 27.3 wt% PbO) (Fig. 3b, c, e, f). The occurrence of smelter-derived slag-like particles, possibly windblown from slag dumps (Fig. 3a, c, e, f, i) and rounded particles characteristic of quenched melt droplets from the smelter emissions (Fig. 3b) implies they are derived from smelting emissions. Metal-bearing droplets in slag particles corresponded to Cu sulfides; slag silicates and glass also containing Cu and Zn are characteristic of Cu smelting waste materials (Ettler et al., 2009; Ettler et al., 2016). By contrast, mine-derived sulfides (commonly identified with weathering rims) were relatively rare in the studied soils (Fig. 3d). Lead phosphates (Fig. 3f, h, i), arsenates (Fig. 3f) and

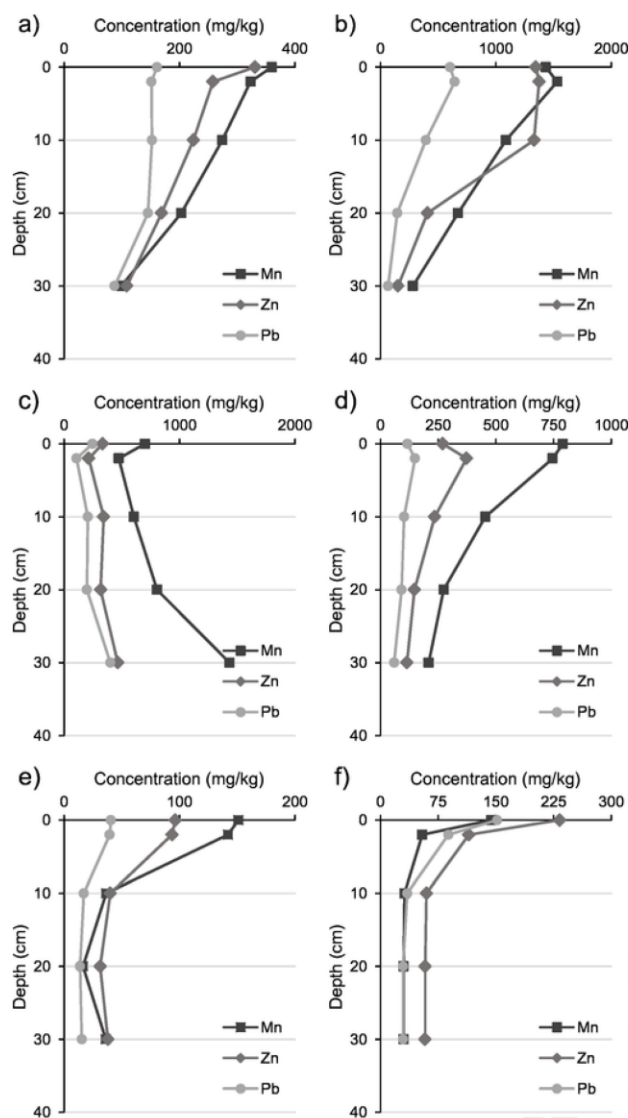


Fig. 2. Soil depth profiles from urban area (a–d) and remnant bushland (e–f) in Newcastle city (Australia) displaying Mn, Pb and Zn soil concentrations. Complete data and site locations are provided in Supplementary data 1.

sulfates (Fig. 3h) were common. Some of these form solid solutions (mimetite – pyromorphite; Fig. 3h) or are associated to similar phases (anglesite – barite) (Fig. 3h). Given that these phases are relatively insoluble, the Pb release from these compounds in aqueous environments would be minimal. Rare particles of Sn oxides were also observed (Fig. 3f) with weathering rims enriched in metal(oids) including Pb, Sb and Zn. Interestingly, metals (mainly Pb and Zn) were found in Fe and Mn oxides formed as weathering products on particles (e.g. Fig. 3b, c), indicating contaminants can be absorbed from surrounding soil during the formation of secondary Fe–Mn oxides.

Lead isotopic compositions were used in this study to delineate potential contaminant sources. The Pb isotopic compositions of soils and reference samples (galena and slag) are presented in Fig. 4 and Supplementary data 3. Surface soils from public and private spaces ($n = 8$) have similar loosely clustered Pb isotopic compositions (range: $^{208}\text{Pb}/^{207}\text{Pb}$: 2.345–2.411, $^{206}\text{Pb}/^{207}\text{Pb}$: 1.068–1.312). Lead isotopic compositions of soils collected from depth profiles ($n = 3$)

within the urban area (profiles b, c, d; Fig. 1) fall within the range of Pb isotopic compositions of surface soils in public and private spaces (range: $^{208}\text{Pb}/^{207}\text{Pb}$: 2.370–2.406, $^{206}\text{Pb}/^{207}\text{Pb}$: 1.093–1.234). Lead isotopic compositions from soil collected from the depth profile in the remnant bushland (profile e; Fig. 1; $n = 1$) are more radiogenic and outside of the range of urban surface soils ($^{208}\text{Pb}/^{207}\text{Pb}$: 2.489, $^{206}\text{Pb}/^{207}\text{Pb}$: 1.198) (Fig. 4, Supplementary data 3). Lead isotopic compositions for the two slag samples reveal divergent Pb isotopic compositions. A black glassy material similar to non-ferrous smelter slag as identified in other studies (Morrison and Gulson, 2007; Piatak et al., 2015) was less radiogenic than surface soils ($^{208}\text{Pb}/^{207}\text{Pb}$: 2.319, $^{206}\text{Pb}/^{207}\text{Pb}$: 1.044) (Fig. 4, Supplementary data 3). A second slag deposit that was more similar to ferrous smelter slag as identified in other studies (Piatak et al., 2015) contained more radiogenic Pb than surface and depth profile soils ($^{208}\text{Pb}/^{207}\text{Pb}$: 2.543, $^{206}\text{Pb}/^{207}\text{Pb}$: 1.318) (Fig. 4, Supplementary data 3). The Wallaroo Mine copper ore that was smelted at the site prior to Newcastle BHP was co-associated with galena Pb ore ($^{208}\text{Pb}/^{207}\text{Pb}$: 2.640, $^{206}\text{Pb}/^{207}\text{Pb}$: 1.477), which has more radiogenic Pb isotopic compositions compared to bushland depth profile soil ($^{208}\text{Pb}/^{207}\text{Pb}$: 2.489, $^{206}\text{Pb}/^{207}\text{Pb}$: 1.198) and slag (Fig. 4, Supplementary data 3). Consequently, we used Wallaroo galena Pb isotopic compositions as an end-member for source apportionment analysis. Although the Pb bearing copper ore associated with the Wallaroo Mine was smelted in large volumes during the early industrial era of Newcastle it appears to have had little influence on soil Pb isotopic compositions (Fig. 4, Supplementary data 3). The other Pb isotopic composition end-member used is the less radiogenic Broken Hill ore ($^{208}\text{Pb}/^{207}\text{Pb}$: 2.319, $^{206}\text{Pb}/^{207}\text{Pb}$: 1.044) (Fig. 4, Supplementary data 3).

The Pb isotopic compositions (Fig. 4, Supplementary data 3) showed an enrichment of public and private space soils (Fig. 4) by the distinct Broken Hill Pb isotope signature ($^{208}\text{Pb}/^{207}\text{Pb}$: 2.319, $^{206}\text{Pb}/^{207}\text{Pb}$: 1.044). Enrichment of public and private soils less radiogenic Pb sources suggests an introduction of Broken Hill type ore bearing material (e.g. black glassy slag waste and fill material) into the Newcastle city environment. Leaded petrol and paint are also potential contributors to Pb in the surface soils of the urban environment (Datko-Williams et al., 2014; Kristensen, 2015; Laidlaw et al., 2012; Mielke et al., 1983; Mielke and Reagan, 1998; Mitchell et al., 2014; Zahran et al., 2013). The Pb isotopic ratios of the soils derived from ICP-MS do not allow discrimination between these tightly clustered sources (Fig. 4). However, despite this limitation, the data show clearly that Pb sources match closely the Broken Hill ore signature in urban soils.

3.3. Soil polycyclic aromatic hydrocarbon concentrations

Soil PAH concentrations are presented in Fig. 1 and Supplementary data 2. Soil samples ($n = 12$) contained 12 of the 15 target PAH compounds (Supplementary data 2). The carcinogenic compound benzo(a)pyrene was detected in all samples with the maximum concentration in Zone 1 (9.8 mg/kg; Fig. 1). Nine samples exceeded the Australian HIL-A guideline (3 mg/kg) for benzo(a)pyrene TEF transformed PAHs (Table 3, Fig. 1, max - 13.5 mg/kg). The maximum TEF transformed PAH concentration (13.5 mg/kg) was detected in Zone 1 sampling area.

3.4. Potential health impacts of soil contamination

Soils in Newcastle are contaminated by a range of metal(oid)s and PAHs. While there is strong evidence to link the soil metal(oid) concentrations to the legacy industrial activities of the city, the source

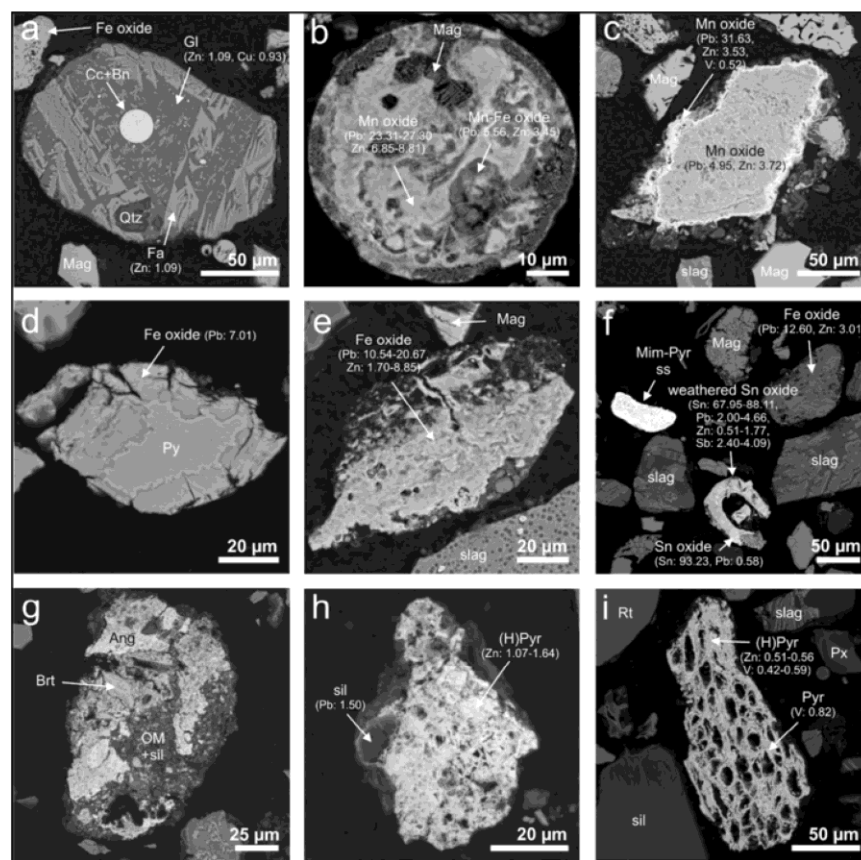


Fig. 3. Scanning electron micrographs in back-scattered electrons (BSE) of representative metal(loid)-bearing particles from two samples (PRS3D and PRS4C from Newcastle city soils (Australia)). Complete data available in Supplementary data 5. a) Smelter slag particle composed of Fe-olivine (fayalite), metal-bearing glass and symplectitic inclusions of chalcocite and bornite (PRS3D); b) Slag-like particle originating probably from smelter dust emissions composed of magnetite sub-micrometric dendrites and newly-formed Mn and Mn-Fe oxides resulting from alteration process in soil (PRS4C); c) Mn oxide particle particularly rich in metals associated with slag particles and magnetite grains (PRS3D); d) Weathered pyrite grain with Fe-oxide rim (PRS3D); e) Metal-bearing Fe oxide associated with slag and magnetite particles (PRS3D); f) Mimetite-pyromorphite solid solution (ss) associated to weathered Sn-oxide and Fe-oxides particles, slag and magnetite grains (PRS3D); g) Complex intergrowth of barite and anglesite with organic matter and unidentified silicate material (PRS3D); h) (hydroxyl)pyromorphite grain associated with unidentified Pb-bearing silicate (probably slag glass) (PRS3D); i) Sponge-like particle composed of hydroxylpyromorphite and pyromorphite associated to rutile and smelter-derived silicates (slag) (PRS4C). The metal(loid) concentrations obtained by EDS are expressed in wt% of oxides. Abbreviations: Ang – anglesite (PbSO_4), Bn – bornite (Cu_5FeS_4), Brt – barite (BaSO_4), Cc – chalcocite (Cu_2S), Fa – fayalite (Fe_2SiO_4), Gl – glass (slag), (H)Pyr – hydroxypyromorphite ($\text{Pb}_5(\text{PO}_4)_3\text{OH}$), Mag – magnetite (Fe_3O_4), Mim – mimetite ($\text{Pb}_3(\text{AsO}_4)_3\text{Cl}$), OM – organic matter, Px – pyroxene ($(\text{Ca,Fe,Mg})_2\text{SiO}_6$), Py – pyrite (FeS_2), Pyr – pyromorphite ($\text{Pb}_5(\text{PO}_4)_3\text{Cl}$), Qtz – quartz (SiO_2), Sil – unidentified silicate; Rt – rutile (TiO_2).

of PAHs in Newcastle is unclear. Steel manufacturing (Yang et al., 2002; Song et al., 2015; Khaparde et al., 2016), coal transport (Wang et al., 2016; Huang et al., 2013), and incomplete combustion of motor vehicle emissions (Jedrychowski et al., 2015; Perer et al., 2006) are known to contribute to PAHs to the soil environment. Regardless of the source, recent studies have demonstrated a link between pre-natal and early life exposure to PAHs (Huang et al., 2013) and a range of cancers (Perer et al., 2006; Mordukhovich et al., 2016; Lemieux et al., 2015; Zhang et al., 2016). Exposure to PAHs in the Newcastle soil environment, combined with the metal(loid) contaminants is likely to generate a significant burden of disease.

Blood Pb (and other metals) analysis of the Newcastle population would be one determinant of exposure to contaminated soils in the City. Unfortunately, no such data exists. Given the paucity of blood Pb (and other metals) data, surrogate indicators are used here to predict the potential health impacts of Pb contaminated soils in the city. Our in-vitro bio-accessibility analysis ($n = 11$) shows that the Pb found in soils would be subject to absorption following ingestion of soils from the study area (Table 4). This is not surprising given that the Pb-bearing Fe- and Mn-oxides identified in Fig. 3b and c are

known to be susceptible to dissolution under acidic conditions. Moreover, the in-vitro bio-accessible Pb concentrations indicate a potential burden of disease associated with Newcastle's soil Pb concentrations, which could be better quantified with a childhood blood Pb survey in Newcastle city linked to participant's home soil and dust metal analysis.

3.5. Managing environmental contaminants in Newcastle city

Some information is available to landowners about the potential risk of environmental contamination in Newcastle, but it is not extensive and it is often site specific. In addition to sites being listed on the NSW Environment Protection Authority's (NSW EPA) list of notified contaminated sites (NSW EPA, 2016) some properties in Carrington have a certificate issued under Section 149 of the *Environmental Planning and Assessment Act 1979* (NSW) advising purchasers:

Land history information indicates that the subject land may be within an area which was once low lying and may have been filled. Limited investigation indicates that the filling material may

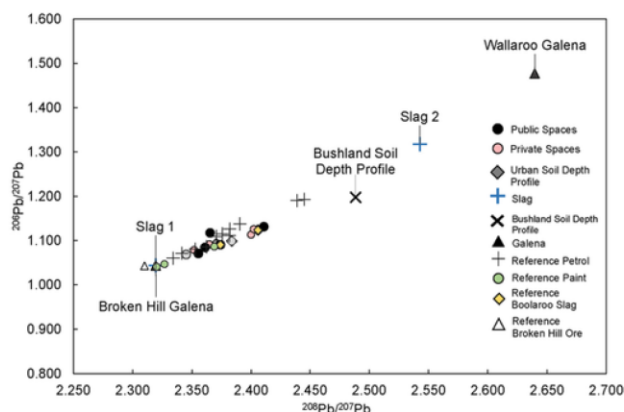


Fig. 4. Lead isotopic compositions of Newcastle city (Australia) soils and slag and galena from Broken Hill and Wallaroo Mines analyzed in this study. Standard deviation from repeat analysis (10 analyses) of one sample: $^{208}\text{Pb}/^{207}\text{Pb}$: 0.004, $^{206}\text{Pb}/^{207}\text{Pb}$: 0.004. Comparison is made to reference Pb isotopic compositions from the nearby Sulphide Corporation's Boolaroo smelter slag (Gulson et al., 2004), Broken Hill ore galena (Gulson, 1984), petrol and paint values (Gulson et al., 1982; Laidlaw et al., 2014).

Table 4
In-vitro bio-accessible soil Pb concentrations.

Sample	In-vitro bio-accessible Pb (mg/kg)	Total Pb (mg/kg)	% bio-accessible ^a
PRS10B	5000	7640	65
PRS7A	732	2830	26
PRS3D	1670	3420	49
Zone 1 S7	1730	1550	112
Zone 2 S17	1410	2770	51
PRS8C	282	1060	27
Zone 1 S20	972	2060	47
Zone 1 S9	607	4650	13
PRS4C	5190	11,570	45
Zone 2 S4	1350	2550	53
Zone 2 S4 ^b	1340	2550	52
NIST2586 (500 mg/kg)	156		31
BLK	< 0.5	-	-
CAL-24 (10 ppm)	10	-	-

NIST2586 is a Pb paint contaminated soil used to assess the extraction efficiency of the in-vitro bio-accessibility method.

BLK = analysis blank.

CAL-24 = 10 ppm 12-element standard reference material used during IPC-MS analysis.

- not applicable.

^a Calculated from pXRF total.

^b Denotes field duplicate sample.

contain ballast and industrial slag which contains some heavy metal including Pb. The Council does not hold information which allows it to say whether or not the subject land contains such filling material and purchasers should make their own enquiries in this respect. If the land does contain such filling material this should be taken into account in the use and the development of the property. Soil sampling and remediation may be required for the further development of the land.

Newcastle City Council (NCC) (1997)

To address the absence of site-specific contamination information, soil metal screening programs such as VegeSafe (Rouillon et al., 2016) could be employed across the city to help inform homeowners of extant soil metal(loid) contamination on their property and the po-

tential associated risks. Such information can be used by homeowners to initiate mitigation strategies, including: engagement of professional trained contractors to remediate soils where appropriate; using raised garden beds for food growing; removing and replacing soils in areas regularly visited by children and pets.

3.6. Industry, smelting and development in global urban cities - environmental implications

This study demonstrates the extensive environmental contamination legacy that can be created by a long history of manufacturing, processing and smelting along with inappropriate waste disposal in urban environments. In 2011 it was estimated that around 30% of the global feedstock for ferrous ore processing was discarded generating ~ 550 million tons of waste slag (Van Oss, 2011). The waste material produced from manufacturing industries, typically rich in a range of environmental contaminants, is often discarded haphazardly in the environment, leading to problematic brownfield sites, requiring costly environmental clean-up (Gallego et al., 2016).

4. Conclusions

This study details a significant potential health risk associated with environmental metal(loid) and polycyclic aromatic hydrocarbon contamination of soils in the industrial city of Newcastle, Australia. Surface soils collected from public (roadside verges and parks) and private (homes) spaces ($n = 170$) show that a range of metal(loid)s are present in the soils above their relevant Australian Health Investigation Level (HIL) concentrations. Lead was the most common contaminant detected in the city soils with a median concentration of 1140 mg/kg in private space soils. Deep soil metal(loid) concentrations across the city were also elevated indicating contamination was laterally and vertically pervasive. In-vitro bio-accessibility analysis of Pb in soils (maximum Pb concentration: 5190 mg/kg; 45% Pb bio-accessibility) revealed these were readily available to the gastric system. Soil Pb isotopic compositions (range: $^{208}\text{Pb}/^{207}\text{Pb}$: 2.345–2.411, $^{206}\text{Pb}/^{207}\text{Pb}$: 1.068–1.312) show that the anthropogenic contamination was sourced from a low radiogenic source, with a Broken Hill type Pb orebody ($^{208}\text{Pb}/^{207}\text{Pb}$: 2.319, $^{206}\text{Pb}/^{207}\text{Pb}$: 1.044) being the most likely origin. Examination of soil mineralogy, particularly the rounded quenched droplet particles extracted during heavy mineral separation (Fig. 3), indicates that Pb and soil metal(loid) contaminants are derived predominantly from smelter emissions and industrial waste slag. An additional burden of contamination in the city soils comes from elevated levels of polycyclic aromatic hydrocarbons compounds, typically associated with coal, ferrous metal smelting and fossil fuels, with 75% of samples ($n = 12$) analyzed exceeding the Australian HIL-A guideline for benzo(a)pyrene.

Source assessment of the environmental contamination indicates that historic and legacy copper and steel smelting industries in the city have contributed substantially to the contamination of soils. This study highlights the health risks faced by communities residing in industrial locations and demonstrates the 'need to know' so that individuals can protect themselves for unnecessary and potential toxic exposures.

Acknowledgements

P.J. Harvey is funded by a Macquarie University Research Excellence Scholarship (MQRES) (2012195) associated with an Australian Research Council Future Fellowship awarded to H.K. Handley (FT120100440). M. Rouillon is funded by an Australian

Postgraduate Award. C. Dong is funded by a Macquarie University Research Excellence Scholarship (iMQRES No. 2014098). Support from the Czech Science Foundation (project no. 13-17501S) is greatly acknowledged. Ernest Ameyaw is thanked for fieldwork and laboratory assistance. The authors thank Tony Morrison and Brian Gulson (Macquarie University) for comments on data interpretation. We would like to thank our colleagues from Charles University and the Geological Institute of the Czech Academy of Science, who helped with the data acquisition and treatment: Petr Drahotka (XRPD), Tomáš Hrstka (TIMA) and Zuzana Korbelová (SEM/EDS).

Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.scitotenv.2017.01.053>.

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5 Discussion

This thesis identifies selected contemporary Australian environmental contamination risks across three spatial scales of magnitude: home environments, agriculture systems and across towns and cities. The approach used demonstrates that there is a significant extant risk of environmental exposures and that management of contamination and pollution incidence at all scales is required to mitigate potential environmental and health related impacts. While the research studies detailed in this thesis are discrete investigations in themselves, they illustrate a broader theme covering a number of over-arching themes that include the:

- prevalence and impact of contemporary and legacy environmental contamination in Australia;
- novel techniques and approaches for identifying the source(s) of environmental contamination.
- success and failure of environmental contamination intervention, mitigation and management, and;

Each of these three themes, along with other key outcomes of this thesis, are examined in further detail in the discussion below.

5.1 The Prevalence and Impact of Contemporary and Legacy Environmental Contamination in Australia

Widespread, pervasive and persistent are words that can be used to describe global environmental contamination. Environmental contaminants have become so ubiquitous that they impact every person on the planet (Lodeiro et al., 2016). Indeed ‘widespread’ (defined by the Macquarie Dictionary (2016) as: (1) spread over or occupying a wide space, (2) distributed over a wide region, or occurring in many places or among many persons or individuals) is used in the title of two papers in this thesis to reinforce that ubiquity. Environmental contamination occurs on a range of scales from localised to much broader regional, national and global scales, and the associated impacts are similarly diverse (Wong et al., 2006). One of the key themes carried through each of the case studies in this thesis is the pervasiveness of legacy and contemporary sources of environmental contamination and the health risks associated with exposure to those contaminants in an Australia perspective. Even though Australia’s European

history is short, this thesis shows that Australia is not immune to environmental contamination, with many instances of environmental contamination arising over many decades. These environmental contamination problems are often unknown or overlooked by the community and relevant regulators (e.g. Paper 2), or alternatively, some problems are ignored in the hope that they will just go away (e.g. Paper 6). The first case study, Paper 1, identifies legacy contamination sources that were contributing to contemporary contamination of drinking water in north-east Tasmania (TAS). This study shows that there was a legacy drinking water lead contamination issue arising from the old degraded supply pipeline infrastructure, built originally to service the tin mining works in the town of Pioneer. Due to the very poor structural condition of the pipeline, there was significant dissolution of metal(loid)s into the drinking water. This pipeline is believed to have been an ongoing source of metal(loid)s in the drinking water for many years, however, this source was ignored by the utility and regulators in their investigations in favour of the spurious but regularly used ‘naturally occurring’ explanation (GHD 2013a, 2013b). The argument of naturally occurring contamination is typically used to obfuscate the true source of environmental contamination in an attempt to divert liability and avoid potential prosecution (Kristensen and Taylor, 2016).

In addition to the contamination sources from the aged pipeline infrastructure in the study location, the data showed that drinking water lead contamination was also occurring from domestic plumbing fixtures and fittings. Plumbing fixtures and fittings, as a contemporary source of drinking water lead (and copper) contamination was further investigated in Paper 2 by analysis of homes from across regional NSW. This study demonstrated that fixtures and fittings were contributing lead to the drinking water. Remarkably, the same generic issue was identified more than two decades earlier by Gulson et al. (1994). Gulson et al.’s (1994) study of Sydney homes showed that Australian plumbing fittings and fixtures contribute to metal(loid)s in drinking water. Moreover, recent international research also concurs with the findings in this thesis by identifying a link between drinking water lead concentrations and the manufacturing material of plumbing products (Edwards et al., 2009, Triantafyllidou and Edwards, 2011, Triantafyllidou et al., 2007). The data presented in Paper 1 and 2 show that the requirements of modern manufacturing standards in Australia are inadequate and have not progressed with the research literature pertaining to human health and environmental contamination. It is important to note here that despite extensive public interest in the outcomes of Paper 1 and 2 in the Australian media and other avenues (Appendix 2.2), regulatory response to this problem of persistent and preventable drinking water metal contamination arising from fixtures and fittings remains unaddressed.

Another example of ongoing, unresolved poor environmental management practice and persistent off-site contamination is described in Paper 5. This study examines contamination associated with a chicken slaughterhouse that has been in the spotlight of environmental regulators for more than four decades. Despite its extended and persistent history of environmental violations and the related investigations by environmental regulators Paper 5 demonstrated unequivocally that the facility was continuing to pollute the environment as it has always done: with contaminated wastewater discharges, which are in breach of its the operating licence.

Recognising and understanding the source(s) of either legacy of contemporary environmental contamination, including its longevity, has implications for the way that contamination is managed and cleaned-up (Wu et al., 2013). Environmental contamination events that are contemporary, have a long history, encompass multiple home environments (Paper 3) or are city-wide (Papers 6 and 7) are typically considered too costly to clean-up comprehensively (Coulon et al., 2016, Naidu, 2013). Subsequently the highest exposure risk areas are targeted to maximise the cost-benefit of clean-up works (Paper 6) (Coulon et al., 2016). If the contamination is derived from a specific isolated legacy source, without further contemporary contributions (Papers 3, 6 and 7), it is possible to do the clean-up just once. If the contamination source is both legacy and contemporary, clean-up likely fails unless the source(s) of ongoing contamination are also curtailed (Paper 5).

5.2 Novel Techniques and Approaches for Identifying the Source(s) of Environmental Contamination

Critical to this thesis is the implementation of the forensic ‘multiple lines of evidence’ approach for determining the source(s) of environmental contamination. This approach seeks to apply the best and most appropriate techniques in, among other disciplines, geochemistry, soil and water science and biology. To highlight this, Paper 5, examining effluent discharge from a chicken slaughterhouse facility, utilised a range of water chemistry techniques to first confirm there was a pollution loading in the waterway. The study then applied arsenic speciation of the water samples to characterise any arsenic fingerprint in the water arising from organo-arsenicals in chicken feed. The detection of arsenic in waters discharging from the facility and not those in adjacent catchments added weight to the hypothesis that the chicken farm was contributing waste water to the catchment. The final stage in this ‘multiple lines of evidence’ approach was to examine the genetic composition of the waters. This lead to the identification of chicken DNA and an anthropogenic marker in the water. By adopting this ‘multiple lines of evidence’

approach, the source(s) of the effluent in the catchment waters became clear and irrefutable. In a pollution and contamination prevention scenario, accurate source identification is paramount as it strengthens the enforcement power of regulators and informs the most appropriate contamination mitigation and clean-up strategies. This was demonstrated by Gulson et al. (2012) who utilised a range of environmental tracers to establish that the source of drinking water lead contamination was the shipping terminal in the Australia coastal town of Esperance, which triggered a large scale clean-up event in that town. This study further adds to this in Paper 8, where smelting and heavy industry have contributed to environmental contamination of an entire city by a range of metal(loid)s and organic compounds. The ‘multiple lines of evidence’ approach, where soils were examined using X-ray fluorescence and identification of source(s) was achieved through lead isotope quantification and soil mineralogy, was used to identify and differentiate all the possible source(s) of environmental contamination in the city and determine the exposure risk through bio-accessibility. From a management and mitigation perspective this is important because it identifies legacy pollution derived from both ferrous and non-ferrous smelting as the primary sources of the contemporary environmental contamination and quantifies the associated health exposure risk. This ultimately has ramifications for the types of clean-up strategies available, as discussed in Paper 6.

5.3 The Success and Failure of Environmental Contamination Intervention, Mitigation and Management

Operating licence requirements for large industrial and mining activities often mandate that an environmental bond is paid prior to commencement of a polluting activity, to be used in the event of extensive environmental contamination arising from the operation (Harris et al., 2009, Naidu, 2013). This stipulation does not apply to older legacy contamination issues that pre-date these requirements. In Western Australia the legislation has moved away from a site-specific bond for the mining industry to payment into a Mining Rehabilitation Fund to better distribute the funding as required following cessation of operations (Gorey et al., 2014). The Lead Abatement Strategy (LAS) in Paper 6 is an example of a legacy environmental contamination strategy that did not have an environmental bond or central funding body. The estimated massive cost (no figure has ever been publically disclosed) of the failed clean-up, funded primarily by the PCCS Administrators highlights the need for effective regulation and management of polluting industries to prevent the contamination of the surrounding environment.

Managing environmental contamination is a delicate balance between cost-benefit, responding to the needs of commercial stakeholders, mitigating negative health outcomes and managing the community perception of the event and clean-up process (O'Faircheallaigh, 2010, Plant et al., 2016). In north-eastern Tasmania (Paper 1) the poor management of drinking water contamination incidence(s) across all of these spheres was executed in a manner that was publicly detrimental for TasWater, the state's water utility. The utility's actions to identify the source and resolve the matter of drinking water lead contamination was heavily criticised by the local community who felt that their needs were being neglected and ignored (Figure 5.4 and Figure 5.4). Instead of dedicating efforts to resolving the contamination problem, the focus of TasWater was negative, combative, dismissive and often strategic to silence what were referred to as 'fringe voices' (Appendix 1.3). The report commissioned by TasWater and compiled by researchers from Water Research Australia (Appendix 1.2) was a clear effort to discredit the peer reviewed outcomes of Paper 1. This attempt to discredit the research and its authors was subsequently shown in documents obtained by the Australian Broadcasting Corporation under a Freedom of Information request (e.g. Appendix 1.3). This approach of targeting the scientist, rather than resolving the contamination problem is a recurring issue in Australian environmental contamination management. For example, Kristensen and Taylor (2016) who have shown that, particularly with respect to the mining industry, the primary aim of the polluters is to discredit the research by suggesting all manner of other possibilities including that the identified contamination is due to a naturally occurring source through to direct attacks on the authors themselves. The approach of attacking the scientist rather than the science is prolific in environmental contamination research and is common in the lead industry (Markowitz and Rosner, 2000, Markowitz and Rosner, 2013), in the debate and forced retraction of work surrounding the carcinogenic effects of the herbicide glyphosate (Séralini et al., 2012) and even in the contaminated drinking water crisis in Flint Michigan (Edwards and Pruden, 2016). This mentality means that the resolution time for environmental contamination events is significantly increased, generating prolonged exposure time and increased health risks. Ultimately communities become disillusioned with the capacity of the environmental managers to resolve the actual problem, in part because they are often blind-sided by spurious and misleading information (Conway and Oreskes, 2012, Taylor et al., 2014a, Taylor et al., 2015a, Taylor et al., 2015b).



Figure 5.4 Signage erected around the town of Pioneer illustrating the heightened emotions of the community, and their anger towards TasWater for its delayed response to drinking water contamination.



Figure 5.5 Community expression of its perceptions of the management approaches to environmental contamination. The example shown is from Pioneer, Tasmania that is discussed in Paper 1.

5.4 Prevention is Better than a Cure

This thesis draws together a number of studies where considered foresight would have prevented a substantial environmental contamination event and an ensuing public health burden. For example, the examination of drinking waters in both Tasmania (Paper 1) and New

South Wales (Paper 2) demonstrate that, although plumbing is a widely documented contamination source in the global literature, this remains overlooked (or ignored) as a potential source of contamination in Australia. By overlooking this contamination source, there is a subsequent oversight generated in public health.

The adage that ‘prevention is better than a cure’ is synonymous with the medical profession. The idea that mitigating risks and using forward planning to prevent an illness occurring in the first instance has resulted in the global introduction of vaccines and the eradication of diseases such as polio and more recently measles in the Americas (Dali, 2017, Graham et al., 2012, WHO, 2013). One of the reasons why this philosophy is so widely applied in medicine is the cost-benefit of prevention over finding a cure (Reyes et al., 2017). Nevertheless, the field of management and mitigation of environmental contamination has been slow to reach the same realisation as the medical profession. This is again particularly evident in the history and use of lead-containing products. In 1904, an Australia physician reported a link between lead-bearing white paints used on hard surfaces in homes and childhood lead poisoning (Lockhart-Gibson 1904), it was not until 1970’s that the addition of lead to paint was finally banned in Australia (Kristensen, 2015). Philippe Grandjean, a prominent contributor to the research of environmental contaminants, comments on the slow response to the science surrounding lead exposure and the toxicological effects (Grandjean, 2013):

“We realized the danger at an inexcusable delay that has resulted in enormous costs in terms of brain toxicity. How we got lured into accepting gigantic amounts of lead pollution reveals naiveté and negligence, at least in retrospect, along with devious misinformation”

But once again, lead is not the only substance to which a huge disparity between identification of the problem and implementing an action applies. This is exemplified in the widespread use and detection in the environment of perfluorinated chemicals (e.g. PFOS/PFOA) (Lindim et al., 2016, Taylor and Cosenza, 2016a, Taylor and Cosenza, 2016b). These chemicals, known to be persistent, bioaccumulative and toxic have recently been banned by many international government agencies and industry bodies (Taylor and Cosenza, 2016b). During the late 1980’s, the chemical manufacturer DuPont was adding PFOA to their Teflon non-stick products at their Washington Works site. Internal managerial documents from that time showed that compared to the expected male incidence, based on long term monitoring, there was a 2-fold increase in incidence of bladder cancer and a 3-fold increase in each of mouth and throat cancers, multiple myeloma, and urinary tract cancers (Appendix 7.9). These critical health statistics indicating that there is a substantial causal link between these chemicals and human health outcomes were withheld by the company, and this was later revealed in a 2016 lawsuit against DuPont (NY

Times, 2016). The debate surrounding the carcinogenic nature of these compounds and the 'safe' level of exposure has intensified (Arrieta-Cortes et al., 2017, Danish EPA 201, Lindstrom et al., 2011, US EPA 2014), and recent lawsuits arising from individuals that have developed an array of cancers following exposure to PFOA that was discharged into drinking water at the DuPont Parkersburg West Virginia plant have resulted in the award of substantial punitive damages (USA District Court, Southern District of Ohio case MDL 2433— IN RE: E. I. du Pont de Nemours and Company C-8 Personal Injury Litigation).

In Australia, off-site migration of the firefighting foam containing PFOS and PFOA from the Williamstown Royal Australian Airforce Base in NSW, 200 km north of Sydney has also directed significant attention to the contaminants (Taylor and Cosenza, 2016a, Taylor and Cosenza, 2016b). The release led to the destruction of grazing lands, aquaculture and contamination of groundwater (Taylor and Cosenza, 2016). The release of these chemicals had been known for many years with very minimal attempt made to clean up the problem (Taylor and Cosenza, 2016a, Taylor and Cosenza, 2016b). Given the debate surrounding the toxicity of these substances, one would argue that prevention of the product entering the environment is more beneficial than remedying any contamination or human health outcomes.

5.5 How Do Communities Protect Themselves?

Often with environmental contamination, the burden is thrust upon the individual to resolve the problem themselves (Gostin, 2016, Leech et al., 2016). While some communities lack knowledge about how to clean-up environmental contaminants, many communities are simply too poor to self-help (Taylor, 2014). Many communities that have no financial security are also those identified as having the greatest health concerns such as the highest blood lead concentrations, the highest incidence of cancer, and increased rate of peri-natal and infant mortality (Briggs, 2003, Lessard et al., 2016). Some communities are fortunate to receive funding for large scale remediation activities, but this often comes after many years of campaigning and exposure to environmental contaminants (Boreland and Lyle, 2006, Gulson et al., 2012, Tirima et al., 2016)

In Australia, there is a legislative framework that separates the power related to environmental matters into federal, state and local council jurisdictions. The state and local council jurisdictions are delegated the role of the majority of contaminated land issues, while the federal jurisdiction applies to land owned by the Australian Commonwealth government or where it is an issue of national interest.

In NSW, where the majority of the studies in this these were conducted, most large-scale environmental contamination events are managed by the Contaminated Sites section of the NSW Environmental Protection Authority (EPA). In order for a site to be listed as an notified contaminated site by the NSW EPA, a section 60 notification pursuant to the *Contaminated Land Management Act 1997* (NSW) must be made to the EPA by the land owner or the person responsible for the contamination, based on the belief that the site may potentially be contaminated. Additionally, the EPA may be notified of a site via other means including assessments conducted by local councils. The total number of contaminated sites in Australia has not been confidently quantified. In NSW, the EPA estimated over 7,000 sites to be contaminated, in 1996, that estimate had increased to approximately 60,000 sites, in 2014 the estimate had decreased 30,000 and by 2016, the number of contaminated sites notified to the NSW EPA was 1617 (Hehir, 2014, Legislative Assembly of NSW, 2016, Smith, 1996 Taylor and Cosenza, 2016).

At the local council level in NSW, contaminated sites are often identified and included on a house title deed as a s149 clause under the *Environmental Planning and Assessment Act 1979* (NSW). Often, homeowners are unaware of the contamination status of their land parcel unless they purchase a s149(2) or s149(5) certificate. A s149(2) certificate informs of land zoning, regulatory controls and contamination status, whereas the s149 (5) certificate informs of regulatory comments and other significant site information. Both of these certificates must be specifically requested by the landowner or purchaser from the relevant local government authority and usually costs between AUD\$50 and \$100 for processing. A vendor under a contract for the sale of land is legally obliged to attach a s149 certificate to the contract before it is signed by or on behalf of the purchaser. This issue is also covered under s52A *Conveyancing Act 1919* (NSW) and *Conveyancing (Sale of Land) Regulation 2010* (NSW) An example of such a notification from the island of Carrington, near Newcastle, NSW, where heavy metal contaminated fill has been identified tells the residents (NCC 1997):

*“Land history information indicates that the subject land may be within an area which was once low lying and may have been filled. Limited investigation indicates that the filling material may contain ballast and industrial slag which contains some heavy metals including lead. The Council does not hold information which allows it to say whether or not the subject land contains filling material and **purchasers should make their own enquiries in that respect.** If the land does contain such filling materials this should be taken into account in the use and the*

development of the property. Soil sampling and remediation may be required for the further development of the land” (bold emphasis added)

The recommendation that “purchasers should make their own enquires”, in conjunction with the examples presented in this thesis, highlights the inadequacy of the current practices in identification and management of contaminated lands in Australia. Indeed, on its face it is contradictory to the legal requirements of the *Conveyancing Act 1919* (NSW) and *Conveyancing (Sale of Land) Regulation 2010* (NSW) along with the Duty to Notify (EPA, 2016). The inference is that landowners, lay-people, are expected to have an awareness and often comprehensive knowledge of environmental contamination problems and management approaches in order to protect themselves and their family from exposure to these contaminants. Conversations with the community undertaken during completion of this thesis revealed that while the community are interested to know about potential risks of harm they are poorly informed about likely contamination sources and its consequences. This is where a program such as VegeSafe (Paper 3) and the sampling approach in the NSW regional water study (Paper 2) are beneficial for the community. These programs help to engage the community, provide free or low cost environmental analysis and offer cost-effective solutions for those people at risk. The ultimate aim is to inform and empower the community on environmental contamination issues. In this sense, the approach applied via the VegeSafe program has received strong community support similar to other international programs (e.g. the Safe Urban Gardening Initiative, Filipelli 2016).

5.6 The Role of Citizen Science and Community Participation in Scientific Research

Citizen science and community participation are instrumental in many of the studies in this thesis. By engaging individuals and the community with the various aspects of the scientific process, they become more alert to the topics studied and more responsive to the conclusions reached in those studies. Paper 3 describes the VegeSafe initiative which provided the citizen science participation model adopted and adapted in other case studies throughout this thesis (Papers 1, 2, 6 and 8). In each of these case studies, members of the community were invited to engage in the data collection phase by either providing samples, promoting the research activity in their local community or more broadly via the media, providing sample site metadata or contributing to the sample collection design. In many instances, participants submitted samples relevant to themselves, and were informed of results relating specifically to those samples.

Interaction with the community and engaging participants through this citizen science approach allowed for the collection of samples that would be otherwise off-limits to a researcher. Specifically, the citizen science approach provided unprecedented access to soil and kitchen tap water in private homes. This was valuable in Papers 2, 3 and 8 where the research questions were focussed around the exposure health risks to individuals in their everyday life. The ability to access soils in private homes in Paper 8 demonstrated that there is a substantially greater health exposure risk associated with soils in the private spaces than in the public spaces.

When engaging community members through citizen science it is important to feed-back the information and research lessons to community members. This allows the community to continue to remain informed and be a part of the research process in an ongoing, practical sense. Many of the case studies in this thesis adopted this philosophy. A number of community engagement events were held to communicate preliminary (Figure 5.6) and final findings (Figure 5.7) to the stakeholder communities. While some of these events were met with hostility by some stakeholders (predominantly those responsible for the pollution or its clean-up), there was broad appreciation from the community that researchers were dismantling the typical disconnect between academic science and the community.



Figure 5.6 Engaging the community of Narrabri, NSW, with drinking water sampling.



Figure 5.7 Communicating science back to the community in an accessible format is essential for engaging citizen scientists. Example show is a community meeting in Pioneer, Tasmania following Paper 1. A recording of the meeting can be viewed at: <http://www.abc.net.au/7.30/content/2015/s4222652.htm>

While there are limitations associated with citizen science, including among others, the potential for samples and metadata to be erroneously collected, the addition of more variables in sampling method as samples are collected by multiple people and the potential for only impacted individuals to respond to and engage in the activity, citizen science has becoming widely applied globally to encourage community engagement with the world around them and promote learning opportunities (Ellwood et al., 2016, Jordan et al., 2016, Le Bourgeois et al., 2016). Understanding and recognising the limitations of citizen science and accounting for those limitations (Paper 2 and Paper 3) allows for efficient, robust and reliable data collection over a much larger sampling area (Dickinson et al., 2010).

5.7 Contaminants on a Global Scale

As populations grow, the demand and strain on resources increases. Urban environments expand and the capacity for the environment to respond to the demands placed on it by humans falters (Fuller et al., 2015). The World Health Organisation estimates that approximately 12.6 million deaths occur each year as a result of environmental factors, with many of these factors associated with environmental pollution and contamination (WHO, 2016, Figure 5.8). The vast majority of these deaths occur in the low- to middle-income nations (Figure 5.8). Some of the key areas where there is global pressure on the environment through contamination include

drinking water, artisanal consumer goods production, artisanal mining and large-scale manufacturing (Figure 5.9).

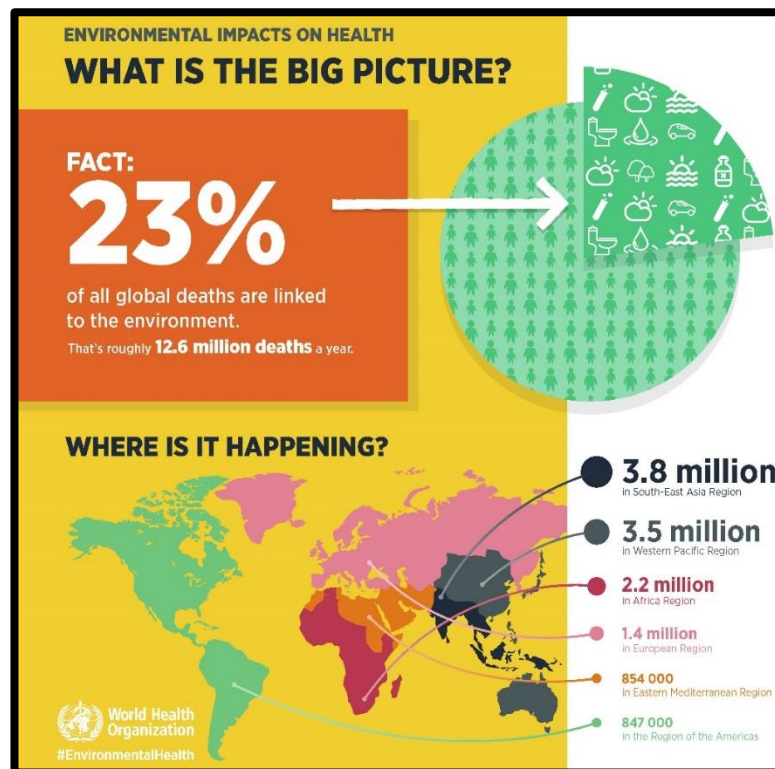


Figure 5.8 Environmental contamination is not a local problem, it impacts every person in the world.

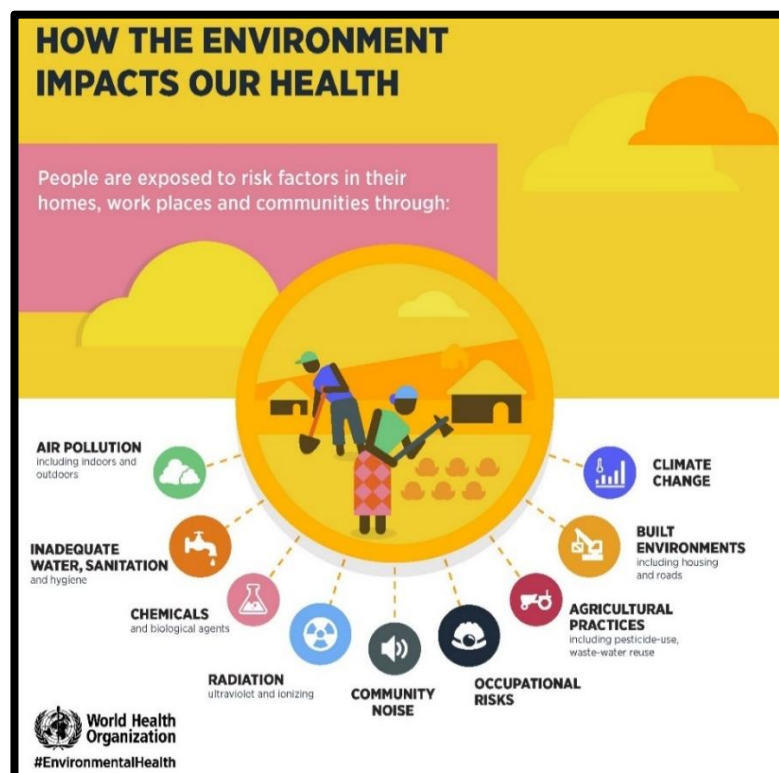


Figure 5.9 Environmental contamination has far-reaching impacts on human health. To combat this, we need to solve the environmental contamination problems on the local scale.

Drinking water contamination, one of those identified sources in Figure 5.9, has recently re-appeared as a major environmental contamination challenge. In higher income nations, lead and copper pose the greatest threats to health, with a startling number of reticulated supplies and schools recently identified as having lead contaminated drinking water (Deshommes et al., 2016, Katner et al., 2016). Drinking water lead research has returned to fashion and this has stemmed from the identification of lead contaminated drinking water in the city of Flint Michigan where drinking water lead concentrations regularly exceeded 1,000 $\mu\text{g/L}$ which when combined with soil lead contamination, generated a public health emergency (Hanna-Attisha et al., 2015, Laidlaw et al., 2016). Metal(loid) contamination of drinking water is only the beginning of the problem. Bacteria, pathogens, radiation and other contaminants are placing strain on global drinking water supplies. The WHO Disability Adjusted Life Years (DALY) index of children <5 years for drinking water (Figure 5.10) shows that globally, there is a massive number of life years subtracted from an expected healthy lifespan, resulting from drinking water contamination. Much of this relates to low- and middle-income countries with some of the key areas of concern including Bangladesh where arsenic contaminates groundwater (Gardner et al., 2011, Kippler et al., 2016, Smith et al., 2000), north Africa where chemicals from cultural crafts contaminate rivers historically used for drinking water in medinas (Figure 5.11) and central Africa where bacteriological contaminants dominate (Kirby et al., 2016). This global environmental management challenge is set to intensify as global climate changes, leading to reduced rainfall in these areas and subsequently fewer clean and safe sources of water. This problem is emerging, and the sections in this thesis that examine water contamination in Australia, particularly Paper 5, are setting the scene for future research on this topic in Australia.

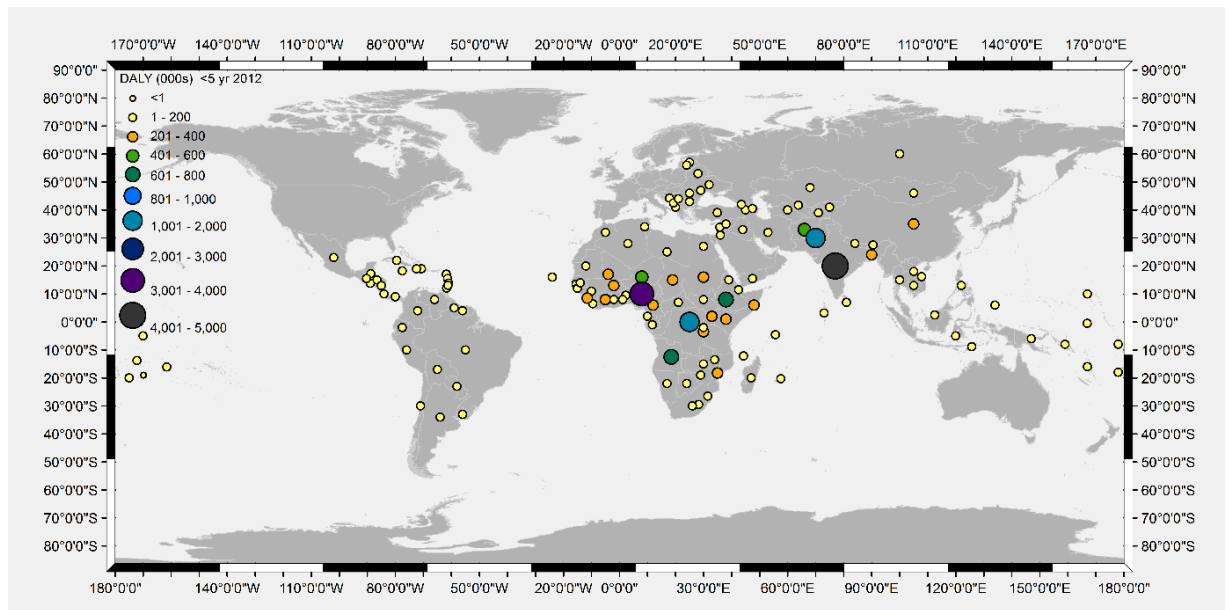


Figure 5.10 The total global distribution of Disability Adjusted Life Years (DALYs), or the years lost from a healthy life span, for children <5 yr in 2012, as an indicator for the global burden of disease from contaminated/polluted water. Notably, the greatest DALY values occur in the low- to middle-income countries.



Figure 5.11 Fez tannery, a source of legacy contamination in the Fez River, Morocco and environmental contamination of the Fez River, Morocco. River remediation projects are working to improve the conditions of this environment.

5.8 Future Contaminants and the Need to Learn from Our Mistakes

Safe and healthy living environments in a world where urbanisation is rapidly changing the global landscape will be progressively rarer in the future unless action is taken now to

effectively and efficiently manage contemporary and legacy contamination exposure sources. Carson (1962) again put it best when she wrote:

“The human race is challenged more than ever before to demonstrate our mastery, not over nature but of ourselves”

Fifty-four years later, we are faced with the same broad issues - an immeasurable challenge of mitigating the environmental and human health risks of environmental pollution and contamination. We are also faced with new challenges. In the July 2016, the United States Toxic Substances Control Act Chemical Substance Inventory contained >67,000 entries for non-confidential toxic chemical substances (US EPA, 2016). Many of these substances have received little to no toxicological assessment so their environmental and health impacts are relatively unknown (Gavrilescu et al., 2015, Wasi et al., 2013). These substances permeate every part of life, from fabrics and textiles to food additives (Bellinger, 2011, Gavrilescu et al., 2015, Grandjean and Clapp, 2014, Lanphear et al., 2005).

In addition to these types of potential environmental contaminants, the shift in the global manufacturing economy to high-tech electronics is creating a demand for previously un-utilised metal(loid)s including rare-earth elements (Chassé et al., 2017). Lithium, used in lithium-ion (Li-ion) batteries for smartphones and other electronics has become a high demand commodity. As large companies such as TESLA, which is developing high-current applications for Li-ion batteries in homes, industry and automobiles, develop their lithium-ion products and demand more ore products, there is an increasing market value for the ore (Figure 5.12). This thesis examines the impacts of ‘traditional’ metal(loid) mining and demonstrates that there is a long lasting legacy from poorly managed extraction and processing of ores. Despite its use in psychiatric treatments the toxicological impacts of lithium are poorly understood, however, studies are beginning to suggest the existence of a tolerance threshold, and exposure in concentrations above which chronic health impacts occur (McKnight et al., 2017, Saad et al., 2017, Thakur et al., 2003). Mining and ore processing typically expose workers and communities to concentrations of environmental contaminants above normal population exposure concentrations (Papers 6 and 8) (Munksgaard et al., 2010, Taylor et al., 2010, Taylor et al., 2014b, Taylor and Schniering, 2010). It is therefore essential that the mistakes of past mining practices, where large urban areas are contaminated by processes stemming from poor environmental regulations, are not repeated with a more contemporary contaminant source such as lithium.

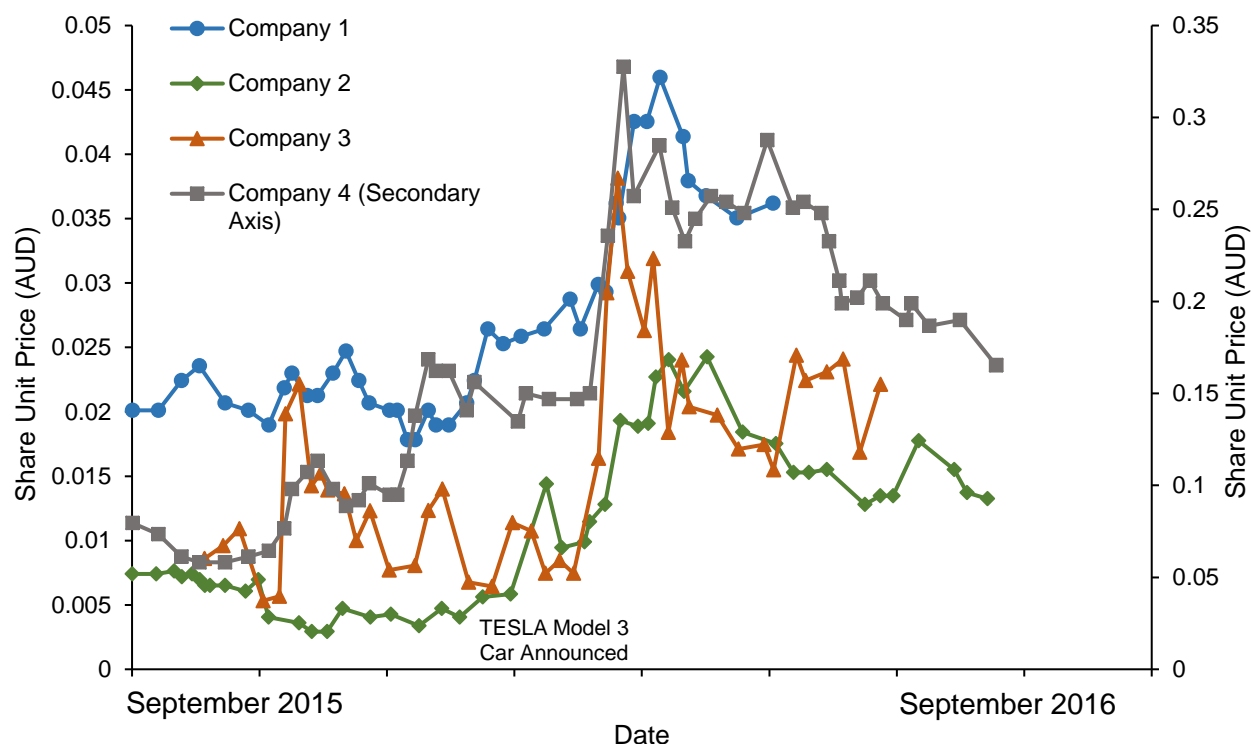


Figure 5.12 Trends in the Australian Stock Exchange share unit price of lithium mining operations prior and post the announcement of the TESLA Model 3 car which utilises lithium ion cells as a power source.

Finally, biological contaminants are also becoming a growing environmental concern with the release of antibiotics into the global environment posing a global threat to ecosystems and human health (Paulson et al., 2016, Rusu et al., 2015). The formation of antibiotic resistance genes in organisms triggered by human and agricultural waste disposal in the environment (e.g. Paper 5) has been identified as one of the most significant environmental contamination hurdles for the near future (Gillings et al., 2008, Li et al., 2016, Wyrsh et al., 2016). Consideration of the successes and failures of past management techniques and lessons learned (Taylor et al., 2011) will assist in the development of robust management plans to mitigate these foreseen future challenges.

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6

Conclusions

Environmental contamination is ubiquitous, pervasive and persistent. It impacts every ecosystem and every person on the planet and with increasing global pressures such as urbanisation, climate change and demand for resources, it is set to become more prevalent in everyday life. This thesis demonstrates, through a series of case studies, that Australia is not resistant to the perils of environmental contamination. Indeed, Australia has many of the environmental contamination problems seen throughout the rest of the world. Environmental contamination does not know jurisdictional boundaries or political borders, it impacts everybody and all environments.

The work presented in this thesis draws together some unique and unrecognised environmental contamination incidents in Australia. By doing this, it has brought to the attention of the national community some of the risk associated with environmental contamination exposure, the successes and failures of environmental contamination management and has provided insights into better ways to detect and manage contamination. The application of multi-disciplinary multiple lines of evidence techniques to sourcing environmental contamination has provided environmental regulators and managers with new insight into how to approach regulatory challenges where a more unique investigatory method may be required. This has had considerable implications in Australia where environmental regulators have already begun utilising some of these techniques.

The role of citizen science in academic investigations has proved to be a valuable tool in this thesis. It is demonstrated that citizen science and engaging the community with research not only empowers the community by providing otherwise inaccessible information, it helps the researcher access sites and data that would otherwise be inaccessible to them.

The real-world quantifiable impacts of this thesis on environmental management and human health can already be observed. In Tasmania, residents of the rural town have received tanks as an alternative safe and clean drinking water source, with a plan to install a new pipeline to the region in the near future. The work also led to improvements in reporting and regulation of drinking water contaminants, and more stringent monitoring of reticulated supplies. Following the VegeSafe program and the NSW water study, communities are becoming more aware of the risks associated with soil and water contaminants and programs are rolling out to highlight these risks to communities. Following the assessment of chicken effluent waters, regulators

have intervened and the discharge has ceased. Additionally, regulators are looking to adopt the approach of DNA marker tracing for other complex environmental problems. The work in Boolaroo, examining soil lead concentrations surrounding the former lead smelter, has led to the formation of a working group which aims to understand the impact that living in this contaminated environment is having on primarily childhood health. Finally, the work in Newcastle has already raised the interest of environmental and health regulators, with an aim to provide the community with a useful solution to this city-wide problem. Through these achievements, this thesis has reduced the risk of human exposure to environmental contaminants in Australia, creating a safer and cleaner environment for future generations.

7 Appendix

7.1 Supplementary Data Study 1

Supplementary Data 1 – Detailed laboratory methods used in the determination of drinking water lead contamination in the Ringarooma catchment.

Procedure	Method Details	Instrument	Procedure	QA/QC
1	Total As, Cu, Pb and Sn quantification in water samples. Analysed at the National Measurement Institute, North Ryde, Australia.	Perkin Elmer Sciex Elan [®] DRC II (Axial Field Technology) Inductively Coupled Plasma Mass Spectrometer (ICP-MS) (As, Cu, Pb and Sn).	A 30 mL aliquot of water sample was digested in 2 mL of analytical grade HNO ₃ . The sample was then placed in a steam digester for 1.5 hours at 110°C and cooled for 1 hour. A 9.5 mL aliquot was then collected and a matrix spike of 0.4 mL of indium and 0.2 mL of HNO ₃ added.	One matrix spike and one duplicate sample were analysed every 10 samples and a blank sample was run at the beginning, middle and end of the ICP-MS procedure. Relative percent differences (RPD), laboratory control spike recoveries (LCS) and matrix spike recoveries (MS): arsenic = not detected RPD, 101 – 105% LCS, 88 – 96% MS, copper = not detected RPD, 96 – 102% LCS, 98 – 105% MS, lead = not detected RPD, 95 – 101% LCS, 95 – 104% MS.
2	Lead isotope determination in water samples. Analysed at the National Measurement Institute, North Ryde, Australia.	Perkin Elmer Sciex Elan [®] DRC II (Axial Field Technology) Inductively Coupled Plasma Mass Spectrometer (ICP-MS)	Samples were digested by adding 2 mL concentrate HNO ₃ to 30 mL sample and heated at 100°C for 1.5 hours.	Samples were bracketed during analysis with the NIST 981 SRM (certified values: ²⁰⁶ Pb/ ²⁰⁴ Pb = 16.94, ²⁰⁶ Pb/ ²⁰⁷ Pb = 1.09, ²⁰⁸ Pb/ ²⁰⁷ Pb = 2.37). Values were collected for ²⁰² Hg, ²⁰⁴ Pb, ²⁰⁶ Pb, ²⁰⁷ Pb and ²⁰⁸ Pb and then corrected for variation based on blank values and the effect of Hg on ²⁰⁴ Pb. Relative standard deviations for the lead isotope ICP-MS procedure are: ²⁰⁶ Pb/ ²⁰⁴ Pb = 0.57%, ²⁰⁶ Pb/ ²⁰⁷ Pb = 0.33%, ²⁰⁸ Pb/ ²⁰⁷ Pb = 0.41%.

3	Soil, sediment or rock screening using a portable X-ray fluorescence spectrometer (pXRF).	Olympus® InnovX Delta series, 4 W 50 kV pXRF.	Soil or Geochem mode using a 30 second beam time for three beams.	The NIST 2711a (Montana, USA, soil) and NIST 2710a (Montana, USA, soil) standards were used for pXRF QA/QC. The following concentration relative percent differences were detected compared to the standards: arsenic = - 27 to + 8%, copper = - 33 to + 5%, lead = - 12 to + 3%, tin = - 65 to – 50%.
4	Soil and sediment analysis by digestion and ICP-MS at the Geochemical Analysis Unit (GAU) in the Department of Earth and Planetary Sciences, Macquarie University, Australia.	Agilent® 7500cs ICP-MS	Samples were digested in a 1:1 solution of HNO ₃ and HF at 120°C for 24 hours then evaporated and repeated. Once evaporated 6N HNO ₃ was added and heated overnight until dry then 10 ml 2% HNO ₃ added followed by heating until residue was dissolved. A dilution factor of 1000 was then applied.	Batch bracketing by standard bcr-2 (USGS, Columbia River, USA, basalt) and sample blank. Standards bhvo-2 (USGS, Hawaiian Volcanic Observatory, USA, basalt) and bir-1 (USGS, Iceland, basalt) included once per 10 samples. The RPD for Pb in bcr-2 duplicates = 13%. The certified values for bir-1 and bhvo-2 was 1.39 mg/kg and 1.19 mg/kg respectively.

Supplementary Data 2 – Detailed element concentrations (As, Cu, Pb, Sn) in water samples collected in the Ringarooma River catchment, including date collected, pH and sample co-ordinates. Data illustrates lead and copper contamination of water is localised to areas of infrastructure. Filtered samples (F), unfiltered (UF).

Sample	Arsenic µg/L	Copper µg/L	Lead µg/L	Tin µg/L	Date Collected	pH	Longitude	Latitude
Pioneer House 1	<1	450	18	<1	Mar-14	Not collected	147.928181	-41.083524
Pioneer House 1 Sample 2	<1	5.5	1.9	<1	Mar-14	Not collected	147.931957	-41.082698
Pioneer House 1 Sample 3	<1	11	27	<1	Mar-14	Not collected	147.931957	-41.082698
Pioneer House 2 Sample 1	<1	1,700	25	<1	Mar-14	Not collected	147.934919	-41.082522
Pioneer House 2 Sample 3	<1	130	4.2	<1	Mar-14	Not collected	147.934919	-41.082522
Pioneer House 4	<1	28	9.3	<1	Mar-14	Not collected	147.935230	-41.082458
Pioneer House 5 Sample 1	<1	52	9.6	<1	Mar-14	Not collected	147.935554	-41.082375
Pioneer House 5 Sample 3	<1	16	2	<1	Mar-14	Not collected	147.935554	-41.082375
Pioneer House 6 UF	<1	1,500	210	<1	Mar-14	Not collected	147.935990	-41.082230
Pioneer House 6 F (household commercial filter)	<1	26	10	<1	Mar-14	Not collected	147.935990	-41.082230
Pioneer House 7	<1	220	17	<1	Mar-14	Not collected	147.936983	-41.081121
Pioneer House 8	<1	4.3	<1	<1	Mar-14	Not collected	147.939618	-41.082329
Pioneer Town End Flow Valve	1.1	550	120	6.1	Mar-14	5.76	147.939240	-41.081400
Pioneer Public Toilet	<1	2,200	220	18	Mar-14	6.29	147.933252	-41.081972
Ringarooma Public Toilet	<1	310	13	1.7	Mar-14	Not collected	147.733799	-41.241857
Gladstone Public Toilet	<1	310	13	<1	Mar-14	Not collected	148.010661	-40.958017
Frome Dam South Input	<1	1.1	1.4	<1	Mar-14	5.67	147.918170	-41.147160
Frome Mid Dam	<1	1.5	<1	<1	Mar-14	6.0	147.915562	-41.145441
Blue Tier Weir At River	<1	<1	<1	<1	Mar-14	7.44	147.941710	-41.169610
Frome Dam Input North	<1	<1	<1	<1	Mar-14	2.86	147.926480	-41.148600
Cesspit Lower Dam	<1	<1	<1	<1	Mar-14	Not collected	147.929435	-41.087238
Cesspit #1	1.8	<1	1.6	<1	Mar-14	Not collected	147.929524	-41.089302
Moorina Power Station Water Race Downstream	<1	1.8	<1	<1	Mar-14	6.81	147.910464	-41.130490

End Of Power Station Pipe	<1	<1	<1	<1	Mar-14	Not collected	147.910860	-41.125700
Greenstone Creek	<1	<1	<1	<1	Mar-14	Not collected	147.916500	-41.114634
Moorina Power Station Below Pipe	23	150	540	11	Mar-14	Not collected	147.911174	-41.130955
Frome Dam Above Wall	<1	<1	<1	<1	Mar-14	3.86	147.911610	-41.145310
Frome Lower Weir	<1	1.1	<1	<1	Mar-14	1.96	147.909713	-41.140334
Ringarooma Post Office	<1	590	36	<1	Mar-14	Not collected	147.735090	-41.241496
Pioneer Town Tank UF	<1	<1	<1	<1	Oct-13	7.20	147.934022	-41.083407
Pioneer Town Tank F	<1	<1	<1	<1	Oct-13	7.20	147.934022	-41.083407
W1 F	<1	<1	<1	<1	Oct-13	6.84	147.766759	-41.151786
W1 UF	<1	<1	<1	<1	Oct-13	6.84	147.766759	-41.151786
W2 F	<1	<1	<1	<1	Oct-13	6.20	147.942950	-41.078760
W2 UF	<1	<1	<1	<1	Oct-13	6.20	147.942950	-41.078760
W3 F	<1	<1	<1	<1	Oct-13	6.20	147.942950	-41.078760
W3 UF	<1	<1	<1	<1	Oct-13	6.60	147.945508	-41.080876
W4 F	<1	<1	<1	<1	Oct-13	6.60	147.945508	-41.080876
W4 UF	<1	<1	<1	<1	Oct-13	Not collected	147.928383	-41.076351
W5 F	<1	<1	<1	<1	Oct-13	Not collected	147.928383	-41.076351
W5 UF	<1	<1	<1	<1	Oct-13	6.30	147.950297	-41.083128
W6 F	<1	<1	<1	<1	Oct-13	6.30	147.950297	-41.083128
W6 UF	<1	<1	<1	<1	Oct-13	6.30	147.950297	-41.083128
W7 F	<1	<1	<1	<1	Oct-13	6.89	147.874078	-41.121171
W7 UF	<1	<1	<1	<1	Oct-13	6.89	147.874078	-41.121171
W7 UF Dup	<1	<1	<1	<1	Oct-13	6.89	147.874078	-41.121171
W8 F	<1	1.2	<1	<1	Oct-13	6.50	147.764306	-41.238298
W8 UF	<1	<1	<1	<1	Oct-13	6.50	147.764306	-41.238298
W9 F	<1	<1	<1	<1	Oct-13	5.93	147.917359	-41.145543

W9 UF	<1	<1	<1	<1	Oct-13	5.93	147.917359	-41.145543
W10 F	<1	<1	<1	<1	Oct-13	6.63	147.833568	-41.143544
W10 UF	<1	<1	<1	<1	Oct-13	6.63	147.833568	-41.143544
W10 UF Dup	<1	<1	<1	<1	Oct-13	6.63	147.833568	-41.143544
W11 F	<1	<1	<1	<1	Oct-13	6.26	147.805451	-41.149861
W11 UF	<1	<1	<1	<1	Oct-13	6.26	147.805451	-41.149861
W12 F	<1	<1	<1	<1	Oct-13	6.49	147.797356	-41.141329
W12 UF	<1	<1	<1	<1	Oct-13	6.49	147.797356	-41.141329
W12 UF Dup	<1	<1	<1	<1	Oct-13	6.49	147.797356	-41.141329
W13 F	<1	<1	<1	<1	Oct-13	Not collected	147.947403	-41.084166
W13 UF	<1	<1	<1	<1	Oct-13	Not collected	147.947403	-41.084166
W13 F Rep	<1	<1	<1	<1	Oct-13	Not collected	147.947403	-41.084166
W14 F	<1	<1	<1	<1	Oct-13	6.44	147.706073	-41.270981
W14 UF	<1	<1	<1	<1	Oct-13	6.44	147.706073	-41.270981
W14 UF Rep	<1	<1	<1	<1	Oct-13	6.44	147.706073	-41.270981
W15 F	<1	<1	<1	<1	Oct-13	6.84	147.871398	-41.124357
W15 UF	<1	<1	<1	<1	Oct-13	6.84	147.871398	-41.124357
W16 F	<1	<1	<1	<1	Oct-13	6.80	147.738545	-41.168475
W16 UF	<1	<1	<1	<1	Oct-13	6.80	147.738545	-41.168475
W17 F	<1	<1	<1	<1	Oct-13	6.50	147.806858	-41.147229
W17 UF	<1	<1	<1	<1	Oct-13	6.50	147.806858	-41.147229
W17 F Rep	<1	<1	<1	<1	Oct-13	6.50	147.806858	-41.147229
W18 F	<1	<1	<1	<1	Oct-13	6.49	147.874635	-41.123280
W18 UF	<1	<1	<1	<1	Oct-13	6.49	147.874635	-41.123280
W18 F Rep	<1	<1	<1	<1	Oct-13	6.49	147.874635	-41.123280
W19 F	<1	<1	<1	<1	Oct-13	6.75	147.736689	-41.157292
W19 UF	<1	<1	<1	<1	Oct-13	6.75	147.736689	-41.157292

W20 F	<1	<1	<1	<1	Oct-13	5.74	147.910472	-41.142239
W20 UF	<1	350	6.5	<1	Oct-13	5.74	147.910472	-41.142239
W20 UF Dup	<1	230	6.1	<1	Oct-13	5.74	147.910472	-41.142239
Gladstone Public Toilet F	<1	3.2	<1	<1	Oct-13	Not collected	148.010661	-40.958017
Gladstone Public Toilet UF	<1	9.7	2.1	<1	Oct-13	Not collected	148.010661	-40.958017
Greenstone Creek F	<1	1.2	<1	<1	Oct-13	6.12	147.916500	-41.114634
Greenstone Creek UF	<1	1.1	<1	<1	Oct-13	6.12	147.916500	-41.114634
Cesspit #1 F	<1	1	<1	<1	Oct-13	Not collected	147.929524	-41.089302
Cesspit #1 UF	<1	1.1	<1	<1	Oct-13	Not collected	147.929524	-41.089302
Cesspit #1 Lower Dam F	<1	15	<1	<1	Oct-13	5.33	147.929435	-41.087238
Cesspit #1 Lower Dam UF	<1	38	7	3.1	Oct-13	5.33	147.929435	-41.087238
Ringarooma Public Toilet F	<1	130	6.6	<1	Oct-13	Not collected	147.733799	-41.241857
Ringarooma Public Toilet UF	<1	150	7.9	<1	Oct-13	Not collected	147.733799	-41.241857
Derby Public Toilet F	<1	56	<1	<1	Oct-13	Not collected	147.797265	-41.142141
Derby Public Toilet UF	<1	71	<1	<1	Oct-13	Not collected	147.797265	-41.142141
Pioneer House 8 F	<1	220	17	<1	Oct-13	5.46	147.937056	-41.082339
Pioneer House 8 UF	<1	390	31	<1	Oct-13	5.46	147.937056	-41.082339
Pioneer House 9 F	<1	190	2.9	<1	Oct-13	Not collected	147.932222	-41.084049
Pioneer House 9 UF	<1	150	2.7	<1	Oct-13	Not collected	147.932222	-41.084049
Field Blank 1	<1	<1	<1	<1	Oct-13	Not collected	No data	No data
Field Blank 2	<1	<1	<1	<1	Oct-13	Not collected	No data	No data
Field Blank 3	<1	<1	<1	<1	Oct-13	Not collected	No data	No data
Field Blank 4	<1	<1	<1	<1	Oct-13	Not collected	No data	No data
Field Blank 5	<1	<1	<1	<1	Oct-13	Not collected	No data	No data
Field Blank 6	<1	<1	<1	<1	Oct-13	Not collected	No data	No data
Field Blank 7	<1	<1	<1	<1	Oct-13	Not collected	No data	No data

Field Blank 8	<1	<1	<1	<1	Oct-13	Not collected	No data	No data
Trip Blank 1	<1	<1	<1	<1	Oct-13	Not collected	No data	No data
Trip Blank 2	<1	<1	<1	<1	Oct-13	Not collected	No data	No data
Trip Blank 3	<1	<1	<1	<1	Oct-13	Not collected	No data	No data
Trip Blank 4	<1	<1	<1	<1	Oct-13	Not collected	No data	No data
Trip Blank 5	<1	<1	<1	<1	Oct-13	Not collected	No data	No data
Trip Blank 6	<1	<1	<1	<1	Oct-13	Not collected	No data	No data
Trip Blank 7	<1	<1	<1	<1	Oct-13	Not collected	No data	No data
Trip Blank 8	<1	<1	<1	<1	Oct-13	Not collected	No data	No data
Trip Blank 9	<1	<1	<1	<1	Mar-14	Not collected	No data	No data

Supplementary Data 3 – Soil and sediment element concentrations for the Ringarooma River catchment. Concentrations of lead are predominantly consistent with local background concentrations. Arsenic and tin are elevated at some sites. Copper is in low concentrations.

Sample	Pb (mg/kg)	Sn (mg/kg)	As (mg/kg)	Cu (mg/kg)	Longitude	Latitude
Blue Tier at Weir Channel Sediment 2	16	53	<5	<10	147.9421	-41.169930
Blue Tier at Weir Sediment Channel Edge	18	139	<5	<10	147.9417	-41.169610
Blue Tier Mining Area Near Weir	38	129	7	264	147.9421	-41.169930
Blue Tier Weir at River Sediment Middle Channel	11	693	<5	<10	147.9421	-41.169930
Cesspit # 1 - Lower Dam	27	14	<5	<10	147.9309	-41.087210
Cesspit #1	23	64	<5	<10	147.9284	-41.091210
Cesspit # 1 - Lower Dam	12	13	<5	<10	147.9309	-41.087210
Channel at PVC Pipe to Pioneer Start	35	18	<1	<10	147.9288	-41.099850
End of Power Station Pipe Sediment	32	48	<5	<10	147.9109	-41.125700
Frome Below Dam Near Old Bottle	16	102	<5	<10	147.9111	-41.145020
Frome Dam Adjacent Sediment	42	43	9	<10	147.9116	-41.145270
Frome Dam Inlet North B	14	56	<5	<10	147.9265	-41.148600

Frome Dam Input North Channel Bank Deposit	23	115	<5	13	147.9265	-41.148600
Frome Dam Lower Weir Sediment	21	250	<5	<10	147.9098	-41.144910
Frome Dam Sediment Above Wall Bank Deposit	43	53	10	<10	147.912	-41.145182
Frome Dam Sediment (Slag Deposit)	20	75	5	<10	147.9165	-41.145727
Frome Dam Sediment 1A	27	34	<5	<10	147.9165	-41.145727
Frome Dam Sediment 1B	25	43	<5	<10	147.9165	-41.145727
Frome Dam Sediment 1C	20	55	5	<10	147.9165	-41.145727
Frome Dam Sediment Above Wall (Near Wall) Out Of Water	32	40	<5	<10	147.9116	-41.145310
Frome Dam Inlet North Sediment C	18	74	<5	<10	147.9265	-41.148600
Frome Downstream Of Dam Wall	14	15	<5	<10	147.9111	-41.145020
Frome Forestry Soil 2	31	17	<5	<10	147.9353	-41.162610
Frome Forestry Soil 1	20	<5	3	<10	147.9353	-41.162610
Frome Lower Weir Sample Deposit Up From Channel	34	222	12	77	147.9098	-41.144910
Greenstone Creek Dam Inflow Sediment	34	26	5	<10	147.9165	-41.116163
Greenstone Creek Sediment	30	15	<5	<10	147.9171	-41.115877
Joint 0 - 0cm	84	<5	<5	<10	147.9309	-41.087210
Joint 0 - 20 cm	47	<5	<5	<10	147.9309	-41.087210
Joint 0 - 5 cm	25	<5	<5	<10	147.9309	-41.087210
Joint 1 - 0 cm	20	<5	<5	<10	147.9309	-41.087210
Joint 1 - 20 cm	23	10	<5	<10	147.9309	-41.087210
Joint 1 - 5 cm	12	<5	<5	<10	147.9309	-41.087210
Joint 2 - 20 cm	9	<5	<5	<10	147.9309	-41.087210
Joint 2 - 5 cm	13	<5	<5	<10	147.9309	-41.087210
Joint 2 - 0 cm	15	<5	<5	<10	147.9309	-41.087210
Mining Area Near Weir	31	564	5	<10	147.9421	-41.169930
Moorina Power Station Upstream Upper Slope Near Cleared Area	34	37	<5	<10	147.9115	-41.131194

Old Pipe Along Drive At Power Station	70	40	<5	<10	147.9092	-41.132029
Water Race Downstream Of Power Station Sediment 1	35	65	<5	<10	147.9105	-41.130279
Water Race Downstream Of Power Station Sediment 2	30	35	<5	<10	147.91	-41.144620
Water Race Entry Gate Sediment 1	27	93	11	19	147.91	-41.144620
Water Race Entry Gate Sediment 2	23	61	<5	11	147.9105	-41.130279

Supplementary Data 4 – ICP-MS and Portable XRF (pXRF) lead comparison table. Concentrations obtained from ICP-MS do not differ significantly from those obtained from the pXRF.

Sample	pXRF Pb Concentration (mg/kg)	ICP-MS Pb Concentration (mg/kg)
Greenstone Creek Sediment	30	38
Frome Dam Inlet North B	14	12
Joint 0 - 20 cm	47	56
Blue Tier Mining Area Near Weir	38	26
Frome Dam sediment 1B	25	25
Cesspit #1	23	26
Cesspit # 1 - Lower Dam	12	24
Frome Below Dam Near Old Bottle	16	19
Frome Dam Sediment Above Wall Bank Deposit	43	46
Z3_W32	18	12

Supplementary Data 5 – Rock sample element concentrations from the Ringarooma River catchment

Element	G1	Run 1	Run 2	Run 3	Average	G2	Run 1	Run 2	Run 3	Average	G3	Run 1	Run 2	Run 3	Average
As (mg/kg)		<5	<5	<5	<5		<5	<5	<5	<5		<5	<5	<5	<5
Cu (mg/kg)		<10	<10	<10	<10		<10	<10	<10	<10		<10	<10	<10	<10
Pb (mg/kg)		54	25	46	42		43	45	52	47		42	8	43	31
Sn (mg/kg)		14	19	16	16		14	11	14	13		10	<5	8	9
Element	G5	Run 1	Run 2	Run 3	Average	G6	Run 1	Run 2	Run 3	Average	G10	Run 1	Run 2	Run 3	Average
As (mg/kg)		6	<5	7	6		<5	<5	<5	<5		<5	5.7	<5	6
Cu (mg/kg)		<10	<10	<10	<10		<10	<10	<10	<10		<10	21	<10	21
Pb (mg/kg)		7	5	5	5		29	17	33	26		ND	ND	8	8
Sn (mg/kg)		<5	<5	<5	<5		<5	<5	10	10		19	14	13	15
Element	G11	Run 1	Run 2	Run 3	Average	G12	Run 1	Run 2	Run 3	Average	GM1	Run 1	Run 2	Run 3	Average
As (mg/kg)		24	37	14	25		12	8	10	10		<5	<5	<5	<5
Cu (mg/kg)		<10	<10	<10	<10		<10	<10	<10	<10		<10	<10	<10	<10
Pb (mg/kg)		40	56	44	47		11	11	24	15		29	32	26	29
Sn (mg/kg)		12	9	14	12		<5	<5	<5	<5		<5	16	8	12
Element	GM2	Run 1	Run 2	Run 3	Average	GM14	Run 1	Run 2	Run 3	Average	GMW 22	Run 1	Run 2	Run 3	Average
As (mg/kg)		<5	<5	<5	<5		10	6	<5	8		<5	<5	<5	<5
Cu (mg/kg)		<10	<10	<10	<10		<10	17	<10	17		<10	<10	<10	<10
Pb (mg/kg)		20	17	27	21		<5	62	<5	33		22	13	22	19
Sn (mg/kg)		9	12	13	11		<5	<5	<5	<5		<5	8	<5	8

Element	Quarry 1	Run 1	Run 2	Run 3	Average										
As (mg/kg)		<5	8	<5	8										
Cu (mg/kg)		<10	<10	<10	<10										
Pb (mg/kg)		26	116	17	20										
Sn (mg/kg)		10	<5	8	9										

7.1.2 Review of Harvey et al. (2015)



Review of Harvey et al (2015) Project 1094-15

Water for the wellbeing of all Australians
JUNE 2015



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Expert Panel Report – Review of Harvey et al (2015)

Report Title: Review of Harvey et al (2015)

Project Leader: Tony Priestley

Researchers: Troy Gaston and Peter Mosse

WaterRA Project No. 1094-15 – Review of Harvey et al (2015)

ABOUT THE AUTHORS

Dr Tony Priestley is a chemical engineer, former CSIRO Sector Manager, and Deputy CEO of the CRC for Water Quality and Treatment. He holds a Bachelor of Engineering from the University of Queensland and PhD in Chemical Engineering from Monash University.

Tony has experience in process design and analysis and in leading multi-disciplinary research groups developing innovative processes for water and wastewater treatment. In CSIRO he was involved in a multi-divisional research program looking at possible alternative approaches to the design of urban water systems.

Tony has interacted widely with the Australian water and waste management industry in Australia through service on various committees of the Victorian Branch of the Australian Water & Wastewater Association and the Environment Management Industry Association of Australia. He participated in high level industry committees which put together a number of papers for the Prime Minister's Science, Engineering and Innovation Council. The papers addressed the topics of "Commercial Opportunities in Waste Management" (1991) and Water for Our Cities (2007).

Dr Troy Gaston is a Lecturer in the School of Environmental and Life Sciences at the University of Newcastle

He has a Bachelor of Science (Honours) and PhD from the University of New South Wales. He also has a Graduate Certificate in University Teaching & Learning from the University of Tasmania.

Troy's Research expertise relevant to this review encompasses:

- Development of environmental monitoring programs addressing water quality, ecosystem health and ecosystem processes;
- Understanding trophic interactions, tracking nutrient sources, elucidating food webs; and
- Application of stable isotope analysis aquatic environments to determine pollution and energy sources.

Dr Peter Mosse is the Director of Hydrological Pty Ltd, a water industry consultancy established in 2002 to provide high-level cost effective specialist technical support and training in the areas of water treatment, wastewater treatment and distribution system management with an emphasis on trouble shooting and process optimisation. The company has developed a niche speciality for the assessment of all aspects of the ability of Water Treatment Plants (WTP) to produce safe drinking water. Clients have included over 25 Victorian and SA water utilities.

Associated with this activity has been the production of a series of Guides to the Operation of key Control Points in WTPs for the Water Industry Operators Association (WIOA). A similar series is also being produced for wastewater treatment.

Most recently in 2013 and 2014, a project has been initiated with the Queensland Water Directorate to provide specialist technical assistance, training and mentoring to a large number of WTPs in rural and remote Queensland.

Peter was project manager for the Water Treatment Alliance in Australia and has been a member of the NHMRC WQAC. He also edits and produces WaterWorks magazine for WIOA.

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1 EXECUTIVE SUMMARY

A report entitled "Identification of the sources of metal (lead) contamination in drinking waters in north-eastern Tasmania using lead isotopic compositions" by Harvey P.J., Handley, H.K. and Taylor, M.P., was published in Environmental Science Pollution Research in April, 2015. The paper concluded that the lead contamination of the drinking water in the town of Pioneer could be attributed to three sources.

1. The water race infrastructure including the large leaded joint pipelines of the Frome Dam, the Moorina Power Station and the Pioneer holding dams;
2. The PVC pipes that connect the Pioneer holding dams to the residential service lines; and
3. Lead contaminated fittings and fixtures within individual properties.

The authors used an isotopic "fingerprinting" technique to differentiate between the different possible sources of this lead contamination. They also analysed water, soil and sediment samples as well as scrapings from various pipelines within the water supply scheme.

TasWater, as operators of the Pioneer water supply scheme, engaged Water Research Australia (WaterRA) to undertake a review of this paper with the intent of assessing the following aspects.

- Review of the science around the application of source identification markers – in particular the use of isotope "fingerprinting";
- Application of the science to the drinking water supply system of Pioneer;
- The study methodology employed by Harvey et al (2015);
- The amount and strength of data gathered;
- The types of data collected and the links between the analysis and the conclusions;
- The conclusions as they relate to the water supply system; and
- Other possible explanations that have not been canvassed but that may be supported by the data.

An expert panel was formed to undertake this work. The work included a close analysis of the paper and its supporting data, an inspection of the water supply system for the town of Pioneer, discussions with TasWater staff with knowledge of the Pioneer system and an analysis of additional data supplied by TasWater.

A visit to the town of Pioneer and an inspection of its water supply system was undertaken by two members of the expert panel on May 18, 2015. This visit also provided the opportunity for extended discussions with TasWater staff. From these discussions it became clear that the Frome Dam and the Moorina Power Station had not been directly connected with the Pioneer water supply scheme since at least 2009. As a consequence, the panel concluded that the data taken by Harvey et al (2015) from the Frome Dam and Moorina Power Station were not relevant to the outcomes of the study. However, the data they took from within the town distribution system and at individual households remained relevant.

A review of the use of isotopic "fingerprinting" techniques to assess the source of lead within the Pioneer water supply system found that, while the technique has the potential for identifying the sources of lead in drinking water, the data contained in the Harvey et al (2015) paper were insufficient to draw any conclusions. One reason for this outcome was the use of a sample from the Moorina Power Station as one potential source of the isotopic composition. Another reason was lack of multiple samples and a consequent inability to estimate the variability of the results obtained.

Another major concern of the review panel was the lack of duplicate analyses and repeat samples from within the town itself. This, combined with an unclear sampling protocol, was seen as a major

REVIEW OF HARVEY ET AL (2015)

weakness of the study. However, when data from both TasWater and Harvey et al (2015) were considered together, there was little doubt that the levels of lead and copper measured at the various locations in the town were intermittently elevated. The source of these metals was the key point in question.

The potential sources of contamination identified by the panel were:

- Water in the Pioneer Dam;
- Sediment in the Pioneer Dam;
- Pipes in the town distribution system; and
- Local plumbing in individual houses.

An analysis of the data available for each of these potential sources found that:

- Based on limited data, there appears to be very low levels of lead and no copper in the raw water sourced from Pioneer Dam. However because of low pH and alkalinity, raw water from the Pioneer Dam would be considered aggressive and likely to leach metals from sediment and pipe materials;
- While the data is limited, it appears that the levels of lead in sediments within the Pioneer dam are low, although there may be some spatial variability;
- It is possible that wind and flood events could raise the turbidity in Pioneer Dam and contribute to lead bearing sediments in the town distribution system; and
- While the aggressive nature of the water could result in the leaching of lead from the PVC pipes, a report from CSIRO, Burn et al (2005) and subsequent discussion with Burn indicate that in well flushed systems this should not result in lead reaching levels dangerous to public health. However, it is possible that in stagnant and dead-end zones within the Pioneer system elevated lead levels could arise. Considering that the PVC pipes in this system are about 35 years old, only further testing could answer this question.

The panel concluded that, while the data contained in the paper by Harvey et al (2015) are sparse and their reproducibility can be questioned, the general conclusions reached in the paper are plausible.

The use of first flush samples in the paper would help explain the strong correlation found between lead and copper levels in their analyses. Consequently, these samples would not necessarily reflect the quality of water in the distribution mains which were well flushed. Calculation of daily flow rates and system volumes indicate that the main east-west distribution pipe in Pioneer should be well flushed. However, the system also contains significant dead-end zones

One option for TasWater to consider is the stabilisation of the raw water by passing it through a bed of limestone particles. This method is being used in various water supplies in both Australia (Bundaberg) and overseas (Scotland). The review panel has also suggested further investigations which would be needed to more accurately identify the source of contaminant metals in the Pioneer water supply.

2 INTRODUCTION

This report is a review of the paper “Identification of the sources of metal (lead) contamination in drinking waters in north-eastern Tasmania using lead isotopic compositions” published in Environmental Science Pollution Research” by Harvey, P.J., Handley, H.K. and Taylor, M.P. in April, 2015. The review was commissioned by TasWater with the intent of covering the following aspects:

- Review of the science around the application of source identification markers – in particular the use of isotope “fingerprinting”;
- Application of the science to the drinking water supply system of Pioneer;
- The study methodology employed by Harvey et al (2015);
- The amount and strength of data gathered;
- The types of data collected and the links between the analysis and the conclusions;
- The conclusions as they relate to the water supply system; and
- Other possible explanations that have not been canvassed but that may be supported by the data.

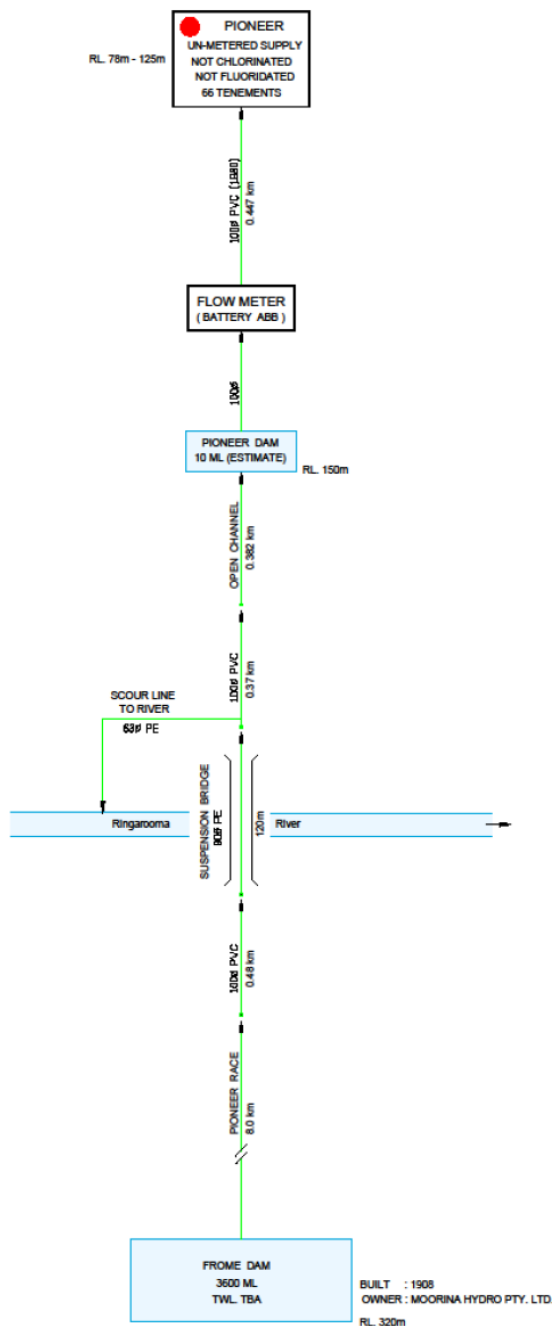
The work included a close analysis of the paper and its supporting data, an inspection of the water supply system for the town of Pioneer, discussions with TasWater staff with knowledge of the Pioneer system and an analysis of additional data supplied by TasWater.

This report:

- 1) Outlines the current water supply system for the town of Pioneer, including both the supply to Pioneer Dam and reticulation of this water to households in the town;
- 2) Reviews the science of “isotopic fingerprinting” and its application to identify sources of lead in the Pioneer system; and
- 3) Discusses the methodology of the paper and, in particular, the quality and quantity of data used to justify the paper’s conclusions.

The potential sources of lead contamination are then discussed in light of the data provided, as well as the strength of the links between the data and the conclusions reached.

2.1 The Pioneer Town Water Supply System



SCHEMATIC DIAGRAM

NOTES:

1. Levels are approximate only - derived from contours.

A schematic diagram of the Pioneer Water Scheme dated 2010, provided by TasWater, is given in Figure 1. The diagram shows that at that time water was supplied to the Pioneer Dam from the Frome Dam via an open raceway and a series of 100mm diameter polyvinyl chloride (PVC) pipes. Water was then transported from the Pioneer Dam via another length of 100mm diameter PVC pipe into a mixture of PVC and polyethylene (PE) pipes, which form the basis of the town's distribution system. Detailed plans of the distribution system within the township indicate that it consists largely of 50mm diameter PVC pipes laid in about 1980, plus further additions of 40mm diameter PE pipes.

Information from TasWater states that, since around 2009, the water supply to Pioneer is no longer sourced from Frome Dam or the raceway infrastructure immediately below it. About 900 metres of the old water race downstream of the dam are no longer used and, since the construction of an irrigation project, it has not been possible to transfer water from the Frome River along that part of the water race. This change also applies to the infrastructure associated with the old Moorina Power Station. Since this change, water for the Pioneer Dam is sourced from the open Pioneer water raceway immediately above the dam, which collects surface water runoff and water from a small water course intercepted by the raceway somewhere between the site of the old Moorina Power Station and the town of Pioneer (a distance of 5 kilometres).

While some of the water samples described in the report were taken within the town's distribution system and in some local houses, a number of samples were also taken from the Frome Dam and the infrastructure associated with the old Moorina Power Station. As these two pieces of infrastructure have not been directly associated with the town's water supply since at least 2009, the data associated with these samples are not applicable to the considerations of the paper.

Figure 1 Pioneer Water Scheme in 2010

2.2 Review of the application of isotope “fingerprinting” to the identification of the sources of lead in drinking water

Lead (Pb) has four stable isotopes: ^{204}Pb (1.4%), ^{206}Pb (24%), ^{207}Pb (23%) and ^{208}Pb (52%) (Larsen et al (2012)). The difference in ratios between the lead isotopes from different locations is a function of the starting composition and decays of the original ore content. Hence the isotope ratios can be attributed to the geological formation time of lead in different mines (Larsen et al (2012)). Although maximum information is obtained from the measurements of all four stable isotopes, generally most tracer work has been done using just the ^{206}Pb : ^{207}Pb ratio because this can be measured most accurately (Maring et al (1987)).

The isotopic compositions of lead ores are retained during natural and man-made processes that contribute to environmental lead. They are also conserved during manufacture of consumer products which subsequently may become sources of human lead exposure (Delves and Campbell (1993)). Fortunately, tracing lead contamination in Australia is often rather simple because almost all of its industrial lead came from a few large, geologically old (w1700 million years) ore deposits that have relatively unique isotopic fingerprints (Gulson (2003)). The principal Australian lead ore deposits are in Broken Hill (New South Wales) and Mount Isa (Queensland), which are among the largest in the world and have distinctive lead isotopic compositions (e.g. Gulson (1985), Gulson and Mizon (1979)).

Measurements of lead isotope ratios have been applied to a number of fields in recent years, covering environmental, geological and forensic applications. Lead isotope ratios have been successfully used to document the contribution of anthropogenic lead from leaded gasoline and mining in the environment and freshwater systems (Larsen et al 2012). It is also possible to use measurements of stable lead isotope ratios to distinguish different sources of lead in drinking water systems.

The major source of lead in drinking water has been identified to be plumbing materials (Lasheen et al (2008)). Lead pipes, lead-based solder, brass fittings and plumbing fixtures such as pipe's jointing faucets are known to be dominant lead sources in public water supply systems (Gulson et al (1994)). Lead pipes have been replaced with other types of pipes such as polymer materials like polyvinyl chloride (PVC), polyethylene (PE) and polypropylene (PP). PVC polymer is mixed with a number of additives including stabilisers in order to provide the range of properties needed in the final products. Stabilisers are often composed of salts of metals like lead and cadmium (Kim (2001)). PVC as a potential source of lead in water is discussed in a later section.

Fingerprinting based on stable lead isotope ratios of environmental samples can help to identify lead exposure pathways (Gwiazda and Smith (2000)). In general, natural sources of lead have Pb isotopic ratios different than those of anthropogenic sources (Komarek et al (2008)). As a result, when environmental lead sources are isotopically distinct from one another, lead exposure pathways may be identified. For example, Cao et al (2014), Gulson et al (1995) and Delves and Campbell (1993) have successfully identified drinking water as a source of lead in children.

However, there have also been studies that failed to identify lead sources using stable isotopes. One reason is the overlap of isotopic compositions of different environmental samples with one another. For example, Soto-Jiménez and Flegal (2011) found that the lead isotope ratios of dust, soil, and aerosols from a smelting area of Mexico were indistinguishable from each other making it impossible to apportion the dominant source of lead. Therefore, investigations require that the different sources of lead be isotopically distinct and that uptake of a given lead source produces a measurable change in the isotopic composition of the receiving water/organism.

In recent years, stable isotopes have increasingly been used as environmental tracers (Lathja and Michener (1994)). One common application uses isotope mixing models to quantify source contributions to a mixture (Phillips et al (2005)). Examples include pollution inputs to air, soil, or water bodies; food sources in animal diets and plant water use from different soil depths. Linear mixing models based on isotopic mass balance have long been used for this purpose (Phillips et al (2005)) as have two-dimensional vector-based source apportionment calculations (Larsen et al (2012)). The benefit of these models is to provide a relative contribution (%) of each source to the final mixture identifying the major pollutant source.

2.3 Application of stable isotopes to the Pioneer drinking water system

There is evidence of elevated lead concentrations in the Pioneer drinking water system. Harvey et al (2015) have used lead isotopic compositions to attempt to differentiate unknown sources of lead contamination of the drinking water supply in north-eastern Tasmania, Australia. The application of the lead stable isotope method is feasible if all potential sources of lead are identified and the isotopic composition of those potential sources are distinct. The potential sources of lead in the local waters were identified as dilapidated drinking water infrastructure, including lead jointed pipelines, end-of-life polyvinyl chloride pipes and household plumbing (Harvey et al (2015)). However, only the lead isotopic ratio of the Moorina Power Station pipe surface and at one joint were determined in this study – a fundamental flaw, as water no longer flows through these pipes. This is considered in more detail below. The study would have benefited from determination of the lead stable isotope composition of PVC pipe and pipes within dwellings of Pioneer.

There is large variability in lead isotopic composition within the first minute of tap flushing which brings into question the reliability of the recommended sampling time of 30 seconds (Gulson et al (1994)). Harvey et al have not described in enough detail how water samples for stable isotope analysis were collected to allow a detailed review of this component of the paper.

Furthermore, only single samples were collected of the potential sources (infrastructure) and mixture (three (3) water samples from three (3) individual locations). This does not provide an estimate of the range or variability of isotope ratios. There can be extremely large lead isotopic differences between dwellings within the one city and between dwellings and the storage tanks for a water supply (Gulson et al (1994)), which can lead to over or under estimation of the correct source apportionment of lead sources. Also, the authors only provide an indicative relative standard deviation for lead isotope values. Collection of replicate samples from the sources and mixture (tap water) would alleviate this uncertainty.

To determine the relative contribution of lead from potential sources, the authors have used a two-dimensional vector-based source apportionment calculation, as described by Larsen et al (2012). This model was used to determine the source of lead to: (1) the Moorina Power Station pipes and (2) water from the Pioneer House and Pioneer public toilets. Using the isotopic composition of the potential sources of local bedrock (included from other sources - see references in Harvey et al 2015) and that of lead ore from Mt Isa/Broken Hill, the joints in the Moorina Power Station pipes have 43% of their lead attributed to Broken Hill/Mt Isa type ores. Whilst this is interesting, it is irrelevant to the study, given that water no longer flows through the Moorina Power Station infrastructure and into Pioneer. It also appears that the authors use the isotopic composition of Broken Hill/Mt Isa lead ore and Moorina Power Station joints as potential sources for the apportionment of lead to the water samples collected. The authors fail to justify the use of different potential sources for each calculation.

Key points

1. The use of the Moorina Power Station joint (as a sampling location pivotal to the isotopic results) is problematic as it is not connected to the Pioneer drinking water system
2. The sampling protocol was poorly described and insufficient for confidence in repeatability
3. Accuracy and reliability of the source apportionment would have been more credible with more appropriate potential sources or “end members” and inclusion of error estimates which would provide a range of values for each lead source. Appropriate “end-members” should include the stable isotope composition of PVC pipes and pipes within dwellings of Pioneer.

3 PAPER METHODOLOGY AND THE AMOUNT AND STRENGTH OF THE DATA

The paper's major aim was to identify sources of metal contamination in drinking water in the town of Pioneer. The approach was to take a series of samples of water, soil/sediment and bedrock in areas closely associated with the water supply infrastructure e.g. transfer channels and pipes, dams and the town reticulation network. Samples were analysed for arsenic (As), copper (Cu), lead (Pb) and tin (Sn), as well as isotopic lead breakdowns for 5 samples. The general conclusions of the paper pointed to lead contamination arising from the local town infrastructure e.g. transfer pipes, PVC reticulation mains and household plumbing.

General comments on the paper's methodology and the amount and strength of the data it contains are outlined below, followed by another section containing more detailed comments on various aspects of the paper.

General Comments

It appears that the paper's authors did not fully understand or appreciate the exact scope of the current water supply system. As described earlier, the Frome Dam and the Moorina Power Station have not been directly connected to the Pioneer town water supply since around 2009. This situation appears to discount data collected by the authors on Frome Dam and the Moorina Power Station infrastructure. Having said this, it is possible that contamination from the Power Station infrastructure may have been transferred to the Pioneer Dam before the disconnection around 2009 and may have accumulated within the sediments contained in Pioneer Dam.

The second general comment relates to the quantity of data collected to support the conclusions of the paper and, in particular, the sampling protocols used to collect that data. An examination of the supplementary data sheets provided with the paper does not provide evidence of significant numbers of duplicate analyses and/or repeat samples. Particularly when it comes to some key results within the town of Pioneer itself, there appear to be no repeat samples or duplicate analyses. Given the importance of these particular results in drawing the paper's conclusions, this is a major weakness and a cause for some uncertainty.

Particular concerns regarding the methodology and the amount and quality of data collected arise from the following observations. This section goes into the details of the working group's thoughts on the Pioneer water supply system and provides the basis for the subsequent discussion.

Specific Comments

3.1 Sampling Sites

The working group was most concerned about the inclusion of samples from Frome Dam, Moorina Power Station and the old lead-jointed pipes associated with the Power Station – none of which have been connected to the Pioneer system since some time in 2009 (Wright 2015).

This major flaw raises significant uncertainty about any conclusions relating to these sites as sources of contamination.

The remaining sample sites are legitimate, although the nomenclature and actual nature of the sample point is at times confusing (see below).

3.2 Samples

Water samples were collected into appropriately prepared sample bottles.

There were two sampling events in October 2013 and March 2014 which the authors state "*encapsulated seasonal variability in the water race of Pioneer*" but they provide no meteorological data to support this statement. There is also no record of the water temperature at the sample sites even though it was stated in the methods that temperature was measured. Examination of the

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temperatures may have made it possible to have made some assessment of the “seasonal variability”. It is also not clear why the authors make the point about seasonal variability.

Further, the authors state that “*water samples were collected as a dissolved (<0.45 µm) and total (unfiltered) fraction. There was limited difference between the two fractions for lead so only total (unfiltered) samples were collected on the second visit. Water pH and temperature were measured at each site*”. In fact, the differences between filtered and unfiltered samples were quite significant for those samples with elevated lead levels. This difference is significant because it relates to the possible mechanisms for lead dispersal into the water. It would also have been useful to include turbidity data to allow the reader to make some independent interpretation of this statement; however there is no turbidity data.

3.3 First Flush and Time Series Sampling

While the authors have recognised the value of obtaining first flush samples, the information relating to the sampling does not make it clear whether the water was sampled from hot or cold water taps.

(Volunteers for kitchen water sampling were sought from the local population. Volunteers were instructed how and when to collect samples. First draw samples were collected after 8 h stagnation time on the first sampling trip.) (Harvey et al (2015))

This is important since levels of metal contaminants are likely to be higher in first draw hot water samples. It is also not clearly stated that the household samples were collected in the pre-prepared sample bottles. It seems reasonable to assume this was the case but it is not stated. There is also a very real risk in taking unsupervised samples. For the integrity of the data it would have been far better if the researchers had collected the samples themselves. There is absolutely no guarantee that all the first draw samples are in fact genuine first draw samples.

3.4 Sample Numbers and Replication

The paper is based on a very small number of samples with essentially no replication. For example, the data in Figure 4 appears to be based on single samples. With only single samples there can be no statement as to the variability of the data and the variability can be significant. Inspection of any NATA inter-laboratory testing report readily reveals significant variation in values returned from identical samples. In support of this statement, in one set of data reported by Harvey et al (2015) where there was replication (see Table 2 in Harvey et al), lead content in Pioneer Cesspit #1 Lower Dam Pipe Joint 2 varied with 94, 484, 274 mg/kg. Clearly this refers to a sediment sample however it emphasises that variations occur. While there do appear to be significant differences between houses in Pioneer the combination of the sampling issues and data variability make interpretation of such differences very difficult.

3.5 Lead and Copper Levels at the Customer Tap

There is little doubt that the levels of lead and copper in drinking water sampled at customer's taps in Pioneer are elevated in some samples. Lead regularly exceeds the ADWG health limit of 0.01 mg/L while copper occasionally exceeds the ADWG aesthetic limit of 1 mg/L and rarely the health limit of 2 mg/L.

The question is what are the sources? While the Harvey et al (2015) paper is entitled “Identification of the sources of metal (lead) contamination in drinking waters in north-eastern Tasmania using lead isotopic compositions” the paper does not provide enough evidence to conclusively identify the exact sources of the contamination.

3.6 Pioneer Demand and Distribution System Turnover

TasWater report that typical monthly usage for the township of Pioneer is in the region of 0.3 ML or around 10 kL/d.

Based on the system drawings provided by TasWater, and only including pipes in the direct line from the primary 100 mm feed main, the calculated volume for the western part of the town (including the 100 mm feed main) is approximately 5.2 kL and for the eastern part of town (including the 100 mm

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feed main) is approximately 3.8 kL. Therefore it seems likely that the water in the mains is fairly well turned over on a daily basis.

There are however a number of quite long dead end mains in the supply system and these may not get turned over. These different parts of the distribution system may also have contributed to some of the variability in the data shown in Figure 4 of Harvey et al (2015).

Key points

1. Data from Frome Dam and Moorina Power Station are not applicable as these sources have not been directly connected to the Pioneer Dam since 2009.
2. A lack of duplicate samples and repeat analyses, combined with an unclear sampling protocol, is a major weakness of the paper.
3. There is little doubt that lead and copper in drinking water sampled at customer's taps in Pioneer are elevated in some samples.

4 POTENTIAL SOURCES OF CONTAMINATION

There are a number of possible sources for contamination of the Pioneer town water supply. These include:

- Water in the Pioneer Dam
- Sediment in the Pioneer Dam
- Pipes in the town distribution system
- Local plumbing in individual buildings

Each of these potential sources will be considered below.

4.1 Water in Pioneer Dam

A description of the water source to the Pioneer Dam as outlined earlier is provided by Wright (2015).

The Pioneer Dam is fed by the open Pioneer water race which originally extended from Frome Dam to Pioneer and supplied water to the Moorina Power Station on the way. Water from the Frome Dam has not now supplied Pioneer for several years. Water that is transferred from the race to the Pioneer Dam is water trapped by the open race from surface water runoff and small watercourse intercepted by the race between the site of the old Moorina PS and Pioneer.

While TasWater refers to the water body supplying the township of Pioneer as Pioneer Dam, Harvey et al (2015) refer to a number of samples as Cesspit #1. By entering the provided latitude and longitude into Google Earth, the sites seem to map to different sites around Pioneer Dam. The use of the term Cesspit to describe part of a water supply system lacks a professional and objective approach to the study. The identification of the sites is made more difficult since in their text they refer to the Cesspit when considering water sample data but then refer to Pioneer Dam when discussing sediment and solid samples. Only the authors know the full details of these sites.

Based on this information, the data for the various Cesspit #1 samples have been interpreted as pertaining to Pioneer Dam. Hopefully this is correct.

There is only limited water quality data available for raw water sampled directly from the Pioneer Dam. The data provided to the panel by TasWater covers the period of 29/8/2012 to 27/6/2013. Sample sites include Dam Inlet, Dam Surface, Dam Bottom and Dam Outlet. Table 1 summarises the lead results for the dam outlet.

TasWater advises that “historically this system has only been sampled from the standard compliance location within the town distribution system. Due to the fact this system receives no form of treatment other than settlement within Pioneer Dam this was taken to reflect the raw water quality”. While this statement is true for some parameters it is not true for all parameters.

The data from the June 2013 sampling period shows the following.

- pH is between 5.7 and 6.0;
- True colour ranges between 5 and 77 CU;
- Turbidity varies between 0.4 and 7.3 NTU; and
- Alkalinity (mg/L CaCO₃) is consistently 2 or less.

Based on the pH and alkalinity, the water would be considered aggressive and would likely leach metals from sediments and pipe materials. However, there is insufficient data to carry out any statistical analysis to demonstrate this point.

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Data from the Harvey et al (2015) paper includes three results from a site called Cesspit #1 and Cesspit Lower Dam which, as outlined above, are assumed to have been taken from the Pioneer Dam. Two separate samples were taken in October 2013 and March 2014. These samples show very low levels of copper and lead, although one sample had unfiltered lead levels of 7 µg/l. The one pH measurement provided of 5.33 indicates that the water is acidic.

While the TasWater data is limited, it appears that all the water sampled from the Pioneer Dam Outlet, with one exception, has very low levels of lead (Table 1). One sample on 21/11/2012 has a value of 1690 ug/L. This is an extremely high value, particularly with respect to all the other measurements. Unless this sample had been retested and the result verified, preferably with a second sample, this value should be considered doubtful.

Table 1. Lead levels in water sampled from the outlet to Pioneer Dam. (Data from TasWater)

Sample Site	Date	Parameter	Value	Units
Pioneer Outlet	29/08/2012	Lead	<0.5	ug/L
Pioneer Outlet	10/09/2012	Lead	<0.5	ug/L
Pioneer Outlet	10/09/2012	Lead	<0.5	ug/L
Pioneer Outlet	18/09/2012	Lead	0.7	ug/L
Pioneer Outlet	25/09/2012	Lead	<0.5	ug/L
Pioneer Outlet	2/10/2012	Lead	<0.5	ug/L
Pioneer Outlet	21/11/2012	Lead	1690	ug/L
Pioneer Outlet	27/11/2012	Lead	<0.5	ug/L
Pioneer Outlet	29/11/2012	Lead	<0.5	ug/L
Pioneer Outlet	3/12/2012	Lead	<0.5	ug/L
Pioneer Outlet	13/12/2012	Lead	1	ug/L
Pioneer Outlet	20/12/2012	Lead	<0.5	ug/L
Pioneer Outlet	6/06/2013	Lead	<0.5	ug/L
Pioneer Outlet	27/06/2013	Lead	1.1	ug/L

There is no data provided by TasWater on copper levels in the raw water. This is consistent with the fact that copper is not normally sampled in raw water. Most copper in water comes from domestic plumbing.

Harvey et al (2015) provide data in their paper and in a supplementary data sheet. The data provided are difficult to interpret partly due to the use of the names of the sample sites, for example W1 to W20 is particularly uninformative. They appear to be from the Ringarooma River. Either way, with the exception of W20, the levels of arsenic, copper, lead and tin are all below detection limits except for one very low positive value. W20, which appears to map somewhere near Frome Dam, had moderate levels of copper and low levels of lead. The source of copper at this point is puzzling. However, this result is not relevant to the considerations of this report since Frome Dam no longer feeds Pioneer Dam.

The issue of copper in the raw water is also raised by the results for one of the Cesspit #1 samples. This sample had low levels of both copper and lead. Once again the presence of copper at this point is puzzling. However the authors do not adequately describe the characteristics of the different sample points. Was it a grab sample or was it sampled through some sort of tap and pipe?

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Based on the limited data available, it appears that there are very low lead levels and no copper (with the exception of the sample described above) in the raw water sourced from Pioneer Dam.

Key points

1. Based on the limited data available, it appears that there are very low lead levels and no copper in the raw water sourced from Pioneer Dam.
2. Based on the pH and alkalinity, the water would be considered aggressive and would likely leach metals from sediments and pipe materials.

4.2 Sediments

Pioneer Dam Sediments

As with the raw water, there is only limited data available on the lead content of Pioneer Dam sediments. In the data provided to the panel by TasWater, there appear to be only two sediment samples. One sample (6/6/2013) returned a value of 416 ug/L and the other (20/6/2013) a value of <7 ug/L. A March, 2013 memorandum to Ben Lomond Water authored by GHD provides a single figure of 117 mg/kg of lead for a soil sample from the dam. The use of ug/L as a concentration unit for a sediment sample in the TasWater data is puzzling. In addressing this question, TasWater state

Attached are the two analytical reports covering these sampling events. Previously unless otherwise specified as a sediment investigation, the laboratory's normal process for "sediment" samples with very low solids was that they were tested as water and hence reported with "water" units. (AST#59735)

Unfortunately this only adds to the problem of interpreting the data. If the samples only had "very low solids" then it is difficult to see how they are in fact sediment samples.

So can any conclusions be reached based on this data? At best they suggest variable amounts of lead in the sediments in Pioneer Dam but this would need to be verified with more and careful sampling and testing.

Harvey et al (2015) report some sediment results from Cesspit #1. The values for lead for this site range between 12 and 27 mg/kg. The authors conclude "*analysis of soil and sediment in and around Pioneer Dam show low lead*".

While the data is limited, it appears that the levels of lead in Pioneer Dam sediments are low. There may however be some spatial variability.

The presence of some lead in the sediments is not surprising given the fact that Pioneer Dam was originally supplied from Frome Dam via the Moorina Power Station and old pipes. Harvey et al (2015) have clearly demonstrated a possible lead contamination along that route.

Potential Impact of Pioneer Dam Sediments on Drinking Water Supplied to Pioneer

There is some lead in the sediments in Pioneer Dam, albeit quite low. However it could be a source of lead under conditions where sediment is carried into the system. Again while the data is very limited, the turbidity of the Dam Outlet during the June 2013 sampling period varies between 0.4 NTU and 1.7 NTU. The slightly higher turbidity could contribute to slightly higher total lead concentrations during the periods of elevated turbidity. It is also possible that the turbidity could be quite a bit higher during flood or wind events which could contribute to variable levels of lead bearing sediment in the mains.

There does appear to be a definite reduction in colour between the Dam Inlet and the Dam Outlet in the June 2013 samples (Figure 1). The difference is significant ($P < 0.05$).

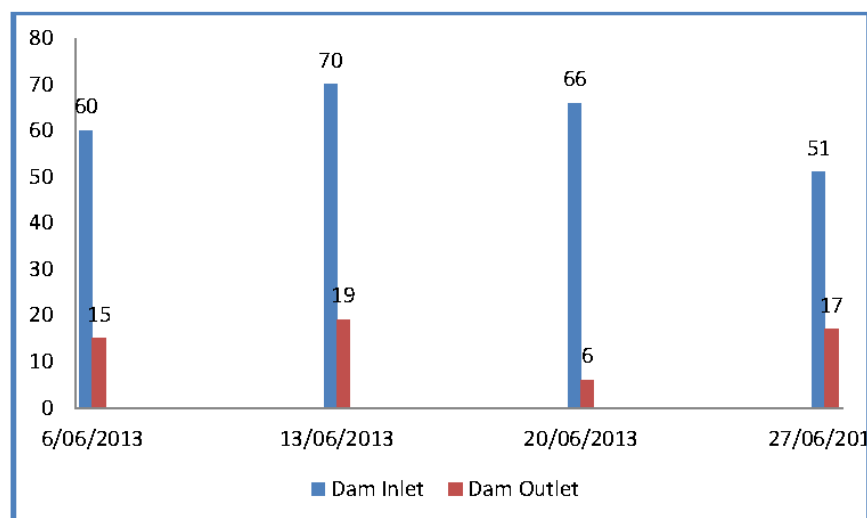


Figure 1. True colour at the Pioneer Dam Inlet and Outlet in June 2013.

This suggests that the sediments may have a significant organic component which in turn might support a range of microorganisms. It is further likely that the redox status of these sediments may change.

In the June 2013 sampling event, Dissolved Oxygen (DO) was also measured in some of the samples. On the 6th June, the DO is recorded as <1 mg/L while on the 13th June the DO is recorded as between 7.8 mg/L and 8.1 mg/L. This is a huge difference, and if correct, suggests the nature of the water can change dramatically. This data needs to be checked carefully. Further, given the apparent variability, it seems unfortunate that TasWater did not continue to collect DO data for the remaining test days.

If the DO results are correct, it is possible that during any anoxic events, metals could be released from the sediment. However, the lead data from the two samples in June 2013 do not support this hypothesis. The DO levels need to be checked carefully and ideally more testing done on the redox status of the water.

Key points

1. While the data are limited, it appears that the levels of lead in Pioneer Dam sediments are low. There may however be some spatial variability.
2. It is possible that the turbidity could be higher during flood or wind events which could contribute to variable levels of lead bearing sediment in the mains.

4.3 Pipes

As a Source of Lead

The pipes that constitute the distribution system in Pioneer are of two types.

1. Unplasticised PVC of 50mm diameter installed in 1980
2. Polyethylene pipes of 40mm diameter installed 1980

Harvey et al (2015) noted that the early PVC pipes were stabilised with lead. The leaching of lead from PVC pipes has been an issue of concern since their introduction into water supply systems in the late 1970s. This concern has led to the elimination of lead from PVC pipes destined for water supply systems in the early 2000s. PVC pipes used for water supply in Australia are now stabilised with calcium/zinc formulations (Burn et al (2005)).

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The PVC pipes used in the Pioneer water supply system were installed in 1980 and thus presumably contain lead stabilisers. Consequently, these PVC pipes are a potential source of lead in the Pioneer water supply system. A number of studies (Al Malack (2001) and Lasheen (2008)) have demonstrated the potential for lead leaching from PVC piping to contribute to lead levels in domestic water supplies. These studies have shown that low pH and alkalinity, combined with extended contact time and high surface to volume ratio can result in increased lead levels in water above the recommended health limit of 10 µg/L. A CSIRO review (Burn et al (2005)) quotes these results, but also states that *“the consensus being that the concentration of lead leached into water does not pose a health hazard to humans, provided proper flushing is conducted during the commissioning of the new pipes”*. Burn’s comments on the Pioneer data are that, in his opinion, only extended contact time of water with the PVC pipe in a stagnant end zone of the pipe could possibly result in the lead levels being observed. Such levels should not occur in situations where there is significant water flow through the pipe (Burns pers comm (2015)).

Clearly the aged PVC pipes in the Pioneer water supply system are a potential source of lead in the water. However, a number of questions surround this possibility. There appear to be no data available in the literature on the long term (over years) leaching characteristics of PVC pipes. The pipes in the Pioneer system are about 35 years old. If lead was being continuously leached from these pipes over that very long time period, then it would be expected that the pipes become increasingly brittle and an increasing rate of pipe bursts should be noted. Discussion with TasWater personnel indicates that this does not appear to be the case.

As a Source of Copper

Global experience has shown that the main source of copper in household drinking water is copper pipes and brass fittings usually confined to domestic plumbing within the property and building.

TasWater reports that *“local operators have indicated that the vast majority of service pipe material post the meter is laid in HDPE. A small number of cast iron galvanized services still remain but no information available on which exact services these are”*. Nothing is known about the internal plumbing, however it seems likely that there may be some copper pipes involved, but this needs to be verified.

During the site visit by the WaterRA panel, copper pipe was observed at the back of the toilet block. The pipe seemed to be pressure jointed rather than soldered. There were also new brass fittings on many of the town hydrants (Figure 2).



Figure 2. Pioneer town hydrants

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Key points

1. While leaching of lead from PVC pipes in Pioneer is possible, a CSIRO report and subsequent discussion with the author state that lead levels should not reach levels dangerous to human health in a well flushed system
2. It is possible that with an aggressive water and stagnant and/or dead-end zones in the distribution system, lead levels could reach elevated levels

5 LINKS BETWEEN DATA AND CONCLUSIONS

Although the data reported by Harvey et al (2015) are sparse and their reproducibility can be questioned, the general conclusions reached in the paper are plausible. There are a number of factors which support this statement.

Firstly, sampling over a period of years by both TasWater and its predecessors has pointed to intermittent levels of lead in the Pioneer distribution system above 10 µg/l. It is the source of this lead which is the main issue under discussion. While data taken from the Pioneer Dam demonstrates very low levels of lead, there is clearly lead entering into the water supply somewhere in the distribution system. The most likely sources of this lead are:

- leaching from accumulated sediment in the distribution system;
- leaching from PVC piping;
- leaching from household plumbing.

An important factor which increases the likelihood of lead leaching is the aggressive nature of the raw water. With a pH ranging from 5.7 to 6.2 and an alkalinity of less than 2 mg/l, the water is clearly aggressive and capable of leaching metals from a variety of substrates. However, there are a number of features of the data provided by Harvey et al (2015) which complicate this picture.

There is the very strong link in the data between copper and lead levels, with copper levels in water samples always significantly higher than lead. The data also show that in the local soils and sediment, copper is always at a lower level than lead. This result suggests that the strong copper/lead relationship in the Harvey et al data is a result of water remaining in contact with brass and copper fittings for lengthy periods of time. If all the samples taken from both town and household taps were first flush samples, then the data would make sense. This conclusion again highlights the need for sampling protocols to include long flush times in order to ensure the samples accurately represent water in the distribution mains.

However, for the samples taken at the Pioneer town end flow valve (cnr of Main Road and Alfred Street) only a 5 second flush time was allowed. This flush time is probably too short to be confident of avoiding any copper/lead accumulation in the piping close to the valve. From examination of all the other town samples where high copper/lead concentrations were measured, it would appear that this lack of adequate flush times before sampling may have impacted on all of the author's results. The other result which adds to this concern is the significant difference between filtered and unfiltered samples for Pioneer houses 6 and 8 and the W20 sample (which appears to be from the Ringarooma River near the Frome Dam). The high particulate or insoluble levels of copper and lead in these samples suggests some copper/lead precipitation in the stagnant zone of the pipe after a long period of stagnation followed by re-suspension after the commencement of sampling. This is one possible explanation of why copper and lead levels went up in Pioneer House 11 after 3 minutes of flushing.

Considering the aggressive nature of the raw water, the leaching of lead from the PVC pipes in the distribution main is a distinct possibility. As described previously, Burn et al (2005), in their report to the Awwa Research Foundation do state that lead leaching from PVC pipes should not represent a health risk to humans, so long as proper flushing procedures are followed during commissioning of the pipe. However, given the aggressive nature of the raw water being supplied in the Pioneer system and the lead leaching results reported by Al Manack (2001) and Lasheen (2008), further testing would be required to validate this conclusion for the Pioneer system.

Key points

1. Although the data reported by Harvey et al (2015) are sparse and their reproducibility can be questioned, the general conclusions reached in the paper are plausible.
2. The strong copper/lead relationship in the Harvey et al data appears to be a result of water remaining in contact with brass and copper fittings for lengthy periods of time.
3. This conclusion again highlights the need for sampling protocols to include long flush times in order to ensure the samples accurately represent water in the distribution mains.

6 CONCLUSIONS

The water supply to the town of Pioneer has been disconnected from the Frome Dam and 900 metres of raceway immediately below the dam since at least 2009. Consequently, the water samples taken by Harvey et al (2015) from these sites are not directly applicable to the water supplied to Pioneer. The samples they took from around Pioneer Dam and within the town itself remain relevant.

Isotopic “fingerprinting” is a widely used tool for identifying the origin of various environmental contaminants and it has potential for identifying the sources of lead in drinking water. However, the paper by Harvey et al (2015) did not report enough appropriate samples of lead in both water and sediments and pipes for isotopic fingerprinting to unequivocally identify the sources of lead in the Pioneer water supply

Water samples from within the town distribution system taken by both Harvey et al (2015) and TasWater have clearly demonstrated the intermittent presence of lead in the water at levels approaching and above the recommended health limit of 10 µg/l. However, application of the data provided by Harvey et al. (2015) is limited by unclear sampling protocols and the lack of duplicate analyses and repeat samples.

Data on the quality of water within Pioneer Dam, although limited in number, indicate that it contains low levels of lead, significantly below those measured in the distribution system. However, the water has a low pH, is very low in alkalinity, and is thus conducive to leaching metals such as lead from any material it remains in contact with for an extended period of time.

Although the data provided in Harvey et al (2015) do not unequivocally identify the sources of lead in the Pioneer water supply, the evidence from all the data made available to the authors of this report indicate that there are three possible sources for the lead.

1. Sediments
 - a. In Pioneer Dam
 - b. In pipes within the town distribution system
2. –Pipes within the distribution system (mostly PVC)
3. – Household plumbing

Much of the data in Harvey et al (2015) appears to be based on first flush sampling of water after extended periods of stagnation in the pipes and local plumbing. These samples do not accurately represent the quality of the main body of water within the distribution system. The high levels of copper found associated with the lead in these samples support these conclusions.

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7.1.3 Australian Broadcasting Corporation Freedom of Information Request Documents

Justin Merrigan

From: Lance Stapleton
Sent: Thursday, 14 May 2015 2:41 PM
To: Justin Merrigan
Subject: RE: Email as per April 13th 2015 - Pioneer

Expires: Friday, 13 May 2016 12:00 AM

Hi Justin,

Re the approach from here on in....

Mac Uni Guys and Panel Review.

I'm proposing to contact Mark Taylor, not get caught in the he said/she said but update him on where we are at and where we're going with the review.
He seems to have some more data that isn't published but could be useful for our review.

Also I think it might be worthwhile us offering to share some of the findings of the panel with him in advance of release and try and invite him to work with us in terms of identification of commonality.
The panel would prefer this doesn't turn into a duelling scientists showdown and there are probably some areas of commonality we can all agree on.

Finding that middle ground that we are all prepared to "sing off the same sheet" is going to do wonders for our credibility and will shut down the media's main weapon.
Their commentary will be back to Jenny B and other fringe voices. They won't have a voice with credibility.

It also gives Mark Taylor a dignified way out of what could be a blow to his credibility.

How do you like this approach? That I call Mark and begin a chat and move on from the past (I'm reasonably clean on this to date) and start working with him on a slightly more consistent message.
I'd like to keep the panel clean from Mark until they've delivered their report.

Community engagement session

Stuart Heggie recommended this and I strongly support the idea that we have a facilitator run this session. The community will be raring to go and it could descend into chaos. Apart from anything else I don't want to expose the panel to abuse, not to mention our own people. I wonder if there's someone we can use who isn't taswater to run this show.

I've seen moderators before hand out numbers to people as they put up their hands and we answer the questions in turn. If number 5 tries to butt in ahead of number 2, the moderator asks them to wait until number 2 is answered etc. I'm not saying we do this at Pioneer but perhaps something similar just for a bit of crowd control. Provide some rules at

the start and we have half a chance of a sensible discussion. This one is probably one we need to run tight rather than all right on the night.

Also that we have a good turnout of TW people because it could get rowdy.

When you've got 5 - lets discuss but I want to get back to Mike Bon the mac uni stuff soon and start making calls.

10

Cheers,

Lance

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Department Manager Product Quality

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Have I been helpful? Please provide feedback by clicking [here](#).

Hi Guys,

Ive read this report and it looks like much is being made of very few samples. The science behind the assertions is very limited, selective samples have been taken and its in my mind a very very weak conclusion they have reached.

It is very wonky science and at this stage I think has truck sized holes in it.

The most obvious cause to me is that lead is leaching from tapware NOT our pipelines and I want to access the full dataset.

The one sample they took from our infrastructure (without our permission) that has high lead is almost certainly due to sediment.

To this end I think we need to contact the researchers and request their full data set and I want to question them on their sampling technique for the samples that the isotopic analysis has been conducted on.

I strongly suspect these were not taken in accordance with Australian standards for sampling.

I also want to engage external reviewers from another university to debunk their assertions.

It looks like they are drawing an incredibly long bow to make the findings they have. There are multiple obvious explanations as to the source of the lead.

In reading this report the MOST likely source is people's taps, not the pipes.

I don't think we can take this lying down and would appreciate your thoughts on coming out with a media statement tomorrow that casts doubt over their findings and criticises their moral legitimacy for sitting on sample results that could constitute a threat to public health

I also think we should contact mac uni management and engage our legal team.

After that roasting on 730 report I think its gone too far and we need to defend ourselves or at very least cast a cloud over their research and their ethics.

Read the ethical statement in their report that talks about "environmental sampling" its a load of hooey and as soon as they start sampling drinking water it is no longer an environmental sample. Morally they should have immediately contacted the director of public health and notified him of their findings and I think this needs to be pointed out.

Talk tomorrow but honestly, this has gone too far and to maintain credibility it might be time to hit back.

Cheer

Lance

Lance Stapleton
Department Manager Product Quality

<image001.png>

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7.2 Supplementary Data Study 2

Supplementary Data 1 – Sample results report mailed to participants of the study.



Healthy Water for Healthy Lives, Department of Earth and Planetary Sciences, Macquarie University

Dear Participant,

Below are the total metal concentrations results from the analysis of your water. Water was analysed by Inductively Coupled Plasma Mass Spectrometry – the gold standard of inorganic water analysis. The analysis of your water is subject to various limitations including collection method and holding time. We advise you that the analyses provided below are subject to technical limitations and should be taken as a guide to the quality of your water. It should be noted that water drawn from hot water heaters will have increased concentrations. Similarly, water drawn during winter may contain higher concentrations of elements.

Element	Analysed Concentration	Australian Drinking Water Guideline* (µg/L)
Arsenic	<1	10
Copper	3.7	2000
Lead	<1	10
Manganese	<1	500

*The complete version of these guidelines is available at <https://www.nhmrc.gov.au/guidelines/publications/eh52>.

Further information: Contact Paul Harvey (paul.harvey@mq.edu.au) or Professor Mark P. Taylor (mark.taylor@mq.edu.au).

Disclaimer All analyses or recommendations contained in this document are made in good faith and on the basis of information, national and international standards, measurements or guidelines supplied to or obtained by Macquarie University at the date of preparation of this document. You are strongly encouraged to seek independent medical advice or obtain independent testing of the water in your property prior to taking any action based on this report. Macquarie University, its officers, employees and contractors are not liable for any loss (including legal costs and expenses), injury or damage including economic loss and consequential loss or liability incurred or suffered by you or your successors in title or any other party acting or purporting to act in reliance on the contents of this report.

Supplementary Data 1 – First draw (unfiltered) water samples. Note: co-ordinates have an error to de-identify the sample site.

Sample	Arsenic (µg/L)	Copper (µg/L)	Lead (µg/L)	Manganese (µg/L)	X-axis co- ordinate	Y-axis co- ordinate
FD1	<1	2640.0	6.2	15.0	150.343940	-35.409588
FD2	<1	730.0	16.0	8.0	150.431931	-35.315843
FD3	<1	89.0	7.5	2.0	150.442593	-35.314636
FD4	<1	93.0	1.3	3.1	150.472059	-35.343324
FD5	<1	79.0	<1	2.3	150.440713	-35.262854
FD6	<1	40.0	<1	10.0	150.408895	-35.345518
FD7	<1	490.0	11.0	6.7	150.442593	-35.314640
FD8	<1	630.0	<1	1.4	150.071612	-35.919135
FD9	<1	440.0	2.9	10.0	150.434817	-35.314832
FD10	<1	150.0	1.3	17.0	150.432581	-35.328906
FD11	<1	140.0	1.4	8.5	150.438051	-35.318598
FD12	<1	220.0	<1	<1	150.077054	-35.920913
FD13	<1	71.0	1.9	2.7	150.469127	-35.331985
FD14	<1	600.0	1.2	<1	150.076641	-35.922310
FD15	<1	530.0	14.0	10.0	150.044420	-36.505064
FD16	<1	2150.0	2.8	9.1	150.070443	-36.431195
FD17	<1	29.0	<1	15.0	149.782514	-36.523828
FD18	<1	220.0	3.9	3.4	150.044308	-36.505146
FD19	<1	1910.0	4.7	3.7	149.890018	-36.388225
FD20	<1	630.0	9.3	140.0	149.840856	-36.676447
FD21	<1	1910.0	7.6	5.0	150.020866	-36.353862
FD22	<1	2.4	<1	<1	149.891141	-36.379686
FD23	<1	1350.0	5.6	4.1	149.892452	-36.390947
FD24	<1	350.0	2.2	<1	150.198155	-35.731706
FD25	<1	26.0	<1	2.5	150.130787	-36.056117

FD26	<1	280.0	<1	1.2	150.082702	-35.910315
FD27	<1	210.0	12.0	<1	150.188040	-35.737528
FD28	<1	790.0	<1	1.5	150.435317	-35.317451
FD29	<1	4.9	<1	6.0	150.230798	-35.686727
FD30	<1	210.0	2.0	<1	150.181553	-35.710412
FD31	<1	1680.0	1.6	6.8	149.765499	-36.826228
FD32	<1	190.0	1.1	42.0	149.976198	-36.727695
FD33	<1	89.0	<1	<1	149.824980	-36.457114
FD34	<1	680.0	8.1	94.0	150.074439	-36.433697
FD35	<1	87.0	<1	10.0	149.841910	-36.447944
FD36	<1	600.0	5.3	19.0	149.840874	-36.675285
FD37	<1	250.0	1.3	2.8	149.838642	-36.488434
FD38	<1	110.0	3.0	110.0	149.844714	-36.678480
FD39	<1	250.0	<1	<1	149.841910	-36.447952
FD40	<1	180.0	1.9	1.6	149.927738	-36.381438
FD41	<1	56.0	2.3	4.3	149.694333	-36.766360
FD42	<1	76.0	<1	6.4	149.845514	-36.680526
FD43	<1	310.0	6.4	6.4	150.076035	-36.429251
FD44	<1	13.0	<1	1.5	149.993514	-36.394847
FD45	<1	1100.0	<1	5.9	149.838656	-36.443005
FD46	<1	370.0	5.4	2.1	149.844401	-36.389422
FD47	<1	1330.0	13.0	7.4	149.887125	-36.388881
FD48	<1	26.0	2.3	11.0	145.828887	-31.510497
FD49	1.3	37.0	<1	<1	141.442619	-31.975145
FD50	<1	1200.0	1.1	23.0	146.723793	-31.553879
FD51	<1	2.5	<1	3.3	145.816474	-31.343677
FD52	1.1	660.0	<1	2.1	141.463958	-31.954178
FD53	<1	54.0	2.4	6.3	145.839069	-31.498373

FD54	1.2	190.0	4.9	1.3	141.452717	-31.943110
FD55	1.1	34.0	<1	1.6	141.467061	-31.938664
FD56	<1	2.3	<1	<1	141.461114	-31.950266
FD57	1.2	30.0	<1	1.2	141.435576	-31.965312
FD58	1.1	39.0	<1	1.5	141.461435	-31.943665
FD59	<1	23.0	5.5	12.0	141.464804	-31.976440
FD60	1.2	88.0	1.4	1.7	141.475123	-31.941873
FD61	1.3	65.0	<1	1.3	141.459518	-31.946421
FD62	1.1	29.0	<1	1.4	141.465373	-31.939887
FD63	1.1	95.0	1.4	1.1	141.464863	-31.976024
FD64	1.3	41.0	<1	<1	141.445274	-31.969690
FD65	1.2	18.0	<1	14.0	141.480179	-31.938754
FD66	<1	840.0	5.3	14.0	141.480179	-31.938754
FD67	<1	58.0	<1	11.0	149.568588	-33.422038
FD68	<1	190.0	<1	16.0	149.571940	-33.433132
FD69	<1	16.0	<1	80.0	149.571940	-33.433132
FD70	<1	6.6	<1	46.0	149.571162	-33.407849
FD71	<1	210.0	1.3	45.0	149.574872	-33.431936
FD72	<1	50.0	<1	46.0	149.590372	-33.431151
FD73	<1	130.0	1.3	70.0	149.581390	-33.408377
FD74	<1	230.0	1.1	36.0	149.580053	-33.425463
FD75	<1	270.0	5.0	20.0	149.573280	-33.420860
FD76	<1	160.0	<1	12.0	149.603889	-33.404677
FD77	<1	180.0	<1	6.1	149.573076	-33.411688
FD78	<1	96.0	1.3	7.8	149.568917	-33.404583
FD79	<1	630.0	1.1	28.0	147.286755	-34.338227
FD80	<1	78.0	1.7	7.7	150.469813	-35.315413
FD81	<1	52.0	<1	4.1	150.436996	-35.316906
FD82	<1	75.0	1.0	6.6	150.471853	-35.318519

FD83	<1	69.0	<1	2.9	150.436996	-35.316906
FD84	<1	670.0	<1	2.9	150.435317	-35.317451
FD85	<1	91.0	<1	5.7	150.468992	-35.350628
FD86	<1	130.0	6.7	3.6	150.458440	-35.349032
FD87	<1	12.0	<1	3.7	150.469813	-35.315413
FD88	<1	86.0	1.1	6.2	150.463386	-35.266902
FD89	<1	16.0	<1	4.3	150.446089	-35.340154
FD90	<1	250.0	2.5	<1	148.646228	-36.404683
FD91	1.1	29.0	1.4	7.4	147.221803	-34.351943
FD92	<1	7.6	6.1	22.0	147.177993	-34.406379
FD93	<1	6.9	<1	13.0	147.177993	-34.406379
FD94	<1	6.4	3.4	19.0	147.177993	-34.406379
FD95	<1	220.0	<1	<1	149.358172	-35.231802
FD96	<1	520.0	4.9	8.7	147.190245	-34.248041
FD97	<1	120.0	<1	10.0	147.221011	-34.354892
FD98	<1	2040.0	3.0	14.0	149.352940	-35.233465
FD99	<1	2040.0	3.8	24.0	149.411724	-35.230556
FD100	<1	28.0	2.2	1.4	149.350552	-35.141837
FD101	<1	710.0	4.4	1.1	148.826206	-36.360957
FD102	<1	43.0	21.0	21.0	148.654669	-36.462721
FD103	<1	360.0	<1	55.0	148.654669	-36.462721
FD104	<1	29.0	<1	<1	148.510537	-36.448503
FD105	<1	37.0	5.7	6.0	148.510537	-36.448503
FD106	<1	5.4	<1	2.3	148.470672	-34.118867
FD107	<1	1510.0	1.8	5.0	148.301744	-34.312691
FD108	<1	140.0	2.0	10.0	149.442852	-35.254299
FD109	<1	11.0	10.0	1.7	147.220323	-34.348012
FD110	<1	200.0	3.4	4.9	152.160553	-32.715804

FD111	<1	98.0	5.3	11.0	149.770182	-30.336249
FD112	<1	410.0	<1	2.5	149.773037	-30.311642
FD113	<1	3.4	<1	57.0	149.792317	-30.319219
FD114	<1	1130.0	6.5	10.0	149.770976	-30.315990
FD115	<1	380.0	<1	6.1	149.770255	-30.317671
FD116	<1	220.0	2.6	21.0	149.768378	-30.314567
FD117	<1	270.0	16.0	4.8	149.787593	-30.326842
FD118	<1	5.8	<1	7.8	149.847915	-30.350228
FD119	<1	100.0	<1	2.0	149.793012	-30.331735
FD120	<1	130.0	3.3	6.6	149.798773	-30.113541
FD121	<1	36.0	<1	<1	150.010208	-30.317059
FD122	<1	2170.0	20.0	15.0	149.782780	-30.319946
FD123	<1	83.0	16.0	9.6	150.043468	-30.703515
FD124	<1	99.0	1.4	1.4	149.212559	-30.482101
FD125	<1	610.0	4.4	5.5	149.783240	-30.326623
FD126	5.8	78.0	<1	2.2	149.783240	-30.326623
FD127	<1	1060.0	89.0	18.0	149.850949	-30.357257
FD128	<1	530.0	1.1	<1	149.850949	-30.357257
FD129	<1	3490.0	5.7	2.0	149.754469	-30.339252
FD130	<1	440.0	<1	<1	149.754435	-30.339220
FD131	<1	180.0	4.1	16.0	149.860769	-30.354218
FD132	<1	230.0	4.0	24.0	149.790766	-30.332255
FD133	<1	320.0	1.7	52.0	148.888478	-30.354665
FD134	<1	70.0	6.7	13.0	148.888478	-30.354665
FD135	<1	3700.0	4.1	4.3	149.832488	-30.361471
FD136	<1	30.0	<1	<1	149.869356	-30.363387
FD137	<1	5.7	<1	56.0	149.792227	-30.319060
FD138	<1	3800.0	3.7	47.0	149.692397	-30.545038
FD139	<1	15.0	<1	4.9	149.740950	-30.372329

FD140	<1	2060.0	35.0	12.0	149.786616	-30.324843
FD141	<1	1120.0	1.5	8.3	149.779336	-30.328984
FD142	<1	20.0	<1	6.2	149.717821	-30.273496
FD143	<1	34.0	3.2	14.0	149.666543	-30.306389
FD144	<1	<1	<1	9.4	149.517832	-30.442995
FD145	<1	8.0	3.2	9.9	149.792957	-29.917639
FD146	<1	130.0	1.8	2.1	149.776057	-30.330082
FD147	<1	270.0	64.0	82.0	149.776057	-30.330082
FD148	<1	100.0	1.6	<1	149.783012	-30.323520
FD149	<1	47.0	20.0	2.5	150.481450	-35.322189
FD150	<1	63.0	<1	4.5	150.471790	-35.346013
FD151	<1	66.0	1.2	<1	150.408837	-35.367217
FD152	<1	8.0	<1	2.5	150.463641	-35.321045
FD153	<1	160.0	<1	1.9	150.472456	-35.343051
FD154	<1	40.0	<1	2.5	150.469223	-35.311572
FD155	<1	29.0	<1	1.1	150.479723	-35.364903
FD156	<1	3.7	<1	<1	150.432191	-35.235998
FD157	<1	39.0	<1	3.0	150.447417	-35.345965
FD158	<1	10.0	<1	<1	150.412110	-35.347426
FD159	<1	280.0	<1	<1	150.459223	-35.281220
FD160	<1	560.0	1.3	1.9	150.380124	-35.380932
FD161	<1	190.0	<1	2.3	150.482123	-35.323437
FD162	<1	29.0	<1	1.2	150.442511	-35.390525
FD163	<1	11.0	1.1	1.3	150.443697	-35.322637
FD164	<1	61.0	3.9	1.6	150.480726	-35.366057
FD165	<1	280.0	1.7	2.8	150.469863	-35.318673
FD166	<1	510.0	7.0	<1	150.480727	-35.366058
FD167	<1	27.0	1.9	<1	150.369191	-35.508351

FD168	<1	660.0	1.2	<1	150.389785	-35.507130
FD169	<1	46.0	3.1	<1	150.442586	-35.314662
FD170	<1	100.0	<1	1.3	150.444750	-35.322799
FD171	<1	11.0	<1	1.3	150.415056	-35.335677
FD172	<1	280.0	<1	2.0	150.407864	-35.292553
FD173	<1	93.0	15.0	4.8	150.464637	-35.350302
FD174	<1	56.0	<1	1.2	150.440955	-35.383541
FD175	<1	162.5	2.0	2.9	150.904590	-31.027604
FD176	<1	1.5	<1	<1	150.744677	-31.262148
FD177	<1	18.8	4.5	17.5	150.904590	-31.027604
FD178	<1	337.5	<1	<1	150.911878	-31.162446
FD179	<1	187.5	<1	2.4	150.744677	-31.262148
FD180	<1	337.5	3.4	6.5	150.952514	-31.168052
FD181	<1	162.5	3.9	9.1	150.976098	-30.996813
FD182	<1	5.5	<1	17.5	151.047445	-31.002647
FD183	<1	1137.5	3.0	12.0	150.877780	-31.039726
FD184	<1	362.5	5.6	7.1	151.002147	-31.119843
FD185	<1	112.5	4.1	7.9	151.065695	-31.017401
FD186	<1	73.8	<1	<1	152.922817	-30.733606
FD187	<1	2325.0	22.5	50.0	150.860757	-31.089743
FD188	<1	62.5	<1	1.5	151.054730	-31.053868
FD189	<1	108.8	4.3	8.6	150.860757	-31.089743
FD190	<1	312.5	27.5	46.3	150.944859	-31.085506
FD191	<1	38.8	<1	<1	151.649452	-30.528794
FD192	<1	675.0	2.3	1.3	151.683457	-30.489914
FD193	<1	137.5	<1	2.8	151.660018	-30.505464
FD194	1.5	121.3	11.6	10.4	151.654343	-30.489226
FD195	<1	450.0	5.0	<1	151.671656	-30.497478
FD196	<1	362.5	2.8	1.5	151.662838	-30.544508

FD197	<1	450.0	<1	<1	151.653895	-30.523866
FD198	<1	20.0	<1	<1	151.686564	-30.492638
FD199	<1	68.8	1.4	<1	151.650735	-30.529161
FD200	<1	175.0	6.1	40.0	151.611283	-30.486215
FD201	<1	187.5	<1	<1	152.911316	-30.730339
FD202	<1	137.5	<1	<1	152.911316	-30.730339
FD203	<1	123.8	1.6	1.4	152.918249	-30.711356
FD204	<1	63.8	<1	<1	152.907061	-30.729446
FD205	<1	37.5	<1	<1	152.906662	-30.729893
FD206	<1	63.8	<1	<1	152.979942	-30.645225
FD207	<1	15.0	<1	2.5	152.799267	-31.066347
FD208	<1	63.8	<1	2.0	152.761629	-31.041494
FD209	<1	61.3	<1	1.3	152.761533	-31.041368
FD210	<1	150.0	<1	<1	153.005481	-30.592626
FD211	<1	406.3	6.3	3.1	148.609537	-32.243800
FD212	<1	843.8	4.8	4.7	148.609537	-32.243800

Supplementary Data 2 – Duplicate water samples.

Sample	Arsenic (µg/L)	Copper (µg/L)	Lead (µg/L)	Manganese (µg/L)	X-axis co- ordinate	Y-axis co- ordinate
FD28	<1	790.0	<1	1.5	150.435317	-35.317451
FD84	<1	670.0	<1	2.9	150.435317	-35.317451
FD201	<1	187.5	<1	<1	152.911316	-30.730339
FD202	<1	137.5	<1	<1	152.911316	-30.730339
FD204	<1	63.75	<1	<1	152.907061	-30.729446
FD205	<1	37.5	<1	<1	152.907061	-30.729446
FD208	<1	63.8	<1	2.0	152.761629	-31.041494
FD209	<1	61.3	<1	1.3	152.761629	-31.041494
FD3	<1	89.0	7.5	2.0	150.442593	-35.314636
FD169	<1	46.0	3.1	<1	150.442593	-35.314636
FD211	<1	325.0	5.0	2.5	148.609537	-32.243800
FD212	<1	675.0	3.9	3.8	148.609537	-32.243800

Supplementary Data 3 – Flushed (2 minutes) water samples.

Sample	Arsenic (µg/L)	Copper (µg/L)	Lead (µg/L)	Manganese (µg/L)	X-axis co- ordinate	Y-axis co- ordinate
FD28	<1	670.0	0.5	2.9	150.435317	-35.317451
FD28FL	<1	230.0	0.5	7.0	150.435317	-35.317451
FD98	<1	2040.0	3.0	14.0	149.35294	-35.233465
FD98FL	<1	280.0	1.9	16.0	149.352939	-35.233464
FD107	<1	1510.0	1.8	5.0	148.301744	-34.312691
FD107FL	<1	1080.0	2.2	6.3	148.301744	-34.312691
FD108	<1	140.0	2.0	10.0	149.442852	-35.254299
FD108FL	<1	20.0	0.5	<1	149.442852	-35.254299
FD109	<1	11.0	10.0	1.7	147.220323	-34.348012
FD109FL	<1	2.1	1.1	4.4	147.220323	-34.348012
FD183	<1	1137.5	3.0	12.0	150.87778	-31.039726
FD183FL	<1	525.0	5.8	10.3	150.87778	-31.039726
FD184	<1	362.5	5.6	7.1	151.002147	-31.119843
FD184FL	<1	187.5	4.6	7.6	151.002147	-31.119843
FD187	<1	2325.0	22.5	50.0	150.860757	-31.089743
FD187FL	<1	108.8	4.3	8.6	150.860757	-31.089743
FD190	<1	312.5	27.5	46.3	150.944859	-31.085506
FD190FL	<1	325.0	150.0	12.5	150.944859	-31.085506
FD189	<1	108.8	4.3	8.6	150.860757	-31.089743
FD189FL	<1	36.3	1.8	9.4	150.860757	-31.089743

Supplementary Data 4 – Case Study Samples.

Sample	Lead (µg/L)
CS1B	90.0
CS1G	9.9
CS1K	36.3
CS1T	<1
CS2B	26.3
CS2G	23.8
CS2K	3.0
CS2T	<1
CS3B	43.0
CS3G	3.4
CS3K1	6.5
CS3K2	13.0
CS3T	<1
CS4T	14.0

Supplementary Data 5 – Laboratory leaching experiment samples.

a. Copper riser and tap.

Sample	Copper (µg/L)						
	Day 1	Day 2	Day 3	Day 4	Day 5	Day 6	Day 7
Fitting 1	448.2	346.5	259.6	308.9	251.9	244.5	267.4
Fitting 2	440.5	460.7	461.7	371.0	293.2	321.7	316.5
Fitting 3	885.3	649.2	602.6	579.3	403.8	515.2	389.7
Fitting 4	378.4	360.0	389.5	481.7	337.1	467.9	754.6
Sample	Lead (µg/L)						
	Day 1	Day 2	Day 3	Day 4	Day 5	Day 6	Day 7
Fitting 1	1437.5	500.0	250.0	225.0	150.0	137.5	107.5
Fitting 2	650.0	512.5	325.0	287.5	200.0	212.5	187.5
Fitting 3	750.0	362.5	237.5	212.5	150.0	175.0	137.5
Fitting 4	712.5	337.5	225.0	237.5	150.0	137.5	175.0

b. Tap only.

Sample	Lead (µg/L)		
	Day 1	Day 2	Day 3
Fitting 1	<1	<1	<1
Fitting 2	59	58	56
Fitting 3	90	71	55
Fitting 4	61	59	62

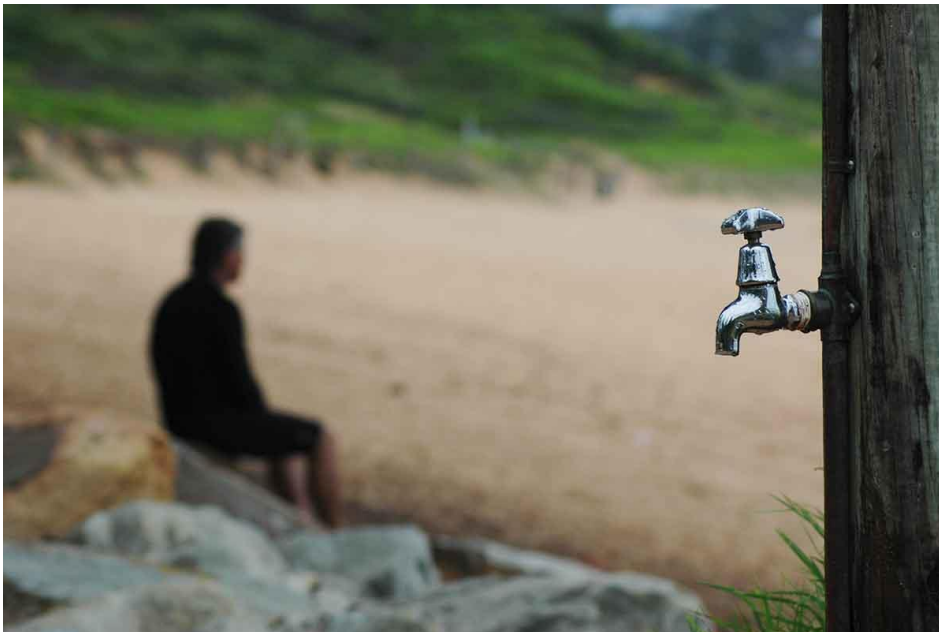
7.2.2 Examples of the extensive media coverage in print, television and radio following the study



<https://www.newscientist.com/article/2100806-lead-in-australias-drinking-water-is-leaching-from-brass-taps/>

11 August 2016

Lead in Australia's drinking water is leaching from brass taps



Jade and Bertrand Maitre/Getty

By **Alice Klein**

The proof is in the plumbing. Low-level lead contamination of water is widespread in Australian homes, with brass tap fittings the most likely source.

This is the finding of a large-scale study on water samples from kitchen taps in 212 homes dotted across New South Wales, the state that contains Sydney.

Authorities around the world routinely monitor drinking water in storage reservoirs and distribution pipes to ensure that lead levels are below guideline limits. But lead from brass kitchen taps can enter water further down the supply chain, says [Paul Harvey](#) at Macquarie University in Sydney.

In their study, Harvey and his colleagues detected lead in 56 per cent of samples – and 8 per cent of those exceeded the limit of 10 micrograms per litre set in the Australian Drinking Water Guidelines.

In a subsequent experiment, the researchers tested water before and after it [passed through brass](#) taps, which contain small amounts of lead, and lead-free stainless steel taps. Lead was only found in water that had passed through brass ones.

“Water sits inside the brass barrel of your tap, where it can be exposed to lead for a long time, especially if it sits there overnight,” Harvey says.

Toxic effects

Lead is a neurotoxin that is particularly harmful to children. Studies have linked low-level lead exposure to [reductions in IQ](#) and attention span, as well as a greater tendency for antisocial behaviour.

Australia’s 10-microgram lead limit for drinking water was set in the mid-1990s, but the World Health Organization has since deemed that [no level of exposure is safe](#).

The researchers are yet to test whether higher lead levels in home water supplies correlate with higher amounts in the blood and adverse health effects, but Harvey believes there is potential for harm.

“Having seen what I’ve seen in terms of the concentrations in the water and the widespread nature of it, it is raising the alarm,” he says.

A handful of samples contained almost 90 micrograms of lead per litre – comparable to the levels recorded during the [recent water crisis in Flint, Michigan](#), after distribution pipes began to corrode.

However, the Australian results may be unrepresentative because they were collected after taps had been off for 9 hours, says [David Simon](#) of the South Australian health department, who sat on the most recent National Health and Medical Research Council lead working committee.

“Generally, people consume water over the whole day,” he says. “Therefore, consumption of contaminants would be lower than indicated by the study, which measured worst-case levels after 9 hours of stagnation.”

Reducing exposure

Evidence of small amounts of lead leaching from taps has also been found in the [US](#) and the [UK](#). In 2014, the US government mandated a [lead limit of 0.25 per cent in plumbing fittings](#).

Taps in Australia and the UK are typically made of brass that contains lead at a level of about [2 to 4 per cent](#).

Introducing similar restrictions to those in the US would be a simple way to cut down on unnecessary exposure levels, even if they are low, says Harvey.

Consumers can also attach lead filters to their taps or switch to stainless steel, he says.

Journal reference: *Environmental Research*, DOI: [10.1016/j.envres.2016.07.041](https://doi.org/10.1016/j.envres.2016.07.041)

Read more: [Toilet to tap: Drinking water at the press of a flush](#)



<http://www.sbs.com.au/topics/science/humans/article/2016/08/11/widespread-lead-contamination-domestic-tap-water-found-nsw>

11 AUG 2016 - 2:46PM

‘Widespread’ lead contamination of domestic tap water found in NSW



(Pixabay)

Researchers believe that similar issues in domestic kitchen taps are likely across Australia.

By
Signe Dean

11 AUG 2016 - 2:46 PM UPDATED 11 AUG 2016 - 2:46 PM

A new study indicates that some Australians could be getting a dangerous level of lead contamination from their kitchen taps.

Researchers from Macquarie University have detected copper and lead contaminants in domestic water samples across New South Wales - and in some cases the levels exceed what's listed as acceptable in the [Australian Drinking Water Guidelines](#) set by the National Health and Medical Research Council.

Back in 2014, the researchers enlisted volunteer help from the community to gather domestic water samples. The results were published this week in [Environmental Research](#).

Problem lies with taps, not water supply

"The water supply is fine. The issue is the water in the house," says study co-author [Professor Mark Taylor](#).

"We were aware that domestic water may be accidentally contaminated through lead use in brass fittings, so to characterise that we wanted to know what the extent of in-house contamination was."

The water samples were collected after water sat in a tap for a nine-hour 'stagnation period', similar to what happens when you run the tap first thing in the morning to fill the kettle.

"My results show that there is quite a significant concentration of lead and copper in the drinking water that is coming out of people's kitchen taps into their morning cup of tea, or baby formula," says lead author of the study, PhD researcher Paul Harvey.

The team tested 212 such 'first draw' samples, and found copper in nearly all of them, while lead was present in 56 per cent of the households tested. According to guidelines, drinking water should contain no more than 10 micrograms of lead per litre - but 8% of the lead samples contained higher levels than that.

"The highest concentration sample that I collected in this study was 89 micrograms [of lead] per litre," says Harvey. "The results of this study demonstrate that along with other potential sources of contamination in households, plumbing products that contain up to 2.84 per cent of detectable lead are contributing to contamination of household drinking water."

A potentially widespread problem

The researchers believe their results could be extrapolated to the population of the whole country - making it a considerable health concern.

"Eight per cent may not sound like a big number, but if it is eight per cent of the Australian population, we're talking about a lot of people," says Harvey.

"The sampling method appears robust and is analogous to that used in the United States," says [Professor Marc Edwards](#) from Virginia Tech in the US, a leading water quality expert who was not involved in this study.

"While at one level collecting about 200 samples and finding 8% above Australian guidelines would appear to be a stretch in identifying a problem as widespread, in this case it merely confirms the obvious - Australian plumbing systems are subject to the exact same problems found elsewhere in the world," he adds.

Health concerns from lead

Lead is well known for its harmful effects on the human body, and [according to the NHMRC](#), infants, children, and pregnant women are at the greatest risk. Drinking water with high levels of lead can [lead to its accumulation in the body](#), posing particular risks to children's brain development even when there are no obvious signs of poisoning. Copper contamination is a less severe problem, but prolonged exposure can lead to liver damage.

"Something should be done to make consumers aware of the problem and avoid irreversible health harm," states Edwards, who was involved in uncovering the [high levels of lead contamination](#) in the water supply of Flint, Michigan.

"The idea is not to set fear in everybody's minds and to tell them to stop drinking water. We want people to be vigilant and be aware that these things can be in the water," Harvey explains.

In response to this study, a NSW Health spokesperson tells SBS Science that "copper and lead dissolving from plumbing fixtures into drinking water has been recognised for a long time and is addressed in fact sheets for these compounds in the Australian Drinking Water Guideline."

According to [NSW Health drinking water recommendations](#), if water is left standing in the pipes for an extended time, it's advisable to flush it from your tap for 2-3 minutes, and use that water for non-food related purposes, such as watering your plants.

"Despite following this established protocol, some of the flushed water samples still contained considerable concentrations of both copper and lead," the researchers write.

Collecting water in rainwater tanks poses a higher risk of contamination, so NSW Health advises consumers to [manage their tanks properly](#) to avoid these risks.

It can be difficult to discern whether the plumbing in your house might be contaminating your water, so researchers are advising consumers to be cautious - especially if the family has young children.

"To mitigate the risks, perhaps you might buy a filter that is efficient at removing heavy metals, for example," suggests Harvey.

They are also advising worried consumers to get in touch with their water supplier or the local council to see whether water testing at the household level is available. If lead is confirmed to be a problem, changing taps to lead-free ones could be a necessary step - however, that incurs a higher cost.

"If you can afford it, replace your taps - at least the faucets where you drink your tap water from," Taylor is blunt. "We know it's a problem, we know lead's not good for you, so why not stop it?"

7.3 Supplementary Data Study 3

APPENDICES

Supplementary Figure 1: VegeSafe soil sampling instructions for the collection of garden soil samples.



VegeSafe - We love your dirt!

Sample locations at your property:

We would like you to take 5 samples from 3 sites (i.e. a total of 5 soil samples) from your garden area. Collect the soils when dry if you can. This will help us speed up the analysis on the day.

Site 1: Identify 3 places for soil sampling either where you already have vegetable beds or where you plan to start your veggie patch.

Site 2: Identify another sample site such as your front or rear garden or if you don't have a these as unpaved areas, a site on or adjacent to the roadside verge.

Site 3: Identify a sample site below the drip line of your gutter, close to your house.

How to sample and store your soil sample:


- At each site, use the trowel or scoop to take soil from the top 2 cm of the sampled area, i.e. the surface soil. If there is grass or other vegetation cover, you will need to remove it and take the soil from underneath.
- Collect about a cricket ball size amount of soil (more is better than less) from an area of ~ 30 x 30 cm, or up to a 1 m² if required.
- Put the soil sample in a **ziplock bag** and seal it. Don't worry about grass or sticks or rocks—we will remove these when we prepare the soil for testing.
- Label the ziplock bags with a permanent marker and include the site location and brief description, date the sample was collected, the address and the postcode. Then put the bagged sample inside another ziplock bag – this protects the label and helps protect against leakage if the inner bags splits.
- Labeling – make sure the label makes sense to you so you can explain it to us when we receive your sample e.g. veggie patch sample 1; veggie patch sample 2, front yard soil; dripline soil north side of house, etc. Below is an example of a labeled bag.



<https://research.science.mq.edu.au/vegesafe/index.html>

Updated August 21st 2015

Supplementary Figure 2: VegeSafe consent form to be completed before samples are screened.



VegeSafe Consent Form

VegeSafe web page:
<http://research.science.mq.edu.au/vegesafe/>
 or our Facebook page:
<facebook.com/pages/VegeSafe/571316076267515>

Thank you very much for participating in the VegeSafe soil testing program provided by Environmental Science Staff at Macquarie University.

As part of your participation in this program, we would like to be able to use the results to produce a map showing the information. The map data will be de-identified to the extent that the locations will only be accurate to within 25 m and no address or other information will be included that can be used to specifically identify the precise locations of the soil samples. We may also use the same results, with your permission, in future publications, again with the aforementioned limitations. Therefore, in signing this form you acknowledge consent for these purposes.

We have run our project past the National Health and Medical Research Council Ethics and they have informed us as follows:

"As collection of the soil samples and proposed analysis do not relate specifically to human health, medicine or human research, the National Statement on Ethical Conduct in Human Research does not apply." (NHMRC Ethics, June 2013).

We will provide you with a summary of the results and some links to documentation to provide guidance on soils and potential lead and other metal exposure.

Donations - We Need Your Support

VegeSafe currently operates as a Community Service and runs on the good will of staff and students. However, tax deductible donations to support our program can be made via the following link: MQ.edu.au/supporting or by sending in with the sample parcel. If your donation is specifically for our program please don't forget to register that fact when giving your support.

Name: _____ **Email address:** _____

Address of soil samples: _____

Primary construction material of property: _____
 (e.g. double brick, timber, fibro, etc.)

Any exterior walls of the property painted?:

Approximate age of property: _____ years

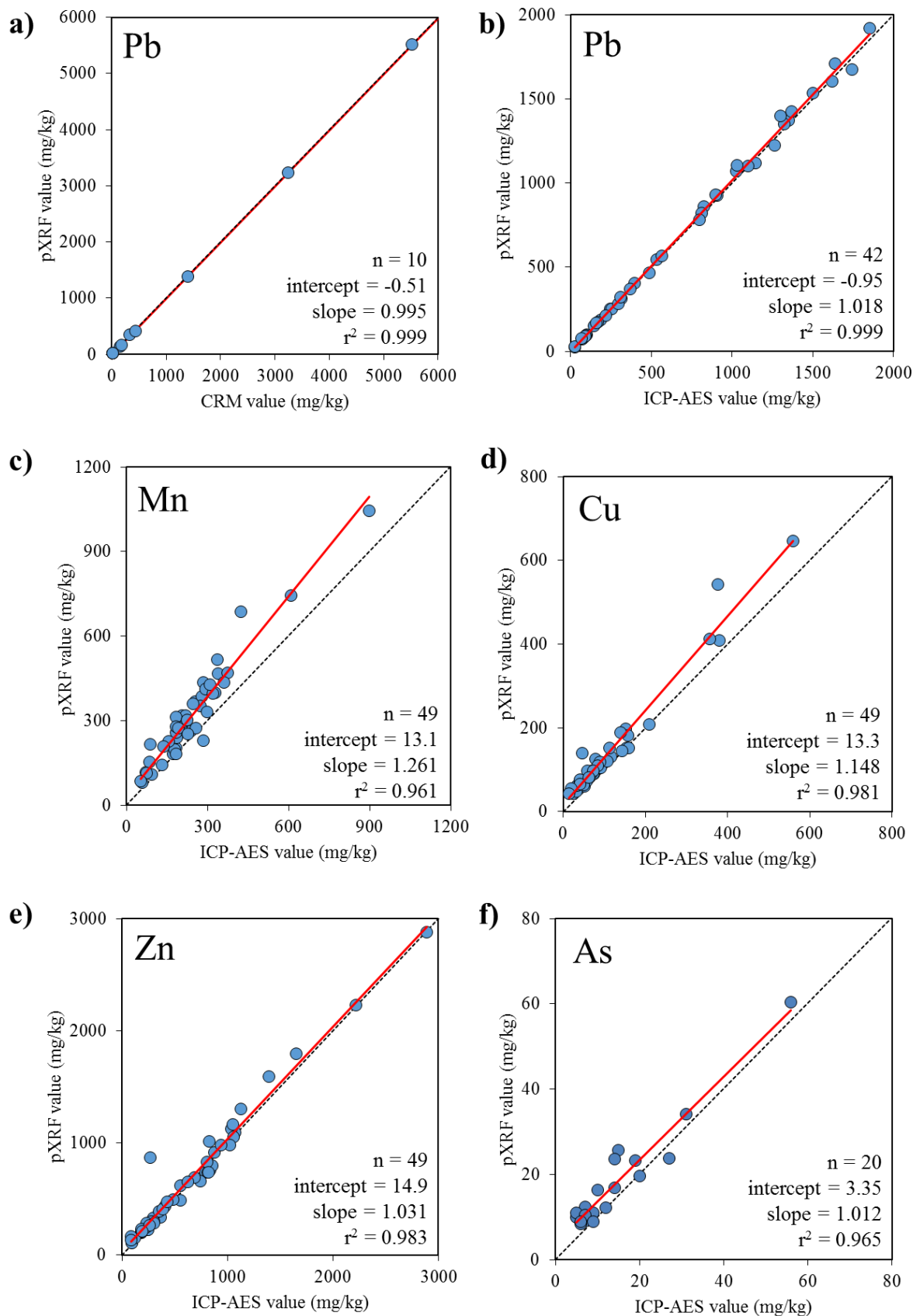
Signature _____

Date: _____

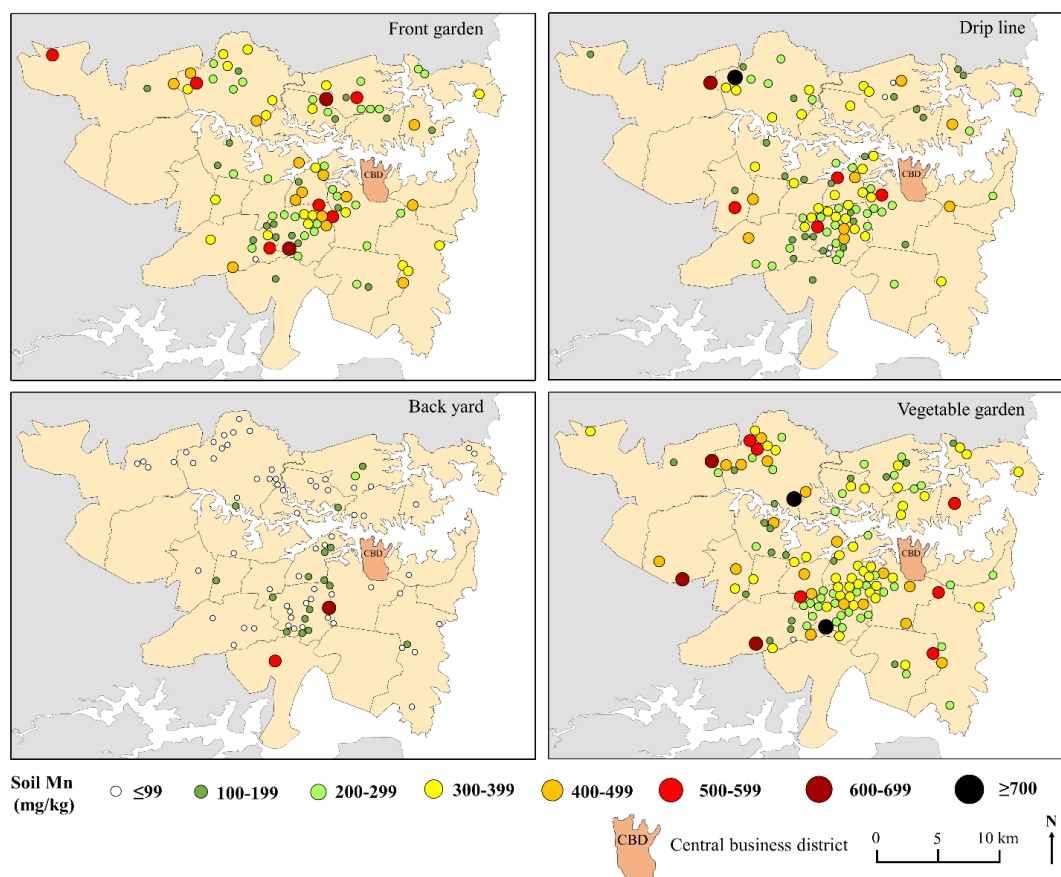
Sample code (A, B, C, D, E)	Sample description (e.g. Veggie patch 1)

Updated August 21st 2015

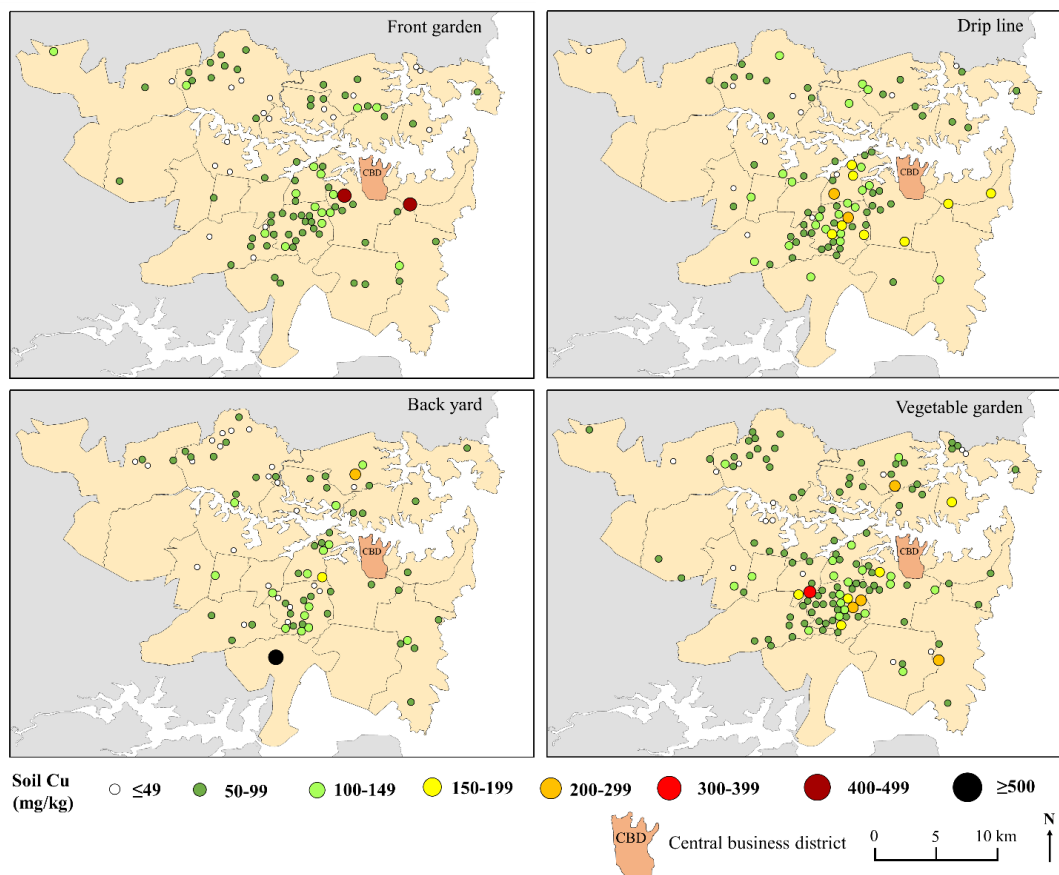
Supplementary Figure 3: Comparison of pXRF soil metal concentration data to a) certified reference materials for Pb and ICP-AES laboratory data for b) Pb, c) Mn, d) Cu, e) Zn and f) As. Regression relationship (solid red line) and 100% recovery (dashed black line) are shown in each plot.



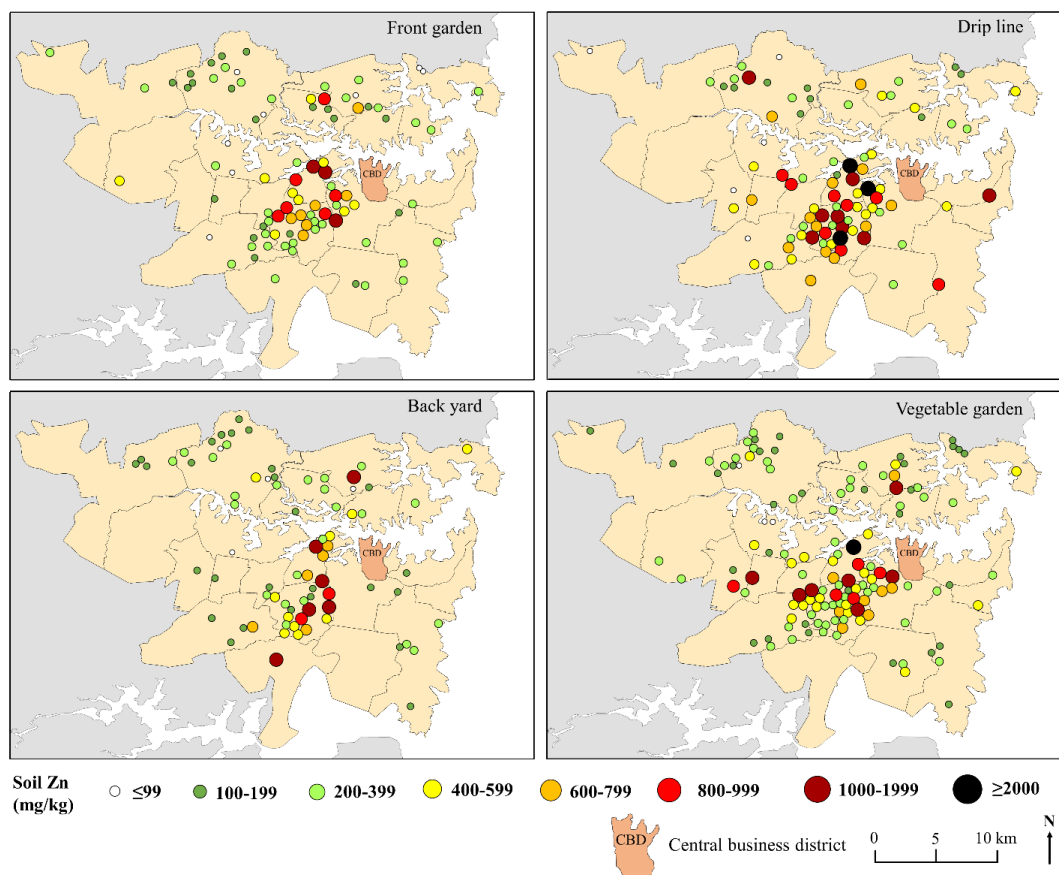
Supplementary Figure 4: Soil manganese concentrations in front yard (n=92), drip line (n=97), back yard (n=80) and vegetable gardens (n=141) areas in private residences of Sydney, Australia.



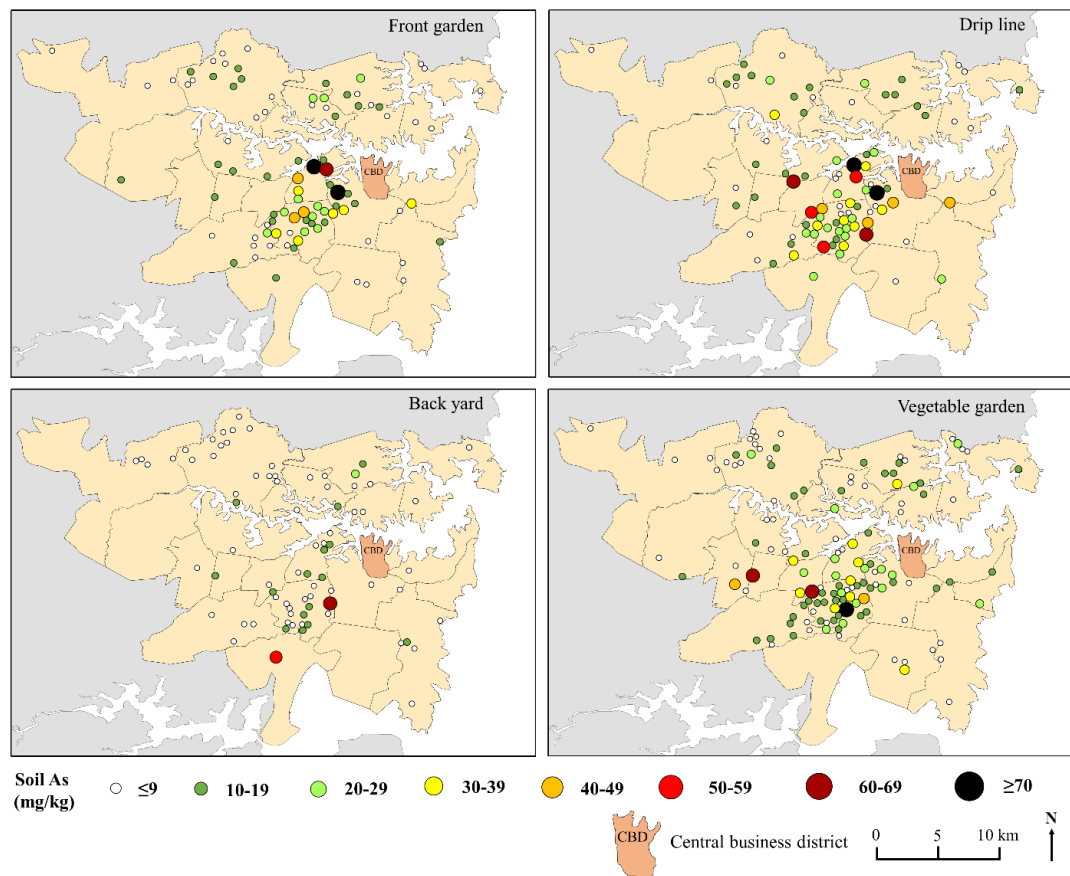
Supplementary Figure 5: Soil copper concentrations in front yard (n=92), drip line (n=97), back yard (n=80) and vegetable gardens (n=141) areas in private residences of Sydney, Australia.



Supplementary Figure 6: Soil zinc concentrations in front yard (n=92), drip line (n=97), back yard (n=80) and vegetable gardens (n=141) areas in private residences of Sydney, Australia.



Supplementary Figure 7: Soil arsenic concentrations in front yard (n=92), drip line (n=91), back yard (n=79) and vegetable gardens (n=140) areas in private residences of Sydney, Australia



Supplementary Figure 8: Example of VegeSafe soil screening report.



VegeSafe, Environmental Science, Macquarie University

Dear VegeSafe participant:

Below are the total metal concentrations results from the X-ray fluorescence analysis of your soils. The analysis of your soil is subject to various limitations including moisture content of the soil, its grain size and organic content. In a standard laboratory test additional and more intensive preparation of your soil samples would occur allowing greater reliability of the results. Therefore, we advise you that the analyses provided below are subject to these technical limitations and should be taken as a guide to the metal content of your soil. If you require more detailed analyses we recommend that you submit your samples to a laboratory accredited by NATA, which is the National Association of Testing Authorities – <http://www.nata.asn.au/facilities-and-labs>. We have made a short guide of how to deal with soil that may be contaminated with elevated lead or other metals – this can be found at the following website: <http://research.science.mq.edu.au/vegesafe/advice-on-soil/>.

Sample ID	Soil Location	As (mg/kg)	Cd (mg/kg)	Cr (mg/kg)	Cu (mg/kg)	Mn (mg/kg)	Pb (mg/kg)	Ni (mg/kg)	Zn (mg/kg)
A	Veggie patch 1	0	0	0	17	119	69.7	0	87
B	Veggie patch 2	0	0	30	42	141	225	0	315
C	Rear yard	12	0	21	46	189	245	0	341
D	Under eave / deck	29	0	45	136	161	609	19	718
E	Front verge (Chandos Street)	12	0	27	86	215	267	13	428

As – arsenic; Cd – cadmium; Cr – chromium; Cu – copper; Mn – manganese; Pb – lead; Ni – nickel; Zn – zinc

Element (mg/kg)	Australia NEPC ¹ (2013)			California CEPA ² (2005)	Canada CCME ³ (2013)	Norway NPCA ⁴ (2009)	United States EPA ⁵ (2013)
	Residential A	Residential B	Recreational C				
Arsenic	100	500	300	0.07	12	12	0.61
Cadmium	20	150	90	1.7	10	12.2	7
Chromium VI	100	500	300	17	0.4	75	0.29
Copper	6,000	30,000	17,000	3,000	63	150	310
Lead	300	1,200	600	80*	140	90	400
Manganese	3,800	14,000	19,000	-	-	-	180
Nickel	400	1,200	1,200	1,600	50	90	150
Zinc	7,400	60,000	30,000	23,000	200	300	2,300

The complete version of these guidelines along with links to the guidelines is available at <http://research.science.mq.edu.au/vegesafe/advice-on-soil/>. Residential A = with garden/accessible soil; Residential B = with minimal access to soil; Recreational C = public open space

Further information: Contact Professor Mark P. Taylor (mark.taylor@mq.edu.au), A/Professor Damian Gore (damian.gore@mq.edu.au) or Marek Rouillon (marek.rouillon@mq.edu.au)

Disclaimer All analyses or recommendations contained in this document are made in good faith and on the basis of information, national and international standards, measurements or guidelines supplied to or obtained by Macquarie University at the date of preparation of this document. You are strongly encouraged to seek independent medical advice or obtain independent testing of the soil in your property prior to taking any action based on this report. Macquarie University, its officers, employees and contractors are not liable for any loss (including legal costs and expenses), injury or damage including economic loss and consequential loss or liability incurred or suffered by you or your successors in title or any other party acting or purporting to act in reliance on the contents of this report.

7.4 Supplementary Data Study 4

Supplementary Data for Environmental contamination of soils from lead joints used in large water supply pipelines, Harvey, P and Taylor, M.P., *Journal of Hazardous Materials*.

Supplementary Data Table S1. Soil metal concentrations along the Chichester Trunk Gravity Main pipeline at Woodberry, NSW, Australia.

Sample description (soil)	Arsenic mg/kg	Chromium mg/kg	Copper mg/kg	Lead mg/kg	Zinc mg/kg
648_0_0-2	22	16	38	20600	84
648_1_0-2	12	11	20	6030	110
648_2_0-2	12	13	15	2250	89
648_3_0-2	9.5	12	13	580	59
648_4_0-2	4.5	9.5	7.4	170	55
648_5_0-2	5.6	10	6.3	99	50
648_6_0-2	5.4	9.2	7.9	96	60
648_7_0-2	3.3	8.2	7.7	49	46
648_7_0-2 DUP	3	8.2	6.5	56	41
648_8_0-2	4.9	9.8	7.9	68	51
648_9_0-2	4.1	8.9	7.2	55	42
648_9_2-10	3.6	8.9	6.7	39	22
648_9_10-20	3.5	8.5	4.3	22	20
648_9_40-50	1.8	6.9	0.68	4.3	4.1
648_9_40-50 DUP	2.4	7.9	0.92	4.6	4
648_11_0-2	3.9	7.9	8.6	32	49
648_13_0-2	12	7.1	19	35	84
648_15_0-2	4.2	9	11	44	44
648_32_0_0-2	9.3	21	12	25	140

663_0_0-2	19	10	19	420	37
663_1_0-2	15	12	19	390	120
663_2_0-2	12	16	14	170	65
663_3_0-2	10	20	11	91	60
663_4_0-2	5.1	15	16	39	180
663_5_0-2	4.1	11	31	48	380
663_6_0-2	6	13	48	94	730
663_7_0-2	5.9	14	48	91	700
663_8_0-2	4.4	12	47	70	610
663_9_0-2	6.2	12	43	73	820
663_9_2-10	7.9	15	63	92	960
663_9_2-10 DUP	7.9	15	52	97	890
663_9_10-20	9	16	76	130	1120
663_9_40-50	3.4	20	12	91	330
663_9_40-50 DUP	4.6	18	12	68	310
663_11_0-2	4.1	12	30	57	660
663_13_0-2	5.2	19	10	26	150
663_15_0-2	11	25	11	33	82
663_53_0-2	7.3	37	30	27	120
663.5_0_0-2	12	19	10	270	28
664_0_0-2	20	11	23	9430	62
662.5_0_0-2	17	21	22	210	38
662_0_0-2	19	25	28	7830	100
Control SE Corner 0-2	2.5	38	26	22	100
Control SE Corner 0-2 DUP	3.1	42	29	27	110
Control SW Corner 0-2	4.9	35	34	30	230

Note: sample codes show the following: joint number_distance from joint_ depth

Supplementary Data Table S3. Soil, paint and joint weld lead and other element concentration values found along a range of large water supply pipelines.

Sample description (grass)	Arsenic mg/kg	Chromium mg/kg	Copper mg/kg	Lead mg/kg	Zinc mg/kg
648_2	0.08	0.13	5.6	50	72
648_5	0.06	0.07	3.6	1.5	49
648_9	0.09	0.16	4.6	1.2	80
648_15	0.07	0.12	4.4	0.55	63
663_2	0.06	0.13	7.9	10	74
663_5	0.20	0.71	4.5	3.1	150
663_9	0.36	0.63	5.1	3.1	180
663_15	0.38	0.93	4.9	1.8	66

Supplementary Data Table S3. Soil, paint and joint weld lead and other element concentration values found along a range of large water supply pipelines.

Forest Pipeline (North Sydney)		
Ethie Road, Allambie Heights, Sydney, NSW Australia		
GPS location - 33.75817 S, 151.142973 E		
Sample	Lead (mg/kg)	Notes
		Instrument: Olympus InnovX Delta 50 kV
Pipe surface paint	2760 mg/kg	Geochem mode
89/16_0_0-2	429 mg/kg	Soil mode; original joint
89/16_1_0-2	31 mg/kg	Soil mode
89/16.5_0_0-2	292 mg/kg	Soil mode
89/17_0_0-2	684 mg/kg	Soil mode; original joint
89/17_1_0-2	38 mg/kg	Soil mode
89/17.5_0_0-2	384 mg/kg	Soil mode
89/17.5_0_0-2 DUP	677 mg/kg	Soil mode
89/18_0_0-2	1020 mg/kg	Soil mode; new joint
89/18_1_0-2	39 mg/kg	Soil mode
Allambie Road, Allambie Heights, Sydney, NSW Australia		
GPS location - 33.76009 S, 151.24490 E		
Sample	Lead (mg/kg)	Notes
		Instrument: Olympus InnovX Delta 50 kV
82/2_0_0-2	2050 mg/kg	Soil mode; new joint
82/2_1_0-2	149 mg/kg	Soil mode
82/2.5_0_0-2	110 mg/kg	Soil mode
82/3_0_0-2	838 mg/kg	Soil mode; original joint
82/3_1_0-2	134 mg/kg	Soil mode

82/3.5_0_0-2	219 mg/kg	Soil mode
Woronora Pipeline (South Sydney)		
Tafft Place, Jannalli, Sydney, NSW, Australia		
GPS location - 34.01440 S, 151.06076 E		
Sample	Lead (mg/kg / %)	Notes
		Instrument: Olympus InnovX Delta 50 kV
Pipe surface	1.75 %	Geochem mode
J1 (actual joint)	21.05 %	Geochem mode
J1_0_0-2	831 mg/kg	Soil mode; original joint
J1_1_0-2	38 mg/kg	Soil mode
J1.5_0_0-2	56 mg/kg	Soil mode
J2_0_0-2	1040 mg/kg	Soil mode; original joint
J2_1_0-2	43 mg/kg	Soil mode
J2.5_0_0-2	49 mg/kg	Soil mode
Woronora Road Reserve, Engadine, Sydney, NSW, Australia		
GPS location - 34.04778 S, 151.00243 E		
Sample	Lead (mg/kg)	Notes
		Instrument: Olympus InnovX Delta 50 kV
Pipe surface	4250 mg/kg	Geochem mode
J1975_0_0-2	1760 mg/kg	Soil mode; new joint
J1975_1_0-2	30 mg/kg	Soil mode
J1975.5_0_0-2	132 mg/kg	Soil mode

J1976_0_0-2	3540 mg/kg	Soil mode; new joint
J1976_1_0-2	25 mg/kg	Soil mode
Goldfields Pipeline (Western Australia)		
Mundaring Weir, Mundaring, WA, Australia		
GPS 31.951245 S, 116.16858 E		
Sample	Lead (mg/kg)	Notes
		Instrument: Olympus InnovX Delta 40 kV
J3 (actual joint)	2330 mg/kg	Geochem mode; Hg 7502, As 565, Cr 443, Cu 741, Co 9721
Pipe surface	48 mg/kg	Geochem mode; As 348, Cu 1322, Cr 111
J1_1_0-2	47 mg/kg	Soil mode
J1.5_0_0-2	630 mg/kg	Soil mode
J2_0_0-2	1350 mg/kg	Soil mode
J2_1_0-2	29 mg/kg	Soil mode
J2.5_0_0-2	87 mg/kg	Soil mode
J3_0_0-2	1760 mg/kg	Soil mode
J3_1_0-2	52 mg/kg	Soil mode

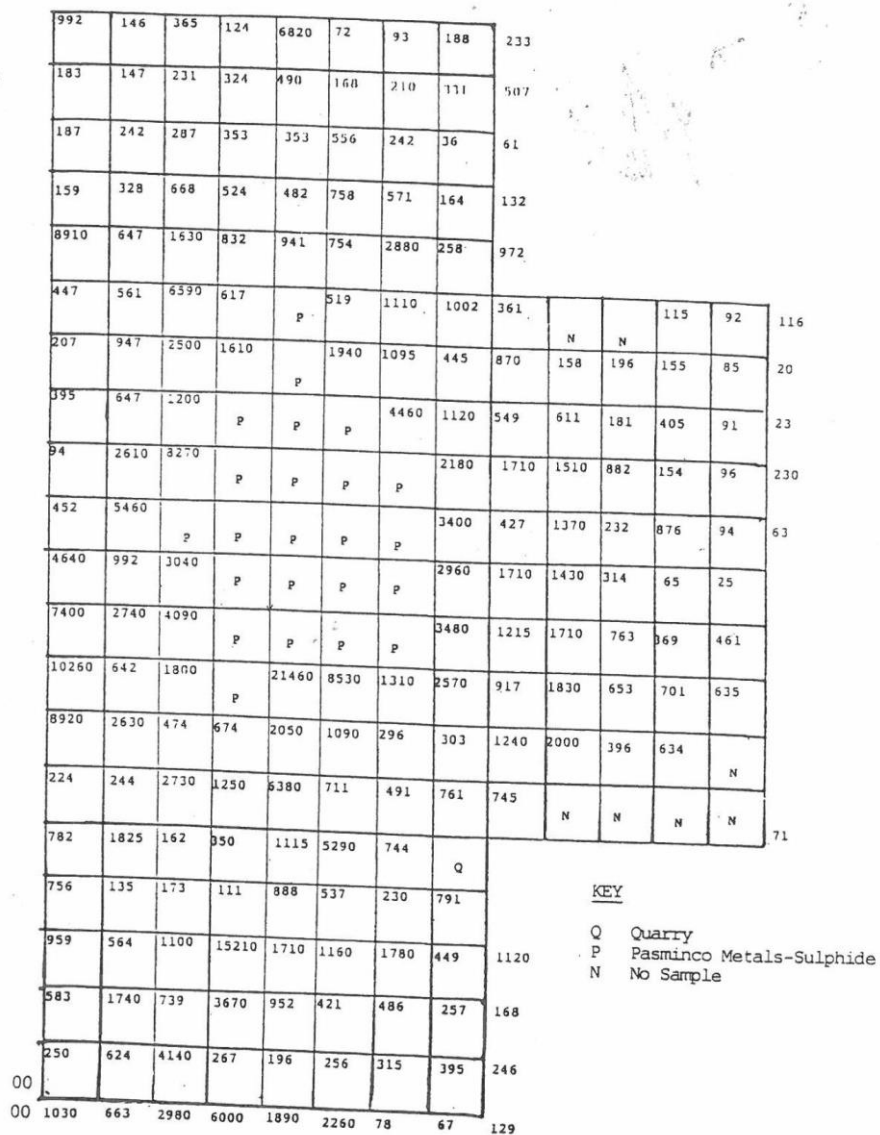
Notes: (1) sample codes show the following: joint number_distance from joint_ depth. (2) The Olympus InnovX Delta series instruments come with NIST 2710a and 2711a standards that were used for measuring machine accuracy during field measurement of soils, paint and joint welds as reported above. The Olympus InnovX Delta returned values within < 1 to 4 % of the reported values of the aforementioned NIST standards.

7.5 Supplementary Data Study 6

Supplementary Figure S1: Soil lead Sample Point Values (a) and Grid Mean Values (b) concentrations collected during the 1992 soil survey. Source Dames and Moore (1994a). Grid cells are 200 x 200 m.

a.

Fig. 16 Soil Lead - Sample Point Values



Note:

1. Sample taken at top left corner of each grid.
2. Results expressed in ppm (parts per million).

b. Grid Mean Values

Fig. 23 Soil Lead - Grid Mean Values

367	222	261	1940	1888	136	331	440										
190	227	299	380	392	294	205	234										
229	381	458	428	537	532	253	98										
2511	818	914	695	734	1241	968	382										
2641	2357	2417	797	738	1316	1513	648										
541	2650	2829	1114	1230	1166	913	670	463	177	155	112	78					
549	1324	1770			2498	1780	746	547	288	236	184	55					
937	3182	4735	P	P		2587	1390	1095	796	406	187	110					
2154	5447		P	P	P	2790	1929	1254	999	536	305	121					
2884	3161		P	P	P	3180	2124	1234	837	372	265	61					
3941	2713	3565	P	P	P	3220	2341	1516	1054	378	230	243					
5261	2318	2945			14995	4920	2453	2046	1418	1239	622	542	548				
5613	1387	983	8061	8283	2807	1120	1258	1497	1220	596	657						
1005	1520	1282	2589	2558	647	463	762	1328	1198	515							
769	1240	1123	2274	3374	1809	665	753										
875	575	200	616	1958	1700	588											
604	494	4150	4480	1074	927	813	787										
962	1036	5180	5386	1068	962	743	499										
799	1811	2204	1271	456	370	363	267										
642	2102	3347	2088	1151	727	214	209										

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P Pasminco

Note:

1. Numbers represent mean values of soil lead samples taken at the 4 intersection points of each grid area.
2. Results expressed in ppm (parts per million)

Supplementary Table S2: Residential soil metal and metalloid concentrations.

GPS Co-ordinates		Site	Antimony mg/kg	Arsenic mg/kg	Cadmium mg/kg	Lead mg/kg	Zinc mg/kg
151.6252042	-32.95001777	S1SF	10	52	38	3,810	2,510
151.6252042	-32.95001777	S1SB	6.6	43	43	2,590	3,130
151.6268233	-32.95098369	S2SF	<0.5	4.9	1.1	45	190
151.6268233	-32.95098369	S2SB	4.5	43	25	2,010	3,160
151.624632	-32.951278	S3SF	5.9	28	29	1,960	2,480
151.624632	-32.951278	S3SB	6.6	55	42	2,510	3,920
151.6208552	-32.95099963	S4SF	19	23	7.3	2,630	2,750
151.6208552	-32.95099963	S4SB	2.7	22	7.2	700	1,190
151.6205994	-32.95100324	S5SF	3	13	7.9	690	960
151.6205994	-32.95100324	S5SB	2	14	6.6	540	970
151.6267028	-32.95251557	S6SF	2.5	16	11	950	1,410
151.6267028	-32.95251557	S6SB	2.1	17	13	880	1,280
151.6265774	-32.95249488	S6aSF	3.4	15	17	1,060	2,040
151.6265774	-32.95249488	S6aSB	3.4	16	19	1,340	1,740
151.6255153	-32.95263217	S7SF	2	8.8	11	950	770
151.6255153	-32.95263217	S7SB	1	9.1	4.2	320	610
151.6255326	-32.95275625	S8SF	2.5	14	16	1,440	1,660
151.6255326	-32.95275625	S8SB	2.2	16	15	1,150	1,860
151.6246207	-32.95271317	S9SF	4	12	16	1,290	1,140
151.6246207	-32.95271317	S9SB	10	42	26	3,410	2,040
151.6225704	-32.95186125	S10SF	2.4	12	7.5	630	1,220
151.6225704	-32.95186125	S10SB	9	16	13	1,610	1,590
151.6205507	-32.95188033	S11SF	1.5	18	5.9	470	610
151.6205507	-32.95188033	S11SB	3.2	23	12	840	1,330
151.6212238	-32.95181716	S12SF	1.6	18	6.2	540	730
151.6212238	-32.95181716	S12SB	2.4	16	8.8	690	900
151.619887	-32.95279439	S13SF	1.2	11	4.6	360	550
151.619887	-32.95279439	S13SB	2.3	13	7.2	580	890
151.6240625	-32.95293802	S14SF	7.1	12	21	2,120	1,120
151.6240625	-32.95293802	S14SB	2.2	15	12	770	1,640
151.62197	-32.954022	S15SF	1.6	14	4.3	360	680
151.62197	-32.954022	S15SB	4	21	11	940	1,860
151.6343456	-32.93635946	S16SF	1.5	17	6.1	420	900
151.6129705	-32.97585992	S17SF	<0.5	3.7	1.1	120	260
151.6129705	-32.97585992	S17SB	0.53	5	1.5	100	350
151.6275967	-32.95079784	S18SF	11	49	55	4,230	5,840
151.6275967	-32.95079784	S18SB	0.78	8.4	6.4	310	940

S = residential site code. SF = front yard soil, SB = back yard soil.

Supplementary Table S3: Residential vacuum dust metal and metalloid concentrations.

GPS Co-ordinates	Site	Antimony mg/kg	Arsenic mg/kg	Cadmium mg/kg	Lead mg/kg	Zinc mg/kg	Bioavailable Lead mg/kg
151.6252042 -32.95001777	S1	4.7	6	13	380	2,900	580
151.6268233 -32.95098369	S2	4.6	13	16	1,070	1,940	850
151.624632 -32.951278	S3	9	270	16	950	2,030	890
151.6208552 -32.95099963	S4	12	11	3.1	150	780	140
151.6267028 -32.95251557	S6	4.1	4.7	2	120	700	86
151.6265774 -32.95249488	S6a	7.5	6.8	9.8	380	1,240	390
151.6255153 -32.95263217	S7	9.4	21	10	800	2,070	720
151.6255326 -32.95275625	S8	6.6	9.2	6.7	480	2,830	350
151.6246207 -32.95271317	S9	2.6	9.3	7	410	940	360
151.6225704 -32.95186125	S10	3.9	9.5	6.1	370	840	300
151.6212238 -32.95181716	S12	33	2.2	1.6	130	660	170
151.619887 -32.95279439	S13	28	2.9	3.8	120	580	130
151.6240625 -32.95293802	S14	14	6	5.4	1,320	1,260	1,010
151.62197 -32.954022	S15	27	15	8.6	750	1,870	680
151.6343456 -32.93635946	S16	13	7.6	4.5	290	1,430	220
151.6129705 -32.97585992	S17	2.2	6.3	1.9	190	730	120
151.6275967 -32.95079784	S18	2.6	32	6.1	500	1,370	460

Supplementary Table S4: Comparison of soil lead concentration in <180 µm fraction to <2 mm fraction.

GPS Co-ordinates		Site	Antimony mg/kg	Arsenic mg/kg	Cadmium mg/kg	Lead mg/kg	Zinc mg/kg
151.6255153	-32.95263217	S7SF	2	8.8	11	950	770
151.6255153	-32.95263217	S7SF2mm	1.9	6.9	8.3	710	550
151.6268233	-32.95098369	S2SB	4.5	43	25	2,010	3,160
151.6268233	-32.95098369	S2SB2mm	3.4	24	14	1,240	2,000
151.619887	-32.95279439	S13SB	2.3	13	7.2	580	890
151.619887	-32.95279439	S13SB2mm	1.3	8	4.5	360	550
151.6208552	-32.95099963	S4SB	2.7	22	7.2	700	1,190
151.6208552	-32.95099963	S4SB2mm	4.5	31	7.9	850	1,370
151.6275967	-32.95079784	S23SF	11	49	55	4,230	5,840
151.6275967	-32.95079784	S23SF2mm	5.9	29	24	2,250	10,200
151.6343456	-32.93635946	S16SF	1.5	17	6.1	420	900
151.6343456	-32.93635946	S16SF2mm	1.6	20	6.5	440	1,090

S = residential site code. SF = front yard soil, SB = back yard soil.

Supplementary Table S5: Non-residential soil metal and metalloid concentrations.

GPS Co-ordinates		Site	Antimony mg/kg	Arsenic mg/kg	Cadmium mg/kg	Lead mg/kg	Zinc mg/kg
151.6229805	-32.9531	PS1	3	14	15	780	960
151.623357	-32.9421	PS2	3.6	13	26	1,520	2,590
151.623357	-32.9421	PS3	4.3	15	19	2,040	1,850
151.623357	-32.9421	PS4	2.1	12	7	700	820
151.631356	-32.9362	PS5	2.6	31	14	970	2,150
151.6227899	-32.96	PS6	2.1	12	11	750	1,220
151.628671	-32.9604	PS7	1	19	7.6	260	460
151.6270332	-32.9677	PS8	2.1	33	7.1	610	1,260
151.634222	-32.9549	PS9	4.9	13	35	1,930	1,970
151.634222	-32.9549	PS10	5	22	37	1,990	2,590
151.634222	-32.9549	PS11	3.8	30	31	1,600	1,930
151.633708	-32.9537	PS12	2.6	38	11	620	780
151.633708	-32.9537	PS13	2.3	30	12	660	870
151.633708	-32.9537	PS14	2.7	39	10	590	770
151.6236194	-32.9616	PS15	<0.5	5	1.6	99	250
151.6236194	-32.9616	PS16	0.71	5.4	2.8	200	290
151.6236194	-32.9616	PS17	2.3	17	9.2	920	1,360
151.6408486	-32.9356	PS18	1.2	8.3	2.8	240	530
151.6401816	-32.9307	PS19	<0.5	6.4	1.5	110	260
151.607615	-32.962	PS20	0.57	7.5	0.69	170	580
151.6411614	-32.9493	PS21	<0.5	8.4	3.3	180	270
151.6202341	-32.962	PS22	2.2	17	5.5	580	1,650
151.6297227	-32.9309	PS23	1.3	11	3.5	380	950
151.6192389	-32.9579	SF1	6.5	33	7.7	1,970	5,990
151.6192389	-32.9579	SF2	41	200	21	17,500	29,900
151.6159087	-32.9602	SF3	1.9	12	4.4	470	1,410
151.6189572	-32.9555	SF4	1.5	24	2.5	580	2,420
151.6163805	-32.9624	PG1	0.59	4.5	1.7	130	490
151.643765	-32.9468	PG2	<0.5	7.3	1.9	100	220
151.6226488	-32.9512	PG3	3.7	18	15	1,080	1,550
151.6226488	-32.9512	PG4	6.6	19	20	1,830	1,330
151.6199415	-32.9541	PG5	4.4	130	9.9	920	6,090

PS = public space, SF = Sporting field, PG = playground.

7.8 Supplementary Data Study 8

Supplementary Data 1. Soil metal(loid) concentrations (pXRF derived) for public (Zone **A SB**; **A** – sampling zone number, **B** – sample site) and private (PR**SXY**; **X** – property code, **Y** – sample number on property) samples. All concentrations mg/kg except where * denotes wt%.

Sample	x co-ordinate	y co-ordinate	Ti	Cr	Mn	Fe*	Zn	As	Cd	Pb
Zone 1 S1	151.754117	-32.921500	4,770	130	1,120	3.51	520	41	<LOD	300
Zone 1 S2	151.757272	-32.917458	10,100	110	1,200	3.88	990	41	<LOD	590
Zone 1 S3	151.755289	-32.918547	3,750	77	820	2.58	800	18	<LOD	280
Zone 1 S4	151.752281	-32.918856	1,930	54	560	2.07	600	17	<LOD	250
Zone 1 S5	151.749133	-32.918808	4,710	120	1,470	3.53	2,010	56	<LOD	1,820
Zone 1 S6	151.745742	-32.914514	4,650	97	3,190	4.15	2,230	60	<LOD	1,310
Zone 1 S7	151.743853	-32.912803	5,130	190	2,110	5.97	1,990	69	<LOD	1,550
Zone 1 S8	151.746250	-32.911394	6,300	190	1,270	5.17	510	37	<LOD	340
Zone 1 S9	151.750283	-32.914881	4,750	170	2,090	5.30	2,660	180	<LOD	4,650
Zone 1 S10	151.753450	-32.916361	4,850	170	1,830	4.27	1,410	25	<LOD	500
Zone 1 S11	151.754900	-32.914300	3,670	59	500	2.20	270	9	<LOD	62
Zone 1 S12	151.753094	-32.910250	4,650	160	3,880	5.69	1,690	83	3	1,650
Zone 1 S13	151.759181	-32.909817	3,660	94	720	2.25	520	19	<LOD	160
Zone 1 S14	151.759392	-32.906031	7,030	260	3,580	10.30	9,470	87	10	1,120
Zone 1 S15	151.756672	-32.904525	5,480	180	1,450	5.22	2,010	55	6	730
Zone 1 S16	151.754450	-32.904392	2,800	90	1,600	3.18	630	17	<LOD	230
Zone 1 S17	151.755342	-32.907642	3,770	57	610	2.51	140	9	<LOD	35
Zone 1 S18	151.750922	-32.907825	3,940	110	1,120	3.39	2,000	130	<LOD	740
Zone 1 S19	151.750175	-32.904172	2,700	75	1,670	2.43	470	15	<LOD	150
Zone 1 S20	151.753067	-32.898278	5,500	210	4,490	14.17	5,090	130	16	2,060
Zone 1 S21	151.748775	-32.895000	3,810	230	3,410	9.71	1,910	35	4	590
Zone 1 S22	151.747103	-32.899675	5,160	200	2,450	9.51	1,230	50	<LOD	790
Zone 1 S23	151.745039	-32.903125	5,640	160	1,200	3.99	400	14	<LOD	240
Zone 1 S24	151.742175	-32.900814	3,900	140	1,040	3.30	1,070	25	<LOD	450
Zone 1 S25	151.744000	-32.896467	4,120	100	2,210	4.85	1,470	34	<LOD	640

Zone 1 S26	151.738069	-32.890289	3,520	92	1,040	3.39	680	12	<LOD	210
Zone 1 S27	151.734183	-32.891683	4,050	140	500	3.22	340	11	<LOD	110
Zone 1 S28	151.731650	-32.893592	6,150	220	1,240	4.08	610	23	<LOD	590
Zone 1 S29	151.727119	-32.889044	4,450	210	1,870	4.27	900	22	<LOD	450
Zone 2 S1	151.763972	-32.907889	6,290	160	1,810	4.80	690	24	<LOD	290
Zone 2 S2	151.765667	-32.908611	4,920	81	1,980	3.94	830	29	<LOD	410
Zone 2 S3	151.766972	-32.907528	4,950	120	1,760	3.57	1,500	14	<LOD	270
Zone 2 S4	151.768639	-32.907889	13,700	710	3,630	17.08	12,100	200	7	2,550
Zone 2 S5	151.762028	-32.910972	5,130	92	546	2.82	140	6	<LOD	33
Zone 2 S6	151.764389	-32.910306	5,110	140	2,600	4.86	1,870	45	4	1,240
Zone 2 S7	151.766500	-32.910806	5,660	140	2,580	4.92	1,860	51	<LOD	1,240
Zone 2 S8	151.768417	-32.909250	5,670	350	3,110	7.18	2,570	61	6	930
Zone 2 S9	151.769806	-32.910778	6,560	330	2,370	4.15	1,130	23	<LOD	280
Zone 2 S10	151.762306	-32.912917	4,300	60	650	2.65	200	14	<LOD	57
Zone 2 S11	151.765167	-32.912889	5,200	130	2,100	4.39	3,050	69	<LOD	1,300
Zone 2 S12	151.767861	-32.912028	3,670	83	1,500	3.09	790	44	<LOD	400
Zone 2 S13	151.765417	-32.914111	3,690	110	1,540	3.00	1,010	32	<LOD	500
Zone 2 S14	151.768333	-32.914083	4,810	180	1,240	5.08	1,950	76	5	930
Zone 2 S15	151.761278	-32.915472	14,800	690	3,250	7.00	4,690	73	<LOD	900
Zone 2 S16	151.764556	-32.916500	4,950	120	1,230	3.54	1,040	36	<LOD	630
Zone 2 S17	151.765389	-32.919556	6,280	190	1,540	4.34	4,260	380	5	2,770
Zone 2 S18	151.766722	-32.918250	4,990	120	1,850	4.11	2,290	42	11	1,220
Zone 2 S19	151.766972	-32.917083	6,040	200	1,530	4.48	4,940	96	13	1,970
Zone 3 S1	151.760889	-32.923492	3,450	59	660	2.21	360	13	<LOD	190
Zone 3 S2	151.762344	-32.924817	4,310	69	1,260	3.69	640	74	<LOD	370
Zone 3 S3	151.765656	-32.925775	3,770	120	770	2.71	690	23	<LOD	220
Zone 3 S4	151.769256	-32.925933	5,110	88	840	3.58	390	44	<LOD	230
Zone 3 S5	151.771983	-32.926467	4,890	91	541	2.79	320	10	<LOD	230
Zone 3 S6	151.775775	-32.925344	3,030	38	790	1.70	240	10	<LOD	39
Zone 3 S7	151.780153	-32.925839	4,500	78	820	2.28	140	6	<LOD	45
Zone 3 S8	151.785872	-32.925525	4,490	88	300	2.03	96	6	<LOD	22
Zone 3 S9	151.789786	-32.924081	4,950	81	730	2.90	380	14	<LOD	150

Zone 3 S10	151.791397	-32.923228	5,740	92	580	2.80	180	9	<LOD	68
Zone 4 S1	151.787817	-32.926988	5,460	140	660	2.78	170	8	<LOD	27
Zone 4 S2	151.785853	-32.927366	4,160	72	360	2.22	250	19	<LOD	110
Zone 4 S3	151.783673	-32.928302	1,750	55	790	1.82	800	21	<LOD	270
Zone 4 S4	151.779448	-32.929244	4,810	68	370	2.03	290	23	<LOD	560
Zone 4 S5	151.782722	-32.932442	5,910	130	680	5.14	220	56	<LOD	50
Zone 4 S6	151.778754	-32.931607	5,240	87	100	4.09	1,220	63	<LOD	3,450
Zone 4 S7	151.775584	-32.930542	3,870	94	660	2.50	170	5	<LOD	23
Zone 4 S8	151.776879	-32.929139	4,280	76	560	2.55	230	6	<LOD	75
Zone 4 S9	151.773684	-32.928170	4,400	74	430	2.12	280	7	<LOD	120
Zone 4 S10	151.770599	-32.929525	3,550	71	1,010	2.17	650	39	<LOD	720
Zone 4 S11	151.770951	-32.932130	8,890	140	280	1.67	98	25	<LOD	41
Zone 4 S12	151.767797	-32.930565	1,920	41	730	1.36	630	13	<LOD	360
Zone 4 S13	151.765789	-32.928902	6,590	160	920	3.27	510	13	<LOD	200
Zone 4 S14	151.766179	-32.927340	4,570	80	950	2.26	230	6	<LOD	150
Zone 4 S15	151.765789	-32.928902	5,390	100	830	3.29	300	23	<LOD	330
Zone 4 S16	151.762763	-32.926629	4,470	91	780	3.62	1,880	22	<LOD	620
Zone 4 S17	151.764658	-32.927587	5,760	110	860	3.58	510	17	<LOD	470
Zone 4 S18	151.789719	-32.926185	6,470	84	560	2.76	230	14	<LOD	69
Zone 4 S19	151.789625	-32.927671	5,880	130	540	2.96	130	9	<LOD	20
Zone 4 S20	151.785676	-32.925958	5,000	98	230	2.45	130	10	<LOD	120
Zone 4 S21	151.783617	-32.930237	5,970	110	540	2.83	460	21	<LOD	450
Zone 4 S22	151.777405	-32.930160	3,810	63	550	2.31	220	11	<LOD	83
Zone 4 S23	151.789632	-32.926502	6,340	93	740	3.41	150	7	<LOD	14
Zone 4 S24	151.784438	-32.927336	5,270	110	920	3.63	1,370	38	3	1,250
Zone 4 S25	151.770352	-32.928371	1,670	52	350	1.59	180	11	<LOD	53
Zone 5 S1	151.755858	-32.926782	3,870	63	780	2.64	420	12	<LOD	210
Zone 5 S2	151.755046	-32.927209	3,970	78	970	2.56	400	11	<LOD	120
Zone 5 S3	151.758133	-32.928261	3,500	96	1,100	2.34	540	18	<LOD	410
Zone 5 S4	151.754604	-32.930872	4,660	77	1,110	2.84	430	17	<LOD	220
Zone 5 S5	151.750771	-32.930600	2,320	77	580	1.84	280	8	<LOD	120
Zone 5 S6	151.753740	-32.933156	4,620	73	1,220	2.81	470	14	<LOD	240

Zone 5 S7	151.750693	-32.934451	4,970	80	700	2.59	280	36	<LOD	220
Zone 5 S8	151.750418	-32.937009	3,890	77	600	2.83	730	22	<LOD	560
Zone 5 S9	151.753171	-32.936175	4,590	68	600	2.34	330	83	<LOD	260
Zone 5 S10	151.755987	-32.938211	3,770	65	870	2.55	240	11	<LOD	180
Zone 5 S11	151.756269	-32.935044	3,440	74	980	2.07	680	13	<LOD	250
Zone 5 S12	151.758250	-32.936281	4,250	86	600	2.53	600	33	<LOD	310
Zone 5 S13	151.760791	-32.937766	4,240	83	180	2.21	270	18	<LOD	440
Zone 5 S14	151.763637	-32.938862	4,790	74	750	2.42	550	42	<LOD	460
Zone 5 S15	151.763484	-32.935614	4,270	79	1,400	3.13	860	34	<LOD	590
Zone 5 S16	151.767670	-32.934845	4,290	57	320	2.06	190	15	<LOD	150
Zone 5 S17	151.765772	-32.934974	2,550	55	540	1.96	230	10	<LOD	140
Zone 5 S18	151.763240	-32.933954	5,360	88	590	2.85	190	10	<LOD	46
Zone 5 S19	151.762349	-32.931880	5,110	75	560	2.58	130	14	<LOD	71
Zone 5 S20	151.758135	-32.932137	3,610	83	1,480	3.08	780	47	<LOD	400
PRS1A	151.755494	-32.931677	2,900	85	760	1.97	640	33	<LOD	780
PRS1B	151.755494	-32.931677	3,940	83	670	2.07	530	39	<LOD	1,460
PRS1C	151.755494	-32.931677	2,520	80	700	2.10	500	25	<LOD	330
PRS2A	151.765575	-32.935832	5,440	73	330	2.09	610	39	<LOD	900
PRS2B	151.765575	-32.935832	5,230	89	460	2.28	400	21	<LOD	510
PRS2C	151.765575	-32.935832	4,140	100	490	2.33	2,090	85	<LOD	2,210
PRS3A	151.753439	-32.910121	4,470	97	790	2.99	880	25	<LOD	840
PRS3B	151.753439	-32.910121	5,670	120	790	3.61	1,030	48	<LOD	1,550
PRS3C	151.753439	-32.910121	5,460	120	1,850	4.32	1,130	63	<LOD	1,740
PRS3D	151.753439	-32.910121	6,790	220	2,220	5.61	6,240	85	3	3,420
PRS4A	151.750841	-32.914811	5,510	110	1,710	4.17	1,630	55	<LOD	1,730
PRS4B	151.750841	-32.914811	6,260	180	1,540	5.56	3,190	280	6	8,280
PRS4C	151.750841	-32.914811	6,610	390	1,650	6.48	8,570	280	5	11,600
PRS5A	151.765773	-32.935121	6,440	96	760	3.02	780	32	<LOD	960
PRS5B	151.765773	-32.935121	5,850	130	790	2.99	1,010	40	<LOD	1,690
PRS5C	151.765773	-32.935121	6,250	140	960	3.64	4,040	56	<LOD	2,090
PRS6A	151.765488	-32.935929	5,920	84	460	2.36	1,030	55	<LOD	1,190
PRS6B	151.765488	-32.935929	4,670	69	400	2.13	550	40	<LOD	740

PRS7A	151.753251	-32.911086	6,330	96	1,350	3.36	1,200	85	<LOD	2,830
PRS7B	151.753251	-32.911086	6,710	200	1,790	5.41	3,210	240	6	5,450
PRS8A	151.735041	-32.891055	4,690	140	650	3.90	1,880	24	<LOD	1,440
PRS8B	151.735041	-32.891055	3,690	110	590	3.47	710	19	<LOD	430
PRS8C	151.735041	-32.891055	4,340	130	600	2.89	660	32	<LOD	1,060
PRS9A	151.733763	-32.891786	3,520	96	340	2.53	560	10	<LOD	110
PRS9B	151.733763	-32.891786	6,210	120	790	3.38	220	10	<LOD	42
PRS9C	151.733763	-32.891786	5,550	110	940	4.26	720	25	<LOD	180
PRS10A	151.747068	-32.899573	5,090	170	1,870	5.86	1,190	46	<LOD	1,170
PRS10B	151.747068	-32.899573	6,040	520	3,360	16.01	5,500	240	<LOD	7,640
PRS10C	151.747068	-32.899573	4,400	120	730	4.69	700	34	<LOD	530
PRS10D	151.747068	-32.899573	5,180	110	617	2.15	830	14	<LOD	510
PRS11A	151.747014	-32.899688	5,180	180	2,070	6.30	2,500	79	<LOD	2,340
PRS11B	151.747014	-32.899688	6,790	310	3,600	10.01	1,720	95	<LOD	2,520
PRS12A	151.765242	-32.917256	12,200	210	960	5.14	1,560	110	<LOD	1,370
PRS12B	151.765242	-32.917256	6,930	180	1,140	3.90	3,560	74	6	1,840
PRS13A	151.764114	-32.917046	2,920	140	740	2.35	1,380	34	<LOD	780
PRS13B	151.764114	-32.917046	4,260	170	970	3.56	3,210	62	9	1,240
PRS13C	151.764114	-32.917046	2,870	190	770	2.28	1,200	35	<LOD	610
PRS13D	151.764114	-32.917046	4,940	140	1,410	4.19	1,360	18	<LOD	400
PRS14A	151.764141	-32.917121	5,550	270	1,280	3.75	2,290	50	5	2,120
PRS14B	151.764141	-32.917121	5,720	180	1,200	3.05	1,420	35	<LOD	1,590
PRS14C	151.764141	-32.917121	5,690	300	960	3.14	2,060	64	<LOD	1,420
PRS14D	151.764141	-32.917121	6,360	410	1,790	5.96	2,800	150	<LOD	4,570
PRS15A	151.756360	-32.931232	4,040	87	700	2.34	1,160	20	<LOD	920
PRS15B	151.756360	-32.931232	2,720	98	910	1.94	793	16	<LOD	360
PRS15C	151.756360	-32.931232	4,690	81	670	2.69	1,320	22	<LOD	1,110
PRS16A	151.754543	-32.930401	4,950	2,400	480	5.59	140	18	<LOD	120
PRS16B	151.754543	-32.930401	3,190	580	500	2.62	420	16	<LOD	210
PRS16C	151.754543	-32.930401	2,880	65	590	2.09	200	21	<LOD	190
PRS17A	151.735603	-32.913977	4,810	98	660	2.66	1,230	42	<LOD	880
PRS17B	151.735603	-32.913977	5,170	160	930	3.98	3,850	160	4	1,450

PRS18A	151.737504	-32.891153	3,190	76	560	2.95	880	12	<LOD	430
PRS18B	151.737504	-32.891153	3,940	100	470	3.30	2,930	12	<LOD	1,230
PRS19A	151.758141	-32.911858	5,740	460	1,840	4.95	3,700	80	4	2,350
PRS19B	151.758141	-32.911858	6,200	160	910	3.22	1,970	30	<LOD	1,810
PRS20A	151.725239	-32.897365	6,350	170	1,100	4.24	2,600	28	<LOD	1,930
PRS20B	151.725239	-32.897365	5,230	160	580	3.40	1,880	24	<LOD	1,020
PRS20C	151.725239	-32.897365	5,310	130	580	3.30	1,320	14	<LOD	830
PRS21A	151.747123	-32.940503	4,740	75	1,000	2.47	1,290	15	<LOD	980
PRS21B	151.747123	-32.940503	3,450	72	920	2.39	970	17	<LOD	1,180
PRS22A	151.711666	-32.926700	3,500	74	410	1.75	1,070	17	<LOD	1,360
PRS22B	151.711666	-32.926700	2,910	54	120	1.31	380	8	<LOD	150
PRS22C	151.711666	-32.926700	3,970	68	480	2.13	380	15	<LOD	170
PRS23A	151.784001	-32.903409	7,080	180	740	5.60	770	47	<LOD	790
PRS23B	151.784001	-32.903409	9,120	210	1,390	8.05	1,090	55	4	1,140
PRS23C	151.784001	-32.903409	6,650	170	1,130	5.23	830	38	<LOD	610
PRS23D	151.784001	-32.903409	5,060	110	990	3.73	820	24	<LOD	450
PRS23E	151.784001	-32.903409	7,320	330	2,130	14.78	3,840	130	4	3,420
Soil Depth Profile a (0 - 2 cm)	151.780194	-32.933708	6170	67	360	2.27	330	20	<LOD	160
Soil Depth Profile a (2 - 10 cm)	151.780194	-32.933708	6370	63	320	2.39	260	28	<LOD	150
Soil Depth Profile a (10 - 20 cm)	151.780194	-32.933708	6400	77	270	2.36	220	30	<LOD	150
Soil Depth Profile a (20 - 30 cm)	151.780194	-32.933708	8400	71	200	2.21	170	21	<LOD	150
Soil Depth Profile a (30 - 40 cm)	151.780194	-32.933708	1.18*	98	99	1.54	110	30	<LOD	88
Soil Depth Profile b (0 - 2 cm)	151.760230	-32.908183	6730	140	1400	4.69	1350	67	<LOD	600
Soil Depth Profile b (2 - 10 cm)	151.760230	-32.908183	6650	170	1500	5.01	1380	62	<LOD	640
Soil Depth Profile b (10 - 20 cm)	151.760230	-32.908183	7280	160	1100	4.62	1330	69	<LOD	390
Soil Depth Profile b (20 - 30 cm)	151.760230	-32.908183	8590	89	670	3.81	410	40	<LOD	150
Soil Depth Profile b (30 - 40 cm)	151.760230	-32.908183	9400	83	280	3.55	150	31	<LOD	66
Soil Depth Profile c (0 - 2 cm)	151.746483	-32.911400	3270	87	700	2.29	330	33	<LOD	250
Soil Depth Profile c (2 - 10 cm)	151.746483	-32.911400	2370	78	470	1.97	210	26	<LOD	110
Soil Depth Profile c (10 - 20 cm)	151.746483	-32.911400	3320	82	600	2.29	340	23	<LOD	210
Soil Depth Profile c (20 - 30 cm)	151.746483	-32.911400	8020	160	810	3.46	320	30	<LOD	200
Soil Depth Profile c (30 - 40 cm)	151.746483	-32.911400	6470	98	1400	3.79	470	32	<LOD	400

Soil Depth Profile d (0 - 2 cm)	151.758508	-32.932803	2640	67	790	1.96	270	29	<LOD	120
Soil Depth Profile d (2 - 10 cm)	151.758508	-32.932803	4270	89	750	2.59	370	36	<LOD	150
Soil Depth Profile d (10 - 20 cm)	151.758508	-32.932803	5460	78	450	2.94	240	85	<LOD	100
Soil Depth Profile d (20 - 30 cm)	151.758508	-32.932803	5860	77	270	2.47	150	44	<LOD	91
Soil Depth Profile d (30 - 40 cm)	151.758508	-32.932803	6450	56	210	1.9	110	32	<LOD	59
Soil Depth Profile e (0 - 2 cm)	151.695386	-32.909801	3270	59	150	1.48	96	3	<LOD	41
Soil Depth Profile e (2 - 10 cm)	151.695386	-32.909801	3570	58	140	1.51	94	6	<LOD	39
Soil Depth Profile e (10 - 20 cm)	151.695386	-32.909801	3510	46	37	1.35	40	4	<LOD	17
Soil Depth Profile e (20 - 30 cm)	151.695386	-32.909801	4130	61	16	1.9	31	6	<LOD	14
Soil Depth Profile e (30 - 40 cm)	151.695386	-32.909801	5240	67	36	3.51	38	13	<LOD	15
Soil Depth Profile f (0 - 2 cm)	151.621760	-32.902235	2830	77	140	1.65	230	14	<LOD	150
Soil Depth Profile f (2 - 10 cm)	151.621760	-32.902235	3040	96	54	1.58	110	13	<LOD	88
Soil Depth Profile f (10 - 20 cm)	151.621760	-32.902235	3010	58	31	1.82	60	12	<LOD	35
Soil Depth Profile f (20 - 30 cm)	151.621760	-32.902235	3260	79	30	1.91	58	9	<LOD	29
Soil Depth Profile f (30 - 40 cm)	151.621760	-32.902235	3260	79	30	1.91	58	9	<LOD	29

Supplementary Data 2. Soil PAH concentrations for the Newcastle city. All concentrations in mg/kg.

PAH: 1 – Naphthalene, 2 – Acenaphthylene, 3 – Acenaphthene, 4 – Fluorene, 5- Phenanthrene, 6 – Anthracene, 7 – Fluoranthene, 8 – Pyrene, 9 - Benz(a)anthracene, 10 – Chrysene, 11 - Benzo(b)&(k)fluoranthene, 12 - Benzo(a)pyrene, 13 - Indeno(1,2,3-cd)pyrene, 14 - Dibenzo(a,h)anthracene, 15 - Benzo(g,h,i)perylene. Soil samples: public (Zone **A SB**; **A** – sampling zone number, **B** – sample site).

Sample marked with ** is a field duplicate.

Sample	x co- ordinate	y co- ordinate	PA H 1	PA H 2	PA H 3	PA H 4	PA H 5	PA H 6	PA H 7	PA H 8	PA H 9	PA H 10	PA H 11	PA H 12	PA H 13	PA H 14	PA H 15	Σ PA H
Zone 5 S1	151.765942	-32.935172	<0.5	<0.5	<0.5	<0.5	0.8	<0.5	2.2	2.1	0.8	0.7	1.4	1.0	0.6	<0.5	0.8	10
Zone 5 S4	151.751481	-32.936583	<0.5	<0.5	<0.5	<0.5	4.4	1.5	10	9.6	3.5	3.5	6.9	5.2	2.9	0.6	3.4	52
Zone 5 S5	151.755347	-32.931136	<0.5	<0.5	<0.5	<0.5	1.7	0.6	4.6	4.5	1.5	1.5	3.1	2.2	1.4	<0.5	1.6	23
Zone 2 S1	151.765858	-32.91974	<0.5	<0.5	<0.5	<0.5	2.4	0.7	6.9	6.4	2.4	2.1	4.5	3.3	2.0	<0.5	2.4	33
Zone 2 S2	151.765405	-32.914048	<0.5	<0.5	<0.5	<0.5	1.5	<0.5	5.7	5.4	2.0	2.0	4.2	2.7	1.9	<0.5	2.2	28
Zone 2 S2**	151.765405	-32.914048	<0.5	<0.5	<0.5	<0.5	2.1	0.7	6.3	6.0	2.3	2.2	4.5	3.2	2.0	<0.5	2.4	32
Zone 2 S3	151.764169	-32.909803	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	1.7	1.6	0.6	0.6	1.5	1.0	0.7	<0.5	0.9	8.6
Zone 2 S4	151.766314	-32.910977	<0.5	0.9	<0.5	<0.5	3.8	1.4	10	10	4.0	3.8	7.3	5.4	3.1	0.7	3.5	54
Zone 1 S5	151.746816	-32.900268	<0.5	<0.5	<0.5	<0.5	1.5	<0.5	5.4	5.3	2.0	2.0	4.4	2.9	2.0	<0.5	2.5	28
Zone 1 S2	151.750378	-32.914897	<0.5	<0.5	<0.5	<0.5	1.6	0.7	5.7	5.6	1.8	2.0	4.3	2.8	1.8	<0.5	2.0	28
Zone 1 S4	151.752958	-32.910561	<0.5	<0.5	<0.5	<0.5	1.9	0.6	5.5	5.3	1.7	1.8	3.7	2.7	1.6	<0.5	2.0	27
Zone 1 S7	151.734307	-32.891691	<0.5	0.8	<0.5	<0.5	8.2	2.4	20	18	7.3	7.5	14	9.8	6.1	1.4	7.6	103

Supplementary Data 3. Lead isotope compositions for soils, slag and galena. Standard deviations determined from repeat analysis (10 analyses) of one sample. Soil samples: public (Zone A SB; A – sampling zone number, B – sample site), private (PRSXY; X – property code, Y – sample number on property) and depth (SDPX X-Y cm; where X is the depth profile number and X-Y is the depth).

Sample	x co- ordinate	y co- ordinate	$^{208}\text{Pb}/^{207}\text{Pb}$	$^{206}\text{Pb}/^{207}\text{Pb}$	$^{206}\text{Pb}/^{204}\text{Pb}$
			+/- 0.004	+/- 0.004	+/- 0.091
PRS4C	151.747014	-32.899688	2.400	1.114	17.483
PRS11A	151.747014	-32.899688	2.365	1.092	16.863
PRS8C	151.735041	-32.891055	2.402	1.126	17.483
PRS20C	151.725239	-32.897365	2.352	1.079	16.474
Zone 1 S20	151.753067	-32.898278	2.411	1.131	17.483
Zone 1 S9	151.750283	-32.914881	2.374	1.091	16.892
Zone 2 S17	151.755342	-32.907642	2.345	1.068	16.584
Zone 2 S4	151.768639	-32.907889	2.361	1.085	16.807
Zone 2 S4*	151.768639	-32.907889	2.365	1.117	17.513
Zone 2 S17**	151.755342	-32.907642	2.355	1.071	16.474
Slag 1	***	***	2.319	1.044	16.077
Slag 2	***	***	2.543	1.318	21.097
SDP b 30-40 cm	151.759338	-32.906211	2.384	1.099	16.949
SDP c 30-40 cm	151.747180	-32.911957	2.370	1.093	16.835
SDP d 30-40 cm	151.758508	-32.932803	2.406	1.124	17.544
SDP e 20-30 cm	151.695323	-32.910189	2.489	1.198	18.727
Wallaroo Galena	***	***	2.640	1.477	24.727
Broken Hill Galena	***	***	2.319	1.044	16.155

*field duplicate

**laboratory duplicate

***GPS co-ordinates not applicable

Supplementary Data 4. Relative abundances of phases were estimated from XRD patterns using the relative intensity ratio (RIR) method.

Group	Phase	Chemical composition	PRS3B	PRS3D	PRS4C	PRS7A	Zone 1 S7	Zone 1 S9	Zone 2 S1	Zone 2 S4	Zone 2 S15
Silicates	Clinopyroxene	(Ca,Mg,Fe,Al) ₂ Si ₂ O ₆		*	***		***	**			*
	Olivine	(Ca,Fe,Mg) ₂ SiO ₄		*	*		*	***			
	Zircon	ZrSiO ₄	*	tr	*	*	*		*	*	*
	Vesuvianite	Ca ₁₀ (Mg,Fe) ₂ Al ₄ (SiO ₄) ₅ (Si ₂ O ₇) ₂				**					
	Melilite	Ca ₂ (Mg,Fe,Zn)Si ₂ O ₇				*			tr	*	
	Amorphous glass	Si-Ca-Fe		***	***	***	**	***		***	*
Oxides	Quartz	SiO ₂	tr						*		*
	Hematite	Fe ₂ O ₃	*	***	**	***	**	**	*	**	tr
	Spinel series	(Fe,Zn,Mg)(Fe,Al,Cr) ₂ O ₄	*	*	*	*	tr	*	*	**	*
	Goethite	FeOOH						tr		tr	
	Wuestite	FeO								tr	
	Rutile	TiO ₂	**	tr	tr		*	tr	*	tr	*
	Ilmenite	FeTiO ₃	tr						***		tr
	Corundum	Al ₂ O ₃	*								

*** abundant, ** common, * minor, tr trace

PRS3B – x co-ordinate: 151.754543; y co-ordinate: -32.930401

PRS3D – x co-ordinate: 151.747068; y co-ordinate: -32.899573

PRS4C – x co-ordinate: 151.750841; y co-ordinate: -32.914811

PRS7A – x co-ordinate: 151.753439; y co-ordinate: -32.910121

Zone 1 S7 – x co-ordinate: 151.743853; y co-ordinate: -32.912803

Zone 1 S9 – x co-ordinate: 151.750283; y co-ordinate: -32.914881
Zone 2 S1 – x co-ordinate: 151.761278; y co-ordinate: -32.915472
Zone 2 S4 – x co-ordinate: 151.768639; y co-ordinate: -32.907889
Zone 2 S15 – x co-ordinate: 151.761278; y co-ordinate: -32.915472

7.9 Additional Materials

2.51 REV. 3/81



E. I. DU PONT DE NEMOURS & COMPANY
INCORPORATED
WILMINGTON, DELAWARE 19898

POLYMER PRODUCTS DEPARTMENT

CC: M. S. DEAK
J. C. DAY

December 14, 1989

PERSONAL AND CONFIDENTIAL

TO: H. V. BRADLEY - WASHINGTON WORKS
H. D. RAMSEY - WASHINGTON WORKS
R. D. LANYON - WASHINGTON WORKS

FROM: H. A. SMITH

WASHINGTON WORKS
CANCER INCIDENCE AND OVERALL MORTALITY RATES

The Corporate Epidemiology Section has just completed an update and review of cancer incidence rates among employees and overall mortality rates among employees and pensioners, including all data through 1987. The basis for comparison is the experience of the entire Du Pont Company. Tables for your site are attached.

This information should be reviewed by your site medical personnel. There is always a chance there could be a question about the numbers in a specific grouping and sometimes our records show the final cause of death and not the primary cause of death. If there are any questions, please contact me so we can keep the records straight.

This information is part of our ongoing program to monitor employee health. It's another tool to help us do our job and, as such, can be used in any way you see fit. It is not the type of thing you would add to the annual communication list or, except in some very special circumstance, use for a special employee communication. This report issues every several years because this type of data has no real meaning for year-to-year trends. In fact, this information seldom stands alone - it's usually used in conjunction with other studies (e.g., animal) or to point out an area deserving of a closer look. Site statistics involve very small numbers, and trying to follow up and explain a small number of cases is exceedingly frustrating because chance plays the biggest role. When you break the data into very small groupings, sooner or later you are going to have a case versus a very small fractional expected number.

BETTER THINGS FOR BETTER LIVING

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SUMMARY OF DATACancer Incidence

No statistically significant excess was observed in the overall cancer experience of employees at this location from 1956-1987. There was, however, a statistically significant excess of cancers of the buccal cavity and pharynx among male wage (8 cases and 2.6 expected) and all male employees. This elevation was reported in the 1956-1983 surveillance and one case has occurred since that time. There was also a statistically significant excess of kidney and other urinary cancers among male salary (5 cases versus 1.8 expected) and all male employees (8 cases versus 3.8 expected). This excess was not present during 1956-1983. Three cases have been diagnosed during 1984-1987 and 1.1 would be expected overall. A bladder cancer excess among male wage employees was also statistically significant (8 cases versus 4.0 expected). No elevation was reported during the earlier surveillance. Three bladder cancer cases were reported during 1984-1987; two of these were initially diagnosed in 1976. An additional new finding was a statistically significant elevation in multiple myeloma among all men (4 observed versus 1.4 expected). Again, three cases were diagnosed during 1984-1987 and 0.5 would be expected. No statistically significant deficits were observed in the overall or site-specific cancer experience of employees at Washington Works during 1956-1987. Cancer experience among female employees was not unusual.

*W. H. 1st floor
C. 2nd floor*

Mortality

Statistically significant deficits were observed in the overall mortality experience of male wage and all male employees at this location from 1957-1987. There was also a statistically significant deficit of deaths from cerebrovascular disease among male wage employees (5 deaths versus 11.1 expected). Significant deficits were seen among all male employees for suicide (3 observed versus 10.1 expected) and "other" accidents (6 observed versus 12.1 expected). No unusual patterns were seen in the mortality tables for female employees.

HAS/is
Attachment
B:19 #7-10

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INTERPRETATION OF SURVEILLANCE DATA

Enclosed are tables showing the (1) numbers of cancer cases reported from 1956 through 1987 among active employees at departmental locations and (2) numbers of deaths reported from 1957 through 1987 among active employees and pensioners at departmental locations. For each specific cause the number of observed cases/deaths (OBS) is compared to the number expected (EXP) based on the experience of the entire Company by the ratio of the observed to the expected numbers of cases/deaths (OBS/EXP). Accompanying each table is a descriptive text which summarizes the main points.

Sources of Surveillance Data

Cancer cases that occur among active employees are recorded in the Company-wide Cancer Registry that was started in 1956. Through 1988, cases have been reported to the Registry primarily by diagnoses entered on Accident and Health Insurance (A&H) claims and by death certificates that accompany life insurance claims filed by beneficiaries of deceased employees. Beginning in 1977, these sources were supplemented by cancer registry report forms submitted by Company physicians. The Cancer Registry includes only cancer cases that occur among active employees and does not include employees whose cancer was first diagnosed after retirement or after

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employment termination resulting from resignation, discharge or other reasons.

Deaths that occur among active and pensioned employees are recorded in the Company-wide Mortality registry that was initiated in 1957. Deaths are identified through life insurance claims filed by beneficiaries of deceased employees. Deaths that occur among employees terminated without pension are not included since there is no uniform mechanism for identification of these deaths.

Methods of Analysis

Methods used for the cancer incidence and mortality tables are basically the same and are discussed concurrently.

To determine expected numbers of cases/deaths for the standardized analysis, cancer incidence and mortality rates for Du Pont employees (and pensioners for mortality), specific for 5-year age categories, sex and payroll class (i.e. wage or salary roll). These rates are computed for each of the major causes shown in the enclosed tables. Then, the Company-wide rates are multiplied by the cumulative midyear population of employees (and pensioners, where applicable) from each plant or office

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location, specific for age, sex and payroll class, over the entire study period. The sum of the products over all age groups is the expected number of cases or deaths.

Standardized analyses are generally preferred because they provide age-adjusted expected numbers and are based on actual plant populations. In some cases, however, they may not be appropriate or informative. In these isolated cases (identified in the attached text and tables), we have provided instead the results of the proportionate incidence or mortality analyses. In these analyses, the observed distribution of cases/deaths by cause at a given site is compared with the expected which is derived from proportions which occur within the entire Company. In these situations, we have no populations for rates and can look only at proportional distributions.

Proportions can be misleading, however, as it is possible to have an unusual distribution of cases/deaths without there being any excess rate for any specific cause. For example, if a plant has a lower death rate from heart disease than the rate in the Company as a whole, the proportion of deaths from other causes would be inflated (compared to that of the entire Company) in order that proportions for all causes add up to 100 percent.

Tests of Significance

To test whether the observed to expected ratios (OBS/EXP) given in the tables differ significantly from 1.00, we determine the probability that the difference between the observed and expected numbers occurred by chance alone. This probability value is obtained from the Poisson probability distribution. The difference is considered statistically significant if the probability value is less than 0.10 using the two-tailed test. In the two-tailed test, statistically significant deficits as well as excesses are denoted. Statistical significance is tested only if either the observed or expected number of cases or deaths is 4 or more.

Interpretation of Statistically Significant Results

The designation of a statistically significant excess often suggests the need for further investigation to determine whether the excess may have occurred because of some agent at the plant. However, an excess may also occur because of environmental and other factors associated with increased risks, such as smoking, diet, alcohol, ethnic origin, socioeconomic status or genetic factors.

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Chance alone may account for a statistically significant difference. When the level of statistical significance is set at 0.10, one should expect to find a statistically significant difference in about 10 out of every 100 comparisons due to chance alone, even when no specific causative factor is responsible.

The magnitude of the difference, expressed as the ratio of the observed to the expected numbers (OBS/EXP), must also be considered in interpretation of the data. The OBS/EXP ratio and corresponding probability value should be considered together in the assessment of the difference between an observed and expected number.

It may be that the observed number for a particular cause is greater than the expected number, but the difference is not statistically significant. In this instance it does not necessarily follow that a particular agent at the plant may not be associated with the moderate excess. If the number of persons at the plant exposed to the agent is small, excess morbidity or mortality in that group would be difficult to detect because of dilution by data from the rest of the plant. Also, it may be too soon for effects of an agent to be manifested by excess morbidity or mortality.

OBSERVED AND EXPECTED CANCER INCIDENCE USING POPULATION BASED RATES - 40C189
WASHINGTON (PARKERSBURG) - PLANT LOCATION CODE = 0131
1956 - 1987, MALES

TYPE OF CANCER	SALARY ROLL			HAGE ROLL			HAGE + SALARY		
	OBSERVED	EXPECTED	OBS/EXP	OBSERVED	EXPECTED	OBS/EXP	OBSERVED	EXPECTED	OBS/EXP
BUCAL CAVITY & PHARYNX	1	1.47	0.68	8	2.64	3.03 *	9	4.11	2.19 *
ESOPHAGUS	0	0.43	0.00	0	0.91	0.00	0	1.34	0.00
STOMACH	0	0.75	0.00	0	1.56	0.00	0	2.33	0.00
SMALL AND LARGE INTESTINE	8	4.97	1.61	6	6.72	0.89	14	11.69	1.20
RECTUM	2	1.77	1.13	3	2.97	1.01	5	4.74	1.05
LIVER AND BILIARY PASSAGES	0	0.43	0.00	2	0.82	2.43	2	1.25	1.59
PANCREAS	1	1.18	0.85	2	1.96	1.02	3	3.14	0.95
PERITONEUM	8	0.10	0.00	1	0.10	9.85	1	0.20	5.06
UNSPEC. DIGESTIVE ORGANS	0	0.03	0.00	0	0.01	0.00	0	0.03	0.00
NOSE, NASAL CAVITIES, ETC.	1	0.13	7.71	0	0.53	0.00	1	0.66	1.52
LARYNX	1	0.69	1.47	3	1.92	1.56	4	2.60	1.54
LUNG, BRONCHUS, & TRACHEA	7	7.06	0.99	10	13.36	0.75	-17	20.44	0.85
MEDIASTINUM & UNSPEC.	0	0.12	0.00	0	0.14	0.00	0	0.26	0.00
PLEURA	0	0.11	0.00	1	0.32	3.16	1	0.44	2.28
BREAST	0	0.11	0.00	0	0.10	0.00	0	0.21	0.00
PROSTATE	1	2.57	0.39	3	3.82	0.79	4	6.39	0.63
TESTIS	2	1.15	1.74	2	1.63	1.23	4	2.76	1.44
OTHER MALE GENITAL ORGANS	0	0.02	0.00	0	0.23	0.00	0	0.24	0.00
KIDNEY & OTHER URINARY	5	1.84	2.72 *	3	1.97	1.52	8	3.81	2.10 *
BLADDER	0	1.91	0.00	8	3.97	2.62 *	8	5.88	1.36
ALIGNANT MELANOMA	7	3.33	2.10	5	5.94	0.84	12	9.27	1.30
EYE	0	0.12	0.00	0	0.22	0.00	0	0.34	0.00
BRAIN & OTHER NERVOUS SYSTEM	0	1.37	0.00	3	2.93	1.02	3	4.31	0.70
THYROID	0	0.49	0.00	1	1.17	0.86	1	1.65	0.60
OTHER ENDOCRINE GLANDS	1	0.21	4.70	0	0.16	0.00	1	0.37	2.72
BONE	0	0.16	0.00	1	0.26	3.80	1	0.42	2.40
CONNECTIVE TISSUE	0	0.72	0.00	1	1.21	0.83	1	1.93	0.82
LYMPHOSARCOMA & RETICULOSARCOMA	0	1.10	0.00	0	1.23	0.00	0	2.33	0.00
HODGKIN'S DISEASE	1	0.89	1.13	2	2.22	0.90	3	3.10	0.97
OTHER LYMPHOMA	1	0.77	1.30	3	1.64	1.81	4	2.43	1.65
MULTIPLE MYELOMA	2	0.45	4.44	2	0.91	2.19	4	1.37	2.93 *
LEUKEMIA	2	1.39	1.44	4	2.69	1.49	6	4.08	1.47
MYCOSIS FUNGOIDES	0	0.12	0.00	0	0.00	-	0	0.12	0.00
OTHER HEMATOPOIETIC SYSTEM	1	0.18	5.81	0	0.22	0.00	1	0.40	2.52
OTHER & UNKNOWN	1	0.95	1.05	1	1.82	0.55	2	2.78	0.72
TOTAL ALL CAUSES	45	39.10	1.15	75	68.34	1.10	120	107.44	1.12

STATISTICALLY SIGNIFICANT EXCESS (*) / DEFICIT (8) AT THE 2-TAILED 0.10 LEVEL
(CALCULATED ONLY WHERE EITHER OBS OR EXP GE 4)

NOTE: OTHER SKIN CANCER NOT INCLUDED IN THIS REPORT

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OBSERVED AND EXPECTED CANCER INCIDENCE USING POPULATION BASED RATES - 30OCT89
WASHINGTON (PARKERSBURG) + PLANT (LOCATION CODE = 013)
1956 - 1987, FEMALES

TYPE OF CANCER	SALARY ROLL			WAGE ROLL			WAGE + SALARY		
	OBSERVED	EXPECTED	OBS/EXP	OBSERVED	EXPECTED	OBS/EXP	OBSERVED	EXPECTED	OBS/EXP
	-----	-----	-----	-----	-----	-----	-----	-----	-----
Buccal Cavity & Pharynx	0	0.04	0.00	0	0.07	0.00	0	0.11	0.00
Esophagus	0	0.80	0.00	0	0.00	-	0	0.00	0.00
Stomach	0	0.09	0.00	0	0.00	-	0	0.09	0.00
Small and Large Intestine	0	0.31	0.00	0	0.12	0.00	0	0.42	0.00
Rectum	0	0.15	0.00	0	0.07	0.00	0	0.22	0.00
Liver and Biliary Passages	0	0.00	-	0	0.05	0.00	0	0.05	0.00
Pancreas	0	0.85	0.00	0	0.01	0.00	0	0.86	0.00
Fertilizer	0	0.00	-	0	0.00	0.00	0	0.00	0.00
Unspec. Digestive Organs	0	0.00	-	0	0.00	-	0	0.00	-
Nose, Nasal Cavities, Etc.	0	0.01	0.00	0	0.00	-	0	0.01	0.00
Larynx	0	0.02	0.00	0	0.01	0.00	0	0.04	0.00
Lung, Bronchus, & Trachea	0	0.44	0.00	0	0.04	0.00	0	0.48	0.00
Mediastinum & Unspec.	0	0.00	-	0	0.00	-	0	0.00	-
Pleura	0	0.00	-	0	0.00	-	0	0.00	-
Breast	2	2.56	0.78	0	1.25	1.60	4	3.81	1.05
Cervix	0	1.45	0.00	3	1.21	2.48	3	2.66	1.13
Other Female Genital Organs	0	0.89	0.00	0	0.30	0.00	0	1.19	0.00
Kidney & Other Urinary	0	0.02	0.00	0	0.07	0.00	0	0.09	0.00
Bladder	0	0.05	0.00	0	0.02	0.00	0	0.07	0.00
Salivary Gland	2	0.42	4.78	0	0.19	0.00	2	0.61	3.27
Eye	0	0.01	0.00	0	0.00	-	0	0.01	0.00
Brain & Other Nervous System	0	0.10	0.00	0	0.00	0.00	0	0.11	0.00
Thyroid	1	0.31	3.20	0	0.02	0.00	1	0.33	3.00
Other Endocrine Glands	0	0.00	-	0	0.05	0.00	0	0.06	0.00
Bone	0	0.08	0.00	0	0.00	-	0	0.08	0.00
Connective Tissue	0	0.09	0.00	1	0.07	13.52	1	0.16	6.18
Lymphosarcoma & Reticulosarcoma	0	0.14	0.00	0	0.03	0.00	0	0.20	0.00
Hodgkin's Disease	0	0.17	0.00	0	0.03	0.00	0	0.18	0.00
Other Lymphoma	0	0.11	0.00	0	0.03	0.00	0	0.14	0.00
Multiple Myeloma	1	0.06	15.88	0	0.00	-	1	0.06	15.88
Leukemia	0	0.10	0.00	0	0.09	0.00	0	0.19	0.00
Myeloid Leukemia	0	0.00	-	0	0.00	-	0	0.00	-
Other Hematopoietic System	0	0.00	-	0	0.00	-	0	0.00	-
Other & Unknown	0	0.06	0.00	0	0.04	0.00	0	0.10	0.00
TOTAL ALL CAUSES	6	7.74	0.77	6	3.05	1.56	12	11.60	1.03

NOTE: OTHER SKIN CANCER NOT INCLUDED IN THIS REPORT

AJP0099994

EID521390

OBSERVED AND EXPECTED DEATHS USING POPULATION-BASED RATES - 50CT69
WASHINGTON (PARKERSBURG) - PLANT (LOCATION CODE = 013)
1957 - 1987, MALES

CAUSE OF DEATH	SALARY ROLL			WAGE ROLL			WAGE + SALARY		
	OBSERVED	EXPECTED	OBS/EXP	OBSERVED	EXPECTED	OBS/EXP	OBSERVED	EXPECTED	OBS/EXP
MALIGNANT NEOPLASMS									
BUCCAL CAVITY & PHARYNX	1	0.72	1.38	2	1.25	1.59	3	1.98	1.52
DIGESTIVE ORGANS	8	9.88	0.81	11	19.79	0.74	19	24.66	0.77
RESPIRATORY SYSTEM	12	10.97	1.09	17	21.64	0.79	29	32.41	0.89
BREAST	0	0.10	0.00	0	0.08	0.00	0	0.13	0.00
GENITAL ORGANS	1	2.29	0.44	2	3.30	0.61	3	5.69	0.54
URINARY ORGANS	0	2.03	0.00	4	2.85	1.40	4	4.87	0.82
LYMPHATIC, ETC.	4	4.20	0.95	6	6.67	0.90	10	10.83	0.92
OTHER & UNSPECIFIED	4	5.23	0.77	8	7.23	1.10	12	12.52	0.96
TOTAL MALIGNANT NEOPLASMS	30	35.41	0.85	50	57.87	0.86	80	93.28	0.86
CEREBROVASCULAR DISEASE	5	5.83	0.86	5	11.11	0.45 *	10	16.94	0.59
DISEASES OF THE HEART									
CHRONIC RHEUMATIC HEART DISEASE	1	0.65	1.53	1	0.71	1.41	2	1.36	1.47
ARTERIOSCLEROTIC HEART DISEASE	44	42.35	1.04	43	71.80	0.68	107	114.15	0.94
CHRONIC ENDOCARDITIS	0	0.24	0.00	0	0.47	0.00	0	0.72	0.00
HYPERTENSIVE HEART DISEASE	0	0.73	0.00	1	1.30	0.77	1	2.03	0.49
OTHER HEART DISEASE	8	6.08	1.32	13	10.05	1.29	21	16.13	1.30
OTHER CARDIOVASCULAR DISEASE									
RHEUMATIC FEVER	0	0.02	0.00	0	0.02	0.00	0	0.04	0.00
HYPERTENSION WITHOUT MENTION OF HEART	1	0.32	3.16	0	0.59	0.00	1	0.90	1.11
GENERAL ARTERIOSCLEROSIS	2	0.28	7.14	0	0.48	0.00	2	0.73	2.75
OTHER	5	2.48	2.01	3	4.09	0.73	8	6.58	1.22
EXTERNAL CAUSES OF DEATH									
MOTOR VEHICLE ACCIDENTS	4	3.36	1.19	8	14.23	0.56	12	17.59	0.68
SUICIDE	8	2.97	0.00	3	7.16	0.42	3	10.12	0.30 *
HOMICIDE	0	0.32	0.00	1	2.48	0.40	1	2.80	0.36
OTHER ACCIDENTS	2	3.38	0.59	4	8.70	0.46	6	12.08	0.50 *
OTHER CAUSES									
INFLUENZA	0	0.02	0.00	0	0.06	0.00	0	0.08	0.00
PNEUMONIA	4	1.61	2.49	3	2.81	1.07	7	4.41	1.59
NEPHRITIS & NEPHROSIS	0	0.51	0.00	1	1.37	0.73	1	1.88	0.63
TUBERCULOSIS OF RESPIRATORY SYSTEM	0	0.03	0.00	0	0.18	0.00	0	0.22	0.00
DIABETES MELLITUS	0	0.68	0.00	2	1.51	1.32	2	2.19	0.91
PEPTIC ULCER	0	0.39	0.00	0	0.59	0.00	0	0.97	0.00
APPENDICITIS	0	0.03	0.00	0	0.01	0.00	0	0.05	0.00
HERNIA & INTESTINAL OBSTRUCTION	0	0.20	0.00	1	0.24	0.18	1	0.44	2.29
CIRRHOSIS OF THE LIVER	2	1.55	1.24	0	2.60	0.00	2	4.15	0.48
EMPHYSEMA	0	0.82	1.22	3	1.86	1.62	4	2.68	1.50
SYMPTOMS & ILL-DEFINED CONDITIONS	0	1.11	0.00	1	1.98	0.51	1	3.89	0.32
RESIDUAL	6	6.70	0.69	13	13.45	0.97	19	22.15	0.86
UNSPECIFIED	0	0.05	0.00	0	0.18	0.00	0	0.23	0.00
TOTAL ALL CAUSES	115	120.12	0.96	176	217.85	0.81 *	291	337.99	0.86 *

STAT SIG EXCESS (+) / DEFICIT (-) AT 2-TAILED 0.10 LEVEL (CALCULATED ONLY WHERE EITHER OBS OR EXP GE 4)

AJPC009995

EID521391

OBSERVED AND EXPECTED DEATHS USING POPULATION-BASED RATES - H00TB9
WASHINGTON (PARKERSBURG) - PLANT (LOCATION CODE = 011)
1957 - 1957, FEMALES

CAUSE OF DEATH	SALARY ROLL			PAGE ROLL			PAGE & SALARY		
	OBSERVED	EXPECTED	OBS/EXP	OBSERVED	EXPECTED	OBS/EXP	OBSERVED	EXPECTED	OBS/EXP
MALIGNANT NEOPLASMS	0	0.01	0.00	0	0.00	-	0	0.01	0.00
BUCCAL CAVITY & PHARYNX	0	0.29	0.00	0	0.12	0.00	0	0.42	0.00
DIGESTIVE ORGANS	0	0.44	0.00	0	0.06	0.00	0	0.60	0.00
RESPIRATORY SYSTEM	0	0.92	0.00	0	0.40	0.00	0	1.12	0.00
BREAST	0	0.26	0.00	0	0.06	0.00	0	0.32	0.00
GENITAL ORGANS	0	0.04	0.00	0	0.03	0.00	0	0.08	0.00
URINARY ORGANS	0	0.28	0.00	0	0.11	0.00	0	0.39	0.00
LYMPHATIC, ETC.	0	0.33	0.00	0	0.00	0.00	0	0.33	0.00
OTHER & UNSPECIFIED	0	2.57	0.00	0	0.56	0.00	0	3.15	0.00
TOTAL MALIGNANT NEOPLASMS	0	0.28	0.00	0	0.32	0.00	0	0.39	0.00
CEREBROVASCULAR DISEASE	0	0.09	0.00	0	0.00	-	0	0.09	0.00
DISEASES OF THE HEART	2	0.55	3.54	0	0.17	0.00	2	0.73	2.72
CHRONIC RHEUMATIC HEART DISEASE	0	0.03	0.00	0	0.00	-	0	0.03	0.00
ARTERIOSCLEROTIC HEART DISEASE	0	0.08	0.00	0	0.00	-	0	0.08	0.00
CHRONIC ENDOCARDITIS	1	0.13	6.15	0	0.01	0.00	1	0.17	5.80
HYPERTENSIVE HEART DISEASE	0	0.00	0.00	0	0.00	-	0	0.00	0.00
OTHER HEART DISEASE	0	0.00	0.00	0	0.00	-	0	0.00	0.00
OTHER CARDIOVASCULAR DISEASE	0	0.00	-	0	0.00	-	0	0.00	-
RHEUMATIC FEVER	0	0.01	0.00	0	0.00	-	0	0.01	0.00
HYPERTENSION WITHOUT MENTION OF HEART	0	0.00	-	0	0.00	-	0	0.00	-
GENERAL ARTERIOSCLEROSIS	0	0.09	0.00	0	0.03	0.00	0	0.12	0.00
OTHER	0	0.00	0.00	0	0.00	-	0	0.00	-
EXTERNAL CAUSES OF DEATH	1	0.48	2.10	0	0.24	0.00	1	0.72	1.59
MOTOR VEHICLE ACCIDENTS	0	0.12	0.00	0	0.09	0.00	0	0.21	0.00
SUICIDE	0	0.11	0.00	0	0.15	0.00	0	0.26	0.00
HOMICIDE	0	0.11	0.00	0	0.04	0.00	0	0.15	0.00
OTHER ACCIDENTS	0	0.00	0.00	0	0.00	-	0	0.00	-
OTHER CAUSES	0	0.00	-	0	0.00	-	0	0.00	-
INFLUENZA	0	0.06	0.00	0	0.00	-	0	0.06	0.00
PNEUMONIA	0	0.00	-	0	0.00	-	0	0.00	-
NEPHRITIS & NEPHROSIS	0	0.00	-	0	0.00	-	0	0.00	-
TUBERCULOSIS OF RESPIRATORY SYSTEM	0	0.02	0.00	0	0.00	-	0	0.02	0.00
DIABETES MELLITUS	0	0.00	-	0	0.00	-	0	0.00	-
PEPTIC ULCER	0	0.00	-	0	0.00	-	0	0.00	-
APPENDICITIS	0	0.00	-	0	0.00	-	0	0.00	-
HERNIA & INTESTINAL OBSTRUCTION	0	0.00	-	0	0.00	-	0	0.00	-
CIRRHOSIS OF THE LIVER	0	0.06	0.00	0	0.00	-	0	0.06	0.00
EMPHYSEMA	0	0.02	0.00	0	0.03	0.00	0	0.03	0.00
SYMPTOMS & ILL-DEFINED CONDITIONS	1	0.09	10.74	0	0.00	-	1	0.13	7.89
RESIDUAL	2	0.43	4.69	1	0.06	15.94	3	0.49	6.13
UNSPECIFIED	0	0.00	-	0	0.03	0.00	0	0.03	0.00
TOTAL ALL CAUSES	7	5.30	1.32	1	1.59	0.63	8	6.89	1.16

AJTP009996

EID521392