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

P.J. Harvey a,*, H.K. Handley a, M.P. Taylor b

a Department of Earth and Planetary Sciences, Faculty of Science and Engineering, Macquarie University, Sydney, NSW 2109, Australia
b Department of Environmental Sciences, Faculty of Science and Engineering, Macquarie University, Sydney, NSW 2109, Australia

A R T I C L E   I N F O

Article history:
Received 3 May 2016
Received in revised form 27 July 2016
Accepted 29 July 2016

Keywords:
Contamination
Brass
Leaching
Exposure

A B S T R A C T

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.

© 2016 Elsevier Inc. All rights reserved.

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;
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:


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) 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 HNO3 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
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 μg/L, copper – 2000 μg/L, lead – 10 μg/L, manganese – 500 μg/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 1.5h 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 μg/L) were treated as 0.5 μg/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 μg/L. Sample bottle blanks contained < 1 μg/L except a small number (n = 10) that contained < 2 μg/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 μg/L, max – 3800 μg/L) and lead (average – 3.9 μg/L, max – 89 μg/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 μg/L, median – < 1 μg/L; manganese: mean – 110 μg/L, median – 3.9 μg/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 μg/L, median – 123 μg/L). Lead was detected in 56% (n = 118) of samples (mean – 3.7 μg/L, median – 1.3 μg/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 24h 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.

<table>
<thead>
<tr>
<th>Region</th>
<th>Element</th>
<th>Mean (µg/L)</th>
<th>Median (µg/L)</th>
<th>Max (µg/L)</th>
<th>Min (µg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Far Western</td>
<td>Arsenic</td>
<td>1</td>
<td>1.1</td>
<td>1.3</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Copper</td>
<td>146</td>
<td>38.0</td>
<td>1200</td>
<td>2.3</td>
</tr>
<tr>
<td></td>
<td>Lead</td>
<td>1.4</td>
<td>&lt; 1</td>
<td>5.5</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Manganese</td>
<td>4.7</td>
<td>1.6</td>
<td>23.0</td>
<td>&lt; 1</td>
</tr>
<tr>
<td>Orana</td>
<td>Arsenic</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
<td>5.8</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Copper</td>
<td>617</td>
<td>115</td>
<td>3800</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Lead</td>
<td>8.2</td>
<td>2.2</td>
<td>89.0</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Manganese</td>
<td>14.0</td>
<td>8.1</td>
<td>82.0</td>
<td>&lt; 1</td>
</tr>
<tr>
<td>New England</td>
<td>Arsenic</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
<td>1.5</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Copper</td>
<td>316</td>
<td>163</td>
<td>2330</td>
<td>1.5</td>
</tr>
<tr>
<td></td>
<td>Lead</td>
<td>4.4</td>
<td>2.5</td>
<td>28.0</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Manganese</td>
<td>9.6</td>
<td>2.8</td>
<td>50.0</td>
<td>&lt; 1</td>
</tr>
<tr>
<td>North Coast</td>
<td>Arsenic</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Copper</td>
<td>90.0</td>
<td>64.0</td>
<td>188</td>
<td>15.0</td>
</tr>
<tr>
<td></td>
<td>Lead</td>
<td>0.6</td>
<td>0.5</td>
<td>1.6</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Manganese</td>
<td>12.0</td>
<td>&lt; 1</td>
<td>2.5</td>
<td>&lt; 1</td>
</tr>
<tr>
<td>Hunter</td>
<td>Arsenic</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Copper</td>
<td>***</td>
<td>***</td>
<td>200</td>
<td>200</td>
</tr>
<tr>
<td></td>
<td>Lead</td>
<td>***</td>
<td>***</td>
<td>3.4</td>
<td>3.4</td>
</tr>
<tr>
<td></td>
<td>Manganese</td>
<td>***</td>
<td>***</td>
<td>4.9</td>
<td>4.9</td>
</tr>
<tr>
<td>Illawarra</td>
<td>Arsenic</td>
<td>***</td>
<td>***</td>
<td>***</td>
<td>***</td>
</tr>
<tr>
<td></td>
<td>Copper</td>
<td>***</td>
<td>***</td>
<td>***</td>
<td>***</td>
</tr>
<tr>
<td></td>
<td>Lead</td>
<td>***</td>
<td>***</td>
<td>***</td>
<td>***</td>
</tr>
<tr>
<td></td>
<td>Manganese</td>
<td>***</td>
<td>***</td>
<td>***</td>
<td>***</td>
</tr>
<tr>
<td>South East</td>
<td>Arsenic</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Copper</td>
<td>376</td>
<td>140</td>
<td>2640</td>
<td>2.4</td>
</tr>
<tr>
<td></td>
<td>Lead</td>
<td>3.2</td>
<td>1.3</td>
<td>21.0</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Manganese</td>
<td>9.0</td>
<td>2.9</td>
<td>140</td>
<td>&lt; 1</td>
</tr>
<tr>
<td>Central West</td>
<td>Arsenic</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Copper</td>
<td>246</td>
<td>180</td>
<td>844</td>
<td>6.6</td>
</tr>
<tr>
<td></td>
<td>Lead</td>
<td>2.0</td>
<td>1.1</td>
<td>6.3</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Manganese</td>
<td>28.0</td>
<td>16.0</td>
<td>80.0</td>
<td>3.1</td>
</tr>
<tr>
<td>Riverina</td>
<td>Arsenic</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
<td>1.1</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Copper</td>
<td>278</td>
<td>29.0</td>
<td>1510</td>
<td>5.4</td>
</tr>
<tr>
<td></td>
<td>Lead</td>
<td>2.8</td>
<td>1.4</td>
<td>10.0</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Manganese</td>
<td>11.0</td>
<td>8.7</td>
<td>28.0</td>
<td>&lt; 1</td>
</tr>
<tr>
<td>Murray</td>
<td>Arsenic</td>
<td>***</td>
<td>***</td>
<td>***</td>
<td>***</td>
</tr>
<tr>
<td></td>
<td>Copper</td>
<td>***</td>
<td>***</td>
<td>***</td>
<td>***</td>
</tr>
<tr>
<td></td>
<td>Lead</td>
<td>***</td>
<td>***</td>
<td>***</td>
<td>***</td>
</tr>
<tr>
<td></td>
<td>Manganese</td>
<td>***</td>
<td>***</td>
<td>***</td>
<td>***</td>
</tr>
<tr>
<td>Total</td>
<td>Arsenic</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
<td>5.8</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Copper</td>
<td>364</td>
<td>123</td>
<td>3800</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Lead</td>
<td>3.9</td>
<td>1.3</td>
<td>89.0</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>Manganese</td>
<td>10.7</td>
<td>3.9</td>
<td>140</td>
<td>&lt; 1</td>
</tr>
</tbody>
</table>

** Cannot be calculated.

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

<table>
<thead>
<tr>
<th></th>
<th>Copper</th>
<th>Lead</th>
</tr>
</thead>
<tbody>
<tr>
<td>n</td>
<td>212</td>
<td>212</td>
</tr>
<tr>
<td>LOD – 50% ADWG</td>
<td>n²=190</td>
<td>78</td>
</tr>
<tr>
<td>% of n²</td>
<td>90.9%</td>
<td>37.7%</td>
</tr>
<tr>
<td>50–100% ADWG</td>
<td>n²=12</td>
<td>23</td>
</tr>
<tr>
<td>% of n²</td>
<td>6.2%</td>
<td>11.1%</td>
</tr>
<tr>
<td>&gt; ADWG</td>
<td>n²=10</td>
<td>17</td>
</tr>
<tr>
<td>% of n²</td>
<td>5.2%</td>
<td>8.8%</td>
</tr>
</tbody>
</table>

**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, R²=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
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,
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 mg/L for copper and 89 mg/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, R²=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 the highest lead concentration in household plumbing while the water is stagnant is known to cause (Schock and Neff, 1988). The decreased concentrations of copper in the samples following a 2-minute flush period suggest that once the stagnant 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 (Trianantafyllidou 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 flushing, 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 flushing 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 (tape 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 derSterren 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; JaneWyatt 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. Che- nyin 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 Rogen, 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.
References

Rajaratnam, G., Winder, C., Am, M., 2002. Metals in drinking water from new...
Wang, Y., 2016. Untold cities across America have higher rates of lead poisoning than Flint. The Washington Post.
Young, A., Nichols, M., 2016. Beyond Flint: Excessive lead levels found in almost 2000 (water) systems across all 50 states. USA Today.