Elements in Tank Water – Comparisons with Mains Water & Effects of Locality & Roofing Materials

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Abstract

Samples from a variety of rainwater tanks and mains water supplies from different regions in the Australian states of NSW, QLD, VIC, and SA, were analysed for a range of elements by ICP-MS. Investigations were performed to determine whether the distributions of elements in stored rainwater varied depending on urban or rural location and roof type (Pre-Painted steel, 55% Al-Zn alloy coated steel, Cement Tiles, or Galvanised Iron). Differences in elemental composition in stored tank water and mains water samples were also investigated. Mains water samples were found to contain significantly higher concentrations of sodium, strontium, barium, magnesium, and molybdenum, and significantly lower concentrations of manganese and zinc compared with tank water samples ($P<0.05$). Urban tank water samples contained significantly higher concentrations of strontium, barium, chromium, and molybdenum than rural tank water samples and significantly lower concentrations of iron, cadmium, manganese and copper compared with rural tank water samples ($P<0.05$). The concentrations of certain elements in samples from rainwater tanks with different roof catchment materials were also found to vary significantly ($P<0.05$). The majority of analysed samples were below Australian Drinking Water Guideline values for metal and non-metal concentrations, although 12% of samples exceeded guideline values for one or more element.

Key words: rainwater tank, elements, metals, non-metals, roofing materials, water quality

Introduction:

The implementation of rainwater harvesting systems on domestic allotments has the potential to mitigate the ongoing water supply crises experienced by many urban centres. External pollution sources, components in the rainwater harvesting system, and biological/chemical reactions in the tank all have the potential to influence rainwater quality. Industry and traffic emissions may contribute to increased concentrations of lead, zinc, cadmium, and arsenic in roof-harvested rainwater (Gould, 1999). Certain roofing materials also have the potential to increase lead and zinc concentrations (Hart & White, 2006; Yaziz et al., 1989; Simmons et al., 2001. Guttering and rainwater tank materials may influence the chemical composition of stored rainwater, while proximity to the ocean may result in higher concentrations of salts such as sodium. Nevertheless, monitoring results have revealed that rainwater collected from roofs of inner city dwellings and stored in tanks can provide acceptable water quality for hot water, toilet, laundry and outdoor uses (Cunliffe, 2004). A review of factors affecting water quality in rainwater harvesting systems highlights the scarcity of knowledge in relation to processes occurring within these systems (Spinks et al., 2003).

The impacts of roofing materials on the concentrations of metals and other elements in stored rainwater have been subject to limited analysis, with some authors finding metal roofs resulting in significantly higher concentrations of zinc (Hart & White, 2006; Yaziz et al., 1989) and lead (Hart & White, 2006; Simmons et al., 2001) compared with cement tiles and wooden roofs, respectively. However, these studies have been conducted in international locations that are subject to different climate and pollution characteristics to Australia with variable roof materials and ages. Lead concentrations were found to exceed Australian Drinking Water Guideline (ADWG, 2004) values in samples from roofs with lead flashing in a study by Magyar et al. (2006), while zinc and copper concentrations were almost three orders of magnitude higher in runoff from roofs consisting of zinc sheeting than in samples affected by atmospheric deposition alone in North America (Forster, 1996). Copper concentrations, however, did not vary significantly for wooden, metal, or asphalt roofs, suggesting that roof type did not affect copper concentrations (Hart & White, 2006).

There is little published data, with the exception of that of Coombes et al. (2000) and Magyar et al. (2006), on the effects on water quality of roofing materials most commonly used in Australian homes: Cement Tiles, pre-painted steel (PPS), Galvanised Iron (GI), and 55% Al-Zn alloy coated steel (ZCS). While the effects of glazed tile, painted metal sheeting, and galvanised iron roofs on elemental composition of tank water have been studied (Magyar *et al.*, 2006), this research was only conducted using new pilot roofing and tank materials, which may not represent actual housing situations and did not take into account the effects of aging processes on roof materials. As the majority of roofs in Australia are greater than 1 year old, this is an important area of research. Additionally, a wider range of metals than those included in the majority of studies to date may be important for human health. The aims of this study were to determine whether differences exist between concentrations of elements in rain and mains water samples, urban and rural tank water samples, and samples collected from tanks with different roof materials used for water catchment. This study did not examine the relative influences of air pollution, roof and tank materials, and proximity to the ocean on the concentration of metals and non-metals in harvested rainwater, but instead provided a snapshot of metal and non-metal contamination in tank water samples from a broad range of sites across rural and urban Australia.

Methodology:

Participant Recruitment and Participating Tank Characteristics:

Participants of this national survey of tank water quality were selected from rainwater industry databases and from the community. Each participating household provided detailed information on their rainwater harvesting system. The participant database extends to 40 tanks, which include sites in Queensland, New South Wales, Victoria, and South Australia. Tables 1a and 1b below summarise the main characteristics of interest of these locations, and provide the numbers of samples obtained from rural and urban regions, along with total numbers of tank water and mains water samples analysed.

Table 1a: Roof and Tank Types

Roof Types	% of Tank Cohort
Galvanised Iron	20.7%
Pre-Painted Steel	37.9%
Cement Tile	23.0%
55% Al-Zn Coated Steel	18.4%
Tank Types	**% of Tank Cohort**
AQUAPLATE® Steel	37.5%
Concrete	42.0%
Plastic	12.5%
Galvanised Iron	6.8%
“Bladder Bag”	1.1%

Table 1b: Sample Types and Tank water Uses

Sample Types	Samples
Urban Samples	34
Rural Samples	53
Total Tank water samples	87
Total Mains water samples	14
Tank water Uses	**% of Tank Cohort**
Garden only	37.5%
Domestic (excluding drinking)	17.5%
All Uses	30%
Backed up by mains water	22.5%

Sample Collection:

Polyethylene (500mL) sampling containers were posted out to participants in foam eskies, along with ice bricks, a sampling protocol, surgical gloves, and an express Australia Post satchel for return postage. Participants were instructed to collect samples from the following outlets, where available: a hot tap supplied by the tank water; a cold tap supplied by the tank water; the external tank tap; and a tap supplied by mains water. Taps were allowed to run for 3 seconds before samples were collected, and surgical gloves were worn to minimise potential contamination of samples. The samples were then placed in eskies, along with frozen ice bricks. Samples were posted back to The University of Newcastle within 24 hours of collection and, in the majority of cases, were received by the laboratory within 48 hours of postage. All samples were prepared for analysis by Inductively Coupled Plasma – Mass Spectrometry (ICP-MS) within 5 days of sample collection.

Sample Preparation for Analysis of Metal and Non-metal Content by ICP-MS:

For each sample 9.7 mL of water was filtered through a 0.45µm membrane filter into a 10mL yellow capped polypropylene tube, and 300µL of concentrated (65% SUPRAPUR ICP-MS grade) nitric acid was added to each sample. Samples were then stored at 4°C prior to analysis by ICP-MS.

Sample Analysis by ICP-MS:

Inductively coupled plasma – mass spectrometry was used to detect and measure a range of elements listed in Table 2. The detection limits for this technique ranged from 0.001 parts per trillion (ppt) for iron and strontium, to 292 ppt for phosphorus.

Statistical Analyses:

Tank Water versus Mains and Urban versus Rural Tank Water: data was tested for normality using Levene's test (Statistica™, Ver. 6.0). In cases where the homogeneity of variance assumption was violated, T Tests were carried out with separate variance estimates.

Effects of Roofing Materials: Tukey's test of Honest Significant Difference for unequal N was carried out to determine whether significant ($P<0.05$) differences existed between different sample groups.

Results:

Comparison of Tank water versus Mains Water:

Rain and mains water samples were collected and prepared for analysis to assess the relative distributions of metals and other elements. This first approach compared metal and non-metal concentrations in tank water samples (N=87) collected from the tank outlets, with concentrations of the same elements in mains water samples (N=14) from the same and/or different experimental sites. The mains water samples had significantly higher concentrations ($P<0.05$) of sodium, strontium, barium, magnesium, and molybdenum, and significantly lower concentrations of manganese and zinc, compared with tank water samples, as shown in Table 2 and Figure 1. Median values are also provided in Table 2, to allow for the possibility of one or two outlier metal or non-metal concentrations skewing the mean.

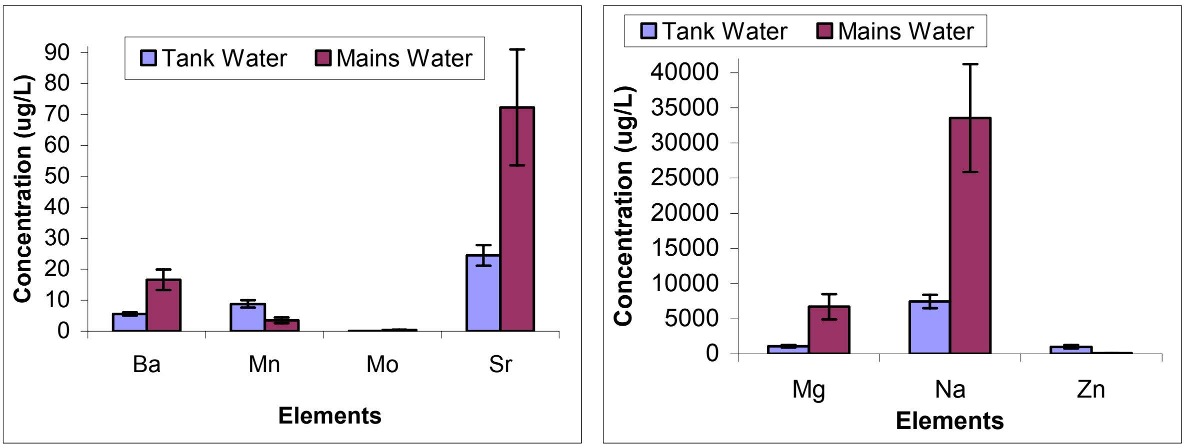


Figure 1: Summary of significant differences in metal and non-metal concentrations between tank water (N=87) and mains water (N=14) samples (±SE).

There were no significant differences in concentrations of other detected elements between mains and tank water samples. All average metal and non-metal concentrations were below Australian Drinking Water Guideline limits. However, 13 (12%) samples exceeded guideline values for one or more elements (Table 3), including 1 mains sample which contained iron concentrations in excess of ADWG (2004) values. Despite these samples being above recommended limits, in all cases where sampling at these sites has been replicated, subsequent samples were shown to be within ADWG limits for all elements. There have been insufficient data published to set a health-based guideline for iron and zinc concentrations (ADWG, 2004). Subsequently, taste thresholds are likely to be overstepped before concentrations are sufficiently high to cause health effects. The single sample which contained elevated arsenic levels was collected from a 'Bladder Tank' which was used for lawn watering purposes only. The same 'Bladder Tank' sample also accounted for one of the two cases of nickel exceeding guidelines, and one of the two cases of iron exceeding ADWG (2004). The next sample taken from this same bladder tank six months later in January 2007, however, complied with ADWG for all elements tested.

Table 3: Comparison of samples with Guideline Values for Elements (ADWG, 2004)

Element	Samples above ADWG	# of Samples above ADWG	Sample Type		Range of Values which Exceeded ADWG (µg/L)	ADWG (µg/L)
			Rain	Mains		
Arsenic	0.90%	1	1	0	7.21	7
Iron	1.80%	2	1	1	338.0 – 871.7	*300*
Lead	7.20%	8	8	0	10.6-32.4	10
Nickel	1.80%	2	2	0	30.2 – 158.95	20
Zinc	1.80%	2	2	0	9774.6 – 11662.3	*3000*

Table 2: Average (±SE) and median concentrations, T, *P*, and ADWG (2004) values for metals and non-metals in tank water and mains water samples.

Element	Tank Means (N=87)	± SE (ug/L)	Mains Means (N=14)	± SE (ug/L)	Tank Median (N=87)	Mains Median (N=14)	T	*P*	ADWG (ug/L)
Arsenic	0.37	0.03	0.38	0.05	0.31	0.42	-0.22	>0.05	7
Barium	**5.56**	**0.53**	**16.65**	**3.30**	**4.06**	**10.46**	**-3.31**	**0.01**	700
Bismuth	0.04	0.01	0.07	0.06	0.01	0.01	-0.61	>0.05	
Cadmium	0.17	0.04	0.09	0.06	0.04	0.02	0.73	>0.05	2
Caesium	0.04	0.01	0.03	0.01	0.03	0.02	0.84	>0.05	
Chromium	0.83	0.30	1.42	0.77	0.19	0.14	-0.73	>0.05	50
Cobalt	0.16	0.05	0.08	0.03	0.06	0.05	0.71	>0.05	
Copper	76.69	10.32	114.58	44.76	36.67	36.38	-1.22	>0.05	*1000*
Iron	46.32	7.50	59.83	26.66	15.77	18.15	-0.63	>0.05	*300*
Lead	3.45	0.44	1.92	0.62	2.12	1.33	1.35	>0.05	10
Magnesium	**1084.13**	**183.97**	**6703.73**	**1805.64**	**619.38**	**2048.86**	**-3.10**	**0.01**	
Manganese	**8.79**	**1.21**	**3.51**	**0.97**	**4.35**	**2.79**	**3.42**	**0.00**	*100*
Molybdenum	**0.11**	**0.02**	**0.38**	**0.12**	**0.06**	**0.07**	**-2.21**	**0.04**	50
Nickel	2.85	1.82	1.74	1.37	0.70	0.23	0.24	>0.05	20
Phosphorus	41.98	7.72	63.53	39.27	24.94	4.02	-0.54	>0.05	
Potassium	1377.40	137.34	2481.12	604.04	947.06	894.36	-1.78	>0.05	
Rubidium	1.33	0.16	2.24	0.46	0.91	1.74	-2.09	>0.05	
Selenium	1.39	0.23	1.70	0.54	0.16	0.96	-0.50	>0.05	10
Silver	0.01	0.00	0.03	0.03	0.00	0.00	-1.00	>0.05	100
Sodium	**7434.88**	**950.69**	**33552.23**	**7690.15**	**5637.92**	**40116.51**	**-3.37**	**0.01**	180000
Strontium	**24.54**	**3.36**	**72.33**	**18.75**	**15.98**	**24.15**	**-2.51**	**0.03**	
Tin	0.59	0.16	0.67	0.40	0.05	0.00	-0.17	>0.05	
Titanium	0.09	0.03	0.11	0.09	0.00	0.00	-0.23	>0.05	
Uranium	0.01	0.00	0.00	0.00	0.00	0.00	1.79	>0.05	
Vanadium	0.47	0.13	0.25	0.05	0.22	0.15	0.69	>0.05	
Zinc	**1017.64**	**261.07**	**77.92**	**28.84**	**218.73**	**47.48**	**3.58**	**0.00**	*3000*

Comparison of Urban & Rural Rainwater Tanks:

Samples were collected from urban (N= 34) and rural (N= 53) rainwater tanks to assess potential impacts of urban pollution. Strontium, barium, chromium, and molybdenum were present in significantly higher concentrations in urban tank water samples than in rural tank water samples ($P<0.05$), while concentrations of iron, cadmium, manganese, and copper were significantly lower ($P<0.05$). The differences between the rural and urban harvested tank water samples are displayed in Figure 2.

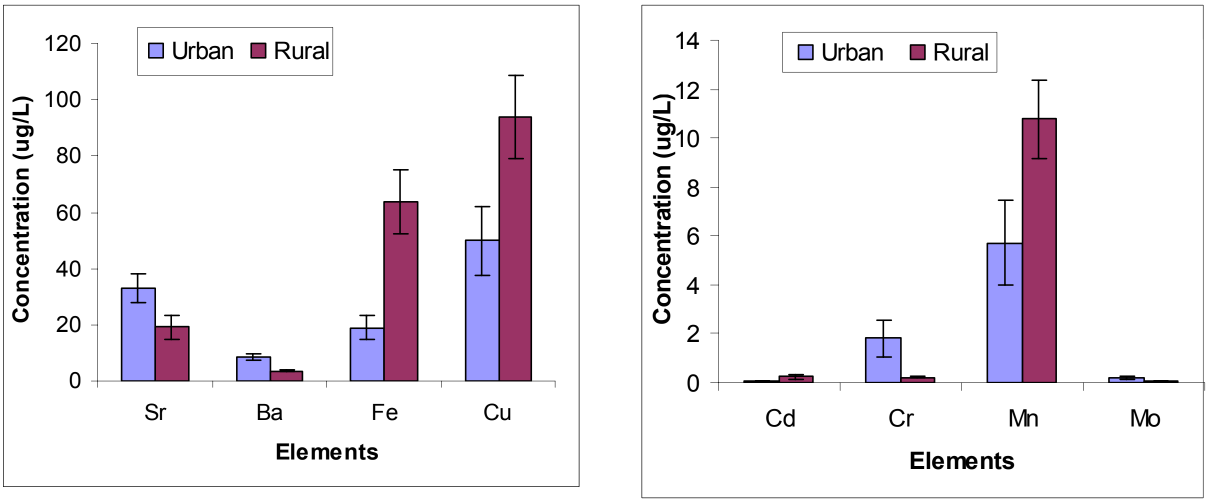


Figure 2: Significant differences between urban (N=34) and rural (N=53) samples.

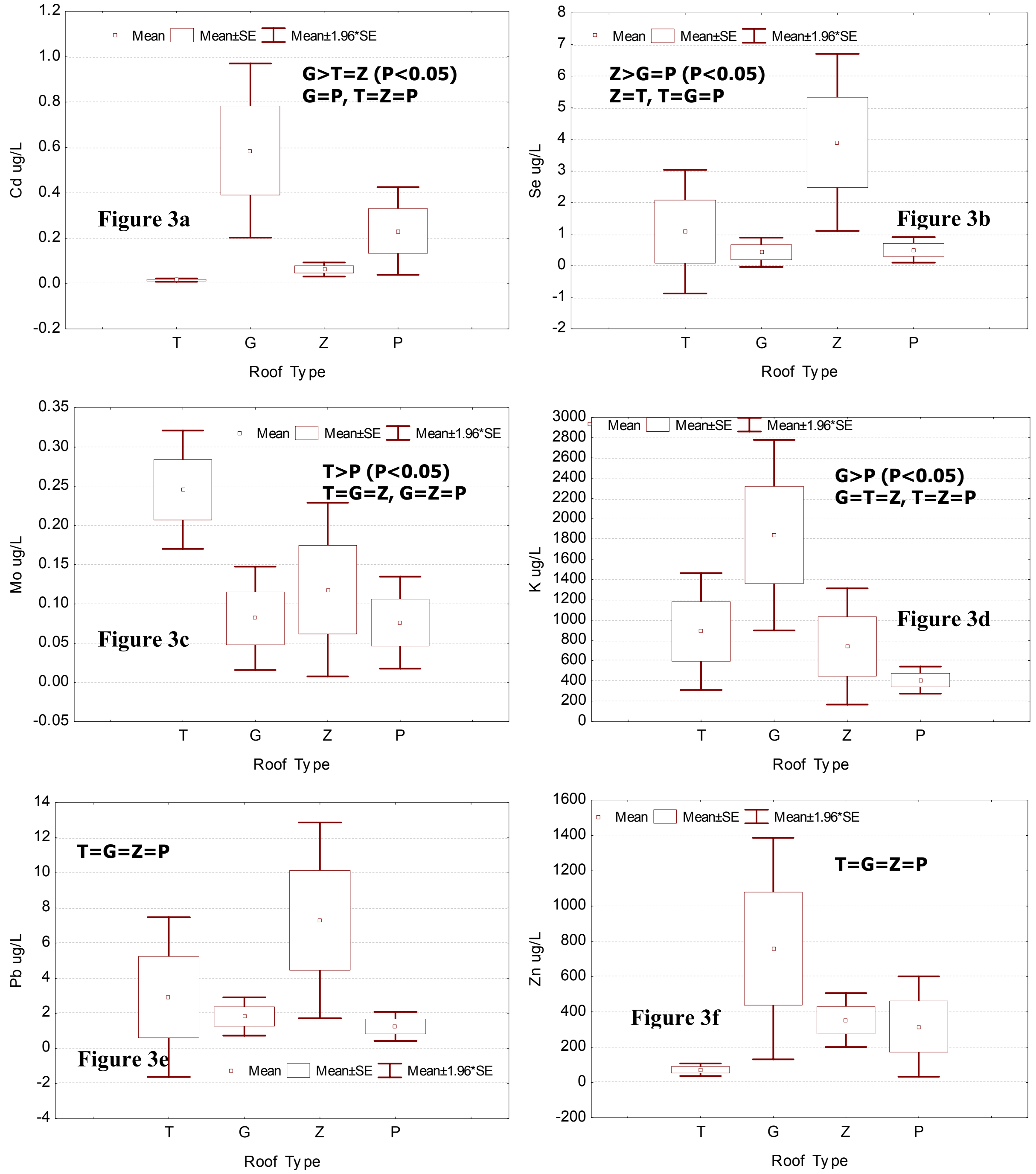
Effects of Roofing Materials on Concentrations of Metals and Non-Metals in Harvested Rainwater:

Samples were taken from tanks attached to roofs made from pre-painted steel, Galvanised Iron, Cement Tiles, and Al-Zn coated steel to determine the potential influence of roofing materials on water quality. In order to minimise confounding variables only samples collected from rainwater tanks made from AQUAPLATE® steel were included in this analysis. These tanks were chosen as they were one of the best-represented groups in the tank cohort. Levels of rubidium, cadmium, caesium, vanadium, molybdenum, potassium, and selenium varied significantly between samples from sites with different roof catchment materials (Table 4).

Table 4: Median concentrations of elements that varied significantly (P<0.05) with roof type.

Mean ±SE, Median		Rubidium	Cadmium	Caesium	Vanadium	Molybdenum	Potassium	Selenium
Tiles								
Mean	**(ug/L)**	0.58	0.02	0.03	2.69	0.25	887.44	1.08
±SE	**(ug/L)**	0.08	0.00	0.01	1.15	0.04	294.15	1.00
Median	**(ug/L)**	0.56	0.02	0.03	0.50	0.21	439.98	0.00
Galvanised Iron								
Mean	**(ug/L)**	1.94	0.59	0.11	0.16	0.08	1838.94	0.43
±SE	**(ug/L)**	0.77	0.20	0.03	0.04	0.03	479.49	0.24
Median	**(ug/L)**	1.95	0.54	0.09	0.19	0.07	1855.73	0.19
Pre-Painted Steel								
Mean	**(ug/L)**	0.55	0.23	0.02	0.11	0.08	408.19	0.50
±SE	**(ug/L)**	0.12	0.10	0.01	0.02	0.03	67.83	0.21
Median	**(ug/L)**	0.36	0.08	0.01	0.08	0.06	477.90	0.10
Al-Zn Coated Steel								
Mean	**(ug/L)**	0.40	0.06	0.04	0.21	0.12	740.21	3.90
±SE	**(ug/L)**	0.13	0.02	0.02	0.03	0.06	292.40	1.43
Median	**(ug/L)**	0.33	0.05	0.03	0.22	0.09	532.77	4.42
***P* value**		<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
ADWG	**(ug/L)**		2			50		10

There were no significant differences in the concentrations of any other tested elements in tank water harvested from different roof types. Box (SE) and whisker (1.96*SE) plots (Figures 3a-f) indicate relative concentrations of a selection of elements between roof types. Cadmium concentrations were significantly higher in samples from sites with galvanised iron roofs, than those from roofs made of cement tiles or 55% Al-Zn coated steel (Figure 3a), and samples collected from sites with roof catchments made of Al-Zn coated steel had significantly higher concentrations of selenium than samples from sites with roofs made of pre-painted steel or galvanised iron (Figure 3b). Molybdenum concentrations were significantly higher in samples from sites with tile roof catchments, than in samples from roofs made of pre-painted steel (Figure 3c), and average potassium concentrations were significantly higher in samples collected from sites with Al-Zn coated steel roofs than those with pre-painted steel roofs (Figure 3d).



Figures 3a-f: Concentrations of elements in samples collected from different roof types. G: Galvanised Iron; P: Pre-Painted steel; T: Tiles; Z: Al-Zn Coated steel. (Significant differences (P<0.05) are marked as ">" or no difference as "=").

Lead and zinc have been included in these plots despite concentrations of these elements not varying significantly between samples from sites with different roof types. Lead is a key element with respect to human health, and zinc is an element which has previously been found to vary in abundance depending on the type of roof catchment (Forster, 1996; Hart & White, 2006; Yaziz *et al.*, 1989). Samples from sites with roofs made from cement tiles or 55% Al-Zn coated steel contained greater variances in lead concentrations, than samples from sites with roofs made from pre-painted steel or galvanised iron (Figure 3e), while galvanised iron roofs resulted in greater variations in zinc concentration than any of the other roof types tested (Figure 3f).

Discussion

Elements in Tank water – Comparisons with Guideline Values:

Concentrations of arsenic, iron, lead, nickel, and zinc were found to exceed ADWG (2004) values in at least one sample. Further research is required to determine the probable sources of these contaminants at each site where an element exceeded guideline values. However, as mentioned previously, in each case where a sample was found to exceed ADWG values for an element, subsequent samples from the same site were found to be within guideline values for the same element. This could be the result of one of several different processes. The concentrations of various elements in tank water are usually rainfall dependent, for example, (Coombes et al., 2000) with concentrations usually higher immediately following rain events. Analysis of rainfall effects has been commenced by the authors. The Australian Drinking Water Guidelines (2004) for metals and non-metals are based on cumulative exposure, assuming average adult water consumption of 2L per day. Subsequently, an elevated concentration of lead, for example, may not pose serious health risks if it occurs only rarely throughout the year, as long as it is not sufficiently high to be toxic in a single dose. Average concentrations of lead, zinc, copper, iron and cadmium were within the range reported in Coombes and Mitchell (in Wong (Ed.), 2006), in which the results from 6 Australian studies on tank water quality are collated.

Elements in Tank water versus Mains Water:

Mains water was found to contain significantly higher concentrations of sodium, strontium, silver, barium, magnesium, and molybdenum, and significantly lower concentrations of manganese and zinc than tank water samples ($P<0.05$). This may be the result of water treatment processes carried out on mains water supplies, or the effect of mains water contact with metal pipes during its transportation. This comparison did not take into account confounding variables, such as roof types and ages, or tank types or ages. Instead, this analysis aimed to provide a snapshot of metal and non-metal contamination in roof-harvested tank water, compared with reticulated water supplies.

Elements in Urban versus Rural Tank water Samples:

Concentrations of certain elements differed significantly between urban and rural tank water samples ($P<0.05$). Increased iron, cadmium, manganese, and copper in rural samples may be the result of contamination from agricultural use or from older roofs or tanks, while increased strontium, barium, chromium, and molybdenum in urban tanks may be a function of air quality in urban areas.

Elements in Samples from Sites with Different Roofing Materials:

Levels of cadmium, caesium, molybdenum, potassium, rubidium, selenium, and vanadium varied significantly between samples from sites with different roof catchment materials ($P<0.05$).Further research is required to determine the relative impacts of roofing materials, and confounding factors such as pollution levels, proximity to the coast, and guttering materials, on the concentrations of elements in harvested tank water. Cadmium levels may have been impacted by air pollution at some sites, the presence of molybdenum may be the result of mining or agricultural practices (ADWG, 2004), while potassium concentrations may have been impacted by the proximity of some sites to the ocean. Research is currently underway to measure levels of metals and non-metals present in direct roof runoff from a variety of roof types, which will provide greater understanding of the contribution of roofing materials on the total metal and non-metal contaminant load in the rainwater tank. As more data becomes available, statistical analysis of the effects of potentially confounding variables on the concentrations of metals and non-metals in tank water will also be possible. However, samples collected from tap outlets, as in this study, provide a greater indication of water quality at the end-point, due to the fact that the quality of tank water has been shown to change from the roof runoff to the tank outlet, as a result of treatment train processes occurring within tanks (Coombes et al., 2000).

Conclusions

- The majority (88%) of tank water samples complied with ADWG (2004) values for metal and non-metal concentrations, while 93% of mains samples complied with guidelines.
- Mains samples had higher concentrations of sodium, strontium, barium, magnesium, and molybdenum, and lower concentrations of manganese and iron than tank water ($P<0.05$).
- Samples from urban tanks contained significantly higher average concentrations of strontium, barium, chromium, and molybdenum, and significantly lower concentrations of iron, cadmium, manganese and copper than those from rural tanks ($P<0.05$).
- Concentrations of cadmium, caesium, molybdenum, potassium, rubidium, selenium, and vanadium varied significantly in tank water samples collected at sites with different roof catchments. Further research is required in order to more fully understand the implications of different roofing materials on the quality of roof-harvested tank water, due to the potential for confounding factors such as air pollution and guttering and tank materials to influence the composition of tank water.
- This study demonstrated differences in concentrations of elements between mains and tank water samples, urban and rural tank water samples, and tank samples fed from different roof catchments.

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