

Rainfall harvesting and coal dust: the potential health impacts of trace elements in coal dust in rainwater

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ABSTRACT

Coal dust contains trace elements of major/minor concern due to their potential impact on human health. Residential households in relatively close proximity to a coal dust source, such as a coal terminal, report that coal dust soils laundry and impacts upon visibility in the atmosphere. One question often asked is - does coal dust pose a threat to human health after being deposited on a rooftop and being washed into the rainwater tank? The answer lies in the likelihood and amount of coal dust (and trace elements) reaching the roof and being transferred to the tank; the potential for leaching of trace elements within the water column in the rainwater tank; and the water quality of rainwater at the point of use (kitchen tap). A range of coal types were supplied by the Dalrymple Bay Coal Terminal (DBCT) which reflected the likely composition of existing coal dust in the area. Leaching experiments were undertaken on a range of coal types (of particle size < 63 µm) to determine the extent of elemental leaching from the coal dust to the rainwater. In addition, water samples from three households (tanks and taps) in the Dalrymple Bay area were obtained and tested for elements considered both a major and minor concern in coal. Leaching experiments suggest that major/minor trace elements in the coal dust are not likely to be a main contributor to trace elements found in rainwater; and in fact may assist in removing some trace elements from the rainwater in the tank. Water samples from all three households recorded concentrations far lower than Australian Drinking Water Guidelines for all tested elements.

Keywords: coal dust, rainwater, health impacts, trace elements.

INTRODUCTION

In residential areas without reticulated water supply, the dominant source of water in Australia is roof-harvested rainfall from the rainwater tank, where it is often used for all indoor/outdoor demands. Airborne particulates are regularly deposited on rooftops in urban areas, including dust, leaves, paint flakes, roof material, etc. Birds, possums and other wildlife also deposit faecal matter onto rooftops. Approximately 3.5 million people in Australia currently use harvested rainwater as their only water

source, and historically, there have been no recorded major health epidemics as a result. However, residents within close proximity of coal loading facilities often question air quality and the impact on rainwater quality.

The Dalrymple Bay Coal Terminal (DBCT) is located within the Port of Hay Point approximately 40 km south of Mackay (Queensland, Australia). The DBCT has a current coal shipping capacity of 70 million tonnes per annum (Mtpa) with plans to expand to 85 Mtpa in the near future. The adjacent townships of Lousia Creek, Salonika and Half Tide are located to the west and south of the Port of Hay Point. These townships use rainwater harvesting as a means of potable supply and some houses are connected to the reticulated town water supply. Based on Australian Bureau of Meteorology data between October 1995 and June 2005, the annual average rainfall was approximately 1170 mm; with a maximum of 1850 mm y⁻¹ and a minimum of 700 mm y⁻¹.

Coal dust has the potential to cause annoyance due to soiling material surfaces and visibility of dust in the atmosphere (Katesstone Environmental 2006). Coal contains (amongst other elements), trace amounts of sulfur (S), chlorine (Cl), fluorine (F), arsenic (As), boron (B), cadmium (Cd), mercury (Hg), molybdenum (Mo), lead (Pb), selenium (Se), chromium (Cr), copper (Cu), nickel (Ni), vanadium (V) and zinc (Zn). However, much of the research and emission guideline development concentrates on the potential effects of trace elements after coal is utilized for combustion, *i.e.* from power generation operations that produce fly-ash and other airborne particulates (National Pollution Inventory 2005).

The impact of these trace elements on human health varies depending on the length of exposure, the range of concentrations encountered and the pathway of ingestion. For example, long-term exposure to arsenic in drinking water can cause cancer in the skin, lungs, bladder and kidney. At low concentrations, mercury, chromium, cadmium, lead and arsenic can damage living organisms and they tend to accumulate in the natural food chain (*i.e.*, bio-accumulate in the tissue of plants and animals).

The aim of this study was to investigate and report on the potential health risks as a result of trace elements of minor and major concern contained in coal dust deposited on rooftops entering rainwater tank systems used for potable supply. Leaching tests were

undertaken on numerous coal types to reflect the potential for trace element release into rainwater in the tank. The Australian Drinking Water Guidelines (NHMRC 2004) provided the threshold levels considered safe for human consumption.

Rainwater samples were collected from three homes within the dust deposition zone in the Dalrymple Bay area to determine a snapshot of current rainwater quality. The concept of coal dust being deposited on rooftops and entering rainwater tank systems used for potable supply suggests there is a likelihood of harmful trace elements in coal being released into rainwater within the tank. However, the processes, pathways and likelihood of occurrence of (1) the entrainment of dust into the atmosphere; (2) coal dust deposition on the roof; (3) the release of trace elements from coal dust into rainwater in the tank and; (4) suitable water quality at the kitchen tap, all need to be understood before results can be appreciated. This is the first study of its kind in investigating the potential human health risks due to deposited coal dust on rooftops and resulting rainwater quality.

MATERIALS AND METHODS

Background data

Coal must first be mined and separated into 'product' by washing and sizing before being exported overseas. Product coal (< 50 mm) is transported to the loading facilities where it is often stockpiled before being loaded onto ships at some time in the future. The coal samples received for analysis represent product coal types exported from the DBCT; and were sampled under standard coal superintending protocols. These consist of a coal-sampler who supervises an automated coal sampling unit. A large primary-cutter samples the coal-stream prior to the buffer bins (before loading onto the ship). The primary-cut increment passes through a series of crushers and slotted-conveyer belts (sub-sampling) to obtain a smaller increment that represents the coal-stream. While coal is loaded onto individual ships, the coal-stream on the main conveyer belt is periodically 'cut' based on coal tonnage; and the final topsize of the product coal (for analysis) is approximately 12 mm after sub-sampling.

A range of trace elements in coal types (in mg kg⁻¹) exported from the DBCT were obtained indirectly from Introspec Consulting. The coal samples sent to the University of Newcastle reflected a range of product coals exported from the

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DBCT. Fifteen different export coal types representing the dominant coal types exported from the DBCT were sent to The University of Newcastle (NSW, Australia) for leaching tests. Actual depositional dust data for the Dalrymple Bay area was obtained from Introspec Consulting and Katestone Environmental to assist with interpretation and discussion of results.

Coal samples and leaching tests

Prior to analysis, Coal samples were dried at 40°C for 24 hours before being sized at 63 µm (0.063 mm). The leaching tests were performed on the < 63 µm size fraction, which was likely to reflect the size of coal-

dust particles that are deposited on rooftops in the Dalrymple Bay area.

Leaching tests were undertaken to determine the potential release of major and minor trace elements, from the received coals, into the water supply (rainwater tank). The leaching test was based on a common soil extraction method (3A1 in Rayment and Higginson 1992); however, rainwater was used as the leaching solution. This process was designed to account for both the chemistry of rainwater and turbulence from the rooftop to the rainwater tank; and to create a known residence time of coal dust in solution.

Rainwater (at pH = 6.6 and initial temperature of 17.2 °C) was used in the leaching tests and was analysed as the sample blank during elemental analysis. Two grams of < 63 µm coal sample and 100 mL of rainwater (1 in 50 ratio; or 20 g of coal dust per litre) were shaken in a conical flask for 1 hour. The coal/rainwater solutions then stood for 24 hours in a non-climate controlled room, to mimic diurnal temperature variation, before filtration through 0.45 µm glass-fibre filters. The supernatant was analysed for major and minor trace elements in coal (i.e. arsenic (As), boron (B), cadmium (Cd), mercury (Hg),

Table 1. Trace elements in coal types exported from DBCT.

Element	Symbol	Concern	Units	BMA	Burton	Blair Atho	Hail Creek	German Creek
Sulfur	S	Major	%	0.56	0.4	0.3	0.35	0.65
Chlorine	Cl	Moderate	mg kg ⁻¹	500	500	200	100	620
Arsenic	As	Major	mg kg ⁻¹	0.4	0.9	2.6	0.4	1.2
Boron	B	Major	mg kg ⁻¹	8	10	20	3	12
Cadmium	Cd	Major	mg kg ⁻¹	0.05	0.28	0.1	0.09	0.1
Lead	Pb	Major	mg kg ⁻¹	6.99	8	7	7.2	3.8
Mercury	Hg	Major	mg kg ⁻¹	0.04	0.05	0.04	0.01	0.21
Molybdenum	Mo	Major	mg kg ⁻¹	ND	1	1.5	0.32	0.4
Selenium	Se	Major	mg kg ⁻¹	0.71	0.8	0.8	0.22	0.77
Chromium	Cr	Moderate	mg kg ⁻¹	4	18	6.8	10	3.3
Copper	Cu	Moderate	mg kg ⁻¹	17	23	8.9	11	16.3
Fluorine	F	Moderate	mg kg ⁻¹	82	146	50	145	120
Nickel	Ni	Moderate	mg kg ⁻¹	7	13	8	9	1.3
Vanadium	V	Moderate	mg kg ⁻¹	43	39	8	22	38
Zinc	Zn	Moderate	mg kg ⁻¹	15	36	44	6	13

Table 2. Trace elements in coal types at DBCT, a range of Australian coals, and a range of coal types from around the world (from CSIRO (2007) and data supplied by Introspec Consulting).

Element	Symbol	Concern	Units	DBCT average (all coals)	Range for Australian coals	Range in coal around the world
Sulfur	S	Major	%	0.55	0.1-0.9	ND
Chlorine	Cl	Moderate	mg kg ⁻¹	400	200-2500	100-3500
Arsenic	As	Major	mg kg ⁻¹	2.18	0.2-2.2	3-26
Boron	B	Major	mg kg ⁻¹	39	5-70	6-146
Cadmium	Cd	Major	mg kg ⁻¹	0.08	0.01-0.32	0.1-0.31
Lead	Pb	Major	mg kg ⁻¹	8.6	2-14	< 1-22
Mercury	Hg	Major	mg kg ⁻¹	0.07	0.1-0.11	0.1-0.19
Molybdenum	Mo	Major	mg kg ⁻¹	1.62	0.1-2.6	0.1-4
Selenium	Se	Major	mg kg ⁻¹	1.6	0.1-1	0.1-5.3
Chromium	Cr	Moderate	mg kg ⁻¹	17	2-25	1-35
Copper	Cu	Moderate	mg kg ⁻¹	10.5	6-27	< 1-28
Fluorine	F	Moderate	mg kg ⁻¹	100	20-225	< 20-340
Nickel	Ni	Moderate	mg kg ⁻¹	11.3	4-23	2-21
Vanadium	V	Moderate	mg kg ⁻¹	23	7-75	1-60
Zinc	Zn	Moderate	mg kg ⁻¹	12.5	3-26	1-55

molybdenum (Mo), lead (Pb), selenium (Se), chromium (Cr), copper (Cu), nickel (Ni), vanadium (V) and zinc (Zn).

Sulfur (S), chlorine (Cl) and fluorine (F) in coal are usually measured due to their harmful properties in air after combustion; however, these elements are relatively abundant in nature and are in relatively low concentrations in coal and rainwater. After the combustion of coal, S, Cl and F are released into the atmosphere and may contribute to 'acid' rain. In this study, the pH of the rainwater after the leach test was deemed a suitable indicator for assessing the potential harm to public health (from non-combusted coal dust) and therefore these elements were not directly measured in the rainwater samples.

Analysis techniques included inductively coupled plasma-optical emission spectrometry (ICP-OES), inductively coupled plasma-mass spectrometry (ICP-MS) and cold vapour-atomic absorption spectrometry (CV-AAS). A range of instruments were utilised to obtain element detection limits considerably less than threshold values in the Australian Drinking Water Guidelines (NHMRC 2004). Detection limits and threshold guideline values will be presented during results.

Water samples from homes in the area and dust deposition rates

Rainwater samples were obtained for analysis from three houses within the Dalrymple Bay area. Samples included reticulated town water, rainwater tank and the hot and cold taps from the kitchen. At one location, unfiltered hot and cold tap samples were also obtained. Water samples were analysed for all major and minor trace elements of concern in coal dust. All results were then compared with the Australian Drinking Water Guidelines (2004) to interpret the potential for causing public health issues.

Annual dust deposition rates ($\text{mg m}^{-2} \text{y}^{-1}$) ranged from 5–40 in residential areas and 40–130 at the DBCT (source). The USEPA's Integrated Source Complex (ISC3) dispersion model was used for the assessment of the DBCT expansion and has been validated against detailed monitoring data from previous studies (Katestone Environmental 2006). The ISC3 model incorporates (amongst other variables) the influence of dominant wind directions, threshold wind velocities, gravitational settling of different particle sizes from the atmosphere and Brownian motion dynamics of dust particles at the air/surface interface in order to determine dust deposition rates. Rainfall data was also incorporated into the simulations undertaken to predict dust deposition.

RESULTS

Trace elements in coal

Table 1 shows typical trace element analysis for coal types exported from the DBCT. These coal types represent approximately 75% of the coal types (thermal and coking) exported from the DBCT. All the coal types contain trace elements of both minor and major concern.

Table 2 shows average trace element concentrations in coal for the DBCT and the range for Australian coals and internationally traded coals. Comparison to coals exported from the DBCT (see Table 1) suggests that the DBCT coal types are relatively low in comparison to trace element concentrations on a world-wide basis.

Whilst the presence of these trace elements in coal indicates a potential for human health impacts, it does not necessarily mean that they will end up being released into rainwater, or indeed even the coal dust will reach the rainwater tank. Pathways and processes identifying the maximum likelihood for these trace elements to be released into rainwater in tank will be discussed later in this paper.

Leaching tests

Table 3 summarises results from leaching tests. The initial chemistry of the rainwater used in the leaching tests and the Australian Drinking Water Guidelines (ADWG) (NHMRC 2004) for each element is also shown.

No trace element was found to exceed ADWG (NHMRC 2004) and the pH of all filtered leaching solutions ranged from 7.3–7.6. It must be noted that the reporting limits were considerably lower than threshold concentrations for all elements (except Se) as stipulated in the Australian Drinking Water Guidelines (NHMRC 2004). The actual detection limits achieved by the respective analytical approach are also shown in Table 3.

The rainwater used in the leaching tests had initial trace concentrations (in $\mu\text{g L}^{-1}$) of V (130), Zn (144), Cd (0.5) and Pb (6). While all these results are at least an order of magnitude lower than the ADWG (NHMRC 2004), there were several decreases in concentration from the leaching solution after the tests. For example, the initial concentration of Zn ($144 \mu\text{g L}^{-1}$) decreased to $< 0.1 \mu\text{g L}^{-1}$ after the leaching tests for all coal types. Similar removal rates were observed for Cd (0.5 to < 0.1), Pb (6 to < 1) and V (130 to < 6) after the leaching tests. Molybdenum (Mo) and Arsenic (As) were the only trace elements that showed an increase in concentration from the initial rainwater concentrations in some coal types. Mo increased from $< 20 \mu\text{g L}^{-1}$ in rainwater to $30 \mu\text{g L}^{-1}$ for coal type GMKA and As increased slightly in coal types GMKA and OCJC.

Rainwater quality from sampled sites

Water samples were taken from rainwater tanks at three locations within the Dalrymple Bay area. Table 4 summarises results from analysis of these water samples.

No trace element was found to exceed ADWG (NHMRC 2004) in all samples. Trace amounts of Cu were detected at Site A (reticulated town supply), Site C cold tap (filtered), Site C cold tap (unfiltered), Site C hot tap (filtered) and Site C hot tap (unfiltered). Trace amounts of Zn were detected at the Site A tank and Site C cold tap (filtered). A trace concentration of Mo was observed at the Site C cold tap (unfiltered).

Total dust deposition and potential coal dust deposition

Annual dust deposition rates in the Dalrymple Bay area ranged from < 5 to $\sim 130 \text{ mg m}^{-2} \text{yr}^{-1}$ (Katestone Environmental 2006). The nearest residential areas receive a maximum of approximately $40 \text{ mg m}^{-2} \text{yr}^{-1}$. However, as previously mentioned, coal dust represents only a percentage of the total dust deposited. Coal dust (as a percentage of total dust loads) monitored in the Dalrymple Bay area ranged from approximately 5% to 80% (Katestone Environmental, 2006). Katestone Environmental (2006) details the methods and dust deposition results due to the forecast expansion of coal shipping capacity at the DBCT to 85 Mtpa. This scenario therefore represents the worse case scenario.

Dust deposition prediction incorporated rainfall regimes indicative of the area and wind roses reflecting dominant wind directions. Therefore, the influence of rainfall and wind has been accounted for in the resulting dust deposition rate. Based on a total dust deposition of $40 \text{ mg m}^{-2} \text{y}^{-1}$, Figure 1 shows the annual mass of coal dust deposited on a range of rooftop areas as a function of (total) dust comprising varying percentages of coal dust (5, 30, 50, 60 and 80%).

Figure 1 indicates that if 80% total dust was coal dust then approximately 9.5 g of coal dust per annum could potentially reach the rainwater tank. Assuming all coal dust reaches the tank, this means the maximum concentration of coal dust in a 10,000 L rainwater tank would be approximately 1 mg L^{-1} (at full capacity) and approximately 10 mg L^{-1} at minimum capacity (1,000 L). Note that the leaching tests were undertaken at 20 g L^{-1} coal dust to represent a worse case scenario. It is also important to note that none of the houses monitored in this study had a first-flush device to prevent dust reaching the rainwater tank. No investigation was made on the trace element contribution of non-coal dust in this study, for example, the percentage of dust that may comprise other natural and/or anthropogenic particulates.

In contrast, if coal dust only represented 5% of total dust then approximately 2 g y^{-1} could potentially reach the rainwater tank. Assuming all coal dust reaches the tank, this means the maximum concentration of coal dust in a 10,000 L rainwater tank would be approximately 0.2 mg L^{-1} (at full capacity) and approximately 2 mg L^{-1} at minimum capacity (1,000 L).

DISCUSSION

Trace elements in coal dust and results from leaching tests

Data from the leaching tests (Table 3) indicate that negligible amounts of trace elements in coal dust were released into the rainwater. All trace element concentrations were below the ADWG (NHMRC 2004). The data also suggest that Zinc (Zn) initially contained in the rainwater used for the leaching tests, was actually removed from

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Table 3. Leaching test results ($\mu\text{g L}^{-1}$).

Analysis technique	ICP-OES	ICP-OES	ICP-OES	ICP-OES	ICP-OES	ICP-OES	ICP-OES	ICP-MS	ICP-MS	CV AAS	ICP-MS	ICP-MS
Coal Type	B	Mo	Cr	Cu	Ni	V	Zn	As	Cd	Hg	Pb	Se
BACS	*	*	*	*	*	*	*	*	*	*	*	*
Foxleigh PCI	*	*	*	*	*	*	*	*	*	*	1	*
GMKA	*	30	*	*	*	*	*	2	*	*	*	*
GMKB	*	*	*	*	*	*	*	*	*	*	*	*
HCKB	*	*	*	*	*	*	*	*	*	*	*	*
HCKC	*	*	*	*	*	*	*	*	*	*	2	*
MBNX	*	*	*	*	*	*	*	*	*	*	*	*
MNBC	*	*	*	*	*	*	*	*	*	*	1	*
OCC	*	*	*	*	*	30	*	*	*	*	4	*
OCJC	*	*	*	*	*	*	*	1	*	*	*	*
OCJO	*	*	*	*	*	*	*	*	*	*	*	*
RSDB	*	*	*	*	*	*	*	*	*	*	2	*
RSDE	*	*	*	*	*	*	*	*	*	*	*	*
RSDL	*	*	*	*	*	*	*	*	*	*	*	*
RSDS	*	*	*	*	*	*	*	*	*	*	*	*
Detection limit	20	20	8	2	7	6	0.1	1	0.1	0.1	1	10
ADWG 2004	4000	50	50	2000	20	50	3000	7	2	1	10	10
¹ Rainwater	< 20	20	< 8	< 2	< 7	130	144	< 1	0.5	< 0.1	6	< 10

¹(leaching solution) pH = 6.6, initial temperature = 17.2 °C
 *(equal to or less than detection limit)

Table 4. Water analysis results ($\mu\text{g L}^{-1}$).

Rainwater site	B	Mo	Cr	Cu	Ni	V	Zn	As	Cd	Hg	Pb	Se
Site A (reticulated town supply)	*	*	*	110	*	*	*	*	*	*	*	*
Site A (tank) ¹	*	*	*	*	*	*	240	*	*	*	*	*
Site B (cold tap) ¹	*	*	*	*	*	*	*	*	*	*	*	*
Site C (cold tap - filtered) ¹	*	*	*	280	*	*	*	*	*	*	*	*
Site C (hot tap - filtered) ¹	*	*	*	120	*	*	*	*	*	*	*	*
Site C (cold tap - unfiltered) ¹	*	30	*	780	*	*	120	*	*	*	*	*
Site C (hot tap - unfiltered) ¹	*	*	*	100	*	*	*	*	*	*	*	*
ADWG 2004	4000	50	50	2000	20	50	3000	7	2	1	10	10*

¹ indicates samples sourced from rainwater
 *(equal to or less than detection limit in Table 3)

solution by the coal dust present ($144 \mu\text{g L}^{-1}$ decreased to $< 0.1 \mu\text{g L}^{-1}$). Compared to initial rainwater concentrations used in the leaching tests, similar trends in removal of Pb, V and Cd were also observed.

The pH and temperature of rainwater in the tank is likely to influence leaching processes and conditions within the tank. This study used rainwater at only one pH value and further studies should involve repeating these experiments with rainwater of varying pH. The pH of rainwater, roof runoff and rainwater in the tank can range from 4.5–7.5 (Coombes 2002), and since pH governs the solubility of many elements

in water, may provide different results than those presented in this study. However, at pH = 6.6 it appears that Zn, Pb, V and Cd were adsorbed to the coal dust during the leaching test and removed from the rainwater. The influence of temperature is not discussed in detail because the rainwater used in the leaching test and the rainwater samples obtained from houses in the area reflected real-world conditions at the time of sampling.

Water samples from homes in the area

Results from rainwater and reticulated town water samples from homes in the Dalrymple

Bay area were below threshold guideline values (NHMRC 2004) (refer Table 4). The absence of a first-flush device at any of the homes suggests that most dust deposited on the rooftops would eventually enter the rainwater tank.

Trace concentrations of copper (Cu) were found in the reticulated town water supply ($110 \mu\text{g L}^{-1}$) at Site A and in the rainwater samples at Site B from the hot and cold taps (filtered and unfiltered). For Site B samples it is interesting to note that the cold tap filtered water sample ($280 \mu\text{g L}^{-1}$) had considerably less Cu than the unfiltered water sample from the cold tap ($780 \mu\text{g L}^{-1}$).

This outcome suggests that there was net removal of Cu from the filtration process. The filtered and unfiltered hot tap water samples (120 and 100 $\mu\text{g L}^{-1}$ respectively) were much less than the filtered cold tap water sample (280 $\mu\text{g L}^{-1}$). This outcome suggests there was net removal of Cu from the hot water system. Trace concentrations of Cu in the reticulated town water, rainwater in the tank and at the tap (Sites A and C) suggest that the source of Cu may be due to plumbing materials such as copper pipes and fittings.

Trace concentrations of Zn were also found at the Site B cold taps (unfiltered) (120 $\mu\text{g L}^{-1}$). The hot water tap samples (filtered and unfiltered) and filtered cold water samples all contained < 100 $\mu\text{g L}^{-1}$ Zn. Again, both filtration and/or the hot water system are implicated in the improved water quality.

Did leaching experiments and rainwater analysis suggest that coal dust in rainwater is a human health risk? A slight increase in Mo was observed at the Site C cold tap (unfiltered) which appears consistent with the slight increase in leached Mo during the leaching tests. However, further discussion using first principles is required to define the maximum likelihood of coal dust to be deposited on rooftops; and to discuss the rainwater tank processes that influence the ability of trace elements to be released into stored rainwater.

Defining the maximum likelihood of coal dust deposition on rooftops

The potential for trace elements in coal dust to be released into harvested rainwater requires an understanding of potential pathways from the source and the likelihood of trace elements to be released into rainwater from coal dust deposited on the roof. Figure 2 summarises the major steps required for trace elements in coal dust to be released into roof-harvested rainwater and potentially impact on human health.

Particle size is an important factor in determining the way in which the particles move through the air. Different dust sources create varying dust particle sizes. For example, motor vehicle emissions are typically 2.5 μm and wind blown particles can be up to 100 μm (average of 30 μm) (Neale and Wainwright 2007). Coal dust particles were assumed to fall within this range of particle sizes (2.5–100 μm).

The entrainment of dust particles into the atmosphere and the subsequent deposition onto a rooftop at a distance from the dust source requires threshold wind velocities and suitable wind directions. Therefore, only certain size particles have the potential to be transported to roof height and be deposited. Wind speed and direction are important variables governing dust deposition and are therefore always incorporated into available models that simulate dust deposition rates. The dust deposition rates are usually validated based on results from dust deposition gauges and high volume air samplers situated at various distances from a dust source at a given site.

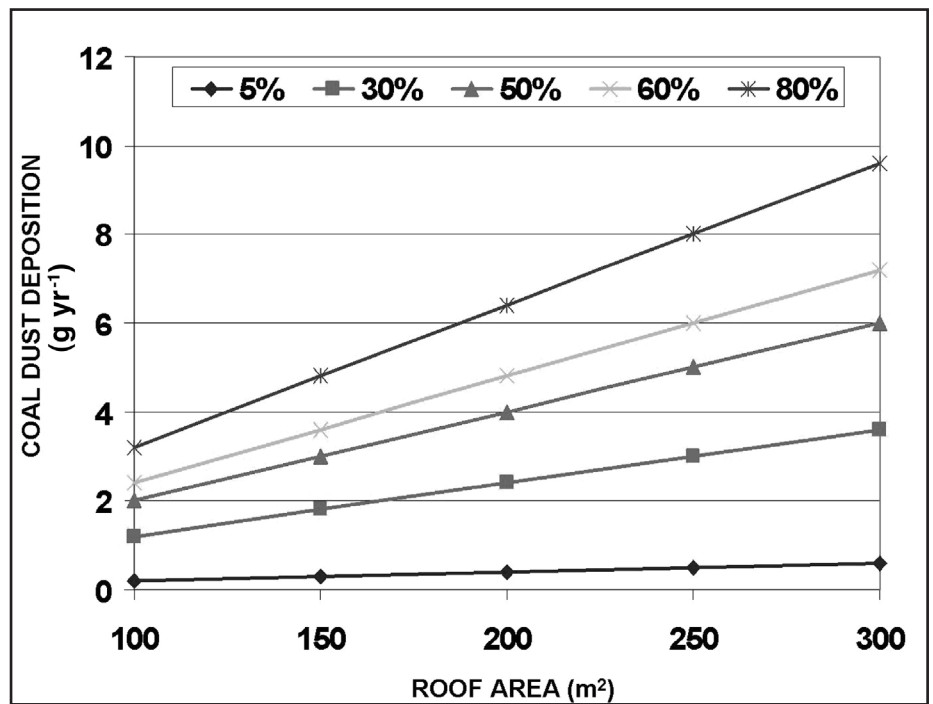


Figure 1. Coal dust deposition to residential rooftops in the Dalrymple Bay area based on 40 $\text{mg m}^{-2} \text{yr}^{-1}$ (total dust). Percentages indicate the proportion of coal dust to total dust.

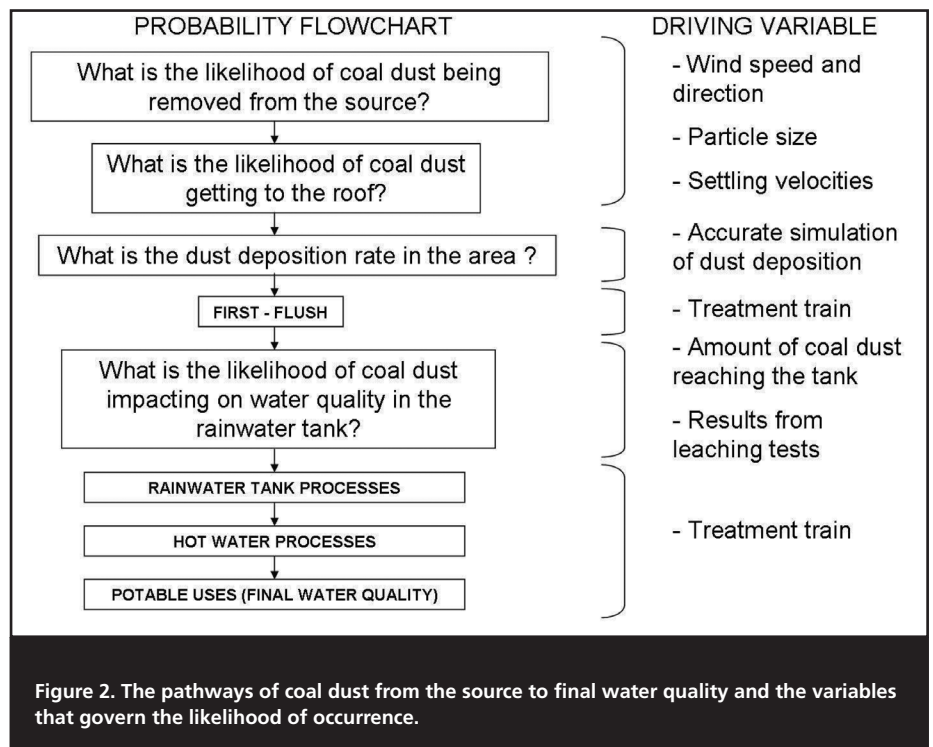


Figure 2. The pathways of coal dust from the source to final water quality and the variables that govern the likelihood of occurrence.

The result is a map of the area highlighting dust deposition rates (as isohyets) at various distances away from the source(s).

The rate of coal dust deposition to the rooftop is most likely governed by wind speed and direction on dry days, as wet dust requires greater forces to be entrained into the atmosphere. The influence of wind direction on microbial counts/species presence in roof-harvested rainwater has been confirmed by Evans *et al.* (2005). Their study directly suggests that wind direction

would also influence dust-particle chemistry. For example, dust-particle chemistry coming from one wind direction (and source) is likely to have a different chemistry to dust particles originating from a different direction (and source). This too was evident in Evans *et al.* (2006) with respect to sodium, calcium, chlorides, sulfates and nitrates, where concentrations in roof-harvested rainwater differed depending on wind direction. The review by Neale and Wainwright (2007) also highlights the difference in dust deposition

rates due to wind direction and therefore is likely to support this hypothesis, particularly when dominant dust sources are known.

Coal dust is likely to be very dry when airborne and when deposited on the roof (as opposed to bird droppings) and therefore is likely to be readily removed by wind from the roof after initially being deposited. For example, dust deposited by winds from one direction may be removed by winds from another direction. However, coal dust is also likely to carry a mild charge and may remain adsorbed to the roof in some instances (depending on climatic conditions and roof material), and be washed into the rainwater tank during rainfall events. The amount of deposited dust is likely to increase with increasing consecutive non-rain days coupled with increasing winds from the main source direction (such as coal stockpile and loading facilities).

Both coal and non-coal dusts are deposited onto rooftops and coal dust is typically only a percentage of the total dust deposited. For example, studies in the Dalrymple Bay area indicate that coal dust comprises approximately 5–80 %, dependent on location and distance from the source (data based on Katestone Environmental 2006). As a result, the deposition of actual coal dust to a rooftop only represents a percentage of the total dust deposition load. Therefore, depending on location and distance from the source, only a percentage of total dust (as coal dust) would be expected to reach a given rooftop.

After rain, any deposited coal dust (and non-coal dust) would most likely be transported to the rainwater tank via guttering and the first-flush device. A first-flush device is a small storage (typically 20 L) that separates the first flows from the roof from rainwater entering the tank. The first-flush device is likely to remove most

of these dust particles that were deposited on the roof (Coombes 2002). In general, dust particles are likely to be washed from the roof during the first-flush and dust concentrations are most likely to decrease throughout a prolonged rain event. In contrast, bird droppings will stick to the roof surface and are likely to release nutrients over successive rain events. However, no first-flush devices were fitted on any of the households in this study.

To become a human health issue the trace elements must be leached from the coal dust into the rainwater while in the tank. No study has directly investigated this phenomena and this research aimed to determine the potential for trace elements in coal dust to impact upon human health, and as a result, improve our understanding of the processes potentially contributing to water quality in the rainwater tank. The leaching tests indicated a low potential for transfer of trace elements in coal dust to the rainwater in the tank, yet other processes contribute to improving the water quality of harvested rainfall.

Rainwater tank processes

Recent studies have highlighted the treatment-train from the rooftop to the tap, including the first-flush device to the rainwater tank, rainwater tank processes that treat rainwater and the influence of the hot water system (Spinks *et al.* 2003, 2004; Coombes *et al.* 2003, 2004). Figure 3 broadly describes the dominant processes occurring in the rainwater tank.

A myriad of interacting chemical, physical and microbial processes operate within the tank that aid in the removal of nutrients (e.g., suspended solids, phosphorus and nitrogen) and heavy metals (e.g., mercury, lead and cadmium). One of the dominant processes in the rainwater

treatment train appears to be flocculation of organic, metallic and chemical parameters at the tank water surface with subsequent settlement of flocs to the bottom of the tank or attachment to walls in the tank (Spinks *et al.* 2003). Ongoing analysis, resulting from this initial observation, has revealed that biofilms do exist in rainwater tanks and that a core group of environmental bacteria such as *Bacillus Spp.* are likely to form biofilms in rainwater tanks (Spinks *et al.* 2003). These bacteria have the potential to remove nutrients, metals and microbes.

Monitoring of the demonstration projects (in Spinks *et al.* 2003) also led to a discovery that domestic hot water services set at temperatures greater than 52°C consistently eliminated bacteria from rainwater (Spinks *et al.* 2004). This discovery led to laboratory experiments into the impact of hot water on the viability of selected pathogens. Potentially pathogenic bacteria were observed to be rapidly eliminated from rainwater at temperatures of 60°C or greater.

PERSPECTIVES AND RECOMMENDATIONS

The use of harvested rainwater may be the only source of water in many regional areas and it is important to identify water quality issues and effectively manage any risks. This study indicates a low risk of coal dust being a source of leached trace elements into rainwater in the tank. A recent study of raintank water in the Brisbane area (Queensland, Australia) indicated a relatively poor correlation between dust deposition (bulk deposition) and tank water concentrations with respect to Lead (Pb) (Huston *et al.* 2008). This indicated that Pb predominantly originated from other sources. Lead flashing was suggested as one possible source of Pb in rainwater in the tanks (Huston *et al.* 2008).

Interestingly, health authorities in New Zealand deem that all reticulated town water has the potential to leach metals from the reticulated town water pipe network and household plumbing (plumbo-solvency) (New Zealand Ministry of Health 2005). No such consideration is made in Australia, where traditionally, poor analyses of rainwater harvesting options coupled with rainwater quality studies that neglect sampling/analysis of reticulated town water at similar sites, frequently act to promote water authority preference for large-scale water management options such as dams, desalination plants and wastewater reuse.

One exception is the study by Morrow *et al.* (2007). Lead (Pb), zinc (Zn) and cadmium (Cd) were detected in harvested rainwater in tanks and also in the reticulated town water supplies in this Australian study. A small number of mains water samples exceeded ADWG. However, detailed investigation of the source of these contaminants remains relatively overlooked. With respect to identifying sources of elemental contamination, Morrow *et al.* (2007) investigated urban versus rural different scenarios as well as the influence of different

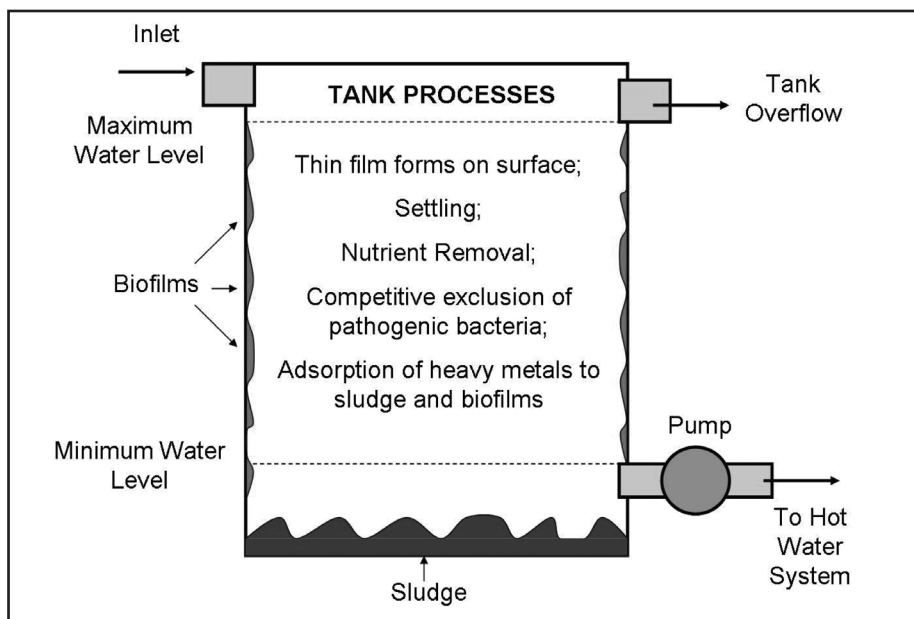


Figure 3. Rainwater tank processes (based on Spinks *et al.* 2003).

Table 5. Ranges and/or means of Zn, Pb and Cd from international rainwater studies, in $\mu\text{g L}^{-1}$, (based on Table 2 in Roy and Nègrel, 2001).

Study Area	Zn	Pb	Cd	Reference
Ontario, Canada	-	23	0.63	Sanderson and Marchand (1988)
Mediterranean basin	-	5-8.6	-	Migon <i>et al.</i> (1993)
SE of USA	1.2-13	3.2-8.4	0.28-0.55	Lindberg (1982), Lindberg (1989)
Rhode Island and urban area, USA	-	4.5	0.05	Heaton <i>et al.</i> (1990)
NW of the United Kingdom	-	14-60	5-18	Heaton <i>et al.</i> (1990)
Paris, France	23.4-235	10-30	0.17-0.65	Roy (1996)
Massif Central, France	14.3	50.2	0.44	Roy and Nègrel (2001)
Australia (rainwater in tank) (n=87)	1017	3.45	0.17	Morrow <i>et al.</i> (2007)
Australia (reticulated town supply) (n=14)	78	1.92	0.09	Morrow <i>et al.</i> (2007)

roof and tank materials. So what is in rainwater before it reaches the roof? Table 5 summarises Zn, Pb and Cd concentrations in rainwater from international rainwater studies. Results from an Australian study comparing water quality from reticulated town water and roof harvested rainwater in tanks are also shown (from Morrow *et al.* 2007).

The studies by Heaton *et al.* (1990; 1992) and Roy and Nègrel (2001) indicate relatively high concentrations of Pb in rainwater; which would exceed water quality guidelines (10 to 60 times greater) if these levels occurred in Australia. The study by Roy and Nègrel (2001) partitioned Pb sources as being from aerosol emissions from vehicles, agriculture, the burning of vegetation and fossil fuels, and the use of fertilisers. Cd concentrations determined by Heaton *et al.* (1992) indicate relatively higher concentrations of Cd in rainwater that would exceed Australian guidelines by 5 to 18 times. Therefore it is apparent that background levels of Pb and Cd in rainwater in some urban/industrialised areas have the potential to be harmful to human health. The composition of rainfall is predominantly influenced by anthropogenic sources in the northern hemisphere (Roy and Nègrel 2001).

In Australia, zinc (Zn) threshold concentrations in drinking water guidelines are set for taste only (NHRMC 2004). The relatively high mean in Morrow *et al.* (2007) suggests Zn originated from sources other than rainfall. Pb was detected in rainwater in tanks and the reticulated town water, which suggests that Pb contamination is likely from a similar source(s).

This study has demonstrated the low potential for trace elements in coal dust to be leached into the rainwater in the tank. Furthermore, Struck *et al.* (1996) concluded that Pb and Cd concentrations in rainwater were lower than their respective solubilities and indicated that soluble Pb and Cd strongly adsorb to particulate matter. This supports the leaching test results in this study, as Pb and Cd concentrations in the rainwater were removed by the coal dust present. Therefore, basic information on background trace element concentrations in reticulated town water, rainwater and

materials contained in the roof/gutter/ rainwater tank/household plumbing (at-a-site) is required before any interpretation on harvested rainwater quality is reported.

Furthermore, the rainwater treatment-train previously discussed will also play a major role in final water quality before use. Very few studies consider many of the pathways, processes and potential trace element sources in interpreting water quality in rainwater tanks. This oversight often results in alarmist reports in the media based on fragmented knowledge.

CONCLUSION

Leaching tests (at 1 part coal dust to 50 parts rainwater at pH = 6.6) resulted in negligible release of trace elements into rainwater. Molybdenum (Mo) and Arsenic (As) were the only trace elements that showed a potential to be leached from coal dust. However Mo and As concentrations in sampled rainwater, tank and tap samples were all below ADWG (NHMRC 2004) and indicated a minimal likelihood of coal dust being an issue with respect to human health. The 1:50 ratio (20 g of coal dust L^{-1}) used in the leaching tests exceeded the potential maximum coal dust/rainwater mixture in the tank. Therefore the test condition exceeded the worse case scenario with respect to coal dust in a rainwater tank in the Dalrymple Bay area.

Trace elements in reticulated town water, rainwater from tanks and kitchen taps in the study area were all less than the ADWG (NHMRC 2004). Results from samples taken after both filtration and the hot water system indicated removal of Cu from the rainwater supply. The quality of rainwater is ensured by a natural treatment-train in the tank that reduces the presence of bacterial and metal contaminants. Bacteria, organics and chemicals form flocculated particles that become biofilms on surfaces or settle to the bottom of the tanks to the sludge. Leaching tests of 100% coal dust indicated a net removal of most elements and suggests that coal dust in the rainwater tank is unlikely to contribute significantly to elemental loads in the rainwater column within the tank. The processes of flocculation, settlement and biofilms in tanks act to improve the quality

of rainwater (Coombes *et al.* 2003, 2004; Spinks *et al.* 2003, 2004).

The pathways of coal dust from the source to final water quality at the tap (Figure 2) indicate that many driving variables must coincidentally occur before any potential risk is realised. Considering that (1) certain sized coal dust particles can only be transported during favourable climatic conditions, (2) the leaching tests indicate negligible transfer of trace elements from coal dust to rainwater, (3) the discussed rainwater quality treatment train, and (4) the continual drawdown and replenishment of rainwater in tanks; the likelihood of trace elements significantly impacting on human health due to coal dust in rainwater tanks is low. Using the data obtained from the leaching tests, an argument could be made that coal dust would in fact remove Zn, Pb, V and Cd (in trace concentrations) from rainwater in the tank. This outcome further highlights the treatment train processes that exist in the rainwater tank. These results should allay major concerns with respect to the potential impact of coal dust on human health through drinking roof harvested rainwater. Coal dust may contain organic compounds that may be of concern in drinking water but analysis of these was outside the scope of this study. Based on the leaching method discussed in this paper, future experiments and analysis should investigate the likelihood of organic compounds in coal dust impacting on human health.

The potential human health implications of trace elements in coal dust in rainwater (at pH = 6.6) appear negligible. However, the variability in rainwater pH was outside the scope of this initial study and is considered an important variable in providing conditions that may enhance or reduce the release of trace elements in coal dust into rainwater in the tank. Furthermore, the variability due to different contact times, *i.e.* coal dust in contact with rainwater for extended periods, was also outside the scope of this study. Both of these considerations warrant further research.

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