The chemical water quality in roof-harvested water cisterns in Bermuda

A. J. Peters, K. L. Weidner and C. L. Howley

ABSTRACT

The chemical quality of water stored in roof-harvested rainwater cisterns and used for domestic consumption in Bermuda was assessed by analyzing a suite of metal and metalloid elements, major ions, pH and conductivity in water and sediment from 112 and 36 private residences, respectively. The chemical quality of water was good, with a low incidence of health-based primary drinking water standards being exceeded: < 1% for Pb and Se and < 3% for nitrate. In nearly all cases these could be accounted for by unapproved use of unregistered groundwater wells to supplement the water supply. Sediment which accumulates in the water cisterns was found to have elevated concentrations of some elements, similar to local soil. In particular, the concentration of arsenic (As) in cistern sediment exceeded the U.S. EPA generic soil screening limits (SSL) for human ingestion of soil at all 36 locations sampled. Additionally, Pb and Hg exceeded the SSL at 4 (11%) and 1 (3%) sites, respectively. However, there are no standards or guidelines in existence with which to fully assess the sediment data. Further research needs are identified which will allow a human health-based risk assessment to be conducted to enable such standards to be developed.

Key words | arsenic, metals, roof-harvested rainwater, sediment, water quality

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INTRODUCTION

Rainwater harvesting as a source of potable water is common in many parts of the world, and likely has been since ancient times (UNEP 1998*a*). It is particularly suitable to locations where the average rainfall exceeds 400 mm/year and other sources of water are scarce and/or of poor quality (Lye 2002). This source of water is currently widely utilized in both developing and developed nations. Rainwater harvesting is common in many countries throughout Asia and Africa (UNEP 1998*a*,*b*). In the Caribbean region, some 500,000 people depend at least in part on rainwater harvesting for their domestic water supply (UNEP 1998*c*). It has been estimated that approximately 40% of households in South Australia use rainwater harvesting as their principal supply of drinking water (Heyworth *et al.* 2006) and rainwater is also an important source of drinking water in some regions of New Zealand (Simmons *et al.* 2001) and the USA (Olem & Berthouex 1989).

Bermuda is a small (54 km^2), highly developed and densely populated sub-tropical island located in a remote position in the North Atlantic ocean at 32°N, 64°W, approximately 900 km from the coast of North America. The population density of the island is 1,155 km^{-2} (Government of Bermuda 2005), comparable to that of some large cities in North America, such as Houston, Texas, USA (US Census Bureau 2006), and the residents of Bermuda enjoy a high standard of living, with an average GDP of $53,414 per capita in 2003 (Government of Bermuda 2005).

The residents of Bermuda have always used rainwater as the principal source of potable water, owing to limited groundwater resources and the absence of rivers and lakes on the island. The average rainfall is 1,410 mm/yr and this is

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distributed evenly throughout the year: there is no "dry" or "wet" season (Government of Bermuda 2006). Under The Public Health (Water Storage) Regulations, 1951, all houses in Bermuda must have at least 80% of their roof area guttered to collect rainwater and be provided with a storage cistern with a capacity of at least 100 Imperial gallons (455 litres) for every 10 square feet (1 m^2) of collection area. The storage cisterns are usually cement-lined cisterns which are located under each house, having been excavated from the limestone bedrock during initial house construction. Average per capita water consumption is about 140 litres per day and the mean total consumption in homes and hotels (other commercial, industrial and agricultural water use is not included in this figure) is approximately 11×10^6 litres per day (Government of Bermuda 2006). Roof-harvested rainwater supplies around 9.6×10^6 litres per day and an estimated 2.6×10^6 litres per day is supplied by other roof and constructed water catchments (Government of Bermuda 2006). Some 2,200 households are connected to a mains water distribution system operated by the government. For all other residents, water supplies are supplemented when needed by commercial deliveries of water acquired from government-controlled groundwater lenses and water treatment plants. Private groundwater wells (which must be registered with the Ministry of the Environment) can only be used for toilet flushing and other non-potable water uses: it is illegal to use them for augmenting potable water supplies.

Owing to the heavy reliance on this source of water, the quality of roof-harvested rainwater is of concern to the residents of Bermuda. Contaminants can be deposited on roof surfaces, either directly incorporated in precipitation (wet deposition) or via fallout of particulate matter (dry deposition). In the absence of control measures (e.g. in situ filters) contaminants in wet deposition will immediately enter the water cistern whereas contaminants subject to dry deposition may accumulate on roof surfaces between precipitation events and then be flushed into the system during the next rain event. In some cases, the roof construction materials themselves can lead to contamination of the water (Forster 1999; Chang *et al.* 2004). This is not a concern in Bermuda where only non-toxic paint and sealant products which are approved by the Department of Health are allowed to be applied to roof surfaces. Approved paints are primarily acrylic and/or latex elastomer based products.

As stated above, particulate matter enters drinking water cisterns via its incorporation in rainwater and/or entrainment of dry deposition by rainwater. Large particles will immediately settle out and finer particles will remain as suspended sediment for some period of time. Over time, a layer of sediment accumulates on the bottom of the cistern. Potential sources of sediment in water cisterns in Bermuda include: soil; soot; dust; pollen; plant material (seeds, leaves and branches); fecal matter from wildlife; and roofing material (eroded cement and paint). The accumulated sediment may act as either a sink for contaminants which may accumulate in the sediment by adsorption from the overlying water or be a source of contaminants as they are released to the water by desorption/dissolution. Once in the cistern, particles and any associated contaminants may then enter directly into the household water supply system in suspension, and particularly after agitation by inflowing rainwater or truck delivered water.

There is thus a potential for roof-harvested drinking water quality to be compromised and to present a human health risk. However, there are relatively few reports in the published literature concerning water quality derived from this source. Of those studies which have been published in mainstream scientific journals, the majority focus has been on microbiological quality. Lye (2002) reviewed a number of cases where adverse health effects have been attributed to the microbiological quality of roof-harvested rainwater used for domestic purposes and Meera & Mansoor Ahammed (2006) have reviewed a number of studies of the microbiological and physico-chemical quality of roof-harvested rainwater and roof runoff. Other studies include: the incidence of *Legionella* species in water cisterns in the U.S. Virgin Islands (Broadhead *et al.* 1988); the incidence of gastroenteritis in young children after consuming untreated cistern rainwater in Australia (Heyworth *et al.* 2006); and chemical contamination of cistern drinking water supplies in the southern USA (Olem & Berthouex 1989).

In Bermuda, the Department of Health is responsible for testing the microbiological quality of cistern water in Bermuda. In 2002 they tested a total of 483 cisterns, of which only 126 (26%) were found to be bacteriologically acceptable (Government of Bermuda 2006). The biological

quality of Bermuda cistern water was further studied by the Atlantis Project's mobile laboratory in 2003 (Atlantis Project 2004). The results from 102 residences showed that 90% of the cisterns analysed were contaminated with total coliforms and approximately 66% of samples showed contamination with *E.coli*. These results suggest that microbiological contamination of cistern water is a frequent occurrence in Bermuda.

However, less is known regarding chemical contamination of roof-harvested water in Bermuda. There are no heavy industrial activities located on the island but other sources of potential water cistern contamination exist, including a municipal solid waste incinerator and an oil-powered electricity generation plant which service the island's population of c. 62,000 (Government of Bermuda 2005), as well as emissions from a large fleet of road vehicles. Other local sources of potential water cistern contaminants include emissions from a small medical waste incinerator and exhaust emissions from shipping and commercial aircraft. In addition, Bermuda is subject to long-range trans-boundary atmospheric pollution (Huang *et al.* 1996).

The aim of this study was to establish the chemical quality of roof-harvested rainwater used for domestic purposes in Bermuda. To achieve this, a suite of chemical parameters was analysed in water and sediment samples from roof-harvested water cisterns.

METHODS

Sampling

Water samples were collected during the summer of 2005 from 112 private households across the island (see Figure 1). The effect on water quality of the physical parameters of rooftop areas (e.g. age, condition, size, dimensions, slope, aspect and external factors such as closeness to adjacent buildings and vegetation) was outside the scope of this study. As far as could be determined during site visits, all of the houses at which water cisterns were sampled had the same style of traditional Bermudian roof, consisting of Bermuda limestone roof slates fixed on a wooden framework. The tiles are coated with a cement-based wash prior to sealing and/or painting with government-approved products (see above). In place of limestone slates, some newer buildings employ different materials for roof slates, such as polymer- or glass-modified cement, but these are coated, sealed and painted with the same types of products. If present, these alternative materials can therefore be assumed to have an identical effect (if any) on roof-harvested water quality.

Water was sampled directly from the water cisterns, prior to any point of contact with plumbing, pumps or filters. The results thus reflect the quality of the water entering cisterns directly from the surface of roofs and not necessarily the quality of water obtained from faucets inside the residence. This provides a better estimation of island-wide water quality as individual residences may vary significantly in terms of the age and quality of plumbing and in the type, if any, of water treatment techniques in place. Residences were excluded from the survey in cases where they had received supplemental water deliveries or had the cistern cleaned within 6 months prior to sampling.

Samples were collected by immersing sample containers into the water cisterns using a grab pole. This enabled the container to be fully submerged below the surface of the water, taking care not to disturb the underlying sediment. All water samples were collected before any sediment sampling was undertaken. At each site, one sample was collected for metal and major cation analysis in a pre-cleaned 250 ml high-density polycarbonate bottle and one sample was collected in a pre-cleaned 250 ml high-density polyethylene bottle for major anion analysis and pH and conductivity determination.

Cistern sediment was sampled at 36 of the locations used for water sampling. The depth and consistency of the sediment was highly variable between sites. In general, sediment was present as a thin (c. 3 cm maximum) and poorly consolidated layer of very fine material with variable amounts of sand and other material (e.g. gravel, cement, paint flakes) present. Typical sediment sampling methods were inappropriate: the shallow depth of sediment and poor consolidation excluded the use of any type of corer and the depth of overlying water and poor consolidation of material excluded the use of manual sampling with hand tools. Sampling was therefore undertaken using an adaptation of the U.S. Geological Survey plate-filter procedure (Wilde

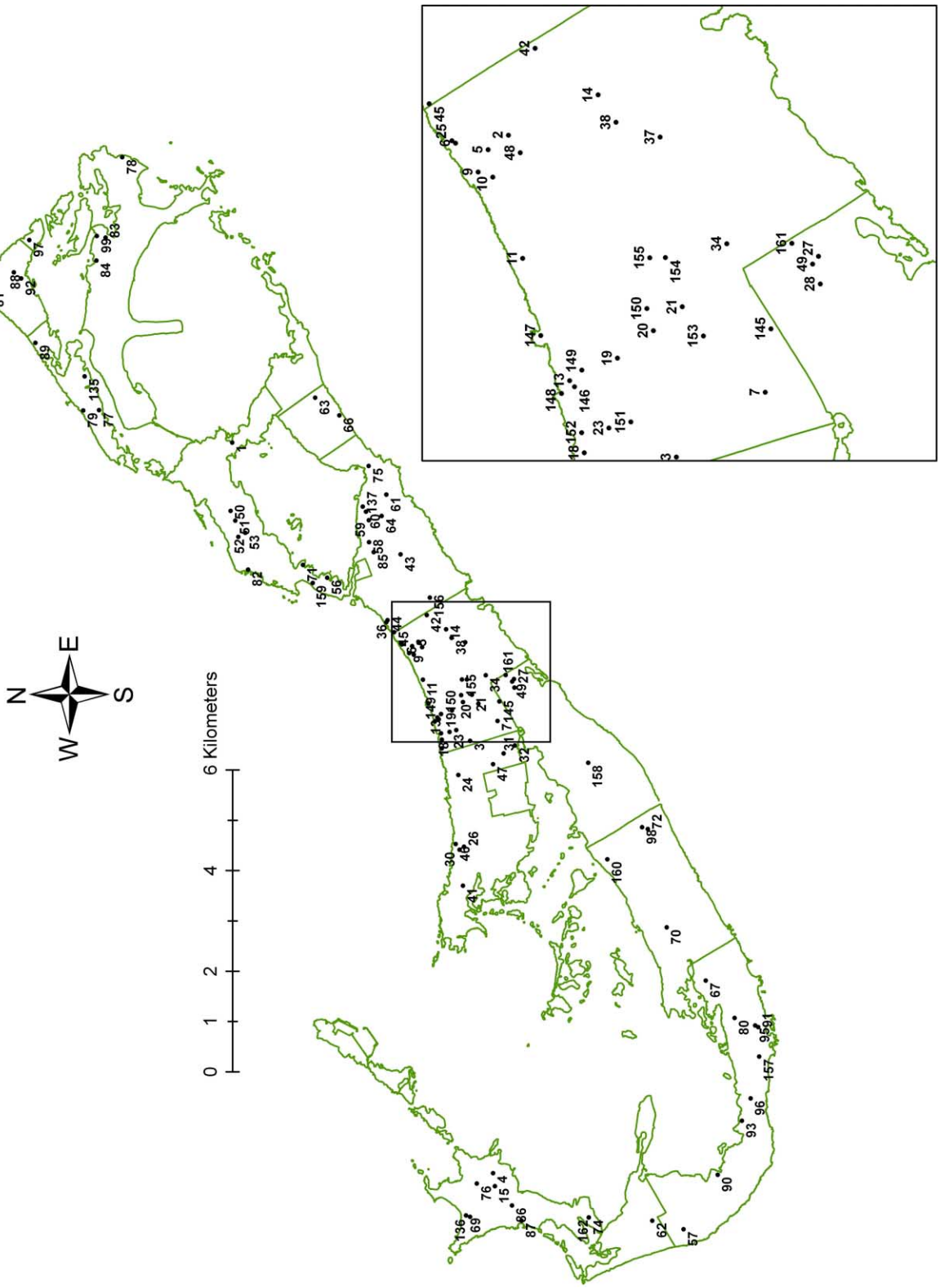

Figure 1 | Map of cistern water sampling locations in Bermuda.

Table 1 | Analytical methods and techniques for water samples

Medium	Parameter	Method	Technique
Water	Major cations, Fe, Mn	APHA* 3120 B	ICP-OES†
Water	Other elements	APHA 3125 B	ICP-MS‡
Water	Major anions	BIOS in-house	Dionex DX-100 ion chromatograph
Water	pH	BIOS in-house	Corning model 345 pH meter
Water	Conductivity	BIOS in-house	Orion model 160 conductivity meter
Sediment	Sb, As, Se	APHA 3114 C	AAS§ hydride generation
Sediment	Hg	APHA 3112 B	AAS cold vapour
Sediment	Other elements	EPA SW 846¶ Method 6010	ICP-OES
Sediment	Loss-on-ignition (LOI)	Loss on combustion at 420°C	Gravimetric

*APHA - American Public Health Association.
†ICP-OES - inductively coupled plasma optical emission spectroscopy.
‡ICP-MS - inductively coupled plasma mass spectrometry.
§AAS - atomic absorption spectroscopy.
¶EPA SW 846 - US EPA Test Methods for Evaluating Solid Waste, Physical/Chemical Methods.

et al. 2004). Cistern water was pumped through a 6.35mm diameter PTFE tube, using an in-line ceramic-piston metering pump (Fluid Metering, Inc., Model Q) powered by a 12 V automotive battery. The flow then continued through PTFE tubing and into a 142 mm diameter polycarbonate in-line filter holder (Geotech Environmental Equipment, Inc.) housing a pre-weighed 142 mm diameter glass fiber filter (Whatman GF/F).

The sample inlet line was attached to a sample grab pole and immersed in the water. Water was pumped through the tubing and pump for *c.* 2 minutes to flush the tubing and wet the filter and then the end of the inlet tube was dragged across the bottom of the water cistern to entrain bottom sediment in the inflowing water. The flow rate was in the range of 25–50 ml/min. Sampling was undertaken over a period time judged to be sufficient based on visual inspection of the cistern bottom, the loading in the inflowing water observed through the sample inlet line and by direct observation of the filter in the transparent filter holder. In general, sampling for *c.* 5 minutes provided sufficient loading of the filter to meet the analytical requirements (0.5 g dry weight minimum). On return to the laboratory, the filters were removed from the filter holder using pre-cleaned HDPE forceps, folded and placed in a labeled self-sealing polyethylene bag. The sediment sampling equipment (tubing, forceps and filter holder) was washed with purified deionised water (Milli-Q water) and cleaned by soaking in 5% HCl overnight.

After sampling, all water and sediment samples were stored in coolers containing freezer packs for transportation from the sampling locations to the laboratory where they were stored in the dark at 4°C. Samples were then dispatched to the analytical laboratory by express courier in cool boxes with freezer packs to maintain refrigerated conditions.

Analysis

With the exception of major anion (Cl^-, SO_4^{2-} and NO_3^-), pH and conductivity determinations which were undertaken at BIOS, all other water analyses and all sediment analyses were performed by EnviroTest Laboratories, in Edmonton, Canada. The laboratory is an ISO/IEC 17025 accredited laboratory. Table 1 details the parameters and methods used.

RESULTS AND DISCUSSION

Cistern water

The data for water samples are presented in Table 2. Two elements (Be and Co) were not detected in any samples and have been designated as "not detected (n.d.)". Of the remaining elements, 20 had low rates of detection (< 75%), and only 6 (Al, Ba, Sb, Sr, V and Zn) had rates of detection > 75%. Following designation of values reported to be less than the detection limit (DL) to a value of

Table 2 | Summary statistics for pH, conductivity (μS/cm), major ions (mg/L) and metals (μg/L) measured in cistern water. Concentration data to 2 sig. figs

N = 112	DL*	N > DL	% > DL	Arith. mean	s.d.	CoV†	95th %ile‡	Median	Geo. mean	Geo. lower 95% CI§	Geo. upper 95% CI¶	Min.	Max.
pH and Conductivity (μS/cm):													
pH	0	112	100%	7.82	0.37	5%	7.89	7.77	**7.81**	7.13	8.55	6.93	9.53
Cond	0	112	100%	130	220	168%	170	96	**100**	37	280	36	2,300
Major Ions (mg/L):													
Cl^-	0.5	112	100%	30	98	323%	48	13	**15**	2.9	77	0.69	920
NO_3^-	0.5	111	99%	5.0	18	358%	8.4	1.2	**1.6**	<0.5	13	<0.5	150
SO_4^{2-}	0.5	111	99%	9.7	22	231%	14	5.5	**6.4**	1.5	27	<0.5	240
Ca^{2+}	0.5	112	100%	15	12	85%	17	12	**13**	5.5	30	5.4	120
K^+	0.1	109	97%	1.2	2.7	229%	1.7	0.50	**0.61**	<0.1	4.0	<0.1	25
Mg^{2+}	0.1	112	100%	1.5	4.4	284%	2.4	1.0	**1.0**	0.29	3.5	0.30	47
Na^+	1	112	100%	13	38	286%	20	7.0	**7.7**	1.8	33	2.0	390
Metals (μg/L):													
Fe	5	70	63%	17	35	207%	24	6.0	**7.8**	<5	73	<5	330
Mn	1	37	33%	Trace‖				Trace	**Trace**			<1	11
Ag	0.4	4	4%	Trace				Trace	**Trace**			<0.4	0.70
Al	20	109	97%	130	85	63%	150	130	**110**	28	430	<20	690
As	0.4	13	12%	Trace				Trace	**Trace**			<0.4	1.8
B	20	17	15%	Trace				Trace	**Trace**			<20	360
Ba	0.2	112	100%	5.0	3.5	70%	5.6	3.9	**4.2**	1.4	13	1.2	31
Be	1	0	0%	n.d.**				n.d.	**n.d.**				
Bi	0.1	8	7%	Trace				Trace	**Trace**			<0.1	1.2
Cd	0.2	1	1%	Trace				Trace	**Trace**			<0.2	0.40
Co	0.2	0	0%	n.d.				n.d.	**n.d.**				
Cr	0.8	43	38%	0.98	1.5	151%	1.3	Trace	**Trace**			<0.8	15
Cu	1	27	24%	1.9	7.4	382%	3.3	Trace	**Trace**			<1	74
Mo	0.1	74	66%	0.20	0.22	110%	0.24	0.10	**0.13**	<0.1	0.75	<0.1	1.3
Ni	0.2	16	14%	Trace				Trace	**Trace**			<0.2	2.0
Pb	0.1	64	57%	0.47	1.7	351%	0.79	0.10	**0.15**	<0.1	1.6	<0.1	17
Sb	0.4	112	100%	0.86	0.34	39%	0.93	0.80	**0.82**	0.46	1.5	0.40	2.9
Se	0.4	57	51%	0.62	2.1	332%	1.0	0.40	**Trace**			<0.4	22
Sn	0.4	2	2%	Trace				Trace	**Trace**			<0.4	0.80
Sr	0.2	112	100%	160	220	137%	200	96	**110**	27	470	30	1,700
Ti	5	6	5%	Trace				Trace	**Trace**			<5	21
Tl	0.1	8	7%	Trace				Trace	**Trace**			<0.1	0.20
U	0.1	17	15%	Trace				Trace	**Trace**			<0.1	0.50
V	0.2	109	97%	2.6	1.9	74%	2.9	2.2	**2.0**	0.40	9.9	<0.2	13
Zn	4	84	75%	23	53	227%	33	8.0	**9.2**	<4	110	<4	480
Hg	0.2	4	4%	Trace				Trace	**Trace**			<0.2	0.50

*DL = detection limit.
†CoV - coefficient of variation = mean/s.d.
‡95th percentile of untransformed data.
§Lower 95% confidence interval of the geometric mean.
¶Upper 95% confidence interval of the geometric mean.
‖Trace = calculated value < DL.
**n.d. = no samples with value > DL.
Bold values = best indicator of central tendency.

0.5 × DL, the measures of central tendency (mean, median and geometric mean) for 15 elements had values less than the analytical DL. They have therefore been designated as "Trace", indicating that they were present on average at Trace levels.

Raw and log-transformed data were subjected to the Shapiro-Wilks test at $P \leq 0.05$ to test for normal or log-normal data distributions. Only one parameter, Ba, was found to conclusively follow a log-normal distribution. Examination of the histograms (not shown) revealed that the distributions of the remaining parameters approximate log-normal distributions, and the geometric mean is therefore the best estimate of central tendency, as indicated in Table 2.

The geometric mean of pH values recorded in cistern water was 7.81. This is higher than the annual mean pH of rainwater observed in Bermuda in 2005, which had a volume weighted average value of pH 5.06 (Peters unpublished data). Similar observations of increases in the pH of roof runoff have been attributed to: dissolution of minerals in roofing material (Querk & Förster 1993); the neutralizing effect of alkaline soil particles deposited on roof surfaces (Thomas & Greene 1993); and the neutralizing effect of cement surfaces in water cisterns (Simmons *et al.* 2001).

The Bermuda Department of Health has issued Drinking Water Standards (DWS) for ensuring healthy supplies of drinking water. These consist of primary standards which specify maximum acceptable limits and are designed to protect human health, and secondary standards which specify limits based on aesthetic quality, i.e. unacceptable colour, taste and odour. The data from the water cisterns is compared with the Bermuda DWSs in Table 3, which also lists the U.S. EPA DWSs for reference.

There were a total of 33 incidences of a Bermuda DWS being exceeded at 27 individual residences, representing 24% of all sites tested. The majority of these cases relate to secondary standards and primary DWS were exceeded in only 5 cases (occurring at a total of 3 residences, 2.7% of total). One residence exceeded 4 DWSs, consisting of 2 secondary (Cl^- and Na^{2+}) and 2 primary DWS (NO_3^- and Se). Three other residences recorded incidences of exceeding 2 DWSs: two of these residences exceeded secondary DWSs only and one exceeded 2 primary DWSs (NO_3^- and Pb). Full details of DWS non-compliance were provided to the Department of Environmental Protection as soon as the results were available so that immediate action could be taken, as appropriate. In nearly all cases, it was found that these incidents could be accounted for by unapproved use of unregistered groundwater wells to supplement the water supply. Groundwater in Bermuda is prone to contamination with NO_3^- from septic tank leakage. Unacceptable levels of Na^{2+} and Cl^- could arise from the use of well water taken from within the freshwater-saltwater transition zone of a groundwater lens, or it could indicate a significant sea salt component arising from deposition of sea spray onto the roof surface.

Table 3 | Drinking water standards (DWS) in effect in Bermuda and the USA for parameters measured in this study. Note different units of measure. U.S. EPA limits from EPA (2002*a*)

	Bermuda DWS	*N* > Bermuda DWS	% > Bermuda DWS	US EPA DWS
Primary Drinking Water Standards:				
As	50 μg/L	0	0%	10 μg/L
Cd	5 μg/L	0	0%	5 μg/L
Cr	50 μg/L	0	0%	100 μg/L
Pb	15 μg/L	1	0.9%	15 μg/L
Hg	1 μg/L	0	0%	2 μg/L
NO_3^-	44.3 mg/L	3	2.7%	44.3 mg/L
Se	10 μg/L	1	0.9%	50 μg/L
Ag	50 μg/L	0	0%	
Secondary Drinking Water Standards:				
Ag				100 μg/L
Al	200 μg/L	17	15.2%	50–200 μg/L
Cl^-	300 mg/L	2	1.8%	250 mg/L
Cu	1,000 μg/L	0	0%	1,300 μg/L
Fe	300 μg/L	1	0.9%	300 μg/L
Mn	100μg/L	0	0%	50μg/L
Na^{2+}	200 mg/L	1	0.9%	
pH	6.5–8.5	7	6.3%	6.5–8.5
SO_4^{2-}	250 mg/L	0	0%	250 mg/L
Zn	5,000 μg/L	0	0%	5,000 μg/L

Cistern sediment

The data for sediment samples are presented in Table 4. Two elements (U and Tl) were not detected in any samples

Table 4 | Summary statistics elements (mg/kg) measured in cistern sediment. Concentration data to 2 sig. figs

N = 36	Mean	s.d.	CoV*	95th %ile†	Median	Geo. mean	Geo. lower 95% CI‡	Geo. upper 95% CI§	Min.	Max.
Sb	0.78	1.7	209%	1.3	0.30	**0.38**	<0.1	2.5	<0.1	7.6
As	5.0	4.2	84%	6.4	3.8	**3.9**	1.1	14	1.3	21
Hg	2.4	5.0	210%	4.0	0.81	**0.98**	0.088	11	0.090	30
Se	0.93	0.53	57%	1.1	0.80	**0.78**	0.23	2.6	0.10	2.3
Al	**18,000**	5,500	30%	2,0000	18,000	17,000	8,100	35,000	6,800	26,000
Ba	220	97	43%	250	190	**200**	90	460	86	530
Ca	**190,000**	6,2000	32%	210,000	190,000	180,000	80,000	380,000	37,000	340,000
Cd	3.9	3.5	89%	5.0	2.9	**2.8**	0.58	14	0.70	15
Co	7.8	3.6	46%	9.0	8.0	**7.0**	2.7	18	3.0	19
Cr	61	27	44%	70	54	**56**	24	130	20	140
Cu	150	150	99%	210	110	**110**	26	490	34	720
Fe	18,000	30,000	162%	28,000	12,000	**14,000**	4,400	42,000	6,800	190,000
K	**700**	210	30%	780	700	670	360	1,300	300	1,200
Mg	9,100	2,800	30%	10,000	8,500	**8,700**	4,900	15,000	4,800	19,000
Mn	260	110	42%	300	230	**240**	110	520	120	680
Na	730	240	32%	810	700	**690**	380	1,300	400	1,600
Ni	**55**	26	46%	64	55	49	18	130	13	130
P	1,300	680	52%	1,500	1,000	**1,100**	440	2,900	490	2,800
Pb	230	220	94%	300	180	**160**	33	810	33	1,100
Sr	1,200	600	51%	1,400	1,000	**1,000**	400	2,700	460	3,000
Ti	**860**	280	32%	960	860	820	410	1,600	310	1,600
V	32	15	47%	37	27	**29**	12	67	12	74
Zn	4,600	4,500	97%	6,100	2,600	**2,800**	380	21,000	390	20,000
Ag	Trace¶				Trace	Trace			<1	2.0
Be	Trace				Trace	Trace			<1	1.0
Mo	Trace				Trace	Trace			<1	3.0
Sn	Trace				Trace	Trace			<5	20
U	n.d.‖				n.d.	n.d.				
Tl	n.d.				n.d.	n.d.				

*CoV - coefficient of variation = mean/s.d.
†95th percentile of untransformed data.
‡Lower 95% confidence interval of the geometric mean = GM/GSD$^{1.96}$.
§Upper 95% confidence interval of the geometric mean = GM × GSD$^{1.96}$.
¶Trace = calculated value < DL.
‖n.d. = no samples with value > DL.
Bold values = best indicator of central tendency.

and 4 elements (Ag, Be, Mo and Sn) had low rates of detection (<75%). Following designation of values reported to be less than the detection limit (DL) to a value of 0.5 × DL, the measures of central tendency (mean, median and geometric mean) for 4 of the elements with low detection rates (<75%) had values less than the analytical DL. They have therefore been designated as "Trace", indicating that they were present at Trace levels.

Sediment data were analysed using the Shapiro-Wilks test as described above. Of the data with detection rates > 75%, 5 parameters were found to conclusively follow a normal distribution and 16 were found to conclusively follow a log-normal distribution. The distributions of Sb and Fe approximate a log-normal distribution, and the geometric mean is therefore the best estimate of central tendency for these elements. The resulting best indicators of central tendency are indicated in Table 4

There are no legislative limits in effect for sediment quality in drinking water cisterns in Bermuda. However, it is of use to compare the concentrations of metals in cistern sediment with the U.S. EPA generic soil screening limits (SSLs) calculated to minimise risk from human ingestion of metals in soil. The results of this comparison are shown in Table 5. It is essential to note that the SSLs are not statutory or cleanup standards: they are used to identify sites which do not require further Federal attention under the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA), also known as "Superfund" (EPA 1996, 2002*b*). They are part of a tiered framework and when considering these generic SSLs it is essential that it is recognised that they are: (1) screening values, used to identify potential contamination and exposure; and (2) they are more stringent (i.e. conservatively protective) than site-specific limits. Where contaminant concentrations equal or exceed SSLs, further study or investigation is warranted but not necessarily cleanup or other remedial action (EPA 1996).

Table 5 | U.S. generic Soil Screening Limits (SSL) for ingestion of soil in residential scenarios and frequency (N > SSL and % > SSL) of sediment samples exceeding an SSL

	Ingestion SSL (mg/kg)	*N* > SSL	% > SSL
Sb	31	0	0%
As	0.4	36	100%
Hg	23	1	3%
Se	390	0	0%
Ba	5,500	0	0%
Be	160	0	0%
Cd	70	0	0%
Cr	230	0	0%
Ni	1,600	0	0%
Pb	400	4	11%
V	550	0	0%
Zn	23,000	0	0%
Ag	390	0	0%
Tl	6	0	0%

It can be seen that 100% of water cisterns sampled contained sediment with levels of As above the generic SSL for human ingestion. Cistern sediment samples also exceeded the SSL for Hg at one location and for Pb at 4 locations. This suggests that ingestion of sediment from Bermuda water cisterns may pose a human health risk. However, further information on suspended sediment levels in water cisterns is required to fully assess any associated risk. At present, there is a lack of data describing the dynamics of particle cycling in roof-harvested rainwater cisterns.

Further analysis of the chemical speciation and (bio)availability of As and other elements in cistern sediment is required to compile a site specific risk assessment. However, the fact that none of the water samples showed As levels above either the Bermuda or U.S. EPA drinking water standards while levels in the sediment were relatively elevated suggests that As is present as a strongly bound residue in the sediment which is unavailable for dissolution to the overlying water. Thus it appears that the sediment is not a significant source of dissolved As in water and the As is most likely strongly bound to Fe- and Al-hydroxides present in the sediment. Similar results for arsenic in soil have been observed, whereby the particulate phase of soil has high concentrations of As relative to the dissolved phase, owing to sorptive processes and formation of an As solid phase via chemisorption (Turpeinen *et al.* 2003).

Comparison of the elemental concentration profile of cistern sediment with that of local soil (Peters unpublished data) indicates that the two components have a similar composition (see Figure 2), indicating that they may be derived from a similar source: i.e. local soil is a significant source of cistern sediment. This suggests that dry deposition of material to roofs is the predominant source of metals in water cisterns in Bermuda and that in contrast to other studies, the roofing material is not the major source of metals in roof-harvested water (Förster 1999; Simmons *et al.* 2001; Chang *et al.* 2004; Rocher *et al.* 2004).

To test for any relationship between cistern sediment and the overlying water, pair-wise correlation was performed on data for those elements in sediment and water

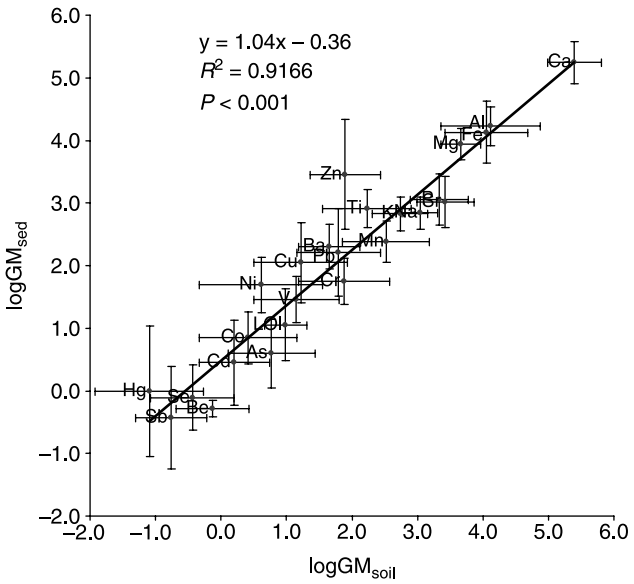

Figure 2 | Correlation of the geometric mean concentrations of elements in sediment ($GM_{sed.}$) and soil (GM_{soil}). Error bars indicate the geometric 95% confidence interval for each value. Note $\log_{10}$ axes scale.

which recorded mean values above Trace level in both Tables 2 and 4 (i.e. Ca, K, Mg, Na, Al, Ba, Fe, Mo, Pb, Sb, Sr, V and Zn). No significant relationships were found at $P \leq 0.05$. This further indicates that the sediment is not a significant source of these elements in the overlying water. This could be a result of the relatively short residence time of water in cisterns compared to that of the sediment or be related to sorptive conditions in the sediment-water system as described above for As. A detailed study of the kinetics of metal adsorption-desorption in the sediment-water system would help to better determine the significance for human health of sediment accumulation in drinking water cisterns.

CONCLUSIONS

It has been shown that the chemical quality of water in roof-harvested rainwater systems in Bermuda is generally good. The incidence of health-based primary drinking water standards being exceeded in water cisterns was low: $< 1\%$ for Pb and Se and $< 3\%$ for nitrate.

However, the data suggest that the potential human exposure to chemical contaminants may be of concern with respect to arsenic, lead and mercury associated with sediment which has the potential to become re-suspended in drinking water. There currently are no standards or guidelines in effect in Bermuda or, to the best of our knowledge, elsewhere in the world, with which to assess the quality of water cisterns with regard to sediment quality. To enable a human health risk assessment to be performed to evaluate the potential risk arising from the consumption of sediment entrained in drinking water, further detailed information is needed to fully characterize the long-term cycling of suspended sediment in roof-harvested rainwater systems. This would provide the first steps to the formulation of such standards.

Any risk presented by consumption of suspended particles in drinking water from roof-harvested water cisterns can easily be reduced by adopting some simple and inexpensive technological adaptations to water cisterns. These can include: design considerations of cistern plumbing to minimize the entrainment of sediment in out-flowing cistern water; use of a first flush diversion device to dispose of the first portion of each rainfall event; and installation of filters on roof gutters and inlet and outlet plumbing (UNEP 1998*a*; Martinson & Thomas 2005*a*,*b*; Meera & Mansoor Ahammad 2006). None of these approaches to cistern water quality improvement are common in Bermuda.

This preliminary study has identified the following recommendations for further research to better characterize the quality of water in domestic water cisterns in Bermuda:

- Examine the extent to which cistern sediment is re-suspended during rainfall and artificial recharge events;
- Investigate the significance of sediment re-suspension on contaminant adsorption/desorption and speciation, particularly for arsenic;
- Determine the amount of sediment which is carried through plumbing to the point of use;
- Determine the relative source contributions of contaminants from rainfall (wet deposition) and from material deposited on the roof surface (dry deposition);
- Investigate the significance of the physical parameters of roofs on contaminant deposition and accumulation.

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