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Research article

Irrigated greywater in an urban sub-division as a potential source of metals to soil, groundwater and surface water

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ABSTRACT

Increased water demands in dry countries such as Australia, have led to increased adoption of various water reuse practices. Irrigation of greywater (all water discharged from the bathrooms, laundry and kitchen apart from toilet waste) is seen as a potential means of easing water demands; however, there is limited knowledge of how greywater irrigation impacts terrestrial and aquatic environments. This study compared four greywater irrigated residential lots to adjacent non-irrigated lots that acted as controls. Accumulation and potential impacts of metals in soil, groundwater and surface water, as a result of greywater irrigation, were assessed by comparing measured concentrations to national and international guidelines. Greywater increased concentrations of some metals in irrigated soil and resulted in As, B, Cr and Cu exceeding guidelines after only four years of irrigation. Movement of metals from the irrigation areas resulted in metal concentrations in groundwater (Al, As, Cr, Cu, Fe, Mn, Ni and Zn) and surface water (Cu, Fe and Zn) exceeding environmental quality guidelines again within four years. These results are unlikely to be universally applicable but indicate the need to consider metals in greywater in order to minimize potential adverse environmental effects from greywater irrigation.

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1. Introduction

Water scarcity is a problem worldwide (Jury and Vaux, 2007; Godfrey et al., 2009). Reuse of greywater has been advocated as one potential solution (Eriksson et al., 2002; Winward et al., 2008; Eriksson and Donner, 2009; Maimon et al., 2010; Zhu et al., 2015). Greywater is wastewater and is typically generated from laundries, bathrooms (showers baths and hand sinks) (Christova-Boal et al., 1996; Eriksson et al., 2002) and sometimes includes kitchen wastewater (sinks and dishwashers) (Nolde, 2000; Friedler, 2004; Maimon et al., 2010), but does not include wastewater generated by toilets (i.e., black-water). Greywater reuse typically occurs via irrigation (Wiel-Shafran et al., 2006; Howard et al., 2007; Travis et al., 2010) although other uses, such as flushing toilets, are becoming more common (Jeppesen, 1996; Godfrey et al., 2009; March and Gual, 2009; Etchepare and van der Hoek, 2015). Despite widespread adoption of the reuse of greywater for irrigation, there is limited research on the impacts of this wastewater on the receiving environment (Donner et al., 2010; Stevens et al., 2011; Turner et al., 2013; Reichman and Wightwick, 2013).

The environmental sustainability of greywater irrigation has been questioned as greywater contains contaminants such as nutrients, cations and anions (e.g., Wiel-Shafran et al., 2006; Turner et al., 2013) and metals (Eriksson and Donner, 2009). Metals in greywater can originate from the source water (Huston, 2010), plumbing (Eriksson and Donner, 2009), household products, household appliances and personal care products (Gray and

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Becker, 2002; Diaper et al., 2008; Tjandraatmadja et al., 2008; Ayenimo et al., 2010). Tjandraatmadja et al. (2008) investigated the presence of As, B, Cd, Cu, Fe, Pb, Ni, Sn and Zn in a range of household and personal care products and found that Fe and Zn were present in the majority of products, B was present in a large range of products (laundry, kitchen and bathroom) and traces of Pb and Ni were also in many products.

Although the potential risks posed by nutrients and salts are commonly addressed in Australian federal and state government guidelines (e.g., ACT Health, 2007 and DoEUS NSW, 2008), the potential hazards posed by metals (Förstner and Wittmann, 2012) in greywater are inadequately addressed. Water quality guidelines for greywater exist in Australia (e.g., EPHC, 2006), but few guidelines are available for metals and those that are included are often derived from a limited dataset (Eriksson et al., 2006; Revitt et al., 2011; Turner et al., 2013). The guidelines for metals that do exist are based on drinking water quality guidelines and irrigation guidelines taken from ANZECC and ARMCANZ (2000). These irrigation guidelines have been developed for the protection of agricultural crops and are not based on ecological considerations. Minimal research has been published assessing metals in greywater and their environmental impacts (Eriksson and Donner, 2009; Reichman and Wightwick, 2013) and no single study has assessed the impacts of metals from greywater irrigation in three connected environmental compartments: soil; groundwater; and surface water (Fig. 1).

Understanding the transport of metals from greywater irrigation in compartments of the environment is important as metals are highly persistent in the environment and some are toxic (Bryan and Langston, 1992; Facchinelli et al., 2001; Rattan et al., 2005; Peralta-Videa et al., 2009). Metal adsorption to soils is complex (Bradl, 2004) and desorption can lead to off-site transport of metals via leaching (Schmidt, 2003) resulting in contamination of groundwater (Rattan et al., 2005) and surface water by leachate. Erosion of contaminated soils, can also result in contamination of surface waterways (Zhong et al., 2015).

It is important that robust government environment regulation and policy is underpinned by sound science. Chartres (2006) emphasized that water resources management policy should not be made without sound evidence – unfortunately this is not always possible. For example, during the Australian millennium drought (late 1996 to mid-2010), water saving policy, including greywater reuse, was urgently needed; however, targeted science supporting greywater reuse and its impacts on the environment was not available. Consequently, in Queensland, and other Australian states, the next best underpinning science (onsite sewerage guidelines (e.g. DIP, 2007)) was used.

The aim of this study was to determine whether metals in greywater that was irrigated to soil could lead to elevated concentrations of metals in soils, groundwater and adjacent surface water. Therefore, this study:

1. assessed the metal composition of the source water and greywater from four households;
2. compared metal concentrations in greywater irrigated soil to those in adjacent non-irrigated soils (controls);
3. compared soil metal concentrations in irrigated soils to national and international soil contaminant guidelines;
4. assessed the transportation of metals from greywater; and
5. compared metal concentrations in groundwater and surface water to national and international contaminant guidelines.

2. Methods

2.1. Research design

The design of this study included the collection and analysis of the following samples for metals:

- greywater;
- soil from four residential urban lots irrigated with greywater and from four adjacent vacant non-irrigated lots (controls);

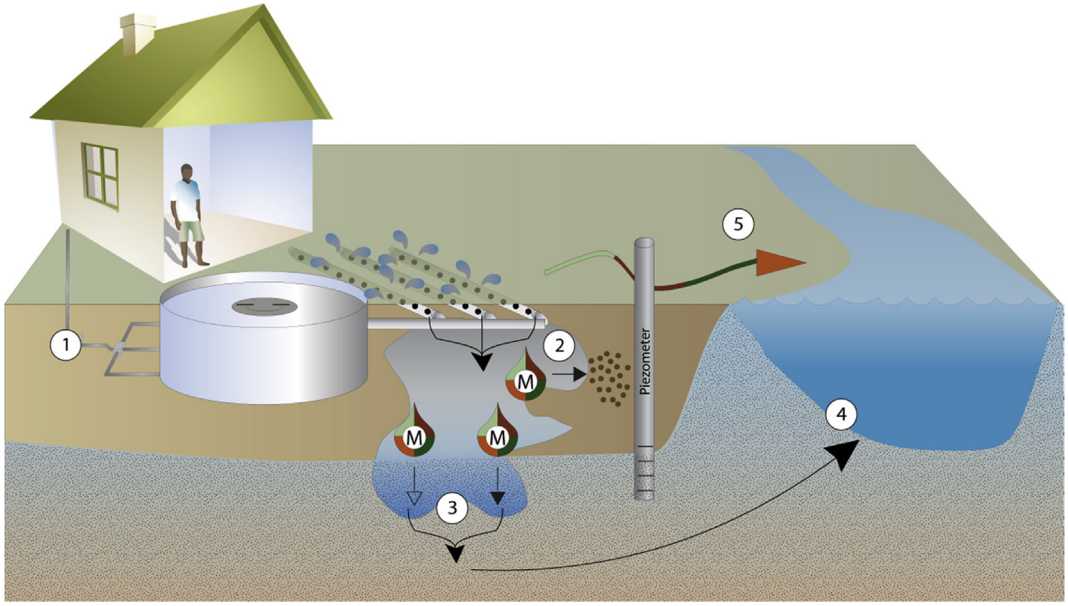


Fig. 1. Conceptual diagram of greywater irrigation and metal movement; Household sources of metals ① stored in a concrete tank then distributed via greywater sub-surface irrigation resulting in direct and indirect metal transportation (metal soil adsorption - direct ②; metal leaching - direct and indirect ③); lateral movement of contaminated groundwater to surface water ④; and surface runoff with soil erosion - (not assessed in this study ⑤) and the impacts in three connected environmental compartments: soil; groundwater (via sampling at the piezometer(s); and surface water.

- perched groundwater and water from an adjacent creek to identify the potential off-site transport of greywater metals from the irrigated lots.

2.2. Study area

The study area is a residential subdivision located approximately 10 km west of Brisbane, Queensland, Australia at the base of a High Ecological Value (HEV) area of Enoggera Reservoir catchment (EHP, 2010). The study area consisted of 22 lots totaling an area of 3.8 ha that borders 200 m of Enoggera Creek (Fig. 2). The subdivision is decentralised from basic utilities and requires each resident to have greywater irrigation. Individual lots ranged in area from 800 to 1800 m^2. Each lot has a 200 m^2 grassed transpiration area that receives the greywater irrigation. The water supply for each household is captured rain water, supplemented when necessary by council potable water. Samples from the rainwater tanks were collected from each household prior to this study; during February, May, June and September 2006 and March and September 2007 and sent to a National Association of Testing Authorities (NATA) accredited laboratory – Queensland Urban Utilities – for dissolved metals analysis using inductively coupled plasma-optical emission spectrometer (ICP-OES).

Four lots (lots A, B, C and D) from the six occupied sites of the subdivision were chosen for this study. They were selected based on household demographics (Turner et al., 2013), workplace health and safety and site access. Control sites were chosen on vacant land next to each lot, approximately 10–15 m away from the irrigated transpiration zones. All lots and controls had identical soil texture and similar carbon content (Turner et al., 2013).

2.3. Greywater

The greywater from the four selected lots was treated (via vermiculture) and stored onsite in concrete greywater treatment systems (Biolytix, 2005) and subsurface drip irrigated into a transpiration zone. Grab samples of greywater from each property were collected six times, over eight years: February 2006, May 2006, June 2006, November 2007, April 2009 and March 2014. Average chemical characteristics of the greywater samples taken during 2006 were presented by Beal et al. (2008). Each greywater sample was a composite 24-h sample taken directly from the irrigation outlet by a capillary tube with a bleed valve. Samples were collected in 2L bottles, stored on ice and retrieved daily. Each sample was subsampled according to the Australian Standard: *AS/NZS 5667.1:1998* (Standards Australia, 1998a, 1998b). All samples were passed through a 0.45 μm cellulose acetate filter (Sartorius) in order to obtain an estimate of the dissolved metal concentrations. Samples were preserved with ultrapure nitric acid to a final sample concentration of 1% (Eaton et al., 2005) and then refrigerated at 4 °C till analysed. All samples were sent to a laboratory accredited by the National Association of Testing Authorities (NATA) for chemical analysis, and therefore, complied with ISO 9001 and ISO17025 as a minimum.

Greywater samples were analysed using a Varian ICP-OES with simultaneous detection analysis (Varian, 2001; Eaton et al., 2005) for Al, As, B, Be, Bi, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sb, Se, Sr, Ti, V, and Zn. Ultra-trace dissolved metals analysis of greywater samples were analysed using a Thermo-Fisher, x-series, inductively coupled plasma-mass spectrometer (ICP-MS) (Thermo Electron Corporation, 2004; Eaton et al., 2005) for Ag, Al, As, B, Be, Bi, Cd, Co, Cr, Cu, La, Mn, Mo, Ni, Pb, Sb, Se, Tl, U, V, and Zn. Results for Al, B, Fe and Mn were combined between the two analytical methods (ICP-OES was used for results >500 μg L^{-1} and ICP-MS was used for results <500 μg L^{-1}). Further information on quality control sample results (filed blank, sample duplicate and certified reference material) and the analytical meta-data including; method setup, method of uncertainty and practical quantitation limits (PQL) for each analyte can be obtained in supplementary information – QA and QC. The accuracy of metal analysis was within the acceptable range of the trace metals Certified Reference Material for all metals analysed.

2.4. Soil

Up to five individual soil cores were taken from the transpiration zone of each irrigated lot using methods based on Rayment and Lyons (2010) and Peverill et al. (1999). Each core was hand augured to a depth of 0.3 m and the core was subsequently divided into 0.1 m segments. A total of 12 control soil cores were collected from the adjacent non-irrigated vacant lots (no more than 15 m away from each transpiration zone) and were divided into 0.1 m segments. These acted as controls, as the study area had been established for four years prior to the sampling. Sampling of the irrigated and non-irrigated lots occurred between 22nd January to 5th February 2009.

Each soil core segment (12–15 per lot and 36 controls) was air dried, ground to <2 mm and stored in air tight plastic containers. Soils were then extracted and analysed for metals. Mehlich3 extracts and ICP-OES (Rayment and Lyons, 2010) were used for Al and B as the laboratory did not have a suitable methods for acid extractable analysis due to typical contamination of ultra-pure nitric acid with Al and B. Microwave extractable metals analysis (metals were extracted with the method outlined in Eaton et al. (2005) and Standard Method No. 3051a (US EPA, 2007) using a Anton Paar 3000 microwave reaction system) was used for As, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Zn and also quantified by ICP-OES (Rayment and Lyons, 2010). Samples for ultra-trace metal analyses of Ag and Cd were digested by a Anton Paar 3000 microwave reaction system using nitric acid extraction based on standard method No. 3051a (US EPA, 2007; Eaton et al., 2005) and analysed by a Thermo-Fisher x-series ICP-MS. Strontium and V were not analysed in soils, as the laboratory used did not have a validated method for these elements (for additional information supplementary information – QA and QC).

2.5. Groundwater

The majority of the study area is steep with slopes up to 20% facing northeast (Fig. 2), with each lot being terraced. Given that the direction of groundwater flow generally follows surface topography (Apgar, 2000), three piezometers were installed in a gradient transect behind lot D (Fig. 2). This impact gradient transect was designed to capture any potential infiltration of metals associated with the greywater irrigation to the groundwater and subsequent transport towards Enoggera Creek. The groundwater depth was approximately 3000 mm, which is around the same level of the water in Enoggera Creek at ambient flow. The piezometer installation was based on the following assumptions. Piezometer one (P1) and two (P2) were located 15 m apart to intercept the groundwater potentially containing metals from infiltrated greywater. It was expected that P2 would have smaller metal concentrations than P1 as it is further from the potential source and would also have less impact by greywater infiltration due to the direction of groundwater flow. Piezometer three (P3) is 18 m from P2 and was located as a control and not expected to be affected by the greywater irrigation (Fig. 2).

The piezometers were installed by a licensed drill operator using a Geoprobe® 6600 series drill rig following the procedures set out

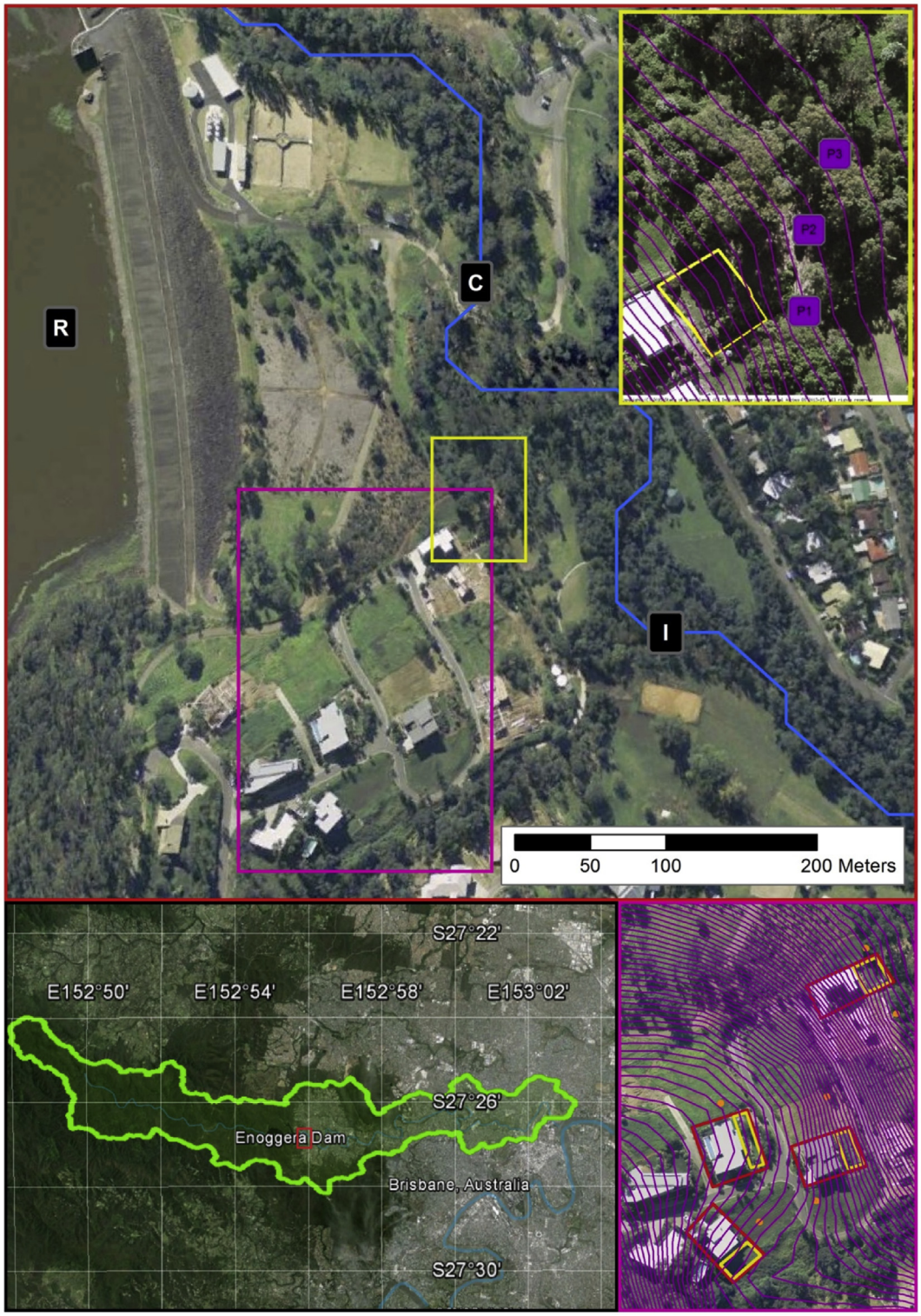


Fig. 2. Ariel photograph with four zoom levels showing: Zoom 1 (in black box); location of study area at the base of the High Ecological Valve - Enoggera Catchment 10 km west of Brisbane. Zoom 2 (red box); study site and Enoggera Creek location (blue line) with the surface water sites marked (black and grey boxes); reference site (R) in Enoggera Dam, upstream control (C) site and downstream impact (I) site. Zoom 3 (pink box); indicates the general topography (pink contours) of the four selected greywater irrigated lots for detailed investigation (red outline), transpiration zones (yellow outline) and control (vacant non-irrigated) sampling sites (orange circles). Zoom 4 (yellow box); lot D with the location of transpiration zone (yellow outline) and the piezometers (purple numbered P1 to P3).

in the "Groundwater Sampling and Analysis – A Field Guide. Geoscience Australia" (Sundaram, 2009) and adhering to *Water Act 2000* (Queensland, 2015). (For further information see supplementary information - groundwater). Samples were collected (between September 2013 to June 2014) from the piezometers in accordance with the Australian Standard: AS/NZS 5667.1:1998 (Standards Australia, 1998a, prepared and analysed for dissolved metal concentrations (as per section 2.3). Figures for each metal in

groundwater are presented in the supplementary information - groundwater.

2.6. Surface water

Enoggera Creek was sampled for metals to determine if there were potential greywater irrigation impacts. Three sites were selected: a reference site located in the Enoggera Creek Reservoir (formed by a dam wall); a control site (140 m downstream of the dam wall) and an impact site (a further 200 m downstream of the control site) (Fig. 2). The reference and control sites are upstream of any influence of groundwater affected by greywater, whereas the impact sites could be influenced. The majority of surface runoff and groundwater originated from the study site in this 200 m section of the creek (Fig. 2). Ambient water quality sampling occurred monthly with an increased frequency of sampling during high flow conditions. Samples were collected between September 2013 and June 2014 and analysed as outlined in section 2.3. Box and whisker plots were produced for the three sites (control, impact and reference) to compare the range of metal concentrations in Enoggera Creek (see supplementary information – surface waters).

2.7. Guideline and trigger value comparisons

Australian long-term irrigation guideline trigger values, which aim to protect plant growth from continual irrigation (ANZECC and ARMCANZ, 2000) were compared to the concentration of metals in the greywater. Metal concentrations in the soils of the transpiration zone were compared to the Australian Ecological Investigation Levels (EILs — soil quality guidelines for contaminated soils) in the National Environment Protection (Assessment of Site Contamination) Measure (NEPM, 2013). Data were also compared to the soil cumulative contaminant loading limit (CCL) for the cumulative amount of contaminant added (ANZECC and ARMCANZ, 2000). The CCLs are specifically designed to assess irrigation hazards.

Groundwater metal concentration data were compared to the groundwater investigation levels (GILS) provided in National Environment Protection Measure (NEPM, 2013) and when no GILS existed, metal concentration data were compared to the Australian surface water quality guidelines – as recommended by ANZECC and ARMCANZ (2000). Surface water metal concentration data were compared to the ANZECC and ARMCANZ (2000) surface water quality trigger values using the 95% level of ecosystem protection for slightly to moderately disturbed ecosystems. Where appropriate Australian guidelines did not exist, the equivalent Canadian Environmental Quality Guideline (CEQG) was used (CCME, 1999). These guidelines are presented in Table 1 and Tables 3–5. The exceedance of any guideline indicates an inappropriate risk to the environment that usually would lead to either immediate remedial or management action or further site-specific investigation.

2.8. Statistical analysis of soil, groundwater and surface water relationships

To assess metal accumulation in the soil from greywater irrigation, box and whisker plots were produced to compare the range of concentrations of metals in the transpiration zone soils to the control soils median concentrations (supplementary information — soils). Typically, environmental data is not normally distributed (Gilbert, 1987); therefore, non-parametric tests are required. The transpiration zone soil metal concentrations were analysed for differences using the non-parametric Kruskal-Wallis test (Hollander and Wolfe, 1999; Tölgyesi et al., 2014). A Grubbs test based on a two-tailed test where the p-value was set to <0.0001 and an alpha of 0.05, was used to identify outlying soil metal concentrations.

Groundwater samples from the piezometers were analysed for differences using the non-parametric Kruskal-Wallis test (Hollander and Wolfe, 1999; Tölgyesi et al., 2014), in order to identify increases in metal concentrations from P3 to P1. Surface water metal concentrations were analysed using the non-parametric Kruskal-Wallis test and Steel-Dwass-Critchlow-Fligner procedure (Hollander and Wolfe, 1999) to test for significant differences ($p \leq 0.05$) between the Impact, Control and Reference (Enoggera Creek Reservoir) sites. Spearman correlations were also performed to look for relationships between surface water concentrations of As and Fe in an attempt to explain results, as As and Fe commonly interact in aquatic environments (Sharma and Sohn, 2009).

3. Results and discussion

3.1. Greywater

Greywater concentrations of Sb, Be, Bi, Mo, Se, Ti, Tl and U were all below their practical quantitation levels (<PQL, refer to supplementary information – QA and QC) and will not be discussed further. General chemical characteristics, i.e., pH, electrical conductivity (EC), sodium absorption ratio (SAR), and concentrations of calcium (Ca), magnesium (Mg), sodium (Na), potassium (K), total nitrogen (TN) and total phosphorus (TP) of the same samples were reported in Turner et al. (2013).

All the greywater metal concentrations – except for B, Mn and Sr – were below typical greywater concentrations (EPHC, 2006) (Table 1). Silver could not be assessed as no published values were available. The rainwater tank samples (source water for greywater) were analysed for 21 of the 25 metals analysed in the greywater (Co, Ag, Sr and V were not analysed in rainwater). The greywater concentrations of Al, As, B, Cr, Cu, Fe, Pb, Mn, Ni, and Zn were greater than the rain tank water, with the maximum concentration of Al in greywater being 60-times larger than the maximum in the rain tank water (Table 1). This indicates that greywater is a potential anthropogenic source of a range of metals to the receiving environment.

Of the metals tested, only the maximum concentrations of B, Cu and Fe in greywater exceeded their long-term irrigation trigger values (Table 1). The Australian and New Zealand guidelines (ANZECC and ARMCANZ, 2000) state that when these trigger values are exceeded, potential environmental risks exist; e.g. off-site transport to groundwater and surface waters via deep drainage and leaching, effects on soil properties and effects on soil biota. In summary, in the greywater, there are elevated levels of Al, As, Mn and Sr compared to the source water and potentially hazardous concentrations of B, Cu and Fe.

3.2. Soil

All the soils were classified as a sandy clay loam. The carbon content at the surface was 3% and decreased down the soil profile. The average pH range of the transpiration zone soils was pH 5.8 to 6.1 ± (0.6) and the soil conductivity average range was 0.006–0.017 ±(0.06) S m^{-1} (see supplementary information - general soil characteristics, based on Turner et al. (2013)). Turner et al. (2013) reported that the control and irrigated transpiration zone soils from this study had similar characteristics in terms of total carbon, soil texture and general pH and EC characteristics. From this, an assumption was made that before the greywater irrigation began, the control and irrigated transpiration zone soils had similar metal concentrations and any subsequent measured differences were due to greywater irrigation.

Table 1
Summary statistics of dissolved metal concentrations in greywater for all lots compared to the Australian and New Zealand long-term irrigation trigger values (ANZECC and ARMCANZ, 2000) and observed greywater concentrations as well as typical rainwater tank concentrations. **Bold** values indicate where a trigger value was exceeded.

Greywater metals	n	Minimum ($\mu g\ L^{-1}$)	Median ($\mu g\ L^{-1}$)	Mean ± (σ) ($\mu g\ L^{-1}$)	Maximum ($\mu g\ L^{-1}$)	Irrigation guideline trigger values ($\mu g\ L^{-1}$)	Australian guidelines for water recycling - observed concentrations ($\mu g\ L^{-1}$)	Rain tank water concentrations supplied to household n = 9–18		Maximum rainwater tank concentrations	
								Mean concentration ($\mu g\ L^{-1}$)	Maximum concentration ($\mu g\ L^{-1}$)	Rodrigo et al. (2009) n = 23 ($\mu g\ L^{-1}$)	Huston et al. (2012) n = 88–363 ($\mu g\ L^{-1}$)
Al	26	50	310	380 ± (30)	1500	*5000*	44,000	6.6	25	114	560
As	19	0.01	0.8	1.6 ± (3)	12	*100*	13	2.5[b]	2.5[b]		3.7
B[c]	52	5	51	83 ± (10)	**520**	*500*	630[a]	6.1	15		
Cd	19	0.02	0.02	0.04 ± (0.04)	0.14	*10*	50	0.5[b]	0.5[b]		
Cr	19	0.3	1	1.2 ± (0.4)	2.8	*100*	5.5	1[b]	1[b]		4.3
Co	19	0.12	0.41	0.35 ± (0.16)	0.62	*50*	1.5				2
Cu	26	8	41	64 ± (81)	**390**	*200*	490	9	33	789	1600
Fe	26	20	70	120 ± (110)	**560**	*200*	28,000	4.6	35	232	4400
Pb	20	0.19	0.85	1.1 ± (1.4)	6	*2000*	150	2.5[b]	2.5[b]	22	85
Mn	26	2	14	20 ± (16)	58	*200*	14.3	4.5	18	6700	140
Ni	20	0.3	4.3	4.4 ± (2.2)	10	*200*	28	1[b]	1[b]		16
Ag	19	0.03	0.12	0.12 ± (0.03)	0.29	-					
Sr	19	10	40	70 ± (0.02)	170	-	60[a]				260
V	19	0.94	1.5	1.7 ± (0.7)	3.5	*100*					10
Zn	26	17	52	65 ± (62)	310	*2000*	13,000	63	113	15,800	26,000

Sigma (σ) = standard deviation.
[a] Result is based on mean only.
[b] Half practical quantitation limit.
[c] A Grubbs test was performed on the B results as apparent outlier was present. The test identified that the control population of samples contained one outlier with B concentrations of 81,000 $\mu g\ L^{-1}$ with a Z-score of 7.1, larger than the critical Z-score of 3.2. The result was thus excluded as an outlier.

When compared to control sites, the metals concentrations in the transpiration zone soils of at least one lot had significantly ($p \leq 0.05$) larger concentrations of As, B, Cr, Pb, Ni and Zn than the control soils (Table 2), most likely due to metals adsorbing onto the soil from the greywater. EILs were only available for As, Cr and Pb. Metal concentrations in the transpiration zone soils were also compared to the CCLs (ANZECC and ARMCANZ, 2000) and CEQGs if CCLs were not available. Despite these significant increases of metal concentrations in the irrigated soils, only the concentration of As exceeded its EIL and cumulative contaminant loading limits (CCL) (ANZECC and ARMCANZ, 2000) and only B exceeded the corresponding Canadian Environmental Quality Guidelines (CEQGs) (CCME, 1999) for park and residential soils (Table 3).

Closer examination revealed that As concentrations in the irrigated soils were similar to the controls, except for Lot D (Table 3), where greater than 75% of the soil samples were above the CCL (20 mg kg^{-1}). This anthropogenic loading of As in Lot D soil occurred after only 4 years of greywater irrigation. When the greywater of Lot D is assessed separately, the As average and maximum concentrations were 4.0-times (2.5 $\mu g\ L^{-1}$) and 9.5-times (12 $\mu g\ L^{-1}$) greater, respectively, than the other lots. Potentially exacerbating the As loadings of Lot D soil is the fact that soils that receive added phosphorus (P) will leach As (Feng et al., 2013; Bolan et al., 2014). Irrigated greywater contains elevated P concentrations (Turner et al., 2013) and consequent leaching of As has the potential to contaminate the ground and surface waters.

Boron was measured at elevated concentrations in the transpiration zone soils in all lots, with lots C and D being significantly ($p \leq 0.05$) larger than the control soils. The elevation of B is consistent with the elevated P concentrations observed previously (Turner et al., 2013), with the likely source being detergents. There are no Australian environmental soil guidelines for B; however, the CEQG (CCME, 1999) is 2 mg kg^{-1} and only 2 samples, from lot D, exceeded this value.

Chromium concentrations in the transpiration zone soils of lots B, C and D were significantly ($p \leq 0.05$) larger than the controls (Table 2). This result is surprising given the Cr concentrations in the greywater were only slightly larger than the rain tank water measured on site. No CCL guidelines exist for Cr so the CEQG of 64 mg kg^{-1} was used (CCME, 1999). Only the soil Cr concentration of Lot C exceeded the CEQG (Table 3).

Lead concentrations in transpiration zone soils of lots C and D were significantly ($p \leq 0.05$) larger than the controls and other lots (Table 2); however, none of the soil Pb concentrations exceeded the EIL or CCL for Pb (supplementary information – soils). Lead was not detected in the rainwater tanks, and was only measured at low concentrations in greywater (Table 1). There were elevated Ni concentrations in the transpiration zone soils of lots B and C compared to the controls (supplementary information – soils) but only Ni concentrations in Lot B were significantly ($p \leq 0.05$) larger than the control (Table 2). None of the soil results were above the EIL or CCL for Ni (supplementary information – soils). Concentrations of Zn in the transpiration zone soils of all lots were larger than the controls (supplementary information – soils), with significant ($p \leq 0.05$) increases in lots B and C (Table 2). No exceedances of either the EIL or the CCL for Zn occurred.

Statistical comparisons also identified significant ($p \leq 0.05$) decreases in Al, Cu, Fe, Pb and Ag concentrations in some transpiration zone soils, presumably due to leaching. It was expected that Al concentrations in transpiration zone soils would be larger than in control soils as Al concentrations in the greywater were elevated (Section 3.1). However, in all lots the soil Al concentrations were smaller than the controls, with the Al concentrations in the transpiration zone soils of lots A, B and C significantly ($p \leq 0.05$) smaller than the control soils (Tables 2 and 3). As

Table 2
Non-parametric Kruskal-Wallis p values results of the transpiration zone soils of lots A, B, C and D. **Bold** values indicate that the irrigated soils had significantly ($p \leq 0.05$) different concentrations to the control soils. An upward arrow (↑) indicates a significant increase in concentration, while a downward arrow (↓) indicates a significant decrease in concentration.

Soil metals	Lot A	Lot B	Lot C	Lot D
Al	**0.012**↓	**0.001**↓	**0.014**↓	0.222
As	0.999	0.199	1	<**0.001**↑
B	0.414	0.175	<**0.001**↑	<**0.001**↑
Cd	0.421	0.955	0.197	0.999
Cr	0.74	<**0.001**↑	<**0.001**↑	**0.009**↑
Co	0.755	0.995	0.755	0.699
Cu	0.271	1	0.085	<**0.001**↓
Fe	0.354	0.65	0.796	<**0.001**↓
Pb	**0.047**↓	0.972	**0.019**↑	**0.025**↑
Mn	0.77	0.65	0.945	0.928
Ni	0.689	**0.009**↑	0.093	0.128
Ag	<**0.001**↓	0.991	0.984	0.987
Zn	0.242	<**0.001**↑	<**0.001**↑	0.992

greywater has elevated concentrations of Ca, Na and N (Turner et al., 2013) a pseudo-podsolization process is likely to be occurring as a result of the greywater irrigation. Increasing soil pH by the addition of base cations (e.g. Ca and Na) can displace Al from binding sites on clay minerals (Reuss, 1983) resulting in leaching. In addition, an increase in nitrate and ammonium concentrations via irrigation can increase Al leaching (Emmett et al., 1995). The greywater has elevated Al concentrations and provides a continual supply of base cations, nitrate and ammonium, displacing Al from the soil into pore-water and then potentially into groundwater. There are no CCL or CEQG values for Al, so it is not possible to determine if the soil Al concentrations pose a risk to soil ecosystems and plants; however, this is unlikely as Al is a key constituent of aluminosilicates (clays).

Copper exceeded the irrigation guidelines and this resulted in Lot C exhibiting transpiration zone soils concentrations above the EIL and CCL. However, concentrations in Lot C transpiration zone soils were not statistically different ($p > 0.05$) to the control soils. Furthermore, Lot D had a significant ($p \leq 0.05$) decrease in concentrations of Cu. Irrigated greywater is a direct and indirect mechanism for Cu adsorption and leaching. The mobility of Cu is highly variable and is influenced by soil pH and P concentrations. Larger soil P concentrations will reduce Cu mobility, thereby reducing leachability; whereas decreasing soil pH will cause an increase in Cu mobility (Kumpiene et al., 2008) and leaching to groundwater. Of the four studied lots, Lot C had the largest average greywater P concentrations (25 mg L^{-1}), the largest soil Colwell extractable P concentrations (>350 mg kg^{-1}) (Turner et al., 2013) and the highest average pH (pH 6.4) causing Cu to adsorb to soil; resulting in Cu increases in the transpiration zone soils. In contrast, Lot D had a smaller average greywater P concentration (16 mg L^{-1}), a similar soil Colwell extractable P concentration (~175 mg kg^{-1}) compared to the control soils (Turner et al., 2013) and the lowest average pH levels (pH 5.8) causing Cu to be mobile and leach; resulting in less Cu in the transpiration zone soils. This Cu leaching is also compounded by the additional large concentration of Cu from irrigated greywater.

There was a significant ($p \leq 0.05$) decrease in concentrations of Ag in Lot A and of Fe in Lot D, which was unusual. Uptake by plants could cause or contribute to some of the observed decreases in transpiration zone soil metal concentrations; however, two factors mitigate the potential role of plants. Firstly, the transpiration

Table 3
Summary statistics and comparisons of metal concentrations in the transpiration zone (TZ) soils for all irrigated and control lots. The TZ soil results were assessed and compared with the Ecological Investigation Levels (EILs) in the National Environment Protection (Assessment of Site Contamination) Measure (NEPM, 2013) and the Australian and New Zealand long-term irrigation trigger values (ANZECC and ARMCANZ, 2000) and Canadian Environmental Quality Guidelines (CCME, 1999). **Bold** lots and values indicate where a trigger value was exceeded.

Soil metals[a]	Soil metal concentration (mg kg^{-1})		Location	n	Soil metal concentration (mg kg^{-1}) except Fe (%)			
	EILs[b]	Soil quality guidelines[c]			Minimum	Median	Mean ± (σ)	Maximum
As	50	20[f]	**Lot A**	12	8	14	14 ± (6.2)	**27**
			Lot B	15	14	17	18 ± (2.9)	**23**
			Lot C	12	12	14	14 ± (2.0)	18
			Lot D	14	17	36	42 ± (23)	**95**
			Controls	36	8	14	16 ± (9.5)	**53**
B	NG	2[g]	Lot A	12	0.13	0.13	0.17 ± (0.18)	0.76
			Lot B	15	0.13	0.13	0.15 ± (0.07)	0.33
			Lot C	12	0.13	0.58	0.93 ± (0.92)	**3.01**
			Lot D	14	0.13	0.20	0.36 ± (0.31)	1.00
			Controls	36	0.13	0.13	0.12 ± (0.01)	0.13
Cr	240[d]	64[g]	Lot A	12	8	24	23 ± (8.7)	37
			Lot B	15	27	35	37 ± (7.5)	56
			Lot C	12	23	39	41 ± (13)	**73**
			Lot D	14	17	26	26 ± (7.7)	48
			Controls	36	12	19	20 ± (5.9)	42
Cu	150[e]	140[f]	Lot A	12	20	50	48 ± (15)	75
			Lot B	15	47	61	64 ± (19)	129
			Lot C	12	61	82	86 ± (28)	**151**
			Lot D	14	28	37	39 ± (12)	71
			Controls	36	40	62	64 ± (19)	106

Sigma (σ) = standard deviation.
[a] Box plots for metal concentrations in the soil of the transpiration zones (TZ) of lots A, B, C and D and the non-irrigated controls as well as the probability values of each lot compared to the control samples using the Kruskal-Wallis test, are provided in the supplementary information.
[b] Values are Ecological Investigation Levels (EILs - Fresh) based on National Environment Protection (Assessment of Site Contamination) Measure (NEPM, 2013); NG = no guideline (either CCL or CEQG).
[c] [f] = cumulative contaminant loading limits (CCLs) (ANZECC and ARMCANZ, 2000).
[d] = Cr value adjusted for average study site soil characteristics, average soil clay content (25%) average background concentration (20 mg kg-1), average Fe content (2.3%).
[e] = Cu value adjusted for average study site soil characteristics, cation exchange capacity (95 meq 100 g −1), average soil pH (5.8) average carbon content (2.4%) average background concentration (64 mg kg-1), average Fe content (2.3%).
[g] = Canadian Environmental Quality Guidelines (CEQGs) (CCME, 1999) for park and residential soils. These were only used when no CCL was available.

Table 4
Groundwater 95th percentile metal concentrations (μg L^{-1}) for piezometers one, two and three (P1, P2 and P3) including the significant differences between piezometers one, and piezometers three. Results were assessed and compared with the Australian and New Zealand water quality guidelines using 95% level of protection (ANZECC and ARMCANZ, 2000) and Canadian Environmental Quality Guidelines (CCME, 1999). **Bold** values indicate where a trigger value was exceeded.

Groundwater metals[a]	95th percentile (μg L^{-1})			Trigger Values[b] PC95 (μg L^{-1})	Probability of comparison of P1 and P3
	P1 (n = 23)	P2 (n = 22)	P3 (n = 23)		
Al	**1780**	**880**	**1080**	55	0.967
As	**41**	11	2.7	24	0.006
B	73	46	30	370	<0.001
Cd	**0.7**	**0.2**	0.1	0.2	<0.001
Cr	**2.3**	**3**	**3.4**	1.0	0.925
Co	**61**	**11**	0.5	1.4[c]	<0.001
Cu	**18**	**17**	**7.6**	1.4	0.647
Fe	**9500**	**2990**	10	300[d]	<0.001
Pb	0.5	0.3	0.3	3.8	0.942
Mn	**4400**	**5000**	200	1900	<0.001
Ni	**31**	8.4	3.8	11	<0.001
Ag[e]	<0.05	<0.05	<0.05	0.05	–
Zn	**521**	**114**	**35**	8	<0.001

[a] Groundwater temporal figures for metal each concentrations and the Australian and New Zealand freshwater quality guideline trigger value for ecosystem protection (ANZECC and ARMCANZ, 2000) are provided in the supplementary information.
[b] Trigger values were obtained from Australian and New Zealand Guidelines for Fresh and Marine Water Quality water quality guidelines using 95% level of protection (ANZECC and ARMCANZ, 2000).
[c] A low reliability Australian guideline was available for Cobalt which was checked against the Canadian Environmental Quality Guidelines (CCME, 1999) Federal Water Quality Guideline of 2.5 μg L^{-1}.
[d] The current Canadian Environmental Quality Guidelines (CCME, 1999) Federal Water Quality Guideline is 300 μg L^{-1} which could be used as an interim indicative working level (ANZECC and ARMCANZ, 2000).
[e] All concentration for silver were below practical quantitation levels.

zones only contain grass which has a typical root depth of 8.3 mm (Connellan, 2002) and the greywater was released into the transpiration zones at 5–18 mm depth. Secondly, the soil transpiration metal concentration values were based on three depth segments taken at 100 mm intervals between the surface and 300 mm depth. Thus the most likely cause of metals decreasing at this site is due to leaching. This is consistent with the view of Mohamed et al. (2014), that soil metal concentration losses (due to grass uptake) would in general be minimal compared to leaching.

There were no differences in the concentration in the transpiration zone and control soils for Cd, Co and Mn and there were no exceedances of guidelines (see supplementary information – soils).

3.3. Groundwater

The 95th percentile concentrations of all the metals that were elevated in greywater (i.e., Al, As, B, Cu, Fe and Mn) decreased from P1 to P2 to P3 (Table 4) with the exception of Al, for which the concentration decreased from P1 to P3 to P2. This general trend confirms the experimental design that assumed that P1 would be most affected by greywater irrigation, P2 to a lesser extent and P3 the least (or as a control). Given the location of the piezometers and the flow of groundwater, the piezometers will mainly be affected by the irrigated greywater from Lot D. Greywater inputs to lots A to C should also affect the composition of groundwater down-flow of their location, but in a manner reflecting their greywater composition.

Table 5
Surface water (SW) 95th percentile metal concentrations (μg L^{-1}) for the control (C), impact (I) and reference (R) sites of Enoggera Creek. Results were assessed and compared with the Australian and New Zealand water quality guidelines (ANZECC and ARMCANZ, 2000) using 95% level of protection and Canadian Environmental Quality Guidelines (CCME, 1999). **Bold** values indicate where a trigger value was exceeded.

Surface water metals	Ennogera creek 95th percentiles (μg L^{-1})			Trigger Values PC95[a] (μg L^{-1})
	Reference n = 7	Control n = 21	Impact n = 21	
Al	12	26	44	55
As	1.8	21.0	6.6	24
B	32	33	34	370
Cd	<0.04	<0.04	<0.04	0.2
Cr	0.13	0.40	0.40	1.0
Co	0.09	0.14	1.13	1.4[b]
Cu	0.30	**2.76**	**2.80**	1.4
Fe	**481**	178	**369**	300[c]
Pb	0.20	0.33	0.43	3.8
Mn	85	214	1130	1900
Ni	<1.0	1.2	<1.0	11
Ag	<0.05	<0.05	<0.05	0.05
Zn	2.6	6.3	**11**	8

[a] Trigger values were obtained from Australian and New Zealand Guidelines for Fresh and Marine Water Quality water quality guidelines using 95% level of protection (PC95) (ANZECC and ARMCANZ, 2000).
[b] A low reliability Australian guideline was available for Cobalt (ANZECC and ARMCANZ, 2000) which was checked against the Canadian Environmental Quality Guidelines (CCME, 1999) Federal Water Quality Guideline of 2.5 μg L^{-1}.
[c] The current Canadian Environmental Quality Guidelines (CCME, 1999) Federal Water Quality Guideline is 300 μg L^{-1} which could be used as an interim indicative working level (ANZECC and ARMCANZ, 2000).

The concentrations of Al, As, B, Cd, Co, Cu, Fe, Pb, Mn, Ni and Zn at P1 were larger than those in the control (P3) but only As, B, Cd, Co, Fe, Mn, Ni and Zn (Table 4) were significantly ($p \leq 0.05$) different. The elevated concentrations of As and B in groundwater can be linked back to the elevated concentrations in the transpiration zone soil and then to the greywater (Tables 3 and 4). The elevated concentrations of Al, Cu and Fe in groundwater can be attributed directly to the greywater and to leaching of these metals from the transpiration zone soils (Table 3, Table 4 and supplementary information – soils). Logically, the elevated concentrations of Ni, Pb and Zn in groundwater must have originated from greywater, as the concentrations in the transpiration zone soil are elevated, even though the greywater concentrations were not highly elevated compared to the source water. The elevated concentrations of Mn in groundwater could be a result of the Mn in greywater not binding to soil (supplementary information – soils) and passing directly to the groundwater. This hypothesis is supported by Khattak and Jarrell (1989) who demonstrated increasing salinity in irrigation waters lead to increased mobility of Mn. The greywater chemistry of the households demonstrated high salinity with maximums of; 2511 μS cm^{-1} for electrical conductivity, 14 mg L^{-1} Ca, 6.3 mg L^{-1} Mg, 630 mg L^{-1} Na and 50 mg L^{-1} K (Turner et al., 2013).

The elevated groundwater concentrations of Cd and Co cannot be directly or indirectly attributed to greywater irrigation. The concentrations of Co in the groundwater were much larger (>10-times) than in the greywater, indicating that there was another source of Co apart from the greywater. A potential source of the elevated Co is fertilisers, where Co is present as a micro-nutrient (Singh, 1993). Similarly, greywater is unlikely to be the main source of Cd and is most likely to be phosphatic fertilisers (Lambert et al., 2007). Mineral fertilizers can also contain trace amounts of various metals as additives or as contaminants (Singh, 1993; Lambert et al., 2007) and may, therefore, have also contributed to the measured groundwater metal concentrations. However, the concentrations of all metals that were elevated in groundwater, apart from Co and Cd, can be explained by the composition of the greywater. Therefore, it can be ascertained that fertilisers are a relatively minor contributor to the observed groundwater metal concentrations.

The 95th percentile values of some piezometers for Al, As, Cd, Cr, Co, Cu, Fe, Mn, Ni, and Zn were larger than their corresponding GILS (Table 4), indicating that they pose a moderate risk to aquatic ecosystems (NEPM, 2013). There were no exceedances for B, Fe, Pb and Ag, and therefore, pose a low risk to aquatic ecosystems. In summary, the concentrations of As, Cd, Co, Fe, Mn, Ni and Zn were significantly elevated in the groundwater and exceeded GILS but only the exceedances of As, Fe, Mn, Ni and Zn could be attributed to greywater irrigation.

3.4. Surface water

Surface water concentrations of Al, B, Co, Cu, Fe, Pb, Mn and Zn were larger at the impact site than at the control site (Table 5) but only those for Co ($p < 0.001$), Fe ($p = 0.002$), Mn ($p = 0.001$) and Zn ($p = 0.007$) at the impact sites were significantly different than control sites. These surface water concentration increases indicate that there is at least one source of these metals up-gradient of the impact sites, which were not similarly influencing the control site. When comparing metal concentrations in surface water and groundwater, only Co, Fe, Mn and Zn were similarly elevated in the groundwater, and of these, only Fe, Mn and Zn can be linked directly back to the greywater irrigation (Table 4). The 95th percentile Fe concentration value at the Enoggera reservoir reference site was 481 μg L^{-1} and 178 μg L^{-1} at the control site immediately below the discharge point. This decrease between the reference and control site may reflect changes in the oxidation state of the water. At depth, water in reservoirs is often anoxic or has a low oxidation state, which once released, is aerated resulting in the Fe being converted from Fe^{2+} to Fe^{3+} and resulting in precipitation and a lowering of the dissolved Fe concentration. The subsequent significant increase at the impact site (369 μg L^{-1}) is thus presumed to be linked to groundwater discharge into Enoggera Creek, and therefore, linked to greywater irrigation.

Unexpectedly, the As concentrations at the impact site were significantly smaller than the control ($p = 0.014$) site. This may be a result of co-precipitation of As with Fe (Feng et al., 2013). The concentrations of As and Fe in surface water at the impact site were highly and significantly correlated (Spearman correlation coefficient = 0.72, p=<0.001), whereas the concentrations at the control site were not correlated (Spearman correlation coefficient = 0.042, $p = 0.86$). Furthermore, the observation of iron flock in the creek during sampling at the impact site, is consistent with this explanation for low concentrations of As at the impact site.

The 95th percentile concentrations of Cu, Fe and Zn at the impact site and Cu at the control site exceeded the corresponding Australian and New Zealand trigger values (ANZECC and ARMCANZ, 2000), indicating they pose a moderate risk to aquatic ecosystems. In summary, the metals that were elevated at the impact site compared to the control site (Co, Fe, Mn and Zn) and that exceeded their trigger values (Cu, Fe and Zn) can each be attributed to greywater irrigation.

3.5. Guideline comparison summary

The concentrations of metals in greywater, transpiration zone soil, groundwater and surface water were compared to relevant guidelines and trigger values (Table 6). Copper consistently exceeded trigger values/guidelines in all the environmental compartments as a result of greywater irrigation (Table 1). A number of metals accumulated in the transpiration zone soils, but only As, B, Cu and Cr exceeded trigger values/guidelines (Table 3). The concentrations of Al, As, Cd, Cr, Co, Cu, Fe, Mn, Ni and Zn in groundwater exceeded their trigger values, whereas the concentrations of only Cu, Fe and Zn exceeded their trigger values in the surface water. All exceedances indicate a moderate risk of environmental harm occurring. The inconsistencies in the metals that exceeded the corresponding trigger values/guidelines indicates the inherent limitation of deriving trigger values/guidelines that only consider effects within a single environmental compartment (e.g. soil, surface water or groundwater) or environmental value (e.g. ecosystem protection or irrigation). In order to use greywater responsibly, specific greywater guidelines should be developed that consider the effects of metals in all environmental compartments that might be directly and/or indirectly impacted. Greywater will also contain a variety of other inorganic (Turner et al., 2013) and organic chemicals (from household products that enter the greywater) and the potential transport and effects of these must also be considered.

3.6. Implications of the study

Greywater irrigation at this site has, within four years, increased concentrations of several metals such that they exceed appropriate environmental quality guidelines and thus pose a moderate risk to ecosystems in transpiration zone soils, groundwater and an adjacent creek. However, it is likely that the effects of greywater will vary at different sites due to variations in greywater composition and site-specific characteristics. For example, the chemical composition of greywater is likely to differ from site to site

Table 6
Comparison of the maximum concentration of each element measured in different media from the greywater irrigated lots compared with the corresponding Australian and New Zealand water quality guideline trigger values (ANZECC and ARMCANZ, 2000). Measured concentrations are less than (<) or greater than (>) the ANZECC and ARMCANZ (2000) guidelines, greater than the CCME (1999) guidelines (>C), or neither guideline was available (–).

Analyte	Greywater[a]	Soil CCL[b]	Groundwater[c]	Surface water[d]
Al	<	–	>	<
As	<	>	>	<
B	>	> C[e]	<	<
Cd	<	<	>	<
Cr	<	> C[f]	>	<
Co	<	<	<	<
Cu	>	>	>	>
Fe	>	–	>	>
Pb	<	<	<	<
Mn	<	<	>	<
Ni	<	<	>	<
Ag	–	–	<PQL	<PQL
Zn	<	<	>	>

[a] Greywater has been assessed with the ANZECC and ARMCANZ (2000) irrigation guidelines in table 9.2.17 using the long-term trigger value.
[b] The transpiration zones has been assessed with the ANZECC and ARMCANZ (2000) guidelines in table 9.2.17 using the soil cumulative contaminant loading limit (CCL).
[c] Groundwater has been assessed with the ANZECC and ARMCANZ (2000) water quality trigger value in table 3.4.1 using 95% level of protection.
[d] Surface water at the impact site has been assessed with the ANZECC and ARMCANZ (2000) water quality guidelines in table 3.4.1 using 95% level of protection.
[e] No Australian guideline was available for Boron therefore the Canadian Environmental Quality Guideline for residential/parkland was used (2 mg kg^{-1}) (CCME, 1999).
[f] No Australian guideline was available for Cadmium therefore the Canadian Environmental Quality Guideline for residential/parkland was used (64 mg kg^{-1}) (CCME, 1999).

reflecting the quantity and types of products used in households, and from region to region due to changes in product formulation to maintain product efficacy. Also, the extent of metal adsorption to soils is likely to vary spatially due to the composition of the soil and the aquifer (e.g. clay content, cation exchange capacity and pH). The ability to detect increased metal concentrations will also depend on the existing concentrations of the metals in the various environmental compartments and the distance to the nearest surface water. Nonetheless, this study clearly indicates the need to carefully consider the metals present in greywater (as a component or a contaminant) and manage the irrigation of water (by controlling the inputs to greywater or the amount and frequency of irrigation permitted) in order to obtain the potential water savings but minimizing environmental harm.

It is recommended that studies similar to the present (e.g. on a larger scale, using different soil and climatic types) are conducted at sites with different characteristics to determine if quantitative relationships similar to those of Broos et al. (2007) and Warne et al. (2008) can be developed and thus generate guidelines for greywater irrigation that account for site-specific characteristics. For example, it would be expected that sandy acidic soils (i.e. with a low clay content (and low CEC) and low pH) and aquifers would not bind metals and essentially all would move into the groundwater.

Further analysis of this data can determine soil enrichment factors and irrigation soil metal loadings. Although greywater irrigation is not always the direct source of the metals observed in the aquatic environment, greywater irrigation is influencing leaching. With this in mind irrigation assessment models such as MEDLI (Gardner et al., 2002) should be enhanced (for sub-surface irrigation) to assess the irrigation site's soil suitability for receiving metals from greywater irrigation and the greywater irrigations impact on soil leaching. This will help ensure future greywater irrigation approvals are designed to be environmentally sustainable. Additional work can also estimate loads of metals delivered to Enoggera Creek assessing the total amount of metal leached to the aquatic environment. Future work should also examine the potential environmental impacts associated with organic contaminants such as pharmaceuticals and personal care products (PPCPs) in greywater.

As more greywater data and scientific understanding is published, government regulation and policy on greywater reuse can use more relevant science and move away from gap filling science from a similar field (i.e. onsite sewerage guidelines). With further research, robust government environment regulation and policy will be underpinned by sound science (a strong science — policy interface). It is inevitable that Australia or other countries will be affected by periods of drought, during which water conservation policies (greywater reuse) will be needed. With ongoing research good science can be expected to underpin greywater reuse policy and the impacts on the environment can be minimised. Furthermore, updating greywater irrigation guidelines with a greater emphasis on metals will ensure that responsible reuse of greywater does not result in environmental harm.

4. Conclusions

Greywater irrigation can help reduce demand on freshwater for external uses such as watering gardens. However, this study provides evidence that greywater irrigation could potentially cause environmental harm due to metal accumulation in soils and the presence of metal contaminants in the aquatic environment.

After four years of commencing greywater irrigation, it has led to an increase in the average concentrations of As, B, Cr, Cu, Pb, Ni and Zn and decreased the concentrations of Al, Cu, Fe, and Ag from soil; increased the concentrations of Al, As, B, Cu, Fe, Pb, Mn, Ni and Zn in groundwater and increased the concentrations of Al, B, Cu, Fe, Pb, Mn, and Zn in an adjacent creek.

The concentrations of As, B, Cr and Cu in soil and Al, As, Cr, Cu, Fe, Mn, Ni, and Zn in groundwater and Cu, Fe, and Zn in the surface water also exceeded the corresponding environmental quality guidelines and thus pose a moderate risk to the exposed ecosystems. Although the composition of greywater and site-specific characteristics will modify the effects at each site, it is clear that the existing greywater reuse guidelines do not provide adequate environmental protection and they should be revised taking into account the concentrations of metals, and other contaminants, in greywater.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvman.2016.09.021.

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