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Assessing environmental risks of laundry detergents in greywater used for irrigation

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ABSTRACT

The objective of this study was to assess the environmental risk posed to Australian and New Zealand ecosystems by the presence of powdered laundry detergents in greywater used for irrigating gardens. Fifty powdered laundry detergents were assessed and all contained hazards which posed moderate to very high risks from increased alkalinity, sodicity and salinity to plants and soils when used at manufacturer-recommended doses and the resulting greywater used for irrigation. A number of detergents had phosphorus and boron concentrations considered to be a high risk for a number of plants. Risk to groundwater quality was also evaluated and found to potentially be a tighter constraint than risk to plants and soil where irrigation reuse is extensive in arid areas. A detergent environmental performance index was composed on risks assessed for three scenarios to compare with a washability performance index for the same powders. Only one detergent exceeded the 80% environmental index (100% = low risk from all hazards assessed) and maintained wash performance above 85%. The analysis suggests that for poorly drained soils greywater reuse is not recommended for most of the powdered laundry detergents evaluated. However the methodology may provide a basis for environmental labelling of detergents.

Key words | detergent, environment, greywater, groundwater protection, risk assessment

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INTRODUCTION

Climate change, prolonged and recurrent drought, population growth and deferred investment in new water supplies have led to water restrictions being imposed on most Australian cities in recent years. Garden watering with mains water has been limited or prohibited leading to the reuse of greywater (waste water from the laundry, showers, baths and taps) by many households to water gardens. Greywater reuse, roof rainwater collection and planting of water-efficient native species or xerophytes have been seen to be ways of sustaining gardens in the future. Greywater reuse may provide 280 to 400 L/household/day for garden use (Loh & Coghlan 2003; Roberts 2004), meeting 60 to 100% of European-style garden water requirements, depending on household water use, garden area, climate and soil type. However irrigation with greywater may have adverse impacts on human health (mainly because of pathogens) and environmental health and reduces the volume of water reaching wastewater treatment plants from which centralised water recycling may occur.

Laundry detergents are ubiquitous in greywater. They are complex commercial products containing a wide range of organic and inorganic compounds including builders, anti-redepositioning agents, corrosion inhibitors, fluorescent whitening agents, processing aids, colorants, fragrances,

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opacifiers, oxygen bleach, enzymes, fillers, surfactants and suds control agents (e.g. Hennes-Morgan & de Oude 1994; Warne & Schifko 1999).

The Australian Guidelines for Water Recycling Phase 1 (AGWR; NRMMC & EPHC 2006) cover the use of greywater in the urban environment and they identified a number of potential hazards (i.e. boron (B), cadmium (Cd), chlorine (Cl), hydraulic loading (from water), nitrogen (N), phosphorus (P), salinity and sodium (Na)) associated with the use of greywater to water gardens. The magnitude of these hazards can vary markedly. For example, Patterson (2004) showed that Na and P concentrations in Australian laundry detergents varied by up to 100-fold when used at manufacturer-specified concentrations. Such differences in the concentration of these hazards in laundry detergents will be reflected in the risks posed by greywater containing different detergents.

The AGWR (NRMMC & EPHC 2006) do not identify any organic constituents of laundry detergents as hazards from the land application of greywater. However, AGWR recommend a watching brief in the area. This is warranted as Warne & Schifko (1999) reported that organic surfactants in Australian laundry detergents were a major contributor (i.e. a mean of 40%) to the toxicity of the detergents and because of current concerns over chemicals in other pharmaceutical and personal care products such as nonylphenol and triclosan (Ying *et al.* 2007). Given the above, inorganic chemicals became the focus of this paper. A more comprehensive understanding of the environmental impact of organic chemicals in detergents is required to complement this study and provide a complete assessment of the risk posed by watering gardens with greywater.

Given the enormous variation in environmental receptors and conditions affecting behaviour and fate of hazards, this study was limited to analyses of constituents of the detergents, calculation of their concentrations in greywater and desk-top evaluations of specific environmental risks based on the AGWR. Prior to this study being undertaken three scenarios were considered:

1. irrigation of greywater from washing machines on domestic gardens;
2. irrigation of horticultural crops by recycled water from sewage treatment plants receiving washing machine water as a typical proportion of treated sewage; and
3. discharge of treated sewage effluent into sensitive aquatic ecosystems.

In all cases, the risks associated with laundry detergent constituents were considered higher for greywater irrigation (1) than for irrigation of water in which detergents had been diluted then subjected to sewage treatment processes (2), hence the latter two were not evaluated further.

The AGWR (NRMMC & EPHC 2006) state that one of the most effective measures to minimise the exposure of garden plants and soil to the hazards from greywater reuse is source control. That is, to prevent or minimise the entry of the hazards into the greywater through the selection of detergents that have low concentrations. The other options are: to divert the water from the wash cycle (which would have the highest concentration of the hazards) into the sewage system and then use water from rinse cycles on gardens; or, to dilute the wash cycle water with rinse water and greywater from other sources such as showers, baths and hand basins.

One system that attempts the former is the Australian Eco-labelling program (GECA 2006) which, for example, sets a maximum limit for Na of 140 mg/L in wash water. Another system is the voluntary industry standard for phosphorus (P) which aims to minimise aquatic environmental impacts. For detergents containing P the limit is <7.8 g P per wash (52 mg/L) or 5% P in the detergent (ACSPA 2002), while for so-called ‘P-free detergents’ the limit is <0.5% P in the detergent (i.e. 5.2 mg/L per wash) (ACSPA 2002). However, neither of these systems addresses all of the potential hazards identified by the AGWR (NRMMC & EPHC 2006).

Risks assessed in this evaluation include those to soils and plants where greywater is irrigated, to groundwater beneath irrigation areas, and to runoff from irrigation areas. Health authorities have not encouraged uncontrolled household recycling of greywater for irrigation because of the potential health risks from increased exposure to human pathogens. The current analysis does not explore disinfection properties of laundry detergents nor take account of human health risks. However, with subsurface drip irrigation, or irrigation that is restricted to night-time applications of non-food plants, or with use of disinfection systems, these risks should be manageable.

Given the above situation the aims of this paper are to:

1. assess the risk posed to Australian and New Zealand ecosystems by the use of greywater that contains powder laundry detergents, to water gardens in terms of the inorganic constituents of the detergents;
2. provide an environmental performance rating index that could be used to inform greywater users whether a specific detergent is appropriate to water their garden; and
3. compare the environmental performance of laundry powder detergents with their wash performance as measured by the Australian Consumers' Association (Choice 2005).

METHODS

Laundry detergent samples

The 50 detergents assessed were selected by the Australian and New Zealand Consumers' Associations so that they were representative of the range of powder laundry detergents used in Australia and were purchased from Australian supermarkets in December 2004 and January 2005.

Greywater composition scenarios

Three greywater composition scenarios that represented the various possible compositions of greywater that would be used for irrigation were assessed in this study. These are where greywater is composed solely of water from:

1. The wash cycle of washing machines assuming deionised water was used as the input water.
2. The wash cycle of washing machines assuming mains water with a Ca concentration of 17 mg/L was used. The only difference from scenario 1 is the source and chemistry of the input water.
3. The wash cycle and water from two rinse cycles, all of which have the same volume. The input water was mains water with a Ca concentration of 17 mg/L. It was assumed that all the water was collected before irrigating the garden and therefore that the resulting greywater had one-third of the concentration of detergent constituents in the wash cycle water (scenario 1 and 2).

Creating greywater representing the wash cycle

The three greywater composition scenarios are either water from the wash cycle or a dilution of this. Therefore, it was only necessary to create a solution for each of the 50 powder laundry detergents that represents the scenario 1 wash cycle in order to quantify the physicochemical properties and chemical composition (i.e. quantifying the hazards) of the three greywater scenarios. Using this information the hazards can be determined for the other greywater scenarios by appropriate calculations. The manufacturer-recommended mass of each detergent powder was dissolved in an appropriate volume of reverse osmosis water (Millipore RO) given the recommended type of washing machine (front or top loading). The volume of water used in washing machines was that defined by Choice (2005): that is, front loaders use on average 75 L per wash and top loaders 150 L/wash. These mixtures represented water from the wash cycle only. No clothes were included in the preparation of these solutions so the effect of adsorption on to clothing in the wash was not considered, as this was assumed to have a minimal effect over the whole wash cycle. All collected greywater samples were stored in the dark at 20 °C prior to analysis.

The distribution of risk for each hazard was calculated by determining the percentage of detergents that fitted into each risk level based on their concentration in each detergent and the trigger values (TVs). These were calculated for each hazard in each of the three greywater composition scenarios. Three dilutions were used when determining this distribution. For Ca 17 mg/L 1:9 dilution, two laundry detergents were low risk for all hazards except pH. Buffer capacity of solutions and mix water were not analysed so it was assumed that dilution would not affect pH of the final mixture.

Chemical analysis of the wash cycle greywater

The concentrations of a range of inorganic constituents, pH, alkalinity and conductivity were analysed using standard procedures (*Standard Methods* APHA/AWWA/WEF 2005) in a laboratory that had National Association of Testing Authorities (NATA) accreditation for the methods (Table 1).

Washing assessment

Washing performance was assessed for each of the 50 detergent powders following a standard procedure as outlined by Choice (2005). Scores were given as a percentage.

Table 1 | Limit of detection and analytical methods used in this study

Analyte	Limit of detection	Method^a
Alkalinity	n.a.	T0101-01
Arsenic (total)	<0.001 mg/L	TIC-003
Bicarbonates (total)	<0.5 mg/L	T0101-01
Biological oxygen demand (BOD)	<2 mg/L	T0153-01
Boron (total)	<0.04 mg/L	TIC-003
Cadmium (total)	<0.0005 mg/L	TIC-003
Calcium	<0.1 mg/L	TIC-001
Carbonates (total)	<0.5 mg/L	T0101-01
Chloride	<4 mg/L	T0104-02
Conductivity	<1.0 μS/cm	T0016-01
Copper (total)	<0.001 mg/L	TIC-003
Fluoride	<0.1 mg/L	T0104-02
Iron (total)	<0.03 mg/L	TIC-001
Lead (total)	<0.0005 mg/L	TIC-003
Magnesium	<0.3 mg/L	TIC-001
Manganese (total)	<0.0005 mg/L	TIC-003
Molybdenum (total)	<0.0005 mg/L	TIC-003
Nickel (total)	<0.0005 mg/L	TIC-003
pH	<1.0 unit	T0010-01
Phosphorus (total)	<0.005 mg/L	T0109-01
Potassium	<1.0 mg/L	TIC-001
Residual sodium carbonate (RSC)		By calculation
Selenium (total)	<0.003 mg/L	TIC-003
Sodium	<0.5 mg/L	TIC-001
Sodium adsorption ratio (SAR)		By calculation
Sulfate	<1.5 mg/L	TIC-001
Total dissolved solids (TDS)	<1.0 mg/L	T0016-01
Zinc (total)	<0.003 mg/L	TIC-003

^{a}*Standard Methods* APHA/AWWA/WEF (2005).

Risk assessment framework

The AGWR (NRMMC & EPHC 2006) use a standard qualitative risk assessment approach (Standards Australia 2004a, b). This study follows a similar approach.

The environmental risk assessment framework used in this paper is a tiered process which consists of a hazard assessment followed by determining the environmental performance index for those hazards that posed an unacceptable hazard.

HAZARD ASSESSMENT

The hazards assessed are the inorganic chemicals and physicochemical properties presented in Table 1. A modified version of the hazard quotient (HQ) method (Urban & Cook 1986) was used to assess each hazard. The modifications were that the predicted environmental concentration (PEC) was replaced by the mean concentration of each hazard in wash cycle water (i.e. greywater composition scenario 1) for the 50 laundry detergents and the lowest toxicity threshold value was replaced by the Australian and New Zealand irrigation water short-term trigger values (STVs) (ANZECC & ARMCANZ 2000). The STVs are designed to protect agricultural systems for a period of 20 years. The HQs were calculated by:

$$\text{HQ} = \text{95th percentile value for hazard/STV} \qquad (1)$$

If the HQ was ≥ 1 then the irrigation water STV for that hazard was exceeded; therefore it was considered to pose an unacceptable hazard. If the $\text{HQ} < 1$ then the hazard was considered acceptable. Only those hazards whose HQ values were ≥ 1 (for any detergent) proceeded further in the risk assessment (for all detergents). The hazard posed by chloride was treated separately for soil water and foliar applications. Thus, a total of eight environmental hazards defined in the AGWR (NRMMC & EPHC 2006) (boron, cadmium, carbonates, chloride, pH, phosphorus, salinity and sodium) were included in the calculation of the environmental performance index (EPI).

ENVIRONMENTAL PERFORMANCE INDEX

Typically in ecological risk assessments, cumulative distributions for the sensitivity of species to each hazard (obtained from toxicity data) and a cumulative distribution of the concentration or values of the hazards measured in, or predicted for, the environment are combined to estimate the probability of a certain size hazard occurring and the probability of a certain size biological effect occurring (i.e. the risk). However, in this case the aim was to derive an EPI and a semi-quantitative approach was adopted. The values for each hazard in the 50 laundry detergents for the three greywater scenarios were compared with numerical values of each hazard that should provide various levels of species protection (i.e. TVs).

The BurrliOZ software (Campbell *et al.* 2000) was used to derive the Australian and New Zealand water quality guidelines for toxicants. It can calculate the concentration of any toxicant that should not be exceeded in order to theoretically protect any selected percentage of species from experiencing toxic effects (i.e. the PC*x* where *x* is the % of species to be protected).

The AGWR (NRMMC & EPHC 2006) use five qualitative measures of the magnitude of environmental impact caused by hazards (Table 2). As part of the present study different levels of protection (i.e. different % of species protected) were assigned to these qualitative measures of the magnitude of environmental impact (Table 2). An environmental ranking value from four to zero was then allocated to each of the AGWR measures of impact (Table 2). The smaller the environmental ranking value the greater the potential environmental impact. The category of risk (i.e. low to very high, column 4 of Table 2) associated with each of the hazards was determined for each detergent and the percentage of detergents that fell into each risk category was determined. This was done for greywater composition scenarios 1 to 3.

The EPI for a detergent was calculated by summing the environmental ranking values for each hazard in the detergent and expressing it as a percentage of the maximum possible score of 32 (i.e. the product of the maximum environmental ranking of four and the eight hazards that had $HQ \geq 1$). The EPI was calculated individually for all 50 detergents for all three greywater composition scenarios. Summary statistics (i.e. mean, median, 5th and 95th percentile values and the standard deviation) of the EPI values of the 50 detergents were then calculated for each greywater composition scenario.

Calculation of trigger values for hazards

The irrigation water STVs (ANZECC & ARMCANZ 2000) were not calculated using a species sensitivity distribution (SSD) method and therefore it was necessary to recalculate them to determine TVs that correspond to various levels of species protection. So for those hazards that had a $HQ \geq 1$,

Table 2 | Percentage of species that should be protected from deleterious effects, the corresponding qualitative measure of impact from hazards used in the Australian guidelines for water recycling and their definitions (NRMMC & EPHC 2006)

Minimum percentage of species to be protected from hazard	Qualitative measure of impact	Definitions	Risk outcome	Environmental ranking value if at least the corresponding % of species is protected
95	Insignificant	Insignificant impact or not detectable	Low	4 (95–100)
90	Minor	Potentially harmful to a local ecosystem with local impacts contained on site	Moderate	3 (90–95)
80	Moderate	Potentially harmful to regional ecosystem with local impacts primarily contained on site	High	2 (80–90)
50	Major	Potentially lethal to local ecosystem. Predominantly local, but potential on and off-site impacts	Very high	1 (50–80)
20	Catastrophic	Potentially lethal to regional ecosystem or threatened species. Widespread, on- and off-site impacts	Very high	0 (0–50%)

TVs had to be calculated from first principles using a SSD method.

The BurrliOZ software (Campbell *et al.* 2000) was used to calculate TVs for the eight hazards identified above in the laundry detergents, using the percentiles presented in Table 2. This software was designed to estimate the percentage of species estimated to be protected from toxic effects. This percentage was then matched to the measures of impact described in the quantitative risk analysis matrix used in the AGWR (NRMMC & EPHC 2006). Percentages and their level of impact vary slightly from those used in the Australian and New Zealand water quality guidelines (ANZECC & ARMCANZ 2000) because of the generalised nature of the risk assessment (i.e. not undertaken for a specific site).

Water of a quality typical for a greywater stream was considered as being applied to irrigate gardens during dry seasons. Further scenarios related to greywater irrigation were considered:

1. Four soil types (i.e. sand, sandy loam, loam and light clay) as outlined in AGWR (NRMMC & EPHC 2006) were adopted.
2. Three vegetation classes were adopted, covering all species (including European species), Australian native species only, and a selected composite of both species groups.

To assess the risk posed to plants and soils in Australia, the initial risk assessment described in this paper used TVs (i.e. if these values are exceeded a toxic response is possible) from the AGWR where data are noted for a diverse range of plants and soils (NRMMC & EPHC 2006). These data contained well accepted TVs for a range of plant and soil endpoints (e.g. salinity, phosphorus, boron).

To determine the risk to soil ecosystems and groundwater, several considerations were made:

- Interactions between climates, soils and plants were neglected in undertaking the analyses of scenarios.
- Irrigation rates were used to calculate trigger values for B and P in soils and salinity in groundwater.
- If plant P demand was exceeded then the potential for environmental impact increased.
- Salinity trigger values were determined for each of the four soil types.
- Where threshold values were quoted as a range, the mean of the range was taken, and where they were indicated as ‘less than’ values, 80% of this figure was used as the mean threshold value.
- Nitrogen and hydraulic load (hazards in the AGWR) were not considered as:

  1. Nitrogen losses on storage and irrigation can be significant (Snow *et al.* 1998), and N levels in greywater were not considered sufficient (averages 15 mg/L total N; NRMMC & EPHC 2006) to pose a significant risk.
  2. Volumes of greywater recycled should have a low risk of exceeding hydraulic loads (i.e. causing water logging or overland flow) if the water is distributed around the garden. However, it is recognised that a more complete risk assessment should consider a more detailed evaluation of organic chemicals found in detergents and pathogens in greywater.

Salinity

Figure 1 gives an example of the distribution of the sensitivity of plant species to hazards; in this case it is the distribution of all plant species to salinity in irrigation water. This shows, for example, that 95% of species are protected with an EC of 0.96 dS/m. Table 3 summarises the distribution modelling for four key hazards considered in the paper (salinity, boron, phosphorus and chloride) for specific groupings of endpoints. For example, there were sufficient data to model plant salinity effects on four different plant groups (i.e. ornamentals, agricultural crops, Australian natives and turf grasses). For comparative purposes irrigation water salinity requirements for sandy soil were used. Two further cases were modelled: a composite for all plant groups in sandy soil and this composite of plant groups in a composite of the four soil types considered in NRMMC & EPHC (2006) (sand, sandy loam, loam and light clay).

These data indicated that the Australian native plants are more tolerant to water salinity (TV for insignificant impact 2.17 dS/m) compared with the other plant groups, with corresponding TVs ranging from 1.01 dS/m for turf grasses, 1.07 dS/m for crops in sand to 1.36 dS/m for ornamentals grown in sandy soils. Although turf grasses had the lowest salinity TV for low risk, it had the highest TV for

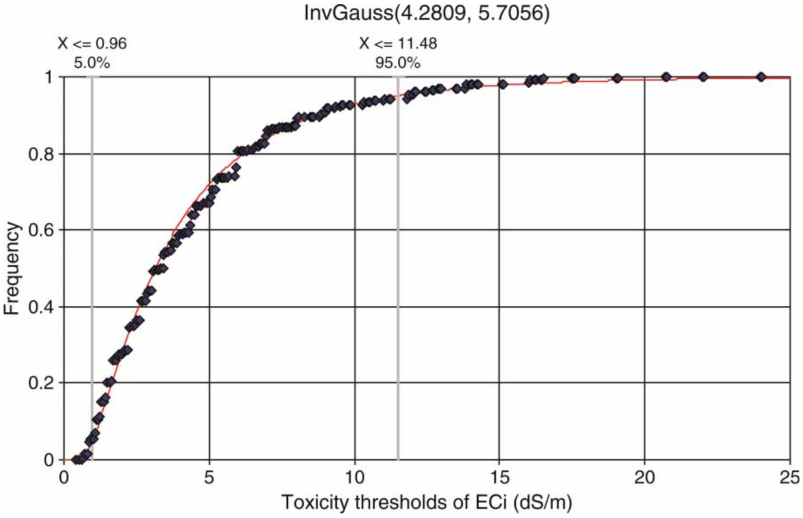

Figure 1 | Cumulative frequency distribution of the electrical conductivity (ECi) of irrigation water that will potentially cause salinity impacts on all plants (native and exotic) (NRMMC & EPHC 2006).

catastrophic impacts, reflecting the range of salinities different turf grass species and cultivars can tolerate (Carrow & Duncan 1998). When all plant and soil types (NRMMC & EPHC 2006) were included the salinity average TVs (Table 3) were lower than sandy soils only TVs. This is because sandy loam, loam and clay soils tolerate less salinity in irrigation water than sandy soils (ANZECC & ARMCANZ 2000).

Boron

Data from two major plant groups (ornamentals and crops) and plants and crops combined were available for determining TVs for B (Table 3). The TVs for ornamental plants were, irrespective of the potential level of impact (i.e. insignificant to catastrophic) more sensitive to B than crop plants (0.5).

Phosphorus

Phosphorus TVs (Table 3) reflect the range of plant requirements for this nutrient. Plant requirements change significantly depending on the species grown, their nutrient requirements and parts of the plants harvested or removed from the area irrigated (i.e. nutrient removal from the system). Once plant requirements are met there is an increased risk of P movement off-site with potential impacts on surface or ground waters. This environmental risk has not been directly assessed in this study as irrigation rates, soil types and rainfall events influence these risks. Therefore, they should be assessed on a site- and location-specific basis. The same logic applies to nitrogen.

Chloride

Chloride TVs (Table 3) vary considerably depending on whether the source of the salinity is soil water or foliar applied irrigation water. These data immediately highlight prevention measures that could be used with sensitive plant species (i.e. surface application of greywater, rather than spraying on to the leaves of the plants). This complies with other reasons for not spraying greywater (i.e. for human health risk from pathogens, spray irrigation of greywater is generally not recommended) (ANZECC & ARMCANZ 2000). The TVs based on the most species for each of EC, B, P and Cl^- were rounded off and used for all further calculations.

Sodicity and carbonates

There were insufficient data to determine TVs for sodium adsorption ratio (SAR), residual sodium carbonate (RSC) and pH using the BurrliOz software. However, well established limiting values for SAR and RSC in irrigation water

Table 3 | Trigger values determined through probability distributions for impact levels and percentiles of specific environmental endpoints protected from hazards in laundry detergents

		Burrlioz output				**Generic plant/soil toxicity data**			**Percentage of specific population (soil or plant) protected**					**Unit**
			Model variables											
									20	**50**	**80**	**90**	**95**	**Percentile**
									Catas-trophic Very high	**Major Very high**	**Moderate High**	**Minor Moderate**	**Insignificant Low**	**(Impact) (Risk[d])**
Hazard	**Observations**	**Distribution type**	**B, alpha or X0**	**C, beta or theta**	**k**	**Effect**	**Types of plant and soil**	**Source table[b]**	**Trigger values**					
ECi	205	BIII	0.25	1.87	70.9	Soil salinity and phytotoxicity	Ornamentals in sand	A5.16	5.49	2.99	1.9	1.57	1.36	dS/m
ECi	107	BIII	0.67	1.95	8.89		Crops in sand	A5.15	4.38	2.42	1.53	1.25	1.07	dS/m
ECi	191	BIII	4.63	2.49	1.48		Australian natives in sand	A5.14	9.58	5.69	3.35	2.73	2.17	dS/m
ECi	74	BIII	9.97	3.48	0.38		Turf grasses in sand	A5.12	10.59	6.17	2.93	1.72	1.01	dS/m
ECi	577	BIII	2.87	2.06	1.64		All plants in sandy soil	A5.12–5.16	7.32	3.39	2.25	1.67	1.29	dS/m
ECi	2,308	BIII	2.54	2.09	1.35		All plant & soil types[a]	A5.12–5.16	5.78	3.08	1.71	1.23	0.93	dS/m
B	40	RW	0.61	1.39		Phytotoxicity	Ornamentals	A5.4	2	0.91	0.5	0.39	0.32	mg/L
B	60	RW	1.04	1.427			Crops	A5.3	2.97	1.33	0.74	0.57	0.47	mg/L
B	100	RW	0.83	1.34			Ornamentals and crops	A5.3, A5.4	2.67	1.15	0.61	0.47	0.39	mg/L
P	369	BIII	4.70	1.88	0.64	Plant toxicity from soil water	All irrigation rates and crops	A5.8, A5.9	7.53	3.32	1.3	0.71	0.4	mg/L
Cl	72	RW	169,598	1.87		Plant toxicity from soil water	Fruit other agricultural crops	A5.18, A5.19	1,405	766	488	403	350	mg/L
Cl	20	RP	700	1.64		Foliar application and direct toxicity to leaves	Range of agricultural crops	A5.17	611	459	262	172	112	mg/L

BurrliOz software used for statistical analyses (Campbell *et al.* 2000, RP = Reciprocal Pareto, BIII = BurrIII, RW = Reciprocal Weibull). [a]sand, sandy loam, loam, light clay; [b]NRMMC & EPHC (2006); [c]Carrow & Duncan (1998); [d]Assumes the likelihood of greywater use is almost certain (NRMMC & EPHC 2006).

applied to soils were related to specific impacts from these hazards.

pH

Trigger values for pH were calculated by digitising data from Anderson *et al.* (2007) which summarised the effects of soil pH on several endpoints (i.e. soil fungi and bacteria, nutrient availability [N, P, K, S, Ca, Mg, Fe, Mn, B, Cu, Zn and Mo] and aluminium toxicity). Graphical representations of minimum and maximum nutrient availability in the pH range 4 to 10 were digitised and the distribution of pH values recorded.

pH 6.87 on a scale of 1 to 14 was used as the 50th percentile. It was taken as the lowest impact (pH 6.8) and increments taken positively and negatively of half standard percentiles to estimate a percentile of the population included in the appropriate pH range.

Estimation of TVs for pH was complicated by the log scale and range of endpoints the pH can impact. However, by combining the rating for all potential impacts (Anderson *et al.* 2007) it was possible to represent the lower and upper TVs for environmental impacts from greywater. These values reflect the impact of the greywater's pH on soil nutrient availability, toxicity to soil fungi and bacteria and aluminium toxicity.

Groundwater protection evaluation

A rigorous generic analysis is not possible for these species, but as will be seen later, the salt concentrations in some detergents are significant and are likely to be a critical hazard in relation to protecting groundwater quality to meet its existing environmental values, as required under the Australian Groundwater Protection Guidelines (ANZECC & ARMCANZ 1995). The water quality criteria are specified in Australian Guidelines for Fresh and Marine Water Quality (ANZECC & ARMCANZ 2000) and the Australian Drinking Water Guidelines (NHMRC & NRMMC 2004).

Environmental values of groundwater that may require protection that have salinity criteria are:

- Drinking water (aesthetic limit 500 mg/L TDS (total dissolved solids) and health limit 1,000 mg/L TDS).
- Irrigation water supplies (limit depends on crop, soil, irrigation method and is site specific).
- Industrial use (limit depends on particular requirements of the industry).
- Ecosystem support (as applies to the aquifer or hydraulically connected ecosystem).

Given the diversity of requirements, the health limit for drinking water was arbitrarily chosen as an indicator target suitable for this generic environmental risk assessment, recognising that this target need not be met everywhere, but it will be a real requirement at some locations. Using assumed rainfall and irrigation rates that span most Australian cities, and typical leaching fractions, the ratio of salt concentration in groundwater recharge to the salt concentration in the greywater irrigation source could be determined using a mass balance method (Cook & Herczeg 1998).

Three climatic zones were considered which were assumed to require 200, 400 and 600 mm/year irrigation during the dry season. Table 4 shows that in the drier climate with higher irrigation application rate, less dilution from rainfall (which is assumed to contribute negligible salt) and lower leaching fraction, the salt concentration in water recharging the aquifer may average six times that of the irrigation water. In the more humid scenario, rainfall

Table 4 | Effect of irrigation, rainfall and leaching fractions relevant for Australian urban areas on the mean ratio of salt concentration in soil water and recharge to that in greywater used for irrigation

Irrigation mm	Rainfall mm	Soil water concentration ratio	Recharge Rate mm	Leaching Fraction %	Recharge Salt concentration ratio[a]
600	400	0.6	100	10	6
400	600	0.4	150	15	2.7
200	800	0.2	200	20	1

[a] Salt concentration ratio of leachate to greywater (= g in Equations (2) and (3)).

dilution is sufficient to compensate for enrichment by evaporation and transpiration.

In all cities mixing of the native groundwater will occur with recharge from areas irrigated with greywater and mains water, and non-irrigated areas. Environmental values ascribed to an aquifer by the Australian Groundwater Protection Guidelines (ANZECC & ARMCANZ 1995) define the water quality objectives for the aquifer including its salinity. It was assumed that the initial groundwater salinity, C_0, may be increased by a fraction, e, while still complying with the water quality objectives. If leachate beneath greywater irrigation contributes a fraction, f, of groundwater in storage, and leachate salinity is a factor, g, greater than that of the greywater applied in irrigation, then the maximum allowable ratio of salinity of greywater (C) to salinity of native groundwater is given by:

$$\frac{C}{C_0} \leq \frac{e+f}{fg} \tag{2}$$

and if $e = 0$

$$\frac{C}{C_0} \leq \frac{1}{g} \tag{3}$$

Table 5 gives examples where in a semi-arid area if there is sufficient contribution to recharge by leachate from greywater irrigation, it is possible that impact on groundwater salinity is more critical than impact on plants or soils. Table 5 shows the maximum acceptable salinity of greywater to adequately protect groundwater where the native groundwater salinity is 500 and 1,500 mg/L. Note that the concentration by evapotranspiration can potentially result in a requirement that salinity of greywater be less than the salinity of native groundwater, although if only a small fraction of recharge is from greywater irrigation leachate, requirements to protect vegetation will be adequate to protect groundwater. For simplicity in the generalised risk assessment that follows, it is assumed that the EC response for all soils and plants correlates strongly with the EC response for groundwater. For this reason no specific metric for groundwater response was evaluated. However, for site-specific risk assessments the approach described above is recommended because groundwater quality protection can be a tighter constraint than soil and plant protection from adverse impacts of high salinity.

Table 5 | Maximum tolerable salinity of greywater (C_{max}) for groundwater protection for various contributions (f) of gross recharge from greywater irrigation areas in a semi-arid area for two examples of native groundwater salinity (C_0)

f	C_{max}/C_0	C_{max} (mg/L)	C_{max} (mg/L)
		$C_0 = 500$	$C_0 = 1,500$
0.001	17	8,500	25,500
0.01	1.8	900	2,700
0.02	1.0	500	1,500
0.1	0.33	165	500

f ratio of recharge from greywater irrigation to gross recharge; C_{max} maximum tolerable salinity of grey water; C_0 native salinity of groundwater. In this table $e = 0.1$ and $g = 6$, as per Equation (2).

Relationship between environmental performance index and washing performance index

In addition, the EPI index values for scenario 1 and 3 greywaters for all 50 laundry detergents were then separately regressed against the washing performance measure (Choice 2005). All statistical analyses were conducted using Genstat (2006).

RESULTS AND DISCUSSION

Analysis of laundry wash water

Summary statistics for the hazards identified in the 50 powdered washing detergents analysed are summarised in Table 6 and are compared with the irrigation water STVs where they exist (NRMMC & EPHC 2006). This recognises that domestic irrigation systems are unlikely to have a life exceeding 20 years and that adopting long-term trigger values (LTVs) would therefore be an unnecessarily onerous objective. Table 6 shows that the 95th percentile of six hazards (i.e. pH, electrical conductivity, SAR, RSC, phosphorus and boron) exceeded the STVs. Comparison of both laboratory data of selected parameters using simple linear regression analysis data averaged a slope of 0.97 ± 0.11 for Na.

Table 6 | Summary statistics of wash performance, environmental rating, cost per wash for 50 laundry detergents and the corresponding values for the chemicals hazards analysed, in water from the first wash. The 95th percentile values were compared with the irrigation water short-term (STVs) trigger values from the Australian and New Zealand guidelines for fresh and marine water quality (ANZECC & ARMCANZ 2000) to determine the hazard quotient

Item	Unit	Mean	Median	Stdev	5th	95th	STV[a]	HQ ≥ 1
Wash performance	Relative	85	90	7.7	74	93		
Environmental ranking	%	42	43	9	27	57		
Detergent: water	g:L	2.3	1.7	1.6	0.9	6.3		
pH		10.8	10.8	0.2	10.4	11.1	6–8.5	Yes
EC	dS/cm	2.67	1.96	1.84	0.95	6.70	0.65–16[b]	Yes
TDS	mg/L	1,590	1,090	1,241	449	4,683		
Ca	mg/L	0.5	0.3	0.6	0.0	1.7		
Mg	mg/L	0.1	0.0	0.4	0.0	1.1		
K	mg/L	0.6	0.0	1.9	0.0	2.9		
Na	mg/L	636	436	499	192	1,852		
SAR[c]	$(\mathrm{mmol_c/L})^{0.5}$	173	129	115	55	379	3[b]	Yes
RSC[d]	meq/L	19	12	14	7	46	0[e]	Yes
$CaCO_3$	mg/L	944	621	679	334	2,294		
HCO_3	mg/L	518	476	240	236	997		
CO_3	mg/L	312	187	309	29	943		
Cl	mg/L	83	7	234	0	664	175[f]	Yes
S	mg/L	303	207	399	1	1,443		
F	mg/L	<0.1					2.0	
P	mg/L	68	57	81	0	245	6.4[g]	Yes
As	mg/L	0.003	0.000	0.008	0.000	0.016	2.0	
B	mg/L	1.0	0.2	5.3	0.0	1.0	7.75[h]	
Cd	mg/L	<0.0005					0. 05	
Cu	mg/L	0.002	0.002	0.002	0.000	0.006	5.0	
Fe	mg/L	0.048	0.032	0.087	0.000	0.299	10.0	
Pb	mg/L	0.000	0.000	0.001	0.000	0.003	5.0	
Mn	mg/L	0.001	0.000	0.003	0.000	0.005	10.0	
Mo	mg/L	0.001	0.001	0.002	0.000	0.003	0.05	
Ni	mg/L	0.002	0.001	0.006	0.000	0.003	2	
Se	mg/L	<0.003					0.05	
Zn	mg/L	0.005	0.003	0.008	0.000	0.015	5	
BOD[i]	mg/L	125	107	119	0	302		

[a]ANZECC & ARMCANZ (2000); [b]Specific to soil, plant and use; [c]SAR = sodium adsorption ratio; [d]RSC = residual sodium carbonate; [e]Carrow & Duncan 1998); [f]the range for the STV is 0 to 350 so the mean value is reported; [g]the range for the STV is 0.8 to 12 so the mean of 6.4 is reported; [h]the range for the STV is 0.5 to 15 so the mean of 7.75 is reported; [i]BOD = biochemical oxygen demand calculated as per NRMMC & EPHC (2006); STV = short-term trigger value for insignificant impact; HQ = hazard quotient.

Sodium and salinity of wash water from the 50 detergents were directly related to the concentrations of detergent recommend by the manufacturer, with P concentrations in the first wash correlated with the amount of detergent used.

The average weight of detergent used per wash in front loading washing machines was 100 ± 8 g compared with 117 ± 64 g for top loading washing machines. However, owing to the lower total volume of wash water used in front loaders for a wash (15 L/wash, excluding rinse water), concentrations of detergents used in front loader washing machines were significantly higher ($p < 0.05$) than top loader washing machines where 60 L/wash (excluding rinse water) was used. Front loader washing machine powders also had significantly higher concentrations of P, Na, SAR and RSC in wash water than top loader washing machines. On average front loaders use ~75 L/full wash cycle and top loaders use ~150 L/full wash cycle.

Environmental rating and ranking of laundry detergents

The relative distribution of these risks for the eight hazards (Table 7) shows that, when washing with good quality water (deionised water) the risks posed by SAR and RSC were very high for all laundry detergents. Similarly, pH, P and EC risks were very high for the majority of detergents (96, 80 and 74% respectively). High risks were posed by B and Cl (both from foliar and soil sources) for a small proportion (6–12%) of detergents; Table 7.

The presence of Ca in wash water and dilution with rinse water (i.e. scenarios 2 and 3) lowered the risk slightly from SAR and RSC risk rating (Table 7). Dilution significantly reduced the risk of EC impacts. Risks associated with other hazards were largely unchanged. This suggests that irrigation of greywater constituted from most of the tested detergents would require remediation strategies to address impacts of SAR, RSC, pH and, for most, also P. For several detergents also EC, B and foliar chloride represent high and very high risks (Table 7).

Table 7 | Distribution of risk categories for eight selected hazards in 50 laundry detergents in greywater composition scenarios 1, 2 and 3

	Percentage of detergents analysed belonging to each risk category for the eight hazards							
Risk category	**EC**	**pH**	**B**	**P**	**Cl–soil**	**Cl–foliar**	**SAR**	**RSC**
Scenario 1								
Low	6	0	92	18	92	88	0	0
Moderate	4	0	2	0	0	0	0	0
High	16	4	0	2	0	0	0	0
Very high	74	96	6	80	8	12	100	100
Scenario 2								
Low	6	0	92	18	92	88	0	0
Moderate	4	0	2	0	0	0	2	0
High	16	4	0	2	0	0	4	0
Very high	74	96	6	80	8	12	94	100
Scenario 3								
Low	74	0	94	20	98	92	2	0
Moderate	0	0	0	4	2	0	10	6
High	18	4	2	0	0	6	60	16
Very high	8	96	4	76	0	2	28	78

EC = electrical conductivity; SAR = sodium adsorption ratio; RSC = residual sodium carbonate.

An overall environmental ranking was determined for each detergent by the addition of the environmental ranking value (Table 8) for each hazard in the detergent. Summary statistics for the environmental ranking of the 50 laundry detergents when they are used to create greywater scenarios 1 to 3 are presented in Table 9. Data are also presented for two additional greywater scenarios: a 1:4 and a 1:9 dilution of scenario 2 greywater. The environmental rankings of the detergents are essentially identical for scenarios 1 and 2. With increased dilution of the laundry detergents (e.g. greywater scenarios 3, 3a and 3b) with rinse water the average environmental ranking increases (an environmental ranking of 100% indicates a low risk for all hazards assessed for a specific detergent). These data indicate that even with considerable dilution, the risks posed by hazards in many detergents are still unacceptable (i.e. risk is high).

Wash performance compared with environmental ranking

Linear regression analysis revealed that there was no significant ($p > 0.05$) relationship between the environmental performances (i.e. the environmental rating index) and wash performance (Figure 2). These data indicate that there were detergents in this study with relatively good wash performance and good environmental performance. This offers some hope for appropriate labelling to result in a shift in the laundry detergent market towards production of more environmentally friendly wash detergents for use when recycling greywater.

Overall environmental performance of powdered laundry detergents

This research highlights that a number of water quality parameters originating in laundry detergents constitute

Table 8 | Summary of trigger values used to determine the environmental risk posed by hazards found in laundry detergent greywater

Hazard	**Unit**	**Trigger values for environmental impacts**[a] **Insignificant**	**Minor**	**Moderate**	**Major**	**Catastrophic**
ECi[b]	dS/m	0.9	1.2	1.7	3.0	5.8
pH[c]	Lower trigger	6.4	5.9	5.0	4.5	4.0
	Higher trigger	7.0	8.0	9.3	10.3	11.4
	50% BurrIII (6.8) ± % BurrIII	10	25	40	45	47.5
B[d]	mg/L	0.4	0.5	0.6	1.2	2.7
P[e]	mg/L	0.4	0.7	1.3	3.3	7.5
Cl[f] (soil water)	mg/L	350	400	500	750	1,400
Cl[f] (foliar)	mg/L	100	170	260	460	600
SAR[g]	$(\text{mmol}_c/\text{L})^{0.5}$	3	6	12	15	20
RSC[h]	meq/L	0	1.25	2.5	5	10
Percentage of population protected from stress from hazard		95	90	80	50	20
Risk if likelihood is almost certain		Low	Moderate	High	Very high	
Environmental ranking value		4	3	2	1	0

[a] If concentration of the hazard does not exceed the trigger value and is higher than the trigger value in the cell to the left.
[b] Rounded off trigger values for electrical conductivity to all plants and all soil types from Table 3.
[c] Calculated from Anderson *et al.* (2007) as described in toxicity data above.
[d] Rounded off trigger values for boron to ornamentals and crops from Table 3.
[e] Rounded off trigger values for phosphorus to all irrigation rates and crops from Table 3.
[f] Rounded off trigger values for chlorine from Table 3.
[g] Table A5.22 (NRMMC & EPHC 2006).
[h] Table 5.5 page 59 (Carrow & Duncan 1998).
Note all impacts are subject to variation with site-specific assessment. Data presented here should be used for a general screening of water quality and risks associated with use of this water for irrigation.

Table 9 | Summary statistics of the environmental rating index scores for all 50 laundry detergents when they are used to create greywater scenarios 1 to 3 (refer to Table 8) and two additional scenarios 3a and 3b where the greywater from scenario 2 is diluted 1:4 and 1:9 respectively

Parameter	**Scenario 1 Deionised water (%)**	**Scenario 2 Ca 17 mg/L (%)**	**Scenario 3 Ca 17 mg/L diluted 1:2 (%)**	**Scenario 3a Ca 17 mg/L diluted 1:4 (%)**	**Scenario 3b Ca 17 mg/L diluted 1:9 (%)**
Average	44	45	61	68	76
Median	44	44	63	69	75
5th percentile	26	26	44	47	61
95th percentile	59	59	75	81	88
Standard deviations	10	11	11	11	8

Note: environmental ranking values from Table 7 were used to determine environmental rating index. The index is the sum of the environmental ranking values for all 50 detergents expressed as a percentage of the maximum possible score of 1,600 (i.e. 50 detergents × 8 hazards × 4 (insignificant impact)).

environmental hazards when irrigating with greywater. Notably in order of priority these are pH, sodium, chloride, phosphorus, electrical conductivity (salinity) and boron, hazards that still require controlling or removing (source control, an exclusion barrier). These constituents may be required to maintain wash performance and removal (an exclusion barrier) as a preventive measure could potentially compromise wash performance. Another preventive measure suggested by the AGWR is an end use restriction barrier, where the user of the greywater must apply on-site

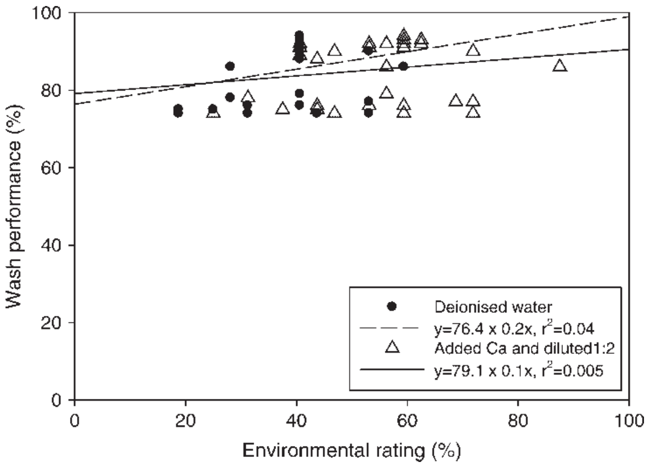

Figure 2 | Relationship between environmental rating (100% = insignificant impact) and wash performance in deionised water (Choice 2005). *Note*: wash performance may vary with different water types. Environmental rating calculated from individual detergent data that were aggregated in Table 6 were ranked for each of 8 hazards shown in Table 8, for waters from scenarios 1 and 3 of Table 7.

restrictions. For example, to manage the high Na concentrations the end user may need to apply gypsum or Ca (Ca amendment) to control soil sodicity (SAR).

Ideally exclusion barriers are most effective from a practical sense in managing risks posed by hazards as they do not rely on actions by the end user (NRMMC & EPHC 2006). In the future the challenge for detergent manufacturers will be to meet increasing demands for high environment standards and low risk while maintaining wash performance. This may be complicated if organic chemicals are used to replace some of the inorganic hazards assessed in this paper.

The next logical step is to include organic hazards in the risk assessment as they are also known to be hazards to the environment (Warne & Schifko 1999; Langenkamp & Part 2001). This risk assessment has been completed using only inorganic hazards. Alkylphenol ethoxylates were also detected at low concentration (μg/L) in only 1 of 50 detergents analysed. In traditional wastewater treatment processes, if the organic compounds used in detergents and surface cleaners are not aerobically and anaerobically degradable, problems of residual levels in biosolids-treated soils may be encountered (Langenkamp & Part 2001). However, detergents are available in some countries which are low in some specified organic compounds. For example, the use of eco-labelled detergents with no linear alkylbenzene sulfonates in Norway have led to much lower concentrations of linear alkylbenzene sulfonate in their biosolids (Langenkamp & Part 2001). Certain levels of organic compounds can be used in Sweden; however, to be considered environmentally friendly a large portion of the organic compounds must be biodegradable (SSNC 2006). Similarly the Australian Voluntary Environmental Labelling Standard states that all surfactants must be readily biodegradable and anaerobically degradable (GECA 2006). If the organic compounds used to replace inorganic hazards are completely degradable and their breakdown products do not pose a risk to the environment, then a laundry detergent which is environmentally friendly for greywater use is possible.

This research also highlights that current methods for assessing environment friendly detergents are inadequate, especially if considerable volumes of greywater are recycled on Australian and New Zealand gardens. The use of P and Na as the only hazards controlled in environmentally friendly detergents would not lower all environmental risks to acceptable levels as defined in the AGWR (NRMMC & EPHC 2006). The TV suggested by the AGWR for the sodium eco-labelling program (GECA 2006) which sets a maximum limit for Na (i.e. 21 g/150 L or 140 mg/L) and total phosphates as sodium tripolyphosphate, shall not exceed 25 ml/L at the recommend dosage (If this $Na_5P_3O_{10}$ is as a salt then this is equivalent to 35 mg/L P). The industry voluntary standard for P to minimise aquatic environmental impacts set by ACCORD is <7.8 g P per wash (i.e. 52 mg/L assuming 150 L/wash-rinse cycle) or 5% P in the detergent, and for a 'no P' logo is <0.5% P in the detergent (5.2 mg/L in a wash-rinse cycle) (ACSPA 2002). With more front loading machines giving enriched wash concentrations and the new guidelines for greywater use in Australia, these values may no longer be appropriate to protect aquatic and terrestrial environments.

Total P concentration in laundry wash water averaged 68 ± 81 mg/L (Table 6); for the total wash (wash–rinse) these values would be diluted to 33%. This suggests most would comply with the voluntary industry standard. However, 22 mg P/L in the complete wash cycle would be catastrophic with respect to oversupply of P to plants and increase the risk of off-site impact. These data should be moderated with the sorption capacity of the soil for P, which is site specific. However, it highlights the potential for excess P to enter the environment when irrigating with greywater.

Sodium concentrations in wash water averaged 636 ± 499 mg/L (Table 6); if this is diluted to 150 L with rinse water the average Na concentration would be 191 mg/L. On average this value exceeds the eco-labelling maximum limit of Na by 51 mg/L. These data indicate many of the powered laundry detergents tested in this study would not meet this standard. When these Na concentrations are converted to SAR values, the average SAR is 173 ± 115, exceeding the catastrophic impact TV (20, Table 8) by at least eight times. Even on dilution with rinse water and assuming a typical Ca concentration in source water, 98% (SAR in Table 7) for detergents, still posed unacceptable risks on soils containing dispersive clays. These data also indicate that in many scenarios SAR values will not meet environmental protection measures identified for greywater recycling in AGWR. These data should also be moderated with the fact that Ca concentrations in the feed water can influence SAR calculations, also water SAR response to soils are moderated by the soil types and the soil ameliorant applied (discussed above).

Another environmental endpoint requiring consideration in AGWR is groundwater. In this evaluation, it is assumed that the hazard posing the tightest constraint is the salinity of greywater used for irrigation. Salt may be regarded as conservative, in that very little is taken up by plants and none is lost by evaporation or transpiration or surface runoff. It is assumed not to be sorbed onto soils and will be mobile in the soil profile in a downwards direction when there is a downward hydraulic gradient in the soil profile. It can also move upwards due to upward hydraulic gradients near the soil surface and in the root zone due to evaporation and transpiration. It will be deposited close to the surface at the point where soil water vaporises during evaporation or around the root where it is unable to enter because of an osmotic gradient. The precipitated salt is available for dissolution and leaching whenever rainfall or irrigation next occurs. Because only a fraction of the irrigation water leaches, it ultimately contains all the salt that has been applied and so the concentration in water recharging an unconfined aquifer will be considerably higher than the concentration in the (greywater) irrigation source, especially in drier climates where there is little dilution by rainfall.

While other considerations such as leaching of nitrate, metals or water with high biological oxygen demand could be considered, they participate in many more biogeochemical reactions, sorption or biodegradation on the soil surface, within the root zone and through the unsaturated zone.

Groundwater quality protection requirements for greywater irrigation will depend on local soil, climate and irrigation management. The evidence presented suggests that in many cases greywater irrigation on a widespread scale would be incompatible with policies that prevented degradation of the groundwater resource. Site-specific investigations are warranted to assess the variability of greywater irrigation in urban areas, and determination of blend-ratios with rainwater to achieve adequate groundwater protection.

CONCLUSION

The concentrations of seven environmental parameters (EC, pH, B, N, P, Cl, Na) commonly found in laundry detergents, were assessed by using the AGWR and assuming the greywater would be used as wash water only or total wash cycled water (wash and rinse water combined) from laundry washes directly on to the garden. No laundry detergent achieved an environmental ranking of 100% (i.e. low risk for all hazards). Overall environmental rankings for the 50 detergents analysed average 44 ± 10% for wash only and 61 ± 11% for wash and rinse (assuming 17 mg/L of Ca in source water).

Most laundry detergents tested passed the voluntary concentration for P set by the industry (ACSPA 2002) but exceeded a voluntary maximum Na concentration (GECA 2006). However, there was one detergent in this study with relatively good wash performance and good environmental performance (environmental ranking >80%). These data indicate that there may be more environmentally friendly ways to produce laundry detergents for greywater recycling or aerobic on-site treatment and recycling systems.

Future work is required to assess:

- nitrogen concentrations in laundry detergent;
- organic chemicals found in laundry detergent;
- the range of other detergents used in domestic households that may enter greywater systems;

- appropriate policies for groundwater protection that account for greywater irrigation;
- environmental performance of liquid detergents and other cleaning agents, soaps and personal care products that enter greywater systems.

The research suggested above, combined with an appropriate labelling system, should allow greywater users to make appropriate choices (i.e. source control preventive measures) for the detergent products they use, ensuring hazards found in greywater that have an impact in the urban environment are minimised. Suitability of detergents at specific sites, with defined soil types, vegetation, hydrology and groundwater conditions and irrigation management systems can be determined; however, the generic approach adopted here is likely to lead to a similar ranking for each detergent in the evaluated cohorts.

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