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Presence and fate of priority substances in domestic greywater treatment and reuse systems

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Abstract

A wide range of household sources may potentially contribute to contaminant loads in domestic greywater. The ability of greywater treatment systems to act as emission control barriers for household micropollutants, thereby providing environmental benefits in addition to potable water savings, have not been fully explored. This paper investigates the sources, presence and potential fate of a selection of xenobiotic micropollutants in on-site greywater treatment systems. All of the investigated compounds are listed under the European Water Framework Directive as either "Priority Substances" (PS) or "Priority Hazardous Substances" (PHS). Significant knowledge gaps are identified. A wide range of potential treatment trains are available for greywater treatment and reuse but treatment efficiency data for priority substances and other micropollutants is very limited. Geochemical modelling indicates that PS/PHS removal during treatment is likely to be predominantly due to sludge/solid phase adsorption, with only minor contributions to the water phase. Many PS/PHS are resistant to biodegradation and as the majority of automated greywater treatment plants periodically discharge sludge to the municipal sewerage system, greywater treatment is unlikely to act as a comprehensive PS/PHS emission barrier. Hence, it is important to ensure that other source control options (e.g. eco-labeling, substance substitution, and regulatory controls) for household items continue to be pursued, in order that PS/PHS emissions from these sources are effectively reduced and/or phased out as required under the demands of the European Water Framework Directive.

Keywords: Organic pollutants; Micropollutants; Water Framework Directive; Greywater; Priority substances; Sludge

1. Introduction

As pressures on potable water supplies continue to increase, so too do the incentives for wastewater reuse and recycling. One option increasingly explored, particularly in water-stressed nations, is the on-site treatment and reuse of household greywater for non-potable purposes such as toilet flushing and garden irrigation. Greywater (used here to refer to domestic wastewater from all sources except toilets, bidets and urinals) typically has a lower pathogen content and organic matter load than combined household wastewater which also contains toilet waste. On-site source separated treatment and reuse of this less polluted wastewater fraction is therefore attractive from a risk management perspective. However, it must also be recognised that domestic greywater can be highly variable in composition, being heavily dependent on the dynamics and behaviour of the households' occupants, and thus reflecting the inhabitants' age distribution, lifestyles, water use tendencies, and consumer choices (e.g. choice of cleaning and personal care products, choice of shower head etc.). The different greywater fractions (i.e. bathroom, laundry and kitchen greywater) also vary in composition (see Table 1 for examples of published values for standard water quality parameters), with different plumbing fixtures (shower, dishwasher, kitchen sink etc.) not only generating different quantities of wastewater, but also differing in terms of characteristic pollutant sources and loads (Friedler, 2004). For example, due to the presence of perishable food waste, kitchen wastewater generally has a higher organic matter loading than other greywater sources and is often omitted from domestic greywater reuse schemes as a result. Existing greywater treatment systems show a wide range of design and sophistication, from simple single-household soil filter systems to community-scale multi-stage rotating biological contactors (RBC) and membrane bioreactors (MBR), but all are based on chemical, physical, and biological processes such as settling, filtration, adsorption, aeration, precipitation,

aerobic/anaerobic digestion, and disinfection. Eriksson et al. (2009) have fully described the structure of a greywater treatment system incorporating a three stage RBC.

Table 1. General characteristics of greywater from different household sources.

Chemical/physical property	Bathroom[a]	Laundry[b]	Kitchen[c]	Mixed greywater[d]
pH	6.4–8.6	8.1–10		5–8.7
TSS (mg l^{-1})	7–207	120–280	235–720	15–112
BOD (mg l^{-1})	26–300	48–380	47–1460	41–500
COD (mg l^{-1})	100–633	725–1815	644–1380	283–549
Total N (mg l^{-1})	3.6–17	6–21	40–74	0.6–11
Total P (mg l^{-1})	0.1–> 49	0.1–> 101	68–74	0.6–> 68

[a] Bathroom: Almeida et al., 1999, Burrows et al., 1991, Christova-Boal et al., 1996, Laak, 1974, Ledin et al., 2006, Nolde, 1999, Rose et al., 1991, Siegrist et al., 1976 and Surendran and Wheatley, 1998.
[b] Laundry: Almeida et al., 1999, Christova-Boal et al., 1996, Laak, 1974, Siegrist et al., 1976 and Surendran and Wheatley, 1998.
[c] Kitchen: Almeida et al., 1999, Günther, 2000, Laak, 1974, Siegrist et al., 1976 and Surendran and Wheatley, 1998.
[d] Mixed: Palmquist and Hanæus, 2005, Casanova et al., 2001, Gerba et al., 1995, Hypes, 1974, Santala et al., 1998, Rose et al., 1991 and Jeppesen, 1993.

Greywater treatment and reuse offers the potential to substantially reduce domestic potable water demand, but care must be taken to ensure this is achieved without detriment to public health and the environment. To date, most studies investigating greywater reuse and associated risks have focussed on conventional water quality monitoring parameters such as those in Table 1 (e.g. biochemical oxygen demand, BOD, chemical oxygen demand, COD, total suspended solids, TSS, nutrients, and microorganisms) with very few studies of the nature, loads and dynamics of micropollutants. Ledin et al. (2006) highlighted the urgent need for broad, well-defined monitoring programmes to fill this knowledge gap, particularly given that initial research had identified as many as 900 xenobiotic organic compounds and compound groups (XOCs) which are commonly used in bathroom and laundry products and hence potentially present in greywater (Eriksson et al., 2002). Subsequent screening of bathroom greywater from a Danish apartment block confirmed the presence of almost 200 such XOCs, including surfactants, fragrances, preservatives, antioxidants, plasticisers, UV-filters, and solvents, a number of which were also suspected endocrine disruptors (Eriksson et al., 2003). With uptake of greywater reuse technologies likely to increase in many areas over the coming decade and dedicated greywater quality guidelines either lacking or still in development in most countries, it is imperative to ensure that greywater reuse risk assessments are thorough and fully accountable. Available data indicates that on the basis of conventional water quality parameters greywater can be successfully treated for non-potable reuse. Further attention is now required to ensure that treatment trains are also optimised as far as practicable for the removal of potentially hazardous micropollutants (particularly those which may form aerosols during toilet flushing or may build up in soils if water is reused for garden irrigation or other outdoor applications). A best case scenario would see greywater treatment and reuse systems not only resulting in significant potable water savings but also limiting the release of household derived water pollutants and thus improving the quality of

combined municipal wastewater (and other receiving environments) by acting as an efficient pollutant barrier. Unfortunately, the current knowledge gaps regarding the presence and fate of micropollutants in greywater make it difficult to adequately determine the level of risk associated with non-standard water quality parameters, and the potential added value of greywater treatment as a micropollutant barrier has effectively remained untested to date.
In this paper, the current state of knowledge with respect to the presence in greywater of a selection of XOCs listed either as Priority Substances (PS) or Priority Hazardous Substances (PHS) under the European Water Framework Directive (WFD) (Directive 2000/EC/60) is presented. These substances have been earmarked for significant reduction (and in the case of PHS for complete cessation) of discharges, emissions and losses to receiving water environments throughout the European Union (EU). Here we address their potential phase distribution and possible fate in greywater treatment and reuse systems, with the primary goal of considering whether the installation of household greywater treatment and reuse systems may be of benefit to EU member states in the quest to reduce PS/PHS emissions. A combination of geochemical modelling and greywater treatment and reuse scenario analyses is used to investigate the potential implications of more widespread deployment of household greywater treatment systems, with a particular emphasis on the potential benefits (or shortcomings) in relation to micropollutant treatment efficiency and pollution control.

2. **Methods**

2.1. Presence of PS/PHS in greywater

A comprehensive review of existing and emerging technologies relevant to greywater treatment, and the presence, concentrations, and removal efficiencies of PS/PHS in greywater systems has been conducted. This review covered the period from 1974 to 2009 and was conducted using scientific literature, government reports and online databases. As the published data were found to be very limited, additional sources of information indicative of the likely presence of PS/PHS in household wastewater (e.g. the United States Department of Health and Human Services "Household Products database" (US DHSS, 2009) and a wide range of material safety datasheets for consumer products were also consulted.

The urban sources of the pollutants listed in the WFD are very extensive with transport, construction sites, waste disposal and households having been identified as major contributors (Holten Lützhøft et al, 2008). With respect to household releases to greywater, quantitative data exists for a number of processes involving benzo(a)pyrene, cadmium, mercury, nickel and trichloroethylene. Pollution sources have also been identified for dichloromethane, endrin, lead, nonylphenols and tributyltin but quantitative release data are not currently available.

2.2. Environmental partitioning and fate

In the absence of specifically tailored greywater treatment models, geochemical modelling applications within EPI Suite™ v3.20 (US EPA, 2007) were used to predict the likely environmental partitioning and fate of the selected PS/PHS in both an aquatic (river/lake) environment (LEVEL3NT–Mackay Level III Fugacity model) and in a conventional activated sludge sewage treatment plant fugacity model (STPWIN). In addition, biodegradation data were sourced from a database collating the key physico-chemical properties of the PS/PHS identified in the WFD (Holten Lützhøft et al., 2007).

The EPI (Estimation Programs Interface) Suite™ tool is a screening level tool developed by the United States Environmental Protection Agency's Office of Pollution Prevention and Toxics and Syracuse Research Corporation. The tool is based on estimation methods which have been developed by government, academic and private sector researchers over many years and which can be used to obtain a cursory insight into the environmental fate of a wide range of pollutants. EPI Suite includes a database of Chemical Abstracts Service (CAS) numbers with associated SMILES (Simplified Molecular Input Line Entry System) notation that can be used for direct import into the EPI Suite interface program that was used in this work. The interface program uses the SMILES notation to run 10 separate structure estimation programs in succession. It then executes the LEVEL3NT and STPWIN modelling applications by transferring the molecular weight, the Henry's law constant, log octanol–water partitioning coefficient and various volatilisation parameters to the models. The LEVEL3NT model predicts substance partitioning between air, water, soil and sediment in a default environment based on the estimated inherent properties of the selected substance and assuming steady state conditions. The model is described in detail in Mackay et al., 1996a and Mackay et al., 1996b. STPWIN predicts the relative importance of biodegradation, sorption, and air stripping in accounting for substance removal in an activated sludge treatment plant. It is a version of the Toronto model originally outlined in Clark et al. (1995).

2.3. Scenario analyses

Scenario analyses have been carried out to demonstrate the potential impacts of greywater treatment and reuse on the wider urban water cycle, including the impacts on wastewater volumes. A range of scenarios have been included. They differ both in the type of greywater being treated (i.e. bathroom, kitchen, and laundry) and in the type of reuse. Scenarios incorporating greywater reuse for irrigation have also been included as this is a very relevant option in warm climates and water-stressed locations. For the purposes of these calculations, input data to the scenario analyses were based on the Danish water use statistics provided below. It is acknowledged that fugitive uses of household chemicals and leaks could influence the overall water balance but in the absence of literature data it has not been possible to include these contributions.

Danish potable water consumption: 119 l $person^{-1}$ day^{-1} (DANVA, 2007).
Proportion of municipal WWTP influent derived from households: 43% (DANVA, 2007).
Proportion of household water (Kjellerup and Hansen, 2007) used in:

Bathrooms: 35–37 (36)%;

Laundries: 13–15 (14)%;

Kitchens: 17–25 (21)%;

Toilet flushing: 20–27 (23)%;

Irrigation: 5–7 (6)%

3. **Results**

3.1. Priority substances in greywater

The results of an extensive literature review investigating the presence and/or concentrations of the selected PS/PHS in greywater are presented in Table 2. It is clear from the limited number of results that monitoring of these pollutants in greywater has thus far been minimal and mainly concentrated in Scandinavia. There are very few studies reporting PS/PHS

Table 2. Summary of data relating to the presence and concentration of organic PS/PHS in raw, untreated, greywater (all values in µg l^{-1}).

Substance name	Treatment plant and location					
	Nordhavnsgården, Denmark[a]	BO90, Denmark[b]	Gals Klint Campingsite, Denmark[c]	Vestbadet I/S, Denmark[a]	Vibyåsen, Sweden[d]	Gebers, Sweden[a] [nd]
Benzene and PAHs						
Benzene		< 1.9				
Naphthalene		< 4.5				0.036 (0.029–0.042)
Anthracene						0.032 (0.023–0.041)
Fluoranthene					0.03	0.034 (0.033–0.035)
Benzo(a)pyrene					0.03 (0.02–0.04)	< 0.01
Benzo(g,h,i)perylene					nd-0.04	< 0.01
Indeno(1,2,3-cd)pyrene						< 0.01
Benzo(k)fluoranthene						< 0.01
Benzo(b)fluoranthene						< 0.01
Chlorinated aliphatics						
Methylene chloride	< 1	< 1.0				
Chloroform	< 0.03 (< 0.1)	< 0.1–250				
Carbon tetrachloride	< 0.02 (< 0.1)	< 0.1–1				
Ethylene chloride	< 0.1	< 0.5				
Chlorinated alkenes						
Trichloroethylene	< 0.02	< 0.1	< 0.050			
Tetrachloroethylene	< 0.02	< 0.1	< 0.050			
Phenols						
Pentachlorophenol	< 0.05	< 0.02–0.04	< 0.050	< 0.05		
Octylphenols	< 0.5	< 0.25	< 0.1	< 0.5		
para-tert-octylphenol		0.2			0.11 (0.08–0.16)	0.10 (0.07–0.15)
Nonylphenols	0.5–0.6	0.5	0.76	0.9		
4-para-nonylphenol					3.8 (2.82–5.95)	0.76 (0.56–1.1)
Organometallic compounds						
Tributyltin cation					0.248 (0.209–	0.004 (0.002–

					0.287)	0.006)
Other						
DEHP	15–16	9.8–39	14	28	57.6 (8.4–160)	15.2 (7.5–20)
Pentabromodiphenylether					0.33 (0.17–0.76)	0.011 (0.0048–0.018)

[a] Andersson and Dalsgaard (2004); [b] Ledin et al., 2006, Eriksson et al., 2003 and Larsen, 2006; [c]Nielsen and Pettersen (2005); [d] Palmquist (2004); [e] Palmquist and Jönsson (2003).

concentrations in raw greywater and only one published study (Nielsen and Pettersen, 2005) reporting both inlet and outlet concentrations relative to a treatment system. Nevertheless, some interesting results have been reported and key findings are summarised below:

- For a treatment plant connected to a Danish housing complex, BO90, with 36 inhabitants (Eriksson et al., 2003, Larsen, 2006 and Ledin et al., 2006), chloroform concentrations up to 250 $\mu g\, l^{-1}$ were recorded in influent greywater. Further investigation indicated that this could be related to the use of household bleach (Ledin et al., 2006) with chloroform being produced through the haloform reaction of chlorine (from bleach) with organic matter (Ivahnenko and Zogorski, 2006).
- Octylphenols (OPs) and nonylphenols (NPs) were detected at most of the treatment facilities despite being banned in both Sweden and Denmark where the reported measurements were taken. The highest recorded value was 5.95 $\mu g\, l^{-1}$ for 4-(para)-NP (Palmquist, 2004). Recent research has shown that these substances are frequently present in imported textiles which then act as emission sources (Höök, 2007 and Testfakta, 2007). Hence, complete source control may not be achievable even though the household use of specific pollutants has been prohibited.
- Di(2-ethylhexyl)phthalate (DEHP) was consistently found in influent greywater, with concentrations ranging from 7.5 to 160 $\mu g\, l^{-1}$ (see Table 2 for details). DEHP is widely used as a plasticiser and may be sourced partly from PVC plumbing fixtures. Other household sources may be linked to its use in cosmetics, perfumes, lacquers, paints, pesticides, and printing inks for paper, plastics and textiles (Økland et al., 2005).
- Pentabromodiphenylether (PBDE) was measured and detected in 2 Swedish greywater treatment plants (Palmquist, 2004 and Palmquist and Jönsson, 2003). The highest recorded concentration was 0.76 $\mu g\, l^{-1}$. The use of penta- and octa-bromodiphenylethers has been restricted in the EU since 2003 (Directive 2003/11/EC: Annex 1, Issues 44 and 45) and their use in new electrical and electronic equipment has been banned since July 2006 (Directive 2002/95/EC). However, these substances are likely to persist in households for many years due to previous extensive use in a broad range of products with extended service life such as computer casings, clothing, upholstery, and construction materials.
- Tributyltin (TBT) was also present in Swedish greywater at the Vibyåsen and Gebers treatment plants (Palmquist, 2004 and Palmquist and Jönsson, 2003). Although the use of TBT biocides (formerly the major use) in all applications should have ceased throughout Europe (Biocides Directive, 98/8/EC), it has previously been reported as occurring in products including tile adhesives and bath caulk (US NIH HPD, 2007). Slow ongoing release from previous applications, such as these, could thus explain the presence of TBT in greywater. It is also possible that imported items continue to be a

source of TBT (used for its antifungal and biocidal properties in laundry sanitisers, hard surface disinfectants and textiles). Contaminated clothing/skin of industrial workers or shipyard workers may also still act as a source of TBT to greywater as this substance was previously extremely widely used as an anti-fouling agent and removal of TBT containing paints from boat and ship hulls is still occurring.

Due to the limited availability of relevant greywater monitoring data, indirect sources of information indicating potential household uses of PS/PHS were also consulted to help determine the likelihood of their presence in domestic greywater. Consultation of the United States Department of Health and Human Services "Household Products Database" (US DHSS, 2009) revealed that although many of the PS/PHS (16 of the 29 organic PS/PHS initially listed in the WFD) were not represented in the database, others were listed as ingredients in a wide variety of products. Although is focused specifically on products for sale on the US market, the database it nevertheless contains useful information regarding the types of products in which PS/PHS may typically be found, including information about ingredients in household products for automobiles, hobby craft, home maintenance, gardening, pest control, indoor use, and pet care. Several of the PS/PHS recorded in the database are present in solvent-based products. For example, benzene, naphthalene, dichloromethane, and chloroform are all listed as being present in various home and automobile cleaning and degreasing products, adhesive removers, paint/varnish removers, and fabric and textile cleaners. The other major PS/PHS usage group indicated in the database includes naphthalene, diuron, alachlor, simazine, atrazine, chlorpyrifos and trifluralin, which are all recorded as ingredients in household pesticides, herbicides, insecticides, algaecides etc. Some of the biocides are no longer authorised for use in products in Europe but may still be present as residues in imported goods and may also be released from long-lasting household goods (e.g. bathroom tile caulking). Unfortunately, some categories of products that are of particular relevance to greywater (e.g. personal care products) are currently not well represented in the database. It should also be noted that not all substance uses in the USA will be relevant in Europe where many of the listed PS/PHS are already heavily regulated.

3.2. Environmental partitioning and fate

The results obtained from the environmental partitioning and fate modelling are presented in Table 3. Although it would be preferable to use models tailored specifically to match greywater composition and treatment conditions, in the absence of suitable models these results do provide some insight into the potential partitioning behaviour of PS/PHS in both soil/wetland based greywater treatment systems and other more mechanised on-site treatment plants. Both the level III fugacity modelling (LEVEL3NT) results and sewage treatment plant (STPWIN) modelling results indicate that for the majority of substances investigated (i.e. 29 out of 33) removal during water treatment is likely to be predominantly due to partitioning to the solid/sludge phase. Only benzene, chloroform, ethylene chloride, and dichloromethane were indicated to differ from this trend, with STPWIN results predicting 7–18% partitioning to the solid phase and the remainder being relatively equally distributed between the aqueous and gaseous phases.

Table 3. Phase distribution modelling results (EPI Suite v3.20, US EPA, 2007) and biodegradability data (Holten Lützhøft et al., 2007) for the selected PS/PHS.

Substance name	CAS no.	STPWIN: Percentage adsorbed to sludge (Total % removed)	Level III fugacity model: Phase distribution (%)			Biodegradation[b]	
			Air	Water	Solid phase[a]	Aerobic	Anaerobic
Alachlor	15972-60-8	13.3 (13.5)	0.0	10.2	89.7	P	
Anthracene	120-12-7	52.5 (54.2)	0.2	9.7	90.1	P–R	P
Atrazine	1912-24-9	3.3 (3.5)	0.0	13.7	86.2	P	
Benzene	71-43-2	1.1 (68.9)	37.6	48.1	14.3	P–R	P
Benzo(a)pyrene	50-32-8	91.9 (92.6)	0.0	2.5	97.5	P–R	P
Benzo(b)fluoranthene	205-99-2	90.2 (90.9)	0.1	3.6	96.2	P–I	
Benzo(g,h,i)perylene	191-24-2	92.8 (93.6)	0.0	1.8	98.2	P–R	
Benzo(k)fluoranthene	207-08-9	91.8 (92.6)	0.0	2.6	97.4	P–I	
Chlorfenvinphos	470-90-6	21.9 (22.2)	0.0	10.7	89.3	P–R	
Chloroform	67-66-3	1.1 (59.8)	42.8	43.2	14.0	P–R	R
Chlorpyrifos	2921-88-2	75.7 (76.4)	0.0	4.5	95.5	P–R	
DEHP	117-81-7	93.2 (94.0)	0.3	3.8	95.9	P–R	P
Diuron	330-54-1	3.6 (3.7)	0.0	14.6	85.5	P–I	P
Endosulfan	115-29-7	22.4 (25.2)	0.3	5.0	94.7	P	
Ethylene chloride	107-06-2	1.4 (34.0)	37.0	44.5	18.4	P–R	P–R
Fluoranthene	206-44-0	81.4 (82.2)	0.4	7.6	92.1	P–R	P
Hexachlorobenzene	118-74-1	88.0 (91.1)	0.5	1.3	98.1	P–I	R
Hexachlorobutadiene	87-68-3	51.9 (88.9)	2.7	3.5	93.8	R	R
Hexachlorocyclohexane	608-73-1	36.4 (37.0)	0.6	6.1	93.2	P–R	P–R
Isoproturon	34123-59-6	4.6 (4.7)	0.0	13.4	86.6		
Dichloromethane	75-09-2	1.0 (56.9)	46.0	47.1	7.0		
Naphthalene	91-20-	8.3 (23.6)	1.0	12.8	86.2	P–I	P–R

Substance name	CAS no.	STPWIN: Percentage adsorbed to sludge (Total % removed)	Level III fugacity model: Phase distribution (%)			Biodegradation[b]	
			Air	Water	Solid phase[a]	Aerobic	Anaerobic
	3						
4-nonylphenol	104-40-5	90.0 (90.8)	0.3	9.5	90.3	I	
Para-tert-octylphenol	140-66-9	84.0 (84.8)	0.2	9.1	90.8	P	P
Pentabromodiphenylether	32534-81-9	93.0 (93.8)	0.2	0.9	98.9	P	
Pentachlorobenzene	608-93-5	79.5 (83.6)	1.0	2.2	96.7	P	R
Pentachlorophenol	87-86-5	80.5 (81.2)	0.0	2.8	97.2	P–I	P–R
Simazine	122-34-9	2.4 (2.5)	0.0	19.6	80.4	P–I	
Tributyltin cation	36643-28-4	74.6 (97.3)	0.4	6.0	93.6	P–I	
1,2,4-trichlorobenzene	120-82-1	26.4 (53.1)	4.0	7.3	88.8	P–I	P
Trifluralin	1582-09-8	84.9 (86.0)	0.1	2.6	97.3	P–I	R
Indeno(1,2,3-cd)pyrene	193-39-5	92.9 (93.7)	0.0	1.7	98.2	P–I	
C10-13-chloroalkanes	85535-84-8	59.2 (98.2)	2.8	11.0	86.2	P	

[a] Solid phase = soil and sediment; [b] P = persistent (< 20%), I = inherently biodegradable (20%–70%), R = readily biodegradable (> 70%).

The relevant biodegradation data are also summarised in Table 3 and indicate that many of the substances are likely to be persistent in the environment. Where a broad range of reported values exists, the biodegradability is reported accordingly e.g. P–R in Table 3 corresponds to reported results indicating persistent to readily biodegradable properties. The apparent range in susceptibility to biodegradation can be attributed largely to variations in experimental conditions as results from both field and laboratory studies have been included. Overall, the biodegradation data support the suggestion that the majority of organic PS/PHS are unlikely to be substantially affected by bioprocesses during treatment, but are instead predominantly affected by phase distribution and related processes such as sorption and volatilisation. This will certainly apply in the case of systems with relatively short hydraulic retention times and systems containing young sludge and biofilms that have not yet adapted to the presence of the relevant xenobiotic substances. These results were also supported by STPWIN predictions that indicated the role of biodegradation as a PS/PHS removal process during activated sludge treatment to be much lower than that of sludge adsorption (results not shown).

3.3. Scenario analyses

A selection of greywater reuse scenario analyses are presented below which have been based on typical reuse scenarios found in the literature (Christova-Boal et al., 1996 and Dixon et al.,

1999; Nolde et al., 1999; Friedler, 2004, Andersson and Dalsgaard, 2004, Pidou et al., 2007 and Li et al., 2009). In the associated calculations 100% implementation of the specified treatment and reuse technology has been assumed. Whilst this may be a future reality for new residential developments, uptake is likely to be substantially lower in more established areas where retrofitting of treatment systems and dual reticulation plumbing would be necessary. It should also be noted that Danish potable water use is relatively low in comparison with many other developed nations, so the potential water savings could well be higher in other areas given sufficient reuse opportunities.

3.3.1. Scenario 1: No greywater reuse

This scenario represents the baseline condition most common in Europe at the present time. Greywater is not treated separately or recycled but discharged directly to the sewer system and transported to a municipal wastewater treatment plant (WWTP). This means that 94% of the water used for domestic purposes (111.9 l $person^{-1}$ day^{-1}) is discharged directly to the sewer system and 6% is used for irrigation (7.1 l $person^{-1}$ day^{-1}).

3.3.2. Scenario 2: Greywater treatment option 1

Bathroom greywater is treated on-site by RBC and reused for toilet flushing. On this basis, it is calculated that 42.8 l $person^{-1}$ day^{-1} of greywater (36% of total potable water used) is treated onsite. Of this volume, 27.4 l $person^{-1}$ day^{-1} can be reused for toilet flushing before being released to the sewer. The surplus treated greywater of 15.4 l $person^{-1}$ day^{-1} could potentially be diverted for other applications such as irrigation rather than being released to the sewer. In a realistic scenario only the amount of water required for specific reuse applications by the community or household hosting the system would actually be diverted for treatment in the system. However, based on the input data used for these analyses a surplus is generated as the average bathroom greywater produced is in excess of the volume required for toilet flushing. Using the average Danish water use input parameters this scenario implies maximum potable water saving of 23%, and a reduction in WWTP influent volume of 11%. Sludge generated by the Nordhavnsgården RBC greywater treatment plant in Copenhagen, Denmark (~ 0.5 l $person^{-1}$ day^{-1}according to 2007 maintenance reports) would be periodically transferred to a municipal WWTP without further pre-treatment.

3.3.3. Scenario 3: Greywater treatment option 2

Both bathroom and laundry greywater, but not kitchen greywater, are treated on-site by RBC and reused for toilet flushing and laundry washing. On this basis, it is calculated that 59.5 l $person^{-1}$ day^{-1} of greywater (50% of the potable water used) is treated onsite. Of this volume, 27.4 l $person^{-1}$ day^{-1} can be reused for toilet flushing before being released to the sewer and 16.7 l $person^{-1}$ day^{-1} can be reused for laundry washing and then returned to the on-site treatment plant for re-processing. Under this scenario the surplus of 15.4 l $person^{-1}$ day^{-1} of treated greywater will be released to the sewer, but is also theoretically available for alternative reuse purposes as discussed in the results for Scenario 2 above. Overall, this scenario implies potable water savings of 37%, and a 17% reduction in WWTP influent volume. Sludge generated by this system would be periodically transferred to a municipal WWTP without further pre-treatment.

3.3.4. Scenario 4: Greywater treatment option 3

Both bathroom and laundry, but not kitchen, greywater is treated using a land-based reed or willow bed system. On this basis, 59.5 l $person^{-1}$ day^{-1} would be treated and reused for

irrigation or groundwater recharge. This would reduce the total WWTP influent volume by 23% but internal household potable water usage would remain the same. Although regular sludge disposal will not be required under this scenario, there will be a steady sediment accumulation over time which will eventually need removal to allow the treatment system to continue to function efficiently.

4. **Discussion**

Greywater reuse is a potentially complex field, encompassing a wide range of potential treatment trains and spatial scales as well as numerous reuse options. Nevertheless, the scenario analyses which have been presented help to illustrate the possible range of impacts of local greywater treatment and reuse on broader-scale wastewater flows and pollutant loads. For example, it is evident that a considerable volume reduction in total WWTP influent can potentially be achieved. This may also increase the pollutant concentration of the combined wastewater entering the municipal WWTP, potentially resulting either in greater treatment efficiency or in operational challenges due to excessive BOD concentrations in the combined influent wastewater. The actual effect is likely to be area specific and difficult to predict, as changes may be intensified by the adoption of additional water saving features in the home (e.g. low flush toilets, water saving shower heads, in-house water meters) but may also be off-set by increases in other areas (e.g. installation of power showers and increasing industrial wastewater discharges). Further research is needed to identify which implemented technologies would most efficiently limit the releases of PS/PHS to the environment.

The scenarios also demonstrate that some reuse options, such as irrigation, completely remove the volume of treated and reused water from the combined wastewater flow destined for the municipal WWTP, whereas other reuse options such as toilet flushing merely divert the water temporarily. This temporary diversion reduces the use of potable water and the overall volume of wastewater discharged to the WWTP, but does not necessarily have a significant impact on the overall load of micropollutants released, particularly if the treatment efficiency of a substance is low and it is primarily removed into the sludge fraction and later transferred to the WWTP. Scenarios 2 and 3 indicate potential potable water savings of 23% and 37%, which compares well with calculated demand reductions of 20–30% for greywater reuse by a 9 l WC depending on the size of the storage tank (Dixon et al., 1999).

Investigations of the chemical quality of greywater have been mainly confined to general water characteristics such as BOD and COD which although highly variable (Table 1) typically demonstrate > 90% removal efficiencies (Jefferson et al., 2004 and Eriksson et al., 2009). The most detailed removal performance study for specific organic micropollutants describes the effective biodegradation of short- and long- chain parabens in a greywater treatment plant incorporating a RBC (Andersen et al., 2007). The fate of selected metals during greywater treatment has been reported by Eriksson and Donner (2009) with up to 50% of Pb, Hg and Ni being removed from bathroom sources but no significant removal occurring for Cd. In the absence of PS/PHS removal efficiency data during greywater treatment, the relevant environmental fate and partitioning data can be used to derive some general conclusions regarding the likely effects of greywater treatment on larger scale pollutant dynamics. It is important to consider that the majority of technology-based greywater treatment systems incorporate the use of one or more settling tanks, and sludge is thus produced as part of the treatment process. Importantly, these systems tend not to include any additional sludge treatment but rely instead on the periodic discharge of sludge (either via the sewer or via tanker) to a WWTP for further processing and disposal.

The geochemical modelling results indicate that the vast majority of PS/PHS (with the exception of the most volatile compounds) will predominantly partition to the solid phase and thus remain in the sludge fraction after treatment. Because this sludge is typically purged to a WWTP, the advantage of greywater treatment and reuse from a PS/PHS source control perspective is questionable, indicating that current greywater treatment technologies cannot necessarily be considered as effective barriers preventing the release of recalcitrant household micropollutants. In contrast, systems based on the reuse of greywater for garden irrigation or infiltration may well result in lower micropollutant loads entering the sewer system, but care must be taken to ensure that unacceptable environmental pollution does not occur due to the release of hazardous substances that are not readily biodegradable in the soil environment. Whilst sludge production and disposal is less relevant for land-based treatment systems such as soil filters/constructed wetlands etc. these systems do present a solid phase to which many pollutants will preferentially partition. Consequently, such treatment systems may act both as pollutant sinks and sources, with bound pollutants potentially becoming re-mobilised under some environmental conditions. Together, these results indicate that additional source control measures focused on decreasing the PS/PHS content of domestic greywater continue to be of importance and should be given further attention in order to minimise the release of PS/PHS and other key micropollutants from household sources. Relevant source control options for consideration include substance substitution, green procurement, eco-labeling of household products, public information campaigns, and regulatory controls.

In most countries, regulations specific to wastewater reuse are relatively undeveloped, especially in relation to the testing requirements for proposed treatment systems and associated water quality standards. Where guidelines/standards do exist, they tend to focus exclusively upon conventional monitoring parameters such as microbial indicator organisms, organic content, turbidity/suspended solids and pH, and do not specifically address significant micropollutants. With regulators increasingly moving to address the issue of wastewater reuse there is a growing need to further understand the pollutant content and composition of greywater. As discharge standards and emission limits become increasingly stringent, the removal of micropollutants (including endocrine disrupting substances, metals, pesticides, personal care products and pharmaceuticals) will become of increasing concern for wastewater utilities and regulators. Hence, it is important to investigate the efficacy of greywater treatment for emerging pollutants and non-standard parameters in order to ensure that treatment can be optimised for the maximum environmental benefit. Many of the WFD PS/PHS are already subject to increasing regulatory restrictions, as well as substitution, and other types of source control. Concentrations of these substances in household greywater should therefore be decreasing, although some substances such as PBDEs and DEHP will continue to be emitted due to their widespread use in household items with long service lives. As numerous other xenobiotic pollutants are also likely to be found in domestic greywater it is possible that the current WFD PS/PHS do not present the greatest potential concern when considering the reuse of greywater. Pathogen related risks will continue to have a high priority when determining the suitability of different greywater reuse options and the removal of microbially related parameters has received considerable attention (e.g. Winward et al., 2008 and Ottoson and Stenstrom, 2003). In comparison, the presence and fate of micropollutants is a relatively understudied area of greywater research which is needed to facilitate appropriate risk assessments and the quality benefits to be gained by on-site treatment.

5. **Conclusions**

By examining a relatively small number of greywater treatment scenarios it has been shown that there are a range of different benefits which can be achieved both with regard to potable water demand and treatment volume arriving at the WWTP. The presence of XOCs in greywater has been demonstrated but significant knowledge gaps still remain and the removal efficiencies of these compounds in greywater treatment systems have so far received little attention. Geochemical modelling results indicate that PS/PHS removal during treatment is predominantly due to sludge/solid phase adsorption and therefore those greywater treatment systems which periodically discharge sludge to sewers will be of limited benefit in terms of comprehensive source control. It is, therefore, essential that other PS/PHS source control options (e.g. eco-labeling, green procurement, information campaigns, substance substitution, and regulatory controls) continue to be pursued in parallel with ongoing efforts to identify key greywater micropollutants and suitable techniques for their treatment.

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