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The implications of household greywater treatment and reuse for municipal wastewater flows and micropollutant loads.

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ABSTRACT
An increasing worldwide interest in water recycling technologies such as greywater treatment and reuse suggests that additional research to elucidate the fate of xenobiotics during such practices would be beneficial. In this paper, scenario analyses supported by empirical data are used for highlighting the potential fate of a selection of xenobiotic micropollutants in decentralised greywater treatment systems, and for investigation of the possible implications of greywater recycling for the wider urban water cycle. Potential potable water savings of up to 43% are predicted for greywater recycling based on Danish water use statistics and priority substance monitoring at a greywater treatment plant in Denmark. Adsorption represents an important mechanism for the removal of cadmium, nickel, lead and nonylphenol from influent greywater and therefore the disposal route adopted for the generated sludge can exert a major impact on the overall efficiency and environmental sustainability of greywater treatment.

Key words – greywater treatment; wastewater influent; recycling; priority substances; scenario analyses; sludge disposal

1. Introduction

With pressures on potable water supplies continuing to increase worldwide, interest in the use of alternative water sources such as recycled wastewater is also growing (Chu et al., 2004; Bixio et al., 2006). In particular, greywater treatment and reuse is receiving increasing attention (e.g. Maimon et al., 2010; Liu et al., 2010). This is because greywater generally has a lower organic pollutant and pathogen content than combined municipal wastewater which also contains toilet waste (Eriksson et al., 2002). Thus, greywater is considered particularly suitable for on-site (i.e. decentralised) treatment and reuse. Greywater treatment and reuse schemes have already been piloted in many countries around the world and are becoming increasingly commonplace in water stressed areas such as Australia and Mediterranean countries (Friedler and Gilboa, 2010; Masi et al., 2010; Pinto et al., 2010). However, related research has largely been restricted to studies of standard water quality parameters such as total organic carbon, biological oxygen demand, chemical oxygen demand and faecal and total coliforms (e.g. Pidou et al., 2008; Paulo et al, 2009). In contrast, there has been very little greywater research investigating the loads and dynamics of micropollutants. Nevertheless, Eriksson et al. (2002; 2003) and Palmquist and Hanaeus (2005) have collectively shown that a large number of xenobiotic substances can find their way into greywater via bathroom and laundry products.

Donner et al. (2010) have reported initial investigations into the fate of a range of pollutants within greywater treatment and reuse systems. However, given the increasing implementation of greywater recycling technology, it is evident that additional research to elucidate the behavior of xenobiotic micropollutants during greywater treatment would be beneficial. It would also be useful to understand the potential implications of more widespread greywater recycling for urban wastewater loads and dynamics. Greywater treatment and reuse is a very diverse field, encompassing a wide range of potential treatment trains and spatial scales, as well as numerous reuse options (Li et al., 2009; Misra et al., 2010). Current treatment options vary widely in sophistication from simple filter systems to constructed wetlands, multi-stage biological treatment systems, and membrane bioreactors. Nevertheless, all systems are based on a combination of chemical, physical and biological processes such as adsorption, coagulation, precipitation, filtration, aeration, biodegradation, and disinfection.

Reuse options cover a wide range of non-potable applications, from those involving a higher risk of human exposure such as spray irrigation and car washing, to lower risk options such as toilet flushing and sub-surface irrigation of non-food crops. Although pathogen transfer is generally considered the most pressing concern, it is nonetheless important to ensure that the lack of information regarding the chemical pollutant dynamics of greywater does not lead to the prevalence of sub-optimal treatment trains or inappropriate reuse practices. This is currently being brought into focus with the development of national standards and codes of practice for both greywater treatment and specific reuse applications (e.g. in the UK and Australia). Fatta-Kassinos et. al. (2010) have recently reviewed the practice of wastewater reuse for irrigation purposes and concluded that the benefits associated with improved water balances and nutritional levels need to be assessed against the current lack of knowledge relating to possible impacts on ecosystems and human health of the applied organic xenobiotics and heavy metals.

In this paper, scenario analyses are used to highlight the potential fate of a selection of xenobiotics in decentralised greywater treatment systems, and to investigate the possible implications of greywater recycling for the urban water cycle. All of the substances investigated are listed under the European Water Framework Directive (WFD) (European Commission, 2000a) as ‘Priority Substances’ (PS) or ‘Priority Hazardous Substances’ (PHS) and are known to be present in greywater. A range of different greywater treatment and reuse scenarios are compared in order to ascertain the likely benefits/shortcomings of the different scenarios in terms of micropollutant persistence and fate, including the possible impacts on municipal wastewater flow dynamics and pollutant source control.

Due to the limited availability of relevant data, the presented results focus on cadmium (Cd), nickel (Ni), lead (Pb), benzene and 4-nonylphenol (4-NP). Cadmium, Ni and Pb are metal pollutants of high concern in the municipal wastewater treatment process, as their tendency to accumulate in sludge can counteract its beneficial reuse for nutrient recovery and soil conditioning. For instance, national and European regulations specify acceptable levels of metal pollutants in sludge destined for recycling to agricultural land (e.g. European Commission, 1986) and sludge not meeting those criteria must be disposed of via alternative means such as incineration or landfilling. Particular focus is given in this paper to the potential for greywater treatment to act as an emission control barrier for Cd. Recognised as a PHS under the WFD and highlighted as a major element of concern in relation to sludge quality, Cd is toxic to humans, has no known biological function and is one of the more mobile metals in soil. It is thus of particular concern in terms of crop uptake potential as it can pose health risks to humans and animals at levels well below phytotoxic concentrations (McLaughlin et al., 2000).

Some sludge regulations (including the Danish national regulations) also specify acceptable levels of key organic pollutants, such as nonylphenols which have been found to accumulate in the sludge fraction during wastewater treatment (e.g. Abad et al., 2005; Koh et al., 2005). For contrast, benzene has also been included among the selected substances because being a relatively volatile substance, it tends to partition predominantly to air rather than sludge or water, and can thus be expected to demonstrate a differing behaviour during greywater treatment. Both benzene and 4-NP are resistant to biodegradation, as is typically the case for substances identified as PS/PHS. This investigation of the fate of selected greywater micropollutants facilitates a good overview of the possible implications of more widespread implementation of greywater reuse technologies.

2. Materials and Methods

2.1. Greywater treatment at Nordhavnsgården

The Nordhavnsgården treatment plant is located in the basement of an apartment block in Copenhagen, Denmark, and consists of a primary settling tank, a three-stage rotating biological contactor (RBC), a secondary settling tank, a sand filter, an ultraviolet disinfection unit, and a service-water storage tank. Eighty-four one-bedroom apartments (~ 117 inhabitants) are connected to this facility which treats bathroom greywater for reuse as toilet flushing water and is automatic and self-cleaning.

2.2. Chemical analysis of PS and PHS in greywater and greywater treatment sludge

The selected PS (benzene, Ni and Pb) and PHS (Cd and 4-NP) were measured both in hot and cold potable water, and in the influent and effluent greywater from the ‘Nordhavnsgården’ greywater treatment system. Sixteen time-proportional samples of influent and effluent greywater were collected over a one-week period (29 November to

5 December 2007) using acid washed bottles. In addition, bottles used to collect samples for organic analysis were pre-heated at high temperature (220ºC for 24 hours). All samples (except for benzene analysis) were filtered prior to analysis (GF/A 1.6 µm for metals analysis and GF/C 1.2 µm for organics analysis).

Cadmium, Ni, and Pb were analysed by Inductively Coupled Plasma - Optical Emission Spectroscopy (Varian Vista-MPX CCD Simultaneous ICP-OES). Benzene was determined by purge and trap (Tekdyn Tekmar Velocity XPT Purge and Trap Sample Concentrator) and gas chromatography (Shimadzu Gas Chromatograph GC-14B, equipped with a Flame Ionization Detector). 4-nonylphenol was isolated and concentrated by solid phase extraction prior to analysis by GC-MS (Agilent 6890N GC-system with an Agilent 5973 Mass Selective Detector). All instrumental analyses were performed in triplicate. Quality control procedures included determination of detection limit, quantification limit, linearity, and precision. The detection limits for the employed analytical procedures were benzene (1.4 µg l^{-1}), 4-NP (0.005 µg l^{-1}), Cd (0.01 µg l^{-1}), Ni (0.1 µg l^{-1}) and Pb (0.03 µg l^{-1}). Internal reference materials were also included in all analyses for quality control purposes.

The total greywater sludge was collected from the primary settling tank and rotating biological contactor on three occasions (separated by 4 monthly intervals) and was initially dewatered by centrifugation (4000 rpm for 20 minutes). The settled material was dried at 105 °C for 1 hour, pulverised and weighed, then acid digested (7 M nitric acid at 125 °C and 2 atmosphere for 30 minutes according to Danish Standards (DS259, 2003; DE/EN15586, 2004) prior to metal analysis by ICP-OES. The sludge was not analysed for benzene and 4-NP. Total solids (TS) were determined according to APHA et al. (2005) to facilitate normalisation of the sludge metal content to the concentration per unit of dry weight (DW).

2.3. Scenario analyses

The twelve greywater treatment and reuse scenarios investigated during this study are documented in Table 1. They range from a baseline scenario of no treatment and no reuse (Scenario A) to full household greywater treatment and recycling (Scenario J; bathroom, laundry and kitchen greywater treated and reused for toilet flushing, laundry washing and irrigation). The identified scenarios differ in terms of the type of treatment plant (e.g. an indoor system using a RBC system and outdoor land-based treatment systems using reedbeds), in terms of the source of the greywater being treated (e.g. bathroom vs. bathroom + laundry) and in terms of the end-use of the recycled water (e.g. toilet flushing vs. toilet flushing + laundry washing). In practice, bathroom greywater is the fraction most commonly recycled and this is the reason for the relative dominance of this fraction in the selected scenarios (Table 1).

2.4. Water use statistics and input data to scenario analyses

The scenario analyses reported in this paper are based on Danish water use statistics. The potential effects of greywater recycling on wastewater flows under the different scenarios (assuming that 100 % implementation of greywater recycling technology is practised) have been calculated based on an average Danish potable water consumption of 119 l $person^{-1}$ day^{-1} and a 43% contribution from households to the influent of municipal wastewater treatment plants (DANVA, 2007). The other major inputs to wastewater treatment plants are from industrial and commercial wastewater, stormwater and sewer infiltration. The proportion of household water used for different domestic purposes (Kjellerup and Hansen, 1994) is identified in Table 2. Similar distributions have been reported by Memon and Butler (2006) for residential properties in the UK although with an increased proportion for toilet flushing and a reduced percentage for general bathroom use.

Table 1: Greywater treatment and reuse scenarios considered for this study.

Scenario	Treatment system	Source of greywater	Reuse of greywater
A	No treatment	-	-
B	Indoor - RBC	Bathroom	Toilet
C	Indoor - RBC	Bathroom	Toilet + Irrigation
D	Indoor - RBC	Bathroom + Laundry	Toilet
E	Indoor - RBC	Bathroom + Laundry	Toilet + Laundry
F	Indoor - RBC	Bathroom + Laundry	Toilet + Irrigation
G	Indoor - RBC	Bathroom + Laundry	Toilet + Laundry + Irrigation
H	Indoor - RBC	Bathroom + Laundry + Kitchen	Toilet + Laundry
I	Indoor - RBC	Bathroom + Laundry + Kitchen	Toilet + Irrigation
J	Indoor - RBC	Bathroom + Laundry + Kitchen	Toilet + Laundry + Irrigation
K	Outdoor - reedbed	Bathroom	Groundwater recharge
L	Outdoor - reedbed	Bathroom + Laundry	Groundwater recharge

Table 2. Proportion of household water used for different domestic purposes (after Kjellerup and Hansen, 1994).

Location/use of household water	Range and average* percentages
Bathrooms	35-37 (36)
Laundry activities	13-15 (14)
Kitchens	17-25 (21)
Toilet flushing	20-27 (23)
Irrigation	5-7 (6)

*Average percentages in parenthesis

2.5. Pollutant fate analysis

The fate of the selected substances during greywater treatment and reuse has also been evaluated under the different scenarios. Hypothetical pollutant removal efficiencies of 10 %, 50 % and 90 % were used for the pollutant fate calculations in order to cover a broad range of potential treatment situations. With such a broad range of treatment systems potentially available and little attention given to optimising these systems for micropollutant removal it is prudent to conclude that many systems may have limited effectiveness in terms of non-standard parameters. Pollutant load data used for the pollutant fate calculations have predominantly been based on the Nordhavnsgården data presented in this paper. However, only bathroom greywater is recycled at the Nordhavnsgården site. Thus, in order to facilitate Cd fate calculations for the full suite of scenarios (Scenarios A-L), additional data on greywater Cd loads for kitchen and laundry greywater was taken from Wall (2002) and Bergstrom (2007) and the Cd load in blackwater (i.e. toilet wastewater including faeces and urine) was taken from Palmquist and Hanaeus (2005). These studies were conducted in Swedish households.

As measured data for laundry and kitchen greywater were not available for benzene, 4-NP, Ni, and Pb only those scenarios involving bathrooms as the source of greywater (Scenarios B and C) have been investigated for these pollutants but a complete scenario analysis has been completed for Cd.

The physicochemical characteristics of the different pollutants have been taken into account in assessing their removal behaviour during the greywater treatment process. For the metals and their compounds the main removal process will be adsorption with negligible removal by biodegradation and no susceptibility to volatilisation. A precise assessment of metal adsorption capability is difficult due to the variety of compounds and complexes which can exist in wastewater samples but in a review of the potential of metals to be removed from stormwater, Revitt et.al. (2008) have identified the highest adsorptive removal to be associated with Pb followed by Ni and with Cd demonstrating the lowest removal potential. The behaviours of benzene and 4-NP can be correlated with the relevant physiochemical parameters such as adsorption coefficients, biodegradation half-lives and Henry's Law constant for volatilisation (Scholes et. al., 2007). These parameters suggest equal, but limited, susceptibilities for both pollutants to aerobic biodegradation but clear differences with regard to adsorption and volatilisation. Benzene is predicted to have the high potential to be removed by volatilisation compared to moderate removal for 4-NP and the reverse is true for adsorption although to a less exaggerated extent.

3. Results and Discussion

3.1. Priority substances in greywater

A summary of relevant pollutant monitoring data for greywater influent to the Nordhavnsgården treatment plant is given in Table 3. All of the selected PS/PHS were detected at measurable concentrations and the results are generally comparable to existing Danish and Swedish greywater monitoring data for these substances (also given in Table 3), with some exceptions such as the high concentration of Cd (2.5 ug l^{-1}) measured at the Gals Clint campingsite (Nielsen and Petersen, 2005). However, a high level of consistency is not to be expected given that greywater flows and pollutant loads are inherently variable and highly dependent on the behaviour of individuals. In addition to the concentrations of the selected PS/PHS in greywater, measured values for these substances in the potable water at Nordhavnsgården, and in the abstraction wells used to supply the potable water distribution network in Copenhagen (Copenhagen Energy 2008a; 2008b) are also presented in Table 3. The abstraction well data clearly demonstrate the low background levels of the monitored substances.

3.2. Flow Calculations

Based on monitored greywater inflow rates and the Danish water use statistics specified in Section 2.4, effluent flow rates (expressed as litres per person per day; l p^{-1} d^{-1}) have been calculated for each of the identified scenarios. Figures 1a and 1b provide diagrammatic representations of the flow pathways associated with Scenarios A and J and serve as examples of the method by which the proportional potable water savings and the proportional reductions in wastewater treatment plant effluent in columns 2 and 3, respectively of Table 4 were derived. It can be seen that under the baseline conditions represented by Scenario A (i.e. no greywater treatment followed by reuse but direct use of greywater for irrigation purposes) a daily potable water use of 119 l p^{-1} d^{-1} results in 111.9 l p^{-1} d^{-1} of household wastewater being released to the municipal wastewater system.

Table 3: Nordhavnsgården monitoring data used in the scenario calculations, and other relevant data from the literature (all values in μg l^{-1}).

	Cd	Ni	Pb	Benzene	4-NP
Influent concentration (Nordhavnsgården) (n=8)	Range: 0.01 – 0.22 Mean: 0.08 Median[1]: 0.07	Range: 5.15 – 26.5 Mean: 9.32 Median: 6.76	Range: 4.89 – 10.2 Mean: 6.95 Median: 6.82	Range: <1.4 – 9.85 Mean[1]: 3.61 Median[1]: 2.51	Range: 0.35 – 1.63 Mean: 0.90 Median: 0.90
Greywater influent concentration (Danish and Swedish greywater literature data)	Range[2]: 0.06-0.66 Mean[2]: 0.22 2.5[3] < 0.1[4] Range[5]: 0.06 – 0.16 Mean[5]: 0.10	Range[2]: 3.86-10.2 Mean[2]: 6.2 1.3[3] 1.5[4] Range[5]: 4.45-28.1 Mean[5]: 11.0	Range[2]: 1.1-6.9 Mean[2]: 3.4 1.8[3] <2[4] Range[5]: 2.14-3.14 Mean[5]: 2.52	All values <1.9[2]	All values <0.5[2*] 0.76[3*] 0.9[4*] Range[5]: 2.85-5.95 Mean[5]: 3.8 Range[6]: 0.56-1.1 Mean[6]: 0.76
Potable water concentration (Nordhavnsgården)	Cold water: <0.01 Hot water: <0.01	Cold water: 0.24 Hot water: 0.35	Cold water: 7.27 Hot water: 6.21	Cold water: <1.4 Hot water: <1.4	No data
Concentration in Copenhagen potable water abstraction wells*	Range: 0.03-0.07 Mean: 0.04	Range: 0.46-8.9 Mean: 2.21	Range: <0.03-0.11 Mean: 0.22	All values <1.4	All values <0.5

[1] 38% of the values for benzene were below the detection limit; for the purposes of calculating mean and median values these were assumed to be equal to half of this value (i.e. 0.7 μg l^{-1} for benzene).

[2] BO90 (apartment block), Copenhagen, Denmark (Ledin et al., 2006)

[3] Gals Klint (campingsite), Denmark (Nielsen and Pettersen, 2005)

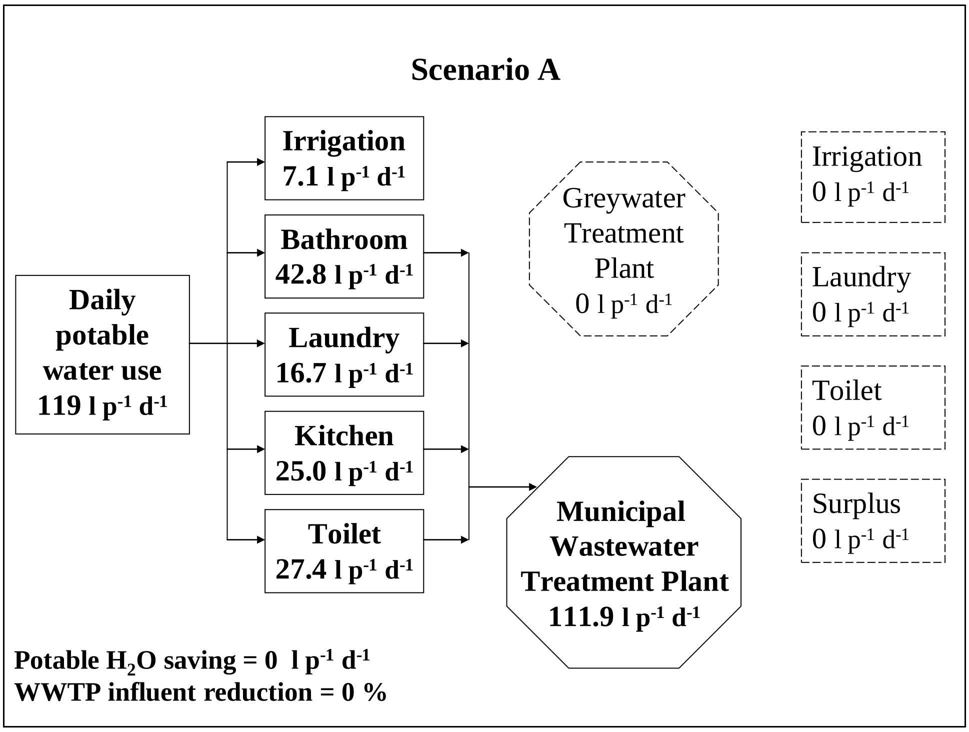
[4] Vestbadet I/S, Denmark (Andersson and Dalsgaard, 2004)

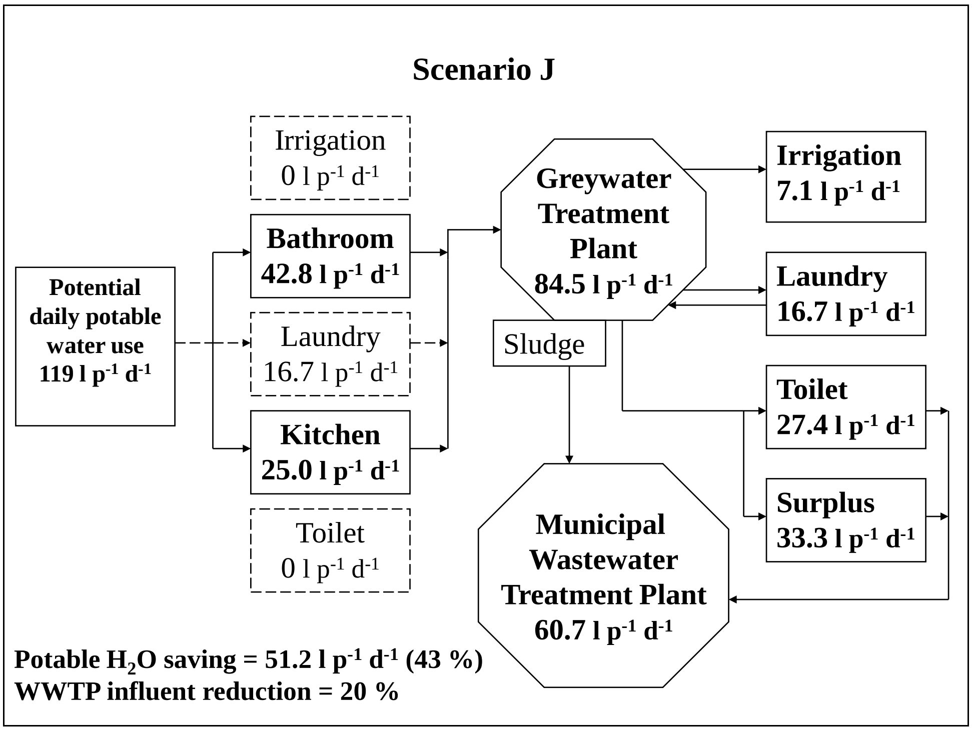
[5] Vibyåsen (housing area), Sollentuna, near Stockholm, Sweden (Palmquist and Hanaeus, 2005)

[6] Gebers (apartment block), Skarpnack, near Stockholm, Sweden (Palmquist, 2004)

[*] Indicates that a measurement includes not only 4-NP but nonylphenols collectively

In contrast, under Scenario J (where bathroom, laundry and kitchen greywater are treated and reused for irrigation, laundry washing and toilet flushing), the effluent volume is reduced to 60.7 l p^{-1} d^{-1}, representing a reduction in the effluent to the municipal wastewater treatment plant (WWTP) of 20 % (when the 43% contribution of households to this wastewater stream is taken into account). This scenario also achieves a potable water saving of 51.2 $l^{-1}p^{-1}$ d^{-1} due to the use of greywater for toilet flushing, the continued recycling of laundry effluents through the greywater treatment system and avoidance of using potable water for irrigation. The effective water use is 67.8 $l^{-1}p^{-1}$ d^{-1} which amounts to a saving of 43% compared to the baseline situation represented by Scenario A. The calculations for Scenario J (Figure 1b) also show that 33.3 $l^{-1}p^{-1}$ d^{-1} of treated greywater will be produced for which there is no identified reuse application. This would represent an inefficient use of treatment resources and the described scenario analysis approach therefore offers a route for optimising the treated volumes according to user requirements.





Figures 1a and 1b: Diagrammatic representation of water flow for Scenarios A and J (dashed borders indicate water use options which are not relevant to that particular scenario).

Table 4: Implications of Scenarios A-L for municipal wastewater flows and Cd loads, assuming onsite greywater treatment Cd removal efficiencies of 10 %, 50 % and 90 %.

Scenario	Potable H_2O saving (%)	Reduction in WWTP influent (%)	Reduction in Cd load to WWTP based on 10 % removal efficiency*		Reduction in Cd load to WWTP based on 50 % removal efficiency*		Reduction in Cd load to WWTP based on 90 % removal efficiency*	
			Assuming sludge is discharged to WWTP	Assuming sludge is removed from WW stream	Assuming sludge is discharged to WWTP	Assuming sludge is removed from WW stream	Assuming sludge is discharged to WWTP	Assuming sludge is removed from WW stream
A	-	-	-	-	-	-	-	-
B	23	11	0	0.31 (1.5 %)	0	1.53 (7.6 %)	0	2.74 (13.5 %)
C†	29	13	0.45 (2.2 %)	0.75 (3.8 %)	0.25 (1.2 %)	1.78 (8.9 %)	0.05 (0.2 %)	2.80 (13.9 %)
D	23	11	0	0.77 (3.8 %)	0	3.85 (19.1 %)	0	6.93 (34.5 %)
E	37	17	0	1.19 (5.9 %)	0	4.56 (22.7 %)	0	7.15 (35.6)
F†	29	13	0.82 (4.1 %)	1.59 (7.9 %)	0.46 (2.3 %)	4.31 (21.5 %)	0.09 (0.4 %)	7.02 (35.0 %)
G†	43	20	1.25 (6.2 %)	2.28 (11.3 %)	0.60 (3.0 %)	5.09 (25.3 %)	0.11 (0.5 %)	7.24 (36.1 %)
H	37	17	0	1.13 (5.6 %)	0	5.16 (25.7 %)	0	8.53 (42.5 %)
I†	29	13	0.69 (3.4 %)	1.62 (8.1 %)	0.38 (1.9 %)	5.02 (25.0 %)	0.08 (0.4 %)	8.43 (42.0 %)
J†	43	20	0.84 (4.2 %)	1.97 (9.8 %)	0.42 (2.1 %)	5.58 (27.8 %)	0.08 (0.4 %)	8.60 (42.8 %)
K	0	27	N/A	N/A	N/A	N/A	N/A	N/A
L	0	17	N/A	N/A	N/A	N/A	N/A	N/A

* Main value given is the reduction in load in $\mu g\ p^{-1}\ d^{-1}$; values in brackets show the reduction in load as a percentage of the total household load).
† Values given show the reduction in load after 5 cycles of the given scenario (i.e. laundry water recycled 5 times).

The flow calculation results provided in Table 4 demonstrate the implications of the different scenarios in terms of both potential potable water savings and reduced wastewater influent volumes at municipal WWTPs. Significant potable water savings (up to 43 % for the described scenarios) can be achieved by recycling greywater. However, subsequent reductions in wastewater flows to large-scale municipal WWTP are predicted to be more modest (up to 27 % for Scenario K) as the assumption has been made that only 43 % of the total WWTP influent volume is derived from households (DANVA, 2007). The most beneficial combination of potable water savings and WWTP influent reductions are achieved when the volume of recycled water is sufficient to cover the requirements for toilet flushing, laundry washing, and outdoor irrigation uses (e.g. Scenarios G and J). It is important to note however that these impacts have been calculated on the basis of 100 % uptake of the relevant greywater recycling scenario. Whilst this is feasible for new developments (or large-scale refurbishments), particularly in water stressed countries where water recycling regulations on new-builds are increasingly likely to be introduced, it should be recognised that implementation of greywater reuse in more established built environments without existing dual reticulation plumbing systems is likely to remain much lower than 100 %.

3.3. Micropollutant fate during greywater treatment and reuse

For each indoor treatment and reuse scenario (Scenarios A-J), the fates of the pollutants have been calculated based on hypothetical greywater treatment removal efficiencies of 10 %, 50 % and 90 %. These hypothetical removal efficiencies span the wide range anticipated for the available treatment options of varying sophistication which can be expected to differ substantially in their ability to remove micropollutants. For example, losses due to volatilisation are likely to be greater in systems incorporating rotating biological contactors, than in simple filtration systems without additional aeration and will therefore exert the greatest influence on the removal of benzene. Treatment systems also vary widely in their ability to remove suspended solids and adsorbed pollutants from greywater (Donner et al, 2010). This is a process which has been identified as being important for the removal of Pb and 4-NP. The composition and condition of the microbial community or biofilm in biological systems will significantly affect the biodegradation potential for organic micropollutants (Donner et al, 2010; Giri et al, 2006) and has been identified as being equally important for the removal of both benzene and 4-NP. Biological greywater treatment systems can take some time to mature and establish reliable performance and may be inhibited by pollutant shock loadings, such as a predominance of bleach or other cleaning products. Treatment efficiencies can be expected to vary over time and the use of hypothetical removal efficiencies of varying effectiveness is thus a useful approach for providing an overview of the possible impacts of different greywater treatment and reuse scenarios on the wider urban water cycle.

In Table 4 the results of the Cd fate calculations for the full range of scenarios are presented. These results also demonstrate how two different hypothetical pathways for sludge disposal will influence the influent Cd load to a WWTP. One set of calculations are based on the assumption that the greywater treatment sludge will be discharged or transferred periodically to the municipal WWTP (as is in fact most commonly the case) with the second set of calculations being designed to investigate the effect of employing a separate sludge disposal route (such as disposal to land).

As an example of the manner by which pollutant pathways have been evaluated for the different scenarios, the fate of household-derived Cd pollution under Scenario B (see Figure 2) is described in detail in Box 1. The different steps in the calculation can be matched to the scenario diagram by means of the square bracketed letters in both Figure 2 and Box 1. According to Scenario B, bathroom greywater is treated on-site using a RBC and reused for toilet flushing, and the results show that treatment and reuse according to this scenario will have no positive effect on WWTP Cd influent loads unless the sludge is removed from the wastewater stream entering the associated WWTP (Table 4). Furthermore, even under conditions of separate sludge disposal, the greatest potential decrease in Cd loading at the treatment plant will be 2.74 $\mu g\ p^{-1}\ d^{-1}$ (assuming 90 % removal efficiency during treatment and 100 % implementation of Scenario B). Compared to the baseline scenario (Scenario A) which incorporates no greywater treatment and reuse, this represents a fairly minor overall reduction (13.5 %) on the influent Cd load at the WWTP, as baseline calculations indicate a total household load of 20.2 $\mu g\ p^{-1}\ d^{-1}$.

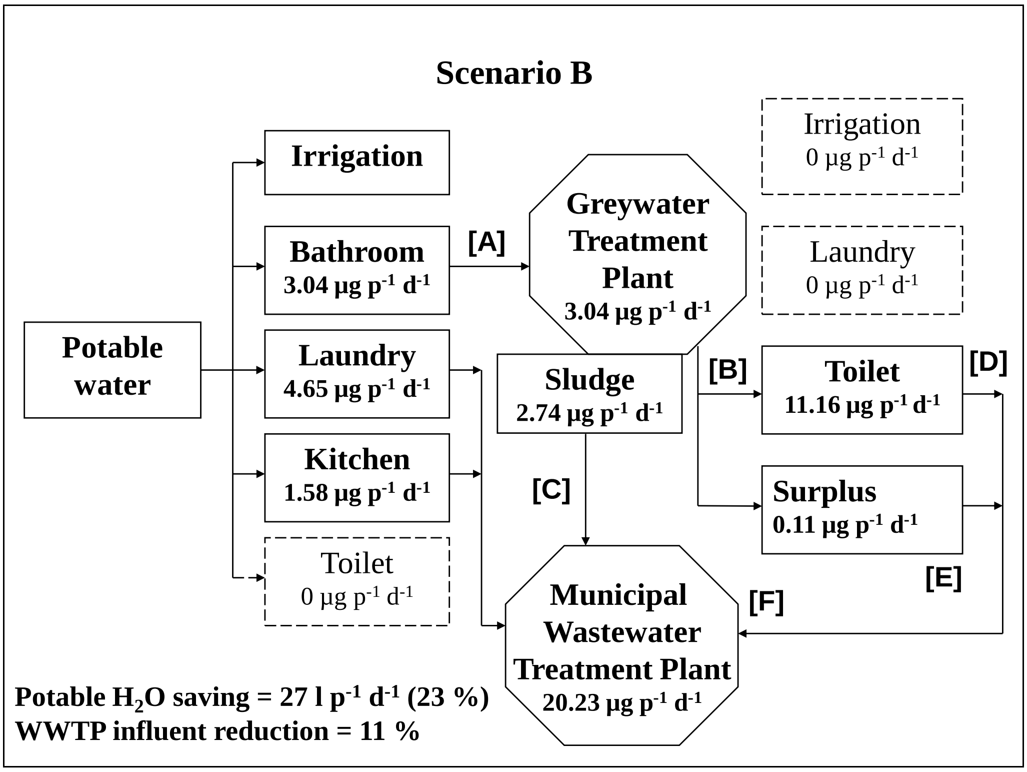


Figure 2: Diagrammatic representation of Scenario B and associated Cd load calculations (based on 90 % removal efficiency during treatment) as described in Box A. Letters in square brackets can be used to match with the associated calculation in Box 1.

It is clear that the incorporation of Cd in the sludge is a critical pathway in controlling the fate of this and similar pollutants. In those situations where the sludge from the greywater treatment process is eventually discharged or transferred to a WWTP, there will be no overall Cd removal unless the scenarios incorporate removal of some of the treated greywater from the municipal wastewater stream by using it for irrigation purposes (i.e. Scenarios C, F, G, I and J). When irrigation is practiced, it is interesting to note that the impact on the WWTP load is not consistent with the increasing treatment efficiency of the greywater plant. Thus for Scenario C, it can be seen that the overall removal of Cd from the wastewater stream in terms of the decrease in total household load arriving at the WWTP decreases from 2.2 % to 1.2 % to 0.2 % as the applied greywater treatment efficiencies increase from 10 % to 50 % to 90 % (Table 4). This can be explained by the fact that the higher treatment removal efficiencies (i.e. 50 % and 90 %) produce treated greywater with lower Cd concentrations, and hence the proportion of Cd removed from the total WWTP system due to losses via irrigation is reduced.

Box 1: Cadmium fate calculations for greywater treatment and reuse according to Scenario B (based on 90 % removal efficiency).

[A] With an estimated bathroom greywater flow rate of 42.8 l p^{-1} d^{-1} (based on DANVA (2007) and Kjellerup and Hansen, 1994) and a median measured Cd concentration in the Nordhavnsgården bathroom greywater of 0.071μg l^{-1}, the median Cd load in untreated bathroom greywater is 3.04 μg p^{-1} d^{-1}.

[B] Assuming a greywater treatment removal efficiency of 90 %, the maximum effluent Cd loading will be 0.30 μg p^{-1} d^{-1}. The remaining Cd (2.74 μg p^{-1} d^{-}1) will be entrained in the sludge produced by the greywater treatment system. The greywater treatment effluent has a Cd concentration of 0.0071 μg l^{-1} (0.30 μg p^{-1} d^{-1} ÷ 42.8 l p^{-1} d^{-1}).

[C] As with most treatment systems of this type the sludge produced at the Nordhavnsgården treatment plant is periodically transferred directly to the municipal WWTP without further pre-treatment.

[D] The Cd loading in the treated water used for toilet flushing is 0.19 μg p^{-1} d^{-1} (27.4 l p^{-1} d^{-1} x 0.0071 μg l^{-1}). Additionally, Cd could be added due to the addition of faeces and urine at this stage. Based on published measurements of Cd in blackwater (Palmquist and Hanaeus, 2005) it is estimated that the concentration of Cd in toilet wastewater would be 0.4 μg l^{-1}. Therefore, in a volume of 27.4 l, the maximum Cd loading contribution from the addition of blackwater would be 10.96 μg p^{-1} d^{-1}. Hence, the total Cd load which would be discharged to the WWTP upon toilet flushing is 11.15 μg p^{-1} d^{-1} (0.19 + 10.96 μg p^{-1} d^{-1}).

[E] Under Scenario B, surplus greywater treatment effluent (i.e. treated greywater not required for toilet flushing) will be discharged directly to the WWTP. The surplus flow rate is 15.4 l p^{-1} d^{-1} and the Cd concentration is 0.0071 μg l^{-1} which equates to a Cd loading of 0.11 μg p^{-1} d^{-1}.

[F] The total Cd load discharged to the WWTP after greywater treatment and reuse is 14.00 μg p^{-1} d^{-1} (2.74 + 11.15 + 0.11). The three contributing sources to this Cd load are sludge [C], reused water after toilet flushing [D] and surplus treated water [E]. Under this scenario, additional household Cd releases will also occur due to laundry washing or kitchen activities as these waste streams are discharged directly to the WWTP. The relevant Cd loads from these sources are estimated to be 4.65 μg p^{-1} $d^{-1}$from the laundry greywater and 1.58 μg p^{-1} d^{-1} from kitchen greywater (1.16 μg p^{-1} d^{-1} for dishwashing + 0.26 μg p^{-1} d^{-1} from sink wiping + 0.16 μg p^{-1} d^{-1} from food preparation) (Wall, 2002). Therefore the total Cd load to the wastewater treatment plant would be 20.23 μg p^{-1} d^{-1} (14.00 + 4.65 + 1.58).

Impact:
The total household Cd load without greywater treatment (Scenario A) is estimated to be 20.23 μg p^{-1} d^{-1} (comprising 3.04 μg p^{-1} d^{-1} from bathroom greywater, 4.65 μg p^{-1} d^{-}1 from laundry greywater, 1.58 μg p^{-1} d^{-1} from kitchen greywater, and 10.96 μg p^{-1} d^{-1} from toilet wastewater). Therefore, as expected, under Scenario B there will be no decrease in Cd loading going to the WWTP unless the greywater sludge is removed from the system and treated separately. If this was practised, it would equate to a decrease in WWTP influent Cd loading of 2.74μg p^{-1} d^{-1} and a potential overall per capita Cd removal efficiency of 13.5 %.

If it is feasible to remove the sludge produced by the greywater treatment system from the external wastewater stream, it can be seen that all scenarios (other than A, K and L) produce overall Cd removal efficiencies which are consistent with the expected results based on the applied greywater treatment values. For 10 % greywater treatment efficiency, the most efficient overall Cd removal is demonstrated by Scenario G (11.3%) whereas for the higher greywater treatment performances Scenario J proves to be most efficient (27.8% and 42.8%). Scenarios G and J both involve continuous recycling of laundry greywater and the results in Table 4 are based on predictions after the completion of 5 cycles. All scenarios incorporating laundry water recycling (Scenarios E, G, H and J) involve micropollutants being continually added to the system and the wastewater being continually circulated and treated for reuse. The calculations indicate that the Cd concentration in these systems initially increases but approaches an equilibrium situation with regard to the greywater Cd loading and an optimal removal efficiency is established within 5 cycles or less. This suggests that there should not be any detrimental impact on washing machine functioning due to micropollutant build-up although the elevated pH levels during typical laundry washing may encourage the precipitation of some constituents and corrosion may occur due to increased salinity.

The annual influent loads of Cd, Ni, Pb, benzene and 4-NP to the Lynetten WWTP, which services the area of Copenhagen where the Nordhavnsgården greywater treatment plant is located, are 21 kg, 386 kg, 1064 kg, 12.6 kg and 178 kg (Lynettefællesskabet I/S, 2008). Because of the differences in influent flows (5.7 m^3/year to Nordhavnsgården greywater treatment plant compared to 74 million m^3/year to the WWTP), the contributions deriving from untreated Nordhavnsgården greywater are very low, typically of the order of 0.001%. Therefore, clearly in terms of assessing the benefits which could be accrued by comprehensive application of greywater treatment, it is more realistic to compare per capita pollutant reductions. On this basis, the results reveal that full implementation of the most effective scenario (i.e. Scenario J with full greywater treatment and recycling and separate sludge disposal) could lead to a calculated reduction in the Cd load to the WWTP of 8.6 $\mu g\ p^{-1}\ d^{-1}$ which is equivalent to a reduction of 14.1 % of the overall Cd influent load at the WWTP (61 $\mu g\ p^{-1}\ d^{-1}$). Although this is relatively low, it is apparent that in areas of low industrial activity and/or with separate stormwater treatment (i.e. where household wastewater is the major contributor to the municipal WWTP influent), the introduction of greywater treatment and reuse technologies may be beneficial in terms of pollutant emission control as well as water conservation. Clearly, the magnitude of the emission control function in relation to micropollutants will be highly dependent on the greywater sludge disposal pathway.The results presented in Tables 4 and 5 show that even when greywater treatment removes a substantial proportion of micropollutants from influent greywater, for elemental pollutants such as Cd, Ni and Pb and for hydrophobic substances such as 4-NP the resulting impact at the WWTP is highly dependent on the fate of the greywater treatment sludge.

In Table 5, the results derived for the bathroom greywater reuse scenarios are presented for two metals (Ni and Pb) and two organic micropollutants (benzene and 4-NP), respectively. Both metals follow similar trends to those described for Cd although with considerably elevated loading values. The magnitude of the differences in pollutant reductions according to the disposal route of the greywater treatment sludge are indicative of the adsorption potentials of different pollutants and are clearly less significant for benzene for which volatilisation plays an important role in controlling pollutant removal from the aqueous phase. The results for benzene and 4-NP shown in Table 5 have been informed by apportioning the contributions to the different removal processes during greywater treatment according to the distribution calculated using a pollutant fate model for an activated sludge WWTP (STPWIN, EPI Suite v 3.20, US EPA, 2007). As expected from a consideration of the physicochemical properties, only 1.1% of benzene is predicted to be removed by adsorption to sludge with volatilisation representing the major removal route (67.8%) in an overall removal capability of 68.9%. This raises concerns regarding the overall environmental effectiveness of greywater treatment as an emission control barrier for benzene. In contrast, 4-NP which has a low volatility (< 1% removal by volatilisation) is predicted to partition predominantly to the sludge (90% removal by adsorption) and therefore behaves in a similar way to the metals placing the fate of this pollutant firmly on the adopted sludge disposal route during greywater treatment. Both benzene and 4-NP are identified as possessing low potentials for removal by biodegradation (<1%).

Table 5: Implications of Scenarios A-C for Ni, Pb, benzene and 4-nonylphenol loads in bathroom greywater treatment sludge and household wastewater, assuming greywater removal efficiencies of 10 %, 50 % and 90 %.

		Reduction in load to WWTP ($\mu g\ p^{-1}\ d^{-1}$)*		
		Scenario A	Scenario B	Scenario C
Ni	10 % removal	-	0 (29.1)	42.59 (71.7)
	50 % removal	-	0 (145.3)	23.66 (169.0)
	90 % removal	-	0 (261.6)	4.73 (266.3)
Pb	10 % removal	-	0 (29.3)	42.98 (72.3)
	50 % removal	-	0 (146.6)	0 (263.9)
	90 % removal	-	23.87 (170.5)	4.76 (268.7)
Benzene	10 % removal	-	10.58 (10.8)	26.33 (26.5)
	50 % removal	-	52.89 (53.8)	61.64 (62.5)
	90 % removal	-	95.20 (96.8)	96.95 (98.5)
4-NP	10 % removal	-	0.03 (3.9)	5.70 (9.5)
	50 % removal	-	0.17 (19.4)	3.32 (22.5)
	90 % removal	-	0.31 (34.8)	0.94 (35.6)

* Main value given is the reduction in load in $\mu g\ p^{-1}\ d^{-1}$ assuming the greywater treatment sludge is discharged to the WWTP; values in brackets show the reduction in load assuming the greywater treatment sludge is removed from the wastewater stream. Removal due to sorption, volatilisation and biodegradation is apportioned according to the distribution calculated using STPWIN (EPI Suite v3.20, US EPA, 2007).

Scenarios K and L investigate the potential implications of land-based greywater treatment systems. Under these scenarios, the greywater is treated using reedbed technology resulting in advantageous overall reductions in terms of the municipal WWTP influent pollutant load, but also raising concerns regarding the possible environmental impacts. For example, under Scenario K, the removal of bathroom greywater for treatment in a reedbed equates to a decrease in Cd WWTP influent loading of 3.04 $\mu g\ p^{-1}\ d^{-1}$. Therefore, the reduction in Cd being directed to the WWTP due to this greywater treatment scenario is 15.0 %. According to Scenario L, in which both bathroom and laundry greywater are treated, the corresponding reduction in WWTP influent load is 38.4 %. In both cases, it is important to consider the environmental implications. Depending on the substrate of the treatment system, Cd may build up in the sediment/soil/solid phase over time and may also leach through to the groundwater. For the Nordhavnsgården greywater treatment plant the annual release of Cd to the environment would be 130.4 mg and 329.0 mg for Scenarios K and L, respectively.

A median wet weather removal efficiency of 84.7 % has been measured for Cd passing through a sub-surface constructed wetland (Revitt et al., 2004). If applied to Scenario K this would indicate that a discharge loading of 3.04 μg p^{-1} d^{-1} could be reduced to 0.46 μg p^{-1} d^{-1} after passing through an appropriately designed vegetated greywater treatment plant. Given the hydraulic loading rate of 42.8 l p^{-1} d^{-1}, this corresponds to a discharge concentration of 0.011 μg l^{-1} which is well below the proposed AA-EQS value (European Commission, 2008) for Cd for the most sensitive inland surface waters (0.08 μg l^{-1}) before any dilution has occurred within the receiving water. By contrast for Scenario L, the discharge of 7.69 μg p^{-1} d^{-1} at a hydraulic loading of 59.5 l p^{-1} d^{-1} corresponds to a discharge concentration of 0.13 μg l^{-1}. Treated greywater with this Cd concentration would require an appropriate dilution on entering a receiving water. More critically, if discharged to ground the adsorption characteristics of the soil would need to ensure that appropriate protection existed for an underlying aquifer.

3.4. Sludge fate and pollutant loading

One of the major drivers for further reducing micropollutant influent loads to municipal WWTPs is to facilitate the beneficial reuse of sewage sludge (i.e. biosolids) for soil conditioning of agricultural land. The European Directive most pertinent to the agricultural use of sewage sludge is Directive 86/278/EEC (European Commission, 1986) which establishes concentration limits for a number of metals that are typically present within sludge. The concentration limits are effectively ceiling limits, meaning that if sludge exceeds the metal concentration limit for any of the listed metals it should not be permitted for land application. Directive 86/278/EEC is currently under revision and the working draft for the revised Directive indicates that future limits will be more conservative (European Commission, 2000b). To enable some member states to achieve the new limits, it is probable that water companies will need to further tighten trade effluent consents for industries as well as seeking further means of reducing WWTP influent loads of key pollutants. The alternative would be an unwanted reduction in land recycling of sludge and a waste of a potentially beneficial resource. Currently, some member states, including Denmark, impose more stringent requirements than those in the EC Directive. For example, the current limit for Cd in the Danish regulations is 0.8 mg/kg DW compared to 20 mg/kg DW in the EC Directive and for nonylphenols the Danish value of 10 mg/kg DW is considerably lower than a proposed European sludge guideline limit of 50 mg/kg DW (Table 6).

Measured concentrations in the greywater treatment plant sludge from Nordhavnsgården are provided in Table 6. The measured metal concentrations in the Nordhavnsgården greywater treatment sludge confirm that adsorption to suspended solids is an important removal process for these substances during treatment. With median sludge concentrations of 1.1, 24 and 34 mg kg^{-1} DW for Cd, Ni and Pb respectively it is evident that removal of greywater treatment sludge from the WWTP influent load could assist in the reduction of metal loadings in municipal WWTP sludge. The separate treatment and disposal of greywater sludge is an attractive prospect because it is unlikely to contain a significant nutrient content, and yet does effectively concentrate unwanted substances such as metals and nonylphenols. The separation of the greywater treatment sludge from community scale treatment and reuse systems is feasible and could effectively reduce WWTP sludge metal loads without significantly impacting on sludge nutrient value. In contrast, sludge separation from single household system designs is unlikely to be practical and currently these systems are typically designed to periodically backwash or flush particulate matter to the sewerage system.

Table 6: Measured concentrations of Cd, Ni and Pb in Nordhavnsgården greywater treatment sludge and Danish wastewater treatment plant sludge, together with Danish and European sludge guideline limits for the relevant substances. All values are given in mg kg^{-1} DW.

Substance	Measured concentration in Nordhavnsgården primary settling tank sludge[1]	Concentration in Danish WWTP sludge[2]	Danish sludge guideline limits (mg kg^{-1})[3]	European sludge guideline limits[4*]	Proposed European sludge guideline limits in working draft[5*]
Cd	Range: 0.7 – 1.2 Mean: 1.0 Median: 1.1	1995: 1.5 (0.8-6.0) 2002: 1.3 (0.3-3.2)	0.8	20 – 40	10
Ni	Range: 22 - 35 Mean: 27 Median: 24	1995: 25.7 (10-141) 2002: 20 (11-42)	30	300 – 400	300
Pb	Range: 34 - 45 Mean: 37.7 Median: 34.0	1995: 72 (26 – 155) 2002: 50 (11-96)	120	750 – 1200	750
Nonylphenols	No data	1995: 8 (0.3–61) 2002: 4 (1-25)	10	N/A	50

[1] n = 3, 1 sample was taken from the primary settling tank and 2 samples were taken from the biological treatment module

[2] Values given are derived from a national survey of sludge quality in Danish WWTPs and are shown as median values, with the 5th and 95th percentiles in brackets (Jensen and Jepsen, 2005).

[3] Cited in Jensen and Jepsen (2005)

[4] Directive 86/278/EEC (European Commission, 1986)

[5] Working document on sludge, 3rd draft (European Commission, 2000)

[*] Limit value applies to the substances nonylphenol and nonylphenolethoxylates with 1 or 2 ethoxy groups.

4. Conclusions

The results of the conducted scenario analyses are important in the face of increasing pressures on potable water supplies, showing that greywater recycling can potentially save significant volumes of potable water. Within greywater treatment plant, the dominant removal process for a particular pollutant is heavily dependent on the physical, chemical and biological properties of that pollutant. For example, some substances will be more readily biodegraded than others, and some substances will be more susceptible to sorption or volatilisation. The potential for the greywater treatment and reuse system to act as a pollutant emission barrier is thus highly substance dependent. In general, a system such as that installed at Nordhavnsgården will only act as a significant pollutant barrier for substances which are readily biodegradable (but this is not the case for most PS/PHS and certainly not for metals). Thus, on the basis of current designs, which typically do not facilitate separate treatment and disposal of greywater treatment sludge, the results indicate that the potential for extra benefits associated with the emission control of xenobiotics are likely to be quite limited. On the other hand, if greywater treatment sludge were to be removed from the wider municipal WWTP load this could potentially improve the sludge quality and hence help meet the requirements of the various national and European sludge regulations.

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