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Alkylphenol and phthalate contamination of all sources of greywater from French households

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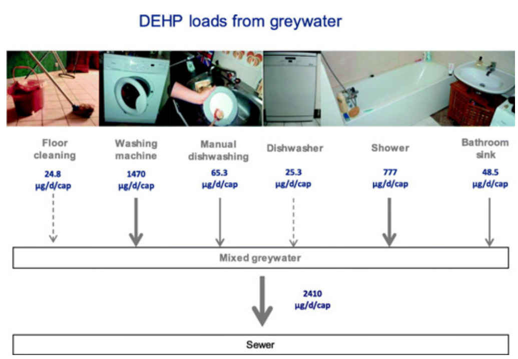
HIGHLIGHTS

- Alkylphenols and phthalates were investigated in six sources of greywater.
- The most contaminated waters were produced by washing machine and floor cleaning.
- Washing machine and shower greywater have the highest phthalate and alkylphenol loads.
- Nature of the products and people habits explained the concentration variability.

GRAPHICAL ABSTRACT



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ABSTRACT

Despite the importance of domestic wastewater, the contribution of greywater to both alkylphenol and phthalate pollution is not yet well documented. Likewise, the detailed emissions of phthalates and alkylphenols by greywater have been insufficiently studied, this work aims to fill this gap. The levels of four phthalates (DEP, DnBP, BBP and DEHP) and two alkylphenols (nonylphenol isomers and octylphenol) were quantified in six different types of greywater, namely that from washing machines, manual dishwashing, dishwashers, bathroom water (from showers and sinks) and floor cleaning. This paper presents the methodology used to characterize all sources of greywater and provides their levels of contamination. The highest concentrations were found in greywater produced by the washing machine and floor cleaning, while washing machine and shower greywater have the highest phthalate and alkylphenol loads because of the volume associated with these two sources of greywater.

1. Introduction

Alkylphenols and phthalates (or phthalate acid ester) are classified as endocrine disruptors. These two families of organic compounds are present in everyday products. Phthalates are commonly used in industry and are components of household products such as adhesives, paints, inks, rubber and surface treatments. Alkylphenols are nonionic

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surfactant agents commonly used as additives in detergents and for the production of alkylphenol ethoxylates. These substances are usually found in cosmetics, personal care products, paints, detergents, herbicides, pesticides, emulsifiers, wetting and dispersing agents, antistatic agents, demulsifiers, and solubilizers.

Few studies have studied alkylphenols and phthalates in domestic wastewater. Paxéus et al. (1992) showed that domestic discharges accounted for >70% of the phthalate loads in wastewater. Besides, Eriksson et al. (2002) and Paxeus (1996) mentioned that alkylphenols in domestic wastewater originate from the chemical products used in households such as detergents. In France, Bergé et al. (2014) showed the significant contribution of domestic wastewater to the load of alkylphenols and phthalates in the Parisian sewer network. Indeed, in this weakly industrialized and highly urbanized catchment, <5% of total loads of these compounds came from industrial discharges (Bergé et al., 2014). Two sources of wastewater are created in a home: greywater and blackwater. To date, only Palmquist and Hanæus (2005) studied phthalates and alkylphenols in these two sources of wastewater separately. They demonstrated that greywater was the most contaminated (Palmquist and Hanæus, 2005). However, this study was performed on a small number of samples (four samples of greywater and three of blackwater) raising the question of the generalization of those results. More recently, Deshayes et al. (2015) focused on greywater originating from showers and washing machines, which make up about 40 and 13%, respectively, of the domestic water consumption in France.

Table S1 provides the values for domestic wastewater obtained in the Paris conurbation (France) by Bergé et al. (2014) and in samples from the area of Maresme (Catalonia, Spain) by Sánchez-Avila et al. (2009). Data from Palmquist and Hanæus (2005), who studied greywater and blackwater separately in two residential areas of Vibyasen (Sweden), are also given. Alkylphenol concentrations, both in greywater and blackwater, were lower than in domestic wastewater (Table S1). This is a striking result since domestic wastewaters are exclusively made up of these two sources of water. A possible explanation for this might be an evolution in the concentrations of alkylphenols in everyday products between 2005 and 2014. Another possible explanation for this is the difference in practices between Sweden, on one hand, and France and Spain, on the other hand. Moreover, Palmquist and Hanæus (2005) demonstrated that, with the exception of DEP, phthalates exclusively originated from greywater. This has been confirmed by Bergé et al. (2014) who reported lower phthalate concentrations in domestic wastewater than those of greywater. It is likely that the explanation lies in the dilution of greywater by blackwater. Unfortunately, despite these three studies on either wastewater or mixed greywater, and the work of Deshayes et al. (2015) on two sources of greywater, we are lacking of consistent information on alkylphenols and phthalates in each source of greywater.

The sources of alkylphenols and phthalates in greywater have not been yet identified. For this purpose, the Central laboratory of the Prefecture of Police (LCPP) in collaboration with the Laboratory water environment and urban system (LEESU) and the Society of water of Versailles and Saint-Cloud (SEVESC) launched a specific research program on alkylphenols and phthalates in greywater. This paper focuses on the monitoring of these emerging pollutants in greywater from six different sources: washing machines, manual dishwashing, dishwashers; bathrooms (from showers and sinks) and floor cleaning.

2. Material and methods

2.1. Sampling methods

The best solution to obtain samples that are as representative as possible of personal habits is to ask the inhabitants themselves to collect the samples. As a consequence, a call for volunteers was done for collecting a maximum of samples. To ensure the representativeness of the samples, volunteers were asked not to change their habits and a guide was published to explain correct sampling practices. Suitable bottles were provided by the Central Laboratory of the Police Prefecture (LCPP). For each type of greywater, samples were initially collected in a single container and, after homogenization, split in four aliquots:

- 500 ml placed in a glass bottle for dissolved and particulate organic carbon (DOC and POC, respectively) and micropollutants;
- 1000 ml placed in a glass bottle for total suspended solids (TSS);
- 500 ml placed in a polyethylene bottle for bio-chemical oxygen demand (BOD_5) and pH;
- And 500 ml placed in a polypropylene bottle for chemical oxygen demand (COD) and total Kjeldahl nitrogen (TKN).

Polyethylene and polypropylene bottles were single use. Blanks were carried out on one sampling bottle, randomly chosen in each lot, in order to verify the absence of contamination. Furthermore, in order to remove all traces of organic contamination, glass bottles were washed with a detergent (Teepol), then rinsed with deionized water before being placed in an oven at 450 °C for 2 h. The sampling method for collecting greywater is detailed below. Samples were kept in a cooler and brought to the laboratory within 24 h of sampling and stored under controlled temperature, i.e., 5 ± 3 °C.

2.1.1. Shower

For this type of greywater, volunteers had to wash themselves in a bathtub. Indeed, it was important to collect the entire volume of water generated by the shower. As a consequence, the volunteers were instructed to plug the bathtub before starting. Then, at the end of the shower, the entire volume was homogenized before collecting subsamples to fill the various bottles previously mentioned.

2.1.2. Washing machine

As with showers, all the water used during a complete cycle of washing machine was sampled. This required a large container (up to 70 l). Once again, the best container proves to be the bathtub. Before turning on the washing machine, volunteers had to rinse their bathtub and put the plug in place. Then, after placing the washing machine's evacuation pipe in the bathtub, they began a laundry cycle as usual. As explained for shower, at the end of laundry, the whole volume was homogenized to get subsamples.

2.1.3. Dishwasher

For dishwashers, the procedure was very similar to that for washing machine. The idea was to collect all the water by placing the standpipe in a 20-l glass bottle. This latter was previously cleaned in the laboratory with detergent (Teepol), rinsed with deionized water before being placed in an oven at 450 °C for 2 h as for sampling bottles. After loading the dishwasher with dirty dishes and dishwashing products, volunteers ran their dishwasher. At the end of the dishwasher cycle, the sample was carefully homogenized by shaking the container and then transferred into the fractionation bottles.

2.1.4. Manual dishwashing

Dishwashing is a two-step process: wash and rinse. Volunteers were asked to scrape dishes to remove leftover food before starting manual dishwashing. Two different procedures were met according to volunteer habits. Indeed, some volunteers cleaned and rinsed dishes in a single kitchen sink (i.e., under a continuous trickle of water), while some others did the dishes in a double kitchen sink. In this latter case, some volunteers preferred filling with hot water both sink for washing and rinsing dishes, whereas some others filled the washing sink with hot water and rinsed dishes under a continuous trickle of water. As a consequence, sampling collection procedures were adapted. For volunteers following the “single-sink” procedure, water used for washing and rinsing was collected in a closed sink. For the “double-sink” procedure,