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To cite this version:
S. Zedek, A. Bressy, S. Deshayes, V. Eudes, E. Caupos, et al.. Triclosan, triclocarban and parabens in greywater: identification of their sources. 26th annual meeting of Society of Environmental Toxicology and Chemistry (SETAC) , May 2016, Nantes, France. hal-01699879

HAL Id: hal-01699879
https://hal.archives-ouvertes.fr/hal-01699879
Submitted on 2 Feb 2018

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Triclosan, triclocarban and parabens in greywater: identification of their sources

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1. Introduction
The use of soaps, shampoos, toothpastes or other personal care products (PCPs) generates discharges of chemicals into the environment through grey- and wastewater. These latter present ecotoxicological potentials (endocrine disruption, fertility reduction, and developmental abnormality). Moreover, chronic and/or possibly synergistic effects of numerous pharmaceuticals and other chemicals even at trace concentrations may also become an issue. Among these chemicals, parabens (PBs), triclosan (TCS) and triclocarban (TCC) are widely used in the composition of PCPs but also in sportive clothes, plastic toys, kitchenware, pharmaceuticals and food products as antiseptic or conservative. Only few researches have published about the occurrence of parabens and triclosan in greywater [1-2]. These previous works indicate that the concentrations can vary in greywater samples from below limit of quantification to few µg.L⁻¹ without information on the different kinds of greywater (shower, washbasin, etc.).

In this context, in a previous study [3] several kinds of greywater from shower, washbasin, manual dishwashing, dishwasher and washing machine have been analyzed. The key lessons highlighted by this work were that (i) greywater strongly contributes to the contamination of wastewater and (ii) washbasins, showers and more surprisingly washing machines are the main contributors (in loads, µg/inhabitant/day) to the contamination of wastewater. PCPs are not therefore the only source of parabens and triclosan in wastewater: dishwashing and washing machine samples appeared as contaminated as shower samples. To reduce micropollutants at source, a better knowledge of the origin of greywater contamination is needed. With these goal and in view of our previous results, new experiments were carried out in order to identify the primary sources of PBs, TCS and TCC in greywater from washing machine and shower.

2. Materials and methods
The identification of the origin of the contamination was assessed through the decomposition of waters from washing machine (n=4) and shower (n=4) into four types of samples (1, 2, 3 and 4) as illustrated in Figure 1. Consequently, several potential sources were investigated:

• **washing machine**: tap water (1), washing machine (2), laundry products (3) and clothes (4);
• **shower**: tap water (1), bathtub (2), personal care products (3) and volunteer (4).

![Figure 1: Decomposition of washing machine and shower](image)

Sampling glassware and a sampling guide for each type of greywater were provided to volunteers for reliable sampling. Hence, volunteers collected their own samples following the given instructions. Experiments were carried out during two days.

Eight compounds were analyzed including six parabens: methylparaben (MeP), ethylparaben (EtP), propylparaben (PrP), benzylparaben (BzP), butylparaben (BuP), isobutylparaben (IsobuP) and TCS and TCC. After filtration (GF/F 0.7 µm) and extraction, the dissolved fraction was analyzed by liquid chromatography coupled to tandem mass spectrometry (LC/MSMS) using MeP-d4 and TCC-d4 as internal
standards. Water quality parameters were also analyzed to ensure the representativeness of samples: pH, suspended solids, organic matter, total Kjeldahl nitrogen etc. 

From the four samples obtained by the decomposition of shower, the contribution (in %) of tap water C1, bathtub C2, volunteer C3 and products C4 to the whole contamination of a complete shower sample (4) were calculated using the following equations (for a given compound X):

\[
C_1 = \frac{([X]_1 - [X]_4)}{[X]_4} \times 100 \\
C_2 = \frac{([X]_2 - [X]_1)}{[X]_1} \times 100 \\
C_3 = \frac{([X]_3 - [X]_2)}{[X]_2} \times 100 \\
C_4 = \frac{([X]_4 - [X]_3)}{[X]_3} \times 100
\]

For washing machine, the contributions were calculated following the same approach. Since isoBuP and BuP were never quantified, no results will be presented thereafter. Since TCS analyzes are still under progress, no result will be presented for that molecule.

3. Results and discussion

This paper focused on the shower decomposition. The averaged dissolved concentrations measured in shower samples for PBs and TCC along with the averaged contributions are given Table 1.

First, whatever the compound, the tap water appears to be not or very slightly contaminated by TCC and PBs. In the same way, the contribution of the bathtub appears to be negligible. Concerning MeP, EIP and PrP, the main contributors are the products (55, 68 and 62 % respectively) and the body (39, 32 and 35 % respectively). For BzP and TCC, products are the only source of contamination.

PBs and TCC are used as antiseptic or conservative in numerous PCPs, including toothpastes, soaps, disinfectants, deodorants and cosmetics products which led to the important contribution of products to the whole contamination. However, the striking contribution of the body was more surprising meaning that an important part of the contaminants (MeP, EIP and PrP) arises from the skin. Three hypothesis have been advanced to explain our results:

- after the use of PCPs, a part remains on the skin;
- a deposition on the skin of air dust containing PBs and TCC;
- and a transfer of contaminants from clothes to skin.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Concentration (ng/L)</th>
<th>Contribution (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1(1)</td>
<td>2(1)</td>
</tr>
<tr>
<td>MeP</td>
<td>1</td>
<td>21</td>
</tr>
<tr>
<td>EIP</td>
<td>&lt; LOQ</td>
<td>&lt; LOQ</td>
</tr>
<tr>
<td>PrP</td>
<td>&lt; LOQ</td>
<td>3</td>
</tr>
<tr>
<td>BzP</td>
<td>&lt; LOQ</td>
<td>8</td>
</tr>
<tr>
<td>TCC</td>
<td>1</td>
<td>9</td>
</tr>
</tbody>
</table>

*Table 1: Averaged concentrations of PBs and TCC in shower samples and their contributions (a= first day ; b= second day)*

4. Conclusions

It has been proved that greywater strongly contributes to the contamination of wastewater by PBs and TCC. The main findings of this study are that personal care products are the main contributor of the contamination of shower water but more surprisingly the body is also a source of contaminants. These results are promising for the promotion of the source reduction linked to changes in consumption practices. Our perspectives are to verify the three hypothesis for the contamination of volunteers body and to interpret the data washing machines. Another perspective would be to improve our knowledge on PCPs contamination by investigating alternative compounds of parabens like methylisothiazolinone.

5. References


