



## Estimation of chemical emissions from down-the-drain consumer products using consumer survey data at a country and wastewater treatment plant level



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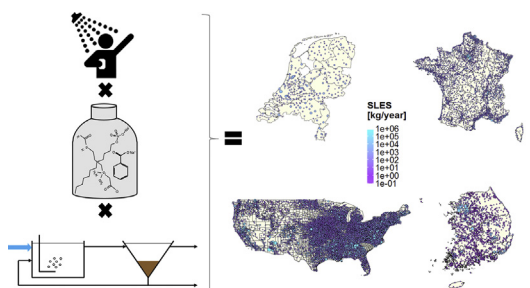
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### HIGHLIGHTS

- Chemical emission loads from the use of personal care products were quantified.
- Consumer use surveys from four countries were used.
- Point source emissions differed up to two orders of magnitude between countries.
- Emission uncertainty (95% confidence interval) was up to 5 times the mean value.
- This approach does not rely on confidential or commercial tonnage data.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Deriving reliable estimates of chemical emissions to the environment is a key challenge for impact and risk assessment methods and typically the associated uncertainty is not characterised. We have developed an approach to spatially quantify annual chemical emission loads to the aquatic environment together with their associated uncertainty using consumer survey data and publicly accessible and non-confidential data sources. The approach is applicable for chemicals widely used across a product sector. Product usage data from consumer survey studies in France, the Netherlands, South Korea and the USA were combined with information on typical product formulations, wastewater removal rates, and the spatial distribution of populations and wastewater treatment plants (WWTPs) in the four countries. Results are presented for three chemicals common to three types of personal care products (shampoo, conditioner, and bodywash) at WWTP and national levels. Uncertainty in WWTP-specific emission estimates was characterised with a 95% confidence interval and ranged up to a factor of 4.8 around the mean, mainly due to uncertainty associated with removal efficiency. Estimates of whole country product usage were comparable to total market estimates derived from sectorial market sales data with differences ranging from a factor 0.8 (for the Netherlands) to 5 (for the USA). The proposed approach is

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suitable where measured data on chemical emissions is missing and is applicable for use in risk assessments and chemical footprinting methods when applied to specific product categories.

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## 1. Introduction

Many of our everyday actions, like household cleaning or taking a shower, involve the use of consumer products (CPs). After use, such products are often released with household wastewater and their constituent chemicals or ingredients may end up in the environment after passing through the sewerage and wastewater treatment system. To assess potential environmental impacts and safe use of the chemicals, techniques such as risk assessment and chemical footprinting methods are employed (e.g. Bjorn et al. (2014); Salvito et al. (2001)). These methods require a reliable quantification of the amount of chemicals used and subsequently released into the various environmental compartments, such as freshwater. Concentrations measured analytically would best reflect the chemical's inflow into a specific catchment (Earnshaw et al., 2014; Kasprzyk-Hordern et al., 2008; Lindim and Cousins, 2015; Whelan et al., 2012). Unfortunately, monitoring data for many chemicals is poor or not available with the exception of certain classes or problematic chemicals. Furthermore, where monitoring data exists, it is often temporally and spatially limited (Petrie et al., 2015). New modelling approaches are therefore required to provide more reliable and realistic estimates of chemical emissions associated with consumer products.

When deriving chemical emission estimates, a measure of their uncertainty is desirable (Ascough et al., 2008; van der Sluijs, 2002) even more since this has been identified as a deficiency in the application of risk assessment and chemical footprinting methods (Bjorn et al., 2014; Chevre et al., 2013; Harbers et al., 2006). For example, in their work on the risk assessment of chemicals from wastewater, Escher et al. (2011) only used a worst-case scenario approach to quantify emissions of pharmaceuticals in wastewater from hospital usage data. In contrast, Oldenkamp et al. (2016) determined the uncertainty in spatially-explicit pharmaceutical emission estimates resulting from substance characteristics and consumption volumes. However, this work did not consider the variability induced by different consumption habits and assumed one absolute per-capita consumption value per pharmaceutical considered. In their top-down approaches where sales data of personal care products at a global level are used to derive smaller scale chemical emissions, Hodges et al. (2014); Price et al. (2010); Whelan et al. (2012) acknowledged the uncertainty of their estimates, but without quantifying it.

Chemical emissions from CPs can be estimated from product use amounts and product formulations. Two approaches exist to derive product use amounts: total market or sales based (top-down) and individual consumption-based (bottom-up). Total market consumption data may be collected directly from manufacturers (Salvito et al., 2001) or derived by commercial market research organisation (e.g. Euromonitor) (Hodges et al., 2014; Keller et al., 2007; Price et al., 2010). However, access to such data is often restricted to commercial companies and the confidentiality of the data and methods used to generate them limits their transparency and reproducibility by others. Although the average consumption approach is described in the OECD Emission Scenario Documents (ECHA, 2000), worst-case default values are generally used to risk assess chemicals. An exception is for pharmaceuticals where consumer usage data were readily used to estimate emissions into

wastewater (Escher et al. (2011), Chevre et al. (2013), and Oldenkamp et al. (2016)). However, to our knowledge, bottom-up approaches have not yet been used for larger scale estimates of more mass market products, such as personal care products, which are typically associated with a wide range of habits and common set of widely used chemicals.

The goal of this paper was to develop a consumer use-based approach to estimate the annual chemical emission loads from the use of personal care products at the country and the WWTP level. We explored how a bottom-up approach starting with easily accessible consumption data derived from consumer surveys can be used to estimate product related chemical emission loads. To illustrate the potential of this approach, three personal care product types were used, namely shampoo, conditioner, and bodywash. These products were chosen because of available consumer survey reports and because they are expected to be almost completely washed off down the drain after use. The study focussed on common chemicals used across the product categories namely three surfactants: sodium lauryl ether sulfate (SLES), cocamidopropyl betaine (CAPB), and cetearyl alcohol (CA) and two preservatives: sodium benzoate (SB) and dimethyloldimethyl hydantoin (DMDMH). As the consumer surveys also report consumer characteristics, variability in consumer behaviour was studied by considering different user category groups, based on age and gender. In addition, the uncertainty in consumer behaviour due to the limited size of the assessed consumer groups was quantified. Combined with uncertainties inherent to chemical inclusion levels in products and to removal efficiencies from wastewater, uncertainty estimates of chemical emissions from WWTPs were quantified.

## 2. Materials and methods

### 2.1. Framework

Chemical emissions from the use of CPs were estimated for each country as well as for every single WWTP using Equation (1). Population data was hereby taken from census data.

$$M_{X,i} = 365 \cdot \sum_{P=1}^m F_{mass,X,P} \cdot F_{X,P} \cdot (1 - E_X) \left( \sum_{c=1}^n A_{P,c} \cdot F_{PU,P,c} \cdot N_{c,i} \right) \quad (1)$$

$M_{X,i}$  Emission from WWTP  $i$  of chemical  $X$  [g/year]

$m$  Number of product classes considered: shampoo, bodywash, and conditioner

$n$  Number of categories  $c$  of persons considered, men and women when possible differentiated by age

$P$  Product class

$C$  Consumer category

$A_{P,c}$  Amount of product  $P$  used per person of category  $c$  per day [g/person/day]

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