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Possible underestimations of risks for the environment due to unregulated emissions of biocides from households to wastewater

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ABSTRACT

The aim of this study was to investigate the role of household products as possible sources of biocidal active substances in municipal wastewater and their regulation under the Biocidal Products Regulation (EU) 528/2012. In 131 households, we investigated the prevalence of products used to control pests, washing and cleaning agents and select personal care products with high release to wastewater. Inventories of these products were established with the help of barcode scanning. All uses of biocidal active substances were evaluated regarding their assessment under the Biocidal Products Regulation.

2963 products were scanned in total, with 48% being washing and cleaning agents, 43% personal care products and 9% products used to control pests. Biocidal active substances were found in each household. These were observed primarily in washing and cleaning agents and personal care products (90%), while only a small percentage of the observations of biocidal active substances was in biocidal products. 64% of the observations of biocidal active substances were in applications that do not fall under the Biocidal Products Regulation and are thus not subject to its environmental risk assessment.

This study shows clearly that risks for the environment are underestimated because unregulated emissions to wastewater occur. It demonstrates that there are gaps in the current chemical legislation that lead to a release of substances into wastewater that were not subject to environmental risk assessment under the Biocidal Products Regulation. This is one example of the limitations of scientific risk assessment of chemicals – its complexity is immense. From our point of view, the results underline the importance of a sustainable use of the substances as this is the only way to decrease yet unidentified risks.

1. Introduction

Contamination of the environment with chemicals is still a challenge, and preventive approaches are needed to mitigate global chemical pollution (Diamond et al., 2015). Emissions of micropollutants from households are one of the threats to the water quality of aquatic systems. Recently, biocidal active substances (BAS), defined as "substance[s] or […] micro-organism[s] that [have] an action on or against harmful organisms" (European Union, 2013), have come into focus since they were first observed in the aquatic environment. Recent studies have shown that BAS can be found in different water bodies (Brausch and Rand, 2011; Buergi et al., 2007; Reemtsma et al., 2006; Weigel et al., 2002), biota (Corcellas et al., 2015; Rüdel et al., 2013) and also in human urine (Frederiksen et al., 2014; Heffernan et al., 2015; Larsson et al., 2014). They can pose a risk for organisms due to their, by definition, intended effects on organisms. These effects are not limited to the environment, but can also be relevant for human health. Potential risks have been identified e.g. for pest control using sprays, spraying of disinfectants or cleaning of surfaces with concentrates (Hahn et al., 2010). BAS can be sensitizing (e.g. methylchloroisothiazolinone/methylisothiazolinone (Geier et al., 2012)) and their contribution to evolving resistances against antibiotics due to cross-resistance is still under discussion (SCENIHR, 2009). The use of the disinfectant benzalkonium chloride for example could trigger antibiotic resistance against fluoroquinolones (Buffet-Bataillon et al., 2016). Especially the use of disinfectants in households has been discussed because the inappropriate use of non-approved disinfectants can lead to risks (Pieper et al., 2014) and the benefit of disinfecting soaps containing triclosan is questioned (Kim et al., 2015).

Approaches to reduce environmental contamination at the source are needed for these kinds of micropollutants instead of end-of-pipe technologies (Kümmerer et al., 2015). The manifold emission routes of biocides due to their diverse applications make, however, this approach

Abbreviations: BAS, biocidal active substance; BP, biocidal products; ERA, environmental risk assessment; BPR, EU Biocidal Products Regulation 528/2012; PCP, personal care products; PPP, plant protection products; PT, product type; STP, sewage treatment plants; WCA, washing and cleaning agents.

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exceedingly difficult. BAS can be released into the environment via direct and indirect emissions. Direct emissions can occur e.g. through run-off from building materials such as roofs or outdoor paints on facades (Bollmann et al., 2014b; Burkhardt et al., 2011; Gromaire et al., 2015). Indirect emissions to the environment can occur through sewage treatment plants (STP), where not all substances are completely removed (Chen et al., 2012; Gasperi et al., 2014; Kupper et al., 2006; Morasch et al., 2010; Singer et al., 2010; Weston et al., 2013; Wick et al., 2010). Households are likely to be major contributors to the total amount of BAS in STP. However, the specific sources within households are not yet fully understood (Bollmann et al., 2014a; Wittmer et al., 2011).

1.1. Regulation

In the EU, biocidal products (BP) are regulated under the *Regulation (EU) No 528/2012 concerning the making available on the market and use of biocidal products* (BPR). Applications for approval of BAS falling under BPR have to be submitted for each of the 22 different product types (PT), as described in Annex V of the BPR, in which the substance is intended to be used for a biocidal purpose. The European Chemicals Agency lists 262 BAS whose risks are currently assessed by the Member States ("under review") or are already approved in the EU. Approval has been sought for 685 active substance-PT combinations (European Chemicals Agency, 2015b). But BAS can simultaneously be used in other product groups, which do not fall under the BPR, e.g. plant protection products (PPP), personal care products (PCP) or washing and cleaning agents (WCA). Article 2 of the BPR defines exemptions for these products falling under the scope of other regulatory instruments. The regulatory differentiation between BP and product groups falling under other regulations is complex (Woutersen et al., 2015). This can lead to borderline cases, for which it has to be decided on a case-to-case basis under which provision a product is regulated (European Commission, 2015). This decision is based on the intended field of application of a product.

The consequence is that emissions of identical substances from applications, which are subject to different regulations, are not aggregated during the separate risk assessments. Thus, possible wastewater emissions of BAS are not completely evaluated and environmental risks are underestimated. Information and data are, therefore, needed to close this knowledge gap regarding these possible emissions of BAS from households into wastewater and to what extent they are regulated under BPR. Only with this knowledge, it is possible to fully understand the environmental risks posed by BAS.

1.2. Information on ingredients

Information on the ingredients of products in Europe is regulated under the respective legislation for product categories. As BAS can be found as ingredients in WCA, PCP and BP, the regulatory background for the labelling of these products is important. This background allows for a qualitative assessment of all BAS used in these categories, but not for quantitative questions:

- In accordance with Annex VII of *Regulation (EC) 648/2004 on Detergents*, manufacturers of detergents have to make available a list of names of ingredients on a website (European Union, 2005). However, no information is given on the amounts of the respective substances in the products.
- For PCP, Article 19(1) of the *Regulation on Cosmetic Products* requires a list of ingredients on the packaging. As in the case of WCA, no information on amounts is required. To gain information regarding the exact amounts of the substances, product testing would be necessary, which is not possible in light of the huge number of products (Dudzina et al., 2014).
- For BP, the authorisation holders are, as stipulated by Article 69(2) of the BPR, required to state the identity of every active substance and its concentration on the label of the products.

1.3. Data collection

Consumption data from households would be the most convenient way to collect information on emissions of BAS from households, as it has been done for pharmaceuticals in the past (Herrmann et al., 2015; Le Corre et al., 2012). However, unlike for pharmaceuticals, consumption data for BAS is currently not available. Besides a chemical characterisation of wastewater from households, the enquiry of consumption data by product inventories is a promising approach to examine emissions from households into wastewater. For a collection of data on the prevalence of BAS, different approaches can be used, e.g. telephone interviews, self-administered (e.g. Internet) surveys or on-site visits (Hertz-Picciotto et al., 2010). Each of these methods has disadvantages. Accurate reporting of used products cannot be expected during telephone interviews (Wu et al., 2010), internet surveys have low response rates and on-site visits such as household visits are intrusive (Hertz-Picciotto et al., 2010) and time-consuming for researchers (Weegels and van Veen, 2001). However, on-site visits are the most promising approach to collect detailed information. During the visits, it is especially important for the acceptance of households, to establish trust between researchers and interviewees and to minimise the time needed (Hertz-Picciotto et al., 2010). A highly sufficient approach to reduce the time required is the use of barcode scanners to inventory present products either by the researcher (Bennett et al., 2012) or by the consumer (Hall et al., 2007). Household investigations that included BAS, but were not focused on them, were conducted in Europe, e.g. in 30 households in one building in Copenhagen by on-site visits (Eriksson et al., 2003), in 2281 households in France by telephone interviews and 23 households close to Angers and Nantes in France by on-site visits (ANSES, 2010). Among other factors, such as age, gender or education, the living conditions are considered to be an important factor for the use of biocidal products (ANSES, 2010). However, until now no studies exist examining the correlation of the use of biocidal products and demographic factors in detail.

1.4. Objectives

Because the studies mentioned above have focused on certain product groups, either on PPP and BP or on PCP and WCA, their results give no overall picture of the sources of BAS in households. In addition, the use of the BAS in households has never been evaluated regarding its coverage by environmental risk assessments. For this reason, the objective of our study was to generate new and urgently needed data on the overall prevalence of BAS in household products in different categories: We aim to identify possible emission sources of BAS from households into wastewater and to examine whether the respective products are subject to environmental risk assessment (ERA) under the BPR. In the following, we present (1) which BAS are used in the studied households and (2) the products they are found in. The uses of BAS in different product categories are then (3) evaluated regarding their regulation under BPR or other legislation.

For this study, we chose on-site visits as a method to collect data and tested the applicability of product inventories using barcode scanners in households of three neighbourhoods in northern Germany. The product inventories were limited to products with a relevant release to wastewater because we considered this to be the most important exposure pathway of BAS from household products into the environment.

2. Material and methods

2.1. Study sites

In total, product inventories in 131 households were recorded. To account for different living conditions, that have been deemed to be an important factor for the use of biocidal products by ANSES (2010), three neighbourhoods were included in this study. These are representative

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