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Risk assessment of triclosan released from sewage treatment plants in European rivers using a combination of risk quotient methodology and Monte Carlo simulation

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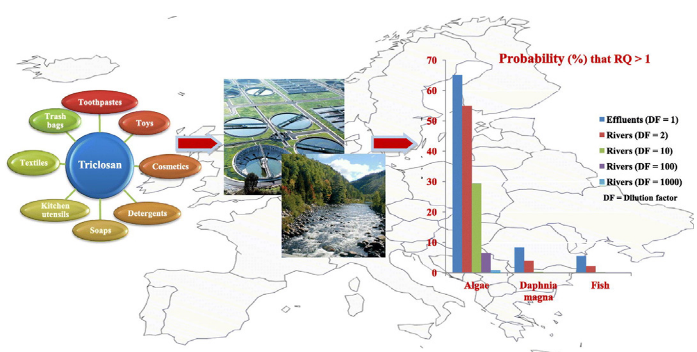
HIGHLIGHTS

- The risk due to triclosan was evaluated in European STPs' effluents and rivers.
- A probabilistic risk assessment, using Monte Carlo simulation, was developed.
- For algae, 95th percentile RQ > 1 was calculated in rivers with DF up to 100.
- The probabilities of RQ to exceed 1 ranged from 0% to 54%, depending on the DF.
- TCS monitoring should be intensified to verify the predicted results.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study a probabilistic risk assessment was applied to investigate the environmental risks for the European aquatic environment associated with triclosan (TCS) occurrence in treated wastewater. The concentrations of TCS in effluents of European Sewage Treatment Plants (STPs) were recorded through literature review, while toxicity data was collected for three groups of aquatic organisms (algae, *Daphnia magna* and fish). The ratio of Measured Environmental Concentration (MEC) and Predicted No Effect Concentration (PNEC), expressed as a Risk Quotient (RQ), was calculated for risk characterization, while Monte Carlo simulation was applied to quantify the associated uncertainty. TCS monitoring data was available for 349 STPs located in 15 out of the 50 European countries. Its mean concentrations in STPs effluents ranged between 2.2 ng L^{-1} and 47,800 ng L^{-1}. Higher TCS concentrations were observed in primarily treated wastewater; whereas no differences among countries or among secondary and tertiary effluents on the basis of the whole set of collected data were found. The 95th percentile of RQ for TCS was higher than 1 (in algae) for rivers with dilution factors (DFs) equal to or lower than 100, when the maximum concentration values were used, whereas the 95th percentile of RQ exceeded 1 for rivers with DFs up to 10, in cases where the calculations were based on mean concentration values. The probability that RQ exceeds 1 in rivers (for algae) ranged from 0.2% (DF = 1000) to 45% (DF = 2), when calculations are based on mean concentration values. The corresponding probabilities in rivers with DFs equal to 2 for *Daphnia magna* and fish were 0.7% and 0.4%, respectively. We propose that TCS monitoring should be intensified, especially on smaller rivers, to verify the findings of this study for possible environmental risks.

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1. Introduction

Sewage Treatment Plants (STPs) are considered major point sources of emerging contaminants into the environment due to their partial removal during applied wastewater treatment processes (Bletsou et al., 2013; Stasinakis et al., 2013; Luo et al., 2014). Triclosan (TCS) is a broad-spectrum antimicrobial agent contained in personal hygiene products, as well as in kitchen utensils, toys, textiles, socks and trash bags (Bester, 2003; Roberts et al., 2014; Gao et al., 2015). It is has been placed on the list of the 10 most frequently detected organic micropollutants in the aquatic environment (Huang et al., 2016; Zhang et al., 2016); it has been detected in 57.6% of U.S. streams (Kolpin et al., 2002), while recently Von der Ohe et al. (2012) detected TCS in 62.7% of water samples from Elbe River. As well as of environmental samples, TCS has also been detected in human body fluids. In a recent study, Han et al. (2016) reported that the detection frequency of TCS in urine samples of U.S. residents for the period 2003–2012 was higher than 72%. Regarding its toxic effects, there are many studies describing TCS endocrine-disrupting properties as well as its toxicity to different (micro)organisms (Bedoux et al., 2012; Lankester et al., 2013). In the United States, a discussion among scientists, authorities and agencies concerning the safety of TCS and the need to regulate it has been in progress during the last decade. Although many scientific and technical gaps still appear to exist, the U.S. Environmental Protection Agency (EPA) has regulated TCS as a registered pesticide under the Federal Insecticide, Fungicide and Rodenticide Act (FIFRA) (Cooney, 2010; Halden, 2014a, 2014b; De Leo and Sedlak, 2014); while recently TCS was also banned by the Food and Drug Administration (FDA) (2016). In the European Union (EU), except for the *Opinion on Triclosan* propounded by the Scientific Committee on Consumer Safety, which recommends the prudent use of TCS (Scientific Committee on Consumer Safety, SCCS, 2010) and the Decision 2016/110, which disapproves the use of TCS in biocidal products, (European Commission, EC, 2016), similar regulations have not been adopted, while TCS has not been included in the list of compounds that should be regularly monitored. On the other hand, Von der Ohe et al. (2012) conducted a risk assessment in Elbe River and suggested that, although monitoring data for many river basins are lacking, this compound should be considered as a candidate for monitoring and prioritization.

Risk quotient (RQ) methodology, wherein RQ is calculated on the basis of Measured Environmental Concentration (MEC) and Predicted No Effect Concentration (PNEC) of the target substance (European Commission, EC, 2003) has been widely applied in the literature to estimate the possible threat posed by micropollutants to the environment. If RQ is lower than 1, the ecological risk posed by the micropollutant is negligible, whereas when values are equal or greater than 1, adverse effects on organisms are probable (Stasinakis et al., 2008; Stasinakis et al., 2012; Cho et al., 2014). Regarding TCS, two approaches consistent with the Technical Guidance Document (TGD) on Risk Assessment have been used for PNEC deduction (European Commission, EC, 2003): the Assessment Factor (AF) method and the statistical extrapolation technique (Species Sensitivity Distribution, SSD) (Capdevielle et al., 2008; Lyndall et al., 2010; Gottschalk and Nowack, 2012). Recently, Thomaidi et al. (2015, 2016) applied the AF methodology on a country level and indicated a presumable threat for the Greek aquatic and terrestrial environment due to the existence of TCS in STP effluents and sludge, respectively. So far, no similar study has been conducted on a European level as the aforementioned methodology has been mainly applied either to specific pollution sources (*e.g.* hospitals, factories) and/or specific water effluent receivers (*e.g.* rivers, lakes) (Stasinakis et al., 2012; Frédéric and Yves, 2014; Carbajo et al., 2015). Additionally, as RQ values are usually calculated based on the maximum MEC and the lowest PNEC values, this methodology provides information for the worst-case scenario. Although this is tempting in its simplicity, no information is given for the uncertainty of the method and the possibility that RQ values exceed 1.

Based on the above, the main objectives of the current study are to collect data for the occurrence of TCS in European treated wastewater and apply the RQ methodology to estimate the possible risk to the European aquatic environment. To achieve this, we compile TCS concentration levels in treated wastewater reported in the literature since 2002 for all European countries and we collect toxicity data from peer-reviewed literature for algae, *Daphnia magna* and fish. For the PNEC estimation we apply the AF method. To estimate the threat associated with the presence of TCS in European rivers, four scenarios are developed, based on different dilutions of the treated wastewater (2, 10, 100 and 1000). In order to underpin the reliability of the RQ methodology, an uncertainty analysis is conducted using Monte Carlo simulation. The findings of the current study will be useful to risk assessors, policy makers and authorities to decide whether there is a need for future TCS monitoring or/and prioritization in surface waters.

2. Materials and methods

2.1. Monitoring data collection

An extended literature review was initially conducted between May and July 2016, to collect monitoring data of TCS in effluents of European STPs. The review was carried out for all European countries (50 in total), including those that are not members of the European Union. Data from 69 international articles, dated from 2002 to 2015, was retrieved using the Scopus database. The search terms were “triclosan” AND “concentration OR occurrence OR monitoring” AND “wastewater OR effluents OR sewage” AND “the name of the country”. The studies covered a total of 349 STPs. The minimum, maximum, mean and median TCS concentration values were recorded for each study, as well as the type (grab or composite) and number of samples, the type of sewage treatment and the affiliation country (Table S1).

In order to determine whether there are any statistically significant differences between the effluents' concentrations determined in different countries, a one-way analysis of variance (Anova) was conducted using IBM SPSS Statistics Base 24. A similar analysis was carried out to investigate the statistically significant differences between TCS concentration in treated wastewater of STPs offering primary, secondary or tertiary treatment.

2.2. Toxicity data collection

The literature data on ecotoxicological acute and chronic effects of TCS on different groups of aquatic organisms was collected from 34 international articles, dated from 1986 to 2016. The search was conducted between July and August 2016, using the Scopus database as the source of publications. The search terms were “triclosan” AND “aquatic toxicity OR EC50 OR LC50 OR NOEC”. Experimental effective/lethal concentration at a 50% level (EC50/LC50) and no-observed effect concentration (NOEC) values obtained for algae, *Daphnia magna* and fish were collected. Additional information, concerning the target aquatic organism species, the type and the duration of the toxicity test was also recorded (Table S2). The selection of the studied organism groups and the dose descriptors was consistent with the Technical Guidance Document on Risk Assessment issued by the EU (European Commission, EC, 2003) and the AF methodology applied in the literature for estimating the ecological threat due to the existence of micropollutants in wastewater (Stasinakis et al., 2012; Frédéric and Yves, 2014; Carbajo et al., 2015; Chen et al., 2016).

2.3. Probabilistic risk assessment

In order to assess the potential risk associated with the presence of TCS in the aquatic environment, the RQ calculations were based on the Measured Environmental Concentration (MEC) of the target compound in effluents, the Predicted No Effect Concentration (PNEC) for 3