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Risk assessment of triclosan in the global environment using a probabilistic approach

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ABSTRACT

The occurrence of antimicrobial agent triclosan (TCS) in the global aquatic and terrestrial environment is an emerging concern. While risk assessment for TCS is available in certain countries, no studies have attempted to assess the risk of TCS worldwide. This could be due to lack of method to characterize the global exposure. The present study therefore proposed a probabilistic-based approach to approximate the percent-ranked measured environmental concentrations (MECs) by estimating exposure concentration distribution (ECD) for different environmental compartments on a global scale, incorporating approximate 1200 single MECs. Hazard of TCS was assessed from species sensitivity distribution as well as predicted no effect concentrations (PNECs) derived from ecotoxicological and toxicological endpoints. We draw on experiences from previous risk assessment exercises and present a holistic approach for characterizing the risk of TCS to microorganism in sewage treatment plant, aquatic and soil organisms, avian and mammalian species, and humans. Using the approach, we estimated risk of TCS to organisms dwelling in sediment and living in surface waters, and the risk quotients (MEC/PNEC) were within the range of 0.95 – 33.3 and 0.49 – 9.5, respectively. While the risk quotients for other environmental compartments were below a value of 1, there are large uncertainties likely due to an insufficient dataset of exposure and hazard of TCS.

1. Introduction

Triclosan (TCS) is an antimicrobial agent formulated in a diverse group of personal care products (PCPs) including soaps, toothpastes, cosmetics, antibacterial soaps and body washes. Unlike pharmaceuticals which are designed for internal use, PCPs are products intended for external use on human body and are hardly subjected to metabolic alterations (Brausch and Rand, 2011). TCS can thus be released into the environment at various stages in their life-cycle. Following the regular usage, TCS will be emitted to sewage treatment plant (STP) and then be released directly into aquatic environment. With a high log Kow of 4.76, TCS is likely to be adsorbed onto organic particles in sewage sludge during STP process and then be transported to terrestrial environment via sludge application (Dhillon et al., 2015). As a consequence, the contamination of TCS in surface water and terrestrial environment has been reported worldwide with concentrations ranging from ng/L to low μg/L levels, i.e. the occurrence of TCS in surface water has been recorded from 0.011 μg/L in Japan to 0.43 μg/L in US (Nishi et al., 2008; Kolpin et al., 2002). The physio-chemical properties of TCS indicate its high bioaccumulation potential to non-target organisms in the environment (Dhillon et al., 2015). Because of the bioconcentration factors (BCF) of TCS ranging from 2532 at 30 μg/L to 4157 at 3 μg/L for zebrafish (*Danio rerio*; Aldous et al., 2009), there is a noticeable bioaccumulation of TCS in aquatic organisms, i.e. a maximum concentration of 150 μg/kg was found in filamentous alga (*Cladophora spp.*) in the receiving stream for a STP (Coogan et al., 2007).

TCS is designed to prevent bacterial propagation and/or eventually results in cell death. It permeates the bacterial cell wall interacting with multiple cytoplasmic and membrane sites, and inhibits RNA synthesis and the production of macromolecules (Dann and Hontela, 2011). However, due to the conservation of similar receptor systems, non-target organisms (e.g., protozoa, algae, rotifers, insects, crustaceans, and fish) in the environment can also be affected (Boxall, 2004). For aquatic organisms, the chronic no observed effect concentration (NOEC) ranged from 0.015 μg/L for effects on the population of algae *Chlamydomonas sp.* to18000 μg/L for bluegill (*Lepomis macrochirus*) based on the endpoint of mortality (EPA, 2017). For terrestrial species, the NOEC and 10% lethal concentration (LC_{10}) values ranged from 0.16 mg/kg dry soil based on a 21d test on the root weight of tomato (*Solanum lycopersicon*) to 250 mg/kg dry soil tested on Springtail (*Heteromurus nitidus*) for the effects on population (EPA, 2017). In view of the occurrence worldwide and the adverse effects on a variety of non-

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target species, TCS has already been a concern of scientific communities and regulatory authorities, especially in western countries (Halden, 2014; HCEC, 2012). As a result, European Union (EU) has disapproved the use of TCS in biocidal products on 27 January 2016 due to its unacceptable environmental risk (EU, 2016). In US, over-the-counter (OTC) consumer antiseptic wash products containing TCS can no longer be marketed after 2 September 2016 (FDA, 2016).

While a few of risk assessment studies of TCS are available for regional environment (e.g. Canada), the environmental risk was characterized by using the standard deterministic procedures (HCEC, 2012). This approach estimated the ratio of predicted environmental concentration (PEC) relative to the predicted no effect concentration (PNEC) based on the published ecotoxicology data of the most sensitive species. The estimation of PEC largely relies on the annual usage (e.g. known as prescription cost analysis (PCA) in UK) which is usually unavailable in most of countries (Guo et al., 2016b). In addition, by the application of this conventional approach, it is unlikely to estimate PECs in each environmental compartment on a global scale. The deduction of PNEC based on the lowest chronic ecotoxicity data has been debated by scientific community and policy makers in viewpoint of whether the selected species could guarantee the safety for other organisms (TGD, 2003).

Probabilistic-based exposure assessment combines all the available measured environmental concentrations (MECs) at both national and / or global level to produce a single cumulative distribution curve, namely exposure concentration distribution (ECD), which characterize the magnitude and frequency of contaminant (EUFRAM, 2005). ECD allows the graphic estimation of overall 50th and 95th percentile MEC values for a direct comparison with the effects (Straub and Stewart, 2007). For hazard assessment, species sensitivity distribution (SSD), known as a statistical extrapolation method deriving PNEC using ecotoxicity data for all taxa rather than a single taxonomic category, has been well established (TGD, 2003; Guo et al., 2016a). By integrating casual relationships between exposure and effect levels of multiple types of animals and plants, the sensitivity distribution of these species can quantitatively assess the effect of human interference (BWN, 2015). An estimated HC5 (hazardous concentration for 5% of species) of a target chemical is deemed to be protective of 95% of species in the environment (Wheeler et al., 2002). Nonetheless, few studies have applied this approach for risk assessment of TCS.

At present little is known about the environmental risk of TCS on a global scale, despite an increasing number of scientific publications focusing on the environmental exposure and toxicity of TCS in each environmental compartment (all types of waters, sludge, and sediment, etc.) are available over the last decade (Dhillon et al., 2015; Dann and Hontela, 2011). No studies have attempted to make use of these data for assessing the emerging health concerns and the potential environmental impact of TCS (Dann and Hontela, 2011). This study, therefore, proposed a probabilistic approach for assessing the risk of TCS by comparing the percent-ranked MECs and PNECs. The aim of this study is to comprehensively evaluate the risk of TCS in microorganism in STP, aquatic and terrestrial species, and humans in the global environment.

2. Methods

To evaluate the risk of TCS in the global environment, we applied risk quotient (RQ) as the primary parameter (Fig. 1). RQ values were estimated by comparing the 50th and 95th percentile of measured environmental concentrations (MEC50 and MEC95, respectively) in various environmental compartments to measurements of potential hazard toward organisms from different trophic levels. This risk assessment approach considered the ecotoxicological and toxicological effects of TCS on aquatic, terrestrial and sediment dwelling organisms, as well as microorganisms in STP, and birds and mammals. In the following section, the estimated exposure concentrations and hazard parameters are described. Methods of specific calculations are given in the Supplemental Data.

2.1. Data collection

Physico-chemical properties (e.g. octanol / water partition coefficient (Kow), organic carbon – water partitioning coefficient (Koc), bioconcentration factor (BCF)) were retrieved from a wide range of literatures and government official reports (e.g. HCEC, 2012; Aldous, 2009); Chronic ecotoxicological and/or toxicological data (e.g. NOEC, no observed adverse effect concentration (NOAEC)) for TCS were retrieved from ECOTOX EPA (2017). MECs of TCS in each environmental compartment were collated from literatures. We searched literatures published after 2002 in Scopus by employing four search terms: 1) Triclosan AND Wastewater AND Occurrence, 2) Triclosan AND Occurrence AND Environment, 3) Triclosan AND Water AND Occurrence, and 4) Triclosan AND soil AND Occurrence. From each retrieve, we identified 130, 104, 187, and 28 papers, respectively. All these papers have been reviewed to collate the global exposure of TCS in each environmental compartment (Table 1).

2.2. Environmental exposure estimation

The MECs of TCS were reported as median, minimum to maximum values, or the individual data of all the analyses in literatures. For literatures with sufficient information, MECs in all the analyses were extracted (Straub and Stewart, 2007; Straub, 2013). For other studies which only reported MECs as range, an average of minimum and maximum concentrations was calculated and applied to derive exposure concentration distribution (ECD). As for data less than the method detection limit (MDL), a value equal to half of the MDL was given as a surrogate (Solomon et al., 2000).

In terrestrial compartment with limited occurrence data of TCS, MECs were estimated by transforming the reported concentrations in STP sludge (Supplemental Data, Equation S1). Here application of STP sludge for soil as fertilizers was considered as the only pathway for TCS entering the terrestrial environment (TGD, 2003). For the compartments of surface water, land, STP effluent, sediment and drinking water, ECDs were derived by cumulative distribution functions in Excel 2010 software. Specifically, all the MECs were percent-ranked and plotted on a logarithmic (concentrations, x-axis) versus probability (percentiles, y-axis) graph. Based on the composite MECs, the MEC50 (50th percentile) and the MEC95 (95th percentile) were estimated from ECD for compartments mentioned above (Straub and Stewart, 2007). The effluent concentration of TCS was applied for the exposure assessment for microorganisms in activated sludge of a STP (TGD, 2003). With the assumption of homogeneous mixing in aeration tank, the really dissolved concentration of a substance in activated sludge approximates effluent concentration. Here, only the dissolved substance is assumed to be bioavailable to the microorganisms in activated sludge (TGD, 2003). PEC of TCS in fish as food (PEC_{fish}) was estimated using the BCF, biomagnification factor (BMF) and the MEC in surface water ($MEC_{sw}50$/$MEC_{sw}95$; Supplemental Data, Equations S2). PEC of TCS in earthworm ($PEC_{earthworm}$; Supplemental Data, Equations S3) on a wet weight basis was estimated using the concentration in porewater (Supplemental Data, Equations S4) and the BCF for earthworms (Supplemental Data, Equations S5), following the approach given in TGD, 2003.

2.3. Hazard characterization

PNECs of TCS were derived from ecotoxicological data, using an appropriate assessment factor (AF) from the technical guidance document (TGD, 2003). When effect data on a wide range of species were available (e.g. direct toxicity to freshwater and terrestrial organisms), SSD approach was applied for hazard characterization. In the cases of organisms such as the microorganism, bird and mammal, whose data of