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Substance flow analysis and assessment of environmental exposure potential for triclosan in mainland China

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HIGHLIGHTS

- 78% of TCS consumed in 2008 unsustainably discharged into the natural environment.
- Most TCS in China is discharged to surface water sediment, ocean, and soil.
- Higher consumption levels increased TCS accumulation in the environment.
- Environmental exposure potential to TCS in China is increasing.
- SFA is a well-established tool to provide information for exposure assessment of TCS.

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ABSTRACT

Triclosan (TCS) is a widely-used antimicrobial agent in many consumer products around the world, and China is a major producer and consumer of TCS. In this study substance flow analysis (SFA) was used to construct a static model of anthropogenic TCS metabolism in China in 2008. The systematic SFA results were used to determine possible exposure pathways and trends in environmental exposure potential through different pathways. TCS discharged in wastewater mainly flowed into surface water sediment, ocean, and soil, where it accumulates in aquatic and agricultural products that may pose a higher risk to human health than brief exposure during consumption. Only 22% of TCS discharged was removed in the built environment with the remainder discharged into the natural environment, indicating that anthropogenic TCS metabolism in China is unsustainable. Per capita TCS consumption increased 209% from 2003 to 2012, resulting in increased discharge and accumulation in the environment. If current trends continue, it will increase to 713 mg capita^{-1} yr^{-1} in 2015 and 957 mg capita^{-1} yr^{-1} in 2020. Accordingly, annual environmental exposure potential will increase from 388 mg capita^{-1} in 2008 to 557 mg capita^{-1} in 2015 and 747 mg capita^{-1} in 2020, indicating an increasing trend of exposure to environmental TCS. Results of Pearson correlation analysis suggested that feasible countermeasures to reduce environmental exposure potential for triclosan would include encouraging the development of small cities, raising awareness of health risks, nurturing environmentally-friendly consumer values, and improving the environmental performance of TCS-containing products.

1. Introduction

Triclosan (TCS) has been marketed for over 40 years (Glaser, 2004) and is produced by manufacturers located in the U.S.A., Switzerland, the Netherlands, China, India, South Korea, and elsewhere (APUA, 2011). It has been suggested that TCS inhibits lipid biosynthesis by acting upon enzymes (Levy et al., 1999; McMurry et al., 1998); as a result, it has become a widely-used antimicrobial agent in pharmaceuticals and personal care products (PPCPs).

However, because TCS is an artificial chemical with no known natural sources (HCEC, 2012), the continuous consumption of TCS creates the risk of "environmental allelopathy", meaning that humans may not be acclimatized to the changing human–microorganisms relationship caused by the continuing environmental occurrence or accumulation of TCS (Gautam et al., 2014; Halden and Paull, 2005; Lindstrom et al., 2002; Miller et al., 2008). Environmental accumulation is a result of TCS migrating from consumer products to the natural environment

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Abbreviations: TCS, triclosan; SFA, substance flow analysis; PPCPs, pharmaceuticals and personal care products; WWTPs, wastewater treatment plants; MeTCS, methyl-triclosan; HCE-TCS, per capita annual urban household consumption expenditure on TCS-related products.

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(Davis et al., 2012; Hua et al., 2005; Ying and Kookana, 2007) where it potentially disrupts critical ecological processes performed by beneficial microorganisms (Dokianakis et al., 2004; Neumegen et al., 2005; Price et al., 2010). TCS is acutely toxic to some aquatic organisms (particularly certain algae species) at low μg L^{-1} levels, and has been shown to modulate thyroid functions in amphibians at concentrations as low as 0.15 μg L^{-1} (Chu and Metcalfe, 2007). Environmental accumulation could also result in the appearance of bacteria strains with TCS-insensitive enzymes, including so-called 'super-bugs' (bacteria that have developed resistance to the effects of antibiotic drugs) such as the new strains of tuberculosis (Phillips, 1998). Thus, a microbial environment which is continually changed by TCS may result in disease outbreaks among vulnerable people. TCS has been found to be omnipresent in human tissues (plasma, milk, liver, adipose and brain) around the world, including in Australia, the U.S.A., Germany, Sweden, Belgium, South Korea, and China (Allmyr et al., 2006; Dann and Hontela, 2011; Geens et al., 2012; Li et al., 2013; NICNAS, 2009; SCCP, 2009); for example, TCS was detected in 74.6% of the urine samples of the general Belgian population (Pirard et al., 2012). TCS has been classified as a potential carcinogen and toxicant (Fang et al., 2010) and can be metabolized to dichlorodibenzo-p-dioxin (Lores et al., 2005). Consumers who use TCS-containing products on a daily basis risk reduced levels of DNA methylation in human hepatocytes and down-regulated MBD2, MBD3, and MeCP2 gene expression (Ma et al., 2013).

TCS has been identified as a chemical which should be a monitoring and research priority due to human toxicity and evidence of adverse environmental effects and endocrine disruption (Clarke and Smith, 2011). However, studies usually focus on independent assessment of individual risk sources, mainly by microexamination of molecules or cells (Binelli et al., 2009; Canesi et al., 2007; Crofton et al., 2007; HCEC, 2012; James et al., 2010; Kawanai, 2011; Lin et al., 2010; Marlatt et al., 2013; Matozzo et al., 2012; Tamura et al., 2012). At present, existing knowledge of the potential environmental and human risks from TCS use is insufficient to carry out a comprehensive risk assessment due to a general lack of data related to TCS distribution in the environment and the amounts of TCS transmitted from the environment to humans, and the most important transmission pathways.

Substance flow analysis (SFA) is usually used to assess the sustainability of socioeconomic development and environmental change, particularly from the perspective of improving substance flow efficiency (Huang et al., 2012; Zhang et al., 2014), but more recently it has also been used to investigate hazardous material for environmental risk assessment and management (Asari et al., 2008; Chèvre et al., 2013; Eriksson et al., 2008; Earnshaw et al., 2013; Herva et al., 2012; Long et al., 2013; Oguchi et al., 2013; Ono, 2013). The integration of SFA and risk assessment can convey a more comprehensive picture than traditional risk assessment; this also facilitates examination of risk distribution in a systemic way, bringing out additional information that can be used to formulate efficient management policies (Ma et al., 2007). In this study, we used a mass balance approach to comprehensively quantify TCS flows and distribution in China to determine possible exposure pathways for TCS. Then, trends in TCS consumption and environmental exposure potential for TCS were analyzed in order to identify appropriate measures to reduce the risks resulting from TCS discharge.

2. Methodology

2.1. Analytical framework

SFA is an analytical method used to systematically assess the flow and stock of a substance through a given system defined in space and time (Brunner and Rechberger, 2004; Huang et al., 2014). It can be used to help construct a systematic database and determine critical links or pathways, and can also facilitate the quantifying of substance flows within the socio-economic and environmental systems (Brunner and Rechberger, 2004; European Communities, 2001; Huang et al., 2012, 2013; Sendra et al., 2007).

To understand TCS flow distribution in the anthroposphere in China, a coupled human and environmental system approach was adopted (Srinivasan et al., 2013; Stannard and Aspinall, 2011). The system boundary is the national border of mainland China, without Hong Kong, Macau and Taiwan (Fig. 1). The system was divided into three groups: 1) TCS flows in the built environment which include imports and exports, production, consumption, discharge and removal (including degradation, landfill, and incineration by built facilities); 2) TCS flows in environment media; and 3) human exposure.

2.2. Data collection

Because it is not known how much TCS is released from domestic or industrial sites (e.g. where TCS is incorporated into plastic and textile items) into the environment, TCS discharge mass balance was based on monitoring of TCS concentration in waterways and effluents. We found that the monitoring data for China in the published literature was more complete in 2008 than in other years, so 2008 was selected for this study. TCS and other related data in China for 2008 were derived from 1) peer-reviewed literature; 2) Chinese government statistical yearbooks or bulletins, which represent the best available data for quantification and forecast of anthropogenic TCS consumption and discharge flows in China (this includes but is not limited to the National Bureau of Statistics, Ministry of Housing and Urban–Rural Development, Ministry of Water Resources, Ministry of Environmental Protection, General Administration of Customs, Ministry of Commerce, National Development and Reform Commission, and Development Research Center of the State Council); 3) industrial reports from chemical associations and research units, especially 'Report of TCS technology and market in China' (S6CCN, 2013), which provides data on national production, consumption, import, and export of TCS; 4) personal communication with some companies (e.g., Weifang Aoyou Chemical Technology Co., Ltd); 5) survey of several major producers and users of TCS; 6) mass balance across the socio–economic–environmental system; 7) proxy data and known scientific relationships; and 8) estimation by industrial association studies.

2.3. Accounting approach

2.3.1. TCS flow distribution

We assumed that 1) the stock of TCS in production or consumption was the same in different years, so TCS stocks need not be considered in 2008, and 2) TCS spreads out in wastes or each environmental medium until an even concentration is achieved in a basin. This allowed us to develop a static physical flow model with detailed TCS balance calculations as indicated by Eqs. (1)–(14) in Table 1.

2.3.2. TCS consumption trends

In order to select a reference indicator for TCS consumption forecasting, Pearson correlation analysis was conducted between TCS consumption and its most likely factors based on the literature (Katz et al., 2013; NICNAS, 2009; Zhao et al., 2013). According to results of Pearson correlation analysis (Table S1, Supplementary data), the highest correlation coefficient was between population and apparent consumption of TCS, so annual increase in per capita TCS consumption (R_a) is taken as an indicator for forecasting; this was calculated by Eq. (15) and validated by Eq. (16).

$$R_a = \left((V_r/V_s)^{1/(y_r - y_s)} - 1 \right) \times 100\% \tag{15}$$

$$R_y = \left(\left(V_{yt} - V_{yt-1} \right) / V_{yt-1} \right) \times 100\% \tag{16}$$