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RESEARCH ARTICLE

Basin-scale emission and multimedia fate of triclosan in whole China

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Abstract This study aimed to investigate the emission and multimedia fate as well as potential risks of triclosan (TCS) in all of 58 basins in China. The results showed that the total usage of TCS in whole China was 100 t/year, and the discharge to the receiving environment was estimated to be 66.1 t/year. The predicted TCS concentrations by the level III fugacity model were within an order of magnitude of the reported measured concentrations. TCS (90.8 %) was discharged into the water compartment and 9.2 % to the soil compartment. The TCS concentration levels in east China were found generally higher than in west China. In addition, the input flux for TCS to seawater was largely attributed to the seasonal variations in advection flows. Preliminary risk assessment showed that medium to high ecological risks for TCS would be expected in the eastern part of China due to the high population density.

Keywords Triclosan · Emission · Multimedia fate modelling · River basin · China

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Introduction

Triclosan is a biocide widely used in various personal care products (PCPs) (SCCS 2010). After use, most of the PCPs containing TCS are washed down the drain and discharged into wastewater treatment plants (WWTPs), with its reported removal efficiencies of 57–99 % (Bester 2005; Halden 2014; Samaras et al. 2013; Singer et al. 2002; Thomas and Foster 2005; Wick et al. 2010; Ying and Kookana 2007), and eventually into the receiving aquatic environment (Halden 2014). The large consumption for TCS has promoted it to be ranked in the list of top contaminants of concern worldwide (von der Ohe et al. 2012). TCS has been detected in various environmental media such as influents, effluents and biosolids in WWTPs (Ying and Kookana 2007), rivers (Halden and Paull 2005; Lindstrom et al. 2002; Sabaliunas et al. 2003; Zhao et al. 2013), lakes (Singer et al. 2002), drinking water (Stackelberg et al. 2004) and ocean water (Xie et al. 2008). Contamination of aquatic environments with TCS may be harmful to aquatic organisms, such as algae (Yang et al. 2008), crustacean (Orvos et al. 2002) and fish (Heidler et al. 2006).

The monitoring work at the catchment scale has been met with limitation of time and cost. There are more than 1500 rivers whose basin area is over 1000 km^2 in China (Statistical Yearbook of China in 2011). Although there have been some field TCS monitoring campaigns in China, TCS concentrations in most rivers are not available so far. Moreover, season variation has been found for TCS concentrations in the Chinese basins (Chen et al. 2014; Zhao et al. 2013). The regional and seasonal variations will certainly affect our knowledge on the fate of TCS in the environment. Modelling approach has provided an alternate method to solve the problem. Combining the chemical usage data, population, removal rate in WWTPs and a parameterized multimedia model describing the fate of chemicals, the chemical

concentrations in the environment can be predicted, and it has been proved to be effective and accurate (Keller et al. 2007; Price et al. 2009, 2010a, b; Whelan et al. 2012; Zhang et al. 2013). Multimedia fugacity models are well established, well documented and widely used for predicting the environmental fate of different chemicals at various scales (single river scale, regional scale and global scale) (Cao et al. 2004; Mackay and Paterson 1991; Prevedouros et al. 2004; Tan et al. 2007; Tao et al. 2003; Wang et al. 2012). We have successfully modelled environmental fate for TCS in Dongjiang River basin (Zhang et al. 2013) and for seven steroids in whole China (Zhang et al. 2014). Based on this, it is thus possible to achieve better understanding of TCS contamination in whole China by using multimedia fugacity modelling approach.

The aim of this study was to evaluate the emission and multimedia fate of TCS at the river basin scale in whole China. The predicted chemical fate includes chemical source, process and output. The "source" (emission data) was derived from market research data. The levels III and IV multimedia fugacity model were applied to characterize the "process" and "output" for TCS in the environment. Based on the modelled exposure concentrations, potential risks posed by TCS and other related biocides were also mapped by using the risk quotient (RQ) approach.

Methodology

Study area

According to the Industry Standard of China (ISC 2000), whole China was divided into 58 basins. Each basin was allocated an ID to simplify the model description (Table 1). All the basins were subject to emission estimation and multimedia fate modelling for TCS. For evaluating the seasonal variations of TCS in China, three representative basins were selected: Liaohe River (basin ID: 7, located in Northern China), Yangtze River Downstream (basin ID: 26, located in Central

Table 1 IDs for all the basins and the first level basins

ID	Name	First level basin	ID	Name	First level basin
1	Heilongjiang	Heilongjiang River	30	Wujiang River	Yangtze River
2	Songhua River		31	Dongting Lake	
3	Wusuli River		32	Hanjiang River	
4	Suifen River		33	Poyang Lake	
5	Tumen River		34	Taihu Lake	
6	Ergun River		35	Qiantang River	Southeast China's coastal area
7	Liao River	Liaohe River	36	Oujiang River	
8	Daling River		37	Minjiang River	
9	Liaodongbandao		38	Mindong-Yuedong	
10	Yalu River		39	Hanjiang River	
11	Luan River	Haihe River	40	Xijiang River	Pearl River
12	Zhangweinan Canal		41	Beijiang River	
13	Yongding River		42	Dongjiang River	
14	Daqing River		43	Pearl River Delta	
15	Ziya River		45	Yueguiqiong	
16	Tuhai-Majia River		44	Hainan	
17	Chaobai-Beiyun-Jiyun River		46	Yuanjiang-Honghe	International rivers of Guangxi,
18	Yellow River	Yellow River	47	Lantsang-Mekong	Yunnan, Tibet, Xinjiang
19	Fen River		48	Nujiang-Irrawaddy	
20	Wei River		49	Brahmaputra	
21	Shandong Peninsula		50	Senggecangbu	
22	Huai River	Huaihe River	51	Ertix River	
23	Yishusi		52	Water system inner of Inner Mongolia	
24	Lixia River		53	Water system inner of Hexi corridor-Alxa	
25	Yangtze River Upstream	Yangtze River	54	Water system inner of Qaidam	
26	Yangtze River Downstream		55	Water system inner of Junggar	
27	Yalong River		56	Water system inner of Ili-Eminhe	
28	Minjiang River		57	Water system inner of Tarim	
29	Jialing River		58	Water system inner of Tibet	

China) and Pearl River Delta (basin ID: 43, located in Southern China). The geographic information layers of the basin system and administrative region of China are both available at National Geomatics Center of China (http://sms.webmap.cn/). And, the map of China basin was obtained using ArcGIS 9.3 software. Detail description of the administrative areas included in each basin can be referred to our previous study (Zhang et al. 2014) and also listed in the Supplementary Information (SI-A).

Emission estimation

Emission of TCS in whole China was derived from market research data. The Euromonitor (www.euromonitor.com) provided us with China market sales data for personal care products (PCPs) in 2011. And, the Mintel's Global New Products Database (GNPD) (www.gnpd.com) provided the ingredient lists associated with these products. As TCS is commonly used as bactericidal additive with a maximum concentration of 0.3 % in PCPs (EC 2009), we were able to estimate the total amount of TCS consumed in the whole China. According to some studies available in literature, the usage of chemical ingredients included in the personal care products in a region corresponded well with the *per capita* chemical consumption (PCC) estimate (mg/cap/year) inputs and populations (Holt et al. 1998; Price et al. 2009; Whelan et al. 2012). Based on the assumption, we distributed the TCS uniformly for the population in China. The emission rate for each of the 58 basins was calculated according to the average consumption data *per capita* and the populations in the region.

For the emission of TCS to the receiving environment, the pathways into the water and soil compartments were considered. Since sewage is the main carrier for TCS, the TCS incorporated in the sewage from urban population can be mostly removed in WWTPs before it enters into surface water (Halden 2014; Halden and Paull 2005; Lozano et al. 2013; Waltman et al. 2006). But, for the TCS in sewage from rural population, it is assumed that the sewage is directly discharged into the receiving environment without treatment based on the report by Ministry of Environmental Protection of China (GOSC 2012). For soil compartment, sewage irrigation and sludge application contribute to the TCS contamination. But, in China, disposal of sewage sludge on agricultural land is still prohibited (Chen et al. 2013). The ratio of sewage irrigation area to the whole agricultural land was 6.65 % according to the second national survey of sewage irrigation (Fang 2011). Assuming that the irrigation water amount per unit area of the agricultural land was the same, the chemical emission into the soil compartment via sewage irrigation can be estimated by the ratio of the sewage irrigation volume to the sewage wastewater discharge volume multiplied by the chemical emission from the sewage wastewater. These calculated emission values for the water and soil compartments of each basin were then used as the model input parameters in multimedia fugacity modelling. Detailed description about the emission estimation methods can also be found in Supplementary Information (SI-B).

Multimedia model description

Based on the approach of Mackay and Paterson (1991), a level III fugacity model was applied to predict the basin scale multimedia fate of TCS. The model applied in this study was based on our previous study (Zhang et al. 2014). In brief, the model includes four bulk compartments and 10 sub-compartments: air (air, particulates), water (water, suspended solids and fish), soil (air, water and solids) and sediment (water and solids). The environmental processes expressed by transfer fluxes in the model include the following: emissions, advective flows, degrading reactions and interphase transport by diffusive and nondiffusive processes. In a level III fugacity model, the mass balance equations were established in terms of transfer fluxes, which express that the total input fluxes from the individual compartments equal to the output fluxes. And, the numerical model simulation was performed using Matlab R.2010a. All parameters used to solve the model equations were from the published literature or reports, which include 14 parameters describing the physicochemical properties of TCS (Table 2) and 42 parameters describing the environment (58 river basins in China) (Supplementary Information SI-B, Table S1–S2). In addition to the sensitivity and uncertainty analysis, the model was validated by comparing the predictions with the observed values collected from published reports. For the Yangtze River, the measured concentrations were determined in our laboratory by using our published method (Zhao et al. 2009). The mass inventories for TCS in different environmental compartments of each basin were calculated by using the product of predicted concentrations and volume of the basin. A detailed description on the modelling parameters and equations can be found in Supplementary Information (SI-B).

A level IV fugacity model was also developed to study the seasonal variation of TCS in the three representative basins (Liaohe River, ID 7; Yangtze River Downstream, ID 26; and Pearl River Delta, ID 43) in Northern, Central and Southern China. In the level IV fugacity model, the fugacity of each compartment changes with time, and the change rate equals to the difference between the input and output rates divided by a product of compartment volume and fugacity capacity. This process is expressed by a differential mass balance equation, which is described in Supplementary Information. A preliminary test showed that the result of level IV fugacity model was almost the same as the result of steady-state fugacity model in summer, and the difference was far less than 1‰. Thus, the initial values of the fugacity were set at steady-state fugacity

Table 2 Physiochemical properties of triclosan

Symbol	Unit	Definition	Mean (range)	References
M	g/mol	Molar mass of the chemical	293	EPI*
ρ	g/ml	Bulk density of the chemical	1.55	EC 2009
MP	K	Melting point of the chemical	330.5	EC 2009
H	Pa m^3/mol	Henry's constant	2.77×10^{-2} (1.52×10^{-2}–5.07×10^{-2})	Lindstrom et al. 2002; USEPA 2008
*P*s	Pa	Vapour pressure	4.87×10^{-4} (5.33×10^{-4}–7.00×10^{-4})	EC 2009; Reiss et al. 2002; Ying et al. 2007; Zhao et al. 2013
K_{OC}	L/kg	Organic carbon normalized partition coefficient	21,982 (11,397–63,096)	Agyin-Birikorang et al. 2010; Wu et al. 2009 Miller et al. 2008; Orvos et al. 2002 Reiss et al. 2002; Zhao et al. 2013
t_1	h	Half-life of the chemical in air	16	EPI
t_2	h	Half-life of the chemical in water	554.5	Singer et al. 2002
t_3	h	Half-life of the chemical in soil	674 (304–1872)	Kwon et al. 2010; Waria et al. 2011; Wu et al. 2009; Xu et al. 2009 Ying et al. 2007
t_4	h	Half-life of the chemical in sediment	13,000	EPI
BCF_f		Bioconcentration factor for fish in water	2500	Orvos et al. 2002; USEPA 2008
B_2	m^2/h	Molecular diffusivity in water	5.09×10^{-7}	Wu and Gschwend 1988
B_4	m^2/h	Molecular diffusivity in sediment	1.03×10^{-7}	Wu and Gschwend 1988
f'	%	Removal efficiency of the chemical in sewage treatment plants	91.9 (56.5–99.9)	Bester 2005; Halden and Paull 2005 Kanda et al. 2003; Lozano et al. 2013; Mcavoy et al. 2002; Nakada et al. 2006, 2007; Reiss et al. 2002; Sabaliunas et al. 2003; Samaras et al. 2013; Singer et al. 2002; Thomas and Foster 2005; Thompson et al. 2005; Waltman et al. 2006; Wick et al. 2010; Ying and Kookana 2007

*No published data available; the value of these parameters were calculated by the software EPI 4.1 from the United States Environmental Protection Agency

values in summer, and the autumn was firstly simulated by using level IV fugacity model.

The parameters used to solve the differential equation were all the same as those for the steady-state modelling (Zhang et al. 2014), except some parameters related to the temperature. Season alternation brings dramatic change in temperature, as well as the temperature-dependent parameters. In our study, temperature and precipitation were monthly values collected from local Statistical Yearbook in 2011. It is noted that the year 2011 is quite typical. Advection water flows and contents of solids in water of each of the three basins were seasonal values based on the reports of Water Resources and Sediment Bulletin of each basin in China. As the published articles can only provide one value for the parameters in a given temperature, the quantitative relationship between the parameter and temperature is more effective for us. And, these derived parameters included degradation rate, Henry's law constant and vapour pressure. Other parameters, which might not change over time or would not have noticeable effects on the modelled results, or with difficulty to get the seasonal values from published reports or quantitative relationships, were kept the values as the inputs of the level III fugacity model. Detailed information on the parameters together with their sources was available in Table 2 and Supplementary Information (SI-B, Table S1–S4).

Results and discussion

Emissions

Based on the market research data, the total sales of PCPs containing TCS in China were summed to 1239 kt in 2011. Thus, the usage of TCS included in these PCPs was estimated

Table 3 Total usage and environmental emission of triclosan in China for the year 2011

Categories	Consumed amount (t/year)[a]	TCS					
		Content level (%)[b]	Inclusion level (%)[c]	Total usage in the PCPs (t/year)	Per person consumption (mg/person/day)[d]	Estimated emission to the receiving environment (t/year)	
Men's skin care Body care	11,743	0.4	0.3	100	0.21	66.1	Water: 60.0
Acne treatments Face masks Facial moisturizers Nourishers/anti-agers Toners	41,440	0.3	0.3				
Liquid soap	35,418	6.3	0.3				
Bar soap	332,159	13.0	0.3				
Body wash/shower gel Men's bath and shower	187,969	4.3	0.3				
Shampoos Men's hair care	396,255	0.8	0.3				Soil: 6.06
Powder detergents Liquid detergents	431	1.4	0.3				
Mouthwashes/dental rinses	4,248	1.3	0.3				
Toothpaste	229,274	16.7	0.3				

[a] Consumed amount, the market research sales data, which was obtained from Euromonitor

[b] Content level, the percentage of consumed products that contained a target chemical, which was obtained from Mintel's New Global New Products Database (GNPD)

[c] Inclusion level, the percentage of the active ingredient in products according to http://ec.europa.eu/health/ph_risk/committees/04_sccp/docs/sccp_o_166.pdf

[d] Per person consumption=Σ(consumed amount×content level×inclusion level×10^9)/population. The total population (1,339,724,852 people) was obtained from the report of the Sixth National Census in China

to be 100 t (Table 3). This is smaller when compared to the reported usage data from USA (300 t; Halden and Paull 2005) and Europe (350 t; Singer et al. 2002). If the total usage of TCS were uniformly distributed to the whole population (1.3 billion) in China, the *per capita* consumption (PCC) of TCS would be 0.21 mg/day. This is far less than the average values of USA and Europe as both have less population than China. For example, Lindstrom et al. (2002) have reported that the PCC for TCS was 0.31–0.76 mg/day in Switzerland. As the sales of PCPs are related to the purchasing power of the population (Hodges et al. 2012) and China's per capita income is only about 9 % of the USA in 2010 (World Bank 2014), it is reasonable that PCC of TCS in China is lower than in those developed countries. As a developing country, China also has an income gap between rural and urban population. Unfortunately, the market research data could not distinguish this, and China has not established a national chemical registration system at the county level. It might create some uncertainty in emission estimation for each basin. However, for simplicity, the nationwide PCC was still applied in the emission estimation and multimedia modelling since each basin is a large region including rural and urban areas.

In China, the emission of TCS to the receiving environment was estimated to be 66.1 t in the year of 2011. The highest emission for TCS (5.05 t) was found in Huai River basin (ID: 22) and the lowest emission (0.001 t) in Senggecangbu basin (ID: 50). From the total TCS usage, only 34 % reduction was observed, mainly due to the fact that more than 50 % of the China's total population are living in the rural area with limited sewage treatment (Zhang et al. 2014). Of the total emission (66.1 t), 60 t of TCS was discharged into the water compartment and the rest was to the soil compartment. The emission values for TCS for each of the 58 basins in China are shown in Table S5 (SI-B). These calculated emission values for the water and soil compartments of each basin were then used as the model input parameters for predicting the multimedia fate of TCS.

Prediction of TCS concentrations in each basin

The concentrations for TCS in all of the 58 basins were modelled for air, water, soil, sediment, suspended particulate and fish. The concentration results are presented in Table S6 (SI-B). The TCS concentrations in the air compartment were all at the low levels, ranging from 8.40×10^{-10} to 7.24×10^{-7} ng/L. This is attributed to the low Henry's constant of TCS (2.77×10^{-2} Pa m^3/mol). The predicted lowest and highest concentrations were found at Senggecangbu basin (ID: 50) and Yishusi river basin (ID: 23). The former basin is located in sparsely populated Tibet while the latter is in densely populated Huaihe Basin. These factors also affect the soil compartment without wastewater irrigation. For soils without wastewater irrigation, the predicted TCS concentrations in soil ranged from 2.79×10^{-9} (ID: 50) to 1.89×10^{-6}

(ID: 31) ng/g. For wastewater irrigated soils, the TCS concentrations were found much higher, ranging between 2.11×10^{-2} (ID: 12) and 9.56×10^{-5} (ID: 19) ng/g. As the subcompartments of water, both suspended particulates and fish showed relatively higher concentrations for TCS when compared with the soil compartments. The highest and the lowest concentrations in suspended particulates and fish were 1.25×10^{-3} (ID: 50) and 8.74 (ID: 42), and 2.96×10^{-2} (ID: 50) and 42.9 (ID: 14) ng/g, respectively. The predicted environmental concentrations (PECs) of TCS in the water and sediment compartments in China are shown in Fig. 1. The TCS concentrations in water ranged from 0.156 to 39.1 ng/L. The most polluted basins were mainly located in North China, including Haihe River and Huaihe River. Dongjiang River (ID: 42) located in Southern China had TCS concentrations of 11.7 ng/L. Figure 1a showed that almost all of the TCS concentrations in the basins of West China were less than 1 ng/L. Compared with other countries, the levels for most of the rivers in China

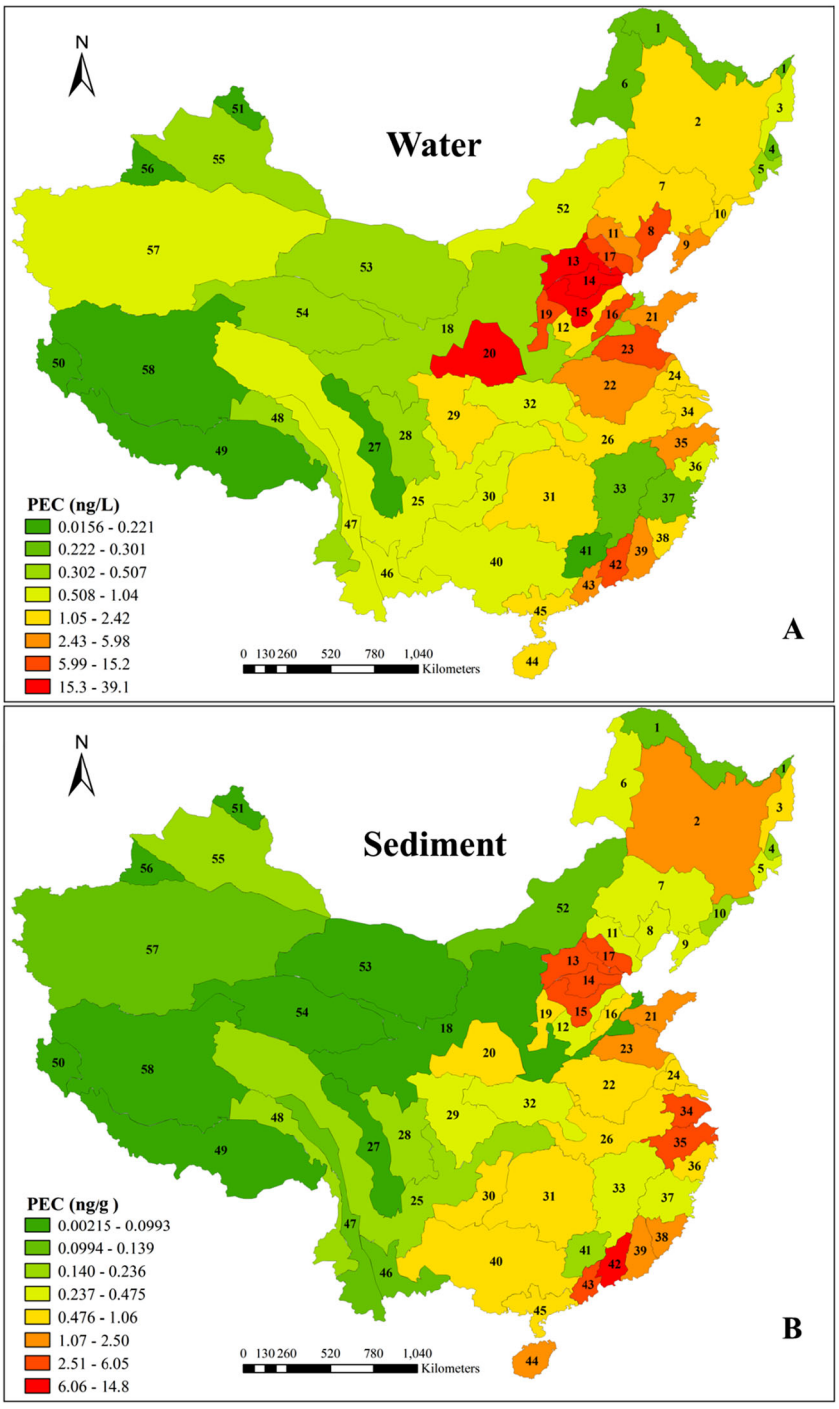

Fig. 1 Exposure mapping for TCS in water (**a**) and sediment (**b**) in China. The predicted environmental concentrations (PECs) for TCS are given in water with the unit of ng/L and in sediment with the unit of ng/g

were slightly lower than some developed countries such as USA (ND-2300 ng/L; Alvarez et al. 2005; Kolpin et al. 2002, 2004; Loraine and Pettigrove 2006; Vanderford et al. 2003), UK (10–15 ng/L, Kasprzyk-Hordern et al. 2008), Korea (1–82 ng/L; Yoon et al. 2010) and Spain (58–138 ng/L; Villaverde-de-Saa et al. 2010) but comparable with Japan (<0.6–59.1 ng/L; Nakada et al. 2008) and Switzerland (<0.4–74 ng/L; Lindstrom et al. 2002). As displayed in Fig. 1b, the PECs of TCS in sediment ranged from 0.002 to 14.8 ng/g. The highest concentration was located in Dongjiang River basin (ID: 42), while the lowest was in Senggecangbu basin (ID: 50). In addition, Haihe River basin (ID: 13–15), Taihu basin (ID: 34) and Qiantangjiang River basin (ID: 35) were also found to have TCS concentrations in sediment more than 3 ng/g.

When compared to the available reported concentrations in Chinese rivers, the differences between the PECs and MECs mostly fell within an acceptable range (less than a log-unit range) (Cao et al. 2004; Liu et al. 1999) (Table 4). The more measured concentrations from a river, the more representative the average value, especially for the large rivers. Yellow River (ID: 18) and Yangtze River (ID: 26) are the two biggest rivers in China, and both of which flow through more than six provinces in China. For both rivers, more than 60 measurement values for water compartment had been used for the calculation of the average concentration. But, only 10 measurement values were available for the sediment compartment of the Yellow River; thus, we had the largest log-diff (1.61) for the sediment in this river. The predicted TCS concentrations in sediment of some other basins like Daqing River (ID: 14), Dongjiang River (ID: 42) and Pearl River Delta (ID: 43) were well verified (Table 4, Table S2 (SI-B)). The predicted TCS concentrations in fish for those basins with big cities such as Beijing (IDs: 13–15, 31.4–42.9 ng/g), Guangzhou and Shenzhen (IDs: 42–43, 10.7–28.6 ng/g) were quite high. Unfortunately, there is no measured concentrations for fish from rivers in China, while in the USA, the measured concentrations in fish from rivers reported by Mottaleb et al. (2009) ranged from 17 to 31 ng/g. Sensitivity analysis showed that TCS concentration in water compartment was more influenced by chemical emission and the area of the water phase. And, most of the basins were within one order magnitude uncertainty by using the SQL measured (the difference between the third and the first quartiles) (SI-B, Fig. S1). Combining the validated results between the MECs and PECs and the uncertainty analysis result, the model presented here was considered to provide a good simulation for the TCS concentrations in the environmental media.

The present study showed regional distribution characters of TCS, with higher concentrations being observed in East China than in West China (Fig. 1). The high modelled TCS concentration levels were distributed in some basins like Haihe River basin in North China and Pearl River basin in South China. This is linked to the high population in the two regions. It should be noted that high chemical concentrations in real aquatic environments are often found in tributaries rather than in the main streams (Wang et al. 2010; Zhao et al. 2010). With rapid economic development and increasing purchasing power in China, it should be expected to see increasing TCS contamination levels in the Chinese riverine environments if there is no improvement in wastewater treatment rate.

Seasonal variation of TCS contamination

Three representative basins were selected to illustrate the seasonal variation for TCS in China, from spring to winter, as showed in Fig. 2. Air and soil compartments were not included in Fig. 2 but separately displayed in Fig. S2 of Supplementary Information (SI-B). The different seasonal variations in TCS concentrations were observed among the three basins. In Liaohe River basin (ID: 7), the TCS concentrations in water varied from 1.39 ng/L in autumn to 2.60 ng/L in spring and in sediment from 0.41 ng/g in summer to 0.60 ng/g in winter. While in Yangtze River Downstream (ID: 26) and Pearl River Delta (ID: 43), the highest TCS concentrations were all in winter: 1.89 and 5.20 ng/L for water, 1.20 and 5.78 ng/g for sediment, respectively. Liaohe River basin is in the North China with a latitude of 40~45° N. The average temperature in winter was lower to −9.2 °C, and the river flow was fairly low in Liaohe basin. As both temperature-dependent half-life and water flow can influence the concentration levels for TCS, although slow degradation rate was available in winter, the combined influence contributed to the highest concentrations in spring (with the average temperature of 7.4 °C) in Liaohe River basin. Compared to the other basins, the suspended particle amount in water compartment in Liaohe River is concentrated in summer (SI-B, Table S4), so more TCS should adsorbed onto the suspend particles (Zhao et al. 2013). And, this contributed to the not-smooth seasonal concentration curve for the basin of Liaohe River. Emission was also an important factor for the TCS seasonal variation; however, temporal-resolved data for TCS emission was unavailable in our modelling. Considering that PCPs are consumed more in summer because of the hot weather, TCS aqueous concentrations in summer would be underestimated. However, in total, hot summer makes TCS of the three basins stay at the lower levels in water and sediment compartments throughout the year.

There was a marked difference in the extent of the TCS concentration variations among the three basins. Since the three basins are located in different regions of China (North China, Central China and South China), the chemical experienced very different environmental changes over the year. Thus, the TCS concentrations in water from the lowest to the highest levels varied by 87.1 % in Liaohe River, 71.8 % in Yangtze River Downstream and 29.0 % in Pearl River Delta.

Table 4 Comparison of the modelled and measured concentrations for TCS in water and sediment

Basin ID	Water					Sediment				
	Modelled concentration (ng/L)	Measured concentration (ng/L)	Log-Diff[a]	N[b]	Ref.	Modelled concentration (ng/g)	Measured concentration (ng/g)	Log-Diff	N	Ref.
7	1.67	23.3	1.14	84	Wang et al. 2011	0.40	5.80	1.16	48	Wang et al. 2011
13	25.7	3.14	−0.91	12	Wang et al. 2010	5.13	46.1	0.95	12	Wang et al. 2010
14	39.1	8.11	−0.68	22	Wang et al. 2010	4.99	17.4	0.54	24	Wang et al. 2010
15	31.0	11.9	−0.41	2	Wang et al. 2010	3.97	23.6	0.77	2	Wang et al. 2010
17	6.92	6.68	−0.02	2	Wang et al. 2010	2.53	–	–	–	–
18	0.43	3.72	0.94	64	Wang et al. 2010	0.05	2.05	1.61	10	Wang et al. 2010
26	1.67	13.7	0.91	142	This study[c]	0.93	1.46	0.20	95	This study[c]
34	2.30	6.43	0.45	5	Zhu et al. 2013	3.03	6.32	0.32	2	Zhu et al. 2013
42	11.7	10.4	−0.05	88	Chen et al. 2014; Zhang et al. 2013	14.8	4.22	−0.54	66	Chen et al. 2014; Zhang et al. 2013
43	5.08	28.1	0.74	36	Zhao et al. 2010	4.87	37.0	0.88	32	Zhao et al. 2010

[a] Log-diff means the log-unit difference between the modelled and measured geometric means; the negative values mean that the modelled results were overestimated

[b] The total number of environmental samples determined

[c] The measured concentration data from this study. A detailed description of the determination method is available in the Supporting Information (Table S7)

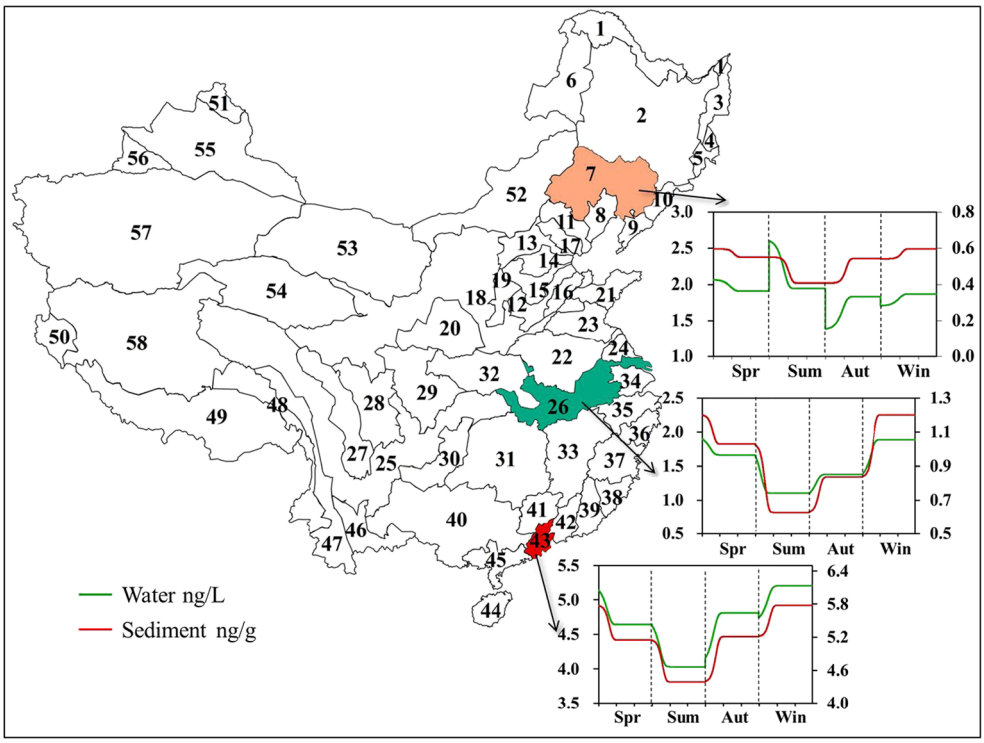

Fig. 2 Seasonal variations for TCS concentrations in water and sediment compartments in basins of Liaohe River (ID: 7), Yangtze River Downstream (ID: 26), and Pearl River Delta (ID: 43). The *left vertical axis* is for TCS concentrations in water, and the *right vertical axis* is for concentrations in sediment

The different variations are mainly attributed to the direct photolysis half-lives and the water flow in different latitudes (Tixier et al. 2002). Since monitoring campaigns are often carried out only in summer and/or autumn (Wang et al. 2011; Zhao et al. 2010), it may create biased monitoring dataset.

Multimedia fate of triclosan in China

The transfer fluxes for all the 58 basins were predicted based on the level III fugacity model, and a summary of the transfer fluxes in air, water, soil and sediment compartments in China is given in Table 5. The most important fluxes for TCS were source emission and degradation in various compartments. Diffusive fluxes between air and water and soil (T_{21d}, T_{31d}) were relatively low because of the low Henry's constant of TCS. In water compartment, sedimentation (T_{24s}) contributed most for the TCS out of the media. This is attributed to the modest hydrophobic nature of TCS (log K_{OW} 4.2–5.4) (Halden and Paull 2005; Lindstrom et al. 2002). Advective flow (T_{20t}) was also the important transfer processes for TCS out of the Chinese rivers. This means that sea water which receives the rivers of China would get the loading of TCS with 3.04 t/year (Table 5). Slow dissipation for TCS has been reported in soils, with its half-lives more than 70 days (Ying et al. 2007). Therefore, TCS would persist in the soil compartments, and the main output flux for soil was erosion from soil to water (T_{32e}). The sedimentation was the main source of TCS in sediment compartment. As the flux of sedimentation accounted for more than 50 % of the total input fluxes for the environment ($T_{02h}+T_{23h}$), it clearly illustrates the distribution characters of TCS in the aquatic environment. Singer et al. (2002) reported the detection of quite high amount of triclosan in the 30-year-old sediment layer. This indicated that sediment would receive more TCS in the future due to its modest hydrophobic nature and slow degradation. When a steady state was achieved in basins of China, the total mass inventory was estimated to be 75.3 t. Of the total mass, 96.7 % was in sediment, 2.4 % in water, 0.9 % in soil and a very small proportion in air.

Three most important fluxes, sedimentation (T_{24s}), degradation in water (T_{20m}) and advective output flow (T_{20t}), were selected to illustrate the seasonal variation of transfer fluxes for TCS in the three representative basins (Liaohe River, Yangtze River Downstream and Pearl River Delta) located in different parts of China (Fig. S3). From summer to winter, T_{24s} displayed a gradual change trend, with its values varying from 0.314 to 0.459 mol/h in Liaohe River basin (ID: 7), 0.634 to 1.21 mol/h in Yangtze River Downstream basin (ID: 26) and 0.386 to 0.508 mol/h in Pearl River Delta basin (ID: 43). In contrast, T_{20m} and T_{20t} remained at highest levels in summer and the lowest levels in winter. The flux of degradation was more influenced by half-life of the chemical, which is a temperature-dependent parameter. For the flux of advective output flow, rainy summer created a higher water flow, resulting in transporting more TCS out of the river system. The difference of seasonal variation for different basins was mainly manifested in the change extent for the fluxes. From the lowest to the highest levels, the T_{24s}, T_{20m} and T_{20t} values in Liaohe River increased by 46.2, 1247, 2482 %,

Table 5 Summary of the transfer fluxes for TCS in and out of the four compartments in China

Environmental compartments	Transfer fluxes (t/year)					
	In			Out		
	Symbol	Definition	Value	Symbol	Definition	Value
Air	T_{21d}	Diffusion from water to air	0.44	T_{12d}	Diffusion from air to water	0.01
	T_{31d}	Diffusion from soil to air	0	T_{12p}	Dry precipitations from air to water	0
				T_{12w}	Wet precipitations from air to water	0
				T_{13d}	Diffusion from air to soil	0
				T_{13p}	Dry precipitations from air to soil	0
				T_{13w}	Wet precipitations from air to soil	0
				T_{10m}	Degradation in air	0.22
				T_{10t}	Advective flows out of the area through air	0.21
Water	T_{12d}	Diffusion from air to water	0.01	T_{21d}	Diffusion from water to air	0.44
	T_{12p}	Dry precipitations from air to water	0	T_{24d}	Diffusion from water to sediment	0.05
	T_{12w}	Wet precipitations from air to water	0	T_{24s}	Sedimentation	34.9
	T_{42d}	Diffusion from sediment to water	0	T_{20m}	Degradation in water	19.4
	T_{42r}	Resuspension	0.01	T_{2f}	Bioaccumulation in fish	3.09
	T_{32e}	Erosion from soil to water in suspended solids	0.44	T_{20t}	Advective flows out of the area through water	3.04
	T_{32l}	Erosion from soil to water in liquid phase	0.48			
	T_{02h}	Chemical entering water from human	60			
	T_{02t}	Advective flows in the area through water	0			
Soil	T_{13d}	Diffusion from air to soil	0	T_{31d}	Diffusion from soil to air	0
	T_{13p}	Dry precipitations from air to soil	0	T_{32e}	Erosion from soil to water in suspended solids	6.05
	T_{13w}	Wet precipitations from air to soil	0	T_{32l}	Erosion from soil to water in liquid phase	0
	T_{23h}	Chemical entering soil during wastewater irrigation	6.06	T_{30m}	Degradation in soil	0.01
Sediment	T_{24d}	Diffusion from water to sediment	0.05	T_{42d}	Diffusion from sediment to water	0.44
	T_{24s}	Sedimentation	34.9	T_{42r}	Resuspension	0.48
				T_{40m}	Degradation in sediment	34

respectively; while in the Pearl River Delta of South China, the T_{24s}, T_{20m} and T_{20t} values changed by 31.6, 145, and 513 %, respectively. In the Yangtze River of Central China, the biggest change among the three basins was 91.1 % observed for sedimentation flux (T_{24s}). Compared to the Pearl River flowing to the South China Sea, the Yangtze River flowing to the East China Sea displayed a smaller seasonal variation (T_{20t}, 392 % (Yangtze River) vs 513 % (Pearl River)) but a larger variation of degradation flux (T_{20m}, 444 % (Yangtze River) vs 145 % (Pearl River)).

The multimedia fate for TCS in China was determined based on the fugacity model. Among the PCPs of 1239 kt consumed in China, 100 t of TCS was used. But, only approximately 34 % of the total TCS usage was removed by WWTPs, and the remaining proportion was discharged into the water compartment (60 t/year) and soil compartment (6.06 t/year). These masses formed the source of TCS in the receiving environment. As a relatively hydrophobic chemical, TCS had a tendency to partition into the sediment compartment. This was demonstrated by its distribution in the environment (mass inventory: 96.7 % in sediment, 2.4 % in water and 0.9 % in soil when a steady state was achieved). Environmental processes such as degradation and advection flows led to the disappearance of TCS from the basins by 53.6 and 3.04 t/year, respectively. As demonstrated by the three representative basins, these two processes are strongly influenced by basin location and season.

Aquatic risk assessment

Based on the predicted aqueous exposure concentrations and literature ecotoxicity data, a preliminary ecological risk assessment was performed for TCS in the Chinese basins by using risk quotient (RQ) approach. RQs are calculated by

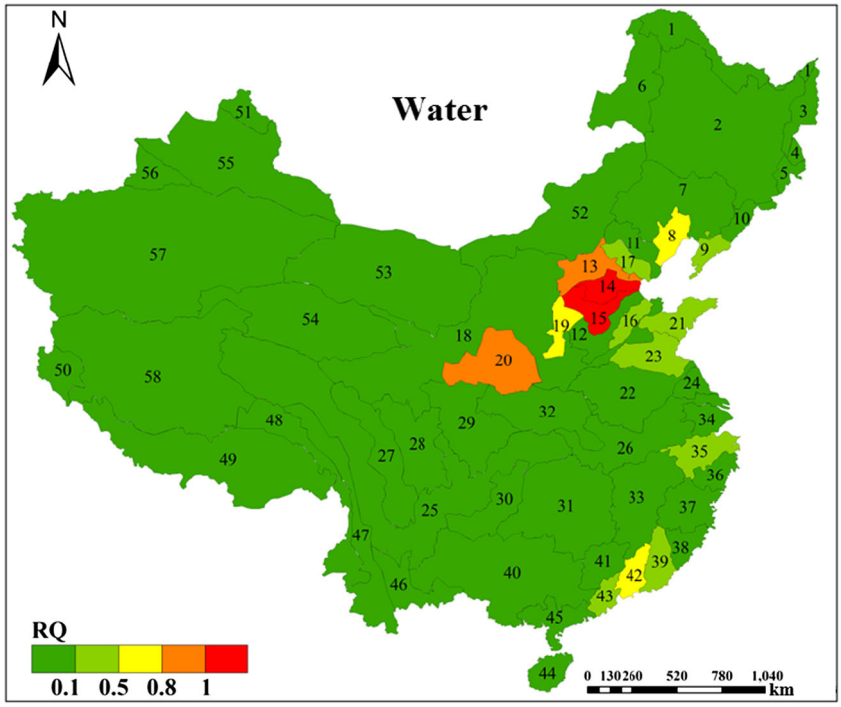

Fig. 3 Risk mapping for TCS in water. Aquatic risks are expressed by risk quotients (RQs)

the PECs and the predicted no-effect concentration (PNEC) according to the European Commission Technical Guidance Document (EC 2003). The PNEC for TCS was 26.2 ng/L, which was derived by using the species sensitivity distribution (SSD) approach (Chen et al. 2014). The criteria for interpreting the RQs are given as follows: RQ<0.1, "low risk"; RQ from 0.1–1, "medium risk"; and RQ>1, "high risk" (Hernando et al. 2006). As shown in Fig. 3, high risks for TCS would be expected in 2 out of 58 basins: Daqing River (ID: 14) and Ziya River (ID: 15). Both basins belong to the densely populated Beijing-Tianjin region. In addition, 13 out of 58 basins were expected to have medium risks, and they are mostly located in the Bohai Bay Rim and Pearl River delta region as well as Taihu Lake with large populations. Low risks would be expected from the TCS exposure for majority of the basins in China. For comparison with reported TCS concentrations in other countries, we expected high risks for some rivers in the USA (Kolpin et al. 2002), Japan (Nakada et al. 2008), Indian (Ramaswamy et al. 2011), Spain (Villaverde-de-Saa et al. 2010) and UK (Sabaliunas et al. 2003).

Conclusions

Environment emissions and multimedia fate of TCS in river basins of whole China were evaluated for the first time. The total usage of TCS in all household and personal care products was estimated to be 100 t/year, while the emission to the receiving environment was 66.1 t/year TCS following wastewater treatment. Among this, majority of the TCS (90.77 %) was discharged into the water compartment and a small proportion (9.23 %) into the soil compartment. A level III fugacity model successfully predicted the TCS concentrations in 58 basins of China, which were validated by some measured concentrations. Seasonal variations of TCS concentrations were also assessed in the three representative basins of North, Central and South China, and it was found that the input flux of TCS to sea water was attributed to seasonal advection flows. Multimedia modelling showed higher TCS concentrations in East China than in West China due to relatively higher population density in East China, which also resulted in medium to high ecological risks being found in East China too. Proper measures should be taken in those regions with high risks in order to reduce potential adverse effects on the ecosystem.

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Conflict of interest The authors declare that they have no conflict of interest.

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