Supporting information

A multimedia fate model to support chemical management in China: a case study for selected trace organics

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31 pages; 15 figures; 8 tables; 9 equations

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concentrations are values of only grid cells which cover the sampling sites of the measured data; the horizontal solid line in the box is the median; the top and bottom of the box are respectively the 75th and 25th percentile; the top and bottom of the whisker are respectively the highest and lowest case within 1.5 times the interquartile range. Extreme circles are max/min values.

Figure S9 Comparison of predicted and measured concentrations in certain river/lake catchment; circles are extreme values for measured and predicted data; there is only one grid cell which covers the limited sampling sites for OC and OMC in water, so the extreme values are not added;

Figure S10 Lognormal distribution and interquartile range (IR, 25th and 75th percentiles) of predicted chemical concentration in freshwater and sediment by Monte Carlo simulation

Figure S11 Predicted concentration for six chemicals in freshwater

Figure S12 Predicted concentration for six chemicals in freshwater sediment

Figure S13 nationally averaged chemical distribution in different media at steady state in the scenario that chemicals are only released to freshwater; the percentage was calculated by the amount of chemical in the bulk of environmental media at steady state

Figure S14 regional TCS distribution in different media around Yangtze River Delta at steady state in the scenario that chemicals are only released to freshwater; the percentage of TCS in pie charts was calculated by the amount of chemical in the bulk of environmental media

Figure S15 Net flux of total molecules of six chemicals (A) and ionic molecules of ionisable chemicals (B) between freshwater and sediment in the scenario that the chemical is released to water.

Model feature, environmental and chemical parameters

Model feature SESAMe v3.3 presented in this study is a further developed version based on a previous version SESAMe v3.0 published by Zhu et al. (2015).[1] The model includes air, freshwater and sediment, sea water and sediment, natural soil, agricultural soil, urban soil, natural vegetation and agricultural vegetation compartments. The model structure and input environmental database are almost the same for the two versions, except for the update with spatially varied freshwater pH and sediment pH across China in SESAMe v3.3. It is a regional multimedia chemical fate model with environmental database for 5468 adjacent but independent grid cells (50×50 km^2, regional scale) covering the mainland China (exclusive of Hainan and Taiwan). So the model does not predict the directionally advective transport between regional grid cells. To take into account the input and effect from surrounding regions, a larger scale, continental scale (150×150 km^2), has been developed, which is composed of 8 adjacent 50×50 km^2 grid cells The continental scale is surrounding and interlinked to regional scale grid cell by advective flow in air and water. Such structure is shown in Figure S1 in a previous paper by Zhu et al.[1]

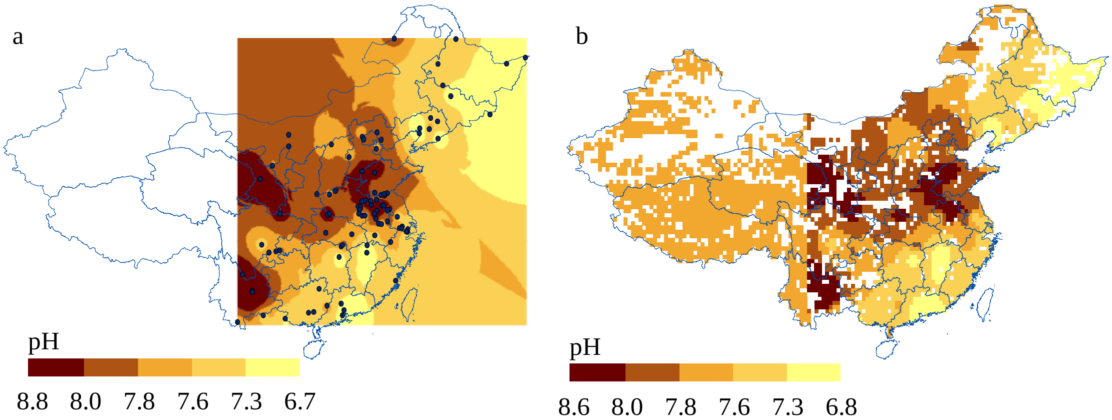

Figure S1 spatial distribution of freshwater pH in China

The formulas for modelling the concentration and fate of ionisable chemicals are the same as the first version of SESAMe model published by Zhu et al. (2014),[2] which have essentially referred to the equations in MAMI III model.[3, 4] For vegetation compartment, which is not considered in MAMI III model, the calculation has been shown in the Supporting Information (SI) of the paper on the first version of the model.[2] It has referred to the relevant calculations for vegetation compartment in Simplebox 3.24a.[5] The vegetation compartment has been considered as a homogenous compartment without separating the different parts of vegetation, e.g. the root and leaf etc. This allows a rough estimation of spatial distribution of chemical levels in vegetation across China and the main source of chemical to vegetation, e.g. from air or soil, which however are not the main topic of this study but worth mentioning.

logD is the log-transformed octanol-water partition coefficient for ionisable chemicals. D has been calculated in the model as below:[3]

$$D = f_{n,pH7} \times K_{ow,n} + (1 - f_{n,pH7}) \times K_{ow,ion} \quad (1)$$

$$K_{ow,ion} = 10^{logK_{ow,n} - 3.5} \quad (2)$$

Notes:

$f_{n,pH7}$: fraction of neutral form of chemicals when solution pH=7 (-);

$K_{ow,n}$: i.e.K_{ow}, the octanol-water partition coefficient of a chemical for the neutral form (-);

$K_{ow,ion}$: the K_{ow} for the ionic form of chemicals (-).

This also has been used in logD calculation in Figure 1.

Kaw (Henry's law constant) can be also pH dependent for ionisable chemicals. However, as (1) the pH dependent Kaw is not calculated and used in SESAMe v3.3 model and (2) it is not significant for target chemicals in this study which have very low concentrations in air either, it is not the point being discussed in this study and, therefore, not shown in Figure 1 with y-direction error bars. In SESAMe v3.3, Kaw (temperature corrected) for neutral molecules has been used for calculating the partitioning of neutral species between air and water with fraction of neutral molecules in water, as only neutral molecules in water can volatile to air. The formulas relevant to diffusive transport flow of chemicals between air and water have referred to MAMI III model and are below:[3]

Water-air mass transport coefficient ($MTC_{water-air}$):

$$MTC_{water-air} = \frac{1}{\frac{f_{dissolved,n}}{MTC_{water-air,w}} + \frac{1}{K_{aw} \times MTC_{air}}} \quad (3)$$

$$MTC_{water-air,w} = 0.01 \times (0.0004 + 0.00004 \times windspeed^2) \times (32/MW)^{0.25} \quad (4)$$

$$MTC_{air} = 0.001 \times (0.3 + 0.2 \times windspeed) \times (18/MW)^{0.335} \quad (5)$$

Gas absorption flow from air to water:

$$GasAbs = f_{aerosolw,n} \times MTC_{water-air} \quad (6)$$

Volatilization flow from water to air:

$$\text{Volatilization} = f_{dissolved,n} \times MTC_{water-air} \quad (7)$$

$f_{dissolved,n}$: fraction of neutral molecules of chemicals dissolved in water phase (-);

$MTC_{water-air,w}$: water side mass transport coefficient between air and water (m/s);

MTC_{air}: air side mass transport coefficient (m/s);

MW: molecular weight (g/mol);

K_{aw}: dimensionless in SESAMe v3.3;

$f_{aerosolw,n}$: fraction of neutral molecules in aerosol water (-);

Freshwater pH generation To aim the ionisable chemicals in this study, spatially varied freshwater pH has been produced. Weekly reported freshwater pH (average taken from gauging data measured every four hours during the week) from 99 gauging stations (located as shown in Figure S1a) in China has been collected from the database of Ministry of Environmental Protection (MEP) of China for the whole year 2012.[6] The annual average freshwater pH has been calculated for each gauging site. A continuous surface water pH layer (1 km^2) has been generated by interpolating these point values by inverse distance weighting (IDW) interpolation method in ArcGIS 10.2.2. The freshwater pH layer can only cover the middle and the east of China (Figure S1a), as the water gauging stations all distribute in these areas. The 1-km^2 freshwater pH has been aggregated to 50×50 km^2 grid with a range of 6.8-8.6 and median 7.8. The median pH has been taken to complement the missing freshwater pH for western China (Figure S1b). The white area in Figure S1b indicates no surface water there.

Soil pH distribution Figure S2 shows the spatial distribution of pH for agricultural soil across China. The white area indicates no agricultural soil there.

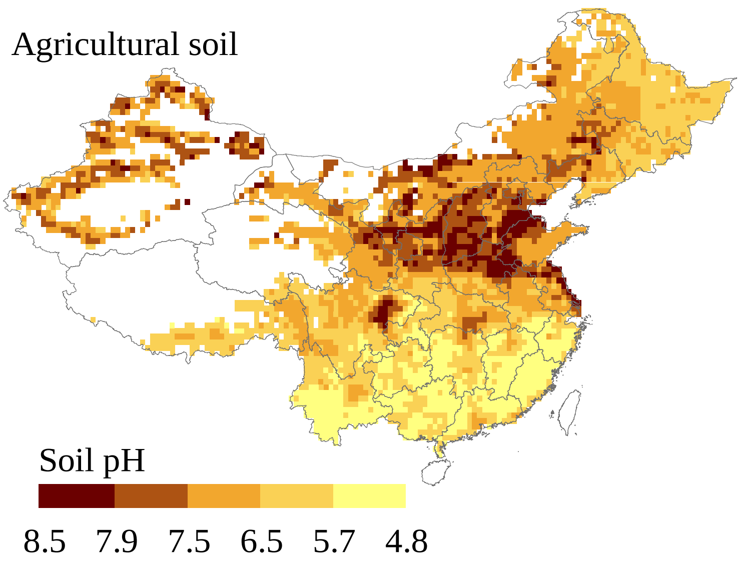

Figure S2 spatial distribution of agricultural soil pH in China

Chemical properties chemical properties related to partitioning were mostly measured values from experimental studies collected from literature.

Table S1 physico-chemical properties of the six chemicals

	Chemical	Molecular weight ($g{\cdot}mol^{-1}$)	Vapor pressure at 25 ˚C (Pa)	Water solubility at 25 ˚C ($g{\cdot}m^{-3}$)	Log K_{OW} (-)	pKa (-)	Half-life in air (hr)	Half-life in water (hr)	Half-life in sediment (hr)	Half-life in soil (hr)
Antibacterial	Triclosan (TCS)	289.54	6.9E-4[a]	8.8[b]*	4.8[b]	7.9[c]	15.9[d]	36[a]	12960[e]	2880[e]
	Triclocarban (TCC)	315.58	4.6E-11[i]	0.0237[b]	5.8[b]	12.7[c]	12.1[d]	1440[e]	12960[e]	2880[e]
	Climbazole	292.8	9.6E-4[b]	58[b]	3.83[b]	7.5[f]	4.87[g]	1440[g]	13000[g]	2880[g]
UV filters	Benzophenone-3 (BP-3)	228.24	1.1E-3[b]	6[b]	3.6[b]	9.54[h]	1.28[g]	900[g]	8100[g]	1800[g]
	Octocrylene (OC)	361.48	4.2E-7[b]	3.8E-3[g]	6.9[g]	neutral	7.9[g]	360[g]	3240[g]	720[g]
	octyl methoxycinnamate (OMC)	290.4	9.9E-3[g]	0.1548[g]	5.8[g]	neutral	4.17[g]	360[g]	3240[g]	720[g]

Notes: MW, molecular weight; VP, vapour pressure; WS, water solubility.
* extrapolated by ECHA experimental data for solubility at 25 °C (3.6 mg/L at 10 °C; 6.5 mg/L at 20 °C; 10.8 mg/L at 30 °C).[7]
a, McMahon et al., 2008;[8]
b, ECHA registered chemical database;[7] c, Loftsson et al., 2005;[9]
d, Zhao et al., 2013;[10]
e, Halden et al., 2005;[11]
f, Chen et al., 2013;[12]
g, EPI Suite;[13]
h, Castro et al., 2003;[14]
i, US EPA, *Initial Risk-Based Prioritization of* HPV chemicals.[15]

Methods of emission estimation

Chemical inclusion levels Inclusion levels of chemicals in products are shown in Table S2.

Table S2 Inclusion levels of chemicals in different sub-categories of products

Chemicals	Categories	Sub-categories	Inclusion level
TCS	Oral Hygiene		0.30%[16]
	Soap & Bath Products		
	Dishwashing Products		
	Fabric Care		
	Skincare		
	Hair Products		
	Diapers & Feminine Hygiene		
	Deodorants		
	Hard Surface Care		
	Toilet Care		
	Fragrances		
	Shaving & Depilatories		
TCC	Soap & Bath Products		0.30%[16]
	Dishwashing Products		
	Fabric Care		
	Hair Products		
	Diapers & Feminine Hygiene		
	Deodorants		
	Hard Surface Care		
	Toilet Care		
	Fragrances		
	Shaving & Depilatories		
Climbazole	Deodorants		0.50%[17]
	Feminine Hygiene Products		2.00%[17]
	Hair products	Conditioner	0.50%[17]
		Hair Styling	0.50%[17]
		Hair Treatments	0.50%[17]
		Shampoo	1.00%[18]
	Skincare products (body, neck and face)	Body Care	0.50%[17]
		Face/Neck Care	0.50%[17]
	Soap & Bath Products	Bar Soap	2.00%[19]
		Shower Products	2.00%[19]
OMC	Deodorants		4.24%[b]
	Dishwashing-hand		4.24%[b]
	Hair products	Conditioner	3.00%[18]
		Hair Styling	3.00%[18]
		Hair Treatments	3.00%[18]
		Shampoo	3.00%[18]
	Shaving & Depilatories	Depilatory	2.94%[20]

		Products	
		Shaving Preparations	2.94%[20]
	Skincare	Body Care	7.14%[18]
		Face/Neck Care	5.58%[a]
		Hand/Nail Care	7.38%[a]
		Lip Care	3.36%[20]
		Sun - After Sun	4.24%[b]
		Sun - Sun/Sunbed Exposure	6.99%[a]
	Soap & Bath Products	Bar Soap	4.24%[b]
		Liquid Soap	4.24%[b]
		Shower Products	4.24%[b]
OC	Deodorants		7.00%[c]
	Hair products	Conditioner	10.00%[18]
		Hair Styling	10.00%[18]
		Hair Treatments	10.00%[18]
		Shampoo	10.00%[18]
	Skincare	Body Care	2.00%[18]
		Face/Neck Care	3.88%[a]
		Hand/Nail Care	2.76%[a]
		Lip Care	7.40%[a]
		Sun - After Sun	7.00%[c]
		Sun - Sun/Sunbed Exposure	5.69%[a]
	Soap & Bath Products	Liquid Soap	7.00%[c]
		Shower Products	7.00%[c]
BP-3	Deodorants		4.84%[d]
	Hair products	Conditioner	6.00%[18]
		Hair Styling	6.00%[18]
		Hair Treatments	6.00%[18]
		Shampoo	6.00%[18]
	Shaving & Depilatories		4.84%[d]
	Skincare	Body Care	3.50%[18]
		Face/Neck Care	4.64%[a]
		Hand/Nail Care	3.00%[18]
		Lip Care	3.62%[a]
		Sun - After Sun	4.84%[d]
		Sun - Sun/Sunbed Exposure	5.40%[18]
	Soap & Bath Products	Bar Soap	4.84%[d]
		Liquid Soap	4.84%[d]
		Shower Products	4.84%[d]

Notes: a, mean value of the inclusion level reported by Manová E et al.[20] and in Mintel database[18];
b, mean value of inclusion levels of OMC in the other products under the same category;
c, mean value of inclusion levels of OC in the other products under the same category;
d, mean value of inclusion levels of BP-3 in the other products under the same category.

Usage The usage (tonnes) of each chemical in products being sold in Chinese market for year 2012 has been calculated as equation (8).

$$\text{Usage} = \sum_i F_i \times T_i \times I_i \quad (8)$$

where i represents (sub)category i; F_i, T_i, I_i are respectively the fraction of products containing a specific chemical sold in Chinese market, the total tonnage of products sold in Chinese market (tonnes) and the inclusion level of the chemical in (sub)category i.

Percentage of wastewater connected to WWTPs Population connected to WWTPs has been estimated as the province-level domestic consumption wastewater discharge (National Bureau of Statistics (NBS) of China, 2009)[21] divided by the daily per capita domestic water use for that province. And then the percentage of the provincial population connected to WWTPs has been calculated and assigned to each county within that province. Due to the lack of specific data from the NBS of China, assumptions have been made such that urban and rural per capita water use is the same; and the urban population has been assigned WWTP connectivity before any rural population. If the estimated total population connected to WWTP exceeds the urban population in the same province, the remaining population have been considered as rural population and the percentage of rural population connected to WWTPs has been assigned to each county in that province. Province-level values used to calculate the population connected are included in Table S3.

Removal ratio of climbazole in WWTPs The predicted removal ratio of climbazole in WWTPs by SimpleTreat 3.2[22] is 12%, which is out of the measured range (18 - 67%) from the literature. Climbazole is set as inherently not biodegradable in SimpleTreat model based on biodegradation tests with initial concentrations of 4 mg/L and 100 mg/L in water reported by European Chemical Agency (ECHA),[7] however its actual concentration in raw wastewater to WWTPs is normally much lower (e.g. 1.4 ug/L[23] in Germany or 0.6-1.0 ug/L[24] in Beijing). It is well documented that under ready test conditions the unrealistic high concentration of test material can cause toxicity in the system and underestimate the removal.[25] Therefore, 40% has been taken for climbazole as stated in the main paper.

Table S3. Table of province level information used to develop estimates of WWTP connectivity

Province	Population (10,000) (2008)[1]	Urban Population (10,000) (2008)[1]	Rural Population (10,000) (2008)[1]	Per Capita Water Use (L/day)[2]	Domestic Consumption Wastewater Discharge (m3/day)[3]	Total Population Connected (10,000) (2008)	Total Population Connected (%)	Urban WWTP Connectivity (%)	Rural WWTP Connectivity (%)
Anhui	6,135	2,485	3,650	123	2,785,288	2,271	37%	91%	0%
Beijing	1,695	1,439	256	252	2,873,753	1,139	67%	79%	0%
Chongqing	2,839	1,419	1,420	169	2,139,342	1,269	45%	89%	0%
Fujian	3,604	1,798	1,806	167	2,637,589	1,579	44%	88%	0%
Gansu	2,628	845	1,783	96	851,096	883	34%	100%	2%
Guangdong	9,544	6,048	3,496	259	12,713,370	4,905	51%	81%	0%
Guangxi	4,816	1,838	2,978	286	3,824,932	1,339	28%	73%	0%
Guizhou	3,793	1,104	2,689	117	1,210,164	1,038	27%	94%	0%
Hainan	854	410	444	202	827,315	409	48%	100%	0%
Hebei	6,989	2,928	4,061	92	3,110,274	3,381	48%	100%	11%
Heilongjiang	3,825	2,119	1,706	135	1,974,959	1,466	38%	69%	0%
Henan	9,429	3,397	6,032	102	4,823,260	4,747	50%	100%	22%
Hubei	5,711	2,581	3,130	148	4,525,671	3,056	54%	100%	15%
Hunan	6,380	2,689	3,691	194	4,328,521	2,233	35%	83%	0%
Inner Mongolia	2,414	1,248	1,166	167	1,130,247	675	28%	54%	0%
Jiangsu	7,677	4,169	3,509	177	6,841,151	3,861	50%	93%	0%
Jiangxi	4,400	1,820	2,580	146	1,924,055	1,316	30%	72%	0%
Jilin	2,734	1,455	1,279	133	1,902,137	1,427	52%	98%	0%
Liaoning	4,315	2,591	1,724	156	3,532,822	2,261	52%	87%	0%
Ningxia	618	278	340	73	479,452	659	100%	100%	100%

Qinghai	554	227	327	165	353,397	214	39%	94%	0%
Shaanxi	3,762	1,584	2,178	102	1,545,370	1,513	40%	96%	0%
Shandong	9,417	4,483	4,935	99	4,984,493	5,046	54%	100%	11%
Shanghai	1,888	1,673	215	327	4,983,014	1,524	81%	91%	0%
Shanxi	3,411	1,539	1,872	79	1,801,671	2,285	67%	100%	40%
Sichuan	8,138	3,044	5,094	116	4,209,397	3,625	45%	100%	11%
Tianjin	1,176	908	268	117	1,117,699	958	81%	100%	18%
Tibet	287	65	222	218	68,384	31	11%	48%	0%
Xinjiang	2,131	845	1,286	158	1,419,863	898	42%	100%	4%
Yunnan	4,543	1,499	3,044	135	1,393,671	1,032	23%	69%	0%
Zhejiang	5,120	2,949	2,171	195	4,106,548	2,101	41%	71%	0%
National	130,827	61,477	69,350	153	90,418,904	59,097	45%	89%	6%

[1] National Bureau of Statistics of China, 2009 (Table 3-4)[21]
[2] National Bureau of Statistics of China, 2009 (Table 11-18)[21]
[3] National Bureau of Statistics of China, 2009 (Table 11-19)[21]
Note: National values for water use and WWTP connectivity are population-weighted

Toxicity data calculation

Orvos et al.[26] has indicated in their article that the concentration of the neutral molecules of TCS for EC50 is relatively constant at four different pH levels, which has been obtained from an exposure testing to daphnia. Roberts et al.[27] has provided toxicity data of TCS at pH levels of 7, 8 and 8.5 as shown in Table S4, which has been obtained from algae exposure testing. It also demonstrates that for each toxicity level, the concentration of neutral forms is relatively constant under different pH levels as shown in Table S4.

Table S4 toxicity data of TCS and its neutral concentration at different pH levels (ug/L)

pH	NOEC	$NOEC_{\cdot n}$*	LOEC	$LOEC_{\cdot n}$*	EC_{10}	$EC_{10 \cdot n}$*	EC_{20}	$EC_{20 \cdot n}$*	EC_{50}	$EC_{50 \cdot n}$*
8.5	25.3	5.1	4.3	0.9	36.5	7.3	65.1	13.1	175.1	35.2
8.0	3.43	1.5	6.0	2.6	5.0	2.2	18.6	8.2	175.9	77.9
7.0	3.92	3.5	1.1	1.0	3.7	3.3	6.4	5.7	16.8	14.9

Notes: $EC_{x.n}$, $NOEC_{.n}$ and $LOEC_{.n}$, the concentration of the neutral form of corresponding toxicity data

Emissions estimated

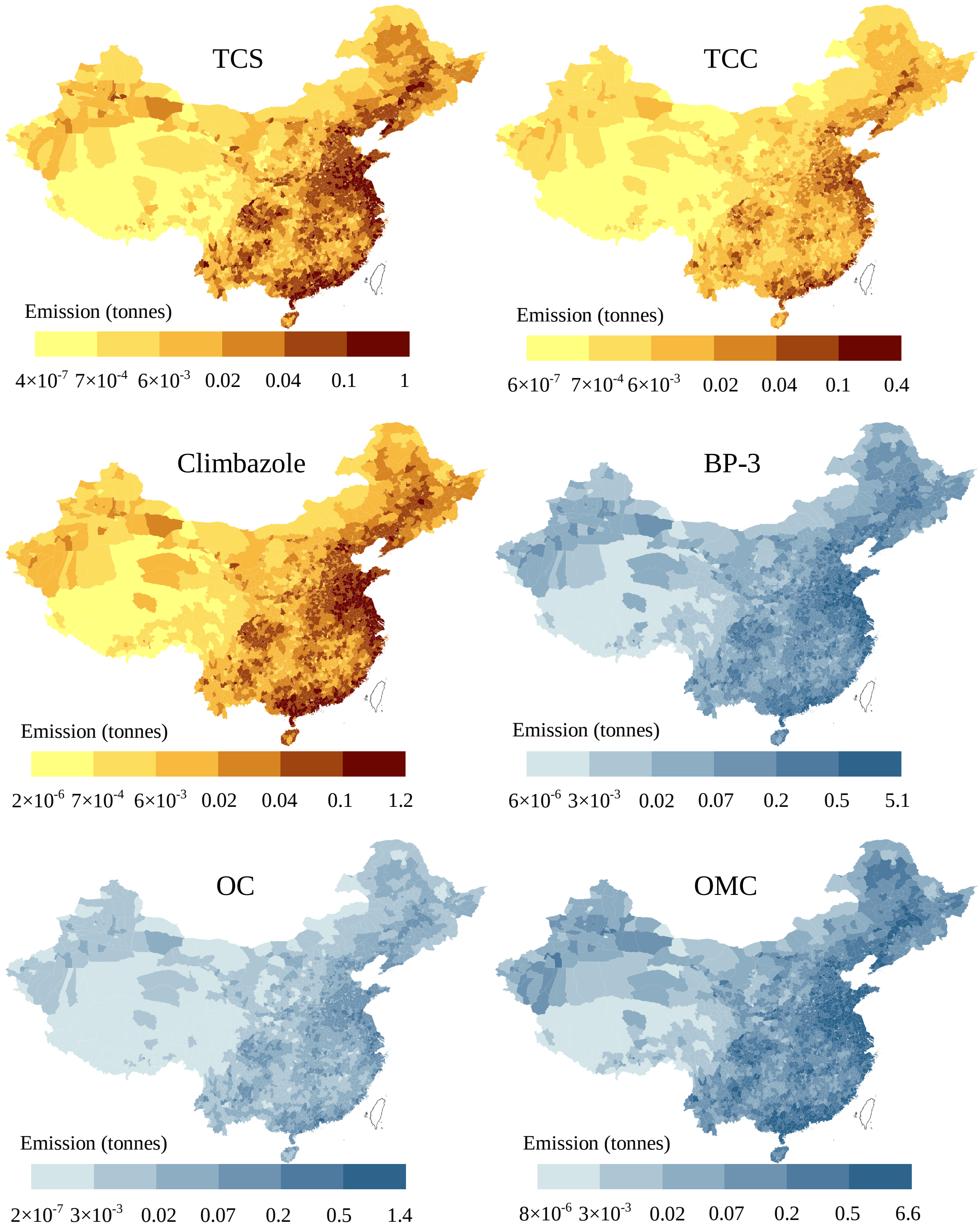

Figure S3 Emissions of three antibacterial agents (TCS, TCC and climbazole) and three UV-filters (BP-3, OC and OMC) by county

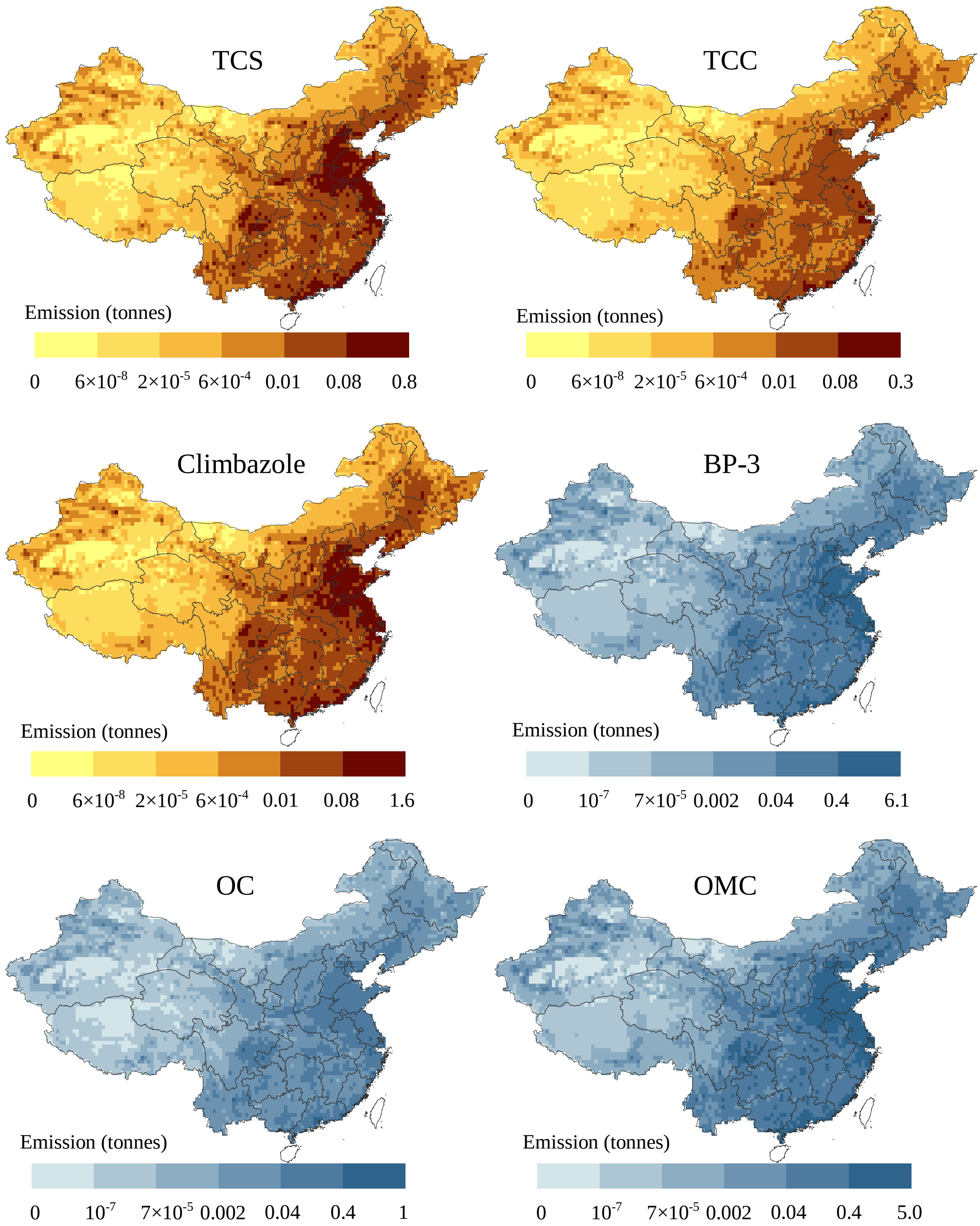

Figure S4 Emissions of three antibacterial agents (TCS, TCC and climbazole) and three UV-filters (BP-3, OC and OMC) by 50 ×50 km^2 grid

Model evaluation Methods

Method of sensitivity analysis (SCV) variability-based sensitivity coefficient has been used for the sensitivity analysis described in below equation.[28]

$$\mathrm{SCV} = \frac{\Delta CV_i^Y / CV_i^Y}{\Delta CV_i^X / CV_i^X} \tag{9}$$

where CV_i^X and CV_i^Y are respectively the coefficient of variance (CV) of values of the ith input variable vector and the corresponding ith predicted output. ΔCV_i^X and ΔCV_i^Y are their differences. The probability distribution of environmental parameters could be found in previous study by Zhu et al.[1] Parameters will be identified to be more influential to the model outputs if the corresponding SCV > 0.1.

Model outputs validation The sampling sites location of measurements from the literature is shown in Figure S5. The sampling size and related references are shown in Table S5.

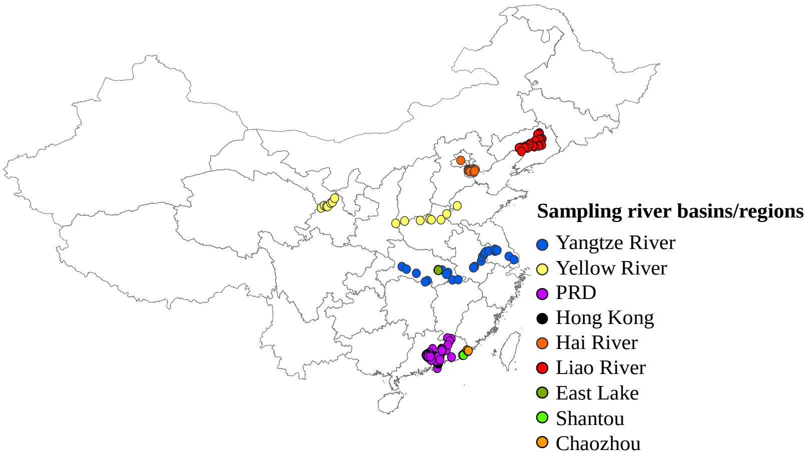

Figure S5 location of sampling sites of measurements

Table S5 Information for measured data collected from the literature

Chemical	Sampling sites	Sampling year	No. of sampling sites	reference
TCS TCC	Liao River	2007-2009	21	Zhao et al., 2013[10]
	Hai River	2007-2009	11	
	Yellow River	2007-2009	15	
	Dongjiang	2007-2009	22	
	Pearl River	2007-2009	17	
Climbazole	Hai River	2009	13	Heeb et al., 2012[29]
	Dongjiang	2012	21	Chen et al., 2014[30]
	Yangtze River	2013	24	Zhang et al., 2015[31]
	Yangtze River	2009-2010	3	Qi et al., 2014[32]
BP-3	East lake	2013-2014	15	Wu et al., 2015[33]
	Shantou	2012	4	Tsui et al., 2014[34]
	Chaozhou	2012	3	Tsui et al., 2014[34]
	Hong Kong	2012	4	Tsui et al., 2014[34]
OC OMC	Shantou	2012	4	Tsui et al., 2014[34]
	Chaozhou	2012	3	
	Hong Kong	2012	4	

Model evaluation results

Figure S6-S9 show the comparison between predictions and measurements.

The results of sensitivity analysis are shown in Table S6.

The probability distribution of the model output by Monte Carlo simulation is shown in Figure S10.

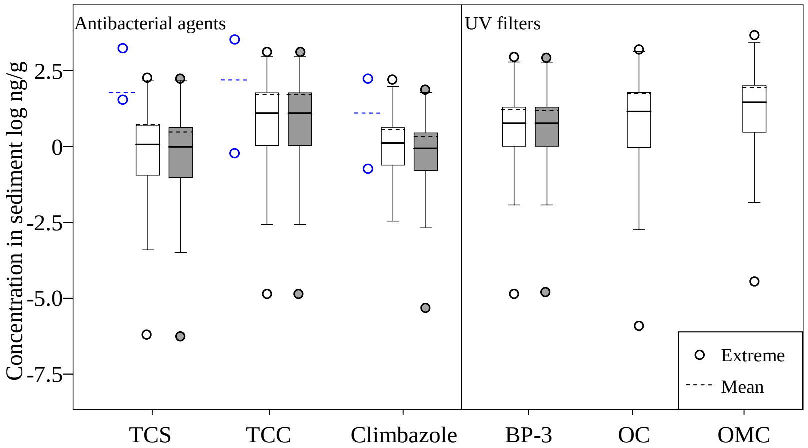

Figure S6 Boxplot (SPSS 18) of predicted total and neutral chemical concentration (predictions from all grid cells), and measurements in freshwater sediment (blue). White-box group indicates predicted total chemical concentrations (neutral plus ionic molecules); grey-box group indicates predicted neutral concentration; the horizontal solid line in the box is the median; the top and bottom of the box are respectively the 75th and 25th percentile; the top and bottom of the whisker are respectively the highest and lowest case within 1.5 times the interquartile range. Extreme circles are max/min values.

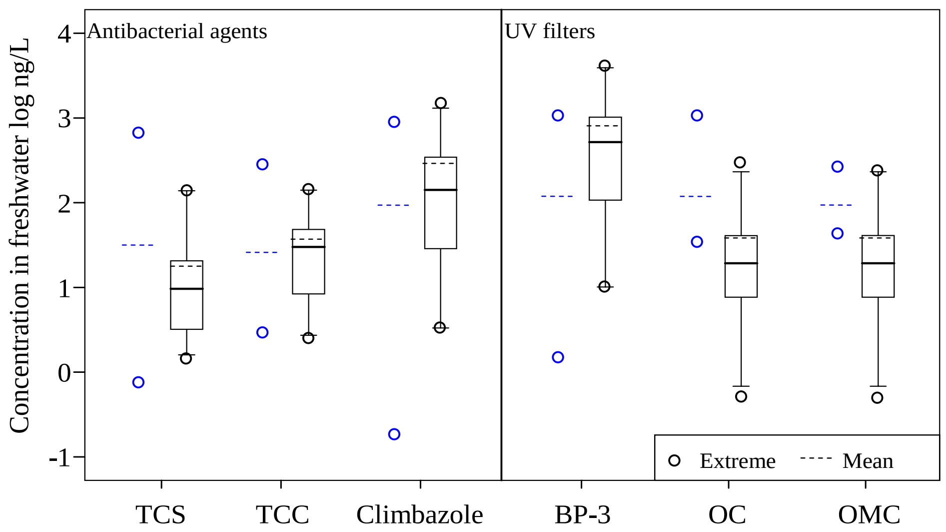

Figure S7 Comparison of statistical distribution of predicted and measured (blue) concentration in water. The white-box group indicates predicted concentrations. Predicted concentrations are values of only grid cells which cover the sampling sites of the measured data; the horizontal solid line in the box is the median; the top and bottom of the box are respectively the 75th and 25th percentile; the top and bottom of the whisker are respectively the highest and lowest case within 1.5 times the interquartile range. Extreme circles are max/min values.

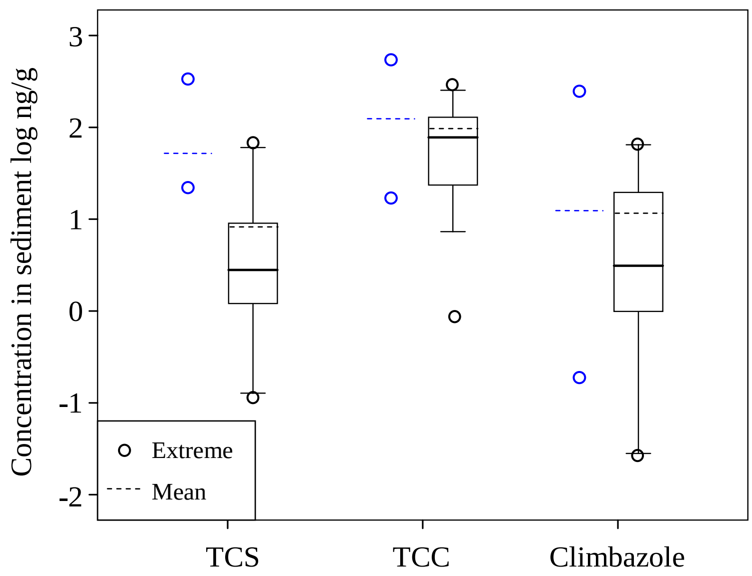

Figure S8 Comparison of statistical distribution of predicted and measured (blue) concentration in sediment. The white-box group indicates predicted concentrations. Predicted concentrations are values of only grid cells which cover the sampling sites of the measured data; the horizontal solid line in the box is the median; the top and bottom of the box are respectively the 75th and 25th percentile; the top and bottom of the whisker are respectively the highest and lowest case within 1.5 times the interquartile range. Extreme circles are max/min values.

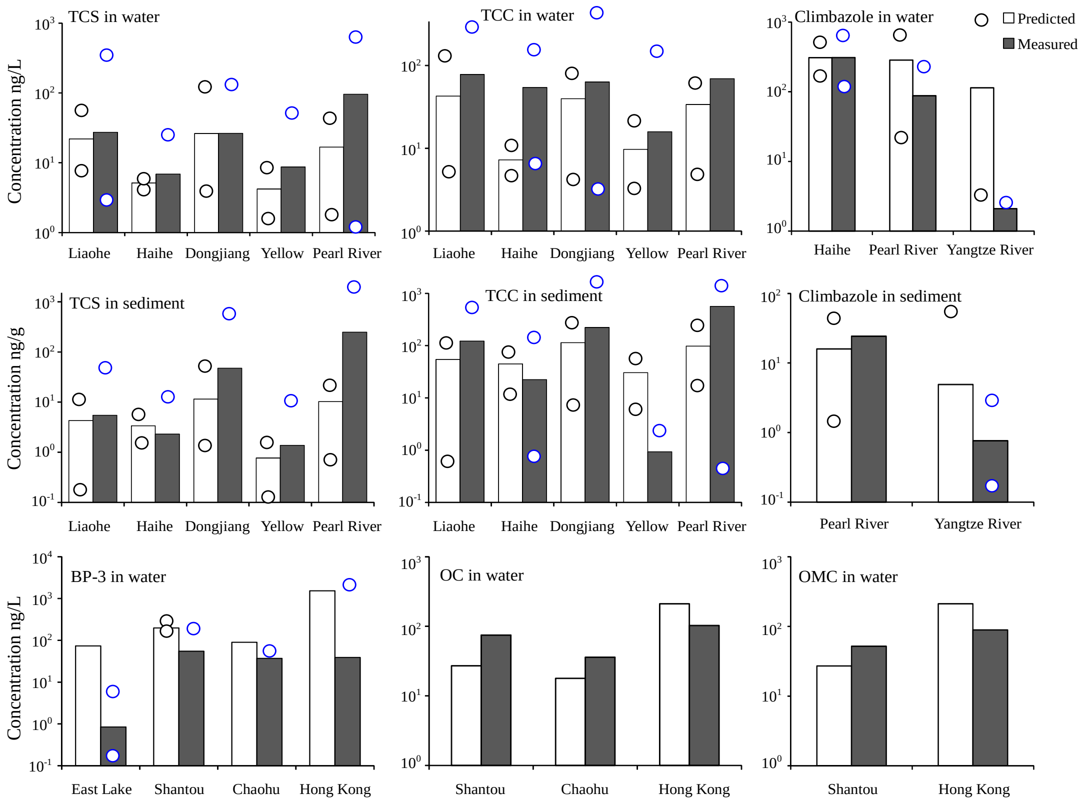

Figure S9 Comparison of predicted and measured concentrations in certain river/lake catchment; circles are extreme values for measured and predicted data; there is only one grid cell which covers the limited sampling sites for OC and OMC in water, so the extreme values are not added;

As the value for each sampling site is normally not provided but only mean is reported in the literature, the measured concentration in Figure S9 is the mean chemical concentration for individual river/lake catchment reported in the literature. If more than one monitoring study focus on the same river/lake catchment, measurements have been averaged for validation. The predicted concentration is the mean value of predicted concentrations for grid cells spatially covering the sampling sites for individual river/lake catchment in literature listed in Table S5 for each chemical.

Table S6 Index of SCV sensitivity analysis (>0.1)

	Triclosan		Triclocarban		Climbazole		Benzophenone-3		Octocrylene		Octyl methoxycinnamate	
Environmental parameters	Water	Sediment	Water	Sediment	Water	Sediment	Water	Sediment	Water	Sediment	Water	Sediment
Temperature	4.4	7.2	11.5	14.9	13.2	15.8	40.7	51.7	47.1	71.3	27.0	41.2
Wind speed	0.2	0.2			0.1		4.5	4.2			1.6	0.6
Precipitation		0.2						0.2				
Surface water depth	10.1	15.7	1.4	1.2	3.6	3.2	10.2	11.6	0.7	0.7	1.9	1.6
Water inflow	3.6	3.8	9.4	9.2	0.1	0.9	6.3	2.8	11.7	12.1	5.5	6.4
Water outflow	19.4	21.6	3.9	4.7	15.2	12.5	27.2	14.0	19.0	14.7	4.3	6.1
Runoff												
Soil water erosion rate												
Soil OC												
Soil Density	1.4	0.5	4.2	0.7	1.5	5.7	14.5	0.4	5.9	0.5	1.7	2.4
Sediment OC	2.3		0.4		0.4	1.0	4.8	1.3	0.2		0.5	0.7
Soil pH					0.2	0.2						
Freshwater pH	0.7	10.5			19.6	79.9	0.3	0.2				
Emission	118.1	210.5	4.9	4.7	3.0	4.7	1.1	1.5	2.7	2.6	6.1	6.1

The parameters with index higher than 0.1 are more influential than those with index lower than 0.1 (not shown in above table).

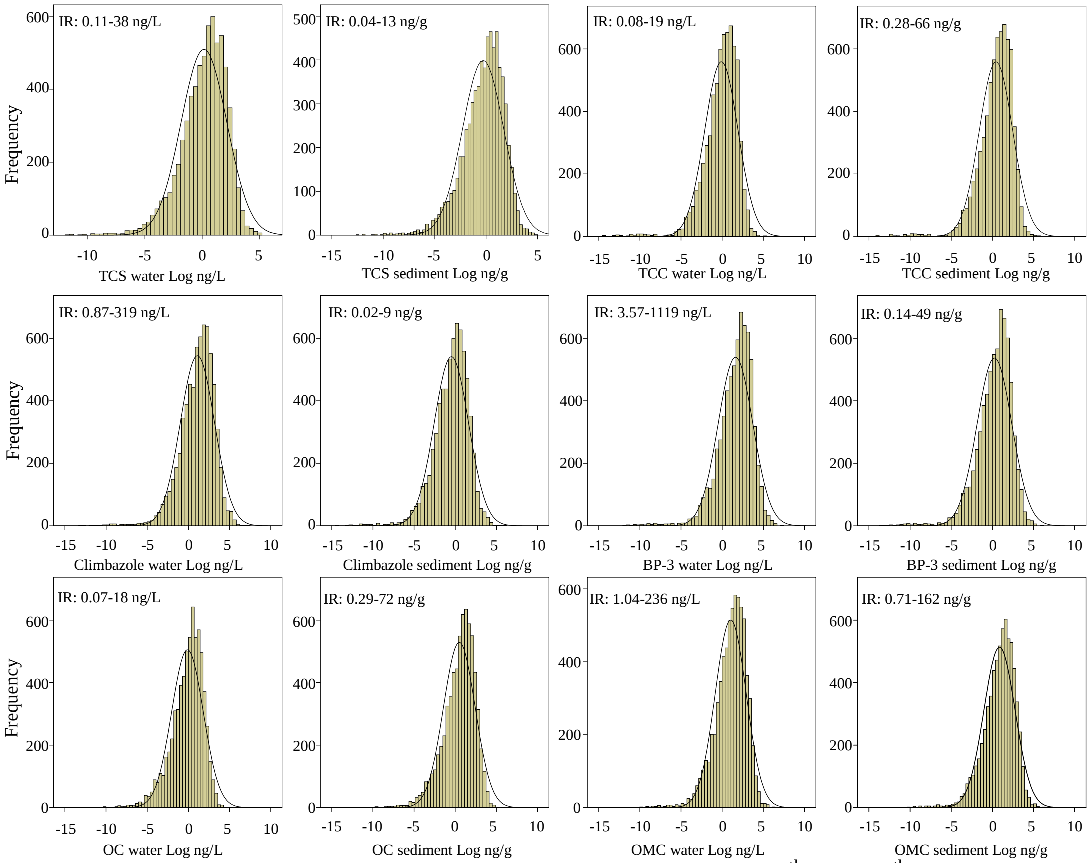

Figure S10 Lognormal distribution and interquartile range (IR, 25th and 75th percentiles) of predicted chemical concentration in freshwater and sediment by Monte Carlo simulation

Geographic distribution of six chemicals in freshwater and sediment across China

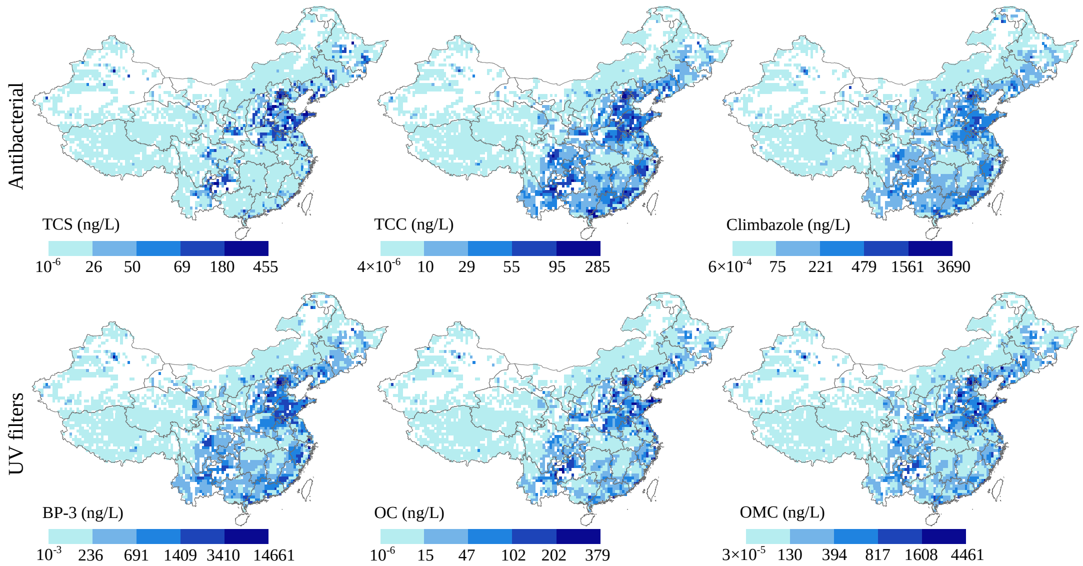

Figure S11 Predicted concentration for six chemicals in freshwater

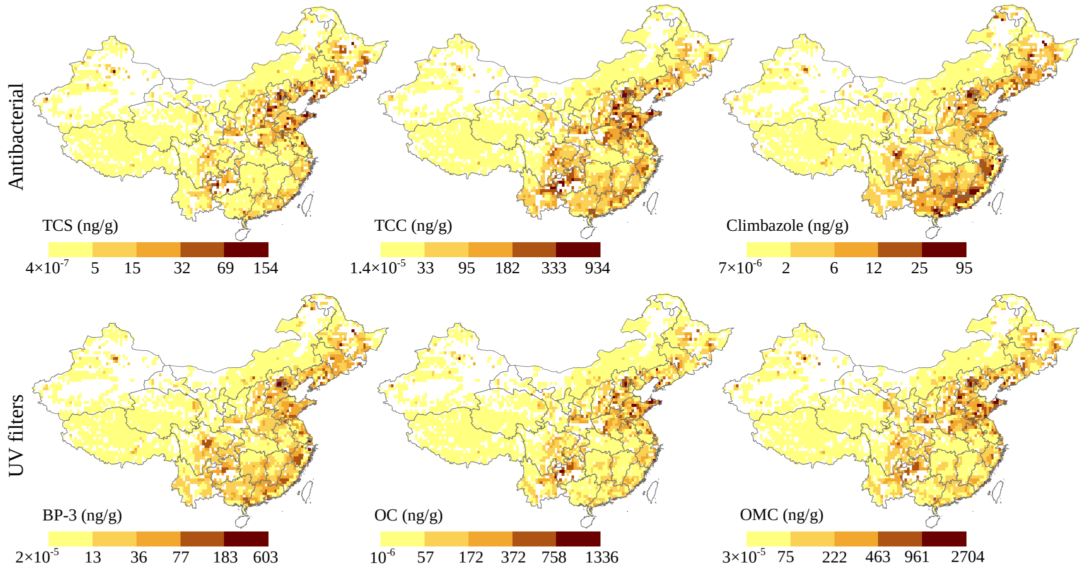

Figure S12 Predicted concentration for six chemicals in freshwater sediment

Range of concentration (5th - 95th, plus mean) at eastern coastal area is, in sea water 2×10^{-3} - 1.7 (0.4) ng/L for TCS, 0.02 - 11 (2.6) ng/L for TCC, 0.6 - 67 (19) ng/L for climbazole, 1 - 200 (55) ng/L for BP-3, 5×10^{-3} - 12 (2) ng/L for OC and 0.1 - 73 (17) ng/L for OMC; sea sediment, 6×10^{-4} - 0.6 (0.1) ng/g for TCS, 0.05 - 31 (6) ng/g for TCC, 0.02 - 3 (0.7) ng/g for climbazole, 0.05 - 9 (2) ng/g for BP-3, 0.01 - 25 (4) ng/g for OC and 0.03 - 22 (4) ng/g for OMC.

Table S7 Comparison of emission and predicted concentration range (Min - Max) in freshwater and sediment for TCS and climbazole between this study and Zheng et al.'s studies

TCS	Emission	Water concentration range	Sediment concentration range
This study	95 tonnes/yr	10^{-6} – 455 ng/L	4×10^{-7} – 154 ng/g
Zhang's study[35]	66.1 t/yr	0.0156 – 39.1 ng/L	0.00215 – 14.8 ng/g
climbazole	Emission	Water concentration range	Sediment concentration range
This study	107 tonnes/yr	6×10^{-4} – 3690 ng/L	7×10^{-6} – 95 ng/g
Zhang's study[31]	254 t/yr	0.2 - 367 ng/L	0.009 – 25.2 ng/g

As the concentration range reported in studies by Zhang et al.[31, 35] is Min - Max (minimum - maximum) instead of 5^{th} - 95^{th} percentiles, range of Min – Max has been taken for this study for comparison in Table S7.

Chemical partitioning in environmental media

Figure S11 and Figure S12 show respectively the nationally averaged chemical distribution of the six chemicals in different media and regional variation of TCS distribution in media around Yangtze River Delta as an example at steady state. It should be noted that they all show the percentage of total amount of a chemical in the bulk of environmental media but do not reflect the relationship of concentration levels of a chemical in different environmental media, e.g. concentrations of chemicals are relatively higher in freshwater and sediment but very low in soil including TCS, climbazole and BP-3 although the amount of the three chemicals distributing in soil is over 50% of all at steady state. And this is under the scenario that chemicals are only released to freshwater.

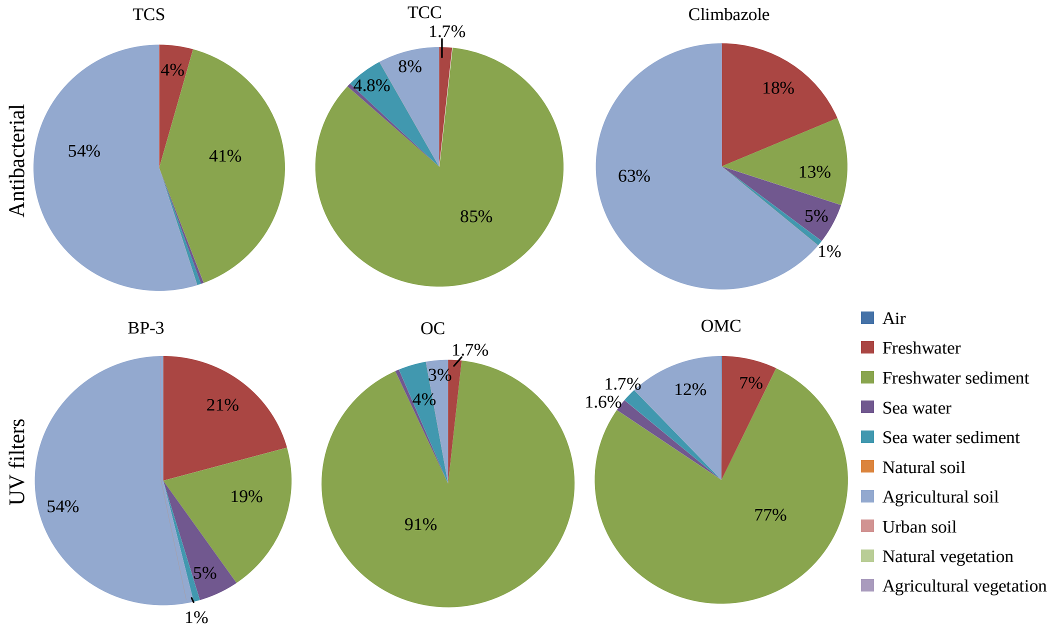

Figure S13 nationally averaged chemical distribution in different media at steady state in the scenario that chemicals are only released to freshwater; the percentage was calculated by the amount of chemical in the bulk of environmental media at steady state

Table S8 Flux (mean ± standard deviation) to agricultural soil from respectively freshwater irrigation, deposition and absorption from air and vegetation litter in national scale (Unit, mol/day/m^2)

Chemical	Irrigation	Deposition and absorption from air	Vegetation litter
TCS	$2.0\times10^{-11}\pm4.4\times10^{-11}$	$1.5\times10^{-15}\pm4.5\times10^{-15}$	$4.5\times10^{-18}\pm1.1\times10^{-17}$
climbazole	$1.1\times10^{-10}\pm1.8\times10^{-10}$	$7.9\times10^{-16}\pm4.5\times10^{-15}$	$1.7\times10^{-16}\pm3.6\times10^{-16}$
BP-3	$5.2\times10^{-10}\pm9.0\times10^{-10}$	$1.6\times10^{-15}\pm1.5\times10^{-14}$	$5.2\times10^{-16}\pm1.1\times10^{-15}$

As there is not any direct wastewater release to soil and sludge application to land, freshwater irrigation, deposition (wet + dry) and absorption from air and vegetation litter are three pathways that transport chemicals to agricultural soil. By comparing the fluxes among different pathways in Table S8, freshwater irrigation is the main way that transports the three chemicals to agricultural soil.

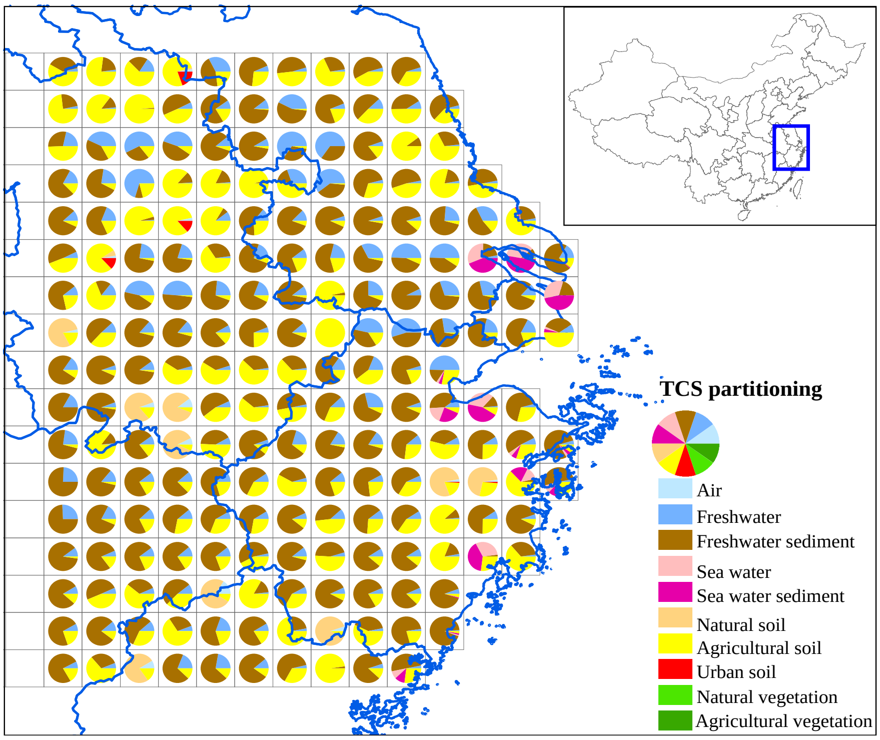

Figure S14 regional TCS distribution in different media around Yangtze River Delta at steady state in the scenario that chemicals are only released to freshwater; the percentage of TCS in pie charts was calculated by the amount of chemical in the bulk of environmental media

Level of chemicals in vegetation It is worth mentioning both the bulk amount and the concentration of chemicals in agricultural vegetation are very low. The percentage of the bulk amount of chemicals is close to zero as shown in Figure S11-S12. The median concentration of chemicals in agricultural vegetation is estimated at 2.3×10^{-9} (TCC) - 1.2×10^{-4} (BP-3) ng/g (dry weight, water contents in vegetation is assumed to be 95%), which is almost undetectable by devices for field samples.

There are evidences on plant uptake of home and personal care product ingredients from soils or under hydroponic conditions with relatively high concentrations in vegetation; however, these are mostly observed in laboratory or greenhouse experiments and concentrations in crops growing in realistic field conditions with biosolids application or treated wastewater irrigation are found very low.[36] This may result from various conditions including the chemical properties and system conditions, such as the medium the vegetation grows in, the pH in the growth medium and pH in cell etc.,[36] and perhaps needs to be studied further with more sophisticated model for vegetation or laboratory experiments.

Chemical transport between freshwater and sediment

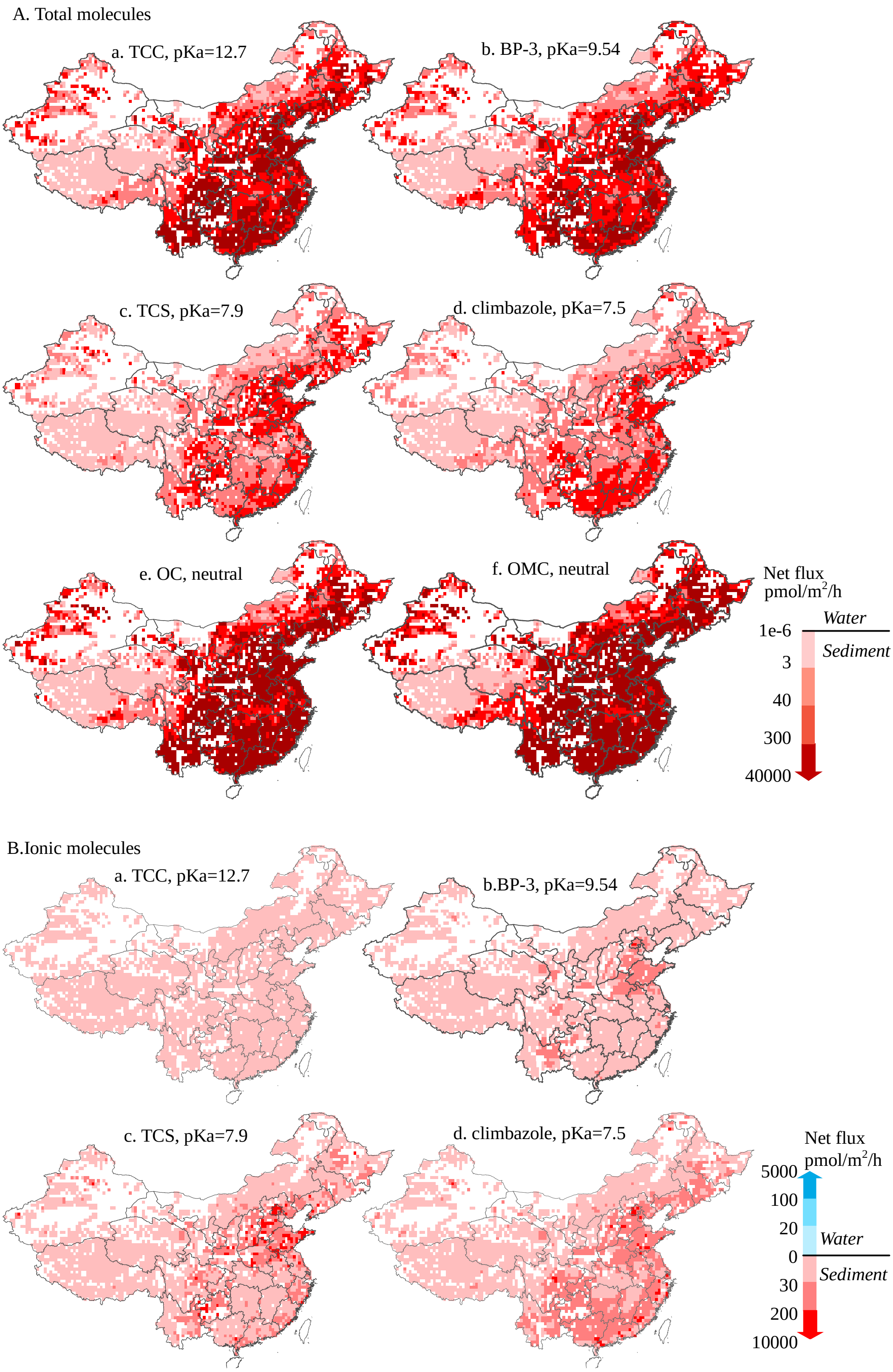

Figure S15 Net flux of total molecules of six chemicals (A) and ionic molecules of ionisable chemicals (B) between freshwater and sediment in the scenario that the chemical is all released to water.

Environmental risk assessment and limitations

It should be noted that due to the model limitations and uncertainty, SESEMe v3.3 should only be used provide information to assist with the identification of areas with higher or lower risk for a specific chemical, e.g. the red and blue areas in Figure 5b. However, this does not imply that all areas in blue are absolutely safe, nor that areas in red represent a definite risk. It can be used to identify areas that may represent higher risk and as such should be investigated in more detail. SESAMe v3.3 can provide average concentrations within each grid cell but cannot identify any hotspots which are on or close to emission sites. Indeed, it is not the intention of the model to provide an assessment of chemical risk on such a fine resolution, but to identify regions where a combination of chemical use/emission and environmental factors require a more detailed investigation.

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