

1 **Drivers of contaminant levels in surface water of China during 2000-2030: relative**
2 **importance for illustrative home and personal care product chemicals**

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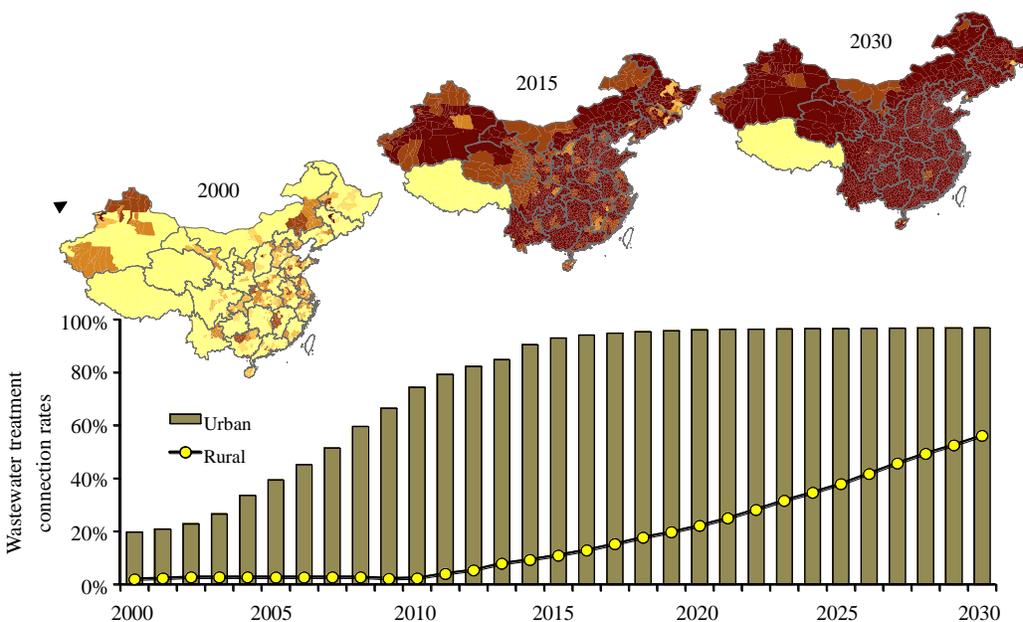
14 **Abstract**

15 Water pollution are among the most critical problems in China and emerging contaminants in
16 surface water have attracted rising attentions in recent years. There is great interest in China's
17 future environmental quality as the national government has committed to a major action plan
18 to improve surface water quality. This study presents methodologies to rank the importance of
19 socioeconomic and environmental drivers to the chemical concentration in surface water during
20 2000-2030. A case study is conducted on triclosan, a home and personal care product (HPCP)
21 ingredient. Different economic and discharge flow scenarios are considered. Urbanisation and
22 wastewater treatment connection rates in rural and urban areas are collected or projected for
23 2000-2030 for counties across China. The estimated usage increases from ca. 86 to 340 tonnes.
24 However, emissions decreases from 76 to 52 tonnes during 2000-2030 under a modelled
25 Organisation for Economic Co-operation (OECD) economic scenario because of the
26 urbanization, migration and development of wastewater treatment plants/facilities (WWTPs).
27 The estimated national median concentration of triclosan ranges 1.5-8.2 ng/L during 2000-2030
28 for different scenarios. It peaks in 2009 under the OECD and three of the Intergovernmental
29 Panel on Climate Change (IPCC), A2, B1 and B2 economic scenarios, but in 2025 under A1
30 economic scenario. Population distribution and surface water discharge flow rates are ranked

31 as the top two drivers to triclosan levels in surface water over the 30 years. The development
32 of urban WWTPs was the most important driver during 2000-2010 and the development of
33 rural works is projected to be the most important in 2011-2030 . Projections suggest discharges
34 of ingredients in HPCPs - controlled by economic growth - should be balanced by the major
35 expenditure programme on wastewater treatment in China.

36 **Keywords:** WWTPs, GDP, urbanization rates, population, surface water concentration, China

37 **TOC art**



38

39 1. Introduction

40 Water contamination can be harmful to human and ecosystem health. Emerging contaminants,
41 such as pharmaceuticals, home and personal care products (HPCPs) have raised growing
42 concerns (Boxall et al. 2012; EPA 2017). Reducing untreated wastewater and protecting
43 aquatic ecosystems are targets of the Sustainable Development Goals set by the United Nations
44 to be reached by 2030 (Hering et al. 2016). China is a country with major challenges of water
45 quality and availability, including: the size and diversity of the country and its rivers; the
46 population size and migration; rapid economic growth, with increased industrial, agricultural
47 and domestic demands for usable water and the effluents that these activities generate. The
48 Chinese Government therefore developed an ‘Action Plan for Water Pollution Prevention’ in
49 mid-2015 (MEP 2015a), which is laid out in the national 13th 5-Year Plan (CPGC 2016).
50 However, in order to make rational, effective and informed decisions which will improve water

51 quality, there is an urgent need for a methodology to identify the potential key drivers to affect
52 the level of contaminants in surface water, especially for those anthropogenic-source
53 contaminants. The results could improve and inform understanding, policy and decision-making.

54 Socio-economic activities and environmental changes have affected water quality in China
55 during the remarkable development over the past decades (Gleick 2008-2009). Ingredients in
56 HPCPs represent an interesting case, because they are closely linked to socio-economic
57 activities and as trace organics they could be markers of sewage and anthropogenic-source
58 ingredients (Gasser et al. 2010; James et al. 2016). Some HPCPs are relatively poorly studied
59 so far and have attracted increasing interest in recent years (Boxall et al. 2012) such as UV
60 filters and parabens, yet some of them are abundant and being considered for environmental
61 limits and more controlled usage, such as triclosan and triclocarban. HPCPs are diffusively
62 discharged in wastewaters. Their consumption increased between ~40~800% in China during
63 the economic boom between 2000 and 2012 (Euromonitor 2015); however, China also
64 increased its wastewater treatment capacity by about 8-fold during the same period ([Supporting
65 Information \(SI\) Figure S1](#)), due to several factors (i.e. rising urbanisation, compliance with
66 discharge standards) (MHURD 2013), potentially counterbalancing the potential release of
67 HPCPs to the environment. Other marked societal and infrastructure trends have occurred and
68 will continue in future. Population growth and migration and rapid urbanization progress (Yang
69 2013) across China make the change of ingredient usage and release more complex
70 geographically. Changing discharge flow, linked to environmental and infrastructure changes,
71 also affects the dilution of chemicals in surface water. These factors have changed / are
72 changing in a way that could be strongly impacting the concentrations and distributions of
73 chemicals in surface water and the water quality (Zhu et al. 2016; Zhu et al. 2014).

74 This study was therefore conceived to develop a modelling approach, to explore the potential
75 influence of several key drivers on past, present and future chemical surface water
76 concentrations in China. We address several drivers which will influence ingredient usage,
77 release and loading in aquatic systems, namely economic development as Gross Domestic
78 Product (GDP), population, urbanization, wastewater treatment capacity and discharge flow
79 rates, to estimate temporal changes in water concentrations. Measurements of pharmaceutical
80 and HPCP ingredients have only become available for a limited number of regions in recent
81 years in China. We chose triclosan as an example ingredient in the calculation as it is well
82 studied and there are more monitoring data available than other ingredients for model
83 validation (Zhu et al. 2016). It enters the aquatic environment diffusively, primarily from

84 domestic wastewaters. Scenarios considered here take account of main drivers discussed above
85 to: (1) model usage, emissions and concentrations in surface water of triclosan in China as an
86 example ingredient between 2000-2030, assuming that it continues to be used in HPCPs in that
87 period; (2) identify and rank the key drivers affecting surface water concentrations. This
88 approach has not been used before, but we believe it can be adapted and applied to a range of
89 chemicals from anthropogenic sources with different usage/release scenarios in future, using
90 the base data and modelling tools assembled here.

91 **2. Methodologies and approaches**

92 **2.1 Ingredient usage under five economic scenarios**

93 OECD (Organisation for Economic Co-operation) per capita GDP (OECD 2014) was found to
94 significantly correlate ($R^2 > 0.88$) to sales volumes (tonnes) of HPCP categories which contain
95 triclosan (SI Table S1 and Figure S2) (Mintel 2014) in the Chinese market for 2000-2019
96 (Euromonitor 2015). As seen in SI Figure S2, most correlations fit linear regression, except
97 those for shampoo, bleach/disinfectant and all-purpose surface care products (SI). Besides
98 these fast moving consumer HPCP categories in Table S1, triclosan is also used in plastic
99 materials, textiles, surface of medical devices, etc. These usages and releases were not taken
100 into account in this study as they are expected to be a small proportion of the total (Euromonitor
101 2015; SCCS 2010). In addition, emission pathways are complex depending on how these
102 materials are disposed of; and triclosan leaching from these materials is likely to be slow (SCCS
103 2010) compared to the daily used HPCP categories.

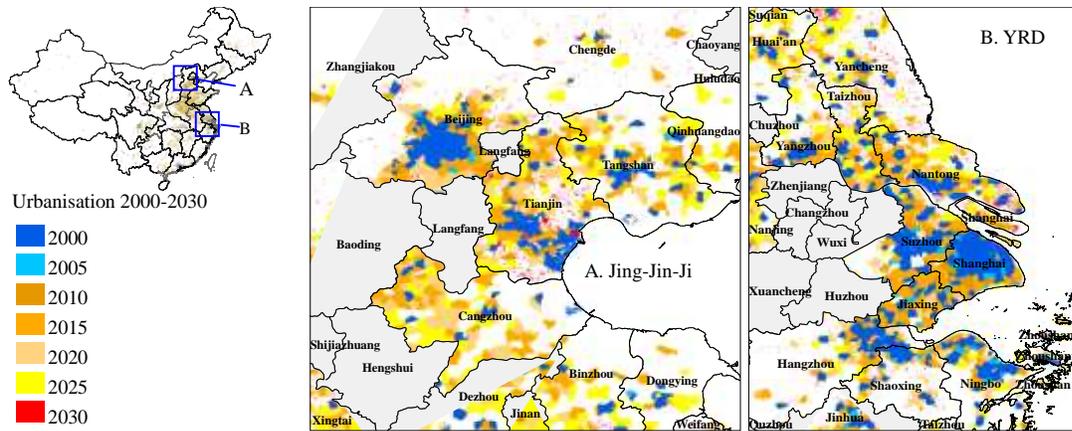
104 Based on above regressions, the annual sales volumes of HPCP categories which contain
105 triclosan were extrapolated for 2011-2030 under five economic scenarios, i.e. one from OECD
106 future GDP outlook (OECD 2014) and four predicted by the Centre for International Earth
107 Science Information Network (CIESIN) under four IPCC marker scenarios (A1, A2, B1 and
108 B2) (CIESIN 2002). OECD is an authoritative economic institution that could provide
109 potentially reasonable economic outlook as reference. IPCC scenarios consider extreme and
110 moderate conditions, which have been widely used in ecology or energy relevant studies. For
111 the feature of the five economic scenarios and their comparison see SI and Table S2. The two
112 ‘A’ economic scenarios were extreme; the two ‘B’ scenarios and the OECD projection are
113 moderate (SI Figure S3). The historical sales volume for 2000-2010 was directly taken from
114 the Euromonitor database (Euromonitor 2015). Products sold in a year were assumed to be
115 consumed within the same year. Triclosan usage was, therefore, estimated from product sales

116 volumes, the inclusion level in products and the percentage of product variants (Mintel 2014)
117 that contain triclosan for all product categories (Eq. 1 in SI). The product categories and
118 triclosan inclusion level (0.3%) (Hodges et al. 2012) were assumed identical during 2000-2030.
119 Based on the usage, emission and surface water concentrations were estimated under these five
120 economic scenarios with the projection of other main drivers described below.

121 It is assumed there will be no bans or elimination of triclosan in this study till 2030 in China.
122 However, future replacement or reduction of ingredient use is possible due to potential
123 government restrictions or if it is phased out by industry.

124 **2.2 Population and urbanization**

125 The gridded (~1 km) Chinese population count and density were projected by CIESIN for the
126 years 2000, 2005, 2015 and 2020 (CIESIN 2016a; b) and by another study for 2030 (Feng and
127 Qi 2016; Qi et al. 2015). For the year 2010, to keep identical to our previous study (Zhu et al.
128 2016), data projected by Landsan (Landsan 2010) was used. The urban population was
129 identified by the population density > 5000 (CIESIN 2011) cap/km² for 2030 and >1000
130 cap/km² for the other 5 years. 5000 cap/km² was suggested in some publications but found too
131 high on the population data by CIESIN. Therefore, 1000 cap/km² was additionally used in this
132 study, which could result in more reasonable results since they are verified with the census of
133 the national urban population (CNSTATS 2000-2015) (SI Figure S4) and the predicted
134 potential annual urban population growth rate for future China (ca. 2%). The projected urban
135 expansion from 2000 to 2030 is shown for every five years in Figure 1. To fill the population
136 data gaps between every two adjacent years with existing projected data (e.g. years between
137 2000 and 2005), it was assumed that the annual change rate of urban and rural population would
138 be steady between the two years. The projected population was applied to the five economic
139 scenarios to spatially allocate ingredient usage and to estimate population connectivity to
140 wastewater treatment facilities installed in rural and urban areas during 2000-2030.

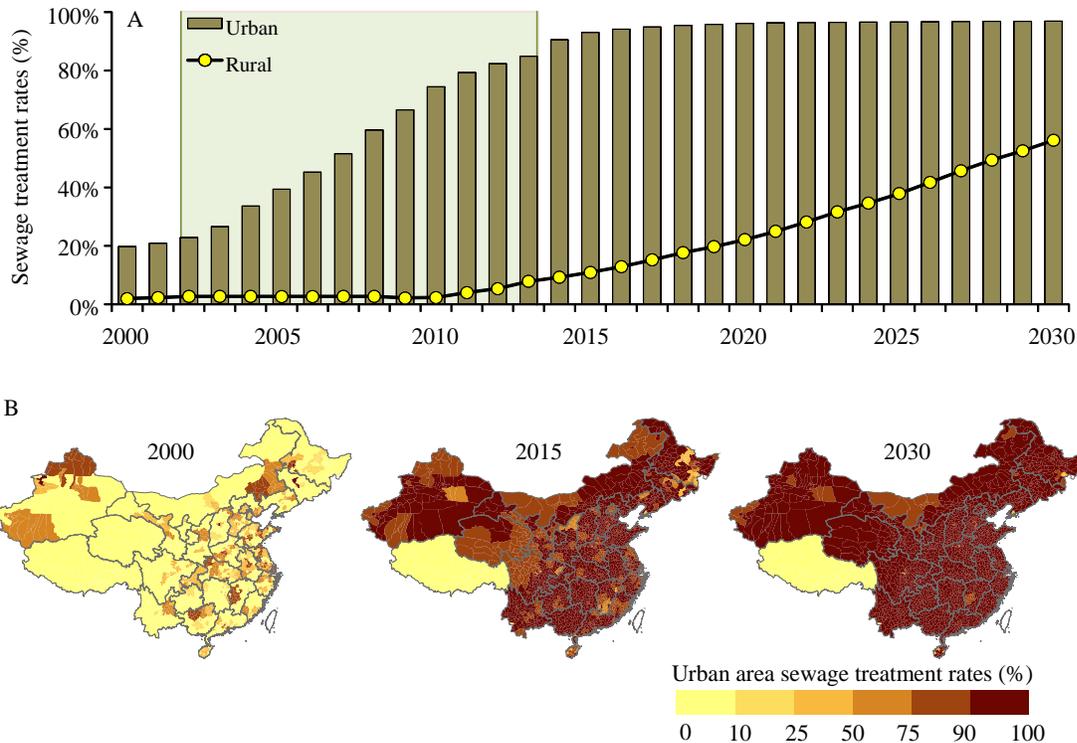


141
 142 Figure 1 Illustrative urban expansion information every five years for 2000-2030 in 2 regions;
 143 A. Jing-Jin-Ji: Beijing-Tianjin-Hebei; B. Yangtze River Delta (YRD). The different colours
 144 represent the urban population (for 2000) and the new urban population compared to that five
 145 years ago (for 2005-2030)

146 **2.3 Wastewater treatment connection rates**

147 The records for estimating the wastewater treatment connection rates, i.e. the proportion of
 148 population connected to wastewater treatment, could be found from the yearbook for 2002-
 149 2013 (covered by light green shade areas in Figure 2A) for urban areas and for 2008-2013 for
 150 rural areas for China (MHURD 2013). The wastewater treatment connection rate was estimated
 151 by total wastewater discharge volumes dividing those volumes treated for individual cities. The
 152 average of city level wastewater treatment connection rates for urban areas increased from 40%
 153 to 89% between 2002-2013; the average of provincial level wastewater treatment connection
 154 rates for rural areas increased from 2.5% to 6.4% between 2008-2013 (Figure 2A) (MHURD
 155 2013). Nationally, urban wastewater treatment plants (WWTPs) developed most rapidly during
 156 2003-2009 with the total national treatment capacity increasing from ca. 0.3 to 1 billion m³
 157 water/day (SI Figure S1) (MEP 2015b). After 2009, the rate of increase decelerated, but the
 158 national investment in WWTP construction was stable during 2009-2013 (MHURD 2013).
 159 Therefore future treatment rates during 2014-2030 were assumed to increase at the same annual
 160 rate of change as that during 2009-2013 for urban areas for individual city and the same as that
 161 during 2008-2013 for rural areas for individual province. Collected historical and predicted
 162 future wastewater treatment connection rates for urban and rural areas were used in all five
 163 economic scenarios for estimating the proportions of domestic sewage treated by wastewater
 164 treatment infrastructures for each county. More details see SI.

165



166

167 Figure 2 A. collected (2002-2013, covered by light green shade area) and predicted national

168 average urban and rural wastewater treatment connection rates during 2000-2030; B. the

169 distribution of wastewater treatment connection rates in urban areas at city level across China

170 for 2000, 2015, 2030

171 **2.4 Releases of ingredient to the environment**

172 As stated above, the use of triclosan is mostly in HPCPs, which is then released with wastewater
 173 to the environment directly or via WWTPs with wastewater effluent to the aquatic environment.

174 The possible release to soil by wastewater irrigation and sludge application regionally was not
 175 considered in this study due to the lack of information. It is the same assumption made in the
 176 previous study by Zhu et al. (Zhu et al. 2016). National triclosan usage was spatially allocated
 177 to counties across China by population for each year during 2000-2030 with assumptions that
 178 the usage per capita was spatially constant across China for individual years. The usage by
 179 respective urban and rural populations was then estimated by urban and rural population for
 180 each county. The wastewater generated by respective urban and rural populations was linked
 181 to WWTPs proportionally as the treatment rates estimated above.

182 The measured removal efficiency of triclosan in urban WWTPs has been reported to vary
 183 between 35 - 98% as a result of different loading mass, treatment technologies, operation

184 conditions, sampling seasons and methods etc. (Aguera et al. 2003; Bendz et al. 2005; Bester
185 2003; Heidler and Halden 2008; Lozano et al. 2013; Ying and Kookana 2007). In rural areas
186 in China, decentralized sanitation systems (household or neighbourhood scale) or centralized
187 wastewater treatment facilities (village level) are used for different conditions (Godavitarne et
188 al. 2011; Qiang et al. 2013). Many studies show that the removal efficiencies of triclosan by
189 constructed wetlands used in rural areas could range 70-100% (Zhao et al. 2016), but the
190 removal efficiency by other facilities/systems used for rural areas in China has not been
191 reported. Due to a lack of information on spatial distribution of different urban WWTPs and
192 rural facilities as well as low wastewater treatment connection rates in rural areas, a removal
193 efficiency of 95% predicted by SimpleTreat 3.2 (Franco et al. 2013) to represent processes in
194 secondary activated sludge plants (most common technology in China (Jin et al. 2014)) was
195 applied across China to estimate triclosan removal in wastewater treatment in this study. The
196 annual emission of triclosan for counties was estimated by combining the urban and rural usage,
197 wastewater treatment connection rates and the ingredient removal efficiency in wastewater
198 treatment (Eq. 2 in SI) for the five economic scenarios.

199 **2.5 Generation of concentrations**

200 Triclosan concentrations in surface waters across China were modelled using the Sino
201 Evaluative Simplebox-MAMI Model (SESAMe) v3.3 (Zhu et al. 2016; Zhu et al. 2015), 50km
202 grid multimedia chemical fate model taking into account of chemical ionization, for 2000-2030
203 under the five economic scenarios, combined with different surface water discharge flow
204 scenarios. The model configuration with input parameters and the chemical properties are
205 detailed in the SI. Table S3 shows the amount of freshwater applied on agricultural irrigation
206 for each province or municipality. The model has been validated on estimating triclosan
207 concentrations in China in a 2012-year scenario and performed well in a previous study (Zhu
208 et al. 2016). The thirty years' emission by counties estimated in the last step was interpolated
209 by population to fit the 50 km grid for the model by ArcGIS 10.4. Future discharge flows
210 predicted by Global Water Availability Assessment model (GWAVA) (Meigh et al. 1999) were
211 acquired under two IPCC marker scenarios (A2 and B1), and the two marker scenarios were
212 modelled by a general circulation model, ECHAM5 model (WATCH 2011). In this study, the
213 A2 discharge flow was assigned to the two 'A' economic scenarios and the B1 discharge flow
214 was assigned to the two 'B' and the OECD economic scenarios as environmental variables in
215 SESAMe v3.3 for 2011-2030. The estimated historical average discharge flow used in a
216 previous study (Zhu et al. 2016) was assigned for 2000-2010. To reflect the influence of the

217 discharge dilution on the water concentration, the historical average discharge flow was again
218 applied all through 2000-2030 under the five economic scenarios for comparison. This is called
219 the ‘constant discharge flow’ below to differentiate with the A2 and B1 discharge flows. The
220 change in other environmental variables through 2000-2030 was not considered, as their
221 influence to the surface water concentration of triclosan was limited (Zhu et al. 2014).

222 **2.6 Ranking the importance of drivers to chemical concentration changes in surface** 223 **water**

224 A variance-based sensitivity analysis, Sobol’s global sensitivity analysis (Saltelli et al. 2008;
225 Zhu et al. 2014) was applied to capture the influence of the full range of spatial and temporal
226 variation of the main drivers to the surface water concentration of triclosan for the whole
227 country. The total-order index was calculated to rank the importance of drivers, which is the
228 total sensitivity of the individual driver contributed by the single driver and its interaction with
229 other drivers (joint effects). A larger index indicates greater importance (see [SI](#)).

230 The six main drivers were: population; urbanization rates; wastewater treatment connection
231 rates for rural and urban areas, respectively; per capita GDP and surface water discharge flows
232 (dilution). For the other environmental parameters, the surface water concentrations are not
233 very sensitive to them and their temporal change is not available yet (Zhu et al. 2014). The
234 sensitivity analysis was conducted twice, for the periods 2000-2010 and 2010-2030
235 respectively, under only the OECD economic scenario, as the changing rate of these drivers
236 may be different in the two periods. For example, urban wastewater treatment connection rates
237 will have grown faster in the first ten years than in the later twenty years, while development
238 of rural wastewater treatment facilities have been faster after 2010 than before (Figure [2A](#)) and
239 is expected to grow in such quicker speed due to a currently stable investment and focus by the
240 government (MHURD 2013). Values of parameters for the grid cells in the SESAMe v3.3
241 model across mainland China for the first ten years and the later twenty years made two original
242 datasets for the six drivers. The Latin Hypercube sampling method (Zhang and Pinder 2003)
243 was used to take 10,000 values from the original datasets for each driver to compose random
244 datasets for the sensitivity analysis. Such sampling method could make sure that the random
245 values cover the full range of parameter values more evenly. For more details on this sensitivity
246 analysis method see the section S5 in [SI](#).

247 **3. Results and discussion**

248 **3.1 Population and urbanization trends**

249 From population projections, China's total population increased by ca. 5% in ten years from
250 2000 (1.26 billion) to 2010 and is predicted to increase by ca. 6% in twenty years from 2010 to
251 2030 (1.41 billion). What is more dramatic is the change in urbanization rates. We estimate this
252 to be as follows nationally: 2000, 31.6%; 2010, 48.5%; 2020, 48.2%; 2030, 74.7% (SI Table
253 S4) - a huge shift over the study decades. Compared to the historical data (2000-2015) in
254 yearbooks (CNSTATS 2000-2015), our estimation is slightly lower, but the trends are very
255 clear (SI Figure S4). Differences between estimates are probably influenced by the assumptions
256 for the threshold-value method used for urban population density, i.e. one identical value for
257 the whole country and the choice of threshold values. The urbanization rate by 2030 predicted
258 by this study is close to the projection by the World Bank and the Development Research
259 Center of the State Council, China (70%) (Guthrie 2007). From this estimate, the annual growth
260 rate of the urban population is ~1.9% from 2015 to 2030.

261 **3.2 Wastewater treatment connection rates**

262 China has made huge investments in urban wastewater treatment infrastructure over recent
263 years. After 2000, the national average urban wastewater treatment connection rate grew fastest
264 during 2003-2010, increasing about 2.8 times in just 7 years; and projections in this study
265 suggest slower but substantial increases during 2010-2030 (Figure 2 and SI Figure S1 and S5).
266 At the beginning of the study period, there was large regional variation for urban areas, but this
267 is reducing as modernisation continues across China (Figure 2B and SI Figure S5). For example,
268 by 2010 the provincial average urban wastewater treatment connection rate was <50% in
269 Heilongjiang and Qinghai; but in contrast, it was >85% in Chongqing, Tianjin, Hebei, Jiangsu,
270 Anhui and Shandong. However, by 2015, the lowest provincial urban wastewater treatment
271 connection rate had already reached about 80%. In the outline of '13th Five-year Plan', the
272 urban wastewater treatment connection rates in cities and county towns (Xian Cheng) are
273 expected to reach 95% and 85% respectively by 2020 (CPGC 2016). From projections by this
274 study, the national urban wastewater treatment connection rate will be $96 \pm 17\%$ (mean \pm STD
275 (standard deviation)) by 2020 (SI Table S5). This is an average of cities and county towns
276 without weighting by population. Almost all provinces will be close to or beyond the national
277 target of 95% by 2020 except for Hainan province (90%) predicted by this study.

278 We could not find any records of the extent of rural wastewater treatment before 2008, so
279 assumed a low and relatively stable wastewater treatment connection rate (1.9-2.7%) for 2000-

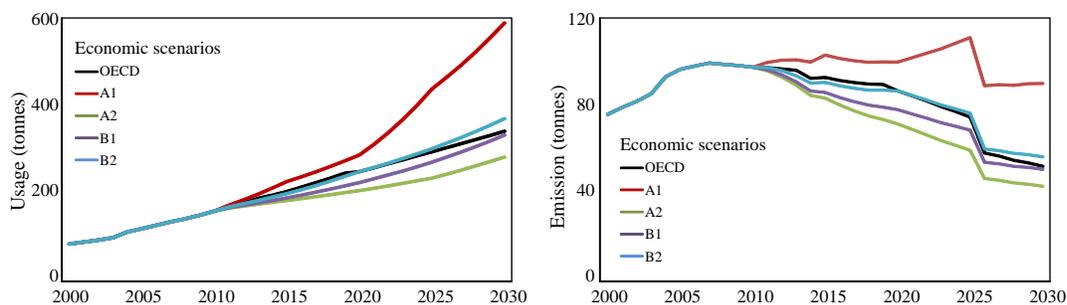
280 2007 for rural areas. After 2010, the national average rural wastewater treatment grows faster
281 and is projected to grow about 24 times during 2010-2030 (Figure 2A). Provincial level
282 variation is low and relatively stable before 2010 but estimated to increase thereafter (SI Table
283 S5). For example, during 2015-2030 rural wastewater treatment facilities are projected to
284 develop much faster in Sichuan, Chongqing, Shanxi, Qinghai, Shaanxi, Ningxia and some
285 coastal provinces than other regions, especially Inner Mongolia and Jiangxi, than in other
286 provinces (SI Figure S6). The Chinese government plans to promote rural wastewater treatment
287 infrastructures gradually (CPGC 2016) but there has not been a specific numerical target such
288 as that for urban areas. However half of the Chinese population are living in rural areas, and
289 considering that in rural villages wastewater produced is normally 60-80% of the water usage
290 and closely linked to the water usage within the same village (Godavitarne et al. 2011), it is
291 crucial to develop infrastructures for wastewater treatment in rural areas.

292 3.3 Projected usage and emissions under five economic scenarios

293 The increase in estimated usage follows that of per capita GDP for the five economic scenarios
294 during 2010-2030 (Figure 3A and SI Figure S3). Usage is predicted to grow relatively
295 moderately under the OECD, B1 and B2 economic scenarios, more sharply after 2020 in the
296 A1 economic scenario and relatively slowly in the A2 economic scenario. Triclosan usage in
297 2000 and 2010 is estimated to be ca. 86 and 160 tonnes, respectively. Under the OECD
298 economic scenario, its usage is estimated to increase about twofold over the twenty years, to
299 reach ~340 tonnes in 2030. Under the A1 economic scenario, usage would increase ~3.7 times
300 during 2010-2030, to reach ~590 tonnes p.a. The relative projected emissions for the five
301 economic scenarios is similar to that for the usage, i.e. emissions are the highest and the lowest
302 respectively under the A1 and A2 economic scenarios, and moderate in the other three
303 economic scenarios. However, the change of emissions with time under the five economic
304 scenarios is more complex than the usage (Figure 3B) due to being driven by the combination
305 of several factors and variation in data sources for different points in time. The detailed annual
306 usage is in SI Table S6.

307 The percentage of total triclosan mass removed by all wastewater treatment infrastructures in
308 China is projected to increase from 11% to 85% during 2000-2030 (SI Figure S7) as a result of
309 urbanization and the development of wastewater treatment infrastructures all over China.
310 National total emissions are projected to be highest in 2007 (100 tonnes) and decrease from ca.
311 76 to 43-57 tonnes in 2030 under four economic scenarios except A1 (SI Table S7). Under A1

312 economic scenario, triclosan emissions remain relatively high and reach the peak in 2025 (110
 313 tonnes), with the projected increased usage after 2020 being somewhat offset by increased
 314 removal with urbanization and infrastructure developments. There is an apparent sharper
 315 change in emissions in 2025 than in other years in the study period (Figure 3B). This is an
 316 artefact of the approach, because we had to use different population data sources for 2020 and
 317 2030, as explained above.



318
 319 **Figure 3** Projected annual total usage (A) and emissions (B) of triclosan in mainland China
 320 from 2000 to 2030 under five economic scenarios

321 **SI Figures S8-S9** show spatial distributions of triclosan usage and emissions at the county scale
 322 for every five years from 2000 to 2030 under the OECD economic scenario. The distribution
 323 of usage is determined by population distribution as stated above. Therefore, most usage over
 324 the whole 2000-2030 period is into eastern or southeast China below the Heihe-Tenchong Line,
 325 a geo-demographic demarcation line that divides the area of China roughly into two equal parts
 326 with about 94% of the Chinese population in the west (**SI Figure S8** for 2000). This covers
 327 almost half of China's land mass area, but is occupied by ~94% of China's population
 328 (surveyed for 2002) (Guthrie 2007). In the west, the usage in some counties in Xinjiang is
 329 estimated to be high. Lowest usage is estimated to be in Tibet. Generally, the spatial distribution
 330 of emissions is very similar to that of usage for 2000-2010. However, after 2015 emissions are
 331 projected to decline steadily in Beijing, southern Hebei, Chongqing, eastern Sichuan, eastern
 332 coastal cities or provinces such as Shanghai, Zhejiang and Guangdong. Until 2030, emissions
 333 in mid-Heilongjiang, south Jilin, east Liaoning, most areas of Hebei, Beijing, Tianjin, Shanxi,
 334 Shandong and most areas in south China are predicted to be rather low and below 0.008 tonnes
 335 per annum in each county. Most counties in Inner Mongolia and some counties in Xinjiang,
 336 eastern Henan and northern Anhui are still estimated to have high emissions. This is because
 337 of low urbanization rates and low rural wastewater treatment connection rates projected for
 338 these areas in 2030.

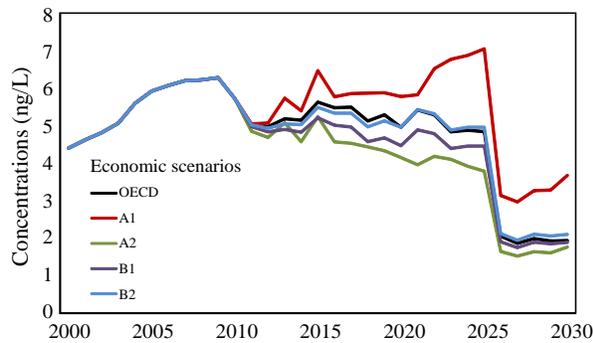
339 3.4 Projected concentrations

340 Triclosan concentrations in surface water are predicted by the SESAMe v3.3 model with ~50
341 km spatial resolution. Figure 4 and SI Figure S10 shows temporal change of the national
342 median concentrations for the 50 km grid cells as a result of the combination of different
343 economic and discharge scenarios. During 2011-2030, the median concentration is estimated
344 to be highest under economic scenario A1 and lowest under A2, and moderate in B1, B2 and
345 OECD economic scenarios. The pattern is similar to that of usage and emissions (Figure 3).
346 When applying different discharge flow scenarios for 2011-2030, the A2 and B1 discharge
347 flows possibly would dilute the chemical in surface water more during 2011-2025 but less
348 during 2026-2030 compared to the constant discharge flow. SI Figure S11-S13 show the
349 histogram of predicted triclosan concentrations in surface water during 2000-2030 under
350 different scenarios. Figure S12-S13 indicates that a wider range of triclosan concentrations
351 across China would be estimated by using the constant discharge flow rather than using A2 and
352 B1 discharge flows under all five economic scenarios.

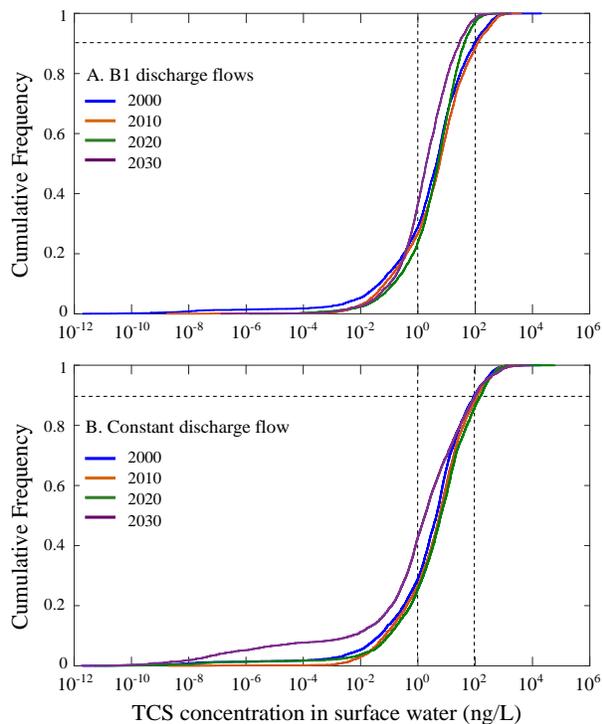
353 When applying A2 and B1 discharge flows for 2011-2030, the highest median concentration
354 of triclosan is estimated to be in 2009 (ca. 6.3 ng/L) under OECD, A2, B1 and B2 economic
355 scenarios, and in 2025 (ca. 7.0 ng/L) under A1 economic scenario (Figure 4). The median
356 concentration generally declines after 2009 in all economic scenarios except A1. When
357 applying the constant discharge flow for 2011-2030, the highest median concentration of
358 triclosan is estimated to be in 2016 under OECD (~6.7 ng/L) and B2 (~6.6 ng/L) economic
359 scenarios, in 2025 under the A1 (~8.2 ng/L) economic scenario, and in 2009 under the A2 and
360 B1 (~6.3 ng/L) economic scenarios (Figure S10). The estimated median concentration has a
361 sharp decline in 2025, which is the same as the emission due to the same reason discussed
362 above. It has declined to ca. 3-3.5 ng/L for A1 economic scenario and ca. 1.5-2 ng/L for all
363 other economic scenarios.

364 Figure 5 shows the temporal evolution pattern of triclosan concentration frequency in water
365 with different discharge scenarios under the OECD economic scenario. Generally, regions with
366 relatively high concentrations increase after 2000 and then decrease in 2020 and 2030. Regional
367 concentrations are more scattered when applying constant discharge flow (Figure 5B) than
368 applying B1 discharge flow (Figure 5A), as more regions having concentrations <1 ng/L
369 or >100 ng/L are estimated when applying constant discharge flow in the model prediction.
370 100 ng/L is the current UK maximum guideline value for total triclosan in surface water

371 (UKTAG 2015). Proportion of areas with estimated concentrations > 100 ng/L is about 10% in
 372 the four years using the constant discharge flow (Figure 5B), but decreases to only ~2% when
 373 using the B1 discharge flow in 2020 and 2030 (Figure 5A).



374
 375 **Figure 4** Predicted national median concentrations from 2000-2030 under five economic
 376 scenarios; the estimated constant discharge flow for 2000-2010 is used for 2000-2010, while
 377 for 2011-2030 the A2 discharge flow is used in A1 and A2 economic scenarios and B1
 378 discharge flow is used in B1, B2 and OECD economic scenarios. Note: the estimated median
 379 concentration has an apparent sharp decline in 2025, as noted earlier for the emission projects.



380
 381 **Figure 5** The cumulative frequency of triclosan concentrations in surface water under the
 382 OECD economic scenario for 2000, 2010, 2020 and 2030. The constant discharge flow is used

383 for 2000 and 2010 in both figures. A. B1 discharge scenario used for 2020 and 2030; B. the
384 constant discharge flow used for 2020 and 2030.

385 SI Figures S14-S15 show the spatial distribution of concentrations estimated under the OECD
386 economic scenario using the B1 discharge flow and the constant discharge flow. When
387 applying the constant discharge flow, estimated concentrations in most regions in Shanxi,
388 North China Plain (NCP), mid-Inner Mongolia and part of the three provinces in the northeast
389 are relatively high all through 2000-2030, due to the low surface water dilution. Estimated
390 concentrations in South China are lower than above regions and decline steadily from 2015 to
391 2030 (SI Figure S15). Such spatial distribution patterns should also be obtained under other
392 economic scenarios, as the constant discharge flow is applied identically. When applying the
393 B1 discharge flow for 2015-2030, estimated concentrations in above high concentration areas
394 are still higher than the other areas, but decline significantly after 2015. Compared to the extent
395 of national median concentration change during the study period, i.e. about threefold (Figure
396 4), the extent of the regional concentration change could reach over 5000 times as much during
397 the period in Zhada, Tibet. The average concentration of triclosan in surface water for 15% of
398 counties across China has changed over 50 times as much over the thirty years.

399 The results on ingredient usage, emissions and concentrations are illustrated and discussed for
400 triclosan. Different temporal patterns for different HPCP ingredients may occur, but the
401 methodology for estimations is adapted.

402 **3.5 Contrasting the spatial distribution of usage, emissions and concentrations**

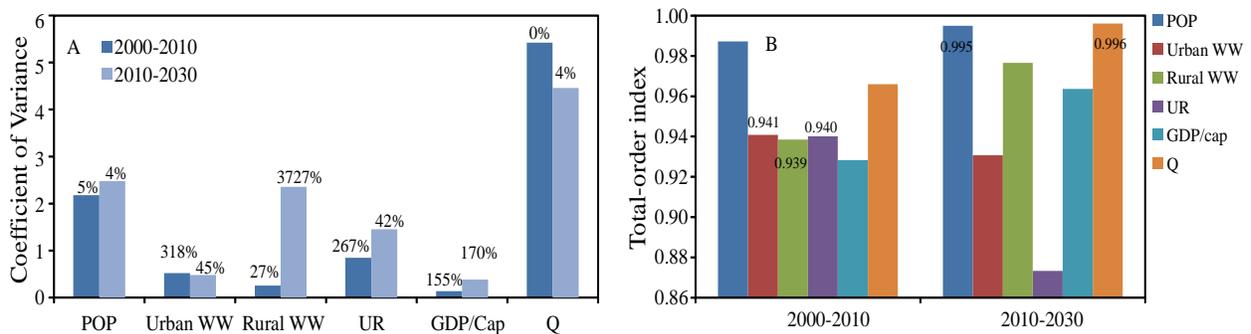
403 Using the B1 discharge flow here for discussion, the spatial distributions of projected usage,
404 emissions and concentrations show geographic contrasts, because of the influence of regional
405 urbanization rates, wastewater treatment connection rates and surface water dilution. For
406 example, the estimated highest usage is in urban Dongguan, Guangdong province from 2000-
407 2025 and in Tailai, Heilongjiang from 2026 to 2030. However, the estimated highest emission
408 is in counties in Guangdong (Dongguan or Puning) during 2000-2010, in Yunnan (Xuanwei)
409 during 2011-2025 and in Henan in 2030. Meanwhile, the estimated highest concentrations are
410 in Shanxi (Yingze) during 2000-2010, but in Hebei (Shanhaiguan) during 2011-2030.

411 The estimated top five counties for the highest usage change from mostly in Guangdong to
412 Beijing (Chaoyang and Haidian districts) and Shanghai (Pudong New Area), and then to
413 Chongqing (Yubei, Yongchuan and Jiulongpo districts) over the 30 years. In contrast, the top

414 five counties that have the highest emission across China change from mostly in Guangdong
 415 until 2010 to counties mainly in the southwest, such as in Guangxi (Guiping and Bobai),
 416 Guizhou (Weining Yi, Hui and Miao autonomous county) and Yunnan (Xuanwei and
 417 Zhenxiong) until 2025, and change to counties all in Henan in 2026-2030. The estimated top
 418 five counties that have the highest concentration are mostly in Shanxi from 2000-2010 and
 419 change to mostly in Hebei from 2015-2030. SI Table S8 shows the top five counties with the
 420 estimated highest usage, emission and concentration for 2000, 2010, 2020 and 2030 for
 421 comparison.

422 SI Figure S16 illustrates the temporal change of average wastewater treatment connection rates
 423 for 11 provinces/cities over 2000-2030. As it is urbanization rates and population weighted
 424 (See SI and SI Eq. 3), it provides direct reasons for the contrast between usage and emissions
 425 as illustrated above. Beijing and Shanghai have relatively high estimated wastewater treatment
 426 connection rates throughout the 30 years and therefore do not have the highest emission in
 427 terms of the highest usage during the same period. Wastewater treatment connection rates in
 428 Yunnan, Guangxi, Guizhou and Heilongjiang are estimated to be much lower and grow slowly
 429 compared to other provinces during 2000-2025, but increase sharply to different extent after
 430 2025. Therefore emissions are estimated to be the highest in some counties in these provinces
 431 during 2010-2025. The average wastewater treatment connection rates in Henan are estimated
 432 to decline after 2025 and be lower than that in Heilongjiang, so emissions in counties there turn
 433 to the highest in 2030. The contrast between the spatial distribution of emissions and
 434 concentrations is a result of the effect from surface water discharge flow.

435 3.6 Ranking of different drivers



436
 437 **Figure 6** A. coefficient of variation of values for 50km grid of the six drivers across China
 438 during 2000-2010 and 2010-2030; the values above the bars are percentages of national average
 439 driver changes from the start to the end year for each period. B. total-order index from the

440 sobol's global sensitivity analysis for respective 2000-2010 and 2010-2030. POP, population;
441 Urban/Rural WW, urban/rural wastewater treatment connection rates; UR, urbanization rate;
442 GDP/Cap, per capita GDP (Gross Domestic Product); Q, surface water discharge flow. The
443 projection is under OECD (Organisation for Economic Co-operation) economic scenario for
444 2000-2030 and constant discharge flow for 2000-2010, B1 discharge flow for 2011-2030. To
445 explicit about their relative importance, the exact values of total-order index are added in figure
446 B for several parameters for which the values are very close to each other.

447 The coefficient of variation (CV) describes the amount of variability relative to the mean
448 (STD/mean). In Figure 6A, CV (bars in the figure) considers both the spatial variation (5469
449 grid cells across mainland China) and the temporal (annually during 2000-2010 and 2010-2030)
450 change of all projected drivers during the two periods. The percentage values (%) above the
451 bars reflect the temporal variation only at national average level. The CV of discharge flows
452 (highest bars marked with Q) and population is mainly contributed by the spatial variation due
453 to the extremely low % values above the bars, and that of per capita GDP is contributed only
454 by temporal change. Discharge flow has the highest variation for the two periods. For the other
455 five drivers, the rank of the variation is in order of population, urbanization rates, urban
456 wastewater treatment connection rates, rural wastewater treatment connection rates and per
457 capita GDP for 2000-2010; and population, rural wastewater treatment connection rates,
458 urbanization rates, urban wastewater treatment connection rates and per capita GDP for 2010-
459 2030. Comparing the variation in the two periods, only projected urban wastewater treatment
460 connection rates and discharge flows have higher variation in 2000-2010 than in 2010-2030.
461 The other four projected drivers all have higher variation in 2010-2030 especially for rural
462 wastewater treatment connection rates. It reveals that the development of urban WWTPs during
463 2010-2030 is estimated to be moderated and approach complete in a national wide compared
464 to the previous ten years; whilst the other four drivers are estimated to have more dramatic
465 development (e.g. rural wastewater treatment facilities and per capita GDP) or spatial variation
466 (e.g. population distribution and urbanization rates) in 2010-2030 than previous years.

467 The total-order index of the global sensitivity analysis in this study (Figure 6B) reflects the
468 relative importance of changes of the six drivers to triclosan concentrations in surface water
469 over the study periods across China taking account of interactions between drivers. The
470 regional population and discharge flow are projected to be the most important drivers to
471 changes in triclosan concentrations for both periods, as the regional population determines the
472 spatial allocation of the usage and discharge flow affects the dilution directly. For the other

473 four drivers in Figure 6B, during 2000-2010, the development of urban wastewater
474 infrastructures and the urbanization progress are projected to be more important to triclosan
475 concentrations across China, followed by rural wastewater treatment infrastructure
476 development and per capita GDP growth in sequence. Contrarily, during 2010-2030, the
477 development of rural wastewater treatment infrastructure probably becomes the most important
478 among the four drivers to triclosan concentrations for China, followed by per capita GDP
479 growth, urban treatment infrastructure development and urbanization progress in sequence.
480 The difference of the estimated relative importance is more significant during 2010-2030 than
481 2000-2010 for the four socioeconomic drivers possibly due to the more significantly different
482 variation of the drivers during 2010-2030 (Figure 6A).

483 The rank of the importance of the drivers to triclosan concentrations does not exactly follow
484 that of variations of the drivers, as (1) it is a non-linear model and (2) changes of multiple
485 drivers will influence triclosan concentrations jointly (interactions of drivers as mentioned
486 above). This implies that it is not reasonable to compare the influence of several changing
487 drivers to surface water concentration of a chemical by simply comparing the extent of the
488 variation/change of drivers. However, if a driver develops more dramatically during a period
489 than any other time, e.g., rural wastewater treatment connection rates during 2010-2030, its
490 relative importance might increase during this period. The global sensitivity analysis was
491 conducted based on triclosan. However, as these main drivers are mostly relevant to sources of
492 the ingredient rather than chemical properties, the conclusion can be generalized to a range of
493 down-the-drain HPCP ingredients that have similar sources with triclosan.

494 **3.7 Uncertainties**

495 Uncertainties of the methodology in this study exist and are common for similar source HPCP
496 ingredients. The uncertainties of using the threshold-value method to identify the urban
497 population have been discussed above. Emissions might be slightly overestimated as a result
498 of following assumptions: (1) The allocation method is applied without differentiating the
499 purchasing power in urban and rural areas. In reality, per capita usage of HPCPs in urban areas
500 might be higher than that in rural areas due to the higher GDP levels in urban areas. Therefore
501 more HPCP ingredients should have been removed as a result of higher wastewater treatment
502 connection rates in urban areas than rural areas. (2) No bans or elimination are assumed in this
503 study till 2030 in China. However, formula change by replacement or reduction of ingredient
504 use is possible if there are restrictions by government or if it is phased out by industry. For

505 triclosan, there is an indication that industry are beginning to phase this ingredient out now. (3)
506 a high level ingredient inclusion level is assumed to be identical in all HPCP categories. In
507 some categories it is actually lower than this. (4) An identical removal efficiency for typical
508 secondary activated sludge plants is applied across China for all wastewater treatment
509 infrastructures. It might overestimate the removal in some rural areas where only primary
510 treatment facilities are used.

511 Slight underestimation of emissions might be caused by the following assumptions: (1) Only
512 sales data of HPCP ingredients that are released down to drain with domestic wastewater is
513 accounted for in this study. Many HPCP ingredients are at the same time contained in solid
514 materials, such as triclosan used in plastic and textiles, might be transported to aquatic system
515 with runoff after the products are disposed in land. However, it might be very slow release. (2)
516 Sludge application in soils is not considered in this study. (3) Possible release from processes
517 of manufacture of HPCPs is not considered. It might exist in some regions but be rather low
518 compared to those accounted in this study.

519 Other uncertainties may also exist. For example, (1) spatial allocation by population might
520 cause skewed spatial distribution of HPCP usage, as the usage might be more relevant to
521 regional GDP levels. (2) As explained above, the relatively sharp change of emissions and
522 national median concentrations in 2025 might be caused by different population data sources.
523 It also reflects uncertainties of future population distribution. The projections from the different
524 sources may be based on distinct assumptions, but both are possible. (3) The projection of
525 population and migration from sources cited in this study probably has not considered the end
526 of one-child policy and permission of a second child for a couple effective in 2016. Such
527 national policy or other economic policies in regional or national scale may all affect future
528 population count and distribution. (4) Per capita GDP might be more important than the
529 prediction in this study considering its spatial variation in reality.

530 **4. Conclusions and implications**

531 For the first time, a methodology is presented for assessing possible future water concentration
532 of contaminants for China under different economic scenarios and the key drivers, which will
533 be useful to policy-makers and stakeholders for mitigation of domestic effluents affecting water
534 quality. Here triclosan is only chosen as an example HPCP ingredient, and the predicted
535 concentrations in this study would be advisable to embark on monitoring work over following
536 a few years to confirm the reliability of this study. All other chemicals which mainly derive

537 from anthropogenic sources that are relevant to economic levels are suitable for this method.
538 HPCPs as trace organics can be marker chemicals for similar anthropogenic-source chemicals,
539 of which the usage is linked to GDP. This framework can provide a preliminary outlook of
540 surface water contamination levels of these chemicals and therefore provide information for
541 chemical management in China.

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544 **Appendix A. Supplementary data**

545 Supplementary data to this article can be found online.

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