The evolution of chemical risk in a Chinese river catchment exposed to untreated wastewater

Abstract

The risks posed by chemicals released with domestic wastewater can be reduced by treating wastewater prior to its release; but in developing countries such as China, many of its settlements lack the infrastructure to treat its domestic wastewater. Improving our understanding of the risk posed by these chemicals in catchments that are partially served by sewage treatment works would enable this risk to be better managed. The spatially explicit chemical risk assessment model, Geo-referenced Regional Exposure Assessment Tool for European Rivers (GREAT-ER), was used to determine how chemical risk varies across the East River catchment, South-East China, in response to the construction of new sewage treatment works and to population change between 2010-2020. We evaluated the risks from two naturally occurring estrogens, estrone and 17β-estradiol, as well as two personal care product ingredients, triclosan and triclocarban. Model predictions suggest that initially, chemical concentrations decrease substantially because of the rapid construction of new sewage treatment works throughout the catchment, but as the rate of construction slowed and population continued to increase, chemical risk increased, with concentrations for most chemicals increasing to levels greater than in 2010. We then explored the potential impact of treating 100% of domestic wastewater within the catchment, which we estimated would sufficiently reduce chemical risk for triclocarban and estrone. Subsequently, to try and reduce the risk posed by triclosan, we theoretically reduced the usage of triclosan until concentrations were reduced to levels below the predicted no effect concentration. The results from this analysis suggested that triclosan usage would need to be reduced by ~90% to reach safe concentrations in the East River catchment.

Keywords

Modelling; Chemical Risk; China; Wastewater; Spatial

1. Introduction

In 2015, the sale of chemicals in China was greater than the total sales in the European Union (EU) and the North America Trade Agreement (NAFTA) countries combined (The European Chemical Industry Council, 2016). This has led to concerns regarding the environmental impact of chemicals, particularly the release of so called “down-the-drain chemicals” to the environment in China (Price *et al.*, 2012). Down-the-drain chemicals are those which are primarily released with domestic wastewater. Examples of down-the-drain chemicals include ingredients within personal care products (PCPs), pharmaceutical ingredients, caffeine and estrogens. Some of these chemicals may be harmful to in-stream organisms, with negative effects on mortality, growth, reproduction and other biological endpoints (Brausch and Rand, 2011).

Many of China’s settlements, particularly smaller populations, lack a sewage treatment works (STW) or are only partially served by a STW (The State Council, 2012). In the case of rural settlements, collection of wastewater is difficult as rural population density is low and population within villages may be scattered (Yu *et al.*, 2015). Chemicals from domestic wastewater sources have been detected at relatively high concentrations in previous surveys, including home and personal care product ingredients (Zhang *et al.*, 2015b), pharmaceuticals (Zheng *et al.*, 2020) and steroid hormones (Huang *et al.*, 2019). Construction of sewage treatment plants should reduce the risk from down-the-drain chemicals in China by reducing the chemical load discharged to water bodies. But constructing all the necessary STWs will take time and as a result STW construction was initially focused on densely populated areas, with an increased focus on towns and small cities (National Development and Reform Commission, 2016). To manage the expansion of STWs across the country, a catchment manager would benefit from an estimate of the spatial distribution of risk posed by down-the-drain chemicals across the river catchment which could help support the prioritisation of new and upgraded STWs. For example, a better understanding on a catchment-by-catchment basis, would enable authorities to determine whether there should be more focus on mitigating the release of down-the drain chemicals from rural areas, or if the focus should remain on improving wastewater infrastructure in urban areas. Alternatively, it may be that a given chemical may be better managed by implementing controls at the source, by reducing the amount that is consumed.

For this study, the Geo-referenced Regional Exposure Assessment Tool for European Rivers (GREAT-ER) model, a catchment scale stochastic-deterministic chemical risk assessment model (Feijtel *et al.,* 1997) was used to estimate the concentration of two personal care product ingredients: triclosan and triclocarban and two estrogens: estrone and 17β-estradiol. The model was applied to the East River catchment, South China for

2010, 2015 and 2020. The model was simulated for the dry season only, during which dilution is at its lowest and concentrations are assumed to be at their highest.

We aimed to use the GREAT-ER model to determine how chemical risk changes over time in response to improvements to wastewater infrastructure and population change, but also to determine whether chemical risk could be mitigated by reducing chemical consumption and by treating previously untreated wastewater prior to release.

2. Materials and methods

2.1 Constituents of interest

Two chemicals commonly found in personal care products were selected: triclosan (TCS) and triclocarban (TCC). In addition, two natural steroidal estrogens were also selected: estrone (E1) and 17β-estradiol (E2). Natural estrogens are endocrine disruptors (Jobling *et al.*, 2002), whereas the primary threat from TCS and TCC is their toxicity to aquatic organisms (Halden, 2014). There have also been concerns that exposure to TCS and TCC may result in endocrine disruption and antibiotic drug resistance (Halden, 2014; Veldhoen *et al.*, 2006).

To evaluate risk, we obtained predicted no effect concentration (PNEC) data for our target chemicals, using the lowest threshold found within the wider literature. For TCC we used a PNEC value of 25 ng/l, which was based on acute toxicity for *D magna* in freshwater and a PNEC of 1.5 ng/l for TCS, based on acute toxicity for algae in freshwater (Musee, 2018). For E1 we applied a PNEC value of 3.6 ng/l based on chronic toxicity for *Danio rerio* and a PNEC value of 0.4 ng/l for E2 based on results from a species sensitivity distribution method used to determine chronic toxicity in water (Čelić *et al.*, 2020).

2.2 Study area

The East River is a major tributary of the Pearl River, South China. The catchment area is 25,325 km^2 (above Bolou gauging station), of which approximately 90% resides in Guangdong Province. The source of the East River is in the North-East of the catchment, in Xunwu county of Jiangxi province; the river then flows south-westerly to the Pearl River delta, where it discharges into the South China Sea. The catchment is subject to a sub-tropical climate which is dominated by a highly seasonal pattern of rainfall and flows. The East River's mean annual discharge is 763 m^3/s, which peaks in July with a mean monthly discharge of 1758 m^3/s and drops to an annual low in January with a mean monthly discharge of 377 m^3/s. Three large reservoirs are located in the upper reaches of the catchment, all of which have a considerable influence upon flow (see online supplementary material, Figure S1); the Xinfengjiang reservoir is the largest reservoir at a capacity of 13.98 billion m^3 (Zhou *et al.*, 2012). Population densities upstream of the reservoirs are low and reservoir residence times (and therefore degradation) were assumed to be high given the large capacities of the reservoirs and likely long residence times; as such the chemical concentration in flow discharging from the reservoirs were assumed to be negligible.

2.3 The GREAT-ER model

The GREAT-ER model (version 3) is a catchment-scale geographic information system (GIS) model. The model estimates exposure concentrations of a chosen chemical within the river network. All outputs and a significant number of inputs are distributions rather than single values (Wagner and Koormann, 2011); these parameters are used within a Monte Carlo component of the model to account for uncertainty and natural variation in the environment. Chemical emissions may be from point or diffuse sources and are calculated on a per capita emission basis (Wagner and Koormann, 2011). Removal of the contaminant can take place prior to emission and in-stream.

The GREAT-ER model has been used for a number of different substances and purposes, although the model's use has been largely restricted to European catchments. Early use of the model focused on household cleaning product ingredients such as Boron and Linear Alkylbenzenesulfonate surfactants. The GREAT-ER model (and its daughter products) has also been used to investigate the fate of active pharmaceutical ingredients (Duarte *et al.,* 2022), personal care products ingredients (Price *et al.*, 2010) and estrogens (Williams *et al.*, 2012). The GREAT-ER model has also been successfully parameterised to simulate nonylphenol and nonylphenol ethoxylates within the Wenyu catchment, an urban catchment in North-East Beijing (Zhang *et al.*, 2015a).

2.4 Modelling approach

To estimate the change in chemical risk overtime, we generated 2010, 2015 and 2020 versions of the East

River catchment for the GREAT-ER model. The model was configured to represent the population and wastewater infrastructure for each represented year, whereas all other aspects of the catchment were assumed to remain the same.

2.4.1 Model parameterisation

The GREAT-ER model requires data that describes chemical properties, chemical usage and catchment data. Data describing chemical properties and removal in-stream and under wastewater treatment were collated from the wider literature (see online supplementary material, Table S6) and where possible a range of possible parameter values was obtained and input into the model as a statistical distribution. For TCS and TCC, limited in-stream removal data was available so the lowest value was selected, but for E1 and E2 sufficient data was available to define the data as a probability distribution. The GREAT-ER model is unable to natively model in-stream removal as a distribution, but the model code, which is written in Python, was modified in order to do so. The Python code was also modified to accommodate different types of probability distributions such as the beta distribution.

Modelling was generally hindered by a lack of high-quality data for the East River catchment, particularly data regarding wastewater emissions. As a result, much of these data had to be estimated, as described by the approach in this section. To verify the model, sampling data were obtained in December 2008 by collaborators from the Guangzhou Institute of Geochemistry (Zhao *et al.*, 2013) and samples were collected by the first author in January 2016, as part of this current study; these data were used to evaluate the 2010 and 2015 model respectively. For the 2016 sampling campaign, a total of 36 sites were sampled, focusing on the densely populated area of Huizhou and Shenzhen, the urban and rural area around the city of Heyuan and the rural region of Longchuan in the North of the catchment. Samples were collected upstream and downstream of STWs where possible. For a full description of the sampling and analytical method used see online supplementary material are described in section S2 (see online supplementary material) with the supporting information and a map of sampling sites is shown within Figure S1 (see online supplementary material).

An estimate for usage (or excretion rate) for all chemicals was calculated based on the concentration of Chinese STW influent concentration, flow and known STW served data. This is based upon the approach of Liu *et al.* (2015) and was determined by acquiring data about STW population served, STW discharge rate and measured concentrations within influent. Usage was then calculated using Equation 1 below:

$$\mathrm{E} = \frac{\mathrm{D}}{\mathrm{P}} * \mathrm{C} \tag{1}$$

where E is the estimated usage/excretion rate per capita per day (kg/cap/year), D is the STW average discharge rate per year (m^3/year), P is the STW population served estimate and C is the measured chemical concentration in effluent (kg/L).

Chinese data were used exclusively for the estimation of estrogen usage, as it has been suggested that the excretion of estrogen varies significantly between western and Chinese populations (Adlercreutz *et al.*, 1994). Usage was estimated for TCC and TCS based on data obtained from STWs in the Pearl River delta (Liu *et al.*, 2017), which may partially account for regional differences in usage (Hodges *et al.*, 2012).

2.4.2 Estimating the spatial distribution of wastewater discharge

A list of STWs operational in China is made available annually from the Chinese government, which has been developed in two different formats. The first format was published on the website of the then named Ministry of Environmental Protection, which provided a list of the names of STWs in each city region, with details regarding the treatment type, the treatment capacity and the average volume of wastewater treated each day. The population served, service area and STW locations are not provided, although the name of the STW may sometimes indicate its approximate location. This list was distributed up until 2014 and is now produced in a different format by the Ministry of Ecology and Environment. The new list provides more information about the location of each STW including the province, city, district and the town or street, but the list does not appear to contain information about STWs that had been built prior to 2014. This information is then used to locate, then identify STWs, with more details regarding the approach used described in section S4 (see online supplementary material) and section S5 (see online supplementary material).

The boundary for the area served by a STW, here described as the wastewatershed, was approximated for each STW. Creation of a wastewatershed enabled an estimate of the population served, which was assumed to equal the population within its boundary and population outside wastewatersheds was assumed to release wastewater to watercourses untreated. It is assumed that a fraction of untreated wastewater will not reach the waterways due to imperfect drainage and degradation. This is incorporated into GREAT-ER by assigning a STW to every reach that was estimated to receive untreated wastewater with a uniform removal rate between 0 to 100%.

To simplify the process of creating a wastewatershed and to be able to generate the necessary spatial data required by GREAT-ER, we generated river reaches and associated reach subcatchments; each subcatchment contained the area that drained to the reach but excluded the catchment that drained to upstream reaches. The river reaches and subcatchments were generated using the stream reach and watershed tool within the Terrain Analysis Using Digital Elevation Models (TauDEM) GIS toolbox (Tarboton *et al.*, 2015), using the 1-arc second (~30m at the equator) digital terrain model, Shuttle Radar Topography Mission (SRTM) v3 (Farr *et al.,* 2007). We then extracted the mean population density to each subcatchment from gridded population data. As it was important to use reliable population data, we used the Worldpop (Gaughan *et al.*, 2013) population datasets for 2010, 2015 and 2020, as well as population projections for each city region of the catchment. This process is described in more detail within section S3 (see online supplementary material).

The wastewatershed delineation process was completed manually. For settlements with a single STW, the outlines of settlements adjacent to each STW were digitised in Google Earth (Google, 2024) and subcatchments that intersected the digitised settlement outline were assumed to be served by the STW, as well as subcatchments that directly drain to river reaches that were located between the STW and the settlement it served.

Wastewatershed delineation within large cities (e.g. Huizhou, Shenzhen and Heyuan) required more precision and additional data. Where multiple STWs serve the same urban area, it is challenging to determine the extent of each wastewatershed as it is often unclear as to which regions of the city are served by each STW (e.g. see online supplementary material, Figure S3). In addition, a proportion of each city might not be served by a STW, so this must be accounted for. Data regarding population served or the service area of STWs within the area were available for some of the STWs within these areas, which was obtained from STW managers or from online reports and news articles (see online supplementary material, Table S4). We expect that these sources may not be entirely reliable but were used due to a lack of any alternative sources of information. The service area of a STW, which describes which regions of the city it serves and/or the total area that it served, was particularly useful to guide the delineation. Where information regarding a STW's service area was available as well as the population served by the STW, the wastewatershed was delineated so that it covered a similar area as described by the service area information, but also so that the underlying population was similar to the population served. Wastewatersheds for STWs with the most information describing population served and service area were usually delineated first, with those without any such data delineated last. All STWs without population served or service area data were delineated by assuming the urban area upstream of the STW was served by this STW, excluding area served by STWs already delineated (e.g. see online supplementary material, Figure S4).

The accuracy of the STW population served estimates and the area served by each STW is uncertain as actual population served data was limited. As a result, the accuracy of the approach used to estimate population served described in this section was tested by applying it to a selection of STWs in South-West England, as data availability is much higher in the region. To do so we utilised the WorldPop dataset for the year 2014. Spatial data were available from European Environment Agency (2023) for STWs in the region, along with estimates of the population served by each STW. Population served estimates were provided in the form of population equivalents, which were calculated by the European Environment Agency based on the biological oxygen demand (BOD) measured in wastewater, assuming 54 g of BOD equalling one person (Organisation for Economic Co-operation and Development, 2001).

Otherwise, the same methodology applied to the East River catchment was applied to the catchments of South-West England. In total, population was estimated for 39 STWs for a variety of different population sizes.

2.4.3 Streamflow estimation

The GREAT-ER model accounts for the variability of flow based on the mean flow and the flow that is exceeded 95% of the time (Q95). This is based on the flow duration curve, which plots discharge against the percentage of time that flow will be exceeded. The flow duration curve may be considered a frequency curve if it is assumed that each data point within a time series for a catchment is independent of one another (Vogel and Fennessey, 1994). This is an approximation, as adjacent data points within a time series will also show strong correlation. Assuming elements within a time series are independent is less appropriate in catchments which experience a strong seasonal variation in rainfall, which includes the monsoonal East River catchment. Calculating low-flow statistics for each month separately, it can be argued, is more appropriate than calculating annual statistics as the influence of within year correlation is reduced within the analysis. As such, we separately estimated statistics for January and December, the months in which sampling took place, for the purpose of validation. For analysing chemical risk overtime, we compared the results for January flows, as it is the driest month on average and therefore should be subject to the highest level of risk.

Daily flow data were manually extracted from physical copies of hydrological yearbooks 2006-2013 for 11 gauges, 5 of which are downstream of one or more major reservoir and 6 which are considered to have limited artificial influence. This study focuses on January and December, the months in which sampling took place. Flow statistics were calculated for flow occurring within December and January for 2006-2013; the median Q95 and mean flow was calculated to determine the long-term average.

The approach used to estimate flow was based on the drainage area ratio methodology (Vandewiele and Elias, 1995), which is primarily used to estimate flow at ungauged basins. The gauges that were sampled were those with no significant artificial influences on flow, which enabled the naturalised flow to be estimated. For every ungauged site, flow normalised by area observed at the nearest gauge was assumed to equal that of the ungauged site (Equation 2).

$$Q_{ug} = \frac{A_{ug}}{A_g} * Q_g \qquad (2)$$

where Q_{ug} is the flow rate statistic (Q95 or mean flow) at ungauged site (m^3/s), A_{ug} is the catchment area at ungauged site (m^2), A_g is the catchment area at gauged site (m^2) and Q_g is the flow rate statistic (Q95 or mean flow) at gauged site (m^3/s).

The main limitation to this method is that flow at the ungauged site is not always mostly strongly related to flow at the nearest catchment (Archfield and Vogel, 2010). We considered alternative methodologies to estimate flow indices including: 1) regional regression which estimates flow in space by relating flow to catchment characteristics such as geology and topography (Smakhtin, 2001), 2) flow contour mapping which involves plotting a flow index onto the centroid of its catchment and flow contour lines are drawn between each centroid using manual or automatic procedures (Smakhtin, 2001), and 3) the use of a hydrological model. The use of regional regression or flow contour mapping approaches was deemed inappropriate given the sample size of 6 gauges, which would likely be insufficient to reliably understand how flow varies in space (Babyak, 2004). Hydrological modelling was also considered, but any outputs would be associated with equifinality as a result of structural and parametric uncertainty (Beven, 2006). As such, the drainage area ratio methodology was selected as it was simple to use, better suited the amount of data available, and avoided any parametric or structural uncertainty.

Following the calculation of natural flow, the influence of the three large reservoirs within the catchment was estimated and applied to reaches downstream of the reservoirs. The influence of each reservoir was estimated by comparing the observed and estimated flow rates at several gauges downstream of the reservoirs. Further detail regarding the flow estimation methodology is provided in section S7 (see online supplementary material).

2.4.4 Model simulations

The GREAT-ER model was configured to determine how chemical exposure for TCS, TCC, E1 and E2 might have changed between 2010-2020, during the dry season. We focused on the dry season as dilution is at its lowest during this period and so concentrations are assumed to be highest, assuming that chemical emissions are constant throughout the year. It is possible that during intense rainfall events that occur during the wet season, raw sewage may be flushed into the waterways untreated leading to concentrations that exceed those in the dry season. The GREAT-ER model is unable to account for the types of mechanisms that occur during high flow conditions, but it is also generally quite difficult to characterise these events without calibration.

Once the GREAT-ER model was parameterised and validated, the model was configured to simulate a range of scenarios designed to assess the reduction of risk posed by all four chemicals. Firstly, we estimated the impact of providing full STW connectivity to the entire catchment. Secondly, we reduced chemical usage to attempt to reduce the risk at the source.

3. Results and discussion

3.1 Model accuracy and uncertainty

The accuracy of the 2010 and 2015 models was assessed by comparing modelled results to observed concentrations; the number of observations is limited and as such only provides an indication of model accuracy. For samples that were below the detection limit, we considered the concentration to be between the limit of detection and zero, therefore any modelled results that were within this range was considered to have an error of zero. In addition, as the GREAT-ER model provides results as a distribution, we assumed any observed concentrations which were between the 10th and 90th percentile had an error of 0 and for all other observations the error was equal to the greater value of either the difference between the observed concentration and the 90th percentile concentration or the difference between the 10th percentile modelled concentration and the observed concentration. We used the Nash-Sutcliffe efficiency metric (Nash and Sutcliffe, 1970) to determine the "goodness-of-fit" of the model results, which is calculated using Equation 3. Plots in Figure 1 show modelled and observed results.

$$NSE = 1 - \frac{\sum(Y_{obs} - Y_{pred})^2}{\sum(Y_{obs} - \bar{Y}_{obs})^2} \quad (3)$$

where NSE is the Nash-Sutcliffe efficiency, Y_{obs} is the observed flow rate (m^3/s), Y_{pred} is the predicted flow rate (m^3/s) and $\bar{Y}_{obs}$ is the mean observed flow rate (m^3/s).

Overall, the observed concentrations were largely within the modelled range, with relatively high Nash-Sutcliffe efficiency values of 0.87, 0.74, 0.66 and 0.89 recorded for TCS, TCC, E1 and E2 respectively. Concentrations at several sites on the mainstream were underestimated for all chemicals other than E2, which could suggest that dilution is overestimated and/or the underestimation of in-stream degradation, the latter being more important with increasing contributing area and residence times. In addition, the model tended to underestimate E1 at various sites across the catchment, the source of this error is difficult to determine given the many possible sources of error but could relate to additional inputs of E1 from non-domestic sources such as livestock (Combalbert and Hernandez-Raquet, 2010).

Model accuracy came at the cost of substantial parametric uncertainty, with concentrations varying by over an order of magnitude in most cases. To determine the largest source of uncertainty in the model parameters we completed the following analysis. We first set the STW removal and untreated wastewater removal to 50%. Then for each chemical, the mean usage was used, which was calculated from the data used to generate the probability distributions described in the model parameterisation section; in the case of E1 and E2, the mean in-stream removal was used, which was also calculated from the data used to generate the in-stream removal probability distributions. Then, for each parameter of interest, we ran the model using a low (5th percentile) and high (95th percentile) value from each parameter's probability distribution. Results are presented as the ratio of the highest and lowest concentration (Table 1). Overall, STW removal was determined to be the most important contributor to uncertainty, followed by usage. In the case of E1 and E2, in-stream removal contributed more to uncertainty than STW removal and untreated wastewater removal, but for E1 usage was by far the most important parameter.

Restraining this level of uncertainty would be difficult to achieve and not always inappropriate, as this would reduce the characterisation of the natural variability of relevant processes. It is possible to constrain uncertainty by calibrating against observations, but this would require a significant increase in the spatial and temporal frequency of sampling, which may not be feasible.

3.2 Evolution of chemical exposure over time

We estimated the risk of the four target chemicals across the East River catchment, characterised by concentrations that exceed each chemical's respective PNEC. For this risk assessment we took a conservative (worst-case) approach and as such we used the 99th percentile from our modelled results. The total catchment population and number of STWs is defined in Table 2 for 2010, 2015 and 2020. A summary of these results is presented in Figure 2 and maps describing the spatial and temporal variation of triclosan is presented in Figure 3; all other chemicals are presented in section S9 (see online supplementary material).

Between 2010-2015, population increased by ~300,000 across the catchment migrated from smaller settlements to the largest cities in the catchment. Also, during this period 17 new STWs were constructed across the catchment, particularly in the large urban areas. As such, the results from model simulations indicate a larger proportion of chemical mass was removed before discharge into receiving waters, resulting in decreased concentrations even in settlements that experienced population rise.

Between 2015-2020, population increased in all regions of the catchment with only 2 additional STWs constructed in this period, which is why the model estimated that a greater mass of chemical was released to waterways in 2020 in comparison to 2015. Between 2010-2020, the percentage of population that was served by a STW increased by 25%, but population change outpaced these improvements with an increase in total catchment population of 38.2%. The modelled median concentration for TCS reduced between 2010-2020, whereas the modelled concentration of all other chemicals increased (Figure 2). This appears to relate to the higher rate of in-stream removal for TCS, which means that wastewater released in the upper reaches of the catchment has less of an impact on downstream loads than other chemicals due to the longer residence time and therefore STWs that were constructed primarily in the lower reaches of the catchment are more impactful.

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3.3 Mitigation measures

Given that the improvements to wastewater connectivity were estimated to be outpaced by population change, we explored the impact of expanding the wastewater network so that the entire catchment is served by a STW. This scenario provides an indication of the maximum benefit that could be obtained from further wastewater improvements, acknowledging that this benefit would decrease if population increased.

Results suggest that for E1 and TCC, scenario A is effective in reducing the number of reaches exposed to concentrations that exceed each chemical's respective PNEC, whereas for TCS and E2, the number of reaches that exceeds the PNEC is only reduced by 2% and 6% respectively. Scenario A also results in the catchment median concentration reducing for each chemical, but the concentration of E2 and TCS still exceeded the PNEC throughout the catchment (Figure 2 and Figure 4).

To try and reduce the risk posed by TCS, we steadily reduced the usage level to determine what usage would be required to reduce the median concentration below the PNEC. It was determined that usage would need to be reduced by 91% to reduce the median concentration below the PNEC (Figure 2). For this scenario, the median TCS concentration was estimated to be reduced to below the PNEC, however the urban area of Shenzhen and Huizhou are still estimated to be exposed to concentrations significantly higher than the PNEC (Figure 5); river reaches downstream of major settlements that were not on the main stream, were also estimated to be exposed to concentrations that exceed the PNEC, but with significant improvements compared to the full STW connectivity scenario. Note that we did not attempt to reduce the usage level for E2, as we assumed that the majority of E2 released to wastewater was produced naturally by humans (Johnson *et al*., 2000) and as such, reducing the E2 load in wastewater influent wasn't feasible.

These results are less surprising when we consider some of the regulatory actions relating to TCS. In 2016, the use of TCS as an ingredient in antiseptic wash products was prohibited in the United States of America (U.S. Food and Drug Administration, 2016) and for use in biocidal products within the European Union (The European Chemicals Agency, 2015) as manufacturers were unable to prove they were safe to use and there was a lack of evidence demonstrating the benefits of their use in comparison to soap and water. Results described here suggest that similar restrictions may be insufficient to enable the safe use of TCS within catchments such as the East River, particularly as there is evidence of reduced but persisting presence of TCS in the United States in samples collected after restrictions came into place, in many cases exceeding the PNEC

(Adhikari *et al.*, 2022). At present TCS is still detected throughout China (Fu *et al.,* 2024) and as far as the authors know, there are no proposals to restrict the use of TCS.

4. Conclusion

Using the catchment scale chemical risk assessment model, GREAT-ER, we estimated the change in chemical exposure within the East River catchment, China, between 2010-2020. The model was then configured in order to perform a number of scenarios aimed at reducing chemical risk.

Similarly to the national picture, in 2010 a significant proportion of the catchment was not served by a STW, and wastewater was released to the waterways untreated. The STWs present in 2010 were primarily located in the largest cities within the catchment, with the majority of smaller settlements discharging wastewater untreated. Between 2010-2015, STWs were constructed rapidly throughout the East River catchment, which resulted in the overall concentration of down-the-drain chemicals decreasing. Between 2015 and 2020, concentrations increased to levels that were worse or similar to those in 2010 as a result of population growth and slower construction of new STWs.

The model was then configured to determine the impact of expanding the wastewater network to serve all population within the catchment, which resulted in the catchment median concentration reducing for all target chemicals. However, the catchment median concentration of TCS and E2 still exceeded the respective PNEC. Finally, we explored the effect of reducing the usage of TCS in order to reduce the median concentration below the PNEC. The usage of TCS had to be reduced by 91%, which suggests that to safely use TCS, its usage must be restricted heavily, ideally restricted to uses that will not result in emission to water.

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Figure 1 – Simulated concentrations for the 2010 and 2015 models, with comparison to observations. Modelled results are displayed for the 10th percentile, the mean and the 90th percentile concentration.

Figure 2 – Summary of modelled results for 2010 – 2020, as well as results for two mitigation scenarios: A) expanding wastewater network to serve 100% of the catchment's population and B) in addition to scenario A, triclosan usage is reduced by 91%.

Figure 3 – Estimated concentration of triclosan, population density and sewage treatment works distribution for 2010, 2015 and 2020.

Figure 4 – Estimated concentration of triclosan, triclocarban, estrone and 17β-estradiol in 2020 for scenario A in which 100% of the catchment is served by a sewage treatment works.

Figure 5 – Estimated concentration of triclosan in 2020 for scenario B in which 100% of the catchment is served by a sewage treatment works and triclosan usage is reduced by 91%.

Table 1 – Each parameter was set to a high (5th percentile) and low (95th percentile) value to determine each parameter's contribution to overall parametric uncertainty. The values below represent the ratio of the highest and lowest concentration estimated as a result of changing each parameter one-at-a-time.

Table 2 – Population, number of sewage treatment works, and the percentage of population served by a sewage treatment works for 2010, 2015 and 2020.