Fate simulation and risk assessment of TBT and TPhT considering water level fluctuations in the TGR before and after AFS Convention implementation in China

Jun-min Gao*, Ping-ting Fu, Xiao-ling Chen, Jin-song Guo, Xian-yu Hou, Jie Zeng and Zhu-man Chen

Abstract

Background: The Three Gorges Reservoir (TGR) is the largest freshwater reservoir in China. Previous studies showed that organotin pollution is present in the TGR. In June 2011, the AFS Convention went into effect in China. In order to explore the pollution evolution processes of tributyltin (TBT) and triphenyltin (TPhT) before and after implementation of the AFS Convention and their variations with water level fluctuations in the TGR, the characteristic parameters of the TGR and the physicochemical parameters of TBT and TPhT were used to develop a level IV multimedia fugacity model considering water level fluctuations to simulate the fate, transfer, and transport of TBT and TPhT in the TGR. Based on the simulation results, exposure concentrations of TBT and TPhT were then used to assess the ecological and health risks in the TGR region (TGRR).

Results: The simulation results showed that the average concentrations of both TBT and TPhT decreased in all compartments except the sediment, whereas the total content of the system continued to increase after the AFS Convention was implemented. The concentration of TBT in the sediment was higher than that in fish, while the concentration of TPhT in fish was much greater than that in the sediment. The total contents of both TBT and TPhT were highest in the sediment phase. In addition, variations in water level of the TGR significantly affected the distribution and transport of TBT and TPhT in the TGR.

Conclusions: Sediment is an important source and sink of TBT and TPhT, and the water level regulation of the TGR strengthened the two roles of sediment. Both TBT and TPhT in surface water, but especially TBT, carried significant chronic exposure risks to the aquatic community of the TGR. Intake of TPhT, mainly through eating fish, posed a potential health risk to the population in the TGRR.

Keywords: Tributyltin, Triphenyltin, The AFS Convention, Fate simulation, Risk assessment

Background

Tributyltin (TBT) and triphenyltin (TPhT) are the only two organometallic compounds among the 67 endocrine-disrupting chemicals listed by the US Environmental Protection Agency, and are the most toxic chemicals artificially discharged into aquatic

environments. Since the 1960s, the global production and use of organotins (OTs), particularly TBT and TPhT, has significantly increased [9, 26]. Because of their bactericidal properties, TBT and TPhT are widely used as antifouling agents, wood preservatives, fungicides, and pesticides [2, 14]. Previous studies have shown that TBT and TPhT can accumulate in some aquatic organisms and cause cumulative poisoning or imposex [3, 48], eventually leading to reproductive disorders, population decline, and potential species

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extinction [54, 62, 80]. In humans, exposure to OTs may induce cell immunotoxicity, neurotoxicity, mutagenicity, and carcinogenicity, resulting in reproductive and immunological diseases [36, 73]. Because of the harmful effects of TBT and TPhT on aquatic organisms and human health, many countries and international organizations have developed water quality criteria and restricted the uses of TBT and TPhT [56]. The International Convention on the Control of Harmful Anti-fouling Systems on Ships (AFS Convention) was adopted by the International Maritime Organization in 2008 and officially went into effect on June 7, 2011, in China. However, TPhT remains widely used agriculturally in China, except in Taiwan, which banned the use of TPhT pesticides in 1999 [45].

The Three Gorges Project is the largest water management project in the world, and the Three Gorges Reservoir (TGR) is the largest freshwater reservoir in China. The TGR provides a habitat for aquatic organisms and a drinking water source for the TGR region (TGRR). After the TGR was created, the hydrologic regimes of the Yangtze River and its tributaries in the reservoir region changed markedly compared with those of the original rivers. The water level of the reservoir alternates between 145 and 175 m on an annual cycle, with periodic fluctuations that are opposite to the seasonal fluctuations of natural rivers. The flow rate and silt concentration in the reservoir also regularly vary with the water level. Thus, the TGR has characteristics of both rivers and lakes, and pollutants in the water column are periodically mixed, suspended, and sunk; the resulting environmental behaviours of pollutants are more complicated than those in rivers or lakes. In addition, the navigation conditions differ based on water levels in the TGR, which may strongly impact the release of OTs. Although China has implemented the AFS Convention, there remain some small and old ships with OT anti-fouling paints used in rivers, and other sources of TBT and TPhT may be present. Moreover, TBT and TPhT are stable in sediments and their half-lives range from several months to several decades [76]. Dredging, eddy currents, and benthic disturbances may cause sediment to be resuspended, and resuspended sediments can act as a source of OTs in water. Previous studies have shown that OT pollution is found in various environmental media (such as water, suspended matter, and sediment) in the TGR. The highest concentration of TBT observed in the water column of the TGR thus far is 393.35 ng Sn L^{-1} [28], and the highest concentration of TPhT is 37.2 ng Sn L^{-1} [29]; both concentrations are sufficient to pose significant ecological risks to aquatic organisms in the TGR. Thus, OT pollution in the TGR after implementation of the AFS Convention requires further study.
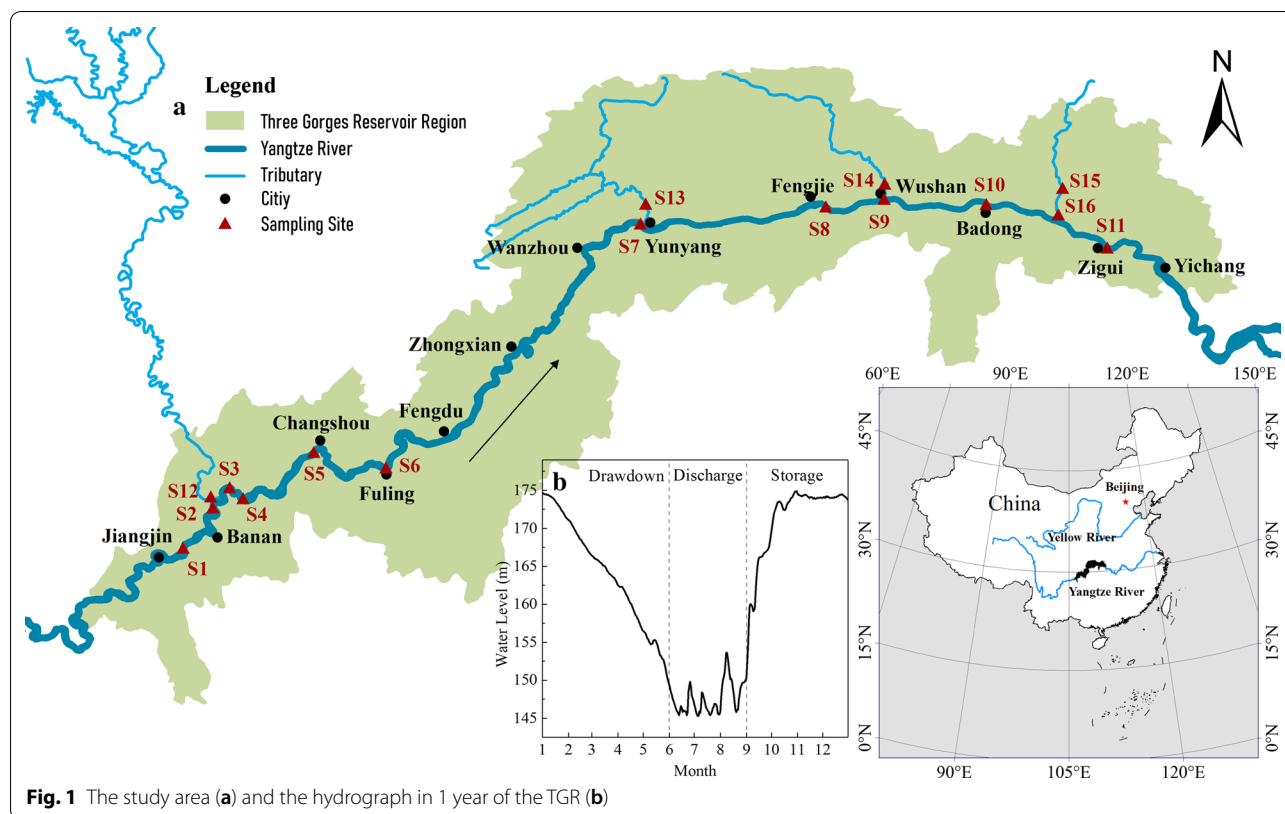
However, field monitoring and tracking investigations are time-consuming and costly, and can only represent limited pollution circumstances at a given time; therefore, it is difficult to assess the pollution level of the overall aquatic environment. The environmental multimedia model offers a simple and economical method to describe the environmental behaviours of chemical substances in various media [17, 19]. Therefore, environmental multimedia modelling, particularly involving the fugacity approach, is widely used for chemical risk assessment and fate simulation studies [60], such as fate simulation of antibiotics in seasonal glacial rivers [61], fate evaluation of polycyclic aromatic hydrocarbons in the Songhua River [69], and the fate and transport of spilled oil in the Arctic region [5]. Although Ranke [50] used a simple water–sediment multimedia model to simulate the persistence of TBT anti-fouling paint in the ocean, few other studies have investigated OTs using the environmental multimedia model, especially in a complicated aquatic environment such as the TGR.

Therefore, this study combined characteristics of the ecological environment and hydrological changes in the TGR to establish a Level IV multimedia fugacity model of TBT and TPhT, in order to explore the fate, transfer, and transport of TBT and TPhT in different media of the aquatic environment based on water level fluctuations; it then assessed risks to aquatic organisms and humans in the TGRR before and after the AFS Convention was implemented in China. The results of this study can be used to evaluate the effectiveness of the implementation of the AFS Convention, provide a scientific basis for OT management and risk assessment, and offer new insights into the environmental behaviours of endocrine disruptors in deep-channel reservoir systems that experience water level fluctuations.

Materials and methods
Study area
The study area (Fig. 1a), the TGRR, is located between 29°16′–31°25′ N and 106°0′–111°50′ E. The reservoir has more than 30 tributaries, a length of approximately 660 km, and an average width of approximately 1100 m. From 2003 to 2008, the TGR experienced water levels of 135 m, 156 m, 172 m and 175 m in the storage stage. Since 2008, the water level of the reservoir has periodically fluctuated between 145 and 175 m (Fig. 1b) to meet the needs of flood control and power generation in a pattern that is opposite to the seasonal fluctuations of natural rivers. After storage began in the reservoir, the flow velocity of the main river decreased from 2 to 0.25–0.50 m s^{-1}, while that of the tributaries was reduced to 0.006 m s^{-1}. The average annual temperature in the TGRR is approximately 16.6 °C and the annual average

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Fig. 1 The study area (**a**) and the hydrograph in 1 year of the TGR (**b**)

rainfall of 1124.5 mm occurs mainly from April to October; this period contributes 80% of total annual precipitation [71].

Level IV multimedia fugacity model with water level fluctuations in the TGR

Model development

The model used in this study was based on the QWASI model developed by the Canadian Environmental Modelling Centre [43]. Four bulk compartments (phases) were considered, including atmosphere, water, sediment, and fish. Each bulk compartment contains sub-compartments. The atmosphere consists of air and aerosols; water consists of water and suspended solids; sediment contains solids and pore water; for simplicity, the biota compartment is represented by fish. Four types of environmental behaviours, including water advection, emission, degradation, and intermedia transfer, were regarded as the main processes affecting TBT and TPhT in the aquatic environment of the TGR, as shown in Fig. 2.

The model assumed that TBT and TPhT were in a homogeneous, unsteady, and non-equilibrium state in all four bulk compartments. Based on the level IV fugacity approach and mass balance, differential equations were established as follows (Eqs. 1–4):

Atmosphere:

$$V_A Z_{TA} \frac{df_A}{dt} = D_{WA} f_W - (D_{AW} + D_{AR} + D_{PW} + D_{DW}) f_A, \tag{1}$$

Water:

$$\begin{aligned} V_W Z_{TW} \frac{df_W}{dt} = {} & G_{OW} C_{OW} + E_W + D_{AW} f_A \\ & + (D_{SW} + D_{Re}) f_S + D_{FW} f_F \\ & - (D_{WA} + D_{WS} + D_{WF} \\ & + D_{RW} + D_{WO} + D_D) f_W, \end{aligned} \tag{2}$$
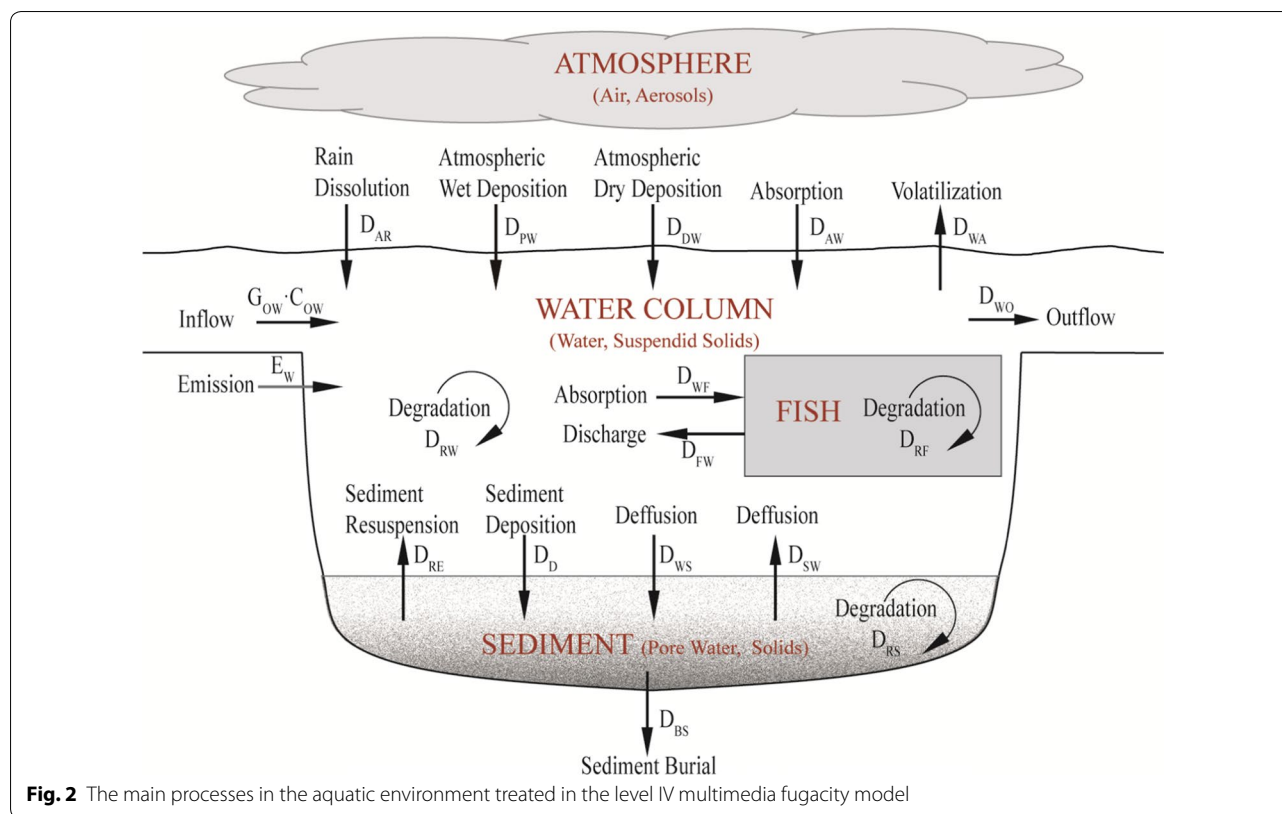
Sediment:

$$V_S Z_{TS} \frac{df_S}{dt} = (D_{WS} + D_D) f_W - (D_{SW} + D_{RS} + D_{Re} + D_{BS}) f_S, \tag{3}$$

Fish:

$$V_F Z_{TF} \frac{df_F}{dt} = D_{WF} f_W - (D_{RF} + D_{FW}) f_F, \tag{4}$$

where t—simulation time (h); A, W, S, F—atmosphere, water, sediment, fish; V_i (i = A, W, S, F)—the volume of each bulk compartment (m^3); Z_{Ti} (i = A, W, S, F)—the fugacity capacity of each bulk compartment (mol m^{-3} Pa^{-1}); f_i (i = A, W, S, F)—the fugacity (Pa); D_{i-j} (i, j = A, W, S, F)—the rate of intermedia diffusion

Fig. 2 The main processes in the aquatic environment treated in the level IV multimedia fugacity model

(mol h^{-1} Pa^{-1}); D_{Ri} ($i =$ A, W, S, F)—the rate of degradation in each bulk compartment (mol h^{-1} Pa^{-1}); D_{AR}, D_{PW}, D_{DW}—the rate of rain dissolution, wet deposition, dry deposition (mol h^{-1} Pa^{-1}); D_D, D_{Re}, D_{BS}—the rate of sediment deposition, sediment resuspension, sediment burial (mol h^{-1} Pa^{-1}); D_{WO}—the rate of effluent in the water compartment (mol h^{-1} Pa^{-1}); G_{OW}—the flow into the reservoir (m^3 h^{-1}); c_{OW}—the concentration of the pollutant of the inflow (mol m^{-3}); E_W—the rate of pollutants emission (mol h^{-1}).

Model parameterization

The definitions and assignments of the original calculation parameters in the TBT and TPhT multimedia

fugacity model are shown in Additional file 1: Tables S1 and S2. The definitions and calculation equations of the intermediate parameters, which are fugacity capacity (Z-values) and the rates of transport and transformation (D-values), are shown in Tables 1 and 2, respectively.

The octanol–water partition coefficient (K_{ow}) and the organic carbon partition coefficient (K_{oc}) were corrected using the formula $K_{oc} = 0.4\ K_{ow}$ [44]. The degradation reaction rate constant and half-life were corrected with the formula $\tau_{1/2} = \ln 2/k$ [44]. The correction formula for Henry's constant (H) is shown in Additional file 1: Table S3. Based on water level data, the corresponding areas and volumes of the TGR, functional relationships between the water level and area, and volumes of the

Table 1 Calculation equations of Z-values (mol m^{-3} Pa^{-1})

Bulk compartment	Sub-compartment	Calculation equation	Z-value
Atmosphere	Air	$Z_A = 1/RT$	$Z_{TA} = Z_A + Z_{PA}\, v_{pa}$
	Aerosols	$Z_{PA} = 6 \times 10^6 Z_A/p^S$	
Water	Water	$Z_W = 1/H$	$Z_{TW} = Z_W + Z_{PW}\, v_{pw}$
	Suspended solids	$Z_{PW} = Z_W\, \rho_{pw}\, K_{oc}\, \omega_{ocw}$	
Sediment	Pore water	$Z_{WS} = 1/H$	$Z_{TS} = Z_{WS}\, v_{ws} + Z_S\, v_{ps}$
	Solids	$Z_S = Z_W\, \rho_{ps}\, K_{oc}\, \omega_{ocs}$	
Fish	Fish	$Z_F = Z_W\, \mathrm{BCF}\, \rho_f$	$Z_{TF} = Z_F$

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Table 2 Calculation equations of *D*-values (mol h^{-1} Pa^{-1})

Media	Process	*D*-value	Calculation equation
Atmosphere → water	Atmosphere dry deposition	D_{DW}	$D_{DW} = U_P A_W v_{pa} Z_{PA}$
	Atmosphere wet deposition	D_{PW}	$D_{PW} = U_R A_W Q v_{pa} Z_{PA}$
	Rain dissolution	D_{AR}	$D_{AR} = U_R A_W Z_W$
	Absorption	D_{AW}	$D_{AW} = 1/(1/K_{VA} A_W Z_A + 1/K_{VW} A_W Z_W)$
Water → atmosphere	Volatilization	D_{WA}	$D_{WA} = D_{AW}$
Water	Degradation	D_{RW}	$D_{RW} = K_{WR} V_W Z_W$
Water → sediment	Diffusion	D_{WS}	$D_{WS} = 1/(1/K_{SW} A_S Z_W + Y_S/B_{SW} A_S Z_W)$
	Sediment deposition	D_D	$D_D = U_{DP} A_S Z_{PW}$
Sediment	Degradation	D_{RS}	$D_{RS} = K_{SR} V_S Z_S$
	Sediment burial	D_{BS}	$D_{BS} = U_{BS} A_S Z_S$
Sediment → water	Diffusion	D_{SW}	$D_{SW} = D_{WS}$
	Sediment resuspension	D_{Re}	$D_{Re} = U_{RS} A_S Z_S$
Water → fish	Absorption	D_{WF}	$D_{WF} = K_{WF} V_F Z_F$
Fish	Degradation	D_{RF}	$D_{RF} = K_{FR} V_F Z_F$
Fish → water	Discharge	D_{FW}	$D_{FW} = K_{FW} V_F Z_F$
Advection	Inflow	D_{ow}	$D_{ow} = G_{ow} Z_{ow}$
	Outflow	D_{wo}	$D_{wo} = G_{wo} Z_W$

TGR were obtained through binomial fitting. Some OT transport and transformation parameters vary with the environmental parameters of the TGR. For instance, the air–water mass transfer coefficient (K_{VA}) is a function of wind speed, and the transfer coefficient of water–air (K_{VW}) varies with the flow velocity and water depth. Both K_{VA} and K_{VW} were corrected using the formulas shown in Additional file 1: Table S3.

In this study, emissions of TBT and TPhT were considered to include both leaching from ship hulls and runoff of TPhT pesticides. With reference to the literature [74] and the Pollution Discharge Coefficient Method [42], the emissions of TBT and TPhT in the TGR were estimated as follows:

$$E_{TBT} = \sum \left(k \cdot 10^{-5} (L_i B_i \varphi_i)(N_a \chi_i) V_i \right.$$
$$\left. \times \left(\frac{\theta_i T_i}{24} + \frac{\Delta S}{S \times 222.24} \right) \right), \tag{5}$$

$$E_{TPhT} = \sum \left(k \cdot 10^{-5} (L_i B_i \varphi_i)(N_a \chi_i) V_i \right.$$
$$\left. \times \left(\frac{\theta_i T_i}{24} + \frac{\Delta S}{S \times 222.24} \right) \right)$$
$$+ (E_{pww} \times S_{ps} \times L_{ps}), \tag{6}$$

where N_a is the number of ships at ports; k is the estimated proportion of ships that continue to use antifouling paints considering ship scrapping and new application, which was 1 prior to the implementation of the AFS Convention and 0.7 thereafter [20]; χ_i is the proportion of each ship type: 28%, 47%, 2%, 20%, and 3% for passenger ships, cargo ships, tugboats, non-transport ships, and fishing boats, respectively; θ_i is the proportion of boats with anti-fouling paint: 1 and 0.7 for the merchant ship and fishing boat classes, respectively, [13]; V_i (µg cm^{-2} day^{-1}) is the leaching rate of OTs (TBT and TPhT): for TBT, this rate is 4.0 (berthing) and 5.1 (navigation) [55], while for TPhT, it is 0.5 (berthing) and 1.0 (navigation); T_i (h) is berthing time, which is 1 h for a passenger ship or fishing boat and 25 h for all others; L_i and B_i are the representative length and width (m), respectively, of the bottom of the ship, referring to the standard sizes of ships in the TGRR; φ_i is a correction factor for the ship bottom area: for cargo ships and fishing boats, this is 0.8, while for other ships, it is 0.6; S (km h^{-1}) is the speed of the ship, which is 27 when only commercial ships are considered; the value of ΔS is 1 km; E_{pww} (kg hm^{-2} a^{-1}) is the pollutant discharge coefficient, with a value of 8.20×10^2 [15]; S_{ps} (hm^2) is the area of cultivated land; and L_{ps} is the coefficient of pollutant loss from cultivation, which is dimensionless with a value of 0.01 [47].

Model calculation and performance

The simulation period was 24 months, from June 2010 to May 2012, corresponding to simulation times of months 1–24, as shown in Figs. 3, 4, and 7. The year prior to the implementation of the AFS Convention was from June 2010 to May 2011, while the year after its implementation was from June 2011 to May 2012. Daily mean values were obtained through cubic-spline

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interpolation and used as the smallest unit of variation; a time step of 1 was used. The initial value of the simulation was the fugacity obtained using the model differential equations at d$f |t = 0$. Input parameters such as temperature, rainfall rate, inflow, and outflow were all defined as functions of time. The model was simulated using MATLAB R2016b, and the fourth-order Runge–Kutta method was used to numerically solve the differential equations to obtain f for each compartment. After the model performance was evaluated, the concentration, transport flux, and mass flux were calculated for each compartment. Because the input parameters were dynamic, the results showed temporal variability. The process of modelling and computing is presented in Additional file 1: Fig. S1.

Water and sediment samples were collected in the reservoir region three times in 2012 (sampling sites are shown in Fig. 1) to obtain the concentrations of TBT and TPhT. Model performance was evaluated using Theil's inequality coefficient method [46] and the percent bias method [31] (Additional file 1: Text S1). In the water phase, the Theil's inequality coefficient values of TBT and TPhT were 0.1090 and 0.1388, respectively, and the percent bias values were $+0.1998$ and $+0.2237$, respectively. In the sediment phase, the Theil's inequality coefficient values of TBT and TPhT were 0.0631 and 0.1283, respectively, and the percent bias values were -0.1345 and -0.2596, respectively. Both of these compartments satisfied the criteria of Theil's inequality coefficient < 0.5 and percent bias $\in [-0.7, +0.7]$ [35], indicating that the prediction error of the model was acceptably small. Although the TBT and TPhT concentrations in fish in the TGR were not observed, the modelled concentrations of TPhT and TBT in the fish phase before implementation of the AFS Convention in this study were 73.51 ± 13.10 ng Sn g^{-1} and 0.31 ± 0.05 ng Sn g^{-1}, respectively, which are similar to the findings by Hu et al. [34] that the concentration of TPhT in the livers of Chinese sturgeon in the TGR was 68.0 ± 31.2 ng Sn g^{-1} and the concentration of TBT was < 1 ng Sn g^{-1}. Therefore, the performance of the multimedia fugacity model developed in this study is considered adequate.

Sensitivity and uncertainty in the model
The sensitivity coefficient (SC) [11, 41] was calculated to identify the influences of individual input parameters (Eq. 7). The corresponding parameter can be regarded as a key input parameter when the SC is > 0.5 [39], showing that it has a strong impact on the model results. The results (Additional file 1: Fig. S2) showed that G_{OW} and C_{OW} had strong impacts on predicted concentrations in all four compartments, with SC values near 1. The main source of OT pollution in the reservoir

is the upper reaches of the river and its tributaries, with the TGR acting as an important sink of OTs. In addition, v_{ps}, U_{BS}, ω_{ocw}, U_{DP}, ρ_{pw}, K_{oc}, and ρ_{ps} have major effects on the predicted concentrations of TBT and TPhT in the sediment compartment. BCF, K_{FW}, and K_{WF} strongly influence the predicted concentrations in the fish compartment. Accuracy in setting these parameters is essential for reliable simulation results:

$$\text{SC} = \frac{|Y(X_{F=1} + X_{F=0.1}) - Y(X_{F=1} - X_{F=0.1})|}{2 \times 0.1} \frac{1}{Y(X_{F=1})}. \tag{7}$$

Here, X is the specified input parameter; Y is the corresponding result obtained from the model with input parameter variation; and $Y(X|F=1)$ and $Y(X|F=0.1)$ are the simulated concentrations when the specified parameter is multiplied by a factor of 1 and 0.1, respectively.

Uncertainty arises from variability and numerical errors in various input parameters. Based on the results of sensitivity analysis, some of the input parameters selected were represented with probability density functions. Then, Monte Carlo simulation was performed 10,000 times for uncertainty analysis [11]. The results are shown in Additional file 1: Fig. S3. The coefficient of variation (CV) of the simulated concentration was $\leq 15\%$, which was smaller than that of the measured concentration.

Risk assessment
The simulated concentrations of TBT and TPhT before and after implementation of the AFS Convention were applied to assess the ecological and health risk in the TGR.

Ecological risk assessment
The risk quotient (RQ) [38] was calculated to assess the ecological risk of TBT and TPhT for organisms in the TGR (Eq. 8).

$$\text{RQ} = \frac{\text{PEC}}{\text{PNEC}} \tag{8}$$

Here, PEC represents the exposure concentration (ng Sn L^{-1}) in the water phase predicted by the model. PNEC represents the Predicted No-effect Concentration (ng Sn L^{-1}) which is from water quality criteria (WQC) and toxicity thresholds in published literature. If RQ ≥ 1, the organisms in the aquatic environment are at significant risk. If $0.3 \leq$ RQ < 1, there is a potential risk. If RQ < 0.3, there is no ecological risk [38].

Health risk assessment
The intake routes of TBT and TPhT including eating fish, drinking water, and skin absorption were considered in

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this study. The hazard index (HI, Eq. 9) [68] and tolerable average residue level (TARL, Eq. 13) [33] of TBT and TPhT in fish were calculated to assess health risks in the TGRR.

(1) HI

$$HI = \sum_i (ADD_i / RfD_j). \tag{9}$$

Here, RfD_j is the reference dose (μg $(kg_{bw}\ d)^{-1}$)a: for TBT, the reference dose is 0.25 [49], while for TPhT, it is 0.5 [63]; ADD_i is the average daily dose (i represents one of the three routes: eating fish (F), drinking water (W) and skin absorption (S)) (μg $(kg_{bw}\ day)^{-1}$), which is estimated by Eqs. 10–12. If HI > 1, there is a significant health risk to the population in the TGRR; if HI < 1, there is no health risk [65]:

$$ADD_F = c_F \times IR_F \times A_F / BW, \tag{10}$$

$$ADD_W = c_W \times TF \times IR_W \times A_W / BW, \tag{11}$$

$$ADD_s = c_W \times k_P \times SA \times F \times A_s / \left(BW \times 24 \times 10^{-3} \right). \tag{12}$$

Here, c_F and c_W are the simulated concentrations in fish (ng g^{-1}) and in water (ng L^{-1}), respectively; IR_F is the average daily consumption of fish, which was calculated according to the annual consumption of fish and the population in the TGRR, and the value of that is 59.84 g day^{-1}; TF represents the OTs purification coefficient of the waterworks, which is 1 for both TBT and TPhT [57, 59]; IR_W denotes the average daily intake of

drinking water, which is 2.3 L day^{-1} for an adult [22]; k_p is the osmotic factor on the skin surface, which is 0.001 cm h^{-1} [68]; SA is the accessible skin surface area, which is 14,150 cm^2 for an adult [66]; F denotes the skin exposure time, which is 1 h [79]; A_i (i = F, W, S) means the absorption factor of eating fish, drinking water, and skin absorption, which are 100% [37], 100% [22], and 1% [78], respectively; BW denotes the body weight (kg), which is 60 kg based on the average weight of Chinese.

(2) TARL

$$TARL = \frac{TDI \times BW}{ADSC} \tag{13}$$

Here, TDI denotes tolerable daily intake (μg $(kg_{bw}\ d)^{-1}$), for TBT, the TDI is 0.25 [49], while for TPhT, it is 0.5 [40]. BW is the average body weight (kg) of Chinese, which is 60 kg. ADSC is average daily consumption of aquatic products, which is 59.84 g day^{-1}.

Results and discussion
Fate simulation of TBT and TPhT with water level fluctuations in the TGR
Concentration variations of TBT and TPhT in different phases
Variations in the concentrations of TBT and TPhT during the simulation periods before and after the implementation of the AFS Convention are shown in Fig. 3. Compared with the concentrations before implementation of the AFS Convention, the average concentrations of TBT and TPhT in the water and fish phases decreased after implementation. The average concentration of TBT in the water phase decreased by 12.27% (from 4.36 to 3.83 ng Sn L^{-1}), while that of TPhT decreased by 6.98%
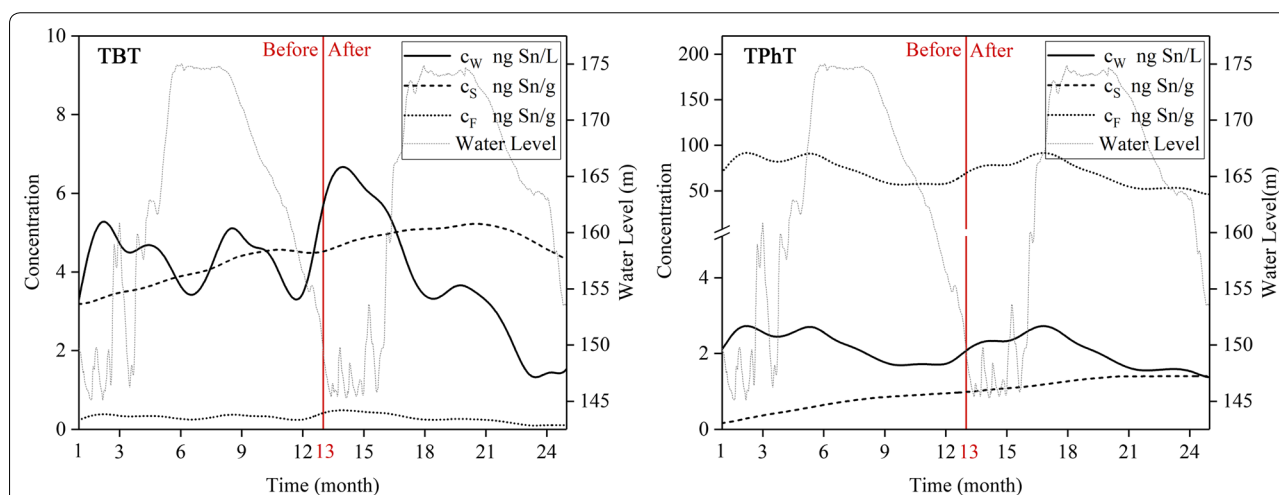
Fig. 3 Variations in the concentrations of TBT and TPhT in different phases during the simulation periods. The concentrations of TBT and TPhT in the atmosphere (c_A) are below 10^{-4} ng Sn L^{-1}, which are not shown in the figure

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(from 2.19 to 2.04 ng Sn L^{-1}). The average concentration of TBT and TPhT in fish showed the same trend as in water, which means the decline of concentrations of TBT and TPhT in aquatic environment resulted in reducing the overall concentration level of TBT and TPhT in fish. These results showed that the reduction of TBT concentration after implementation of the AFS Convention was more obvious than that of TPhT. Notably, for TBT, high concentrations in water phases occurred mainly during low-water periods. This pattern may be due to the inflow, outflow, and flow velocity increasing during low-water periods, resulting in greater turbulence. The resuspension of sediment is a source of TBT that increases the concentrations in other phases. Small peaks of TBT concentration also appeared in water phase during the winter high-water period, which may be due to the combined effects of more navigation, low degradation rate of TBT, and other factors. For TPhT, concentrations in the water and fish phases fluctuated slightly with water level variations and seasonal changes, but were generally similar regardless of implementation of the AFS Convention. Aside from resuspension of sediment induced by turbulence during the flood season, fluctuations in concentrations of TPhT might be also related to the seasonal application of TPhT pesticides, soil erosion, and surface runoff [27, 28].

Notably, the TBT and TPhT concentrations in the sediment phase fluctuated little during the simulation period, although the concentration of TPhT in the sediment phase showed a gradual increase throughout the simulation period. For TBT, the concentration in the sediment phase showed an increasing trend before the AFS Convention and continued to increase for 10 months after implementation of the AFS Convention. This delay may be due to the long half-lives of TBT and TPhT in sediment and their slow degradation rates. Choi et al. [16] reported that TBT has a half-life of up to 17 years in sediment. The concentration of TBT was greater in the sediment phase than in the fish phase, while the concentration of TPhT was much greater in the fish phase than in the sediment phase. This difference indicates that TPhT accumulates more easily in fish because of bioaccumulation and biomagnification. The concentration simulated for the fish phase in this study represents the average level in a general distribution of fish species and is related to the overall resource availability and growth rate of fishes in the TGR. Seasonally mediated physiological changes, such as growth and metabolic compensation [21], as well as hydrologic alteration of the TGR (e.g., alternating water discharge and storage operations), can lead to variations in fish resources and their physiological activities, thereby affecting the pattern of OT accumulation in fish.

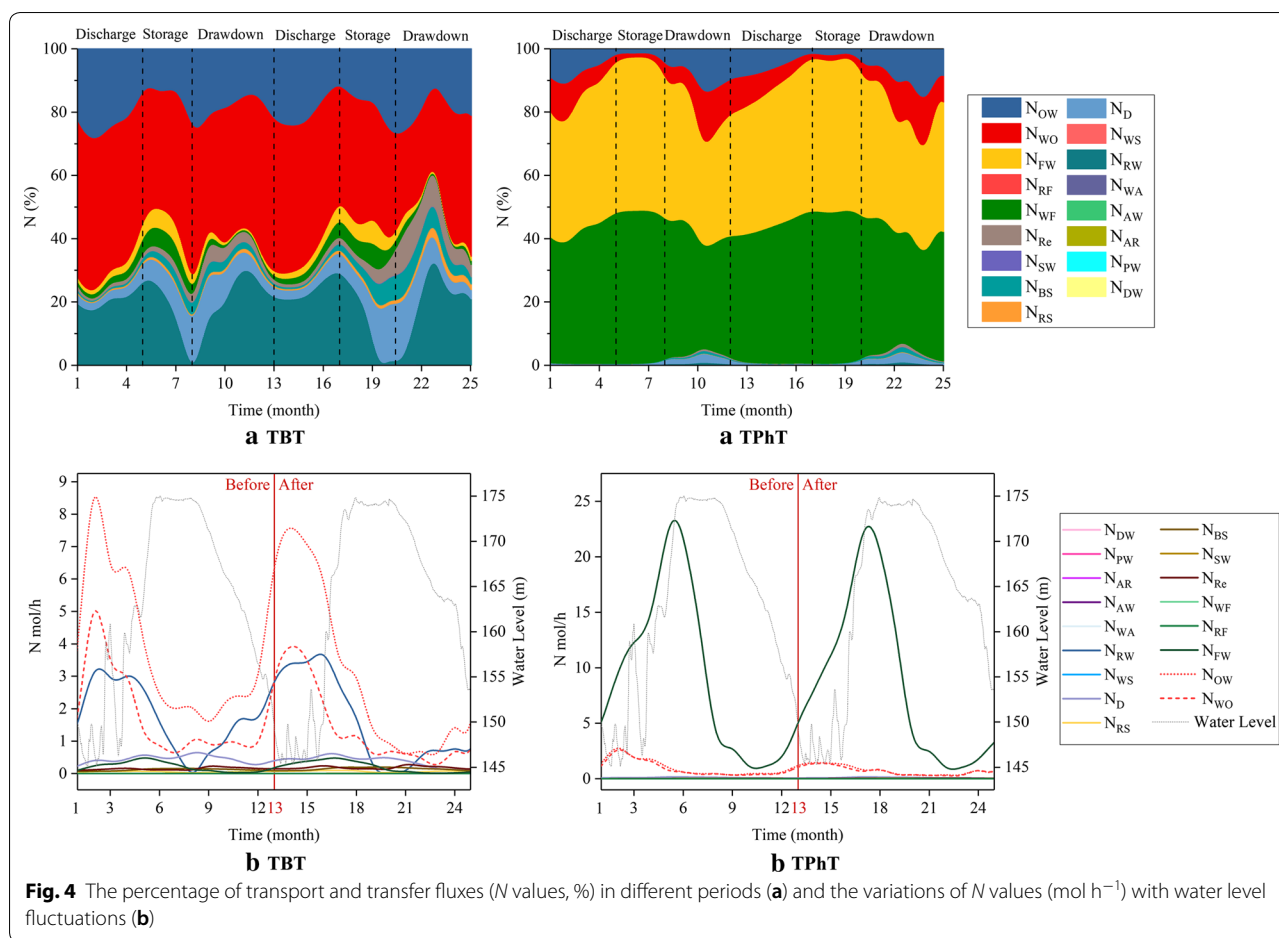### Variations in transport and transfer fluxes (N) among phases

The percentage of N (%) for all environmental processes is shown in Fig. 4a. The N_{OW}, N_{WO}, and N_{RW} values were highest for TBT during the simulation period, with average proportions of 43.32%, 20.04%, and 21.13%, respectively, indicating that advection and degradation in water were important multimedia transfer and transport processes affecting TBT. The percentage of N between water and sediment was also high, comprising 9.92% of the total. TBT accumulated in sediment mainly through the deposition of particles, while resuspension was the main process by which TBT was released from the sediment into the water. These two processes were both significantly affected by the operation of the TGR. Despite the fact that advection, degradation in water, and transport between water and sediment were important for both TBT and TPhT, for TPhT N_{FW} and N_{WF} were more dominant than that for TBT. This may be because TPhT has greater biosorption capacity and lipid solubility than TBT [6, 7], and fish have a certain ability to metabolize TBT in contrast to TPhT [10], which allow it a preferential accumulate of TPhT in fish to exhibit more significant bioaccumulation and biomagnification effects [58, 72].

The N values of TBT and TPhT varied regularly with fluctuations of the water level in the TGR (Fig. 4b). The peak N values for TBT occurred during the discharge period at low water; there are two possible reasons for this phenomenon. First, the low-water period was during summer in the TGR, when high temperatures led to an increase in the degradation rate constant of TBT in the water column. In addition, flooding resulted in greater inflow and outflow, leading to enhanced turbulence. For TPhT, N_{FW} and N_{WF} increased during the discharge period, then decreased when the storage period began. This pattern may have manifested because fish resources and their physiological activities were most abundant during the discharge period of the TGR, resulting in increased transport between the fish and water.

Mass fluxes and residue levels of TBT and TPhT before and after implementation of the AFS Convention

The cumulative mass fluxes and residue levels for the year before and the year after implementation of the AFS Convention are shown in Fig. 5. For the year after the AFS Convention was implemented, most of the cumulative mass fluxes of TBT and TPhT decreased. In contrast, cumulative mass fluxes from the sediment to water and for sediment burial both increased slightly. The calculated residue levels for the four phases before and after the AFS Convention exhibited the same sequence: sediment > water > fish > atmosphere. Residues of TBT and TPhT in the sediment after implementation of the AFS

Fig. 4 The percentage of transport and transfer fluxes (*N* values, %) in different periods (**a**) and the variations of *N* values (mol h^{-1}) with water level fluctuations (**b**)

Convention increased by 24.67% and 92.91%, respectively. This result may be related to the properties of TBT and TPhT, which are readily adsorbed onto particles and have long half-lives. This result also revealed that the sediment is an important sink of both TBT and TPhT. This sink can be transformed into a source that is non-negligible under certain conditions. After the TGR began operation, more sediment was produced due to the water storage, thereby emphasising the sink and source properties of the sediment and increasing the levels of OTs in the aquatic environment.

After the AFS Convention was implemented, the residue levels of the atmosphere, water, and fish were reduced by 21.21%, 11.24%, and 12.23% for TBT and 18.68%, 5.87%, and 6.91% for TPhT, respectively; however, the total contents of TBT and TPhT in the system increased by 15.20% and 38.10%, respectively. This result suggests that although implementation of the AFS Convention had the effect of reducing OT pollution, the residue levels of TBT and TPhT in the overall TGR system remained high and continued to increase. In particular,

the sediment properties of both sink and source areas affect the concentrations of OTs in aquatic systems for an extended duration. The calculation results also showed that 1460.51 kg TBT and 716.48 kg TPhT entered the lower reaches of the TGR within 1 year after implementation of the AFS Convention, which may pose risks to downstream aquatic organisms and populations.

Ecological risk assessment of TBT and TPhT for organisms in the TGR

Considering the high toxicity and bioaccumulation characteristics of TBT and TPhT, many countries and researchers have established standards and risk thresholds for OTs. Previous studies showed that the acute thresholds for both TBT and TPhT were greater than 48 ng Sn L^{-1} [25, 51, 53, 64, 70, 77], while the maximum concentrations predicted for TBT and TPhT during the simulation period in this study were 6.67 and 2.73 ng Sn L^{-1}, respectively; therefore, the acute risk for organisms in the TGR is negligible.
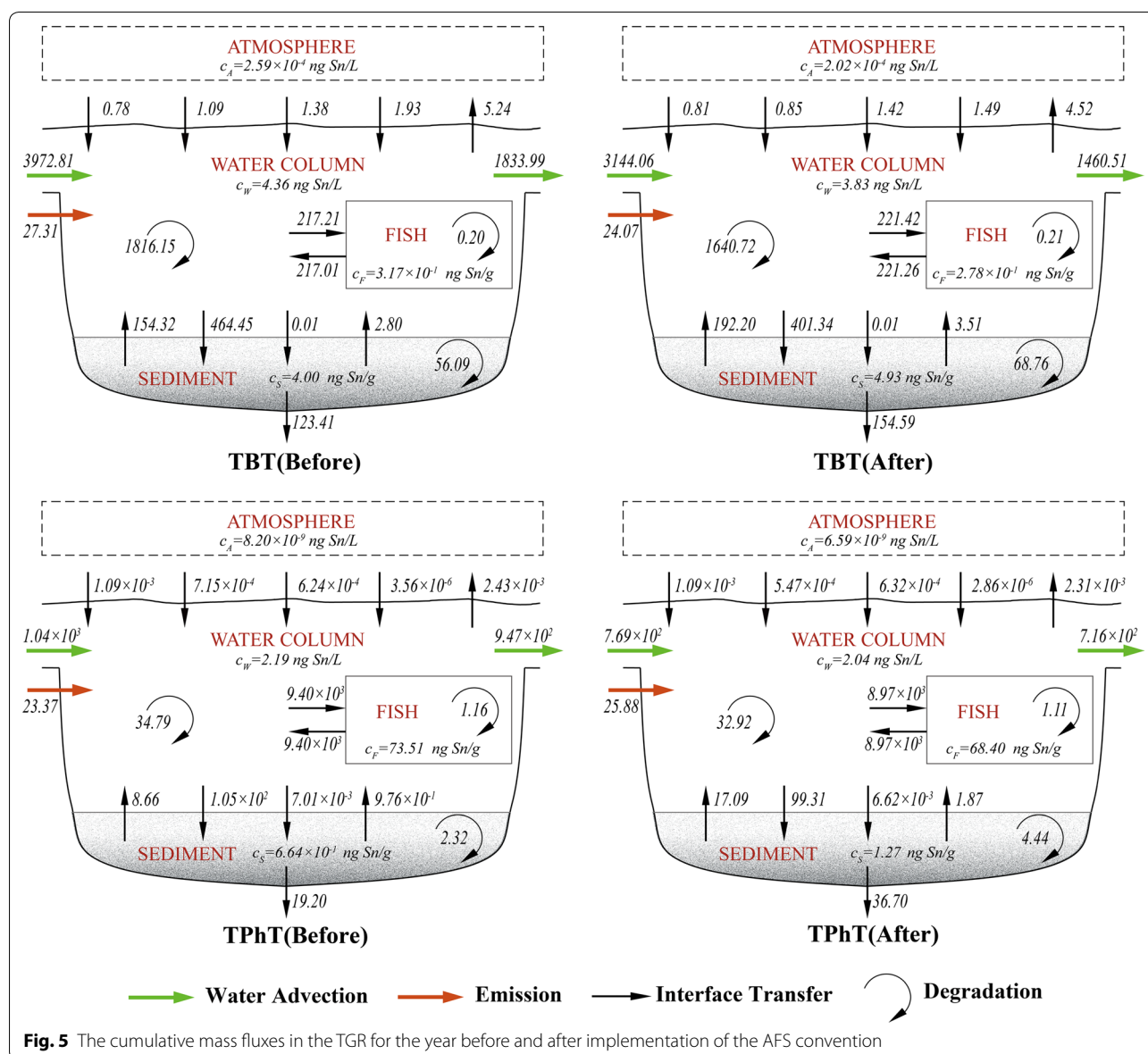
Fig. 5 The cumulative mass fluxes in the TGR for the year before and after implementation of the AFS convention

In contrast, the chronic ecological risks of TBT and TPhT to aquatic organisms in the TGR were more severe. RQ values calculated using different criteria and chronic exposure thresholds are shown in Fig. 6. These results show that the decline in chronic ecological risk from TBT and TPhT after implementation of the AFS Convention has not been robust. For TBT, all RQ values were greater than 1, with a maximum of 59.84 [51]; therefore, TBT posed a significant chronic ecological risk to aquatic organisms in the TGR, and may cause chronic toxicity in aquatic organisms [4, 8, 32, 51, 64], such as oyster calcification abnormalities [1] and weight loss and imposex in snails [30]. For TPhT, most RQ values were greater than 1, with a maximum of 16.83 [52]. In particular, RQ values

calculated using the endpoints of suppressed spawning frequency [80], eye defects in embryos or larvae [80], and imposex in snails [77] were all greater than 1, suggesting that the chronic ecological risk of TPhT in the TGR cannot be ignored.

Health risks from TBT and TPhT to the population of the TGRR

Calculation of the average daily dose (ADD) in the TGRR showed that for TBT, ADD_s (47.01%) > ADD_f (34.65%) > ADD_w (18.34%), indicating that the intake of TBT occurs mainly through skin absorption when swimming or bathing. For TPhT, ADD_f (99.59%) > ADD_s (0.29%) > ADD_w (0.11%), indicating that the intake of
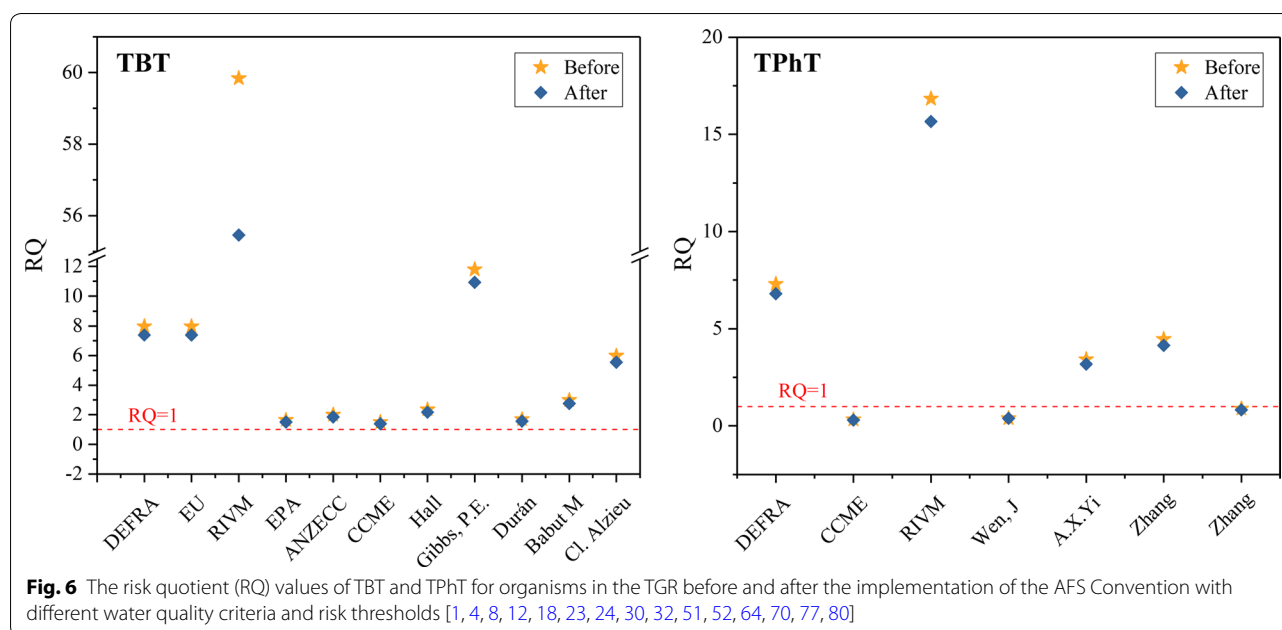
Fig. 6 The risk quotient (RQ) values of TBT and TPhT for organisms in the TGR before and after the implementation of the AFS Convention with different water quality criteria and risk thresholds [1, 4, 8, 12, 18, 23, 24, 30, 32, 51, 52, 64, 70, 77, 80]

TPhT occurs mainly through eating fish. The results of the health risk assessment are shown in Fig. 7. The hazard index values of TBT ranged from 0.0032 to 0.0161, and those of TPhT ranged from 0.34 to 0.60. The hazard index values of both TBT and TPhT were less than 1, suggesting that health risks to the population in the TGRR from TBT and TPhT were low during the simulation period [67]. Compared with TBT, TPhT carried a higher health risk for the population, as its highest hazard index value (0.60) during the simulation period was near 1.

The calculation of tolerable average residue level (TARLs) showed that the TARL of TBT was 92.09 ng Sn g^{-1}, while that of TPhT was 154.38 ng Sn g^{-1}. The maximum simulated concentration (0.48 ng Sn g^{-1}) of TBT in fish during the simulation period was much lower than the TARL of TBT, while the maximum TPhT level (91.59 ng Sn g^{-1}) was close to the TARL of TPhT. These results indicate that TPhT contamination of aquatic products is a serious problem in the TGRR. The TARLs of TBT and TPhT in Taiwan were calculated by Lee et al. [37]; these were 88.6 ng Sn g^{-1} and 149.4 ng Sn g^{-1}, respectively. The TARL of total butyltin ($\sum$BTs = TBT + dibutyltin + monobutyltin) in seafood from Chinese Bohai coastal waters, calculated by Yang et al. [75], was 90.8 ng Sn g^{-1}. The previous results are similar to those of the present study.

Although the health risk of TBT in the TGRR was low, a previous study found that, in some locations, the concentration of TBT reached 161.82 ng Sn L^{-1} in 2010 and 393.35 ng Sn L^{-1} in 2012 [29]; thus, TBT posed much greater health risks to the populations of some areas
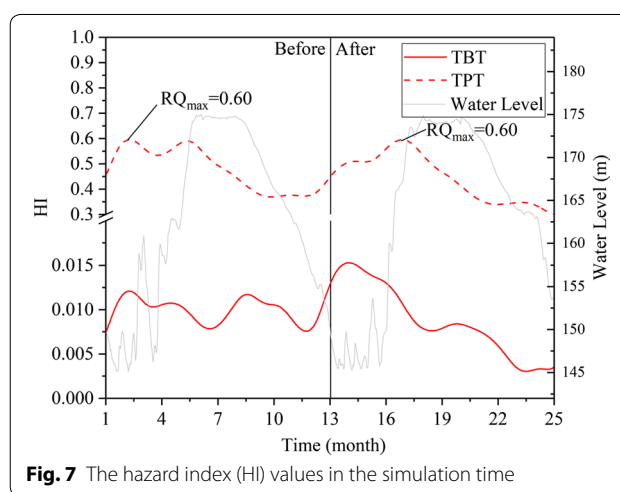
Fig. 7 The hazard index (HI) values in the simulation time

within the TGRR. Moreover, for TPhT, the average daily intake of aquatic products (ADSC) in the TGRR was 59.84 g day^{-1}. Fisheries in the TGRR have expanded, and the ADSC of some populations may be much greater than the reported value; fishermen or those who have a preference for fish will experience greater health risks. In addition, the simulated concentration of TPhT represents the average level in the TGRR. For some specific fish or regions, the concentration of TPhT in the fish phase may be greater and therefore pose an increased health risk to the population.

With the rapid development of agriculture and aquaculture in China, the application of TPhT as a fungicide

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and insecticide may increase over the next few years or decades [14]; notably, TPhT is not yet regulated by the Chinese government. Thus, comprehensive monitoring and strategic management of TPhT pollution are essential to controlling the contamination level in the TGRR, especially in sensitive areas such as source regions of potable water, in order to reduce the health risks from TPhT to the population of the TGRR.

Conclusions

A multimedia fugacity model was developed to simulate the fate, transfer, and transport of TBT and TPhT before and after implementation of the AFS Convention while considering water level fluctuations in the TGR. The simulation results were used to assess ecological and health risks in the TGRR. The simulation results revealed that implementation of the AFS Convention reduced both TBT and TPhT concentrations in three of the four phases considered in this study, but not in sediment. The reduction of TBT was more robust than that of TPhT. However, TBT and TPhT in sediment and the overall system remained at high levels and continued to increase. The concentrations of TBT and TPhT, as well as the transfer and transport fluxes in each phase, varied periodically with the cycle of reservoir regulation and water level. Sediment is an important source and sink of TBT and TPhT, and the alternation of these two roles became more apparent with water level fluctuations, affecting the concentrations of OTs in the aquatic environment over an extended period. TBT and TPhT pose significant chronic risks to aquatic organisms in the reservoir area, and the risk from TBT is greater than that from TPhT. Based on average levels, TBT and TPhT pollution in the TGRR do not pose significant health risks to the population. However, the health risk from TPhT, mainly due to intake of contaminated fish, is of concern, especially to populations that engage in greater intake of aquatic products in some areas of the TGRR. Therefore, continuous monitoring and effective management measures are essential for controlling OT pollution in the TGRR.

Supplementary information

Supplementary information accompanies this paper at https://doi.org/10.1186/s12302-020-0298-6.

Additional file 1: Table S1. Input parameters: physical and chemical properties. **Table S2.** Input parameters: characteristic parameters of the TGR. **Table S3.** Correction formulas for original parameters. **Text S1.** Model performance evaluation method. **Fig. S1.** The process of modeling and computing. **Fig. S2.** Sensitivity analysis of TBT and TPhT. **Fig. S3.** Frequency distribution and coefficient of variation (CV) of TBT and TPhT concentrations.

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