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EARTH SCIENCES

Three Gorges Dam: friend or foe of riverine greenhouse gases?

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ABSTRACT

Dams are often regarded as greenhouse gas (GHG) emitters. However, our study indicated that the world's largest dam, the Three Gorges Dam (TGD), has caused significant drops in annual average emissions of CO₂, CH₄ and N₂O over 4300 km along the Yangtze River, accompanied by remarkable reductions in the annual export of CO₂ (79%), CH₄ (50%) and N₂O (9%) to the sea. Since the commencement of its operation in 2003, the TGD has altered the carbonate equilibrium in the reservoir area, enhanced methanogenesis in the upstream, and restrained methanogenesis and denitrification via modifying anoxic habitats through long-distance scouring in the downstream. These findings suggest that 'large-dam effects' are far beyond our previous understanding spatiotemporally, which highlights the fundamental importance of whole-system budgeting of GHGs under the profound impacts of huge dams.

Keywords: Three Gorges Dam, greenhouse gas, spatiotemporal variation, equilibrium, Yangtze River, whole system analysis

INTRODUCTION

Most rivers worldwide are supersaturated with greenhouse gases (GHGs) owing to inputs of carbon (C) and nitrogen (N) from land, and become net sources of GHGs for the atmosphere [1]. To meet the growing global demand for water and energy, more than 70 000 large dams have been constructed [2]. Such dams are regarded as a source of excessive GHG emissions [3–5]. The estimated annual emissions are 48 Tg C as CO₂ and 3 Tg C as CH₄ from global hydropower reservoirs, and 0.03 Tg N as N₂O from all reservoirs in the world [4,6].

Previous studies on the effects of dams on GHGs have been mostly limited to the vicinity of reservoirs [7–10]. Although these considerations hold for small dams (reservoir capacity < 10 km³), the impact of large dams on GHGs (reservoir capacity ≥ 10 km³) is much greater because the original physical and biochemical equilibria are disrupted over large spatiotemporal scales. Firstly, a large dam alters the hydrodynamic conditions and material fluxes of a river: after operation commences, the

peak flood discharge decreases and fluxes of nutrients and sediments exported to the sea are often reduced [11–14]. Secondly, the river regime tends to remain stable, but increasing longitudinal erosion of the riverbed beyond the dam causes long-term readjustment over a considerable distance [15]. Thirdly, changes to water and sediment fluxes significantly affect the functioning of microbial communities [16–18] (e.g. photosynthesis, methanogenesis and denitrification) and GHG emissions (Supplementary Table 1).

As the world's largest dam, the Three Gorges Dam (TGD) has been regarded as a significant source of GHG emissions [3,4,19]. For example, CO₂ and CH₄ emissions from the 25 km² core reservoir area upstream of the TGD in 2008 were estimated to be 40 and 20 Gg yr⁻¹, respectively, ~40- and 20-fold larger than before impoundment [20]. Similar findings [4,21] reported that the total CH₄ emission rate in the Three Gorges Reservoir (TGR) was 0.315 Gg yr⁻¹. However, the impact of the TGD extends far beyond the reservoir area. The TGD has

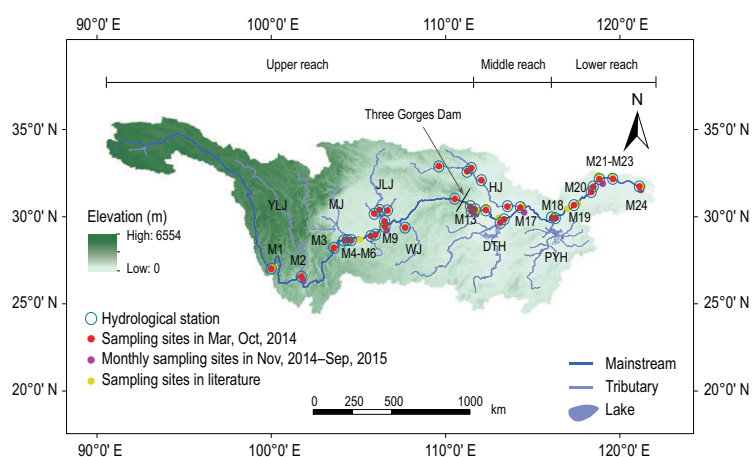


Figure 1. The Yangtze River Basin and sampling sites. Lines indicate the mainstream river and its tributaries, the former having a length of 4300 km (i.e. the actual sinuous channel length, equivalent to 2.05 times the straight-line distance of 2102 km from the start to the end of the sampling sites). Yellow solid circles show the locations of previous sampling sites (see Supplementary Tables 2–3); red solid circles show the locations of our recent simultaneous sampling sites in March and October 2014 (for details see Supplementary Table 4); purple solid circles show the locations of our monthly sampling sites from October 2014 to September 2015; blue open circles show the locations of the hydrological stations. The upper reach is from Shigu (M1) to Yichang (M13), the middle reach from Yichang to Hukou (M18) and the lower reach from Hukou to Xuliujing (M24). The major tributaries include Yalongjiang (YLJ), Minjiang (MJ), Jialingjiang (JLJ), Wujiang (WJ) and Hanjiang (HJ); two river-regulated lakes are Dongting (DTH) and Poyang (PYH).

altered hydrodynamic conditions along almost the entire length of the Yangtze, as physical and biochemical processes have readjusted both upstream and downstream of the dam, most notably the long-distance, long-term scouring of the riverbed downstream of the dam [15,22,23]. This highlights the necessity of whole-river analysis in order to properly assess the changes in GHG fluxes caused by large dams.

Here, we estimate changes in dissolved and emitted fluxes of GHGs in the Yangtze River before and after the TGD became operational in 2003. Based on the time series of 30 water quality indices monitored over 312 months (1990–2015) and the measured GHGs (Supplementary Tables 2–4) along 4300 km of the Yangtze River (Fig. 1), CO_2 is calculated using the well-known CO2SYS model, while CH_4 and N_2O are estimated with artificial neural networks (ANNs; see Methods).

RESULTS AND DISCUSSION

Temporal effect of the TGD on CO_2 fluxes

The mean annual $p\text{CO}_2$ between 1990 and 2002 was $2526 \mu\text{atm}$ (Fig. 2). Subsequently, $p\text{CO}_2$ declined greatly to $1336 \mu\text{atm}$ once the TGD began operation over the whole mainstream (Fig. 2a). This de-

clining trend is particularly significant in the middle and lower reaches, though annual $p\text{CO}_2$ in the upper reach remained relatively steady before and after 2003 (Fig. 2b–d). The spatially averaged annual $p\text{CO}_2$ was $2205_{-925}^{+2497} \mu\text{atm}$ (where the numbers display the mean and range of values) in the middle reach. $p\text{CO}_2$ increased to $2974 \mu\text{atm}$ during the 1990s, peaked in 1996 and declined significantly to $1720 \mu\text{atm}$ after TGD impoundment [24] (Fig. 2c). In the middle reach, $p\text{CO}_2$ decreased from 2907 to $1446 \mu\text{atm}$ in the wet season and from 2196 to $1377 \mu\text{atm}$ in the dry season (Supplementary Fig. 1a–d).

From 1990 to 2015, CO_2 exported to the East China Sea exhibited substantial inter-annual variations (Supplementary Fig. 2). The mean annual value increased from $\sim 469 \text{ Gg C yr}^{-1}$ in 1993 and reached a peak of $3354 \text{ Gg C yr}^{-1}$ during the 1998 flood before declining to pre-1993 levels by 2003 (Supplementary Fig. 2). The mean exported CO_2 flux from 1991 to 2015 was $1128 \text{ Gg C yr}^{-1}$, corresponding to 5.6% of dissolved inorganic carbon transported by the Yangtze River (Supplementary Table 5). The annual averaged CO_2 outgassing flux and CO_2 exported to the sea over the Yangtze experienced remarkable drops of 55% and 79% since 2003, suggesting a much stronger effect, due to TGD impoundment, on $p\text{CO}_2$ than that from other influencing factors (such as the anthropogenic discharge of sulfur and nitrogen containing pollutants) reported previously [24].

Monthly and annual CO_2 emission fluxes from the upper, middle and lower reaches were on average lower after 2003 than before, indicating that the entire mainstream progressively became a smaller emission source (Supplementary Fig. 3). The largest change occurred in the middle and lower reaches, where CO_2 emission flux dropped from $2723 \text{ Gg C yr}^{-1}$ before TGR impoundment to $1087 \text{ Gg C yr}^{-1}$ after. Annual averaged CO_2 emission flux from the Yangtze mainstream was estimated as $2420_{-1200}^{+2590} \text{ Gg C yr}^{-1}$ (Supplementary Table 6), which accounts for emissions from 1.3% of global rivers and 4.8% of temperate rivers [1,25] between 25°N and 50°N . These results were convincing with uncertainty analysis based on representative stations as described in the Supplementary Data.

Temporal effect of the TGD on CH_4 fluxes

To estimate dissolved and emitted CH_4 over the Yangtze River before and after impoundment of the TGR, monthly observed data of chemical oxygen demand, dissolved oxygen, water temperature, pH and nitrogen during 1990–2015 were used for validation and verification as input variables of ANN

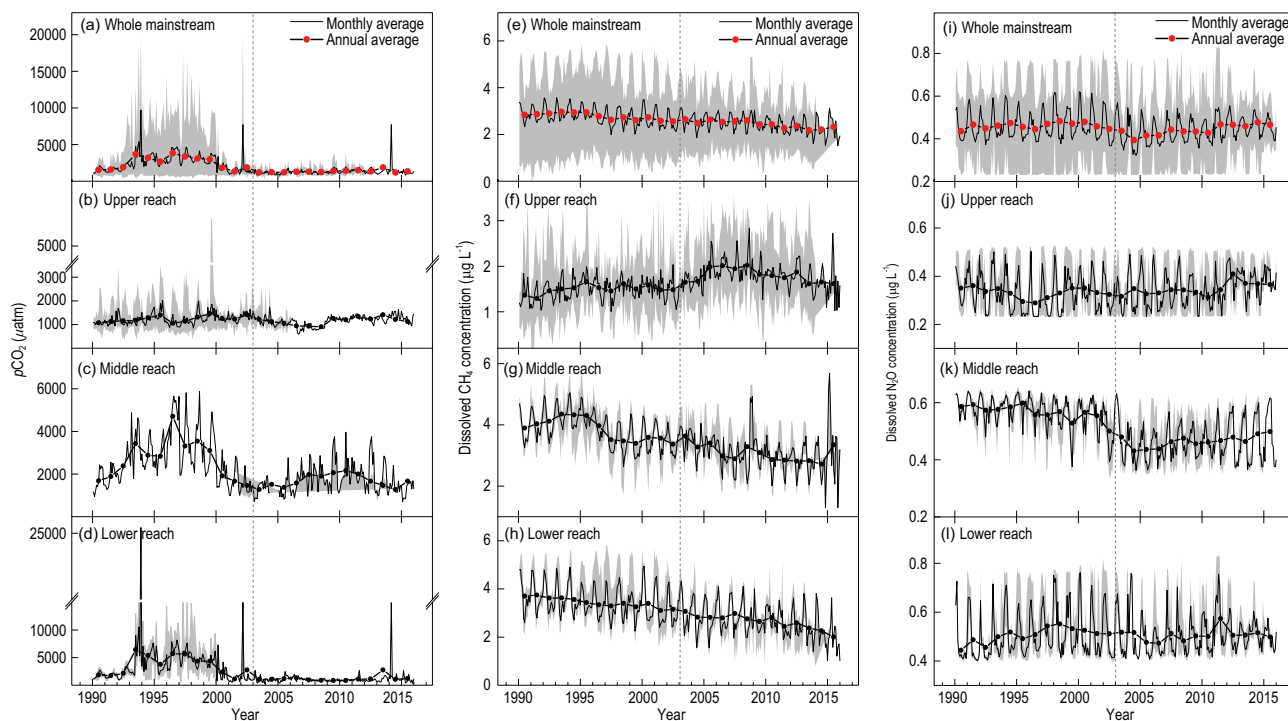


Figure 2. Temporal variations in monthly and annual averages of dissolved-GHG concentrations from 1990 to 2015: (a–d) $p\text{CO}_2$, (e–h) dissolved CH_4 , and (i–l) dissolved N_2O . The shadow areas represent the range of dissolved-GHG concentrations at different monitoring stations in the corresponding reaches. Vertical dashed lines denote 2003, when the TGD commenced operation.

models (see Methods). Supplementary Fig. 4 shows spatiotemporal variations in dissolved nitrogen (NH_4^+ , NO_3^- , NO_2^-) in the whole mainstream during the period 1990–2015.

After the TGR impoundment in 2003, both dissolved and emitted CH_4 concentrations increased in the upper reach, decreased in the middle reach and hardly changed in the lower reach (Fig. 2f–h, Supplementary Fig. 5b–d). The annual averaged CH_4 concentration from 1990 to 2015 over the whole mainstream was $2.22^{+0.54}_{-0.65} \mu\text{g L}^{-1}$ (Fig. 2e), comparable to that for the Amazon River (Supplementary Table 7) [26]. The mean dissolved CH_4 was $3.15^{+0.62}_{-0.56} \mu\text{g L}^{-1}$ in the dry season and $2.57^{+0.59}_{-0.72} \mu\text{g L}^{-1}$ in the wet season in the Yangtze (Supplementary Fig. 1). A major change in seasonal cycles of dissolved CH_4 occurred in 2003. In the wet season, the mean dissolved CH_4 increased from 1.45 to 1.95 $\mu\text{g L}^{-1}$ in the upper reach but decreased from 3.51 to 3.02 $\mu\text{g L}^{-1}$ in the middle reach. Based on the parameters derived from representative stations (Supplementary Table 8), temporal variation in CH_4 flux exported to the East China Sea decreased from 3.1 to 1.5 Gg C yr^{-1} after 2003 (Supplementary Fig. 6). Emitted CH_4 flux decreased from 3.3 to 2.7 Gg C yr^{-1} along the whole mainstream, having increased from 0.4 to 0.5 Gg C yr^{-1} upstream of the dam and decreased from 2.9 to 2.2 Gg C yr^{-1}

downstream of the dam since the operation of the TGD (Supplementary Fig. 7).

Temporal effect of the TGD on N_2O fluxes

Input variables in the ANN model for estimation of N_2O emissions included dissolved oxygen, water temperature, pH and nitrogen. Total dissolved nitrogen ($\text{NH}_4^+ + \text{NO}_3^- + \text{NO}_2^-$) increased during the period of interest, while NH_4^+ and NO_2^- had much lower concentration levels than NO_3^- (Supplementary Fig. 4). This is consistent with increasing nitrogen input from fertilizers to the Yangtze River basin in the past few decades, enhanced by population and economic growth in central and east China [27,28]. After training and verification of the ANN, the modeled results showed a slight reduction of dissolved and emitted N_2O owing to the dam's operation since 2003. Over the Yangtze mainstream, the annual average concentration was $0.45^{+0.38}_{-0.22} \mu\text{g L}^{-1}$ (Fig. 2i), demonstrating a moderate dissolved N_2O concentration compared with other large rivers (Supplementary Table 9). Dissolved N_2O reached a maximum of 0.55 $\mu\text{g L}^{-1}$ at the Xuliujing station in the river mouth (Fig. 2l), and a minimum of 0.32 $\mu\text{g L}^{-1}$ at the Luzhou station in the upper reach (Fig. 2j). Impoundment of

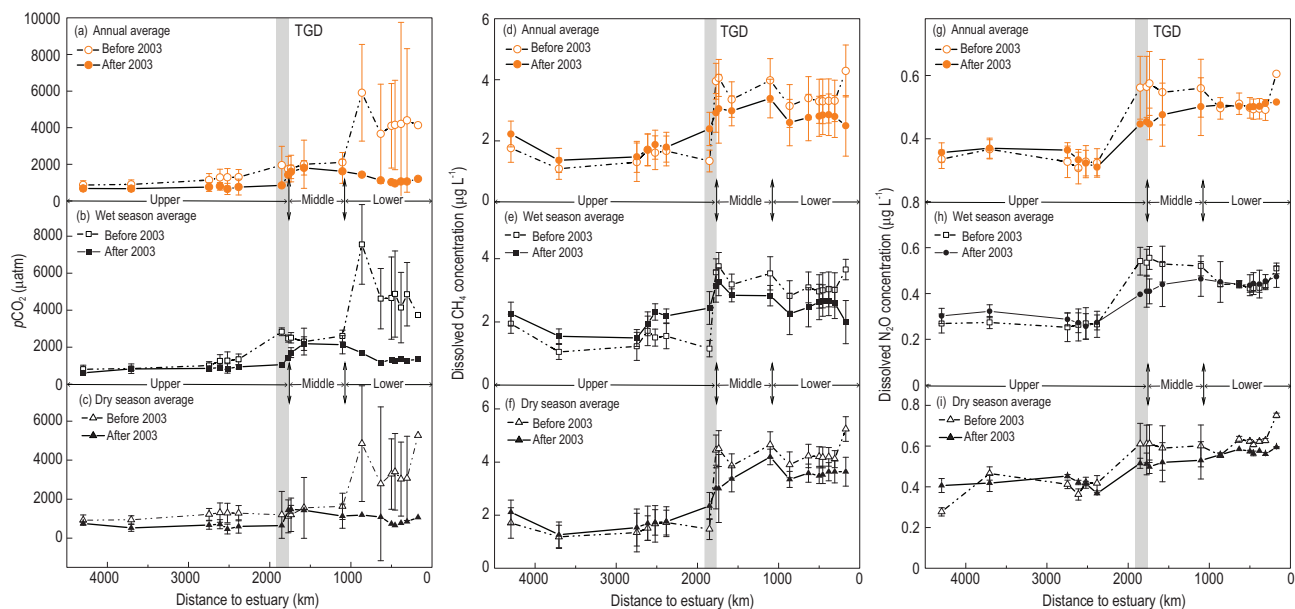


Figure 3. Spatial variations in annual and seasonal dissolved-GHG concentrations in the 4300-km stretch of the Yangtze River: (a–c) dissolved- CO_2 concentration, (d–f) dissolved CH_4 concentration, and (g–i) dissolved N_2O concentration. The error bars are the annual standard deviations at the given monitoring stations. The shaded area indicates where the TGD reservoir is located.

the TGR operation caused dissolved N_2O to decrease from 0.56 to $0.46 \mu\text{g L}^{-1}$ in the middle reach after 2003 (Fig. 2k). Large amplitude variations in seasonal N_2O patterns also occurred in the middle reach (Supplementary Fig. 1k). After 2003, the average dissolved N_2O concentration declined from 0.61 to $0.51 \mu\text{g L}^{-1}$ in the dry season and from 0.54 to $0.41 \mu\text{g L}^{-1}$ in the wet season in the middle reach. Seasonal differences of N_2O emission rates were also calculated (Supplementary Fig. 8e–h). The long-term average (1990–2015) displayed higher N_2O emission rates at Yichang and Wuhan in the wet season than in the dry season, in all cases indicating the Yangtze was a net source of N_2O (Supplementary Fig. 8). Meanwhile, N_2O emission rates at Yichang have fallen from 39.3 to $19.2 \mu\text{g m}^{-2} \text{h}^{-1}$ during the wet season and from 18.4 to $11.6 \mu\text{g m}^{-2} \text{h}^{-1}$ during the dry season (Supplementary Fig. 8g). Based on monthly dissolved N_2O and flow discharge, the highest values of N_2O fluxes to the estuary occurred in 1998, the year with historical floods. Mean annual dissolved N_2O fluxes to the estuary decreased from 0.46 to $0.41 \text{ Gg N yr}^{-1}$ after TGD impoundment in 2003 (Supplementary Fig. 9), because of the disruptive effect on the physical and biochemical equilibria of the river. The annual N_2O outgassing in the mainstream was estimated as $0.43 \text{ Gg N yr}^{-1}$ (Supplementary Fig. 10).

Spatial effect of the TGD on GHG emissions

Before 2003, $p\text{CO}_2$ ranged from 880 to $4399 \mu\text{atm}$ in the mainstream channel of the Yangtze River (Fig. 3a). A trend of increasing $p\text{CO}_2$ was evident along the mainstream, rising from $1314 \mu\text{atm}$ in the upper reach to $4111 \mu\text{atm}$ in the lower reach, along with the decreasing pH level of the lower reach and dilution by water entering from Poyang Lake during the period 1990–2002. After 2003, $p\text{CO}_2$ was almost constant upstream of the TGD, but rose immediately downstream of the dam, affected by flow regulation and sediment trapping [29]. It has been estimated that reservoir sedimentation caused by the presence of a dam results in an average carbon accumulation rate of $400 \text{ g m}^{-2} \text{ yr}^{-1}$ globally [30]. Carbon burial therefore results in a potential available carbon source for biological respiration and might increase $p\text{CO}_2$ in a reservoir, particularly in the early years after impoundment [31]. Other human activities might also increase exchanges between water and mineral, thus increasing $p\text{CO}_2$ [32]. Similar trends of increasing $p\text{CO}_2$ were observed along the mainstream in both wet and dry seasons (Fig. 3b–c). The higher values of $p\text{CO}_2$ in the wet season compared to the dry season, especially in middle and lower reaches, might be due to the efficient production of soil-originated CO_2

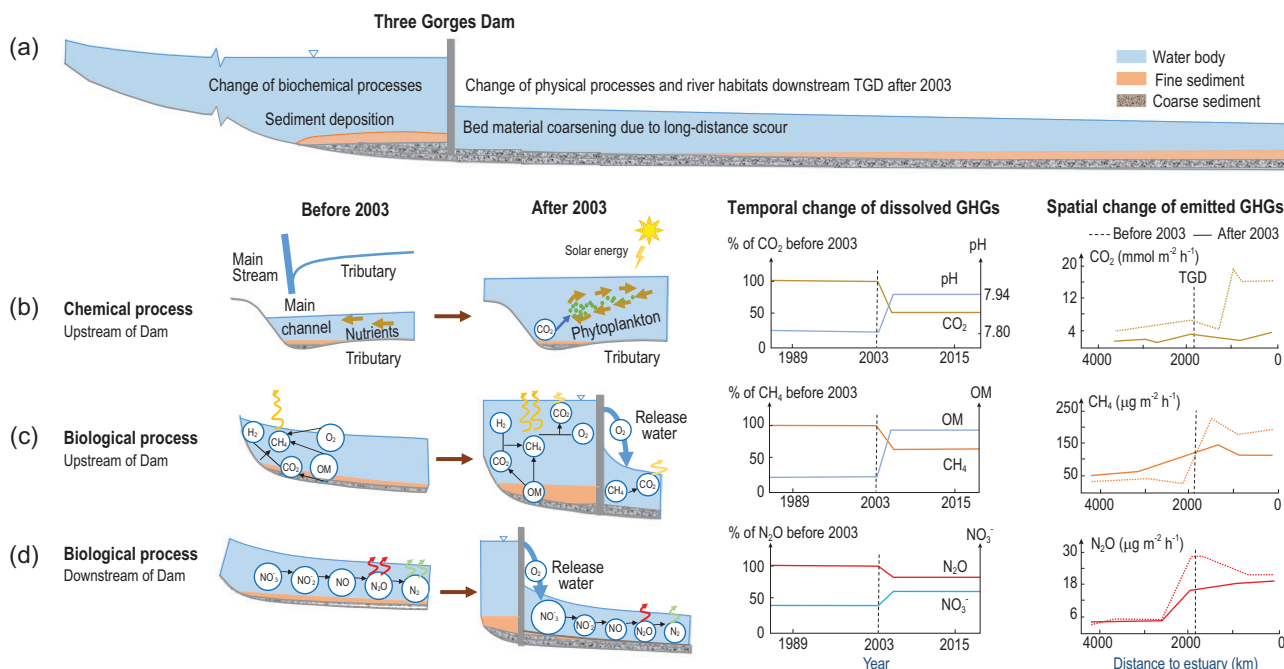


Figure 4. (a–d) Whole-system analysis concerning readjustment of physical and biogeochemical equilibria involved in the regulation effects of the TGD on GHG emissions from the Yangtze River.

and its transportation by surface run-off [31]. Supplementary Fig. 11 shows the CO₂ emission rate profiles along the mainstream before and after operation of the TGD. These are qualitatively very similar to the dissolved CO₂ profiles. After 2003, the mean CO₂ emission rate along the mainstream was $3.0 \pm 1.7 \text{ mmol m}^{-2} \text{ h}^{-1}$. Degassing rates were higher in the middle and lower reaches than in the upper reach, controlled by $p\text{CO}_2$.

CH₄ concentration was lowest in the upper reach of the Yangtze in both wet and dry seasons (Fig. 3d–f), primarily because of lower levels of organic matter. After 2003, CH₄ concentration increased slightly from 1.50 to 1.83 $\mu\text{g L}^{-1}$ in the upper reach, and decreased from 3.13 to 2.74 $\mu\text{g L}^{-1}$ in the lower reach (Fig. 3d). The TGD impoundment influenced the CH₄ emission rate in a trend similar to that of its dissolved concentration (see Supplementary Fig. 5).

The TGD influenced N₂O distributions both upstream and downstream of the dam, especially in the middle reach of the Yangtze (Fig. 3g). After 2003, annual averaged N₂O concentrations decreased slightly from 0.42 to 0.38 $\mu\text{g L}^{-1}$ in the wet season and from 0.55 to 0.50 $\mu\text{g L}^{-1}$ in the dry season (Fig. 3h–i). The most remarkable decrease in N₂O concentration occurred at Yichang, immediately downstream of the TGD (Supplementary Fig. 12a). At Yichang, monthly averaged N₂O emission rates fell both in the wet and dry seasons, and the amplitude of the fluctuations in N₂O emission rate also declined (Supplementary Fig. 12a) with smaller

seasonal differences (Supplementary Fig. 12b) after TGD impoundment.

GHG fluxes in response to readjustment of physical and biochemical equilibria

Our study indicated that the TGD has caused significant drops in the overall annual GHG fluxes emitted to the atmosphere and exported to the sea since 2003 (Supplementary Table 10). To interpret such changes, a whole-river analysis (Fig. 4) must be made of the readjustments to hydrodynamic conditions (Fig. 4a) and biogeochemical equilibria (Fig. 4b–d) over the broader spatiotemporal scale of the river.

Cause of CO₂ drop

Due to TGD impoundment, a backwater zone developed upstream of the dam wherein water exchanges took place between the mainstream and tributaries (Fig. 4b). Water retention time significantly increased in the reservoir in addition to the significantly decreased flow velocity ($<0.2 \text{ m s}^{-1}$) in some tributaries entering into the reservoir. Such changes replenish nutrients in the tributaries via circulation with the mainstream [33]. Accumulated nutrients and restricted vertical mixing in the backwater area of the tributaries favored phytoplankton growth [34,35], causing algae to flourish [36] (Supplementary Table 11). Algae's

photosynthetic removal of CO_2 and bioaccumulation of NO_3^- , H_2PO_4^- , HPO_4^{2-} and PO_4^{3-} resulted in a higher pH in the tributaries, promoting acceleration of eutrophication [37,38]. The higher pH in the tributaries helped neutralize hydrogen ions in the mainstream, breaking the carbonate equilibrium of the river and ultimately leading to a sharp drop in CO_2 in the mainstream (Supplementary Fig. 13).

Cause of CH_4 drop

Although CH_4 increased upstream, a net reduction of CH_4 emissions ($\sim 17\%$) happened along the whole mainstream after the TGR impoundment, due to a decrease in CH_4 downstream of the TGD. The input of dissolved CH_4 into the ocean decreased by 50%, primarily because the TGD modified the GHG regime and disrupted the biotic equilibrium of the Yangtze (Fig. 4c). Upstream of the TGD, both dissolved and emitted CH_4 increased after the reservoir impoundment, owing to the effects of flow regulation and sediment trapping. Such carbon burial promotes heterotrophic methanogenesis, thus increasing the dissolved CH_4 content of the reservoir [29]. Anoxic conditions due to increased water depth in front of the dam would also be beneficial to methanogens locally [11]. However, both dissolved and emitted CH_4 declined downstream of the dam, mainly because of riverbed scouring, which damaged the habitat of anaerobic *Archaea* responsible for heterotrophic methanogenesis [39,40]. In addition, the pre-impoundment clearance also reduced decomposition of organic carbon and inhibited the significant increase in CH_4 emissions in the TGR. During reservoir flushing, degassing would occur because of rapid depressurization and strong aeration, resulting in increased emissions of dissolved CH_4 and lowering of CH_4 concentration downstream [6,41]. Overall, the TGD regulated the CH_4 emission regime of the Yangtze, causing dissolved CH_4 to increase in the upper reach and decrease in the lower reach.

Cause of N_2O drop

N_2O flux emissions over the mainstream decreased from 0.44 to 0.41 Gg N yr^{-1} , and N_2O exports to the sea fell from 0.46 to 0.41 Gg N yr^{-1} after TGD operation commenced. Land use changes and water quality protection measures resulted in low nitrogen loading to the TGR. Formation of hypoxia or even anoxia in the reservoir was generally restricted (Fig. 4d). The promoted denitrification, whereby N_2O was transformed directly to N_2 , caused N_2O to decrease slightly upstream of the dam [42–44]. On the other hand,

riverbed scouring downstream of the TGD altered the habitat of heterotrophic denitrifiers, slowing down denitrification. This is consistent with our findings of high NO_3^- concentration but low NO_2^- concentration in the river [45] (Supplementary Figs 4 and 14a–b). Again, reservoir flushing would have raised degassing of N_2O and N_2 . Discharge of cooler, high-pressure bottom water, supersaturated with gases, from the 175-m-deep reservoir to the warmer, low-pressure downstream river would enhance N_2O emissions [14]. Riverine microbial communities require phosphorus as a nutrient, and pH to regulate nitrification and denitrification processes. The estimated annual mass of reactive phosphorus retained by dams along the Yangtze was 0.5 Gmol yr^{-1} in 2010, and it will rise to 2.9 Gmol yr^{-1} by 2030; this would alter denitrification, thus decreasing N_2O production. Hence, the influence of phosphorus is likely to be significantly less than riverbed scouring on the nitrogen cycle downstream of the TGD. Field observations also exhibited an increase in pH downstream of the TGD since 2003; this encouraged nitrification, as evidenced by the very low levels of ammonium that were recorded (Supplementary Fig. 14).

Lastly, the key concern becomes how the enlargement of CO_2 (1.8×10^2 – 3.4×10^2 Gg C yr^{-1}), CH_4 (0.18–0.37 Gg C yr^{-1}) and N_2O (0.0072–0.01 Gg N yr^{-1}) emissions caused by the reservoir itself would be finally offset by the reduction of GHG emissions resulting from downstream habitat modification. According to pre-impoundment estimates of GHG fluxes from the reservoir and post-impoundment measurements on possible GHG pathways, such a balancing out would be expected at 766–819 km (for CO_2), 124–180 km (for CH_4) and 18–53 km (for N_2O) downstream of the TGD, respectively (Fig. 5). Under the practical scenarios for TGD operation [46] (Supplementary Table 12), the overall net reduction in GHG emissions would still be significant (38.43%–44.60% for CO_2 , 14.51%–19.70% for CH_4 and 0.21%–2.50% for N_2O) in the entire Yangtze. In the reservoir area, the river-valley geomorphology restricted the rise of the littoral shallow area (< 10 m), resulting in less CH_4 and CO_2 emissions from ebullition ($< 8\%$ in the gross GHG emission estimates of the TGR, see Supplementary Table 13). Sensitivity analysis confirmed the availability of the study results under uncertainties from the models and those induced by the TGR (Supplementary Figs 15 and 16). In the balance, the net change in GHG emissions directly caused by the TGR could alter neither the dominant GHG emission pathways from the reservoir nor the general GHG reduction trend from the

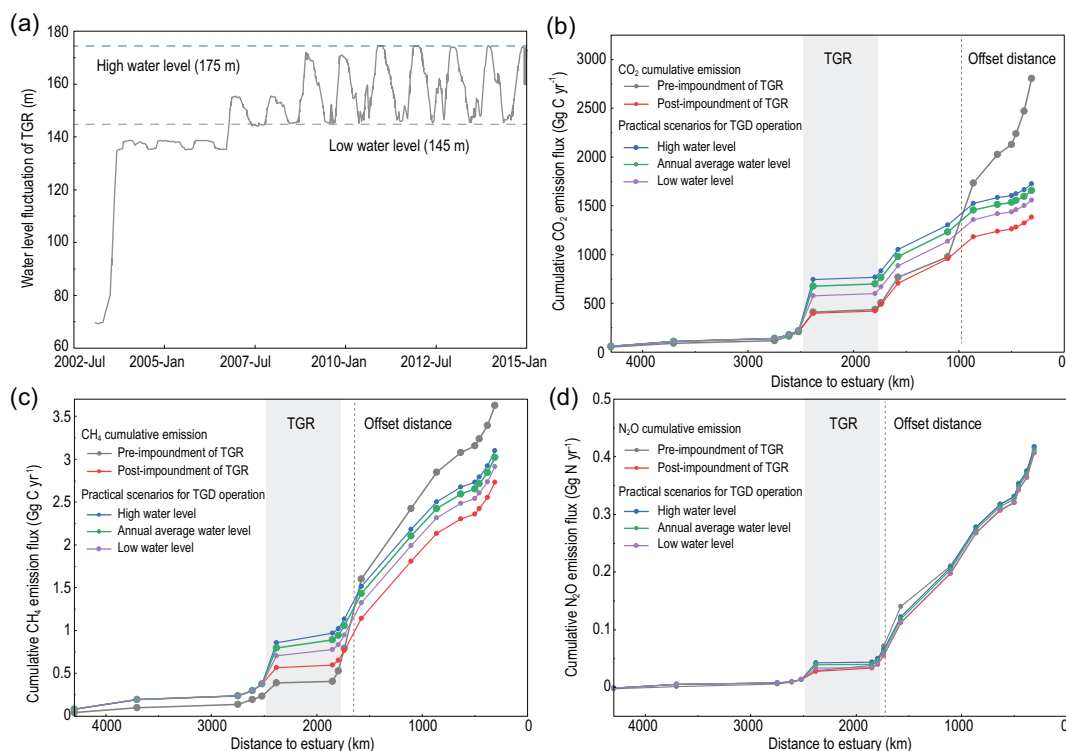


Figure 5. The balance of GHG emission fluxes enlarged by the reservoir itself and those reduced by habitat modification downstream from the dam under practical TGD operation. According to (a) different scenarios for the annual variation of the TGD’s operating water level, the offset distance was (b) 766–819 km for CO₂, (c) 124–180 km for CH₄ and (d) 18–53 km for N₂O downstream from the dam, respectively. Under the averaged operating water level, the vertical dotted lines indicate the locations where the changed GHG emission fluxes, due to the reservoir, were offset by the decreased GHG emissions in the downstream of the dam.

perspective of the full 4300 km along the mainstream of the Yangtze River (for details see Section 9 in the Supplementary Data).

CONCLUSIONS

In contrast to the general claim that dams increase emissions of GHGs from rivers, we found that the TGD, the world’s largest dam, caused a significant reduction in annual average emissions of CO₂, CH₄ and N₂O over a 4300-km stretch of the Yangtze River. Meanwhile, a remarkable drop occurred in the annual export of CO₂ (79%), CH₄ (50%) and N₂O (9%) to the sea from the river. These findings suggest that more profound impacts are produced by the ‘large dams’ than are expected from ‘small dams’, whose effects are limited to the vicinity of reservoirs, either spatially or temporally. The impoundment of a large reservoir not only altered the environment in the reservoir area, but also resulted in significant changes to riverine habitats downstream. In particular, long-term and long-distance riverbed erosion downstream of the large dam essentially changes the processes of photosynthesis, methanogenesis and denitrification, commencing the re-establishment of

the biogeochemical equilibrium over the whole river system. This highlights the primary importance of whole-system analysis in understanding the complex effects of large dams on readjustments of physical, chemical and biological equilibria in large rivers globally.

METHODS

Water quality was monitored monthly at 43 hydrological stations (blue open circles, Fig. 1). Simultaneous sampling of hydrological, environmental and all GHG constituents was undertaken in the spring and autumn of 2014 along the 4300-km stretch (i.e. the actual sinuous channel length, equivalent to 2.05 times the straight-line distance of 2102 km from the start to the end of the sampling sites; red circles, Fig. 1). Further monthly sampling took place from November 2014 to September 2015 at six stations (purple solid circles, Fig. 1). Given the limited data available for model establishment (Supplementary Tables 2–3), we included data from previous studies conducted at certain sites along the Yangtze River. Details of model verification are given in Supplementary Tables 14 and 15. All samples were

collected in triplicate. Dissolved CO₂, CH₄ and N₂O were determined using the headspace equilibration technique [47]. CO₂, CH₄ and N₂O emission rates were measured using the static floating chamber technique [47,48]. CO₂, CH₄ and N₂O concentrations were obtained using a gas chromatograph.

Water chemistry monitoring was conducted by the Changjiang Water Resources Commission on a monthly basis from 1990 to 2015. pH, total alkalinity, HCO₃⁻, water temperature (T), pCO₂ and dissolved CO₂ concentrations were determined at 18 stations (Supplementary Table 16). As described in Supplementary Figs 17 and 18, ANNs based on backward propagation were used to calculate dissolved CH₄ (with inputs of chemical oxygen demand, dissolved oxygen, water temperature, pH, NO₃⁻ and NH₄⁺) and N₂O (with inputs of NH₄⁺, NO₂⁻, NO₃⁻, dissolved oxygen, water temperature and pH). The model validation of dissolved CH₄ and N₂O concentrations (including data from previous studies conducted at certain sites along the Yangtze River) is shown in Supplementary Figs 19 and 20. Sensitivity analysis was performed by changing input variables (Supplementary Figs 15 and 16). For comparison, calculated dissolved N₂O concentrations from previous regression models are listed in Supplementary Table 17. The GHG emission rate across the air–water interface was calculated using a two-layer diffusive gas exchange model [49]. Herein, *k*₆₀₀ is an important parameter for calculating the gas emission rate from the dissolved gas concentration. Based on the re-examination of existing empirical formulas for *k*₆₀₀ (Supplementary Table 18), *k*₆₀₀ was determined for the monitoring sites at different reaches of the Yangtze River (Supplementary Table 19). Wind speed data near the hydrological stations were extracted from the China Meteorological Data Sharing Service System (<http://data.cma.gov.cn>). The atmospheric CH₄ concentration was assumed to be equivalent to the monthly averaged global background concentration at six monitoring stations across the world (NOAA/CMDL/CCGG air sampling network, <http://www.cmdl.noaa.gov/>). Model validation and parameter (e.g. *k*₆₀₀) determination are detailed in the Supplementary Data.

SUPPLEMENTARY DATA

Supplementary data are available at [NSR](#) online.

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AUTHOR CONTRIBUTIONS

J.R.N. designed the research. J.R.N., H.Z.W., T.M. and R.H. performed the research. H.Z.W., T.M., R.H., Z.L. and J.F.C. analyzed the data. P.C., A.G.L.B., Y.Y., B.L., Y.C.W., M.S.Z. and T.W. contributed new ideas and information. J.R.N., H.Z.W., Z.L. and Y.Y. wrote the paper with help from A.G.L.B. and P.C. All authors read, commented on and approved the final version of this article.

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