Response to Associate Editor Decision: Reconsider after major revisions (03 Oct 2018) by David Butman
Comments to the Author:
1. After reviewing all of the interactive comments on bg-2018-227, it is recommended that the authors return to the calculated annual emissions and possibly remove this from the discussion. As outlined from all comments, the appropriate data sets may not exists to provide a meaningful annual emission estimate given the restricted sampling period. The discussion could be limited to monsoonal periods only. If the authors would like to keep this in, it is suggested that they develop a stronger presentation on what changes across seasons related to gas transfer and concentrations.

Response: Thank you for your helpful comment. According to your suggestion, annual emission estimate was deleted and CO_2 evasion upscaling was conducted during the monsoonal period in main text as follows.

"We used our measured CO_2 emission rates by FCs for upscaling flux estimates during monsoonal period given the sampling in this period and it was found to be 0.70 TgCO_2 for all rivers sampled in our study (Table 3a). The estimated emission in the monsoonal period was close to that of the revised model (0.71 ± 0.66 (95% confidence interval: 0.46-0.94) Tg CO_2), and using the determined k average, i.e., 0.69 ± 0.65 (95% confidence interval: 0.45-0.93) Tg CO_2, but slightly higher than the estimation using water-depth based model (0.54 ± 0.51 Tg CO_2) and Alin's model (0.53 ± 0.50 Tg CO_2) (Table 3b). The higher emission, i.e., 1.66 ± 1.55 (1.08-2.23) Tg CO_2,in the monsoonal period only using flow velocity based model may be over-estimated when compared to other models, flux from determined k (Table 3b) and previous annual estimates, i.e., our earlier annual evasion of 0.64-2.33 Tg CO_2/y using TBL on the TGR river networks (Li *et al.*, 2018). Moreover, our estimated CO_2 emission in the monsoonal period also suggests that CO_2 annual emissions from rivers and streams in this area were previously underestimated, i.e., 0.03 Tg CO_2/y (Li *et al.*, 2017) and 0.37-0.44 Tg CO_2/y (Yang *et al.*, 2013) as the former used TBL model with a lower k level, and the latter employed floating chambers, but they both sampled very limited tributaries (i.e., 2-3 rivers). Therefore, measurements of k must be made mandatory along with pCO_2 measurement in the river and stream studies." (Last paragraph in section 4.3)

Further, we strongly discussed the monsoonal sampling effects on gas transfer and pCO_2 concentrations, as well as annual evasion in the SOM. This could help do the comparison with other studies.

Seasonal changes related to gas transfer and concentrations

We used our measured CO_2 emission rates by FCs for upscaling flux estimates and it was found to be 1.39 TgCO_2/y for all rivers sampled (Table S3a). The estimated emission was close to that of the revised model (1.40 ± 1.31 (95% confidence interval: 0.91-1.87) Tg CO_2/y), and using the determined k average, i.e., 1.37 ± 1.28 (95% confidence interval: 0.89-1.84) Tg CO_2/y, but slightly higher than the estimation using water-depth based model (1.08 ± 1.01 Tg CO_2/y) and Alin's model (1.06 ± 1.00 Tg CO_2/y) (Table S3b). The estimate was within the range of our earlier work using TBL on the TGR river networks (0.64-2.33 Tg CO_2/y) (Li *et al.*, 2018). The higher emission, i.e., 3.29 ± 3.08 (2.15-4.43) Tg CO_2/y, using flow velocity based model may be over-estimated (Table 3b). Therefore, this study suggests that CO_2 emissions from rivers and streams in this area may be underestimated, i.e., 0.03 Tg CO_2/y (Li *et al.*, 2017) and 0.37-0.44 Tg CO_2/y (Yang *et al.*, 2013) as the former used TBL model with a lower k level, and the latter employed floating chambers, but they both sampled very limited tributaries (i.e., 2-3 rivers). Therefore, measurements of k must be made mandatory along with pCO_2 measurement in the river and stream studies.

As our sampling was limited to monsoonal periods only, which could not provide a meaningful annual emission estimate given the restricted sampling period. Thus, we developed a stronger discussion on what changes across seasons related to gas transfer and pCO_2 concentrations. As outlined in the main text, riverine pCO_2 in the monsoonal season in this region was 81% the level in the dry season, and current velocity was 1.7-fold higher in monsoonal season (Li *et al.*, 2018), thus k_{600} was 1.6-fold higher in the monsoonal period based our model. This could be defensible due to that prior study on the Yellow River reported that k600 level in the wet season was 1.8-fold higher than in the dry season (Ran *et al.*, 2015), another study on the Wuding River demonstrated that k level in the wet season was 83%-130% of that in the dry season (Ran *et al.*, 2017). Moreover, a factor of 1.4 for water area was designated based on other monsoonal rivers. Then annual emission could be estimated at 1.59 Tg CO_2/y, slightly higher than the estimation using the data in the monsoonal period only.

Table S3. CO_2 emission from total rivers sampled in the study.

(a) Upscaling using CO_2 areal flux by FC.

	Catchment Area km^2	Water surface km^2	CO_2 areal flux mmol/m^2/d	CO_2 emission Tg CO_2/y
Daning	4200	21.42	122.0 ±239.4	0.042
Qijiang	4400	30.8	50.3 ±177.2	0.025
TGR river	50000	377.78	217.7 ±334.7	1.321
Total				1.39

(b) Upscaling using determined k_{600} average and models (whole dataset are used here).

	From determined k_{600} mean	Flow velocity-based model (Fig. 4b) (numbers in bracket is from the revised model; Fig. 4c)	Water depth-based model (Fig. 4a)	Alin's model

Mean		1.37	3.29 (1.40)	1.08	1.06
S.D.		1.28	3.08 (1.31)	1.01	1.00
95% CI for Mean	Lower Bound	0.89	2.15 (0.91)	0.71	0.69
	Upper Bound	1.84	4.43 (1.87)	1.46	1.43

A total water area of approx. 430 km^2 for all tributaries (water area is from Landsat ETM+ in 2015).

Table S4. Monsoonal sampling effects on annual emission

	Monsoonal season	Dry season	Monsoonal/Dry
pCO_2 (μatm)	846	1043	0.81
ΔpCO_2 (μatm)	446	643	0.69
k_{600} (cm/h)	48.4	31.3	1.55
CO_2 areal flux ($mmol/m^2/d$)	196.9	183.4	
Water area (km^2)	602	430	
Emission (Tg CO_2)	0.96	0.63	

2. Furthermore, it is recommended that the authors provide a stronger discussion of the bias that may occur using static chambers in a river environment. There are estimates of this bias in the literature and those values should be referenced. This method is concerning, unless in very small streams with significant turbulence induced from both surface and subsurface features.

Response: We presented strong discussion (please refer to section 4.4), and the following text was added.

Efforts have been devoted to measurement techniques (comparison of FC, eddy covariance-EC and boundary layer model-BLM) for improving CO_2 quantification from rivers because of a notable contribution of inland waters to the global C budget (), which could have a large effect on the magnitude of the terrestrial C sink. Prior studies reported inconsistent trends of CO_2 area flux by these methods. For instance, CO2 areal flux from FC was much lower than EC (Podgrajsek et al., 2014), while areal flux from FC was higher than both EC and BLM elsewhere (Erkkilä et al., 2018), however, Schilder et al (2013) demonstrated that areal flux from BLM was 33-320% of in-situ FC measurements. Albeit unsatisfied errors of varied techniques and additional perturbations from FC exist, FC method is currently a simple and preferred measurement for CO_2 flux because that choosing a right k value remained a major challenge and others require high workloads (Martinsen et al., 2018).

Recent study further reported fundamental differences in CO_2 emission rates between ACs and freely DFs (Lorke *et al.*, 2015), i.e., ACs biased the gas areal flux higher by a factor of 2.0-5.5. However, some studies observed that ACs showed reasonable agreement with other flux measurement techniques (Galfalk *et al.*, 2013), and this straightforward, inexpensive and relatively simple method AC was widely used (Ran *et al.*, 2017). Water-air interface CO_2 flux measurements were primarily made using ACs in our studied streams and small rivers because of relatively high current velocity; otherwise, floating chambers will travel far during the measurement period. In addition, inflatable rings were used for sealing the chamber headspace and submergence of ACs was minimal, therefore, our measurements were potentially overestimated but reasonable. We could not test the overestimation of ACs in this study, the modified FCs, i.e., DCs and integration of ACs and DCs, and multi-method comparison study including FCs, ECs and BLM should be conducted for a reliable chamber method.

3. Furthermore, the inclusion of data that is not relevent to the calculation and interpretation of k and CO2 emissions was identified as distracting. This is in reference to the DOC/TN/TP. As outlined in two of the reviews, either identify why these are included (DOC is useful to know that you are not overestimating Alkalinity from organic acids) or remove from the manuscript.

Response: The parts of TN and TP were removed.

4. This effort will provide new knowledge and data from understudied rivers in SE Asia. That alone is a strong contribution. After addressing in detail these and the reviewer comments, this would be suitable for publication.

Response: Thanks for your very positive comment.

END of Review

Response to Anonymous Referee #1

1. General comments: The paper "Gas transfer velocities of CO2 in subtropical monsoonal climate streams and small rivers" appears to be something of a companion piece to "Riverine CO2 supersaturation and outgassing in a subtropical monsoonal mountainous area (Three Gorges Reservoir Region) of China" https://doi.org/10.1016/j.jhydrol.2018.01.057 published in the Journal of Hydrology. In this current submission, the authors present k calculated from floating chamber flux measurements and using models, and discuss the implications of the differing approaches to k for making regional scale flux estimates. Using chambers to determine CO2 fluxes, the authors then use pCO2 to derive the gas transfer velocity. These flux-derived k values are compared to modeled k values. It is good to see the spatial aspects of the gas transfer velocity addressed. However, I do not feel that there is an adequate consideration of the uncertainty in the estimates/calculations provided. For the flux-derived k values, there is little provided in terms of uncertainty assessments.

Response: We thanked the referee for the comment. In our previous article, we studied the *p*CO_2 and emission rate as well as their controls from fluvial networks in the TGR area, which is based on two field works in the TGR region, and the diffusive models from other studies were used. In this study, we attempted to derive k levels and develop the gas transfer model in this area (mountainous streams and small rivers) for more accurate quantification of CO_2 areal flux, and also to serve for the fluvial networks in the Yangtze River or others with similar hydrology and geomorphology. In addition, we did more detailed field study in the two contrasting rivers Daning and Qijiang for developing models (see the sampling locations map). This study clearly showed original contribution to the current literature and this study is different than the article published in the Journal of Hydrology. We clearly state the new contributions and significances in the last paragraph of the "Introduction" as follows.

"Our recent study preliminarily investigated *p*CO_2 and air – water CO_2 areal flux as well as their controls from fluvial networks in the Three Gorges Reservoir (TGR) area (Li *et al.*, 2018). The past study was based on two field works, and the diffusive models from other continents were used. In this study, we attempted to derive k levels and develop the gas transfer model in this area (mountainous streams and small rivers) for more accurate quantification of CO_2 areal flux, and also to serve for the fluvial networks in the Yangtze River or others with similar hydrology and geomorphology. Moreover, we did detailed field campaigns in the two contrasting rivers Daning and Qijiang for models (Fig. 1). The study thus clearly stated distinct differences than the previous study (Li *et al.*, 2018) by the new contributions of specific objectives and data supplements, as well as wider significance."

We added a section (4.4.) for "**Uncertainty assessment of *p*CO_2 and flux-derived k values**" in the part of "Discussion".

The uncertainty of flux-derived k values mainly stem from air–water gradient of CO_2 (ΔpCO_2 in ppm) and flux measurements (Golub et al., 2017; Lorke et al., 2015; Bodmer et al., 2016). Thus we provided uncertainty assessments caused by dominant sources of uncertainty from measurements of aquatic pCO_2 and CO_2 areal flux since uncertainty of atmospheric CO_2 measurement could be neglected.

In our study, aquatic pCO_2 was computed based on pH, alkalinity and water temperature rather than directly measured. Recent studies highlighted pCO_2 uncertainty caused by systematic errors over empiric random errors (Golub et al., 2017). Systematic errors are mainly attributed to instrument limitations, i.e., sondes of pH and water temperature. The relative accuracy of temperature meters was ±0.1 ^{0}C according to manufacturers' specifications, thus the uncertainty of water T propagated on uncertainty in pCO_2 was minor (Golub et al., 2017). Systematic errors therefore stem from pH, which has been proved to be a key parameter for biased pCO_2 estimation calculated from aquatic C system (Li et al., 2013; Abril et al., 2015). We used a high accuracy of pH electrode and the pH meters were carefully calibrated using CRMs, and in situ measurements showed an uncertainty of ±0.01. We then run an uncertainty of ±0.01 pH to quantify the pCO_2 uncertainty, and an uncertainty of ±3% was observed. Systematic errors thus seemed to show little effects on pCO_2 errors in our study.

Random errors are from repeatability of carbonate measurements. Two replicates for each sample showed the uncertainty of within ±5%, indicating that uncertainty in pCO_2 calculation from alkalinity measurements could be minor.

The measured pH ranges also exhibited great effects on pCO_2 uncertainty (Hunt et al., 2011; Abril et al., 2014). At low pH, pCO_2 can be overestimated when calculated from pH and alkalinity (Abril et al., 2014). Samples for CO_2 fluxes estimated from pH and alkalinity showed pH average of 8.39±0.29 (median 8.46 with quartiles of 8.24-8.56) (n=115). Thus, overestimation of calculated CO_2 areal flux from pH and alkalinity is likely to be minor. Further, contribution of organic matter to non-carbonate alkalinity is likely to be neglected because of low DOC (mean 6.67 mg/L; median 2.51 mg/L) (Hunt et al., 2011; Li et al., 2013).

Efforts have been devoted to measurement techniques (comparison of FC, eddy covariance-EC and boundary layer model-BLM) for improving CO_2 quantification from rivers because of a notable contribution of inland waters to the global C budget (), which could have a large effect on the magnitude of the terrestrial C sink. Prior studies reported inconsistent trends of CO_2 area flux by these methods. For instance, CO2 areal flux from FC was much lower than EC (Podgrajsek *et al.*, 2014), while areal flux from FC was higher than both EC and BLM elsewhere (Erkkila *et al.*, 2018), however, Schilder et al (Schilder *et al.*, 2013) demonstrated that areal flux from BLM was 33-320% of in-situ FC measurements. Albeit unsatisfied errors of varied techniques and additional perturbations from FC exist, FC method is currently a simple and preferred measurement for CO_2 flux because that choosing a right k value remains a major challenge and others require high workloads (Martinsen *et al.*, 2018).

Recent study further reported fundamental differences in CO_2 emission rates between ACs and freely DFs (Lorke *et al.*, 2015), i.e., ACs biased the gas areal flux higher by a factor of 2.0-5.5. However, some studies observed that ACs showed reasonable agreement with other flux measurement techniques (Galfalk *et al.*, 2013), and this straightforward, inexpensive and relatively simple method AC was widely used (Ran *et al.*, 2017). Water-air interface CO_2 flux measurements were primarily made using ACs in our studied streams and small rivers because of relatively high current velocity; otherwise, floating chambers will travel far during the measurement period. In addition, inflatable rings were used for sealing the chamber headspace and submergence of ACs was minimal, therefore, our measurements were potentially overestimated but reasonable. We could not test the overestimation of ACs in this study, the modified FCs, i.e., DCs and integration of ACs and DCs, and multi-method comparison study including FCs, ECs and BLM should be conducted for a reliable chamber method.

Sampling seasonality considerably regulated riverine pCO_2 and gas transfer velocity and thus water-air interface CO_2 evasion rate (Li *et al.*, 2012; Ran *et al.*, 2015). We sampled waters in wet season (monsoonal period) due to that it showed wider range of flow velocity and thus it covered the k_{600} levels in the whole hydrological season. Wet season generally had higher current velocity and thus higher gas transfer velocity (Ran *et al.*, 2015), while aquatic pCO_2 was variable with seasonality. We recently reported that riverine pCO_2 in the wet season was 81% the level in the dry season (Li *et al.*, 2018), and prior study on the Yellow River reported that k level in the wet season was 1.8-fold higher than in the dry season (Ran *et al.*, 2015), while another study on the Wuding River demonstrated that k level in the wet season was 83%-130% of that in the dry season (Ran *et al.*, 2017). Thus, we acknowledged a certain amount of errors on the annual flux estimation from sampling campaigns during the wet season in the TGR area, while this uncertainty could not be significant because that the diluted pCO_2 could alleviate the overestimated emission by increased k level in the wet season (stronger discussion please refer to SOM).

2. pCO2 was not measured, but rather was computed based on pH, alkalinity and temperature. This would have large uncertainties that then propagate into k estimates. Golub et al. (2017, doi: 10.1002/2017JG003794) note that "freshwater researchers must make significant efforts to standardize and reduce errors in pCO2 predictions". I encourage the authors to undertake a more systematic uncertainty analysis for their pCO2 values and propagate this error into uncertainty estimates for k.

Response: Thanks for the comment. We reported the quality control such as systematic errors and random errors of pH and alkalinity, water temperature, as well as non-carbonate alkalinity effects. Please refer to the response above.

3. Further, the authors here excluded deriving k values for samples that did not have a very large gradient in CO2 across the air-water interface. The authors chose 110 uatm as the threshold for excluding data, but this was presented without any indication of choice of threshold, making it appear rather arbitrary. Given the pH of the rivers sampled and the pCO2 that was at times undersaturated, this appears rather problematic *in that it introduces bias that carries through to the regional estimates provided.*

Response: We addressed this issue as follows.
"Prior to statistical analysis, we excluded k_{600} data for samples with the air-water pCO$_2$ gradient <110 µatm, since the error in the k_{600} calculations drastically enhances when $\triangle p$CO$_2$ approaches zero (Borges et al., 2004), and datasets with $\triangle p$CO$_2$ >110 µatm provide an error of <10% on k_{600} computation (see the Fig. as follows)"

The additional section 4.4 **Uncertainty assessment of pCO$_2$ and flux-derived k values** included uncertainty of pH and scaling-up estimation, for example, effects of chemical enhancement for quite high pH values.

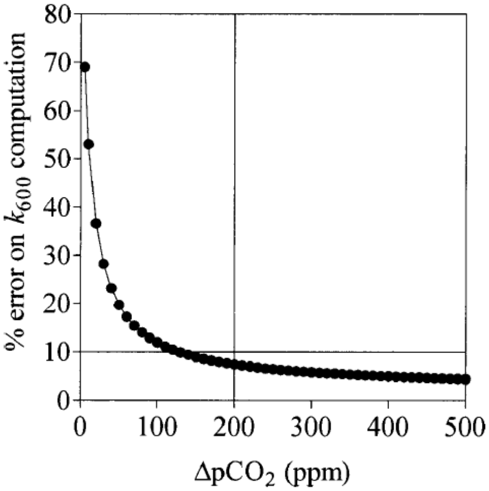

Figure Theoretical error (±%) on the computation of the gas transfer velocity of CO$_2$ (k_{600}) as a function of the air–water gradient of CO$_2$ (ΔpCO$_2$ in ppm), assuming a constant uncertainty on ΔpCO$_2$ of ±3% (Borges et al., 2004).

Borges, A.V., Delille, B., Schiettecatte, L.S., Gazeau, F., Abril, G., Frankignoulle, M., 2004a. Gas transfer velocities of CO2 in three European estuaries (Randers Fjord, Scheldt, and Thames). Limnology & Oceanography 49, 1630-1641.

4. The authors in this paper refer to their k values as "observed", but these are in fact derived, and so need to have uncertainty better characterized. Upscaling from X floating chamber measurements to a river network draining 58000 km2. How many flux measurements were made with floating chambers is not clearly stated, but it appears to be about 100 all made during summer 2016. Going from summer measurements for 100 points to annual estimates for 58000 km2 also requires some consideration of error propagation and bias. Fluxes were only retained when the floating chambers yielded linearly increasing CO2 against time, which again biases against low flux locations.

Response: We have changed "observed, or measured" to "flux-derived or derived", and discussed the uncertainty of k values as mentioned above. We agreed that more samples could improve the CO$_2$ estimates, while our sampling locations were much more or at least comparable to the other publications.

A total of 115 discrete grab samples were collected (each sample consisted of three replicates). Floating chambers with replicates were deployed in 101 sites (32 sampling sites in Daning, 37 sites in TGR river networks and 32 sites in Qijiang). The sampling period covers spring and summer season, our sampling points are reasonable considering a water area of 433 km^2. For example, 16 sites were collected for Yangtze system to examine hydrological and geomorphological controls on pCO$_2$ (i.e., Liu et al., 2017), and 17 sites for dynamic biogeochemical controls on riverine pCO$_2$ in the Yangtze basin (Liu et al., 2016, Global Biogeochemical Cycles).

In our sampling points, all measured fluxes were retained since the floating chambers yielded linearly increasing CO$_2$ against time following manufacturer' specification.

Liu, S., Lu, X.X., Xia, X., Yang, X., Ran, L., 2017. Hydrological and geomorphological control on CO2 outgassing from low-gradient large rivers: An example of the Yangtze River system. Journal of Hydrology 550, 26-41.

5. Of the attempted flux measurements, what fraction was discarded?

Response: We revised as follows.
"Prior to statistical analysis, we excluded k$_{600}$ data for samples with the air-water pCO$_2$ gradient <110 µatm, since the error in the k$_{600}$ calculations drastically enhances when △pCO$_2$ approaches zero (Borges *et al.*, 2004; Alin *et al.*, 2011), and datasets with △pCO$_2$ >110 µatm provide an error of <10% on k$_{600}$ computation. Thus, we discarded the samples (36.7% of sampling points with flux measurements) with △pCO$_2$ <110 µatm for k$_{600}$ model development, while for the flux estimations from diffusive model and floating chambers, all samples were included."

6. Finally, a minor point is that the authors state several times that theirs is the first determination of k for subtropical streams and small rivers. I would point the authors towards global syntheses on CO2 evasion as well as individual studies that include k estimates.

Response: We agree the comment, and addressed this issue. Several sentences for implication of k determination and comparison with other k studies were added in the part of Discussion. We also re-organised and added sub-headings of "Discussion".

"4.1. Determined k values relative to world rivers; 4.2. Hydraulic controls of k600; 4.3. Implications for large scale estimation; 4.4. Uncertainty assessment of pCO$_2$ and flux-derived k values"

Minor comments

7. Finally, a minor point is that the authors state several times that theirs is the first determination of k for subtropical streams and small rivers. I would point the authors towards global syntheses on CO2 evasion as well as individual studies that include k *estimates.*

Response: We have presented k levels related to global rivers (please refer to 4.1) and implications of k for large scale estimation (see section 4.3).

8. The figure S1 does not show the sample locations within the Daning or Qiijiang basins. These may be the same locations as Figure 1 in Li et al. (2018) Journal of Hydrology doi: 10.1016/j.jhydrol.2018.01.057?

Response: The sampling sites and study aims are different than previous study (Please refer to the section of 2.1 and the last paragraph in the "Introduction").
In the revised Ms, we supplied the map of sampling locations in the main text as Fig. 1.

We added several sentences in the section of "INTRODUCTION" to highlight the differences between our study and previous study, as well as what is advanced by this study (please refer to the first Comment).

9. There are a number of grammatical issues throughout the paper that the authors should address.

Response: We carefully edited English, and also get helps from a native English scientist.

The additional Table was added to the Table S2 in SOM.

	Current velocity m/s	Water depth m/s	Wind speed m/s	k_{600} cm/h	Reference
Mekong tributary	0.39 ±0.28	0.9 ±0.6	0.7 ±0.6	23.3 ±17.3	Alin et al., 2011
Yellow	1.8		1.8 (1.2-2.3)	42 ±17	Ran et al., 2015
Yangtze	1.2 ±1.5		1.2 ±1.1	38 ±40	Liu et al., 2017
Mekong stem	0.92 ±0.42		1.8 ±1.2	15 ±9	Alin et al., 2011

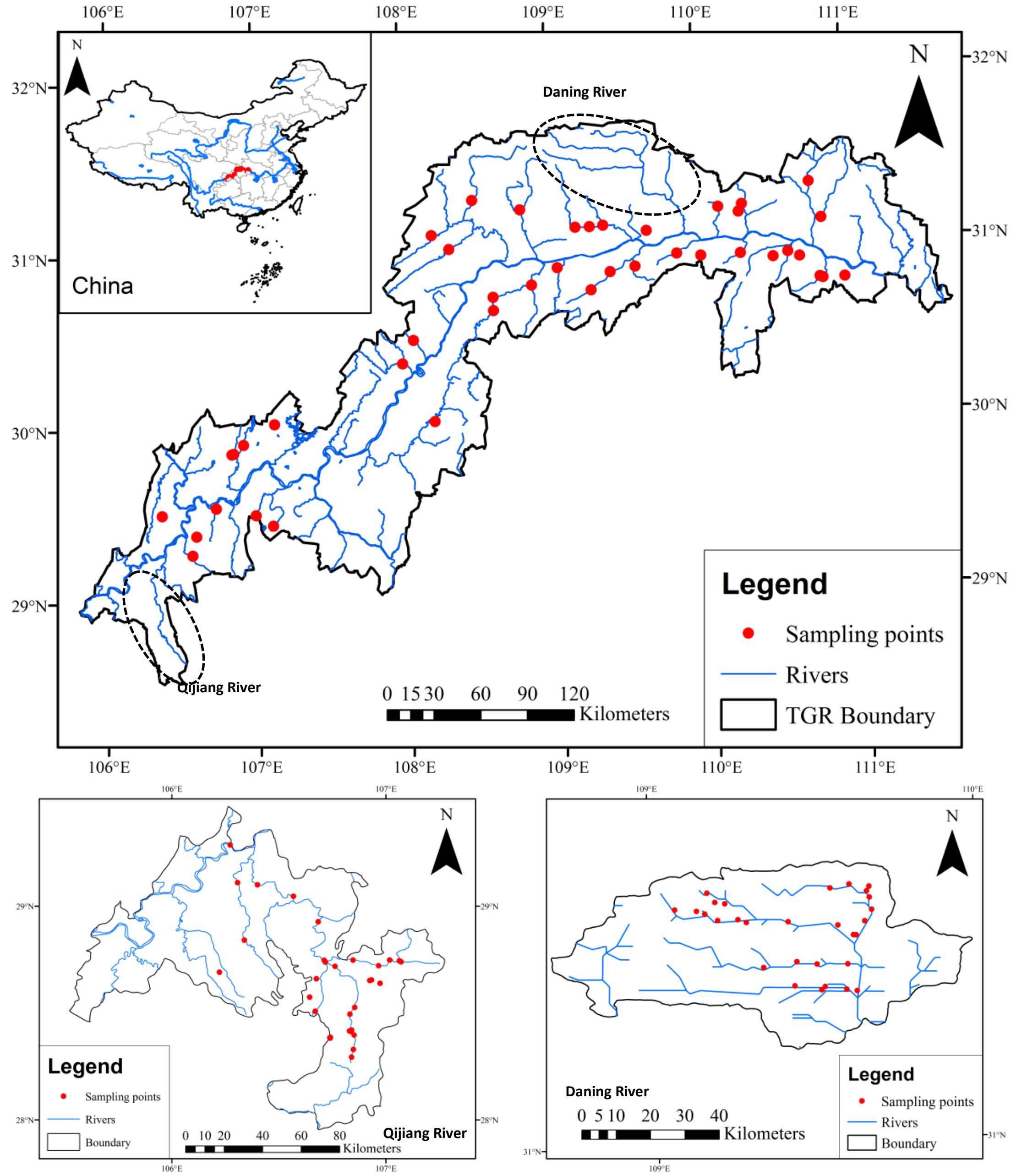

Fig. 1. Map of sampling locations of major rivers and streams in the Three Gorges Reservoir region, China (main text).

Response to Anonymous Referee #2

The manuscript reports on transfer velocities of CO2 (K) in streams and small rivers for assessing the gas fluxes. CO2 released from lakes and rivers has been recently recognized as an important component in the global carbon cycle. The accurate estimation of CO2 flux is still challenging primarily due to the difficulty in obtaining an appropriate K value. The topic would be of great interest for the community of scientists working on carbon cycles and can be considered for publication. However, the current version need to revised (see below).

Response: We thanked the referee for the positive comment. We revised the Ms based on the comments and suggestions as follows.

General comments

1. As emphasized by the authors, the study focuses on the subtropical monsoonal streams and small rivers which are characterized by large seasonal variations in climate and discharge. Hence, the K value in these rivers should also have obvious seasonality. Unfortunately, the samples presented in this study were collected in the rainy season. The K value were calculated based on the one-time sampling campaign, which might result in a certain amount of errors on the annual flux estimation. Regarding this, the uncertainty of the sampling data and the calculations, as well as the reliability of the argument should be sufficiently discussed.

Response: We agreed the comment. Sampling seasonality largely impacted riverine $p\mathrm{CO}_2$ and gas transfer velocity and thus water-air interface CO_2 evasion rate. In our Ms, we sampled waters in the rainy season due to that it showed wider range of flow velocity and thus rainy season covered the k levels in the whole hydrological season. Rainy season generally had higher current velocity and thus higher gas transfer velocity, while aquatic $p\mathrm{CO}_2$ was variable with seasonality. Thus, we estimated CO2 emission in monsoonal period instead of annual emission (please refer to section 4.3).

We added a section of "**4.4. Uncertainty assessment of $p\mathrm{CO}_2$ and flux-derived k values**" in the part of "Discussion". In this section, effects and uncertainty of sampling seasonality on errors of annual CO_2 flux estimation were included.

Sampling seasonality considerably regulated riverine $p\mathrm{CO}_2$ and gas transfer velocity and thus water-air interface CO_2 evasion rate (Li *et al.*, 2012; Ran *et al.*, 2015). We sampled waters in wet season (monsoonal period) due to that it showed wider range of flow velocity and thus it covered the k_{600} levels in the whole hydrological season. Wet season generally had higher current velocity and thus higher gas transfer velocity (Ran *et al.*, 2015), while aquatic $p\mathrm{CO}_2$ was variable with seasonality. We recently reported that riverine $p\mathrm{CO}_2$ in the wet season was 81% the level in the dry season (Li *et al.*, 2018), and prior study on the Yellow River reported that k level in the wet season was 1.8-fold higher than in the dry season (Ran *et al.*, 2015), while another study on the Wuding River demonstrated that k level in the wet season was 83%-130% of that in the dry season (Ran *et al.*, 2017). Thus, we acknowledged a certain amount of errors on the annual flux estimation from sampling campaigns during the wet season in the TGR area, while this uncertainty could not be significant because that the diluted pCO_2 could alleviate the overestimated emission by increased k level in the wet season (stronger discussion please refer to SOM).

2. In my point of view, the variation in K value of the rivers studied are obvious and need to be discussed. In addition, the spatial difference of K values is only sorted out for the three river systems (Daning, Qijiang and TGR). I would suggest the authors examine the variations of K following the physical characteristics of rivers (such as the current velocity, slope and the water depth) or/and the river orders.

Response: Spatial differences of k values were discussed for the three rivers systems (please refer to section 4.2). We discussed the controls of physical characteristics of current velocity, slope and the water depth, while river orders were not extracted.

"This could substantiate the higher k_{600} levels and spatial changes in k_{600} values of our three river systems. For instance, similar to other turbulent rivers in China (Ran *et al.*, 2015; Ran *et al.*, 2017), high k_{600} values in the TGR, Daning and Qjiang rivers were due to mountainous terrain catchment, high current velocity (10 – 150 cm/s) (Fig. 4b), bottom roughness, and shallow water depth (10 - 150 cm) (Fig. 4a). It has been suggested that shallow water enhances bottom shear, and the resultant turbulence increases k values (Alin *et al.*, 2011; Raymond *et al.*, 2012). These physical controls are highly variable across environmental types (Figs. 4a and 4b), hence, k values are expected to vary widely (Fig. 3). The k_{600} values in the TGR rivers showed wider range (1-177 cm/h; Fig. 3; Table S1), spanning more than 2 orders of magnitude across the region, and it is consistent with the considerable variability in the physical processes on water turbulence across environmental settings. Similar broad range of k_{600} levels was also observed in the China's Yellow basin (ca. 0-123 cm/h) (Ran *et al.*, 2015; Ran *et al.*, 2017).

Contrary to our expectations, no significant relationship was observed between k_{600} and water depth, and current velocity using the entire data in the three (TGR, Danning and Qjiang) river systems (Fig. S4). There were not statistically significant relationships between k_{600} and wind speed using separated data or combined data, and it is consistent with earlier studies (Alin *et al.*, 2011; Raymond *et al.*, 2012). Flow velocity showed linear relation with k_{600}, and the extremely high value of k_{600} was observed during the periods of higher flow velocity (Fig. S4a) using combined data. Similar trend was also observed between water depth and k_{600} values (Fig. S4b). The lack of strong correlation between k_{600} and physical factors are probably due to combined effect of both flow velocity and water depth, as well as large diversity of channel morphology, both across and within river networks in the entire catchment (60, 000 km^2). This is further collaborated by weak correlations between k_{600} and flow velocity in the TGR rivers (Fig. 4), where one or two samples were taken for a large scale examination. k_{600} as a function of water depth was obtained in the TGR rivers, but it explained only 30% of the variance in k_{600}. However, model using data from Qijiang could explain 68% of the variance in k_{600} (Fig. 4b), and it was in line with general theory. Nonetheless, k_{600} from our flow velocity based model (Fig. 4b) was potentially largely overestimated with consideration of other measurements (Alin *et al.*, 2015; Ran *et al.*, 2015; Ran *et al.*, 2017). When several extremely values were removed, k_{600} (cm/h) was parameterized as follows (k_{600} = 62.879FV + 6.8357, R^2= 0.52, p=0.019, FV-flow velocity with a unit of m/s), and this revised model was in good agreement with the model in the river networks of the Yellow River (Ran *et al.*, 2017), but much lower than the model developed in the Yangtze system (Liu *et al.*, 2017) (Fig. 4c). This was reasonable because of k_{600} values in the Yangtze system were from large rivers with higher turbulence than Yellow and our studied rivers. Furthermore, the determined k_{600} using FCs was, on average, consistent with the revised model (Table 2). These differences in relationship between spatial changes in k_{600} values and physical characteristics further corroborated heterogeneity of channel geomorphology and hydraulic conditions across the investigated rivers."

3. The pCO2 calculated in this paper is between 50-4830ppm, which indicates that the river pCO2 value is sometimes much lower than that of the atmosphere, that is, the studied rivers can sometimes absorb CO2 from the atmosphere. It seems that the annual CO2 flux for the whole basin was calculated in this paper based on the averaged K value from the observed results using floating chamber method. The question is that is it reliable to estimate both directions of the CO2 flux at the air-water interface (including river CO2 outgassed to the atmosphere and the atmospheric CO2 input to rivers) by using the same K value? Or what uncertainty will it cause?

Response: We thanked for your critical comment. Worldwide studies reported the dependence of k on flow dynamics; while k average from statistical analysis with $\triangle p\mathrm{CO_2}$ >110 µatm was normally used in riverine CO_2 flux estimation in rivers where a broad of range of $p\mathrm{CO_2}$ occurred (Alin et al., 2011). Considering that k was largely dependent on hydraulic characteristics, this uncertainty was not discussed. The method employed here for scaling up CO_2 estimation was widely used (e.g., Alin et al., 2011). We added one section 4.4 (see Discussion part) on systematic errors and empiric random errors of $p\mathrm{CO_2}$, CO_2 areal flux and k from $p\mathrm{CO_2}$ determinations, sampling seasonality, flux measurements etc.

4. This study measured DOC, DTN, and DTP, but the authors did not mention these measurements in the discussion section. What is the relationship between these variables and the K values?

Response: k values are reported to be dominated by physical characteristics of river systems (Borges et al., 2004; Alin et al., 2011; Raymond et al., 2012). We re-wrote this part and pattern of nutrients in the "Result" section was removed.

Specific comments

5. L 94-97: I would suggest rephrase these sentences, since they cannot convey clearly the real contribution or scientific merit of this study.

Response: We revised as follows.

"Our new contributions to the literature include (1) determination and controls of k levels for small rivers and streams in subtropical areas of China, and (2) new k models developed in the subtropical mountainous river networks"

6. L 111-112, 117: The classification method of the river order used here should be clarified. The number of a river order defined by different classification system may represent different size or hierarchy of a river.

Response: We addressed this issue, and provided the map of sampling location (Fig. 1). We also supplied methodology of water areal extraction in section 2.5.

7. L 214-216: This statement is problematic. Clearly, the studied rivers are not always supersaturated reference to atmospheric CO2 as the pCO2 in rivers is between 50- 4830 uatm.

Response: We changed to "*p*CO_2 varied between 50 and 4830 µatm with mean of 846 ± 819 µatm (Table 1). There were 28.7% of samples that had *p*CO_2 levels lower than 410 µatm, while the studied rivers were overall supersaturated with reference to atmospheric CO_2 and act as a source for the atmospheric CO_2."

8. L 274-285 These arguments need more solid evidence to support. As mentioned in the general comments, I would suggest that the authors focus on discussions on relationship between spatial change in K values and physical characteristics of rivers or/ and the river orders.

Response: Spatial differences of k values were discussed for the three rivers systems (please refer to section 4.2). We discussed the controls of physical characteristics of current velocity, slope and the water depth.

9. L 497-498 The water area is a very critical parameter for the calculation of CO2 flux in a basin, so the acquisition of water area is essential and should be described more in detail. For example, what is the resolution of the satellite image? In addition, the variation of surface area of water between wet and dry seasons should be considered.

Response: We provided the information for the acquisition of water area and citation was included (please refer to Methodology section).

2.5. Estimation of river water area

Water surface is an important parameter for CO_2 efflux estimation, while it depends on its climate, channel geometry and topography. River water area therefore largely fluctuates with much higher areal extent of water surface particularly in monsoonal season. However, most studies do not consider this change, and a fraction of the drainage area is used in river water area calculation (Zhang *et al.*, 2017). In our study, a 90 m resolution SRTM DEM (Shuttle Radar Topography Mission digital elevation model) data and Landsat images in dry season were used to delineate river network, and thus water area (Zhang *et al.*, 2018), whilst, stream orders were not extracted. Water area of river systems is generally much higher in monsoonal season in comparison to dry season, for instance, Yellow River showed 1.4-fold higher water area in the wet season than in the dry season (Ran *et al.*, 2015). Available dry-season image was likely to underestimate CO_2 estimation.

Ran, L.S., Lu, X.X., Yang, H., Li, L.Y., Yu, R.H., Sun, H.G., Han, J.T., 2015. CO2 outgassing from the Yellow River network and its implications for riverine carbon cycle. Journal of Geophysical Research-Biogeosciences 120, 1334-1347.
Zhang, T., Li, J., Pu, J., Martin, J.B., Khadka, M.B., Wu, F., Li, L., Jiang, F., Huang, S., Yuan, D., 2017. River sequesters atmospheric carbon and limits the CO2 degassing in karst area, southwest China. Science of The Total Environment 609, 92-101.
Zhang, J., Li, SY., Dong, RZ., Jiang, CS., 2018. Physical evolution of the Three Gorges Reservoir in Holocene using advanced SVM on Landsat images and SRTM DEM data. Environ Sci Pollut Res 25, 14911-14918.

10. Finally, I would suggest the authors polish the English grammar and writing, as well as the figs presenting.

Response: We carefully edited the Ms and re-organized the Tables and Figures presentation.

Response to Anonymous Referee #3

General comments
The manuscript of Li et al. presents measured CO2 fluxes, transport coefficients based on CO2, and calculated pCO2 data of running waters in a subtropical monsoonal climate zone. These data are complemented by among others water chemistry parameters such as DOC, DTN, DTP, as well as hydrogeomorphology data (e.g. water depth, flow velocity). They provide data and insights about transport coefficients for a so far understudied region and highlight the spatial variability and subsequent uncertainty for regional upscale estimates.
By investigating the key parameter for CO2 flux estimates - the transport coefficient - in an understudied region, Li et al. address a very relevant topic. Narrowing down the uncertainties of regional upscaling estimates of riverine CO2 fluxes is of wide interest, hence this study would make a good contribution to the literature and the subject matter is thus of interest to Biogeosciences readers.

Response: We thank you for your overall positive comments, and accordingly revised the Ms.

However, in my opinion, the manuscript has some problems:
(1) The terminology used in this manuscript is quite confusing to me. It seems to me that "streams“, "rivers“, "river networks“ are used interchangeably (without definition and consistency), which makes it hard to follow the red line of the story. The terminology needs to be clarified and unified.

Response: Based on the delineation of river systems by Alin et al., 2011 (JGR), small rivers and streams encompass rivers with channels < 100 m. We have clarified the term in the method.

(2) The sampling design is not very clear to me. All investigated running waters seem to be in the Three Gorges Reservoir (TGR) region, but in addition two larger streams (Daning and Qijiang) were sampled. In the results and discussion, these investigated running waters are combined, sometimes split, which makes it hard to follow (in the main text and tables). In my opinion, these three "regions“ need to be presented in a unified way (always separated or combined, possibly both in each table and figure), and presented more clearly in the text.

Response: We provided both separated and combined data in Tables and Figures, p*lease refer to Figs 2 and 3, as well as Figs. S2 and S3. In addition, we also clearly stated this in the “Method” part.*

“Spatial differences (Daning, Qijiang and entire tributaries of TGR region) were tested using the nonparametric Mann Whitney U-test. Multivariate statistics, such as correlation and stepwise multiple linear regression, were performed for the models of k_{600} using potential physical parameters of wind speed, water depth, and current velocity from separated data and combined data (Alin *et al.*, 2011).**”**

(3) One of the main messages is the presentation of transport coefficients in a subtropical monsoonal climate zone, which is interesting, but I can imagine that there is a large difference in the wet and dry season. However, all the measurements were done in the wet season. I think this issue should be clearly acknowledged and discussed.

Response: We agreed your opinion and addressed this issue in an additional section 4.4. We

sampled in the monsoonal period as it covered the flow velocity in the whole hydrological year. Albeit k_{600} is higher in wet season than dry season, our main objectives are to develop models of k rather than the annual evasion. In this section, effects and uncertainty of sampling seasonality on errors of annual CO_2 flux estimation were also included.

"Sampling seasonality considerably regulated riverine $p\mathrm{CO}_2$ and gas transfer velocity and thus water-air interface CO_2 evasion rate (Li *et al.*, 2012; Ran *et al.*, 2015). We sampled waters in wet season (monsoonal period) due to that it showed wider range of flow velocity and thus it covered the k_{600} levels in the whole hydrological season. Wet season generally had higher current velocity and thus higher gas transfer velocity (Ran *et al.*, 2015), while aquatic $p\mathrm{CO}_2$ was variable with seasonality. We recently reported that riverine $p\mathrm{CO}_2$ in the wet season was 81% the level in the dry season (Li *et al.*, 2018), and prior study on the Yellow River reported that k level in the wet season was 1.8-fold higher than in the dry season (Ran *et al.*, 2015), while another study on the Wuding River demonstrated that k level in the wet season was 83%-130% of that in the dry season (Ran *et al.*, 2017). Thus, we acknowledged a certain amount of errors on the annual flux estimation from sampling campaigns during the wet season in the TGR area, while this uncertainty could not be significant because that the diluted $p\mathrm{CO}_2$ could alleviate the overestimated emission by increased k level in the wet season (stronger discussion please refer to SOM)."

(4) There are two technical issues: (i) The measurements with the floating chambers are poorly described. The only information Li et al. provide is that the floating chambers were "deployed". If the flux measurements are done in an anchored or free floating manner is critical (see e.g. Lorke, A., Bodmer, P., Noss, C., Alshboul, Z., Koschorreck, M., Somlai-Haase, C., Bastviken, D., Flury, S., McGinnis, D. F., Maeck, A., Müller, D., and Premke, K.: Technical note: drifting versus anchored flux chambers for measuring greenhouse gas emissions from running waters, Biogeosciences, 12, 7013-7024, https://doi.org/10.5194/bg-12-7013-2015, 2015.). Hence, this issue needs to be addressed clearly. (ii) It seems that all the flux and pCO2 measurements were done distributed during the day. The fact that there is a diurnal cycle of CO2 was not considered (see e.g. Pascal Bodmer, Marlen Heinz, Martin Pusch, Gabriel Singer, Katrin Premke, Carbon dynamics and their link to dissolved organic matter quality across contrasting stream ecosystems, Science of The Total Environment, Volume 553, 2016, Pages 574-586, ISSN 0048-9697, https://doi.org/10.1016/j.scitotenv.2016.02.095.), and values directly compared. This issue should at least be discussed.

Response: A new section of "4.4. Uncertainty assessment of $p\mathrm{CO}_2$ and flux-derived k values" was added. We assessed systematic errors and random errors of measurements, drifting chamber and anchored chambers, sampling time. We carefully read the two article and the two citations were included. In addition, the following text was added in the Method section.

"Similar to other studies (Alin et al., 2011), sampling and flux measurements in the day time would tend to underestimate CO_2 evasion rate (*Bodmer et al., 2016*)."

(5) Developing models to estimate transport coefficients is meaningful, but the process of the model development is poorly described. Additionally, which data were used for the models, and which not is confusing to me (goes along with my comment (2) above).

Response: The issue was addressed as follows.

Water samples from a total of 115 sites were collected. Floating chambers with replicates were deployed in 101 sites (32 sampling sites in Daning, 37 sites in TGR river networks and 32 sites in Qijiang). The sampling period covered spring and summer season, our sampling points are reasonable considering a water area of 433 km^2. For example, 16 sites were collected for Yangtze system to examine hydrological and geomorphological controls on pCO_2 (Liu *et al.*, 2017), and 17 sites for dynamic biogeochemical controls on riverine pCO_2 in the Yangtze basin (Liu *et al.*, 2016). Similar to other studies, sampling and flux measurements in the day would tend to underestimate CO_2 evasion rate (Bodmer *et al.*, 2016).

Prior to statistical analysis, we excluded k_{600} data for samples with the air-water pCO_2 gradient <110 μatm, since the error in the k_{600} calculations drastically enhances when $\triangle pCO_2$ approaches zero (Borges *et al.*, 2004; Alin *et al.*, 2011), and datasets with $\triangle pCO_2$ >110 μatm provide an error of <10% on k_{600} computation (please refer to the Fig. 1 in the bottom). Thus, we discarded the samples (36.7% of sampling points with flux measurements) with $\triangle pCO_2$ <110 μatm for k_{600} model development, while for the flux estimations from diffusive model and floating chambers, all samples were included.

Multivariate statistics, such as correlation and stepwise multiple linear regression, were performed for the models of k_{600} using potential physical parameters of wind speed, water depth, and current velocity as the independent variables from both separated data and combined data (Alin *et al.*, 2011). k models were obtained by water depth using data from the TGR rivers, while by flow velocity in the Qijiang.

We also highlighted separated or combined data used in the Tables and Figures.

(6) From what I see in these data, there are several running waters undersaturated with respect to CO2 (Fig. S2 and Fig. 1), and hence a sink of CO2. This aspect is totally neglected and the investigated running waters are generalized as CO2 sources to the atmosphere. In my opinion, this aspect of influxes of CO2 is very valuable and should be properly discussed.

Response: We addressed this issue by revision in the parts of "Result" and "Discussion". ***Firstly, we revised in the "Result" section as follows.*** "pCO_2 varied between 50 and 4830 μatm with mean of 846 ± 819 μatm (Table 1). There were 28.7% of samples that had pCO_2 levels lower than 410 μatm, while the studied rivers were overall supersaturated with reference to atmospheric CO_2 and act as a source for the atmospheric CO_2."

Secondly, the under-saturated pCO_2 levels were further examined in the "Discussion" part.

"The calculated pCO_2 levels were within the published range, but towards the lower-end of published concentrations compiled elsewhere (Cole and Caraco, 2001; Li *et al.*, 2013). The total mean pCO_2 (846 ± 819 μatm) in the TGR, Danning and Qijiang sampled was lower than one third of global river's average (3220 μatm) (Cole and Caraco, 2001). The lower pCO_2 than most of the world's river systems, particularly the under-saturated values, demonstrated that heterotrophic respiration of terrestrially derived DOC was not significant. Compared with high alkalinity, the limited delivery DOC particularly in the Daning and Qijiang river systems (Figs. S2 and S3) also indicated that in-stream respiration was limited. These two river systems are characterized by karst terrain and underlain by carbonate rock, where photosynthetic uptake of dissolved CO_2 and carbonate minerals dissolution considerably regulated aquatic pCO_2 (Zhang *et al.*, 2017).*”*

(7) As far as I see, there is some arbitrariness regarding data handling/processing. The cut-off at 110 µatm (line 198) for the air-water CO2 gradient for k600 calculations, as well as “When several extremely values are removed: : :” (line 303), needs to be described/demonstrated/justified much more clear.

Response: We provided more details on this concern.

Prior to statistical analysis, we excluded k_{600} data for samples with the air-water pCO_2 gradient <110 µatm, since the error in the k_{600} calculations drastically enhances when $\triangle pCO_2$ approaches zero (Borges et al., 2004; Alin et al., 2011), and datasets with $\triangle pCO_2$ <110 µatm provide an error of <10% on k_{600} computation” (see the Fig. 1 in the bottom)

Regarding “**“When several extremely values are removed: : :” (**L 303) for revised model, we supplied data including extremely data and excluding extremely data in Fig. 4 (the original Fig. 3).

(8) CO2 fluxes were measured, while pCO2 and transport coefficients were calculated. This should be clearly stated throughout the manuscript to be transparent.

Response: We highlighted this in the section of “Method” (see section 2.4), and changed “measured k or observed k to flux-derived k or derived k or calculated k”.

(9) I am not a native English speaker, but I think that the manuscript should be revised for the English language. (see exemplarily in the specific comments and technical corrections below)
I think this is a valuable study, but the combination of the points mentioned above make the manuscript hard to follow and the conclusions and main messages drawn in the current state of the manuscript too general. In a revised version, the study would get more shaped, more detailed and informative, and the conclusions and main messages can be specified and more related to the investigated region.

Response: We edited the English and revised the Ms based on comments and suggestions.

Specific comments
Abstract:
Line 20: Indicate how many river networks (see general comment (2))

Response: 60 rivers

Line 24: As far as I understood not when all data were included. Please be more specific here.

Response: Corrected and provided details in “Method” section.

Lines 30 – 33: This is not really new. Maybe you can specify this statement for the investigated region?

Response: Addressed. The sentence was revised as follows.

“We concluded that simple parameterization of k as a function of morphological characteristics was site specific and hence highly variable in river systems of the upper Yangtze. k models should be developed for stream studies to evaluate the contribution of these regions to atmospheric CO_2.”

Introduction:
Line 41: Bastviken et al., 2011 totally focuses on CH4. I suggest replacing this reference with a more suitable one.

Response: Bastviken et al., 2011 was replaced by “Raymond et al., 2013; Butman and Raymond, 2011”

Line 42: But you did not present "new accurate measurement techniques“ in your study, what are your reasons to mention this in the introduction?

Response: “new” was removed.

Lines 50 – 58: This equation is pretty standard knowledge and can just be described in words here. The equation can be moved to the methods.

Response: Yes, it is very standard knowledge, while the text would be more shape, and easily to follow with equations here.

Line 63: The standardized transport coefficient (k600) should be explained here.

Response: k_{600} (the standardized transfer coefficient at a temperature of 20 ^{0}C)

Line 80: You set the scene of seasonal precipitation, but in the study, you only measure in the wet season. This is contradictory. This issue should be discussed.

Response: We changed “concentrated seasonal precipitation” to “hydrological seasonality”

Lines 84-89: Kind of repetition and partially contradictory to the text in lines 43-49.

Response: The topic is different. The former focuses on flux determination, while the latter talked about k measurement method. We clarified the main text.

Lines 92-99: The relevance of the study (first time in this region, etc.), the objectives and how these objectives were approached should be written more clearly. At this point, the input parameters for the model development is totally unclear.

Response: We re-wrote this part as follows, we also provide details for k models.

To contribute to this debate, extensive investigation was firstly accomplished for determination of k in rivers and streams of the upper Yangtze using FC method. Models of k were further developed using hydraulic properties from flux measurements and TBL model.

Our recent study preliminarily investigated pCO_2 and air – water CO_2 areal flux as well as their controls from fluvial networks in the Three Gorges Reservoir (TGR) area (Li *et al.*, 2018). The past study was based on two field works, and the diffusive models from other continents were used. In this study, we attempted to derive k levels and develop the gas transfer model in this area (mountainous streams and small rivers) for more accurate quantification of CO_2 areal flux, and also to serve for the fluvial networks in the Yangtze River or others with similar hydrology and geomorphology. Moreover, we did detailed field campaigns in the two contrasting rivers Daning and Qijiang for models (Fig. 1). The study thus clearly stated distinct differences than the previous study (Li *et al.*, 2018) by the new contributions of specific objectives and data supplements, as well as wider significance.

Our new contributions to the literature include (1) determination and controls of k levels for small rivers and streams in subtropical areas of China, and (2) new k models developed using hydraulic parameters in the subtropical mountainous river networks.

Materials and methods:
Line 105: In my opinion, Figure S1 should go to the main text. There are no sampling points for Daning and Qijiang, which is confusing to me.

Response: We supplied the map in the main text as Fig. 1 (see Fig. 2 in the bottom).

Lines 105-109: Please add a reference for this statement.

Response: "Li et al., 2018" was cited here.

Lines 110-118: Please see my general comment (2): Please restructure this, make clear how many running waters were sampled where, the size of the sampled running waters (Strahler stream order is fine), and why in these three regions. Otherwise, it is hard to follow your storyline.

Response: We provided details in the section 2.2.

Water samples from a total of 115 sites were collected. Floating chambers with replicates were deployed in 101 sites (32 sampling sites in Daning, 37 sites in TGR river networks and 32 sites in Qijiang). The sampling period covered spring and summer season, our sampling points are reasonable considering a water area of 433 km^2. For example, 16 sites were collected for Yangtze system to examine hydrological and geomorphological controls on pCO_2 (Liu *et al.*, 2017), and 17 sites for dynamic biogeochemical controls on riverine pCO_2 in the Yangtze basin (Liu *et al.*, 2016). Similar to other studies, sampling and flux measurements in the day would tend to underestimate CO_2 evasion rate (Bodmer *et al.*, 2016). In our sampling points, all measured fluxes were retained since the floating chambers yielded linearly increasing CO_2 against time following manufacturer' specification.

Prior to statistical analysis, we excluded k_{600} data for samples with the air-water pCO_2 gradient <110 µatm, since the error in the k_{600} calculations drastically enhances when $\triangle pCO_2$ approaches zero (Borges *et al.*, 2004; Alin *et al.*, 2011), and datasets with $\triangle pCO_2$ >110 µatm provide an error of <10% on k_{600} computation. Thus, we discarded the samples (36.7% of sampling points with flux measurements) with $\triangle pCO_2$ <110 µatm for k_{600} model development, while for the flux estimations from diffusive model and floating chambers, all samples were included.

Line 141-142: What is "PP“?

Response: EGM-4 (Environmental Gas Monitor; PP SYSTEMS Corporation, USA)

Line 148: I don’t really understand what you mean by this sentence, please revise.

Response: Changed to “All the solvents and reagents used in experiments were of analytical - reagent grade”

Lines 155-156: This sentence is confusing to me, please revise.

Response: Changed “The relationship was yielded when z=1 (U_{10}=1.208×U_1). ” to **“U_{10}=1.208×U_1 as we measured the wind speed at a height of 1 m (U_1). ”**

Line 158: Do you mean CO2SYS? If yes, please add the corresponding reference.

Response: “Lewis et al., 1998” was cited.

Lewis, E., Wallace, D., Allison, L.J., 1998. Program developed for CO2 system calculations.; Brookhaven National Lab., Dept. of Applied Science, Upton, NY (United States); Oak Ridge National Lab., Carbon Dioxide Information Analysis Center, TN (United States), p. Medium: ED; Size: 40 p.

Line 167: What was the brand of the tubing?

Response: Changed to “CO_2 impermeable rubber-polymer tubing”

Line 170: What is DC?

Response: “DC” was removed.

Line 173: Please see my general comment (4) (i)

Response: Uncertainty of chambers was discussed in the additional section 4.4. In our study, both drifting chamber and anchored chambers were used, which is dependent on *in situ* current velocity. The following text was added.

“In sampling sites with low and favorable flow conditions (Fig. S1), freely drifting chambers (DC) were executed, while sampling sites in rivers and streams with higher flow velocity were conducted with anchored chambers (AC) (Ran *et al.*, 2017). AC would create overestimation of CO_2 emissions in our studied region (Lorke et al., 2015).”

Line 177: The units are confusing to me. Why is there two times pressure and temperature? Please double check if the units match up in the end, to me they do not.

Response: We have carefully checked and revised.

Line 187: Please be more specific: k was calculated by reorganizing Eq (1)

Response: Revised.

Line 192: Sc to the power of 0.5? This seems weird. What do you mean here?

Response: Corrected.

Line 198: Please justify the cut-off at 110 _atm. Maybe add a figure to the supplementary material.

Response: Revised. Please refer to general comment (7).

Line 203: I read about water depth and current velocity the first time here. These measurements need to be described before.

Response: Measurements of water depth and current velocity were added in the part of "Methods".

Line 213: The pH is quite high. This in combination with influxes of CO2 requires at least a short discussion about chemical enhancement.

Response: We revised the text in the section of "Result" (see the first paragraph below), and added text (see the second paragraph below) in the Discussion section (4.1).

"*p*H varied from 7.47 to 8.76 with exceptions of two quite high values of 9.38 and 8.87 (mean: 8.39 ± 0.29 from total dataset). Much lower *p*H was observed in TGR rivers (8.21 ± 0.33) (Table 1; $p<0.05$; Fig. S2)."

"Higher pH levels were observed in Daning and Qijiang ($p<0.05$ by ANOVA), where more carbonate rock exists that are characterized by karst terrain. Our pH range was comparable to the recent study on the karst river in China (Zhang *et al.*, 2017). Quite high values (i.e., 9.38 and 8.87) were recorded in the investigated sites, where chemical enhancement would increase the influx of atmospheric CO_2 to alkaline waters (Wanninkhof and Knox, 1996), while 1.7% of sampling sites that were strongly affected by chemical enhancement were not significant on a regional scale. This chemical enhancement of CO_2 influx was also reported to be limited in high-pH rivers (Zhang *et al.*, 2017)."

Wanninkhof, R., Knox, M., 1996. Chemical enhancement of CO2 exchange in natural waters. Limnology and Oceanography 41, 689-697.

Line 214: Please see my general comment (6)

Response: Addressed. Please refer to general comment (6)

Lines 218-222: This paragraph belongs to the discussion section.

Response: We moved this to Discussion (see 4.1)

Lines 223-227: This paragraph should be revised because it is not very clearly written. Please add the p-values to the text in case of significances.

Response: We re-wrote this part, and nutrients were removed.

Lines 235-237: What is the meaning of this ratio? Please add a few words what the reader can get from this information.

Response: Removed.

Line 242: These models and how you developed them should be explained better (in the method section).

Response: We provided details in the section of "Method".

Lines 246-248: I do not understand this sentence. What do you mean by "binned"?

Response: Changed to "combined"

Discussion:

Lines 270-274: How does this paragraph support the discussion of your study?

Response: These texts could support the discussion on relationship between spatial change in k values and physical characteristics (i.e., current velocity, slope and the water depth) of three river systems.

Spatial differences of k values and their controls of physical characteristics of current velocity, slope and the water depth were discussed for the three rivers systems (please refer to section 4.2).

"This could substantiate the higher k_{600} levels and spatial changes in k_{600} values of our three river systems. For instance, similar to other turbulent rivers in China (Ran *et al.*, 2015; Ran *et al.*, 2017), high k_{600} values in the TGR, Daning and Qjiang rivers were due to mountainous terrain catchment, high current velocity (10 – 150 cm/s) (Fig. 4b), bottom roughness, and shallow water depth (10 - 150 cm) (Fig. 4a). It has been suggested that shallow water enhances bottom shear, and the resultant turbulence increases k values (Alin *et al.*, 2011; Raymond *et al.*, 2012). These physical controls are highly variable across environmental types (Figs. 4a and 4b), hence, k values are expected to vary widely (Fig. 3). The k_{600} values in the TGR rivers showed wider range (1-177 cm/h; Fig. 3; Table S1), spanning more than 2 orders of magnitude across the region, and it is consistent with the considerable variability in the physical processes on water turbulence across environmental settings. Similar broad range of k_{600} levels was also observed in the China's Yellow basin (ca. 0-123 cm/h) (Ran *et al.*, 2015; Ran *et al.*, 2017).

Contrary to our expectations, no significant relationship was observed between k_{600} and water depth, and current velocity using the entire data in the three (TGR, Danning and Qjiang) river systems

(Fig. S4). There were not statistically significant relationships between k_{600} and wind speed using separated data or combined data, and it is consistent with earlier studies (Alin *et al.*, 2011; Raymond *et al.*, 2012). Flow velocity showed linear relation with k_{600}, and the extremely high value of k_{600} was observed during the periods of higher flow velocity (Fig. S4a) using combined data. Similar trend was also observed between water depth and k_{600} values (Fig. S4b). The lack of strong correlation between k_{600} and physical factors are probably due to combined effect of both flow velocity and water depth, as well as large diversity of channel morphology, both across and within river networks in the entire catchment (60, 000 km^2). This is further collaborated by weak correlations between k_{600} and flow velocity in the TGR rivers (Fig. 4), where one or two samples were taken for a large scale examination. k_{600} as a function of water depth was obtained in the TGR rivers, but it explained only 30% of the variance in k_{600}. However, model using data from Qijiang could explain 68% of the variance in k_{600} (Fig. 4b), and it was in line with general theory. Nonetheless, k_{600} from our flow velocity based model (Fig. 4b) was potentially largely overestimated with consideration of other measurements (Alin *et al.*, 2015; Ran *et al.*, 2015; Ran *et al.*, 2017). When several extremely values were removed, k_{600} (cm/h) was parameterized as follows (k_{600} = 62.879FV + 6.8357, R^2= 0.52, p=0.019, FV-flow velocity with a unit of m/s), and this revised model was in good agreement with the model in the river networks of the Yellow River (Ran *et al.*, 2017), but much lower than the model developed in the Yangtze system (Liu *et al.*, 2017) (Fig. 4c). This was reasonable because of k_{600} values in the Yangtze system were from large rivers with higher turbulence than Yellow and our studied rivers. Furthermore, the determined k_{600} using FCs was, on average, consistent with the revised model (Table 2). These differences in relationship between spatial changes in k_{600} values and physical characteristics further corroborated heterogeneity of channel geomorphology and hydraulic conditions across the investigated rivers.”

Lines 286-288: No significances: : : But still, you developed the models considering all data? This is not at all clear to me. Did you split/separate the data set for the models? This is not clear in Table 2. Please see my general comment (2). I think these data/models are valuable, but at the moment they seem arbitrary and should be better explained. This would help to give them more weight.

Response: We now provided details of model developing in the “Method” (see the response to general comment (2), (5) and (7). We also clearly stated the separated or combined data for models in the captions of Tables and Figs.

Lines 286-309: I see a lot of results here, which are presented in the discussion for the first time. The part presenting pure results should be moved to the results section.

Response: Revised. The following text was moved to the “RESULT”.

“Contrary to our expectations, no significant relationship was observed between k_{600} and water depth, and current velocity using the entire data in the three river systems (TGR, Danning and Qjiang) (Fig. S4). There were not statistically significant relationships between k_{600} and wind speed using separated data or combined data. Flow velocity showed linear relation with k_{600}, and the extremely high value of k_{600} was observed during the periods of higher flow velocity (Fig. S4a) using combined data. Similar trend was also observed between water depth and k_{600} values (Fig. S4b). k_{600} as a function of water depth was obtained in the TGR rivers, but it explained only 30% of the variance in k_{600}. However, model using data from Qijiang could explain 68% of the variance in k_{600} (Fig. 4b), and it was in line with general theory. ”

Line 303: Please justify the removal of "extremely values". Maybe add a figure to the supplementary material. If there is no objective criteria and justification, I do not see why data should be removed.

Response: We provided details in the Method and also supplied figure with or without extremely values in the main text (Fig. 4).

Lines 327-333: Why discussing k values and not k600 values? I think this needs to be unified/consistent throughout the manuscript.

Response: We unified to k_{600} though k_{600} and k were widely discussed in previous studies.

Conclusion:
Lines 358-360: Very general, but actually, the regions had to be separated/ split, no?

Response: The words were removed.

Lines 368-369: I think you should focus the conclusion on the investigated region.

Response: "in the river systems of the upper Yangtze River" was added.

Tables:
Table 3: b) Why not presenting k600 values here which can be directly compared with other studies?

Response: We unified to "k_{600}"

Figures:
Fig. S1: Was always everything sampled at each point? "Samples" should be replaced by "sampling point"

Response: We moved it to the main text as Fig. 1, and "Samples" was changed to "Sampling point".

Fig. S4: There is no reference to this figure in the main text.

Response: Fig. S4 was cited in the main text.

Technical corrections
Line 22: Delete "were"
Line 44: add "by" or "via" before "floating chambers"
Line 59: Replace "precisely" with "well"
Line 127: "consisted" instead of "consists"
Line 139: "pH sonde" "was" instead of "is"
Line 225: "Daning" instead of "Danning"

Response: All the typos were corrected.

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Gas transfer velocities of CO_2 in subtropical monsoonal climate streams and small rivers

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Abstract

CO_2 outgassing from rivers is a critical component for evaluating riverine carbon cycle, but it is poorly quantified largely due to limited measurements and modeling of gas transfer velocity (k) in subtropical streams and rivers. We measured CO_2 flux rates, and calculated k and partial pressure (pCO_2) in 60 river networks of the Three Gorges Reservoir (TGR) region, a typical area in the upper Yangtze River with monsoonal climate and mountainous terrain. The ~~observed~~ determined k_{600} values (~~k_{600}=~~48.4±53.2 cm/h) ~~were~~ showed large variability due to spatial variations in physical controls on surface water turbulence. Our flux-derived k values ~~measurements~~ using chambers were comparable with model derived from flow velocities. Unlike in open waters, k is more pertinent to flow velocity and water depth in the studied ~~small~~ river systems. Our results show that TGR river networks emitted approx. 0.7~~1.4~~ Tg CO_2~~/y~~during monsoonal period using varying approaches such as chambers, derived~~measured~~ k values and developed k model. This study suggests that incorporating scale-appropriate k measurements into extensive pCO_2 investigation is required to refine basin-wide carbon budgets in the subtropical streams and small rivers. We concluded that simple parameterization of k as a function of morphological characteristics was site specific and hence highly variable in rivers of the upper Yangtze. k models should be developed for stream studies to evaluate the contribution of these regions to atmospheric CO_2.

Key words: CO_2 outgassing, riverine C flux, flow velocity, physical controls, Three Gorge Reservoir, Yangtze River

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1. Introduction

Rivers serve as a significant contributor of CO_2 to the atmosphere (Cole *et al.*, 2007; Tranvik *et al.*, 2009; Li *et al.*, 2012; Raymond *et al.*, 2013). As a consequence, accurate quantification of riverine CO_2 emissions is a key component to estimate net continental carbon (C) flux (Raymond *et al.*, 2013). More detailed observational data and ~~new~~ accurate measurement techniques are critical to refine the riverine C budgets (Raymond and Cole, 2001; Li and Bush, 2015). Generally two methods are used to estimate CO_2 areal fluxes from the river system, such as direct measurements floating chambers (FCs), and indirect calculation of thin boundary layer (TBL) model that depends on gas concentration at air-water gradient and gas transfer velocity, k (Guerin *et al.*, 2007; Xiao *et al.*, 2014). Direct measurements are normally laborious, while the latter method shows ease and simplicity and thus is preferred (Butman and Raymond, 2011; Li *et al.*, 2012; Li *et al.*, 2013; Lauerwald *et al.*, 2015; Ran *et al.*, 2015).

The areal flux of CO_2 (F, unit in $mmol/m^2/d$) *via* the water–air interface by TBL is described as follows:

$$F = k \times K_h \times \triangle pCO_2 \qquad (1)$$

$$K_h = 10^{-(1.11 + 0.016 * T - 0.00007 * T^2)} \qquad (2)$$

where k (unit in m/d) is the gas transfer velocity of CO_2 (also referred to as piston velocity) at the *in situ* temperature (Li *et al.*, 2016). $\triangle pCO_2$ (unit in µatm) is the air-water gradient of pCO_2 (Borges *et al.*, 2004). K_h ($mmol/m^3$/ µatm) is the aqueous-phase solubility coefficient of CO_2 corrected using temperature (T in ℃) (Li *et al.*, 2016).

△$p\mathrm{CO}_2$ can be measured ~~precisely~~ well in various aquatic systems, however, the accuracy of the estimation of flux is depended on the k value. Broad ranges of k for CO_2 (Raymond and Cole, 2001; Borges *et al.*, 2004; Raymond *et al.*, 2012) were reported due to variations in techniques, tracers used and governing processes. k is controlled by turbulence at the surface aqueous boundary layer, hence, k_{600} (the standardized gas transfer velocity at a temperature of 20 ^{0}C) is parameterized as a function of wind speed in open water systems of reservoirs, lakes, and oceans (Borges *et al.*, 2004; Guerin *et al.*, 2007; Wanninkhof *et al.*, 2009). While in streams and small rivers, turbulence at the water-air interface is generated by shear stresses at streambed, thus k is modeled using channel slope, water depth, and water velocity in particular (Alin *et al.*, 2011; Raymond *et al.*, 2012). Variable formulations of k have been established by numerous theoretical, laboratory and field studies, nonetheless, better constraint on k levels is still required as its levels are very significant and specific due to large heterogeneity in hydrodynamics and physical characteristics of river networks. This highlights the importance of k measurements in a wide range of environments for the accurate upscaling of CO_2 evasion, and for parameterizing the physical controls on k_{600}. However, only few studies provide information of k for riverine CO_2 flux in Asia (Alin *et al.*, 2011; Ran *et al.*, 2015), and those studies do not address the variability of k in China's small rivers and streams.

Limited studies demonstrated that higher levels of k in the Chinese large rivers (Alin *et al.*, 2011; Ran *et al.*, 2015; Liu *et al.*, 2017; Ran *et al.*, 2017), which contributed to much higher CO_2 areal flux particularly in China's monsoonal rivers that are impacted by ~~concentrated seasonal precipitation~~hydrological seasonality. The monsoonal flow pattern and thus flow velocity is expected to be different than other rivers in the world, as a consequence, k levels should be different than others, and potentially is higher in subtropical monsoonal rivers.

Considerable efforts, such as purposeful (Jean-Baptiste and Poisson, 2000; Crusius and Wanninkhof, 2003) and natural tracers (Wanninkhof, 1992) and FCs (Borges *et al.*, 2004; Guerin *et al.*, 2007; Alin *et al.*, 2011; Prytherch *et al.*, 2017), have been carried out to estimate accurate k values. The direct determination of k by FCs is more popular due to simplicity of the technique for short-term CO_2 flux measurements (Raymond and Cole, 2001; Xiao *et al.*, 2014; Prytherch *et al.*, 2017). Prior reports, however, have demonstrated that k values and the parameterization of k as a function of wind and/or flow velocity (probably water depth) vary widely across rivers and streams (Raymond and Cole, 2001; Raymond *et al.*, 2012). To contribute to this debate, extensive investigation was firstly accomplished for determination of k in rivers and streams of the upper Yangtze using FC method. Models of k were further developed using hydraulic properties by flux measurements and TBL model. Our new contributions to the literature include~~are~~ (1) ~~providing first~~ determination and controls of k levels for small rivers and streams in subtropical areas of China, and (2) ~~comparisons of two methods for CO_2 areal fluxes by FCs and models developed~~new models developed in the subtropical mountainous river networks. The outcome of this study is expected to help in accurate estimation of CO_2 evasion from subtropical rivers and streams, and thus refine riverine C budget over a regional/basin scale.

Our recent study preliminarily investigated pCO$_2$ and air – water CO$_2$ areal flux as well as their controls from fluvial networks in the Three Gorges Reservoir (TGR) area (Li *et al.*, 2018). The past study was based on two field works, and the diffusive models from other continents were used. In this study, we attempted to derive k levels and develop the gas transfer model in this area (mountainous streams and small rivers) for more accurate quantification of CO$_2$ areal flux, and also to serve for the fluvial networks in the Yangtze River or others with similar hydrology and geomorphology. Moreover, we did detailed field campaigns in the two contrasting rivers Daning and Qijiang for models (Fig. 1). The study thus clearly stated distinct differences than the previous study (Li *et al.*, 2018) by the new contributions of specific objectives and data supplements, as well as wider significance.

2. Materials and methods

2.1. Study areas

All field measurements were carried out in the rivers and streams of the Three Gorges Reservoir (TGR) region (28°44′–31°40′N, 106°10′–111°10′E) that is locating in the upper Yangtze River, China (Fig. 1~~S1~~). This region is subject to humid subtropical monsoon climate with an average annual temperature ranging between15 and 19 ℃. Average annual precipitation is approx. 1250 mm with large intra- and inter-annual variability. About 75% of the annual total rainfall is concentrated in April through September (Li *et al.*, 2018).

The river sub-catchments include large scale river networks covering the majority of the tributaries of the Yangtze in the TGR region, i.e., ~~32 first-order tributaries and 16 second-order~~48 tributaries were collected. These tributaries have drainage areas that vary widely from 100 to 4400 km^2 with width ranging from 1 m to less than 100 m. The annual discharges from these tributaries have a broad spectrum of 1.8 – 112 m^3/s. Detailed samplings were conducted in the two largest rivers of Daning (35 sampling sites) and Qijiang (32 sites) in the TGR region. These two river basins drain catchment areas of 4200 and 4400 km^2~~-~~.~~with maximal third-order tributaries~~The studied river systems had width < 100 m, we thus defined them as small rivers and streams. The Daning and Qijiang river systems are underlain by widely carbonate rock, and locating in a typical karst area. The location of sampling sites is deciphered in Fig. 1~~S1~~. The detailed information on sampling sites and primary data are presented in the Supplement Materials (Appendix Table A1). The sampling sites are outside the Reservoirs and are not affected by dam operation.

2.2. Water sampling and analyses

Three fieldwork campaigns from the main river networks in the TGR region were undertaken during May through August in 2016 (i.e., 18-22 May for Daning, 21 June-2 July for the entire tributaries of TGR, and 15-18 August for Qijiang). A total of 115 discrete grab samples were collected (each sample consisted~~s~~ of three replicates). Running waters were taken using pre acid-washed 5-L high density polyethylene (HDPE) plastic containers from depths of 10 cm below surface. The samples were filtered through pre-baked Whatman GF/F (0.7-µm pore size) filters on the sampling day and immediately stored in acid-washed HDPE bottles. The bottles were transported in ice box to the laboratory and stored at 4 ℃ for analysis.

Concentrations of dissolved organic carbon (DOC) and nutrients~~, dissolved total nitrogen (DTN), and dissolved total phosphorus (DTP)~~ were determined within 7 days of water collection (Mao *et al.*, 2017).

Water temperature (T), *p*H, DO saturation (DO%) and electrical conductivity (EC) were measured *in situ* by the calibrated multi-parameter sondes (HQ40d HACH, USA, and YSI 6600, YSI incorporated, USA). pH, the key parameter for *p*CO$_2$ calculation, was measured to a precision of ±0.002, and pH sonde was~~is~~ calibrated by the certified reference materials (CRMs) before measurements with an accuracy is better than 0.2%. Atmospheric CO$_2$ concentrations were determined *in situ* using ~~PP Systems~~ EGM-4 (Environmental Gas Monitor; PP SYSTEMS Corporation, USA). Total alkalinity was measured using a fixed endpoint titration method with 0.0200 mol/L hydrochloric acid (HCl) on the sampling day. DOC concentration was measured using a total organic carbon analyzer (TOC-5000, Shimadzu, Japan) with a precision better than 3% (Mao *et al.*, 2017). ~~DTN and DTP concentrations were determined using a continuous-flow autoanalyzer (AA3, Seal Analytical, Germany) and/or spectrophotometer following peroxodisulfate oxidation (Ebina *et al.*, 1983).~~ All the solvents and reagents used in experiments were of a~~A~~nalytical ~~-~~reagent grade. ~~chemical were used for all experiments.~~

Concomitant stream width, depth and flow velocity were determined along the

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cross section. Wind speed at 1 m over the water surface (U_1) and air temperature (Ta) were measured with a Testo 410-1 handheld anemometer (Germany). Wind speed at 10 m height (U_{10}, unit in m/s) was calculated using the following formula (Crusius and Wanninkhof, 2003):

$$U_{10} = U_Z\left[1+\frac{(C_{d10})^{1/2}}{K}\times\ln\left(\frac{10}{z}\right)\right] \qquad (3)$$

where C_{d10} is the drag coefficient at 10 m height (0.0013 m/s), and K is the von Karman constant (0.41), and z is the height (m) of wind speed measurement. ~~The relationship was yielded when z=1 (~~U_{10}=1.208×U_1 as we measured the wind speed at a height of 1m (U_1)~~)~~.

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Aqueous pCO_2 was computed from the measurements of pH, total alkalinity, and water temperature using CO_2 system (k_1 and k_2 are from Millero, 1979) (Lewis *et al.*, 1998), which have been identified as high quality data (Borges *et al.*, 2004; Li *et al.*, 2012; Li *et al.*, 2013).

2.3. Water-to-air CO_2 fluxes using FC method

FCs (30 cm in diameter, 30 cm in height) were deployed to measure air-water CO_2 fluxes and transfer velocities. They were made of cylindrical polyvinyl chloride (PVC) pipe with a volume of 21.20 L and a surface area of 0.071 m^2. These non-transparent, thermally insulated vertical tubes, covered by aluminum foil, were connected *via* CO_2 impermeable rubber-polymer tubing (with outer and inner diameters of 0.5 cm and 0.35 cm, respectively) to a portable non-dispersive infrared

CO_2 analyzer EGM-4 (PPSystems). Air was circulated through the EGM-4 instrument *via* an air filter using an integral ~~DC~~ pump at a flow rate of 350 ml/min. The chamber method was widely used and more details of advantages and limits on chambers were reviewed elsewhere (Borges *et al.*, 2004; Alin *et al.*, 2011; Xiao *et al.*, 2014).

Chamber measurements were conducted by deploying two replicate chambers or one chamber for two times at each site. In sampling sites with low and favorable flow conditions (Fig. S1), freely drifting chambers (DCs) were executed, while sampling sites in rivers and streams with higher flow velocity were conducted with anchored chambers (ACs) (Ran *et al.*, 2017). ACs would create overestimation of CO_2 emissions in our studied region (Lorke *et al.*, 2015). Data were logged automatically and continuously at 1-min interval over a given span of time (normally 5-10 minutes) after enclosure. The CO_2 area flux (mg/m^2/h) was calculated using the following formula.

$$F = 60 \times \frac{dpco2 \times M \times P \times T_0}{dt \times V_0 \times P_0 \times T} H \qquad (4)$$

Where dpco_2/dt is the rate of concentration change in FCs (µl/l/min); M is the molar mass of CO_2 (g/mol); P is the atmosphere pressure of the sampling site (Pa); T is the chamber absolute temperature of the sampling time (K); V_0, P_0, T_0 is the molar volume (22.4 l/mol), P_0 is atmosphere pressure (101325 Pa), and T_0 is absolute temperature (273.15 K) –under the standard condition; H is the chamber height above the water surface (m) (Alin *et al.*, 2011). We accepted the flux data that had a good linear regression of flux against time ($R^2 \geq 0.95$, $p<0.01$) following manufacturer’ specification. In our sampling points, all measured fluxes were retained since the

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floating chambers yielded linearly increasing CO_2 against time.

Water samples from a total of 115 sites were collected. Floating chambers with replicates were deployed in 101 sites (32 sampling sites in Daning, 37 sites in TGR river networks and 32 sites in Qijiang). The sampling period covered spring and summer season, our sampling points are reasonable considering a water area of 433 km^2. For example, 16 sites were collected for Yangtze system to examine hydrological and geomorphological controls on pCO_2 (Liu *et al.*, 2017), and 17 sites for dynamic biogeochemical controls on riverine pCO_2 in the Yangtze basin (Liu *et al.*, 2016). Similar to other studies, sampling and flux measurements in the day would tend to underestimate CO_2 evasion rate (Bodmer *et al.*, 2016).

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2.4. Calculations of the gas transfer velocity

The k was ~~computed with~~calculated by reorganizing E~~e~~q (1). To make comparisons, k is normalized to a Schmidt (Sc) number of 600 (k_{600}) at a temperature of 20 ℃.

$$k_{600} = k_T\left(\frac{600}{S_{CT}}\right)^{-0.5} \qquad (5)$$

$$S_{CT} = 1911.1 - 118.11T + 3.4527T^2 - 0.04132T^3 \qquad (5)$$

Where k_T is the measured values at the *in situ* temperature (T, unit in ^oC), S_{CT} is the Schmidt number of temperature T. Dependency of ~~k proportional to $Sc^{-0.5}$~~ -0.5 was employed here as measurement were made in ~~highly~~ turbulent rivers and streams in this study (Wanninkhof, 1992; Borges *et al.*, 2004; Alin *et al.*, 2011).

2.5. Estimation of river water area

Water surface is an important parameter for CO_2 efflux estimation, while it depends on its climate, channel geometry and topography. River water area therefore largely fluctuates with much higher areal extent of water surface particularly in monsoonal season. However, most studies do not consider this change, and a fraction of the drainage area is used in river water area calculation (Zhang *et al.*, 2017). In our study, a 90 m resolution SRTM DEM (Shuttle Radar Topography Mission digital elevation model) data and Landsat images in dry season were used to delineate river network, and thus water area (Zhang *et al.*, 2018), whilst, stream orders were not extracted. Water area of river systems is generally much higher in monsoonal season in comparison to dry season, for instance, Yellow River showed 1.4-fold higher water area in the wet season than in the dry season (Ran *et al.*, 2015). Available dry-season image was likely to underestimate CO_2 estimation.

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2.6~~5~~. Data processing

Prior to statistical analysis, we excluded k_{600} data for samples with the air-water pCO_2 gradient <110 µatm, since the error in the k_{600} calculations drastically enhances when ΔpCO_2 approaches zero (Borges *et al.*, 2004; Alin *et al.*, 2011),~~.~~ and datasets with ΔpCO_2 >110 µatm provide an error of <10% on k_{600} computation. Thus, we discarded the samples (36.7% of sampling points with flux measurements) with ΔpCO_2 <110 µatm for k_{600} model development, while for the flux estimations from diffusive model and floating chambers, all samples were included.

Spatial differences (Daning, Qijiang and entire tributaries of TGR region) were tested using the nonparametric Mann Whitney U-test. Multivariate statistics, such as correlation and stepwise multiple linear regression, were performed for the models of k_{600} using potential physical parameters~~, such as~~ of wind speed, water depth, and current velocity –as the independent variables (Alin *et al.*, 2011). Data analyses were conducted from both separated data and combined data of river systems. k models were obtained by water depth using data from the TGR rivers, while by flow velocity in the Qijiang, whilst, models were not developed for combined data. All statistical relationships were significant at $p < 0.05$. The statistical processes were conducted using SigmaPlot 11.0 and SPSS 16.0 for Windows (Li *et al.*, 2009; Li *et al.*, 2016).

3. Results

3.1. $\mathrm{CO_2}$ partial pressure and key water quality variables

The significant spatial variations in water temperature, pH, $p\mathrm{CO}_2$~~,~~, and DOC~~ and nutrients (DTN and DTP)~~ were observed among Daning, TGR and Qijiang rivers whereas alkalinity did not display such variability (Fig. S2). *p*H varied from 7.47 to 8.76 ~~9.38 (mean of 8.39 ±0.29)~~ with exceptions of two quite high values of 9.38 and 8.87 (total mean: 8.39 ±0.29). ~~and~~ Much lower *p*H was observed in TGR rivers (8.21 ±0.33) (Table 1; $p<0.05$; Fig. S2). $p\mathrm{CO}_2$ varied between 50 and 4830 μatm with mean of 846 ±819 μatm (Table 1). There were 28.7% of samples that had $p\mathrm{CO}_2$ levels lower than 410 μatm, ~~, suggesting that all three~~while the studied rivers were~~are~~ overall supersaturated with reference to atmospheric CO_2 and act as a source for the atmospheric CO_2. The $p\mathrm{CO}_2$ levels were 2.1 to 2.6-fold higher in TGR rivers than

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Daning (483 ± 294 µatm) and Qijiang Rivers (614 ± 316 µatm) (Fig. S2). ~~The calculated *p*CO_2 levels were within the published range, but towards the lower-end of published concentrations compiled elsewhere (Cole and Caraco, 2001; Li *et al.*, 2013). The total mean *p*CO_2 (846 µatm) in the TGR, Danning and Qijiang sampled is lower than one third of global river's average (3220 µatm) (Cole and Caraco, 2001).~~

The much higher concentration~~s~~ of dissolved organic carbon (DOC) ~~and dissolved nutrients (DTN and DTP) (Fig. S3)~~ was~~were~~ observed in the TGR rivers ($p<0.01$ by Mann-Whitney Rank Sum Test; Fig. S3) ~~than Qijang and Danning~~. In comparison to Daning, Qijiang showed significantly~~Relatively~~ higher concentration~~s~~ of DOC ~~and DTN were observed in Qijang than Daning River but mean TDP was much higher in latter than former river~~ ($p<0.05$; Fig. S3~~; Table 1~~).

3.2. CO_2 flux using floating chambers

The calculated CO_2 areal fluxes were higher in TGR rivers (217.7 ± 334.7 mmol/m^2/d, n = 35), followed by Daning (122.0 ± 239.4 mmol/m^2/d, n = 28) and Qijiang rivers (50.3 ± 177.2 mmol/m^2/d, n = 32) (Fig. 2~~1~~). The higher CO_2 evasion from the TGR rivers is consistent with high riverine *p*CO_2 levels. The mean CO_2 emission rate was 133.1 ± 269.1 mmol/m^2/d (n = 95) in all three rivers sampled. The mean CO_2 flux differed significantly between TGR rivers and Qijiang (Fig. 2~~1~~). ~~The ratio of mean to median of CO_2 areal flux ranged between 1.4 (Qijiang) and 2.6 (TGR rivers).~~

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3.3. k levels

~~Samples with $\Delta p\mathrm{CO}_2$ less than 110 µatm were excluded for k_{600} calculations, thus a~~ A total of 64 data were used (10 for Daning River, 33 for TGR rivers and 21 for Qijiang River) to develop k model after removal of samples with $\Delta p\mathrm{CO}_2$ less than 110 µatm (Table 2). No significant variability in k_{600} values were observed among the three rivers sampled (Fig. 3~~2~~). The mean k_{600} (unit in cm/h) was~~is~~ relatively higher in Qijiang (60.2 ±78.9), followed by Daning (50.2 ±20.1) and TGR rivers (40.4 ±37.6), while the median k_{600} (unit in cm/h) was higher in Daning (50.5), followed by TGR rivers (30.0) and Qijiang (25.8) (Fig. 3~~2~~; Table S1). Combined ~~Binned~~ k_{600} data were averaged to 48.4 ±53.2 cm/h (95% CI: 35.1-61.7), and it is 1.5-fold higher than the median value (32.2 cm/h) (Fig. 3~~2~~).

Contrary to our expectations, no significant relationship was observed between k_{600} and water depth, and current velocity using the entire data in the three river systems (TGR, Danning and Qjiang) (Fig. S4). There were not statistically significant relationships between k_{600} and wind speed using separated data or combined data. Flow velocity showed linear relation with k_{600}, and the extremely high value of k_{600} was observed during the periods of higher flow velocity (Fig. S4a) using combined data. Similar trend was also observed between water depth and k_{600} values (Fig. S4b). k_{600} as a function of water depth was obtained in the TGR rivers, but it explained only 30% of the variance in k_{600}. However, model using data from Qijiang could explain 68% of the variance in k_{600} (Fig. 4b), and it was in line with general theory.

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4. Discussion

4.1. Determined k values relative to world rivers

We derived first-time the k values in the subtropical streams and small rivers. Our determined~~measured~~ k_{600} levels with a 95% CI of 35.1 to 61.7 (mean: 48.4) cm/h is compared well with a compilation of data for streams and small rivers (e.g., 3-70 cm/h) (Raymond *et al.*, 2012). Our determined k_{600} values are greater than the global rivers' average (8 - 33 cm/h) (Butman and Raymond, 2011; Raymond *et al.*, 2013), and much higher than mean for tropical and temperate large rivers (5-31 cm/h) (Alin *et al.*, 2011). These studies evidences that k_{600} values are highly variable in streams and small rivers (Alin *et al.*, 2011; Ran *et al.*, 2015). Though the mean k_{600} in the TGR, Daning and Qijiang is higher than global mean, however, it is consistent with k_{600} values in the main stream and river networks of the turbulent Yellow River (42 ± 17 cm/h) (Ran *et al.*, 2015), and Yangtze (38 ± 40 cm/h) (Liu *et al.*, 2017) (Table S2).

The calculated $p\mathrm{CO}_2$ levels were within the published range, but towards the lower-end of published concentrations compiled elsewhere (Cole and Caraco, 2001; Li *et al.*, 2013). The total mean $p\mathrm{CO}_2$ (846 ± 819 μatm) in the TGR, Danning and Qijiang sampled was~~is~~ lower than one third of global river's average (3220 μatm) (Cole and Caraco, 2001). The lower $p\mathrm{CO}_2$ than most of the world's river systems, particularly the under-saturated values, demonstrated that heterotrophic respiration of terrestrially derived DOC was not significant. Compared with high alkalinity, the

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limited delivery of DOC particularly in the Daning and Qijiang river systems (Figs. S2 and S3) also indicated that in-stream respiration was limited. These two river systems are characterized by karst terrain and underlain by carbonate rock, where photosynthetic uptake of dissolved CO_2 and carbonate minerals dissolution considerably regulated aquatic pCO_2 (Zhang *et al.*, 2017).

Higher pH levels were observed in Daning and Qijiang river systems ($p<0.05$ by Mann-Whitney Rank Sum Test), where more carbonate rock exists that are characterized by karst terrain. Our pH range was comparable to the recent study on the karst river in China (Zhang *et al.*, 2017). Quite high values (i.e., 9.38 and 8.87) were recorded in some investigated sites, where chemical enhancement would increase the influx of atmospheric CO_2 to alkaline waters (Wanninkhof and Knox, 1996), while 1.7% of sampling sites that were strongly affected by chemical enhancement were not significant on a regional scale. This chemical enhancement of CO_2 influx was also reported to be limited in high-pH rivers (Zhang *et al.*, 2017).

4.2. Hydraulic controls of k_{600}

It has been well established that k_{600} is governed by a multitude of physical factors particularly current velocity, wind speed, stream slope and water depth, of which, wind speed is the dominant factor of k in open waters such as large rivers and estuaries (Raymond and Cole, 2001; Crusius and Wanninkhof, 2003; Borges *et al.*, 2004; Alin *et al.*, 2011). In contrast k_{600} in small rivers and streams is closely linked to flow velocity, water depth and channel slope (Alin *et al.*, 2011; Raymond *et al.*, 2012).

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Several studies reported that the combined contribution of flow velocity and wind speed to k is significant in the large rivers (Beaulieu *et al.*, 2012; Ran *et al.*, 2015). Thus, k_{600} values are higher in the Yellow River (ca. 0-120 cm/h) as compared to the low-gradient River Mekong (0-60 cm/h) (Alin *et al.*, 2011; Ran *et al.*, 2015), due to higher ~~wind speed and~~ flow velocity in the Yellow River (1.8 m/s) than Mekong river (0.9 ±0.4 m/s), resulting in greater surface turbulence and higher k_{600} level in the Yellow (42 ±17 cm/h) than Mekong river (15 ±9 cm/h). This could substantiate the higher k_{600} levels and spatial changes in k_{600} values of our three river systems. For instance, similar to other turbulent rivers in China (Ran *et al.*, 2015; Ran *et al.*, 2017), ~~The~~ high~~er~~ k_{600} values in the TGR, Daning and Qjiang rivers were~~are~~ due to mountainous terrain catchment, high current velocity (10 – 150 cm/s) (Fig. 4~~3~~b), bottom roughness, and shallow water depth (10 - 150 cm) (Fig. 4~~3~~a). It has been suggested that shallow water enhances bottom shear, and the resultant turbulence increases k values (Alin *et al.*, 2011; Raymond *et al.*, 2012). These physical controls are highly variable across environmental types (Figs. 4~~3~~a and 4~~3~~b), hence, k values are expected to vary widely (Fig. 3~~2~~). The k_{600} values in the TGR rivers showed wider range (1-177 cm/h; Fig. 3~~2~~; Table S1), spanning more than 2 orders of magnitude across the region, and it is consistent with the considerable variability in the physical processes on water turbulence across environmental settings. Similar broad range of k_{600} levels was also observed in the China's Yellow basin (ca. 0-123 cm/h) (Ran *et al.*, 2015; Ran *et al.*, 2017).

~~Contrary to our expectations, no significant relationship was observed between~~

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~~k_{600} and water depth, and current velocity using the entire data in the TGR, Danning and Qjiang rivers (Fig. S5). There were not statistically significant relationships between k and wind speed and it is~~ Absent relationships between riverine k_{600} and wind speed were consistent with earlier studies (Alin *et al.*, 2011; Raymond *et al.*, 2012). ~~Flow velocity showed linear relation with k, and the extremely high value of k was observed during the periods of higher flow velocity (Fig. S5a). Similar trend was also observed between water depth and k values (Fig. S5b).~~ The lack of strong correlation between k_{600} and physical factors using the combined data were~~are~~ probably due to combined effect of both flow velocity and water depth, as well as large diversity of channel morphology, both across and within river networks in the entire catchment (60, 000 km^2). This is further collaborated by weak correlations between k_{600} and flow velocity in the TGR rivers (Fig. 4~~3~~), where one or two samples were taken for a large scale examination. We provided new insights into k_{600} parameterized using current velocity. ~~k_{600} as a function of water depth was obtained in TGR rivers, but it explained only 30% of the variance in k_{600}. However, model using data from Qijiang could explain 68% of the variance in k_{600} (Fig. 3b), and it was in line with general theory.~~ Nonetheless, k_{600} from our flow velocity based model (Fig. 4~~3~~b) ~~is~~ was potentially largely overestimated with consideration of other measurements (Alin *et al.*, 2015; Ran *et al.*, 2015; Ran *et al.*, 2017). When several extremely values ~~are~~ were removed, k_{600} (cm/h) ~~is~~ was parameterized as follows (k_{600} = 62.879FV + 6.8357, R ²= 0.52, p=0.019, FV-flow velocity with a unit of m/s), and this revised model ~~is~~ was in good agreement with the model in the river networks of

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the Yellow River (Ran *et al.*, 2017), but much lower than the model developed in the Yangtze system (Liu *et al.*, 2017) (Fig. 4~~3~~c). This ~~is~~ was reasonable because of k_{600} values in the Yangtze system ~~are~~ were from large rivers with higher turbulence than Yellow and our studied rivers. Furthermore, the ~~measured~~ determined k_{600} using FCs was, on average, consistent with the revised model (Table 2). These differences in relationship between spatial changes in k_{600} values and physical characteristics further corroborated heterogeneity of channel geomorphology and hydraulic conditions across the investigated rivers.

The subtropical streams and small rivers are biologically more active and are recognized to exert higher CO_2 areal flux to the atmosphere, however, their contribution to riverine carbon cycling is still poorly quantified because of data paucity and the absence of k in particular. Larger uncertainty of riverine CO_2 emission in China was anticipated by use of k_{600} from other continents or climate zones. For instance, k_{600} for CO_2 emission from tributaries in the Yellow River and karst rivers was originated from the model in the Mekong (Zhang *et al.*, 2017), and Pearl (Yao *et al.*, 2007), Longchuan (Li *et al.*, 2012), and Metropolitan rivers (Wang *et al.*, 2017), which are mostly from temperate regions. Our k_{600} values will therefore largely improve the estimation of CO_2 evasion from subtropical streams and small rivers, and improve to refine riverine carbon budget. More studies, however, are clearly needed to build the model, based on flow velocity and slope/water depth given the difficulty in k quantification on a large scale.

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4.3. Implications for large scale estimation

We compared CO$_2$ areal flux by FCs and models developed here (Fig. 4~~3~~) and other studies (Alin *et al.*, 2011) (Tables 2 and 3). CO$_2$ evasion was estimated for rivers in China with k values ranged between 8 and 15 cm/h (Yao *et al.*, 2007; Wang *et al.*, 2011; Li *et al.*, 2012) (Table S2). These estimates of CO$_2$ evasion rate were considerably lower than using present k_{600} values (48.4±53.2 cm/h). For instance, CO$_2$ emission rates in the Longchuan River (e.g., k=8 cm/h) and Pearl River tributaries (e.g., k=8-15 cm/h) were 3 to 6 times higher using present k values compared to earlier estimates. We found that the determined k_{600} average was marginally beyond the levels from water depth based model and the model developed by Alin et al (Alin *et al.*, 2011), while equivalent to the flow velocity based revised model, resulting in similar patterns of CO$_2$ emission rates (Table 2). Hence selection of k values would significantly hamper the accuracy of the flux estimation. Therefore k must be estimated along with *p*CO$_2$ measurements to accurate flux estimations.

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We used our measured CO$_2$ emission rates by FCs for upscaling flux estimates during monsoonal period given the sampling in this period and it was found to be 0.70~~1.39~~ TgCO$_2$~~/y~~ for all rivers sampled~~–~~ in our study (Table 3a). The estimated emission in the monsoonal period was close to that of the revised model (0.71~~1.40~~ ± 0.66~~1.31~~ (95% confidence interval: 0.46~~91~~-0.94~~1.87~~) Tg CO$_2$~~/y~~), and using the determined k average, i.e., 0.69~~1.37~~ ±0.65~~1.28~~ (95% confidence interval: 0.45~~89~~-0.93~~1.84~~) Tg CO$_2$~~/y~~, but slightly higher than the estimation using water-depth based model (0.54~~1.08~~ ±0.51~~1.01~~ Tg CO$_2$~~/y~~) and Alin's model (0.53~~1.06~~ ±0.50~~1.00~~

Tg CO_2~~/y~~) (Table 3b). ~~The estimate was within the range of our earlier work using TBL on the TGR river networks (0.64-2.33 Tg CO_2/y) (Li *et al.*, 2018).~~ The higher emission, i.e., 1.66~~3.29~~ ± 1.55~~3.08~~ (1.08~~2.15~~-2.23~~4.43~~) Tg CO_2~~/y~~, using flow velocity based model may be over-estimated when compared to other models, flux from determined k (Table 3b) and previous annual estimates, i.e., our earlier annual evasion of 0.64-2.33 Tg CO_2/y using TBL on the TGR river networks (Li *et al.*, 2018). Moreover, our estimated CO_2 emission in the monsoonal period ~~Therefore, this study~~ also suggests that CO_2 annual emissions from rivers and streams in this area ~~may be~~were previously underestimated, i.e., 0.03 Tg CO_2/y (Li *et al.*, 2017) and 0.37-0.44 Tg CO_2/y (Yang *et al.*, 2013) as the former used TBL model with a lower k level, and the latter employed floating chambers, but they both sampled very limited tributaries (i.e., 2-3 rivers). Therefore, measurements of k must be made mandatory along with *p*CO_2 measurement in the river and stream studies.

4.4. Uncertainty assessment of *p*CO_2 and flux-derived k_{600} values

The uncertainty of flux-derived k values mainly stem from Δ*p*CO_2 (unit in ppm) and flux measurements (Lorke *et al.*, 2015; Bodmer *et al.*, 2016; Golub *et al.*, 2017). Thus we provided uncertainty assessments caused by dominant sources of uncertainty from measurements of aquatic *p*CO_2 and CO_2 areal flux since uncertainty of atmospheric CO_2 measurement could be neglected.

In our study, aquatic *p*CO_2 was computed based on pH, alkalinity and water temperature rather than directly measured. Recent studies highlighted *p*CO_2

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uncertainty caused by systematic errors over empiric random errors (Golub *et al.*, 2017). Systematic errors are mainly attributed to instrument limitations, i.e., sondes of pH and water temperature. The relative accuracy of temperature meters was ±0.1 ^{0}C according to manufacturers' specifications, thus the uncertainty of water T propagated on uncertainty in pCO$_2$ was minor (Golub *et al.*, 2017). Systematic errors therefore stem from pH, which has been proved to be a key parameter for biased pCO$_2$ estimation calculated from aquatic carbon system (Li *et al.*, 2013; Abril *et al.*, 2015). We used a high accuracy of pH electrode and the pH meters were carefully calibrated using CRMs, and *in situ* measurements showed an uncertainty of ±0.01. We then run an uncertainty of ±0.01 pH to quantify the pCO$_2$ uncertainty, and an uncertainty of ±3% was observed. Systematic errors thus seemed to show little effects on pCO$_2$ errors in our study.

Random errors are from repeatability of carbonate measurements. Two replicates for each sample showed the uncertainty of within ±5%, indicating that uncertainty in pCO$_2$ calculation from alkalinity measurements could be minor.

The measured pH ranges also exhibited great effects on pCO$_2$ uncertainty (Hunt *et al.*, 2011; Abril *et al.*, 2015). At low pH, pCO$_2$ can be overestimated when calculated from pH and alkalinity (Abril *et al.*, 2015). Samples for CO$_2$ fluxes estimated from pH and alkalinity showed pH average of 8.39±0.29 (median 8.46 with quartiles of 8.24-8.56) (n=115). Thus, overestimation of calculated CO$_2$ areal flux from pH and alkalinity is likely to be minor. Further, contribution of organic matter to

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non-carbonate alkalinity is likely to be neglected because of low DOC (mean 6.67 mg/L; median 2.51 mg/L) (Hunt *et al.*, 2011; Li *et al.*, 2013).

Efforts have been devoted to measurement techniques (comparison of FC, eddy covariance-EC and boundary layer model-BLM) for improving CO_2 quantification from rivers because of a notable contribution of inland waters to the global C budget, which could have a large effect on the magnitude of the terrestrial C sink. Whilst, prior studies reported inconsistent trends of CO_2 area flux by these methods. For instance, CO_2 areal flux from FC was much lower than EC (Podgrajsek *et al.*, 2014), while areal flux from FC was higher than both EC and BLM elsewhere (Erkkila *et al.*, 2018), however, Schilder et al (Schilder *et al.*, 2013) demonstrated that areal flux from BLM was 33-320% of in-situ FC measurements. Albeit unsatisfied errors of varied techniques and additional perturbations from FC exist, FC method is currently a simple and preferred measurement for CO_2 flux because that choosing a right k value remains a major challenge and others require high workloads (Martinsen *et al.*, 2018).

Recent study further reported fundamental differences in CO_2 emission rates between ACs and freely DFs (Lorke *et al.*, 2015), i.e., ACs biased the gas areal flux higher by a factor of 2.0-5.5. However, some studies observed that ACs showed reasonable agreement with other flux measurement techniques (Galfalk *et al.*, 2013), and this straightforward, inexpensive and relatively simple method AC was widely used (Ran *et al.*, 2017). Water-air interface CO_2 flux measurements were primarily made using ACs in our studied streams and small rivers because of relatively high

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current velocity; otherwise, floating chambers will travel far during the measurement period. In addition, inflatable rings were used for sealing the chamber headspace and submergence of ACs was minimal, therefore, our measurements were potentially overestimated but reasonable. We could not test the overestimation of ACs in this study, the modified FCs, i.e., DCs and integration of ACs and DCs, and multi-method comparison study including FCs, ECs and BLM should be conducted for a reliable chamber method.

Sampling seasonality considerably regulated riverine $p\mathrm{CO}_2$ and gas transfer velocity and thus water-air interface CO_2 evasion rate (Li *et al.*, 2012; Ran *et al.*, 2015). We sampled waters in wet season (monsoonal period) due to that it showed wider range of flow velocity and thus it covered the k_{600} levels in the whole hydrological season. Wet season generally had higher current velocity and thus higher gas transfer velocity (Ran *et al.*, 2015), while aquatic $p\mathrm{CO}_2$ was variable with seasonality. We recently reported that riverine $p\mathrm{CO}_2$ in the wet season was 81% the level in the dry season (Li *et al.*, 2018), and prior study on the Yellow River reported that k level in the wet season was 1.8-fold higher than in the dry season (Ran *et al.*, 2015), while another study on the Wuding River demonstrated that k level in the wet season was 83%-130% of that in the dry season (Ran *et al.*, 2017). Thus, we acknowledged a certain amount of errors on the annual flux estimation from sampling campaigns during the wet season in the TGR area, while this uncertainty could not be significant because that the diluted $p\mathrm{CO}_2$ could alleviate the overestimated emission by increased k level in the wet season (stronger discussion please refer to SOM).

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5. Conclusion

We provided first determination of gas transfer velocity (k) in the subtropical streams and small rivers in the upper Yangtze. High variability in k values (mean 48.4 ±53.2 cm/h) was observed, reflecting the variability of morphological characteristics on water turbulence both within and across river networks. ~~The determined k using floating chambers (FCs) was comparable to our newly water depth based model, while substantially lower than flow velocity based model.~~ We highlighted that k estimate from empirical model should be pursued with caution and the significance of incorporating k measurements along with extensive $p\mathrm{CO}_2$ investigation is highly essential for upscaling to watershed/regional scale carbon (C) budget.

Riverine $p\mathrm{CO}_2$ and CO_2 areal flux showed pronounced spatial variability with much higher levels in the TGR rivers. The CO_2 areal flux was averaged at 133.1 ± 269.1 mmol/m^2/d using FCs, the resulting emission in the monsoonal period was around 0.7~~1.39~~ Tg CO_2~~/y~~, similar to the scaling up emission with the determined k, and the revised flow velocity based model, while marginally above the water depth based model. More work is clearly needed to refine the k modeling in the river systems of the upper Yangtze River for evaluating regional C budgets.

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Table 1. Statistics of all the data from three river systems (separated statistics please refer to Figs. S2 and S3 in the Supplementary material)~~s~~.

		Water T (^{0}C)	pH	Alkalinity (μeq/l)	pCO$_2$ (μatm)	DO%	DOC (mg/L)
Number		115	115	115	115	56	114
Mean		22.5	8.39	2589.1	846.4	91.5	6.67
Median		22.8	8.46	2560	588.4	88.8	2.51
Std. Deviation		6.3	0.29	640.7	818.5	8.7	7.62
Minimum		11.7	7.47	600	50.1	79.9	0.33
Maximum		34	9.38	4488	4830.4	115.9	37.48
Percentiles	25	16.3	8.24	2240	389.8	84.0	1.33
	75	29	8.56	2920	920.4	99.1	9.96
95% CI for Mean	Lower Bound	21.4	8.33	2470.8	695.2	89.1	5.26
	Upper Bound	23.7	8.44	2707.5	997.6	93.8	8.09

CI-Confidence Interval.

Table 2. Comparison of different model for CO_2 areal flux estimation using combined data (unit is mmol/m^2/d for CO_2 areal flux and cm/h for k_{600}).

CO_2 areal flux[a]		From FC	Flow velocity-based model (Fig. 4~~3~~b)	Water depth-based model (Fig.3a)	Alin's model
k_{600}		48.4[b]	116.5[c]	38.3	37.6
Mean		198.1	476.7	156.6	154.0
S.D.		185.5	446.2	146.6	144.2
95% CI for Mean	Lower Bound	129.5	311.5	102.3	100.6
	Upper Bound	266.8	641.8	210.8	207.4

CI-Confidence Interval

a-CO_2 areal flux is based on TBL model.

b-mean level that is determined using floating chambers (FC).

c-This figure is revised to be 49.6 cm/h if the model (k_{600} = 62.879FV + 6.8357, R^2= 0.52, p=0.019) is used (Fig. 4~~3~~c), and the corresponding CO_2 areal flux is 203±190 mmol/m^2/d.

Table 3. CO_2 emission during monsoonal period (May through Oct.) from total rivers sampled in the study.

(a) Upscaling using CO_2 areal flux by FC during monsoonal period.

	Catchment Area km^2	Water surface km^2	CO_2 areal flux $mmol/m^2/d$	CO_2 emission $Tg\ CO_2/y$
Daning	4200	21.42	122.0 ± 239.4	~~0.042~~0.021
Qijiang	4400	30.8	50.3 ± 177.2	~~0.025~~ 0.0125
TGR river	50000	377.78	217.7 ± 334.7	~~1.321~~0.666
Total				~~1.39~~0.70

(b) Upscaling using determined k_{600} average and models (whole dataset are used here).

		From determined k_{600} mean	Flow velocity-based model (Fig. 4~~3~~b) (numbers in bracket is from the revised model; Fig. 4~~3~~c)	Water depth-based model (Fig. 4~~3~~a)	Alin's model
Mean		0.69 ~~1.37~~	1.66 (0.71)~~3.29 (1.40)~~	0.54 ~~1.08~~	0.53 ~~1.06~~
S.D.		0.65 ~~1.28~~	1.55 (0.66)~~3.08 (1.31)~~	0.51 ~~1.01~~	0.50 ~~1.00~~
95% CI for Mean	Lower Bound	0.45 ~~0.89~~	1.08 (0.46)~~2.15 (0.91)~~	0.36 ~~0.71~~	0.35 ~~0.69~~
	Upper Bound	0.93 ~~1.84~~	2.23 (0.94)~~4.43 (1.81)~~	0.74 ~~1.46~~	0.72 ~~1.43~~

A total water area of approx. 430 km^2 for all tributaries (water area is from Landsat ETM+ in 2015); CO_2 emission upscaling was conducted during the monsoonal period because of the sampling in this period.

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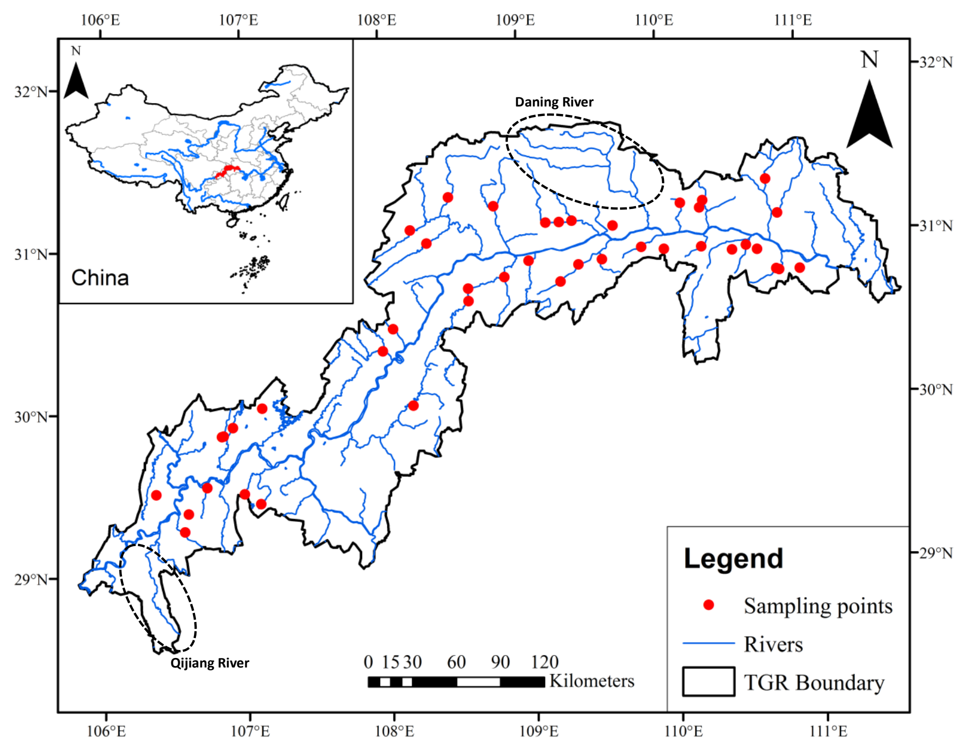

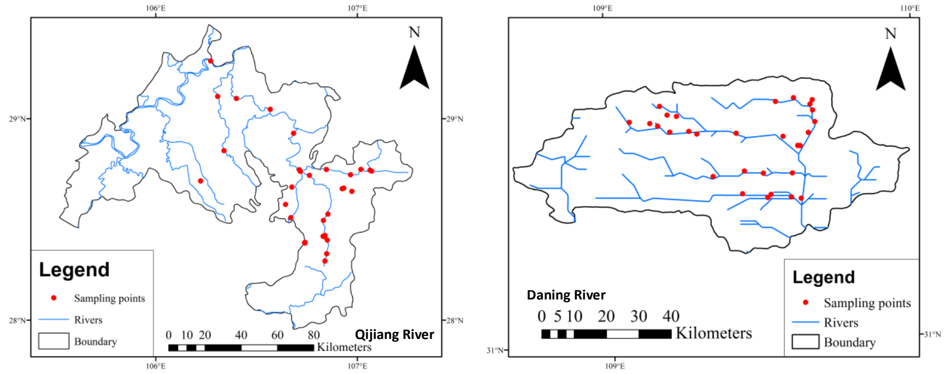

Fig. 1. Map of sampling locations of major rivers and streams in the Three Gorges Reservoir region, China.

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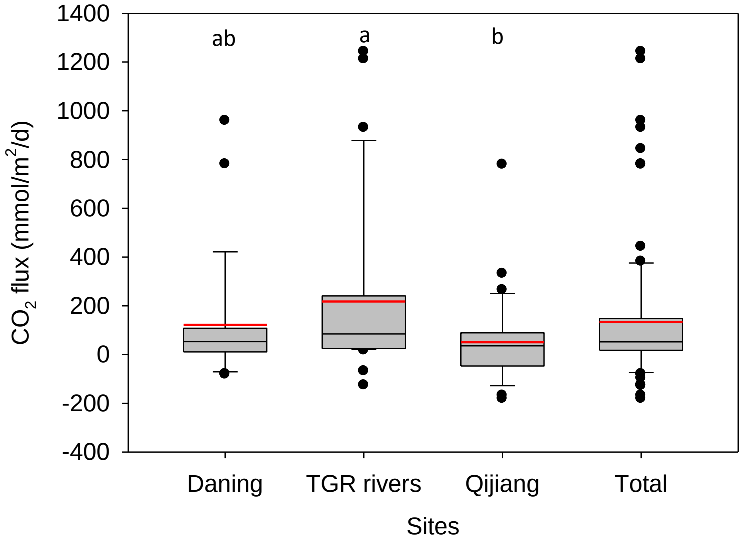

Fig. 2~~1~~. Boxplots of CO_2 emission rates by floating chambers in the investigated three river systems~~subtropical rivers~~ (different letters represent statistical differences at $p<0.05$ by **Mann-Whitney Rank Sum Test**). (the black and red lines, lower and upper edges, bars and dots in or outside the boxes demonstrate median and mean values, 25th and 75th, 5th and 95th, and <5th and >95th percentiles of all data, respectively). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article) (Total means combined data from three river systems).

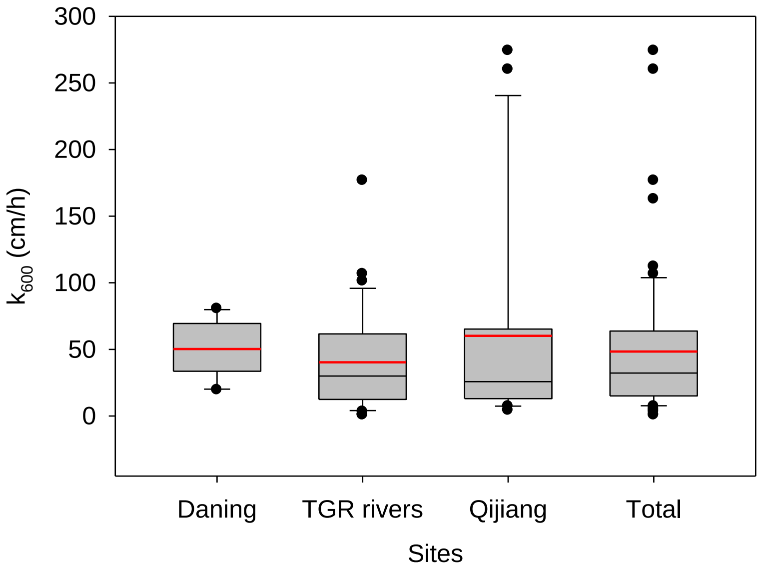

Fig. **3~~2~~.** Boxplots of k_{600} levels in the investigated three river systems~~subtropical rivers~~ (there is not a statistically significant difference in k among sites by Mann-Whitney Rank Sum Test). (the black and red lines, lower and upper edges, bars and dots in or outside the boxes demonstrate median and mean values, 25th and 75th, 5th and 95th, and <5th and >95th percentiles of all data, respectively). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article) (Total means combined data from three river systems).

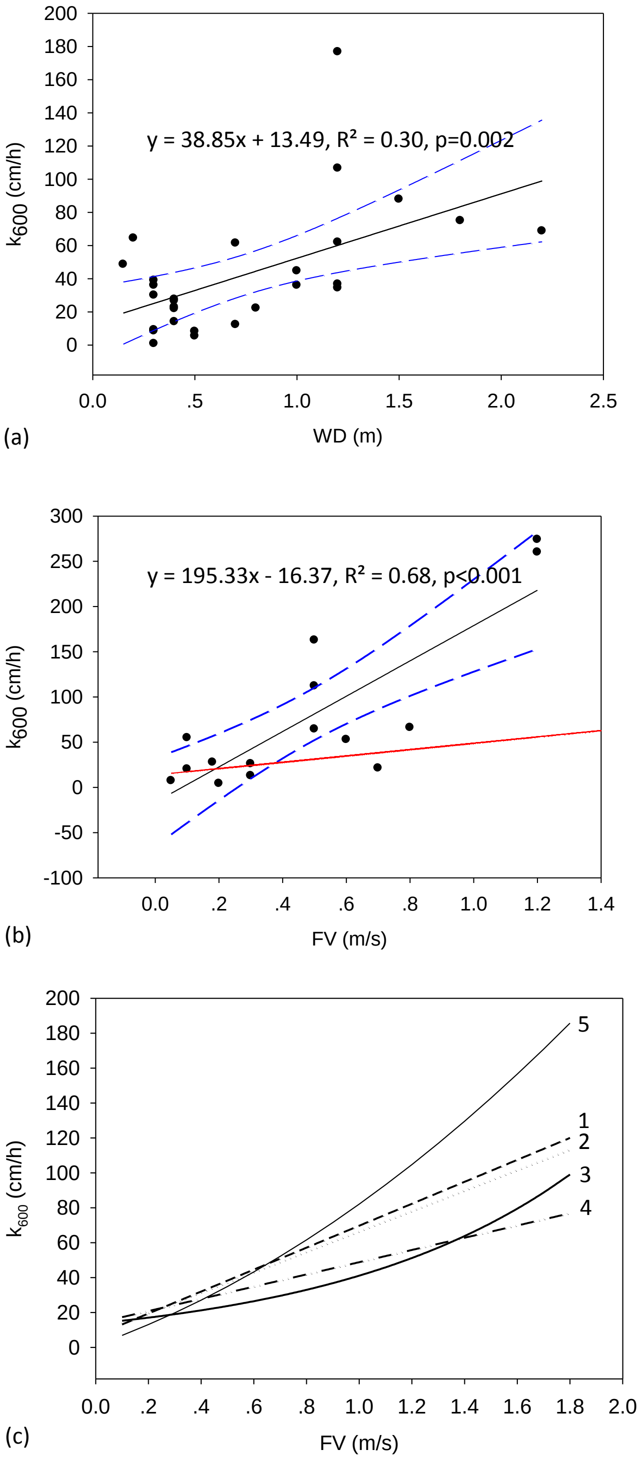

Fig. 4~~3~~. The k_{600} as a function of water depth (WD) using data from~~in~~ TGR rivers (a), flow velocity (FV) using data from ~~in~~ Qijiang (b), and comparison of the developed model with other models (c) (others without significant relationships between k and physical factors are not shown). The solid lines show regression, the dashed lines represent 95% confidence band, and the red dash-dotted line represents the model developed by Alin et al (2011) (~~in panel b,~~ if several extremely values are removed in panel b, the revised model would be k_{600} = 62.879FV + 6.8357, R^2 = 0.52, p=0.019) (in panel c, 1-the revised model, 2-model from Ran et al., 2017, 3-model from Ran et al., 2015, 4-model from Alin et al,, 2011, 5-model from Liu et al., 2017) (1- k_{600} = 62.879FV + 6.8357; 2- k_{600} = 58.47FV+7.99; 3- k_{600} = 13.677exp (1.1FV); 4- k_{600} = 35 FV + 13.82; 5- k_{600} = 6.5FV^2 + 12.9FV+0.3) (unit of k in models 1-4 is cm/h, and unit of m/d for model 5 is transferred to cm/h).

Supporting material

Gas transfer velocities of CO_2 in subtropical monsoonal climate streams and small rivers

Siyue Li*, Rong Mao, Yongmei Ma, V.V.S.S. Sarma

Appendix Table A1. Dataset in relation to our study.

Seasonal changes related to gas transfer and concentrations

We used our measured CO_2 emission rates by FCs for upscaling flux estimates and it was found to be 1.39 TgCO_2/y for all rivers sampled (Table S3a). The estimated emission was close to that of the revised model (1.40 ± 1.31 (95% confidence interval: 0.91-1.87) Tg CO_2/y), and using the determined k average, i.e., 1.37 ± 1.28 (95% confidence interval: 0.89-1.84) Tg CO_2/y, but slightly higher than the estimation using water-depth based model (1.08 ± 1.01 Tg CO_2/y) and Alin's model (1.06 ± 1.00 Tg CO_2/y) (Table S3b). The estimate was within the range of our earlier work using TBL on the TGR river networks (0.64-2.33 Tg CO_2/y) (Li *et al.*, 2018). The higher emission, i.e., 3.29 ± 3.08 (2.15-4.43) Tg CO_2/y, using flow velocity based model may be over-estimated (Table 3b). Therefore, this study suggests that CO_2 emissions from rivers and streams in this area may be underestimated, i.e., 0.03 Tg CO_2/y (Li *et al.*, 2017) and 0.37-0.44 Tg CO_2/y (Yang *et al.*, 2013) as the former used TBL model with a lower k level, and the latter employed floating chambers, but they both sampled

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very limited tributaries (i.e., 2-3 rivers). Therefore, measurements of k must be made mandatory along with pCO_2 measurement in the river and stream studies.

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As our sampling was limited to monsoonal periods only, which could not provide a meaningful annual emission estimate given the restricted sampling period. Thus, we developed a stronger discussion on what changes across seasons related to gas transfer and pCO_2 concentrations. As outlined in the main text, riverine pCO_2 in the monsoonal season in this region was 81% the level in the dry season, and current velocity was 1.7-fold higher in monsoonal season (Li *et al.*, 2018), thus k_{600} was 1.6-fold higher in the monsoonal period based our model. This could be defensible due to that prior study on the Yellow River reported that k600 level in the wet season was 1.8-fold higher than in the dry season (Ran *et al.*, 2015), another study on the Wuding River demonstrated that k level in the wet season was 83%-130% of that in the dry season (Ran *et al.*, 2017). Moreover, a factor of 1.4 for water area was designated based on other monsoonal rivers. Then annual emission could be estimated at 1.59 Tg CO_2/y, slightly higher than the estimation using the data in the monsoonal period only.

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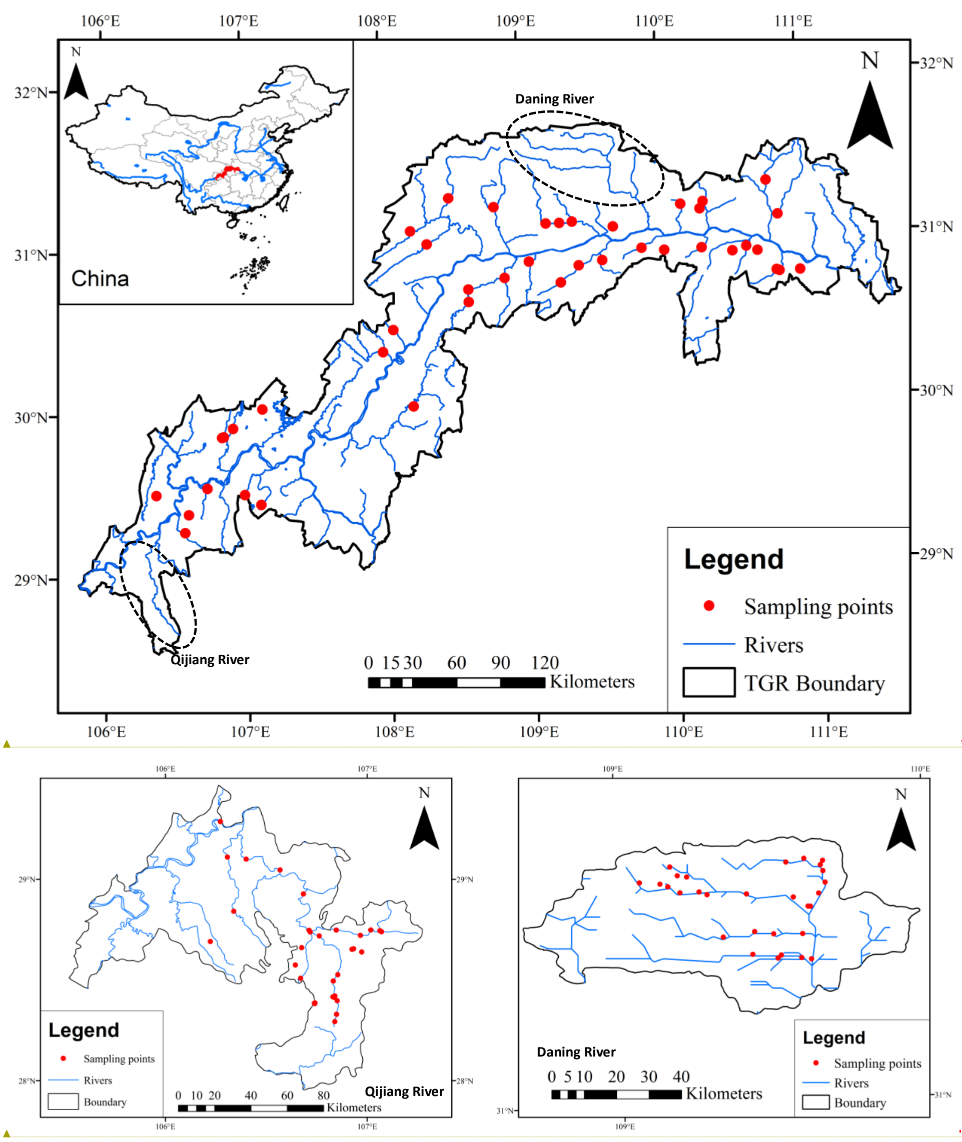

~~**Fig. S1.** Map of sampling locations of major rivers and streams in the Three Gorges Reservoir region, China.~~

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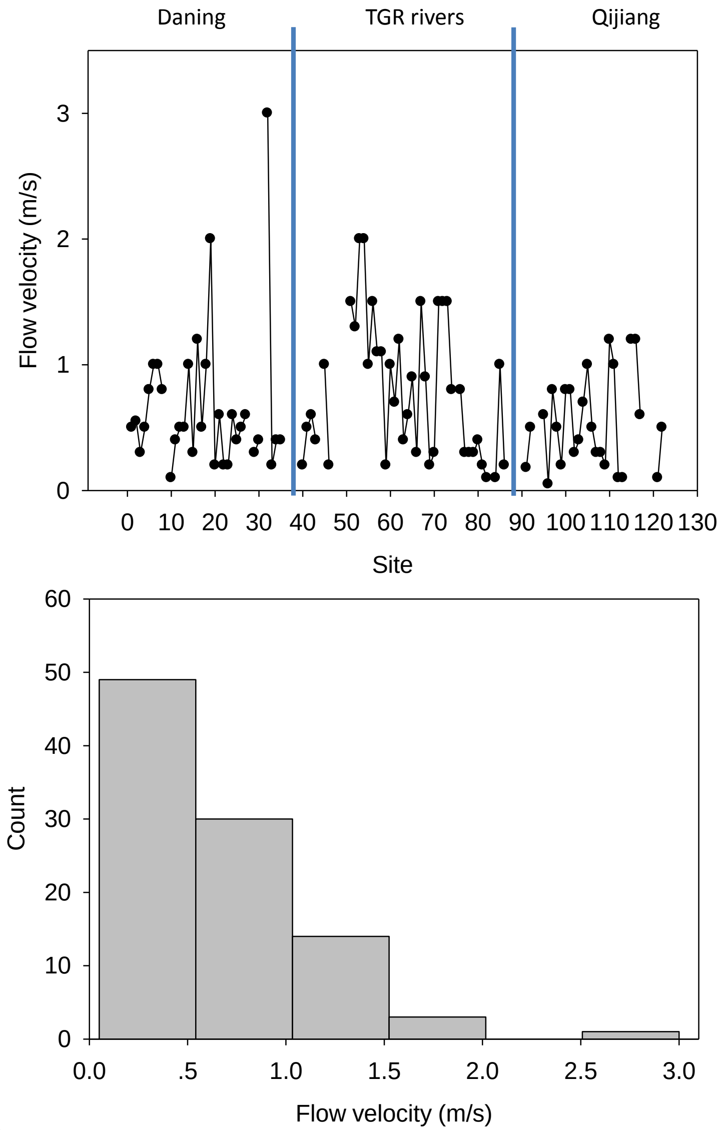

Fig. S1. Current velocity in our studied rivers.

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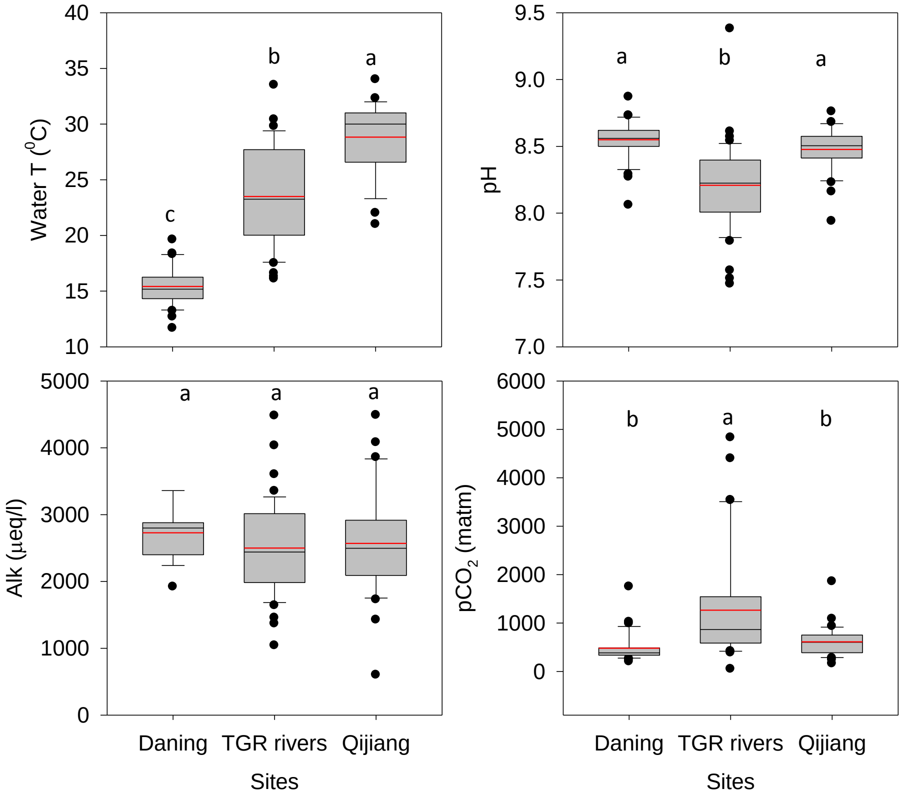


Fig. S2. Boxplots of water temperature, pH, alkalinity and pCO2 in the subtropical rivers (different letters represent statistical differences at $p<0.05$). (the black and red lines, lower and upper edges, bars and dots in or outside the boxes represent median and mean values, 25th and 75th, 5th and 95th, and <5th and >95th percentiles of all data, respectively). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

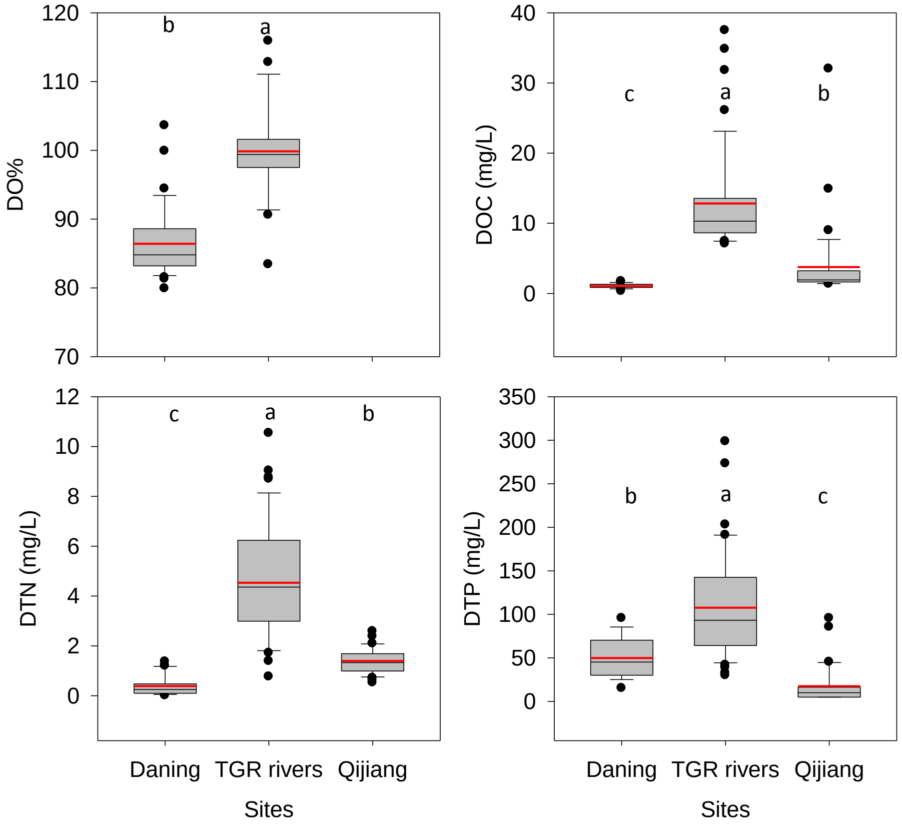
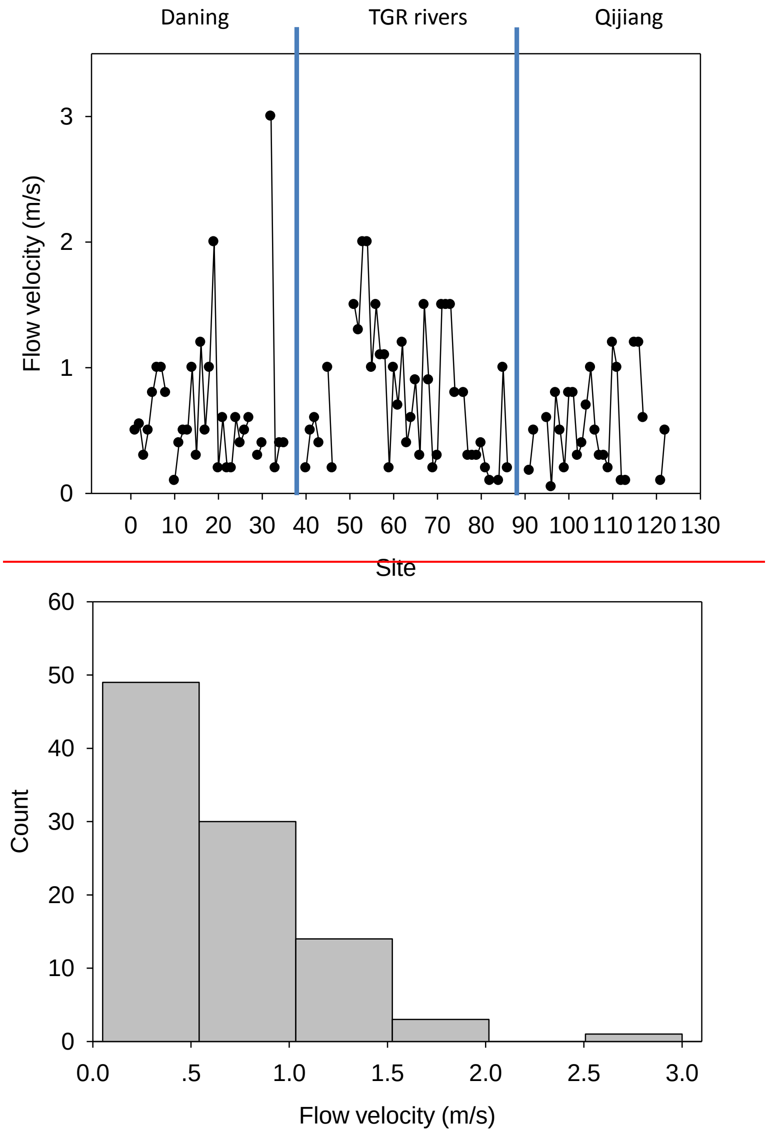


Fig. S3. Boxplots of organic carbon and nutrients in the subtropical rivers (different letters represent statistical differences at $p<0.05$). (the black and red lines, lower and upper edges, bars and dots in or outside the boxes demonstrate median and mean values, 25th and 75th, 5th and 95th, and <5th and >95th percentiles of all data, respectively). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).



~~**Fig. S4.** Current velocity in our studied rivers.~~

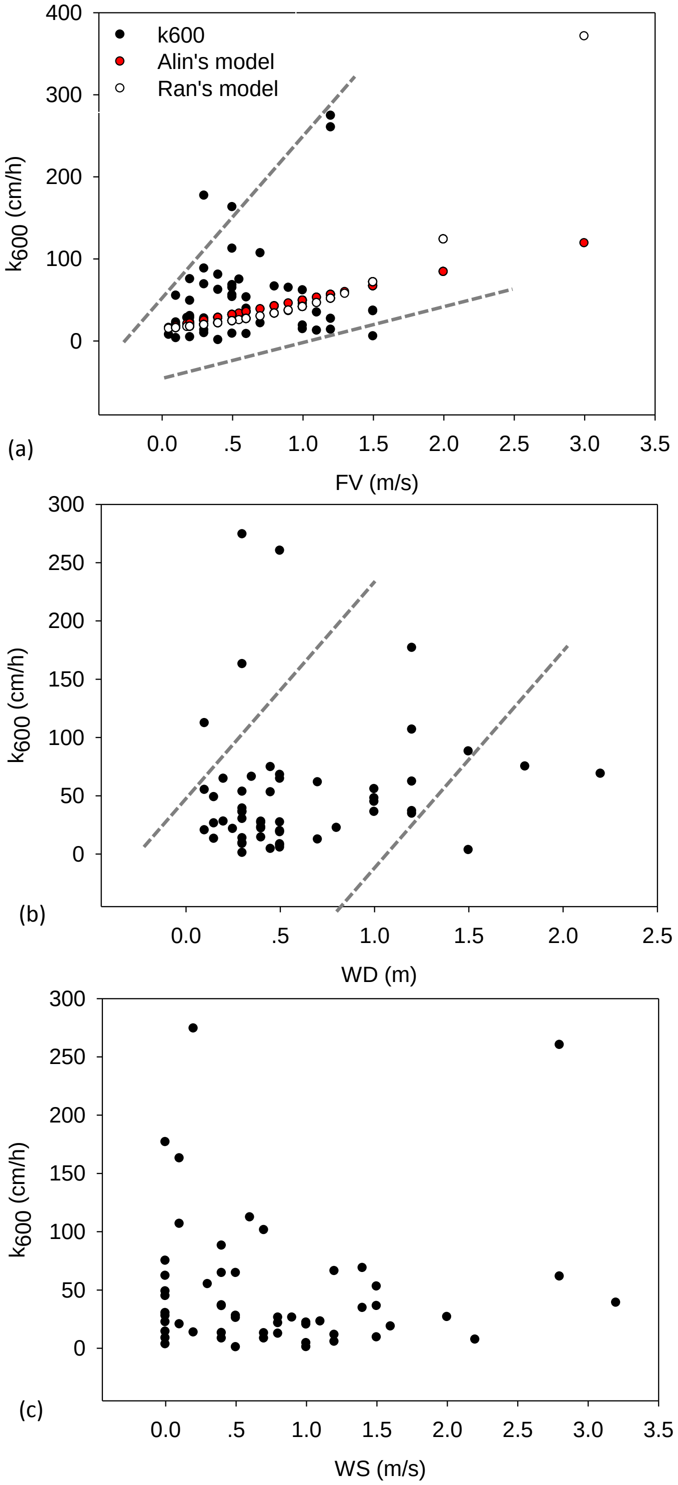


Fig. S4~~5~~. Plots of k_{600} and flow velocity (FV, unit in m/s) (a), WD (water depth, unit in m) (b), and WS (wind speed, unit in m/s) (c). The circle and red-dotted lines in Fig. 3a represent the model developed by Ran et al. (2015) and Alin et al (2011), respectively.

Table S1. Estimated k_{600} levels in our rivers.

		Daning	TGR rivers	Qijiang	Total
Mean		50.23	40.36	60.20	48.41
Median		50.46	30.04	25.81	32.22
Std. Deviation		20.10	37.60	78.85	53.20
Minimum		19.48	0.75	4.22	0.75
Maximum		80.50	176.71	274.12	274.12
Percentiles	25	35.809	12.628	13.213	16.303
	75	67.754	61.537	64.827	63.194
95% CI for Mean	Lower Bound	35.86	27.02	24.31	35.12
	Upper Bound	64.61	53.69	96.10	61.70

CI-Confidence Interval

Table S2. Comparison of k values adopted by previous studies.

River	Sites	Climate	k~~K~~$_{600}$ cm/h	Mean pCO_2 (µatm)	CO_2 degassing flux mmol/m^2/d	References
Longchuan River	China	Subtropic	8	2100	156.2	Li et al., 2012
Upper stream of Maotiao River	China	Subtropic	10	3740	294.5	Wang et al., 2011
Pearl River	China	Humid subtropic	8-15	2600	189-356	Yao et al., 2007
Yangtze (Datong)	China	Subtropic	8	1297	38-148	Wang et al., 2007
Rivers in TGR area*	China	Subtropic	48.4±53.2	846 ±819	133.1 ±269.1	This study
Yangtze*	China	Subtropic	38 ±40	1000-2035		Liu et al., 2017
Yellow river network*	China	Temperate	42.1± 16.9	2810±1985	856±409	Ran et al., 2015
Wuding*	China	Temperate	29-37	881	116-218	Ran et al., 2017

*k levels are from measurements

带格式的: 下标

	Current velocity m/s	Water depth m/s	Wind speed m/s	k~~K~~$_{600}$ cm/h	Reference
Mekong tributary	0.39±0.28	0.9±0.6	0.7±0.6	23.3±17.3	Alin et al., 2011
Yellow	1.8		1.8 (1.2-2.3)	42±17	Ran et al., 2015
Yangtze	1.2±1.5		1.2±1.1	38±40	Liu et al., 2017
Mekong stem	0.92±0.42		1.8±1.2	15±9	Alin et al., 2011

带格式的: 下标

Table S3. CO_2 emission from total rivers sampled in the study.

(a) Upscaling using CO_2 areal flux by FC.

	Catchment Area km^2	Water surface km^2	CO_2 areal flux $mmol/m^2/d$	CO_2 emission Tg CO_2/y
Daning	4200	21.42	122.0 ± 239.4	0.042
Qijiang	4400	30.8	50.3 ± 177.2	0.025
TGR river	50000	377.78	217.7 ± 334.7	1.321
Total				1.39

(b) Upscaling using determined k_{600} average and models (whole dataset are used here).

		From determined k_{600} mean	Flow velocity-based model (Fig. 4b) (numbers in bracket is from the revised model; Fig. 4c)	Water depth-based model (Fig. 4a)	Alin's model
Mean		1.37	3.29 (1.40)	1.08	1.06
S.D.		1.28	3.08 (1.31)	1.01	1.00
95% CI for Mean	Lower Bound	0.89	2.15 (0.91)	0.71	0.69
	Upper Bound	1.84	4.43 (1.87)	1.46	1.43

A total water area of approx. 430 km^2 for all tributaries (water area is from Landsat ETM+ in 2015).

Table S4. Monsoonal sampling effects on annual emission

	Monsoonal season	Dry season	Monsoonal/Dry
pCO_2 (μatm)	846	1043	0.81
ΔpCO_2 (μatm)	446	643	0.69
k_{600} (cm/h)	48.4	31.3	1.55
CO_2 areal flux ($mmol/m^2/d$)	196.9	183.4	
Water area (km^2)	602	430	
Emission (Tg CO_2)	0.96	0.63	

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Dataset APPENDIX TABLE A1

(Daning River)

Time	River	Latitude	Longitude	Air pressure	a.s.l.	T(water)	pH	EC(Cond)	EC(SPC)	TDS	DO	DO	River width	River depth	Water velocity	Alkalinity	DOC	TDN	TDP
				hPa	m	^{0}C		μs/cm	μs/cm	mg/L	%	mg/L	m	m	m/s	μeq/L	mg/L	mg/L	μg/L
2016.5.19;9:21	XJH1	31^{0}23'54"	109^{0}31'25"	979	290	15.58	8.27	266.8	325.3	211	81.3	8.09	5	0.5	0.5	3200	1.09	1.37	30.15
2016.5.19,10:25	XJH2	31^{0}23'48"	109^{0}30'48"	982.1	240	18.294	8.06	353.1	405	263	88.9	8.36	10	0.45	0.55	3360	1.57	1.21	25.13
2016.5.19,11:00	XJH3	31^{0}24'20"	109^{0}31'26"	988	160	18.138	8.29	347.2	399.5	260	86.1	8.12	10	0.5	0.3	3360	1.74	1.07	45.23
2016.5.19,11:42	XJH4	31^{0}23'54"	109^{0}33'33"	989	210	18.275	8.39	349	400.4	260	83.7	7.87	10	0.3	0.5	3360	1.32	1.20	30.15
2016.5.19,15:20	HXH1	31^{0}27'48"	109^{0}21'14"	967	340	14.326	8.58	151.5	190.2	124	87	8.91	15	0.5	0.8	2320	1.15	0.54	30.15
2016.5.19,16:00	HXH2	31^{0}28'49"	109^{0}26'0"	973	320	14.136	8.53	159.1	200.7	130	84	8.63	8	2	1	2240	0.71	0.16	75.38
2016.5.19,16:32	HXH3	31^{0}27'54"	109^{0}30'8"	979	260	14.473	8.53	162.9	203.8	132	84.4	8.61	10	0.5	1	2240	0.67	0.38	55.28
2016.5.19,17:07	HXH4	31^{0}27'46"	109^{0}36'49"	987	260	15.177	8.55	211.7	260.6	169	84.8	8.51	20	0.8	0.8	2400	1.28	0.36	40.20
2016.5.19,17:32	DNH1	31^{0}27'35"	109^{0}38'6"	1000	230	15.221	8.56	220.4	271.1	176	84.6	8.48				2400	1.39	0.12	85.43
2016.5.20;10:19	DXH1	31^{0}39'48"	109^{0}33'7"	958	540	16.248	8.53	218.4	262	170	84.1	8.25	0.5	0.1	0.1	2800	1.41	0.18	40.20
2016.5.20;10:55	DXH2	31^{0}39'46"	109^{0}33'6"	965	560	15.106	8.55	226.3	279	181	84.4	8.49	50	0.3	0.4	2800	1.21	0.33	50.25
2016.5.20;11:20	DXH3	31^{0}39'3"	109^{0}39'46"	970	430	14.55	8.62	222.6	278.1	181	83.6	8.51	2	0.3	0.5	2880	0.89	0.26	75.38
2016.5.20;11:54	DXH4	31^{0}38'3"	109^{0}40'11"	973	130	16.153	8.6	241	289.5	188	82.5	8.12	25	0.5	0.5	2800	1.20	0.17	55.28
2016.5.20;14:09	DXH5	31^{0}36'16"	109^{0}40'34"	974	360	16.389	8.71	242.2	289.9	188	87.6	8.57	20	1	1	2720	0.98	0.66	15.08
2016.5.20;14:40	DXH6	31^{0}34'23"	109^{0}39'11"	981	320	16.417	8.71	238.5	285.2	185	88.6	8.66	25	0.5	0.3	2800	1.34	0.33	30.15
2016.5.20;15:03	DNH2	31^{0}31'31"	109^{0}37'18"	985	270	14.886	8.62	189.4	234.7	153	89.7	9.07	50	1	1.2	2880	0.66	0.12	40.20
2016.5.20;15:35	DNH3	31^{0}28'1"	109^{0}38'9"	987	260	16.689	8.73	193.5	229.9	149	94.4	9.16	50	1	0.5	2480	0.97		40.20
2016.5.20;16:23	XXH1	31^{0}32'16"	109^{0}37'5"	983	260	14.772	8.67	176.2	219	142	92.8	9.4	25	1.5	1	2400	1.02	0.21	95.48
2016.5.20;16:53	DXH7	31^{0}32'13"	109^{0}36'39"	984	280	15.984	8.61	228.9	276.5	180	89	8.78	8	1.5	2	2560	0.99	0.08	50.25
2016.5.20;18:15	DXH8	31^{0}39'48"	109^{0}40'15"		500	15.84	8.56	279.1	338.2	220	82.8	8.2	2	0.2	0.2	3120	1.19	0.24	75.38

2016.5.21;10:18	XXH2	31°39'48"	109°10'8"	922	860	11.671	8.54	131.5	176.4	115	81.5	8.84	4	0.5	0.6	2640	0.95	0.04	45.23
2016.5.21;10:57	XXH3	31°38'11"	109°11'58"	888		15.278	8.73	239.1	293.5	191	82	8.22	1.5	0.1	0.2	2400	0.78	0.42	30.15
2016.5.21;11:11	XXH4	31°37'46"	109°14'17"	947	670	13.934	8.71	187.7	238	155	85.8	8.85	15	1	0.2	2880	1.62	0.46	55.28
2016.5.21;13:15	XXH5	31°37'12"	109°4'31"	912	930	13.93	8.57	187.6	237.9	155	79.9	8.24	4	0.2	0.6	2800	0.87	0.45	70.35
2016.5.21;13:47	XXH6	31°36'10"	109°7'53"	934	780	12.688	8.5	135.8	177.6	115	83.2	8.82	8	0.5	0.4	2800	0.33	0.17	85.43
2016.5.21;14:24	XXH7	31°37'7"	109°8'27"	929	780	13.358	8.56	200.5	257.9	168	82.8	8.65	3	0.5	0.5	2960	1.59	0.07	85.43
2016.5.21;14:55	XXH8	31°36'29"	109°9'59"	939	700	14.696	8.46	210.7	262.4	171	83	8.42	3	0.3	0.6	2880	0.53	0.08	25.13
2016.5.21;15:21	XXH9	31°35'21"	109°12'16"	944	660	14.319	8.57	151.7	190.6	124	83.9	8.58	25	0.3		2240	0.87	0.11	30.15
2016.5.21;15:43	XXH10	31°35'23"	109°16'4"	951	600	15.092	8.6	155.3	191.6	125	85	8.55	25	0.4	0.3	2240	0.69	0.08	25.13
2016.5.21;16:16	XXH11	31°35'23"	109°17'28"	945	660	13.219	8.46	125.9	162.5	106	84.9	8.9	4	0.2	0.4	1920	0.70	0.01	35.18
2016.5.21;17:05	XXH12	31°34'37"	109°25'54"	967	490	16.142	8.87	198.9	239.5	156	103.6	10.19				2720	0.95	0.01	65.33
2016.5.21;18:07	XXH13	31°33'54"	109°34'13"	982	340	14.455	8.46	176.3	220.8	144	92.1	9.39	8	2.5	3	2560	1.13	0.33	65.33
2016.5.22;9:26	DNH4	31°23'25"	109°37'27"	997	210	16.187	8.5	213.2	256.2	166	86.8	8.54	25		0.2	2800	1.19	0.09	30.15
2016.5.22;9:44	XJH5	31°23'26"	109°37'25"	997	180	18.37	8.35	358.7	410.7	267	86.1	8.08	8		0.4	3360	1.32	1.16	70.35
2016.5.22;10:15	DNH5	31°23'38"	109°37'49"	997	230	19.618	8.68	292	325.5	212	99.9	9.15	2	0.2	0.4	3200	1.10	0.72	45.23

(TGR rivers)

Time	Code	Latitude	Longitude	asl	T(air)	Wind speed	T(water)	pH	ORP	EC	DO	DO	River width	River depth	Water velocity	Alk	DOC	DTN	DTP
				m	°C	m/s	°C		mv	μs/cm	mg/L	%	m	m	m/s	μeq/L	mg/L	mg/L	μg/L
2016.6.21;13:41	BLXX	29°30'30"	106°59'26"	432	39.1	0.6	29.1	7.57		233						1880	26.08	3.88	76.12
2016.6.21;15:00	BLTH	29°26'47"	107°06'11"	700	33.2	1.1	24.7	7.47		225			15	0.4	0.2	1976	37.48	7.03	120.89

2016.6.22;09:04	BNB	30°01'50"	108°11'32"	616	28.2	0	20	7.96		125.8			11	0.3	0.5	1040	14.31	1.94	82.84
2016.6.22;10:00	BLH	29°17'53"	108°55'00"	518	29.9	0.4	23.5	7.84		202.3			80	0.5	0.6	1840	22.03	3.09	143.28
2016.6.22;12:00	BHJH	30°22'07"	107°59'10"	181	33.5	1	29	7.79		418			4	0.3	0.4	3128	13.56	3.09	138.81
2016.6.22;13:14	BRXH	30°30'10"	108°03'45"	215	36.1	0.5	33.5	7.82		344						3112	31.80	7.19	108.95
2016.6.22;18:00	BDH	31°18'07"	108°28'52"	193	32.5	2.8	29.3	8.25	128	976	8.49	115.9	6	0.7	1	2152	10.30	1.71	44.78
2016.6.23;10:00	BNH	31°06'21"	108°12'02"	43	31.9	0	30.4	8.06	159.3	359	6.61	90.6	6	0.15	0.2	4032	34.82	3.45	140.30
2016.6.23;11:25	BPLH	31°01'18"	108°18'58"	152	33.3	0.9	29.4	8.15	155.9	269	6.07	83.4				1688	10.47	2.96	140.30
2016.6.23;15:47	BMDX	30°44'15"	108°36'07"	217	31.6	0.8	22.2	8.42	115.2	203	8.34	101				1640	7.54	3.22	29.85
2016.6.23;16:22	BSBH	30°39'42"	108°36'07"	256	32.5	0	28.9	8.61	81	240	7.63	104.1				2536	16.56	3.58	62.69
2016.6.23;18:56	BDXH	30°48'04"	108°51'47"	132	31.2	0.7	29.4	8.21	58.9	870	8.36	112.8				3256	10.00	1.81	41.79
2016.6.24;14:34	BCTH	30°53'48"	109°02'20"	138	25.4	1.2	22.2	7.89	99.1	102.3	8.43	99.2	4	0.5	1.5	1368	9.95	2.66	95.52
2016.6.25;12:00	BMXH	30°45'40"	109°15'37"	558	22.3	2.8	17.7	8.14	85.5	211	8.98	98.9	20	0.4	1.3	2488	7.30	6.34	77.61
2016.6.25;13:30	BMXH-2	30°51'44"	109°23'34"	317	23.5	0.6	17.8	8.19	92.2	320	9	96.7	25	0.5	2	2704	8.68	4.43	71.64
2016.6.25;15:10	BJPH	30°53'19"	109°33'38"	149	23.3	1.4	17.6	8.38	64.1	265	8.89	99.4	30	1	2	2392	10.23	4.34	50.75
2016.6.26;12:23	BGDH	30°57'15"	109°50'36"	366	27	0	16.1	8.26	38.3	278	9.44	98.3	30	1.5	1	3016	9.92	1.97	59.70
2016.6.26;15:36	BBLH	30°56'09"	110°00'17"	197	25.1	0	16.6	8.36	7.9	279	8.54	94.3	30	0.5	1.5	3112	7.45	4.66	98.51
2016.6.27;11:32	BWFH	30°56'26"	110°16'15“	402	28.1	1.4	16.3	8.4	9.03	303	9.03	101.2	6	1.2	1.1	2520	8.53	5.42	57.46
2016.6.27;14:07	BLFD	30°56'16"	110°35'19“	216	30.4	0.8	21.3	8.35	48.6	290	8.56	99.5	4	0.7	1.1	2216		4.39	68.66
2016.6.27;15:00	BQGH	30°54'45"	110°29'20“	237	31.2	0	22.8	8.45	29.1	324	8.61	101.4	7	0.4	0.2	2904	9.81	6.88	80.60
2016.6.27;16:48	BJZH	30°54'30"	110°39'58“	220	29.5	0	22.1	8.45	33.5	320	8.14	101.2	7	0.4	1	2664	8.64	6.18	47.76
2016.6.27;18:00	BJWX	30°46'41"	110°49'00“	152	28.3	0.1	18.3	8.39	25.5	280	9.52	103.8	25	1.2	0.7	2616	8.33	4.76	104.48
2016.6.27;18:30	BJWX-2	30°47'04"	110°47'44“	242	29.2	2	20.1	8.33	21.8	314	8.63	98.2	3	0.4	1.2	2616	8.65	4.07	73.13
2016.6.28;10:20	BMPH	30°46'47"	110°57'44“	137	29.3	1	21.1	8.35	32.5	353	8.63	96.8	1.2	0.4	0.4	3352	11.20	3.61	161.19
2016.6.28;14:40	BGLH	31°07'27"	110°49'21“	120	33.9	3.2	21.5	8.52	-1.6	321	8.3	101.8	15	0.3	0.6	3008	22.38	8.76	44.78
2016.6.28;16:20	BXXH	31°20'07"	110°45'02“	185	33.1	1.5	19.9	8.51	31.9	330			12	0.3	0.9	3200	11.82	2.34	176.12
2016.6.29;10:44	BLTH-XXGQ	30°13’32"	110°32'47“	745	26.9	1.1	17.6	8.57	64.3	208	8.26	98.7	7	1.2	0.3	2616	7.08	1.81	191.04

2016.6.29;11:32	BSNX	31°13'22"	110°17'39"	125	33.7	0.4	19.3	9.38	8.6	232	22	1.2	1.5	2368	7.44	1.91	32.84
2016.6.29;14:32	BLXB	31°10'45"	110°16'07"	171	37.8	0.4	29.8	8.49	-2	378	6	0.2	0.9	3600	11.09	4.63	58.21
2016.6.29;16:31	BBCH	31°12'47"	110°08'03"	631	33.8	0	27.7	8.24	3.5	438	5	0.3	0.2	4480	21.81	9.03	86.57
2016.6.29;17:29	BCaoT	31°05'31"	109°38'46"	193	33.6	1.5	28.7	8.54	96.8	501	4	0.3	0.3	3176	11.23	8.70	202.98
2016.6.30;10:40	BCJH	31°07'56"	109°21'21"	129	30.7	0.4	22.8	8.5	20.3	371	22	1	1.5	2616	8.35	3.61	128.36
2016.6.30;11:10	BMeiXH	31°07'37"	109°15'54"	140	27.3	5	21.6	8.38	22.7	218	30	0.4	1.5	1968	9.53	4.60	85.82
2016.6.30;11:40	BCJB	31°07'35"	109°10'06"	288	27.8	0.5	22.9	8.34	23.8	162.8	35	1.2	1.5	1456	7.22	6.24	73.13
2016.6.30;15:21	BTXH	31°14'17"	108°48'00"	164	25	0.8	17.5	8.3	9.7	162	25	1,5	0.8	1936	8.28	0.76	38.81
2016.7.01;10:31	BGaoTH	30°17'31"	107°27'33"	331	25.9	0.7	23.3	7.93	32.3	249				2290	9.28	6.93	273.13
2016.7.01;11:28	BGJH	30°12'35"	107°29'47"	354	28.5	0	23.3	8.14	29.9	274	15	2.2	0.8	2270	12.18	3.52	101.49
2016.7.01;12:30	BQXH	30°06'21"	107°31'06"	378	29.2	1.4	23.4	8.17	69.3	258	25	2.2	0.3	2260	11.80	4.57	147.76
2016.7.01;14:41	BLXH	29°50'30"	107°08'58"	190	33.3	0	25.9	7.51	54.6	379	30	1.2	0.3	2340	12.30	8.08	191.04
2016.7.01;15:54	BTXH	30°01'59"	107°07'23"	328	29.6	0.4	24.7	7.86	40.7	377	15	1.5	0.3	2100	13.95	10.54	298.51
2016.7.02;10:50	BWTH	29°51'42"	106°50'08"	194	32.2	0	23.3	8.16	36.3	300	30	1.2	0.4	2664	10.59	2.01	89.55
2016.7.02;11:17	BYLH	29°51'54"	106°50'49"	149	34.6	0	24.4	8.17	37.6	264	45	1.8	0.2	1880	9.13	1.38	144.78
2016.7.02;11:37	BDH-YB	29°54'59"	106°54'51"	134	33.9	0	27.6	7.99	59.1	322	45	1.5	0.1	2296	10.86	4.93	94.78
2016.7.02;14:49	BCTH-YL	29°32'59"	106°43'35"	181	35.5	0.4	27.7	8.18	44	270				2008	10.72	4.57	135.82
2016.7.02;16:04	BHXH	29°23'19"	106°35'37"	208	31.2	0	23.2	8.08	73.2	303	20	0.8	0.1	2176	9.86	6.37	144.78
2016.7.02;17:25	BYPH	29°16'46"	106°34'01"	118	31	0	24.2	8.1	74.9	232	35	1	1	1912	8.43	3.55	91.79
2016.7.02;18:23	BLTH-BSY	29°30'35"	106°22'00"	271	31.3	0.4	28.1	7.87	12.6	462	6	0.6	0.2	3144	14.15	6.24	159.70

(Qijiang River)

Date	Time	River	T(air) ^{0}C	Wind speed m/s	T(water) ^{0}C	pH	EC µs/cm	TDS mg/L	River width m	River depth m	Water velocity m/s	Alk µeq/L	DOC mg/L	TDN mg/L	TDP mg/L
2016.8.15	13:30	BZD1	39.6	0.5	30	8.41	351.6	229	3	20	0.18	2920	1.34	2.58	10.05
	14:30	BZD2	40.5	1	25	8.59	317	206	20	40	0.5	2472	1.61	2.09	10.05

	15:30	BZD3	37.3	0.7	31	8.67	301.8	196				2656	2.32	1.99	5.03
	15:40	BZD4	34.4	0.6	31	8.52	269.1	175				1824	14.88	0.53	5.03
	16:10	BZD5	33.7	1.6	32	8.52	282.5	184	20	80	0.6	2336	1.48	0.87	5.03
	16:45	BZD6	34.4	2.2	30	8.3	448.2	292	2	50	0.05	2216	1.65	1.89	5.03
2016.8.16	9:10	BSY1	31	0.4	22	8.64	322.3	210	3	60	0.8	3056	2.00	1.01	5.03
	9:50	BSY2	33.1	0.5	23.3	8.49	345	224	6	50	0.5	2896	1.65	0.97	10.05
	10:25	BSY3	31.6	1	23.5	7.94	374.7	244	20	45	0.2	2520	1.39	1.12	
	13:40	BSK1	32.2	1	26.8	8.63	513.8	334	1	15	0.8	2552	4.46	1.99	5.03
	14:20	BSK2	36.1	1.2	21	8.27	338.6	220	2	35	0.8	2288	32.01	2.05	10.05
	15:00	BSK3	41.8	0.5	30.5	8.67	286.7	186	12	50	0.3	2600	3.72	0.97	10.05
	15:40	BSK4	35.4	1.3	31	8.16	416.2	271	1.5	60	0.4	1896	1.88	0.99	5.03
	16:00	BSK5	38.3	0.8	26.5	8.68	334.5	218	3	25	0.7	2048	2.53	1.57	10.05
	16:20	BSK6	40	0.8	29	8.56	381.2	248	2	50	1	1728	1.50	1.29	20.10
	17:00	BSK7	38.1	0.6	32.3	8.76	335.2	218	2	10	0.5	1984	1.65	0.86	5.03
2016.8.17	9:30	BSK8	30.2	0.8	23.3	8.48	368.9	240	11	15	0.3	3112	1.64	1.55	10.05
	10:00	BSK9	32.3	0.4	26	8.4	362.9	236	11	15	0.3	2632	1.72	1.45	45.23
	11:00	BSK10	34.4	0.2	29	8.56	367.4	239	20	35	0.2	2368	1.86	1.29	
	11:24	BSK11	35.5	2.8	29.1	8.52	410.8	207	5	50	1.2	2544	1.40	0.71	5.03
	12:45	BSK12	38.5	1.6	30	8.46	474.8	309	12	50	1	2904	1.44	1.33	40.20
	15:00	BYD1	35	0.3	29.5	8.45	663.6	431	1	10	0.1	2240	2.25	2.38	10.05
	16:30	BYD2	32.7	0.8	30	8.58	217.6	142	4	10	0.1	1808	4.74	1.03	5.03
	17:45	BYD3	33	0.5	29	8.45	1087.9	707				4488	2.00	1.62	15.08
	18:14	BYD4	32.6	0.2	32	8.55	1099.5	716	5	30	1.2	3592	2.49	1.58	15.08
	17:00	BYD5	32	0.2	30.5	8.53	1051.9	684	4	30	1.2	4080	1.73	1.26	5.03
	19:30	BQJ1	32.3	1.5	30	8.56	623	405	40	45	0.6	3784	1.83	1.43	15.08
2016.8.18	10:00	BQJ2	37	1.2	29.5	8.35	396.4	258				2416	2.99	1.26	95.48

11:20	BPH1	33.2	0.7	31.9	8.49	1350.3	878				3856	8.98	1.47	25.13
15:00	BSX1	41.4	0.1	32	8.42	150.9	98				600	3.46	0.64	5.03
16:00	BSX2	41.2	1	34	8.23	179.9	117	8	10	0.1	1424	3.31	1.33	30.15
16:30	BQG3	37.1	0.1	32	8.43	422.2	274	15	30	0.5	2376	2.46	1.71	85.43