

Supporting Information

The carbon footprint of a Malaysian tropical reservoir: measured versus modeled estimates highlight the underestimated key role of downstream processes

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Table S1: Summary of the mean, standard deviation (SD), range (min and max), and number of observation (n) of physical and chemical variables measured at the surface of the reservoir and its inflows.

Variables	Units	Reservoir					Inflows				
		Mean	SD	Min	Max	n	Mean	SD	Min	Max	n
Water temperature	°C	31	0.8	24	32	134	27	2.4	24	30	10
DOC	mg.L ⁻¹	0.9	0.2	0.3	1.8	115	0.8	0.4	0.4	1.3	8
TP	µg.L ⁻¹	5.9	2.4	2.8	23.3	114	20.7	7.6	11.7	36	8
TN	mg.L ⁻¹	0.11	0.04	0.06	0.33	115	0.14	0.04	0.07	0.19	8
Chla	µg.L ⁻¹	1.3	0.7	0.3	4.0	112	2.1	1.7	0.1	4.4	9
CO ₂ diffusion	mmol.m ⁻² .d ⁻¹	7.7	18.2	-30.8	80.0	129	137.3	192.4	9.7	593.9	8
CH ₄ diffusion	mmol.m ⁻² .d ⁻¹	0.6	0.6	0.03	3.7	129	30.7	37.4	1.2	113.4	8
N ₂ O diffusion	nmol.m ⁻² .d ⁻¹	-0.2	2.1	-7.1	2.7	15					0
[CO ₂]	µmol.L ⁻¹	16.3	5.2	6.8	36.2	132	60.1	34.9	10.1	113.5	10
[CH ₄]	µmol.L ⁻¹	0.2	0.2	0.0	1.2	137	6.6	4.3	0.6	14.8	10
[N ₂ O]	nmol.L ⁻¹	5.6	1.2	3.9	7.8	15	6.1		5.2	7.0	2

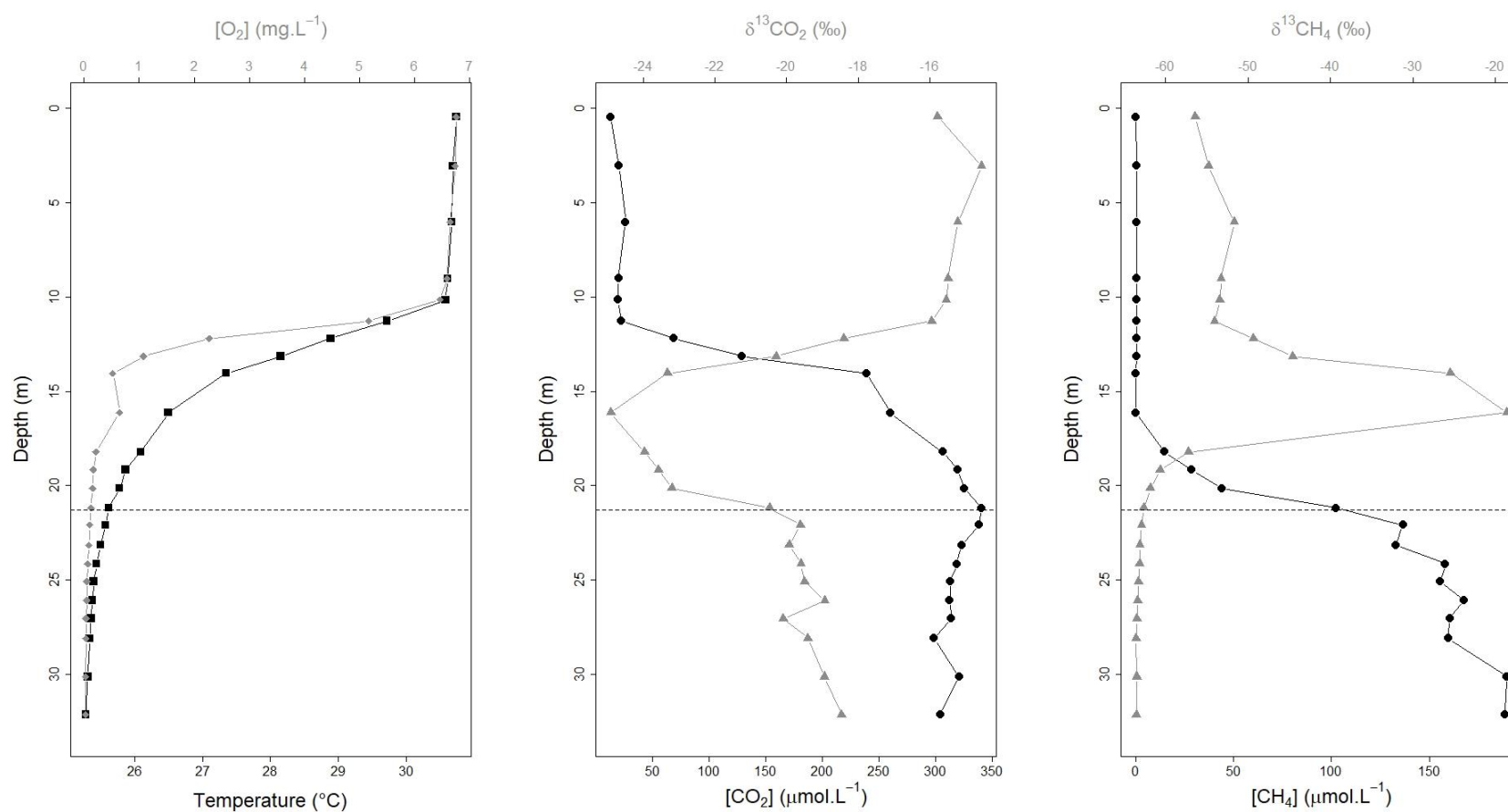


Figure S1: Profile example of water temperature (black squares), dissolved oxygen concentration (gray diamonds), CO_2 and CH_4 concentrations (black circles) and isotopic $\delta^{13}\text{C}$ signature (gray triangles) in the main basin of the reservoir right upstream of the dam in Aug 2018. The horizontal dashed line represent the water withdrawal depth.

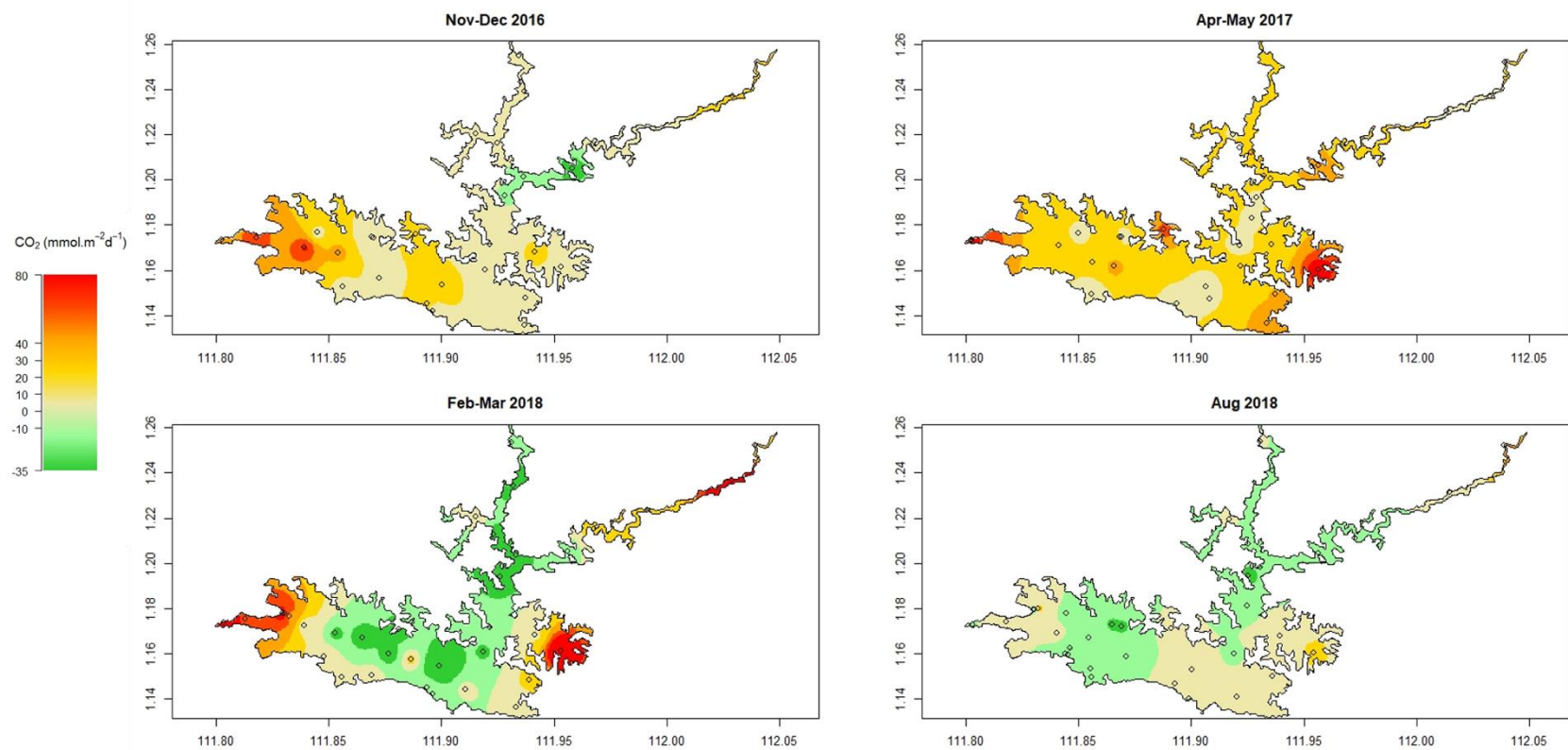


Figure S2: Maps of the spatially interpolated surface CO₂ diffusive flux in Batang Ai reservoir for each sampling campaign. Unit is in $\text{mmol.m}^{-2}.\text{d}^{-1}$. Graph axes are the spatial coordinates (latitude and longitude).

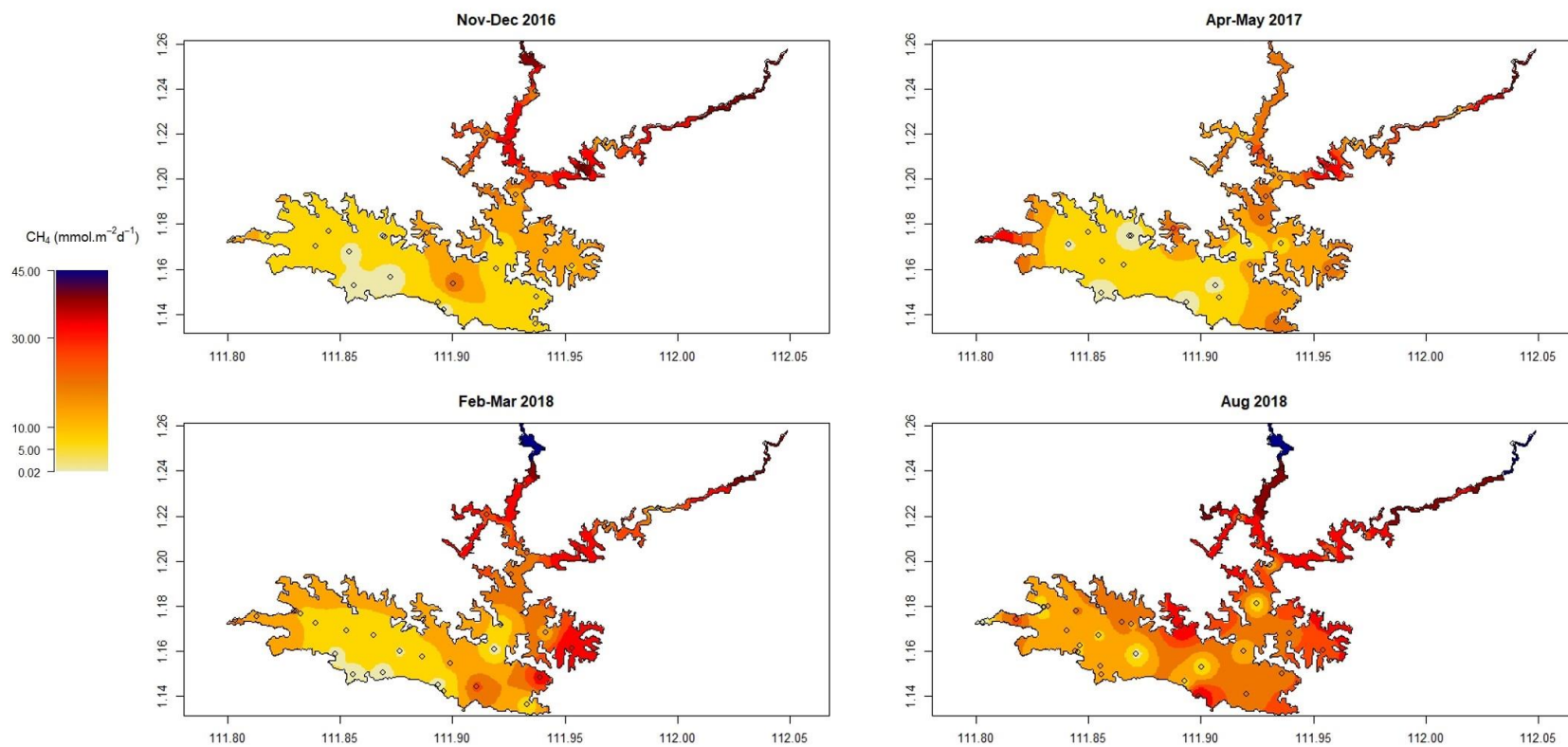


Figure S3: Maps of the spatially interpolated surface CH_4 diffusive flux in Batang Ai reservoir for each sampling campaign. Unit is in $\text{mmol.m}^{-2}.\text{d}^{-1}$. Graph axes are the spatial coordinates (latitude and longitude).

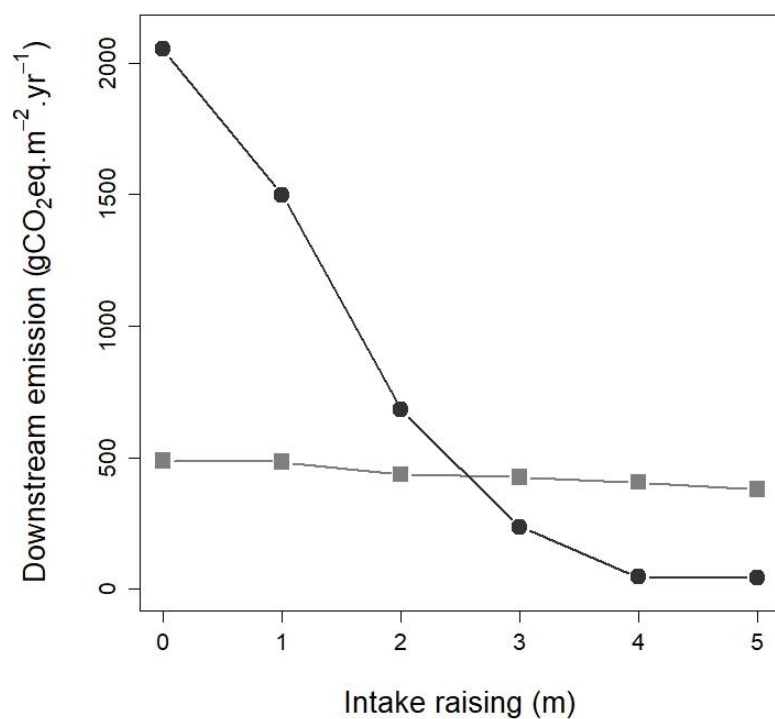


Figure S4: Simulated downstream emissions (including degassing) of CO₂ (gray squares) and CH₄ (dark circles) under different water withdrawal depth raise. Simulated emissions do not take into account CH₄ oxidation in the outflow river.

Table S2: Equations used to derive modeled CO₂ and CH₄ emissions based on the G-res and Barros et al. models. Input variables are described in Table S3.

	G-res model	Barros et al. model
log₁₀ (CO₂ diffusion)	1.7892 - 0.3364 log ₁₀ (Age) + 0.0400 Effective temperature CO ₂ + 0.06918 log ₁₀ (Reservoir area) + 0.0216 Soil C content + 0.1472 log ₁₀ (TP)	3.06 - 0.16 log ₁₀ (Age) - 0.01 Latitude + 0.41 log ₁₀ (DOC input) - log ₁₀ (400)
log₁₀ (CH₄ diffusion)	0.8804 - 0.0116 Age + 0.6068 log ₁₀ (% littoral area / 100) + 0.04828 Effective temperature CH ₄	1.33 - 0.36 log ₁₀ (Age) - 0.32 log ₁₀ (Mean depth) + 0.39 log ₁₀ (DOC input) - 0.01 Latitude
log₁₀ (CH₄ ebullition)	-0.98574 + 1.0075 log ₁₀ (% littoral area / 100) + 0.04928 (Cumulative global horizontal radiance)	-
CH₄ degassing	10 ^{(-5.5029 + 2.2857 log₁₀ (Modeled CH₄ diffusion) + 0.9866 log₁₀ (Water residence time))} × Discharge / Reservoir area	-

Table S3: Input variables values, units, and source used in the modeling of Batang Ai CO₂ and CH₄ emissions.

Input variable	Value	Unit	Source
% Littoral area	5.15	%	$(1 - (1 - (3 / \text{Maximum depth})) ((\text{Maximum depth} / \text{Mean depth}) - 1)) \times 100$
Age	33	year	Mean reservoir age at sampling years (2016 - 2018)
Annual runoff	2219	mm.yr ⁻¹	UNH/GRDC Composite Runoff Fields V1.0 ⁵²
Catchment area	1142	km ²	GranD database V1.3 ¹
Cumulative global horizontal radiance	56.4	kWh.m ⁻² .yr ⁻¹	Surface meteorology and Solar Energy ⁵³
Discharge	106	m ³ .s ⁻¹	Average during the four sampling times (provided by Sarawak Energy)
DOC	0.8	mg.L ⁻¹	Average of measured surface values in this study
DOC input	81.93	mg.m ⁻² .d ⁻¹	$(\text{DOC} \times \text{Volume}) / (\text{Reservoir area} \times \text{Water residence time} \times 1000 \times 365)$
Effective temperature CH ₄	26.37	°C	Mean annual temperature corrected for the effect on CH ₄ from G-res tool ⁵⁴
Effective temperature CO ₂	26.37	°C	Mean annual temperature corrected for the effect on CO ₂ from G-res tool ⁵⁴
Latitude	1.16	Decimal degrees	Google earth
Maximum depth	85	m	GranD database V1.3 ¹ – Height dam wall
Mean depth	34.4	m	GranD database V1.3 ¹
Modeled CH ₄ diffusion	134	gCO ₂ eq.m ⁻² .yr ⁻¹	Average over 100 years of CH ₄ diffusion modeled based on the G-res equation (Table S2)
Reservoir area	68.4	km ²	GranD database V1.3 ¹
Soil C content	2.41	kgC.m ⁻²	Estimated based on Wasli et al. 2011
TP	6.77	µg.L ⁻¹	Average of measured surface values in this study
Volume	2360 x 10 ⁶	m ³	GranD database V1.3 ¹
Water residence time	0.93	year	$(\text{Mean depth} \times \text{Reservoir area}) / (\text{catchment area} \times \text{Annual runoff}) \times 1000$

Modeling downstream river oxidation

The following section describes the conceptual framework underlying the isotopic model used to estimate CH₄ oxidation in the outflow of the reservoir. For that, we assume that the only source of water and CH₄ to the outflow (starting right downstream of the power house) is the reservoir discharge. We also assume oxidation and evasion to the atmosphere are the only two loss processes for CH₄ in the outflow, and that both reactions have a constant specific rate (they are a linear function of CH₄ concentration). When following a parcel of water travelling along the river, the change in CH₄ concentration can thus be described as:

$$\frac{d[CH_4]}{dt} = -k_{oxi}[CH_4] - k_{eva}[CH_4] \quad (Eq.S1)$$

with k_{oxi} and k_{eva} the rate coefficients of CH₄ oxidation and evasion respectively, and $[CH_4]$ the CH₄ concentration. Integration of Eq.S1 yields the following generic time-depend solution:

$$[CH_4]_t = [CH_4]_0 e^{-(k_{oxi}+k_{eva})t} \quad (Eq.S2)$$

The instantaneous oxidation rate at a given time (t) or at a given point in the river is equal to:

$$oxidation\ rate = k_{oxi} [CH_4]_t \quad (Eq.S3)$$

Combining Eq.S2 and Eq.S3 yields the following equation:

$$oxidation\ rate = k_{oxi} [CH_4]_0 e^{-(k_{oxi}+k_{eva})t} \quad (Eq.S4)$$

Hence, the total amount of CH₄ oxidized between time 0 and time t (or between km 0 and t in the river) is derived from the integration of Eq.S4:

$$amount\ oxidized = \int_0^t k_{oxi} [CH_4]_0 e^{-(k_{oxi}+k_{eva})t} dt \quad (Eq.S5)$$

Simplifying Eq.S5 yields:

$$amount\ oxidized = [CH_4]_0 \left(\frac{k_{oxi}}{(k_{oxi} + k_{eva})} \right) (1 - e^{-(k_{oxi}+k_{eva})t}) \quad (Eq.S6)$$

The amount oxidized can also be calculated as the product of the original CH₄ concentration and the fraction of CH₄ oxidized (F_{ox}):

$$\text{amount oxidized} = [CH_4]_0 F_{ox} \quad (Eq. S7)$$

According to Eq.S6 and Eq.S7, we can derive F_{ox} from the following equation:

$$F_{ox} = \left(\frac{k_{oxi}}{(k_{oxi} + k_{eva})} \right) (1 - e^{-(k_{oxi} + k_{eva}) t}) \quad (Eq. S8)$$

In Eq.S8, the second term of the product can be rearranged using Eq.S2 to represent the remaining fraction of CH₄ concentration at time or km t:

$$F_{ox} = \left(\frac{k_{oxi}}{(k_{oxi} + k_{eva})} \right) \left(1 - \frac{[CH_4]_t}{[CH_4]_0} \right) \quad (Eq. S9)$$

Given a fractionation factor α for CH₄ oxidation, the behavior of the two CH₄ isotopes (¹²CH₄ and ¹³CH₄) can be described based on Eq.S2 as following:

$$[^{12}CH_4]_t = [^{12}CH_4]_0 e^{-(k_{oxi}^{12} + k_{eva}) t} \quad (Eq. S10)$$

$$[^{13}CH_4]_t = [^{13}CH_4]_0 e^{-\left(\frac{k_{oxi}^{12}}{\alpha} + k_{eva}\right) t} \quad (Eq. S11)$$

With [¹²CH₄] and [¹³CH₄] the concentrations of the two isotopes, and k_{oxi}¹² the rate coefficient of oxidation for the ¹²CH₄ isotope. The isotopic ratio of CH₄ concentration at t (R_t) can be derived from Eq.S10 and Eq.S11 as following:

$$R_t = \frac{[^{13}CH_4]_t}{[^{12}CH_4]_t} = \frac{[^{13}CH_4]_0 e^{-\left(\frac{k_{oxi}^{12}}{\alpha} + k_{eva}\right) t}}{[^{12}CH_4]_0 e^{-(k_{oxi} + k_{eva}) t}} \quad (Eq. S12)$$

By simplifying Eq.S12, R_t can be written as:

$$R_t = R_0 e^{(1 - \frac{1}{\alpha}) \cdot k_{oxi} t} \quad (Eq. S13)$$

The term t can be isolated from equation Eq.S13:

$$t = \frac{\ln(R_t/R_0)}{(1 - \frac{1}{\alpha}) \cdot k_{oxi}} \quad (Eq. S14)$$

Similarly, the term t can be isolated by rearranging Eq.S2:

$$t = \frac{\ln([CH_4]_t/[CH_4]_0)}{-(k_{oxi} + k_{eva})} \quad (Eq. S15)$$

Combining Eq.S14 and Eq.S15 results in:

$$\frac{\ln(R_t/R_0)}{(1 - \frac{1}{\alpha}) \cdot k_{oxi}} = \frac{\ln([CH_4]_t/[CH_4]_0)}{-(k_{oxi} + k_{eva})} \quad (Eq. S16)$$

Rearranging E.S16 yields:

$$\frac{-\ln(R_t/R_0)}{(1 - \frac{1}{\alpha}) \cdot \ln([CH_4]_t/[CH_4]_0)} = \frac{k_{oxi}}{(k_{oxi} + k_{eva})} \quad (Eq. S17)$$

Using Eq.S9 and Eq.S17, we can derive F_{ox} as:

$$F_{ox} = \frac{k_{oxi}}{(k_{oxi} + k_{eva})} \left(1 - \frac{[CH_4]_t}{[CH_4]_0} \right) = \frac{-\ln(R_t/R_0) \cdot (1 - [CH_4]_t/[CH_4]_0)}{(1 - \frac{1}{\alpha}) \cdot \ln([CH_4]_t/[CH_4]_0)} \quad (Eq. S18)$$

The isotopic ratios (R_t and R_0) can be converted to $\delta^{13}CH_4$ expressed in ‰ using the following standard isotopic equation calculation:

$$\delta^{13}CH_4 = \left(\frac{[^{13}CH_4]:[^{12}CH_4]}{[^{13}CH_4]_{std}: [^{12}CH_4]_{std}} - 1 \right) \times 1000 \quad (Eq. S19)$$

with $[^{13}CH_4]_{std}$: $[^{12}CH_4]_{std}$ the standard reference Pee Dee Belemite isotopic carbon ratio. The conversion of R_t and R_0 to $\delta^{13}CH_4$ in Eq.S18 results in the following equation for F_{ox} calculation:

$$F_{ox} = \frac{-[\ln(\delta^{13}CH_{4_t} + 1000) - \ln(\delta^{13}CH_{4_0} + 1000)] \cdot (1 - [CH_4]_t/[CH_4]_0)}{(1 - \frac{1}{\alpha}) \cdot \ln([CH_4]_t/[CH_4]_0)} \quad (Eq.S20)$$

Eq.S20 was used to determine F_{ox} and CH_4 oxidation (product of F_{ox} and $[CH_4]_0$) in Batang Ai river outflow using km 0 and 19 as a start and end points of the river stretch. Sampling of CH_4 concentration and isotopic signature was conducted at two other points within this river stretch (km 0.6 and 2.7). This additional sampling resolution can help test the adequacy of the model and its conceptual assumptions by recalculating CH_4 oxidation in individual segments (between 0-0.6 km, 0.6-2.7 km, and 2.7-19 km). If the model assumptions are correct (absence of other CH_4 source in the river, constant specific oxidation rate, and constant river velocity), the amount of CH_4 oxidized in the entire stretch (Ox_{total}) should equal the sum of the amount oxidized in each individual segment (Ox_{sum}). In two of the sampling campaigns (Apr-May 2017 and Feb-Mar 2018), the difference between Ox_{total} and Ox_{sum} was very small ($CV < 5\%$). In the two other campaigns, this difference was larger (CV of 12 and 35 % for Nov-Dec 2016 and Aug 2018 respectively), due to an additional CH_4 source causing occasional increases of CH_4 concentration along the stretch. Overall, model assumptions appear reasonable, however, to avoid overestimating the amount oxidized in cases of additional CH_4 sources, we considered Ox_{total} as the more suitable estimate, since it is less influenced by CH_4 addition in individual segments.