SUPPORTING INFORMATION

A gas-tracer experiment for evaluating the fate of methane in a coastal plain stream: Degassing versus in-stream oxidation

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Supplementary Materials Text

Previously published evasion rates range from 0.01-10^4 mmol m^{-2} d^{-1}.Total evasion rates (mass transfer of dissolved CH_4 plus ebullition-driven bubble transfer) measured by floating gas-flux collection chambers on eight Amazonian Rivers ranged from 0.01-40.0 mmol m^{-2} d^{-1}, with diffusive-flux calculations of evasion of dissolved methane ranging from 0.01-19 mmol m^{-2} d^{-1} [Sawakuchi et al., 2014]. Diffusive-flux calculations of evasion based on seasonal dissolved- CH_4 measurements on three rivers of the Ivory Coast ranged from 0.05 to 0.49 mmol m^{-2} d^{-1} [Kone et al., 2010]. Calculated diffusive-flux evasion from the Kuparuk River in arctic Alaska was 0.36 mmol m^{-2} d^{-1} [Kling et al., 1992]. Evasion rates of 0.03 to 0.82 mmol m^{-2} d^{-1} were measured in streams in eastern Tennessee using conservative solute and volatile gas tracers [Jones and Mulholland, 1998]. Total evasion rates measured with floating chambers at three tropical rivers downstream of reservoirs in French Guiana and Brazil ranged from about 10-100 mmol m^{-2} d^{-1} [Guerin et al., 2006]. Evasion rates estimated with eddy models and direct measurements of CH_4 loss for several Pacific Northwest rivers ranged from 0 to 20 mmol m^{-2} d^{-1} [Lilley et al., 1996]. An average evasion rate estimated for the Ogeechee River (Georgia) using statistical models was 6 mmol m^{-2} d^{-1} [Pulliam and Meyer, 1992]. A study of a 1st order peatland stream in Scotland determined that CH_4 evasion rates were inconsequential compared with other carbon sinks (<0.2% of CO_2 evasion) [Hope et al., 2001]. Total evasion measured with floating chamber seasonally at four sites along the Sitka Stream in the Czech Republic ranged from 0.09 to 2.3 mmol m^{-2} d^{-1} [Rulik et al., 2013].

Previously published methane oxidation (MOX) rates range from 1 to 47,000 nmol L^{-1} d^{-1}. MOX rates for the Hudson River using sequential CH_4 decrease measurements ranged from 1-167 nmol L^{-1} d^{-1} [de Angelis and Scranton, 1993]. Along the Sinnamary River of French Guiana, MOX rates were 8-473 μmol L^{-1} d^{-1} [Guerin and Abril, 2007]. MOX rates calculated from $^{14}CH_4$ at Randers Fjord in Denmark were 15 nmol L^{-1} d^{-1} [Abril and Iverson, 2002]. CH_4 flux measurements, along with CH_4 concentration and isotopic ration ($\delta^{13}CH_4$) dissolved in water and bubbles in the sediment, of tropical large rivers in the Amazon Basin showed that MOX accounted for 57-82% of CH_4 loss (conversely indicating 18-43% loss as evasion) [Sawakuchi et al., 2015].

Complete stream methane budgets have only been calculated for a few rivers. For the Sinnamary River (French Guiana) CH_4 evasion accounted for about 50 % of total CH_4 loss, MOX accounted for 40% of CH_4 loss, with the remaining loss (10%) as downstream advection [Guerin and Abril, 2007]. A mass-balance analysis based on benthic and floating chamber measurements along a 45-m long (4.4 m wide) experimental section of Sitka

Stream (Czech Republic) indicated a benthic production rate accounted for 45% of total stream CH_4, with surface-water inflow and in-stream CH_4 production rate accounting for the remaining 55% of stream CH_4; evasion accounted for 40% of the loss of this stream CH_4, with the remainder being transported downstream (no in-stream net MOX was observed) [Rulik et al., 2013]. MOX in the Columbia River (Oregon and Washington) accounted for 25% of CH4 loss, but was insignificant in the more turbulent Wenatchee River [Lilley et al., 1996].

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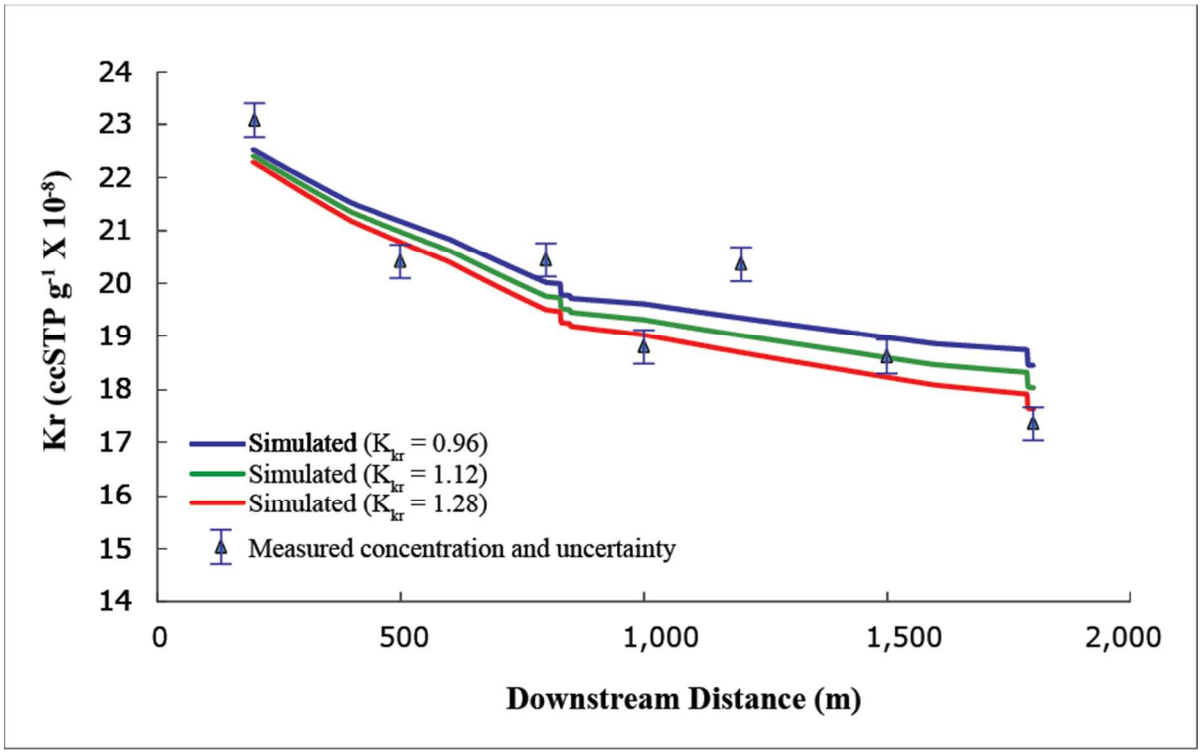
Sawakuchi, H. O.; Bastviken, D.; Sawakuchi, A. O.; Ward, N. D.; Borges, C. D.; Tsai, S. M.; Richey, J. E.; Ballester, M. V. R.; Krusche, A.V. Oxidative mitigation of aquatic methane emissions in large Amazonian rivers. *Global Change Biology* **2015**; doi:10.1111/gcb.13169.

Supplementary Materials Figure A. Observed and simulated stream water krypton (Kr) concentrations in ccSTP g^{-1} (cubic centimeters of gas at standard temperature and pressure per gram of stream water) using a gas transfer velocity of 1.12 ± 0.2 m d^{-1}. Atmospheric Kr was defined in the model as 8.7 x 10^{-8} ccSTP g^{-1}, which is negligible compared to stream injection concentrations and did not have a material impact on atmospheric gas transfer.



Supplementary Materials Table 1. Model parameter values and uncertainty for simulation of CH_4 loss from West Bear Creek

Model parameter	Symbol	Value	Units	Coefficient of Variation (Uncertainty)
[a]Stream discharge at 200 m	Q	480 ± 20	L s^{-1}	4%
[b]Combined tributary and groundwater inflow	$q_{trib} + q_l$	67 ± 10	L/s	15%
[c]Net injected stream CH_4 concentration (injected – naturally occurring) at 0 m	C_s	3120 ± 125	nmol L^{-1}	4%
[d]Stream width	w	6.7 ± 0.76	m	11%
[e]Stream depth	b	0.37 ± 0.05	m	28%
[f]K_{CH4} atmospheric gas transfer rate	K_{CH4}	1.17 ± 0.17	m d^{-1}	15%

[a]Based on a flowmeter measurement at 0 m and Br^- dilution at 200 m; [b]Combined total gain from 0 to 1800 m (28 L s^{-1} of tributary inflow plus 39 L s^{-1} of groundwater inflow; RSD determined from LINEST Excel function goodness of fit to linear trend; [c]Estimated based on measurements at 200 m; [d]Value and uncertainty based on mean and standard deviation of width at 8 locations; [e]Value and uncertainty based on mean and standard deviation of depth at 8 locations; [f]Determined by calibrating to injected krypton; Uncertainty based on Monte Carlo simulations (n=1000)