



Very large fluxes of methane measured above Bolivian seasonal wetlands

James L. France^{a,b,c,1}, Mark F. Lunt^d, Marcos Andrade^{e,f}, Isabel Moreno^e, Anita L. Ganesan^g, Thomas Lachlan-Cope^a, Rebecca E. Fisher^b, David Lowry^b, Robert J. Parker^h, Euan G. Nisbet^b, and Anna E. Jones^a

Edited by Simon Levin, Princeton University, Princeton, NJ; received April 29, 2022; accepted May 13, 2022

Methane (CH₄) mole fractions from the large semiseasonal Llanos de Moxos wetlands (~70,000 km²) in northern Bolivia were measured by aircraft flights and ground sampling during early March 2019 (late wet season). Daily fluxes of CH₄ determined from the measurements using box models and inverse modeling were between 168 (± 50) and 456 (± 145) mg CH₄·m⁻²·d⁻¹ for the areas overflowed, very high compared with those of previous Amazon basin studies. If the seasonality of the CH₄ emissions is comparable to other parts of the Amazon Basin, the region could contribute as much as 8% of annual Amazonian CH₄ emissions.

methane | climate | wetlands | greenhouse gases

Here, we present results from an aircraft and ground-based campaign to measure methane (CH₄) emissions from the seasonal Llanos de Moxos and Rio Mamoré floodplain wetlands of northeastern Bolivia. This was a wide-area-scale airborne measurement of CH₄ emissions from these major wetlands in the Bolivian component of the Amazon Basin. Previous studies focusing on the Brazilian Amazon Basin found large CH₄ fluxes, averaging 27 mg CH₄·m⁻²·d⁻¹ (maximum ~80 CH₄·m⁻²·d⁻¹) (1), while work using multiyear aerial measurements found an average of 17.4 ± 3.9 mg CH₄·m⁻²·d⁻¹ from the Amazonian region as a whole (2). Although the importance of the Bolivian “*sabana inundable*” (flood-prone savannah) as a major CH₄ source has been inferred from large underestimates of CH₄ emissions by land-surface models compared with satellite data (3), there have been no previous aircraft studies in this part of Amazonia to test for large-scale emissions to the atmosphere.

The Llanos de Moxos are extensive (~70,000 km²) seasonal wetlands in northeastern Bolivia, flanking the Rio Mamoré at a latitude of about 12 to 15°S with an outertropical wet season from December through March. We measured atmospheric CH₄ mole fractions during aircraft sorties on 2 days in March 2019, aiming to determine bulk wetland-scale CH₄ fluxes. Early March is in the region’s late wet season, with temperatures still warm and water levels high from the cumulative rainfall. Air samples were captured during flights and in a parallel campaign at ground level adjacent to the wetlands. These air samples were analyzed for δ¹³C_{CH₄} and their isotopic source signatures used to indicate the pathway of CH₄ formation and thus whether the source is likely to be from wetland (4).

Globally averaged atmospheric CH₄ mole fractions have risen persistently since 2007, with further acceleration from 2014 and record growth in 2020 (5). Concurrently, atmospheric δ¹³C_{CH₄} has become isotopically lighter since 2007 in all latitude bands, most plausibly explained by increasing emissions from biogenic sources such as wetlands and cattle (6). Large-scale tropical wetlands are major sources of CH₄ emissions, both in South America (1, 2) and Africa (7). Saunio et al. (8) estimated that wetlands account for ~30% of the total CH₄ flux to the atmosphere, but those wetlands with the greatest emissions (in the tropics) are the least well-characterized, and climate change feedbacks, especially in southern Amazonia, are poorly understood (9). Methane emissions from tropical wetlands respond to increasing temperature and precipitation, both key factors in interannual variation. The warming may be feeding warming, driving sustained growth in the global CH₄ burden (5).

Results

Two flights were conducted with both continuous measurement and bag sampling on board. Flight A was in the late afternoon (8 March 2019), with a planetary boundary layer (PBL) thickness of at least 1,400 m; Flight B was a morning flight (9 March 2019) with an average PBL thickness of 620 m. The PBL depths are estimated from

Author affiliations: ^aBritish Antarctic Survey, Natural Environment Research Council, Cambridge, CB3 0ET, United Kingdom; ^bDepartment of Earth Sciences, Royal Holloway University of London, Egham, TW20 0EX, United Kingdom; ^cEnvironmental Defense Fund, London, EC3M 1DT, United Kingdom; ^dSchool of GeoSciences, University of Edinburgh, Edinburgh, EH9 3FF, United Kingdom; ^eLaboratory for Atmospheric Physics, Institute for Physics Research, Universidad Mayor de San Andres, 8635 La Paz, Bolivia; ^fDepartment of Atmospheric and Oceanic Sciences, University of Maryland, College Park, MD 20742; ^gSchool of Geographical Sciences, University of Bristol, Bristol, BS8 1SS, United Kingdom; and ^hNational Centre for Earth Observation, University of Leicester, Leicester, LE4 5SP, United Kingdom

Author contributions: J.L.F., T.L.-C., E.G.N., and A.E.J. designed research; J.L.F., M.A., and I.M. performed research; R.J.P. contributed analytic tools; J.L.F., M.F.L., A.L.G., R.E.F., and D.L. analyzed data; M.A. and E.G.N. contributed to the preliminary fieldwork campaign; M.A. contributed to planning; E.G.N. contributed to conceptualization; A.E.J. was the project principal investigator and contributed fieldwork organization; and J.L.F., M.F.L., and A.E.J. wrote the paper.

The authors declare no competing interest.

Copyright © 2022 the Author(s). Published by PNAS. This open access article is distributed under Creative Commons Attribution License 4.0 (CC BY).

¹To whom correspondence may be addressed. Email: james.france@rhu.ac.uk.

This article contains supporting information online at <http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.2206345119/-/DCSupplemental>.

Published August 1, 2022.

vertical profile measurements of air temperature during flights. Flight tracks, CH₄ atmospheric mixing ratios, and locations are shown in Fig. 1 *A* and *B*.

Maximum CH₄ enhancement above background mixing ratios was 200 ppb on Flight A and 550 ppb Flight B. Using Keeling plot analysis on sampled air, the overall δ¹³C_{CH₄} source signature of CH₄ enhancements during Flight A was determined to be $-56.1 \pm 2.9\text{‰}$ and on Flight B was $-58.6 \pm 4.1\text{‰}$ (95% confidence), consistent with the results of the parallel ground-based downwind sampling campaign which gave a δ¹³C_{CH₄} source signature of $-55.5 \pm 4.0\text{‰}$.

Methane fluxes from the aerial surveys of the Llanos de Moxos were calculated using three methods.

- 1) Using a simple boundary layer box-model mass balance approach (10) assuming that the wetland surface emits at a constant rate per square kilometer, giving a flux of $168 (\pm 50) \text{ mg CH}_4 \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ and $456 (\pm 145) \text{ mg CH}_4 \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ for the areas overflowed by Flights A and B, respectively.
- 2) Using a Bayesian inversion of measurement data and a nested version of the GEOS-Chem model, an average flux of $384 (\pm 48) \text{ mg CH}_4 \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ was determined along the path of the main Rio Mamoré river channel corresponding to the flight path of Flight B (Fig. 1).
- 3) Using a hierarchical Bayesian inversion with the high-resolution Numerical Atmospheric dispersion Modeling Environment (NAME) regional Lagrangian transport model (11),

an average flux of $156 (\pm 48) \text{ mg CH}_4 \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ over the same Flight B main river channel.

To establish whether the measured CH₄ enhancements are a regular regional feature during the wet season, we compared GOSAT (Greenhouse gases Observing SATellite) satellite retrievals over the Llanos de Moxos with mole fractions simulated by the GEOS-Chem model over the period from January to March from 2010 to 2020 (Fig. 1 *E* and *SI Appendix, Supplementary Materials and Methods*). To scale the model column enhancements to the GOSAT-observed data, average required CH₄ flux from the Llanos de Moxos wetland would be $\sim 210 (\pm 55) \text{ mg CH}_4 \cdot \text{m}^{-2} \cdot \text{d}^{-1}$. For 2019 only, the required flux was $\sim 355 (\pm 115) \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, consistent with the fluxes determined from Flight B (although with high uncertainty).

Discussion

CH₄ enhancements observed in flight were very large (>500 ppb for Flight B) and prolonged through the measurement period. The variability in the measured atmospheric mixing ratios of CH₄ is a function of the depth of the PBL and of the emission flux of methane from the wetland below. These measurements are well-simulated by the models, with close correspondence of box-model and three-dimensional modeling results. As the flights were predominantly over the main river channel, there are larger uncertainties for fluxes over the wider Llanos de Moxos. These

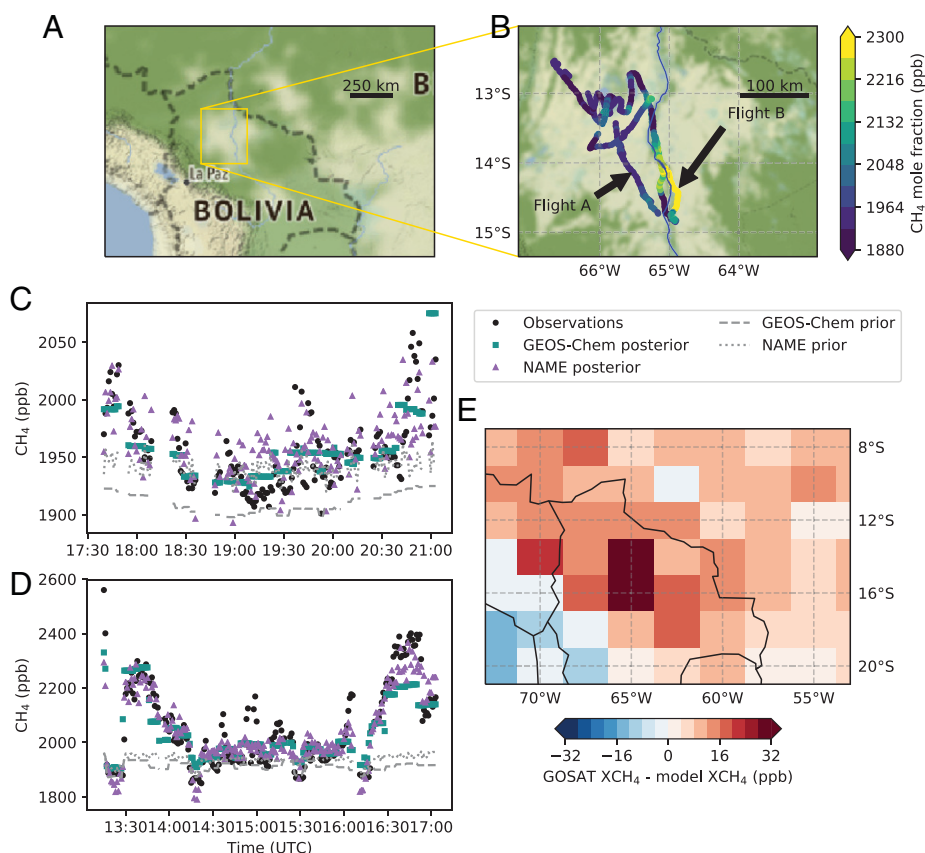


Fig. 1. (A) Location of campaign, flights starting and ending at Trinidad, Bolivia. (B) Flight tracks and observed atmospheric methane mixing ratios. (C) Comparison between observations of methane mixing ratios with model prior and posterior derived methane mixing ratios for Flight A, 8 March 2019. Gaps in the data are due to in-flight calibrations and profile climbs outside of the boundary layer. (D) As C, for Flight B, 9 March 2019. Flight A is primarily over the seasonal wetland to the northwest of Trinidad, and Flight B follows the main river channel (Rio Mamoré) to $\sim 13^{\circ}\text{S}$ and then resamples regions from Flight A. Note the different y axis scales in C and D. Wind direction was primarily from the northwest for Flight A and from the north for Flight B. (E) Mean excess methane measured from GOSAT satellite data compared with simulated methane from the GEOS-Chem atmospheric chemistry transport model over the period from January to March from 2010 to 2020.

CH₄ fluxes are greater than found in other Amazonian region-scale measurements, with the largest comparable previously determined flux of ~80 mg CH₄·m⁻²·d⁻¹ for eastern Amazonia in mid-February (1). We note, however, that early March would be around the expected late rainy seasonal peak of emission. The results of our comparison of modeled wetland emissions with satellite enhancements over the Llanos de Moxos are similar to those of Parker et al. (3). They found wetland modeling underestimated fluxes from a broader region including northern Bolivia, particularly during peak inundation and flux (January to March).

The isotopic results are closely comparable to results from parallel southern outer tropical wetlands in Zambia (12) and to previously reported overflight measurements from Brazilian Amazonian wetlands that gave a δ¹³C_{CH₄} source signature of -58.8‰ (13). Such sources, if increasing, would help drive the global methane burden to more negative δ¹³C_{CH₄} emissions. Our measurement observations hint at two modes of emission from the region: a seasonally inundated wetland flux from the plains (Flight A, Fig. 1C) and a more intense flux centered on tree-lined permanently inundated area along the major river channel (Flight B, Fig. 1D). This interpretation is supportive of findings of large CH₄ fluxes across a number of rivers in the central Amazon Basin where they calculated that tree-mediated fluxes were equal to, if not greater than, all other combined ground-level fluxes (14).

In the 11-year GOSAT satellite record, consistent large (albeit coarse-resolution) enhancements over the Llanos de Moxos recorded add confidence that the observed CH₄ enhancement is not a one-off result (Fig. 1E). Assuming that the fluxes from the Llanos de Moxos region have a methane flux seasonality comparable to the Rio Branco site (67.9°W, 9.3°S) (2), scaling up the observations from Flight A (as this flight is the most representative of the region as a whole) to annualized emissions yields rates of ~3.6 Tg CH₄/y for the whole Llanos de Moxos wetland region. This flux would be equivalent to ~8% of the estimated total Amazonian CH₄

emission (2), showing that the northern Bolivian wetlands are likely to be a significant contributor to global emissions. This is a preliminary result from a brief campaign of opportunity and further study is required to assess the seasonality of emissions and the possibility of disproportionately large emissions from riverine forests and wetlands.

Materials and Methods

Measurements were made from the BAS twin-otter aircraft using a Los Gatos Research uGGA instrument calibrated in-flight. Spot samples were collected in Tedlar bags in-flight over wetlands and at ground level downwind from wetlands and returned to the United Kingdom for analysis. Fluxes of CH₄ were determined from the measurements three ways, using simple box-model mass-balance methods (10) and inverse modeling using the GEOS-Chem (7) and NAME models (15). GOSAT satellite column XCH₄ data from 2010 to 2020 were compared with XCH₄ enhancements simulated by GEOS-Chem over the wetlands to determine whether CH₄ enhancements seen during this campaign were consistent with previous wet seasons. For further details see *SI Appendix, Supplementary Materials and Methods*.

Data Availability. All original data within this manuscript have been deposited in the publicly accessible Mendeley Data repository (doi: [10.17632/pm4sw7hy3b.1](https://doi.org/10.17632/pm4sw7hy3b.1)).

ACKNOWLEDGMENTS. This work was part of Methane Observations and Yearly Assessments, the United Kingdom's global methane budget study. The Natural Environment Research Council supported this work through grants to E.G.N. (NE/N016238/1 and NE/S00159X/1), A.E.J. (NE/N015584/1 and NE/N015916/1), R.E.F. (NE/V000780/1), and R.J.P. (NE/N018079/1), and through Fellowship NE/L010992/1 to A.L.G. This research used the Advanced Leicester Information and Computational Environment at the University of Leicester for the GOSAT retrievals. We thank the Japanese Aerospace Exploration Agency, National Institute for Environmental Studies, and the Ministry of Environment for the GOSAT data. We acknowledge very helpful discussions with John Miller, Luciana Gatti, and Emanuel Gloor. We especially thank Ambassador Ross Denny and the UK Foreign, Commonwealth & Development Office. We are very grateful to the pilots, Mark Beasley and Andy van Kints, and Dan Beeden for logistics.

1. J. B. Miller et al., Airborne measurements indicate large methane emissions from the eastern Amazon basin. *Geophys. Res. Lett.* **34**, L10809 (2007).
2. L. S. Basso et al., Amazon methane budget derived from multi-year airborne observations highlights regional variations in emissions. *Commun. Earth Environ.* **2**, 246 (2021).
3. R. J. Parker et al., Evaluating year-to-year anomalies in tropical wetland methane emissions using satellite CH₄ observations. *Remote Sens. Environ.* **211**, 261–275 (2018).
4. A. L. Ganesan et al., Spatially resolved isotopic source signatures of wetland methane emissions. *Geophys. Res. Lett.* **45**, 3737–3745 (2018).
5. X. Lan, E. G. Nisbet, E. J. Dlugokencky, S. E. Michel, What do we know about the global methane budget? Results from four decades of atmospheric CH₄ observations and the way forward. *Philos. Trans. A Math. Phys. Eng. Sci.* **379**, 20200440 (2021).
6. E. G. Nisbet et al., Very strong atmospheric methane growth in the 4 years 2014–2017: Implications for the Paris Agreement. *Global Biogeochem. Cycles* **33**, 318–342 (2019).
7. M. F. Lunt et al., An increase in methane emissions from tropical Africa between 2010 and 2016 inferred from satellite data. *Atmos. Chem. Phys.* **19**, 14721–14740 (2019).
8. M. Saunio et al., The global methane budget 2000–2017. *Earth Syst. Sci. Data* **12**, 1561–1623 (2020).
9. J. C. Espinoza, J. Ronchail, J. A. Marengo, H. Segura, Contrasting north-south changes in Amazon wet-day and dry-day frequency and related atmospheric features (1981–2017). *Clim. Dyn.* **52**, 5413–5430 (2019).
10. R. V. Hiller et al., Aircraft-based CH₄ flux estimates for validation of emissions from an agriculturally dominated area in Switzerland. *J. Geophys. Res. Atmosph.* **119**, 4874–4887 (2014).
11. A. Jones, D. Thomson, M. Hort, B. Devenish, "The U.K. Met Office's next-generation atmospheric dispersion model, NAME III" in *Air Pollution Modeling and Its Application XVII*, C. Borrego, A.-L. Norman, Eds. (Springer, Boston, 2007), pp. 580–589.
12. E. G. Nisbet et al.; MOYA/ZWAMPS Team, Isotopic signatures of methane emissions from tropical fires, agriculture and wetlands: The MOYA and ZWAMPS flights. *Philos. Trans A Math. Phys. Eng. Sci.* **380**, 20210112 (2022).
13. V. Beck et al., Methane airborne measurements and comparison to global models during BARCA. *J. Geophys. Res. Atmosph.* **117**, D15310 (2012).
14. S. R. Pangala et al., Large emissions from floodplain trees close the Amazon methane budget. *Nature* **552**, 230–234 (2017).
15. A. L. Ganesan et al., Characterization of uncertainties in atmospheric trace gas inversions using hierarchical Bayesian methods. *Atmos. Chem. Phys.* **14**, 3855–3864 (2014).