



Global Methane Budget 2013

Three Decades of Global Methane
Sources and Sinks : from global to
regional

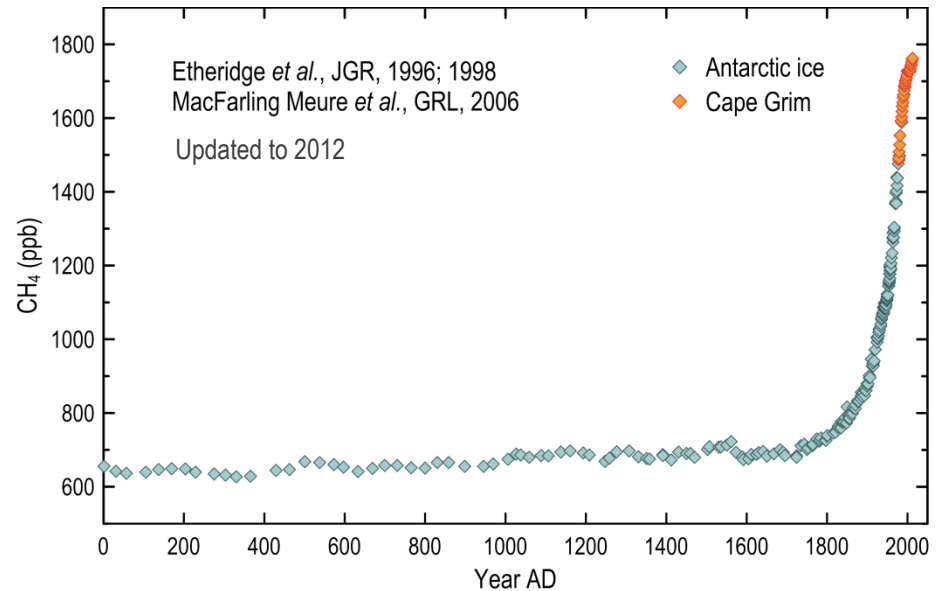


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SINO-FRENCH INSTITUTE FOR EARTH SYSTEM SCIENCE
PEKING UNIVERSITY CEA VERSAILLES ST QUENTIN UNIVERSITY



The Context

- After carbon dioxide (CO₂), methane (CH₄) is the second most important well-mixed greenhouse gas contributing to human-induced climate change.
- In a time horizon of 100 years, CH₄ has a Global Warming Potential 28 times larger than CO₂.
- It is responsible for 20% of the global warming produced by all well-mixed greenhouse gases.
- The concentration of CH₄ in the atmosphere is above 150% from the levels prior to the Industrial Era (cf. 1750).
- The atmospheric life time of CH₄ is approximate 10±2 years.



- Methane also contributes to ozone production in the troposphere, which is a pollutant with negative impacts on human health and ecosystems.
- Increasing emissions of methane are transformed into water in the stratosphere by chemical reactions.

Atmospheric
Observations

Emission
Inventories
(B-U)

Biogeochemistry
Models
(B-U)

Inverse Models
(T-D)

OH Sink

The Tools and Data

Ground-based
data from
observation
networks (AGAGE,
CSIRO, NOAA,
UCI).

Airborne
observations.

Satellite data.



Agriculture and
waste related
emissions, fossil
fuel emissions
(EDGAR, EPA,
IIASA).

Fire emissions
(GFED, GICC,
FINN, RETRO).



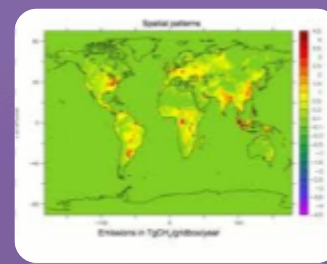
Ensemble of
different wetland
models, (LPJ-
WHyMe, LPJ-wsl,
ORCHIDEE).

Data and models
to calculate
annual flooded
area.



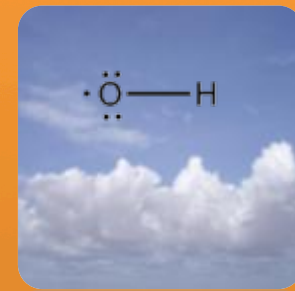
Suite of different
atmospheric
inversion models
(TM5-4DVAR,
LMDZ-MIOP,
CarbonTracker-
CH₄, GEOS-Chem,
LMDZt-SACS,
MATCH, TM2,
GISS).

TransCom
intercomparison.

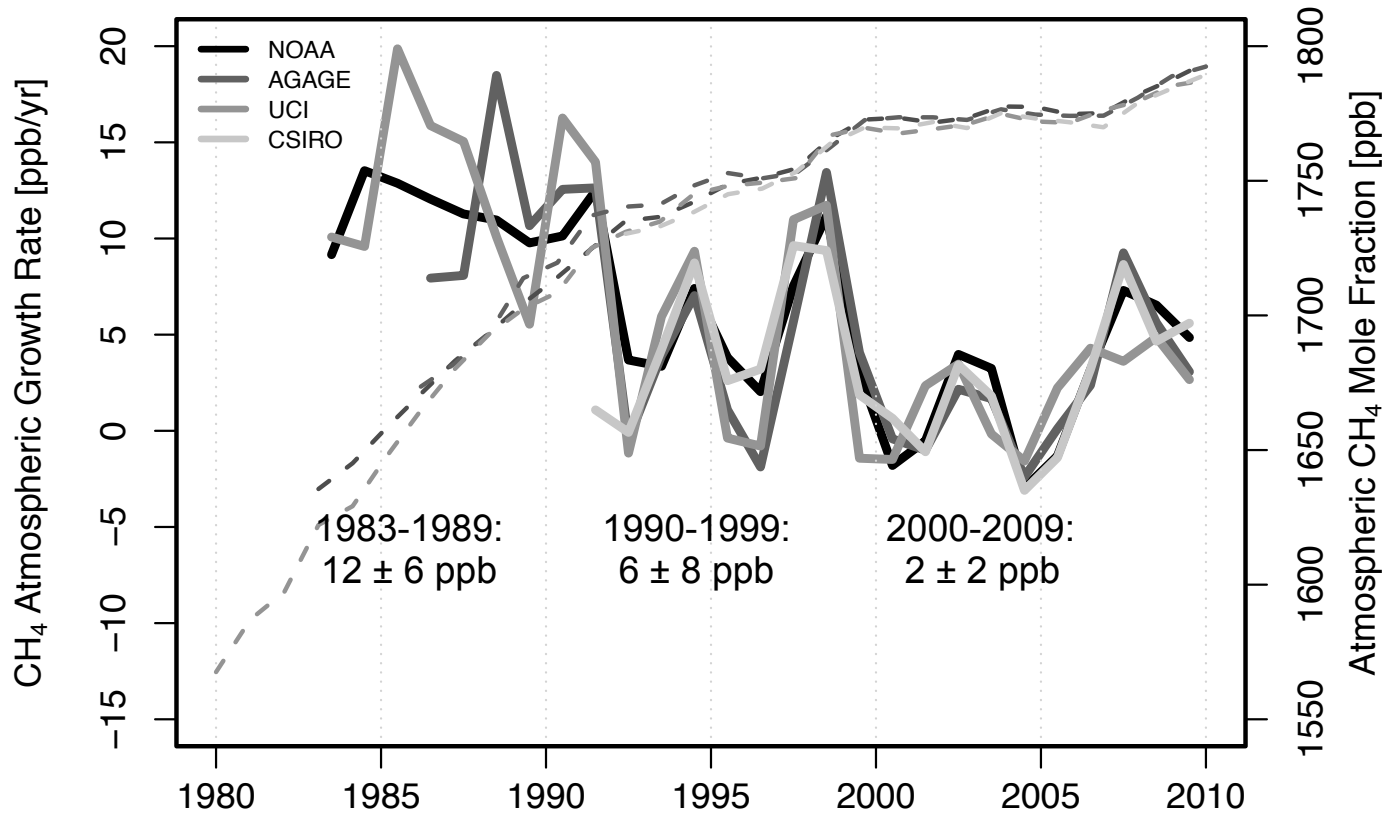


Long-term trends
and decadal
variability of the
OH sink.

ACCMIP CTMs
intercomparison.



CH₄ Atmospheric Growth Rate, 1983-2009



- Slowdown of atmospheric growth rate before 2005
- Resumed increase after 2006

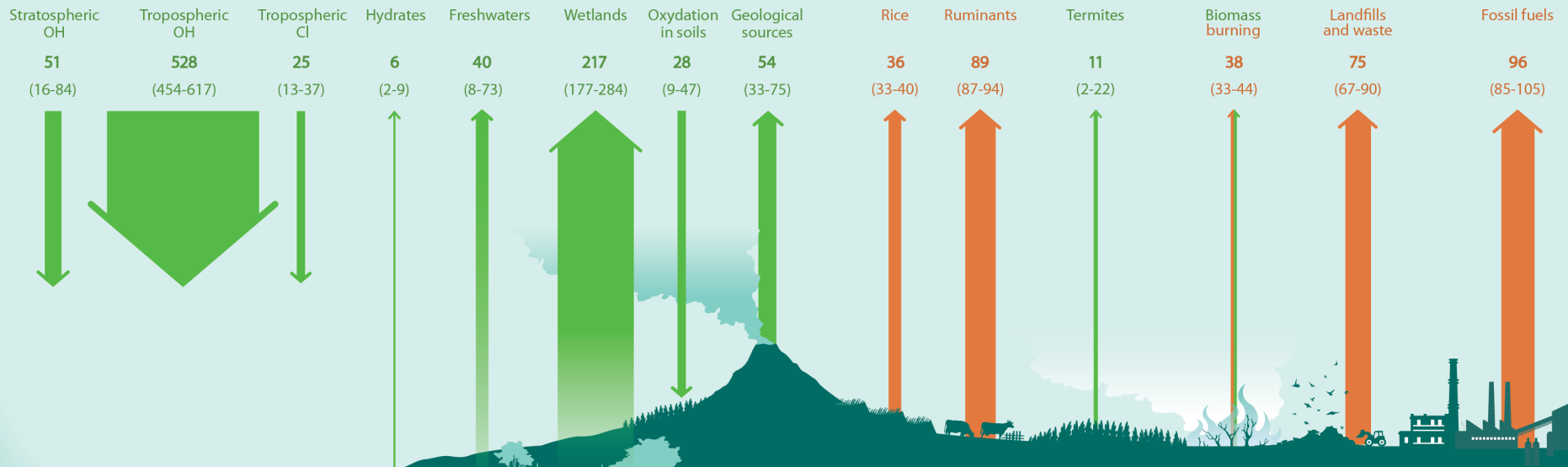
Decadal Budgets

From B-U models & data :

METHANE BUDGET : 2000-09

ATMOSPHERE

Methane reservoir in atmosphere prior to the Industrial Era (in TgCH₄)







EXCHANGES BY SOURCE

in teragrams CH₄ / year



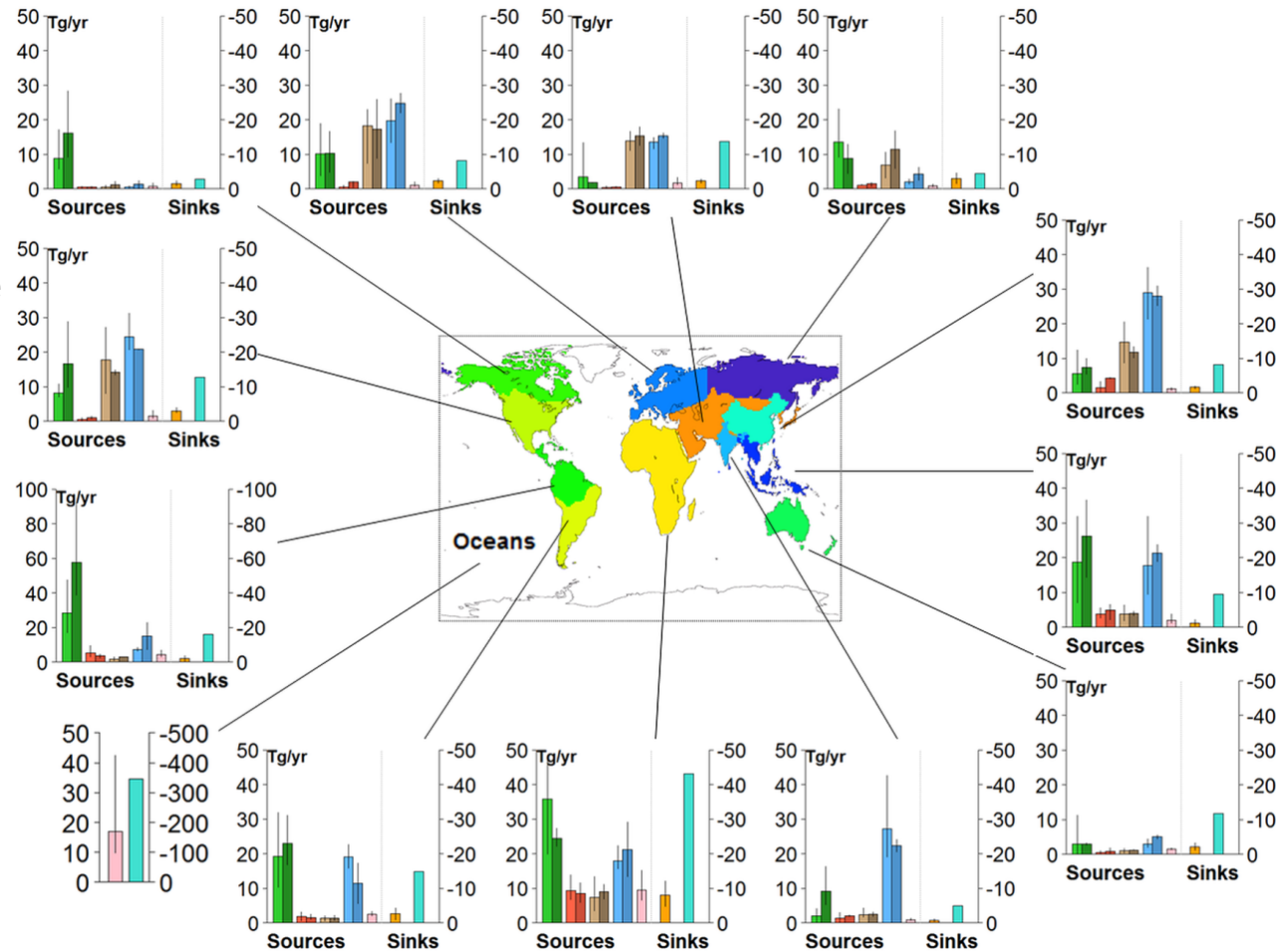
Tg CH ₄ yr ⁻¹	1980–1989		1990–1999		2000–2009	
	Top-Down	Bottom-Up	Top-Down	Bottom-Up	Top-Down	Bottom-Up
Sources						
Natural Sources	203 [150–267]	355 [244–466]	182 [167–197]	336 [230–465]	218 [179–273]	347 [238–484]
Natural Wetlands	167 [115–231]	225 [183–266]	150 [144–160]	206 [169–265]	175 [142–208]	277 [177–284]
Other Sources	36 [35–36]	130 [61–200]	32 [23–37]	130 [61–200]	43 [7–65]	130 [61–200]
Anthropogen. Sources	348 [305–383]	308 [292–323]	372 [290–453]	313 [281–347]	335 [273–409]	331 [304–368]
Agriculture & Waste	208 [187–220]	185 [172–197]	239 [180–301]	187 [177–196]	209 [180–241]	200 [187–224]
Rice		43 [41–47]				36 [33–40]
Ruminants		85 [81–90]				89 [87–94]
Landfills & Waste		55 [50–60]				75 [67–90]
Biomass Burning	46 [43–55]	34 [31–37]	38 [26–45]	42 [30–45]	30 [24–45]	35 [32–39]
Fossil Fuels	94 [75–108]	89 [89–89]	95 [84–107]	84 [66–96]	96 [77–123]	96 [85–105]
Sinks						
Total Chemical Loss	490 [450–533]	539 [411–671]	525 [491–554]	571 [521–621]	518 [510–538]	604 [483–738]
Global						
Sum of Sources	551 [500–592]	663 [536–789]	554 [529–596]	649 [511–812]	548 [526–569]	678 [542–852]
Sum of Sinks	511 [460–559]	539 [420–718]	542 [518–579]	596 [530–668]	540 [514–560]	632 [592–785]
Imbalance (Sources-Sinks)	30 [16–40]		12 [7–17]		8 [-4–19]	
Atmospheric Growth Rate	34		17		6	

Inland waters
Geological leaks

-  Larger global total emissions from Bottom-Up (inventories, models) than Top-Down (atmospheric inversions) because of larger natural emissions
-  Large uncertainties remain for wetland emissions (min-max range)
-  ~50 Tg global imbalance in B-U approaches (T-D constrained by atmosphere)
-  Increasing OH loss between decades in B-U (not clear in T-D)

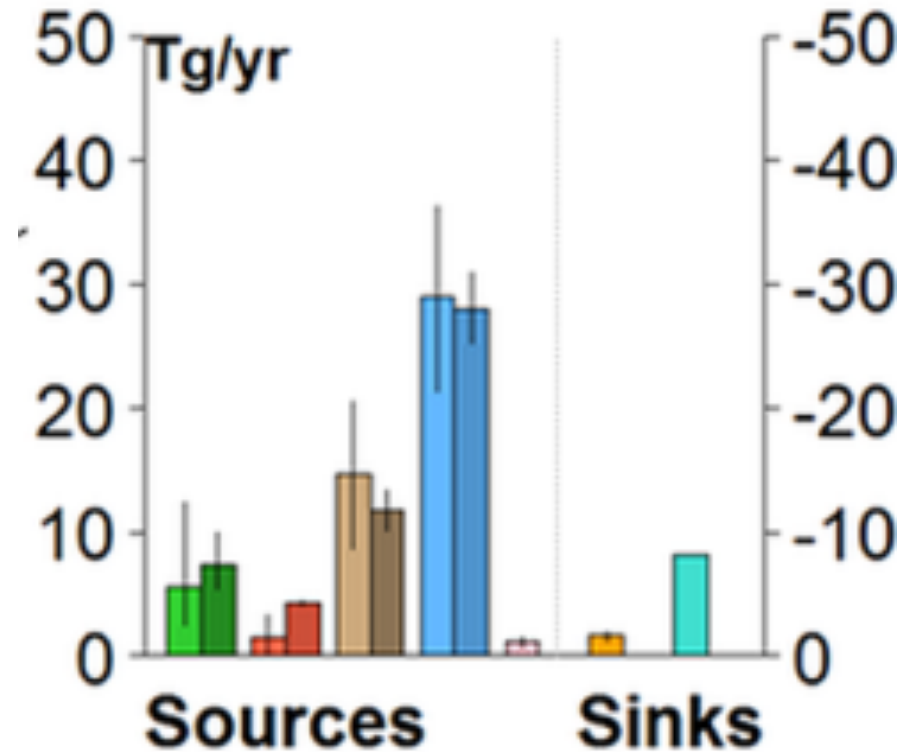
Regional Methane Budget

- Dominance of wetland emissions in the tropics and boreal regions
- Dominance of agriculture & waste in India and China
- Balance between agriculture & waste and fossil fuels at mid-latitudes
- Uncertain magnitude of wetland emissions in tropical South America between T-D and B-U



Regional Methane Budget : China

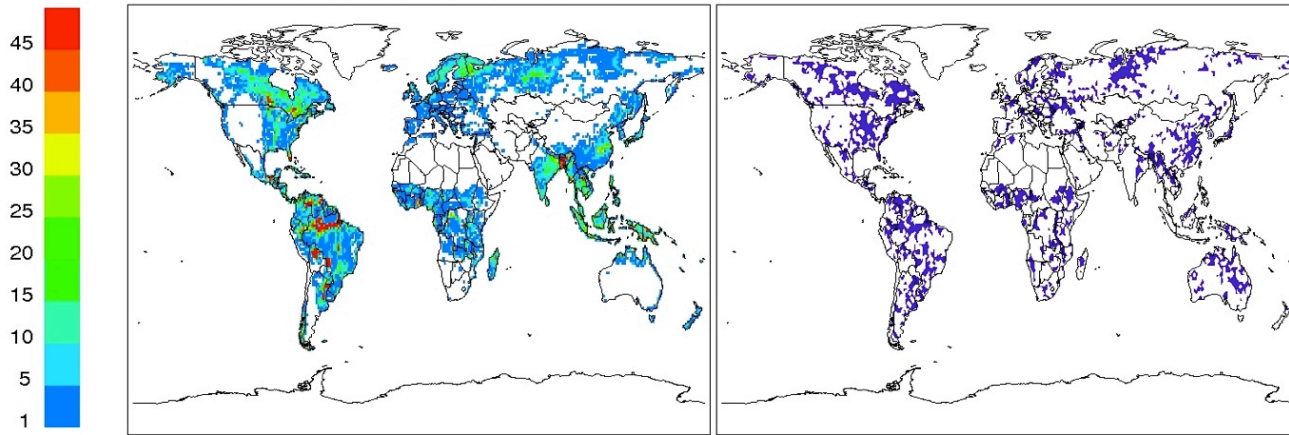
- Decadal China emissions for the 2000s (% of total source for each category) :
- Nat. wetlands : ~5 % [2-7%]
- BBG & biofuels : ~7 % [0-10%]
- Fossil fuels : ~15 % [8-20%]
- Agri. & waste : ~15 % [10-18%]



Spatial Distribution of Fluxes

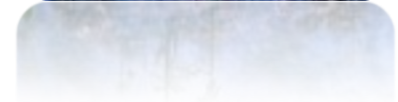
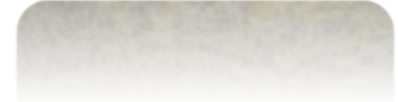
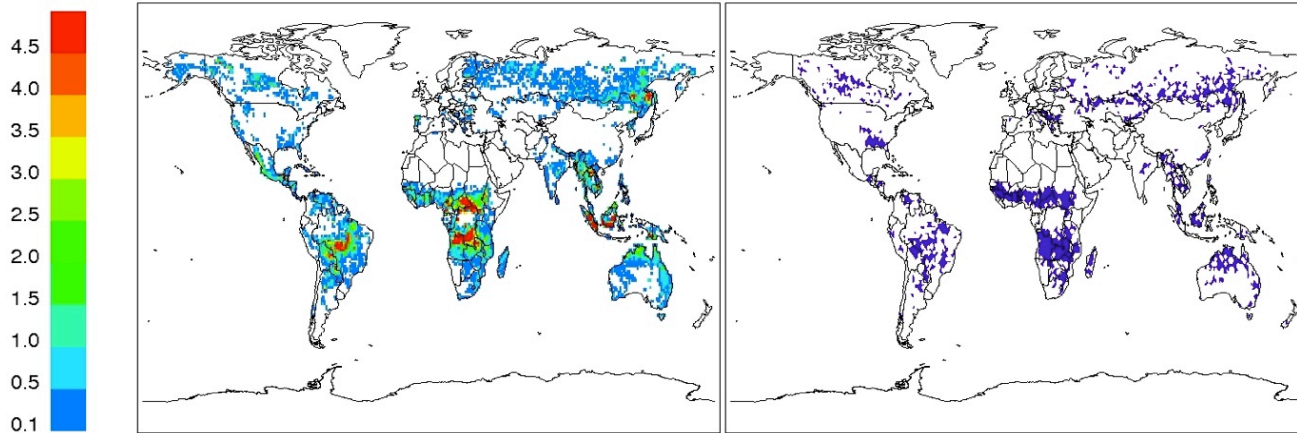
mg.m⁻².d⁻¹

Wetland emission flux 1990-2006



mg.m⁻².d⁻¹

Fire emission flux 1997-2000



Kirschke et al. 2013, Supplementary Information, Nature Geoscience

Data sources: Wetland emissions (ORCHIDEE, LPJ-WHyMe, LPJ-wsl), Biomass burning emissions: GFED2, GFED3, RETRO, GICC).

Scenarios of Temporal Change

(Results of the) Scenario Analysis for IAV



Stabilisation period (1999-2006):

→ Decreasing to stable fossil fuel emissions and stable to increasing microbial emissions are more likely

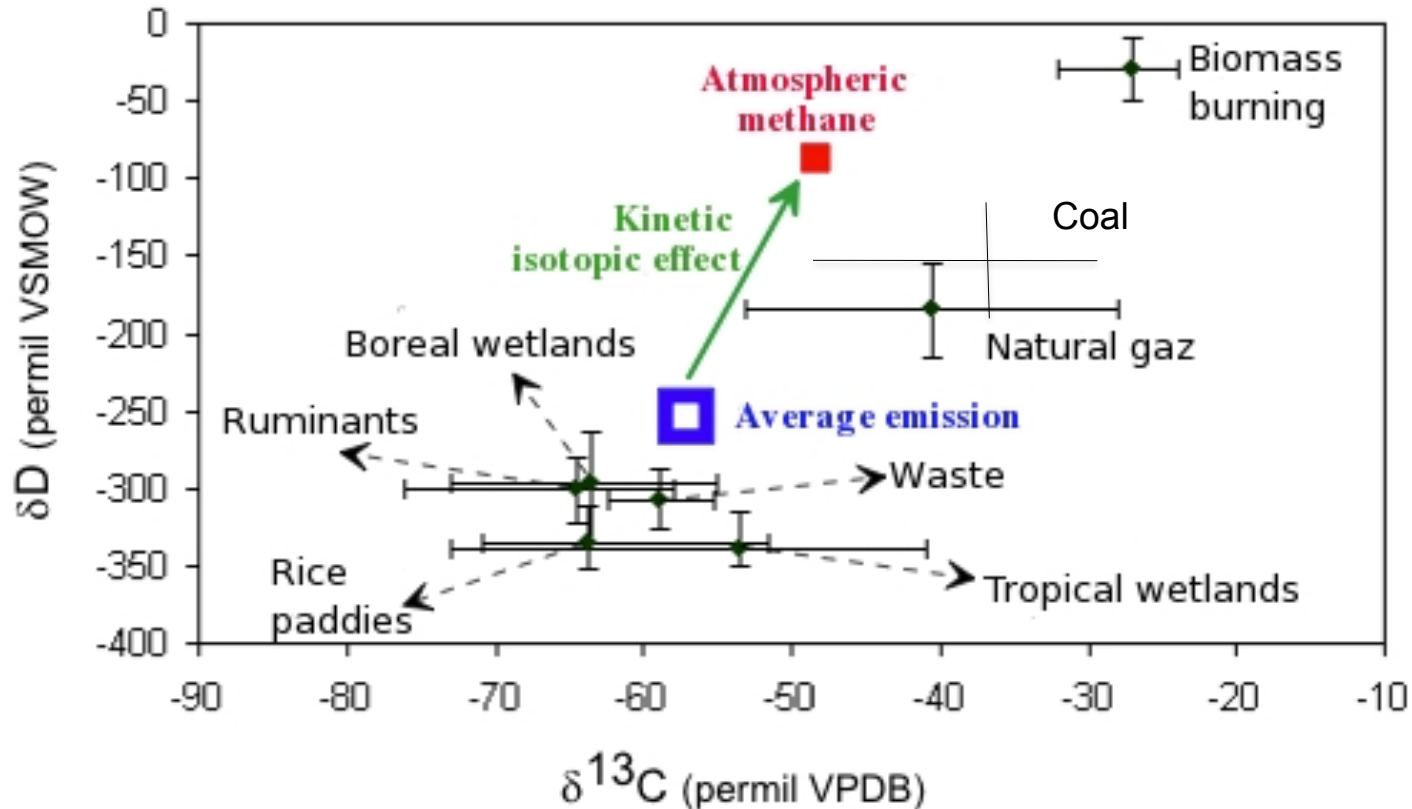
Resumed atmospheric increase (>2006) :

→ Mix of fossil fuel and wetland emissions increase, but relative magnitude remains uncertain

→ What about isotopes ?

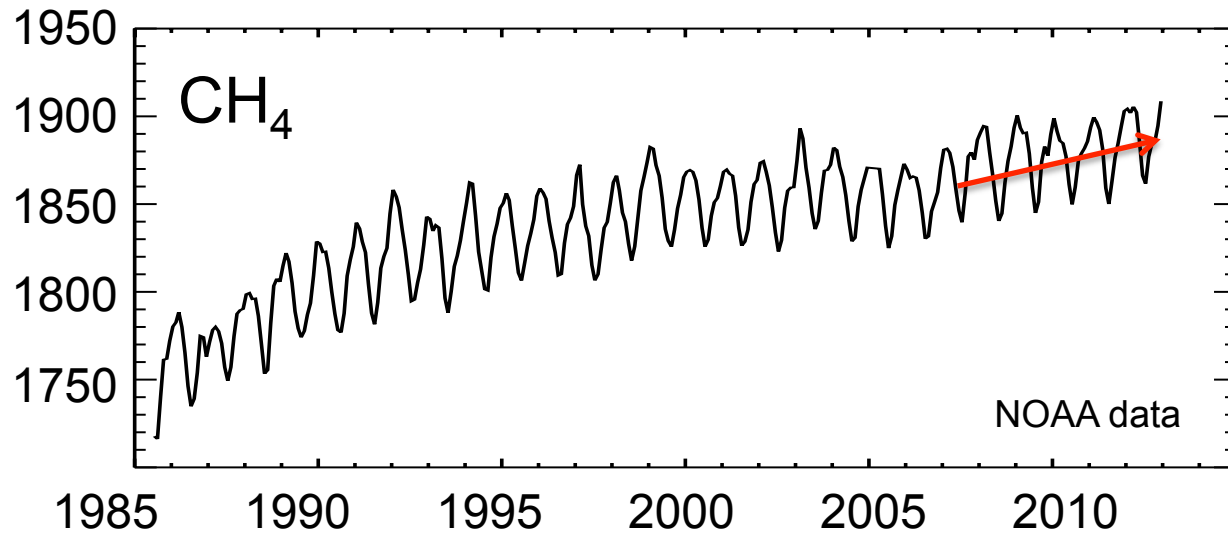
Can isotopes help to partition emission types ?

What is the interest of δD and $\delta^{13}C$ in CH_4 ?

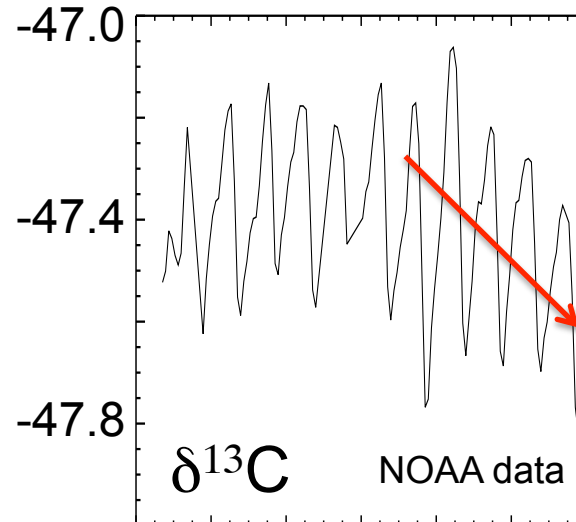


Double isotopic signature of various sources of methane determined by experimental studies. Adapted from Marik 1998.

Can isotopes help to partition emission types ?



Example :
ALERT station
(Canada)



First-order impact of a simple isotopic analysis



- 2007-2012 period (data analysis) :
 - Increasing CH_4 ($\sim 5,7$ ppb/yr) decreasing $^{13}\text{CH}_4 \sim -0,037$ ‰/yr
 - Atmosphere $\sim -47,4$ ‰, Coal (China) ~ -35 ‰, Wetlands ~ -60 ‰
 - Isotopic signature of total anthropogenic emission changes (EDGAR4.2 or EPA – 2000-2008) : from -45 ‰ to -52 ‰ depending on changes in coal emissions in China and gas in the US

---> Wetland increase (almost) required !

---> The increase in coal emissions has to be compensated by the stabilisation/decrease of another source with a less depleted signature in ^{13}C than the atmosphere (gas, BBG ?) !

--- > 1-Box model calculation, 2 equations : Wetland changes (70-90%) dominate anthropogenic changes (10-30%, increasing Coal, agriculture&waste, but decreasing BBG, assuming no change in OH)

Further improvements



- The large uncertainties in natural wetlands limit our ability to fully close the CH_4 budget ---> Improved parametrisations, WETCHIMP intercomparison ?
- Other natural emissions are also highly uncertain (inland waters, geological) ---> proxy tracers ?
- Little ability of the top-down atmospheric inversions to partition emissions among source types ---> Use of isotopes ?
- Large uncertainties in the OH mean values ---> proxy methods & isotopes ?
- Changes after 2006 still debated between ↗ wetlands and ↗ fossil fuels --> use of isotopes, refine IAV of emission inventories ?
- Uncertainty on transport modelling significant ---> Refine models

Acknowledgements



The work presented here has been possible thanks to the enormous observational and modeling efforts of the institutions and networks below

Atmospheric CH₄ datasets

- NOAA/ESRL (Dlugokencky et al., 2011)
- AGAGE (Rigby et al., 2008)
- CSIRO (Francey et al., 1999)
- UCI (Simpson et al., 2012)

Top-down atmospheric inversions

- TM5-4DVAR (Bergamaschi et al., 2009)
- LMDZ-MIOP (Bousquet et al., 2011)
- CarbonTracker-CH₄ (Bruhwiler et al., 2012)
- GEOS-Chem (Fraser et al., 2013)
- TM5-4DVAR (Beck et al., 2012)
- LMDZt-SACS (Pison et al., 2009; Bousquet et al., 2011)
- MATCH model (Chen & Prinn, 2006)
- TM2 model (Hein et al., 1997)
- GISS model (Fung et al. 1991)

Bottom-up studies data and modeling

- LPJ-wsl (Hodson et al, 2011)
- ORCHIDEE (Ringeval et al., 2011)
- LPJ-WhyMe (Spahni et al., 2011)
- GICC (Mieville et al., 2010)
- RETRO (Schultz et al., 2007)
- GFEDv2 (Van der Werf et al., 2004)
- GFEDv3 (Van der Werf et al., 2010)
- FINNv1 (Wiedinmyer et al., 2011)
- IIASA (Dentener et al., 2005)
- EPA, 2011
- EDGARv4.1 (EDGAR4.1, 2009)
- EDGARv4.2 (EDGAR4.2, 2011)
- Description of models contributing to the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP, Lamarque et al., 2013; Voulgarakis et al., 2013; Naik et al., 80 2013)
- TM5 full chemistry model (Williams et al., 2012; Huijnen et al., 2010)

Three decades of global methane sources and sinks

Stefanie Kirschke *et al.**

Methane is an important greenhouse gas, responsible for about 20% of the warming induced by long-lived greenhouse gases since pre-industrial times. By reacting with hydroxyl radicals, methane reduces the oxidizing capacity of the atmosphere and generates ozone in the troposphere. Although most sources and sinks of methane have been identified, their relative contributions to atmospheric methane levels are highly uncertain. As such, the factors responsible for the observed stabilization of atmospheric methane levels in the early 2000s, and the renewed rise after 2006, remain unclear. Here, we construct decadal budgets for methane sources and sinks between 1980 and 2010, using a combination of atmospheric measurements and results from chemical transport models, ecosystem models, climate chemistry models and inventories of anthropogenic emissions. The resultant budgets suggest that data-driven approaches and ecosystem models overestimate total natural emissions. We build three contrasting emission scenarios — which differ in fossil fuel and microbial emissions — to explain the decadal variability in atmospheric methane levels detected, here and in previous studies, since 1985. Although uncertainties in emission trends do not allow definitive conclusions to be drawn, we show that the observed stabilization of methane levels between 1999 and 2006 can potentially be explained by decreasing-to-stable fossil fuel emissions, combined with stable-to-increasing microbial emissions. We show that a rise in natural wetland emissions and fossil fuel emissions probably accounts for the renewed increase in global methane levels after 2006, although the relative contribution of these two sources remains uncertain.

Reconstructions of atmospheric methane (CH_4) concentrations between 1850 and the 1970s have been made using air trapped in polar ice cores and compacted snow. The data reveal an exponential increase in CH_4 levels in the atmosphere from 830 ppb to 1500 ppb in the late 1970s¹. Direct measurements of CH_4 in the atmosphere began in 1978², and reached global coverage after 1983. Today, CH_4 concentrations can be assessed using discrete air samples collected regularly at the surface, continuous measurements made at the surface^{3–4} or in the troposphere^{5–9}, and remotely sensed measurements of atmospheric CH_4 columns retrieved from the surface or from space^{10–12} (see Supplementary Section ST1). Surface-based observations from four networks (National Oceanic and Atmospheric Administration, NOAA¹³; Advanced Global Atmospheric Gases Experiment, AGAGE¹⁴; Commonwealth Scientific and Industrial Research Organization, CSIRO¹⁵; and University of California Irvine, UCI¹⁶) show consistent changes in the global growth rate of annual CH_4 concentrations since 1980 (Fig. 1 and Supplementary Section ST1). The agreement between these networks has improved with increasing coverage. The standard deviation for the global annual growth rate decreased from ± 3.3 ppb yr^{-1} in the 1980s to ± 1.3 ppb yr^{-1} in the 2000s. These data reveal a sustained increase in atmospheric CH_4 levels in the 1980s (by an average of 12 ± 6 ppb yr^{-1}), a slowdown in growth in the 1990s (6 ± 8 ppb yr^{-1}), and a general stabilisation from 1999 to 2006 to 1773 ± 3 ppb. Since 2007, CH_4 levels have been rising again¹⁴, and reached 1799 ± 2 ppb in 2010. This increase reflects a recent imbalance between CH_4 sources and sinks that is not yet fully understood¹⁴.

Previous reviews of the global CH_4 budget have focused on results from a few studies only^{17–19}. These studies covered different time windows and employed different assumptions, making it difficult to interpret the decadal changes presented. Only very few studies addressed multi-decadal changes in CH_4 levels^{20,21}. Here we construct a global CH_4 budget for the past three decades by combining bottom-up and top-down estimates of CH_4 sources and the chemical CH_4 sink (Box 1). We use chemical transport models — constrained by atmospheric CH_4 measurements — to estimate CH_4 fluxes using top-down atmospheric inversions. We compare these

fluxes with those simulated by ecosystem models of wetland and biomass burning emissions and by data-driven approaches for other natural sources (Methods and Supplementary Section II). We also gather recent data from fossil fuel CH_4 emission inventories based on energy use statistics, and from agricultural and waste inventories based on livestock and rice paddy statistics.

Sources and sinks

The global atmospheric CH_4 budget is determined by many terrestrial and aquatic surface sources, balanced primarily by one sink in the atmosphere. CH_4 emissions can be broadly grouped into three categories: biogenic, thermogenic and pyrogenic. Biogenic sources contain CH_4 -generating microbes (methanogens)²², and comprise anaerobic environments such as natural wetlands and rice paddies, oxygen-poor freshwater reservoirs (such as dams), digestive systems of ruminants and termites, and organic waste deposits (such as manure, sewage and landfills). Thermogenic CH_4 is formed over millions of years through geological processes, is a fossil fuel. It is vented from the subsurface into the atmosphere through natural features (such as terrestrial seeps, marine seeps and mud volcanoes), and through the exploitation of fossil fuels, that is, through the exploitation of coal, oil and natural gas. Pyrogenic CH_4 is produced by the incomplete combustion of biomass and soil carbon during wildfires, and of biofuels and fossil fuels. These three types of emissions have different isotopic $\delta^{13}\text{C}$ signatures ($\delta^{13}\text{C} = [({}^{13}\text{C}/{}^{12}\text{C})_{\text{sample}} / ({}^{13}\text{C}/{}^{12}\text{C})_{\text{standard}}] - 1) \times 1000$): -55 to -70% for biogenic emissions, -25 to -55% for thermogenic emissions, and -13 to -25% for pyrogenic emissions^{23,24,25}. The isotopic composition of atmospheric CH_4 — measured at a subset of surface stations — has therefore been used to constrain its source^{26–28}. CH_4 emissions by living plants under aerobic conditions do not seem to play a significant role in the global CH_4 budget (Supplementary Section ST8); some very large²⁹ estimates of this source published in 2006 have not been confirmed²⁸.

The primary sink for atmospheric CH_4 is oxidation by hydroxyl radicals (OH), mostly in the troposphere, which accounts for around 90% of the global CH_4 sink. Additional oxidation sinks include methanotrophic bacteria in aerated soils^{27,28} ($\sim 4\%$), reactions with

Stefanie Kirschke, Philippe Bousquet, Philippe Ciais, Marielle Saunoy, Josep G. Canadell, Edward J. Dlugokencky, Peter Bergamaschi, Daniel Bergmann, Donald R. Blake, Lori Bruhwiler, Philip Cameron-Smith, Simona Castaldi, Frédéric Chevallier, Liang Feng, Annemarie Fraser, Martin Heimann, Elke L. Hodson, Sander Houweling, Béatrice Josse, Paul J. Fraser, Paul B. Krummel, Jean-François Lamarque, Ray L. Langenfelds, Corinne Le Quéré, Vaishali Naik, Simon O'Doherty, Paul I. Palmer, Isabelle Pison, David Plummer, Benjamin Poulter, Ronald G. Prinn, Matt Rigby, Bruno Ringeval, Monia Santini, Martina Schmidt, Drew T. Shindell, Isobel J. Simpson, Renato Spahni, L. Paul Steele, Sarah A. Strode, Kengo Sudo, Sophie Szopa, Guido R. van der Werf, Apostolos Voulgarakis, Michiel van Weele, Ray F. Weiss, Jason E. Williams & Guang Zeng (2013) **Three decades of global methane sources and sinks**. *Nature Geoscience*. doi:10.1038/ngeo1955. Published online 22 September 2013.

<http://www.nature.com/ngeo/journal/vaop/ncurrent/full/ngeo1955.html>

*A full list of authors and their affiliations appears at the end of the paper.

Global Methane Budget Website

<http://www.globalcarbonproject.org/methanebudget>

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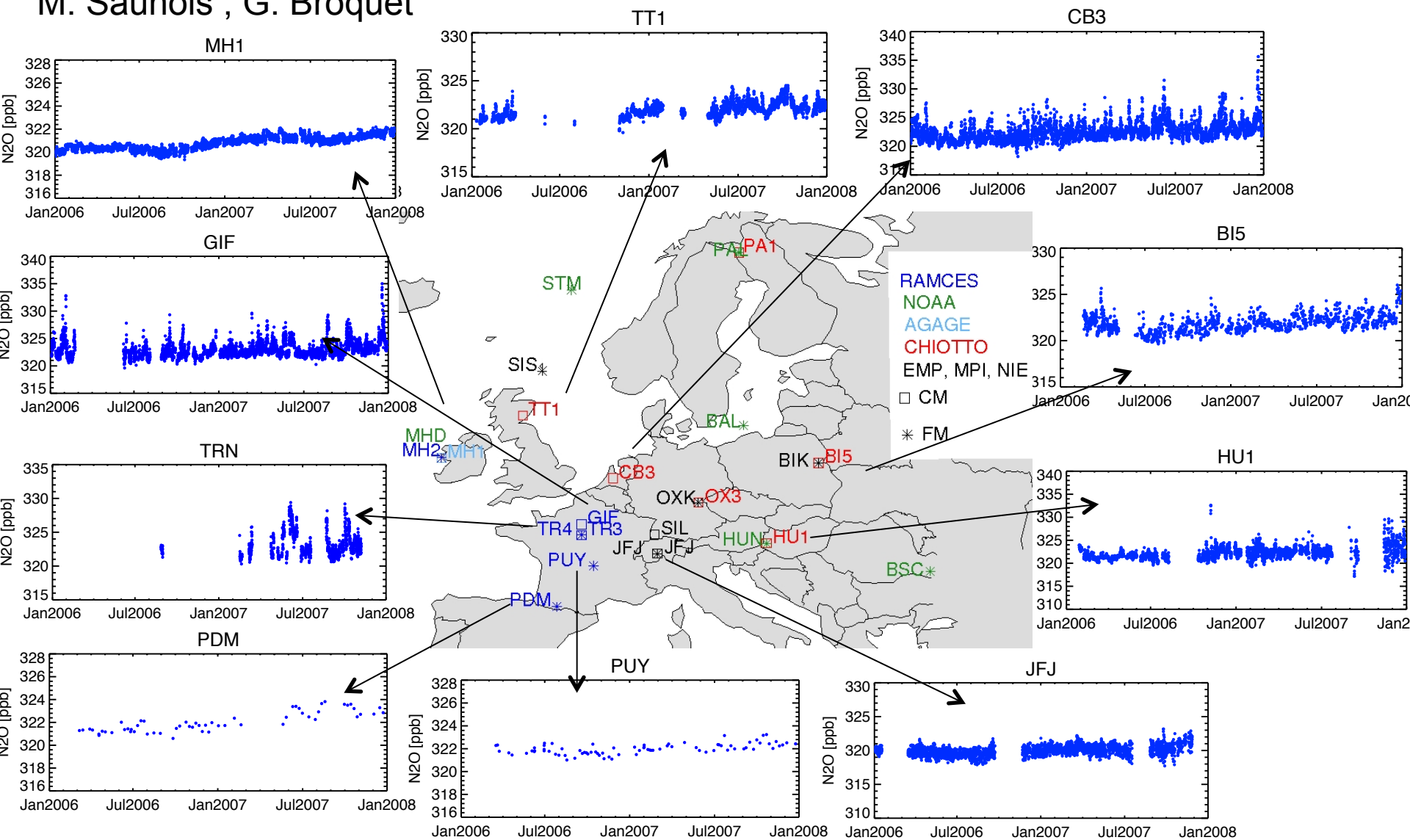
First N₂O inversions

M. Saunois , G. Broquet

N₂O inversions following IMAGINE project

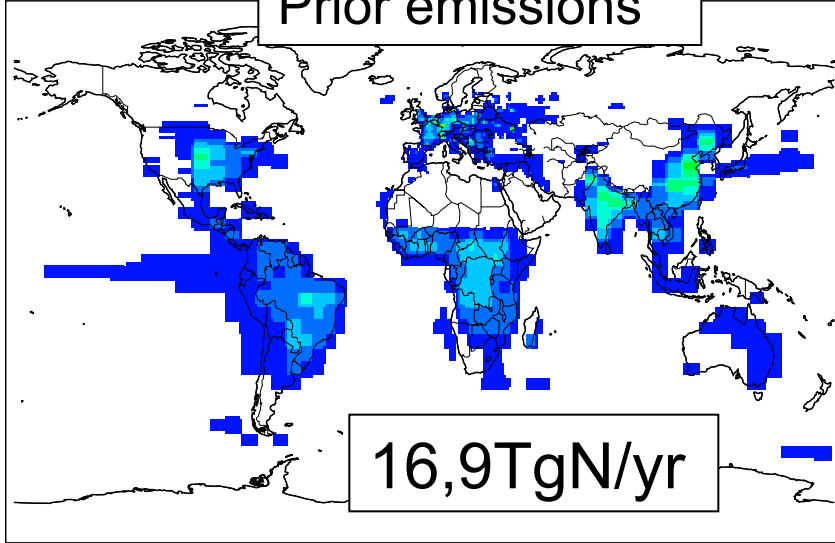


M. Saunois , G. Broquet

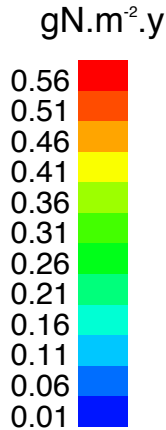
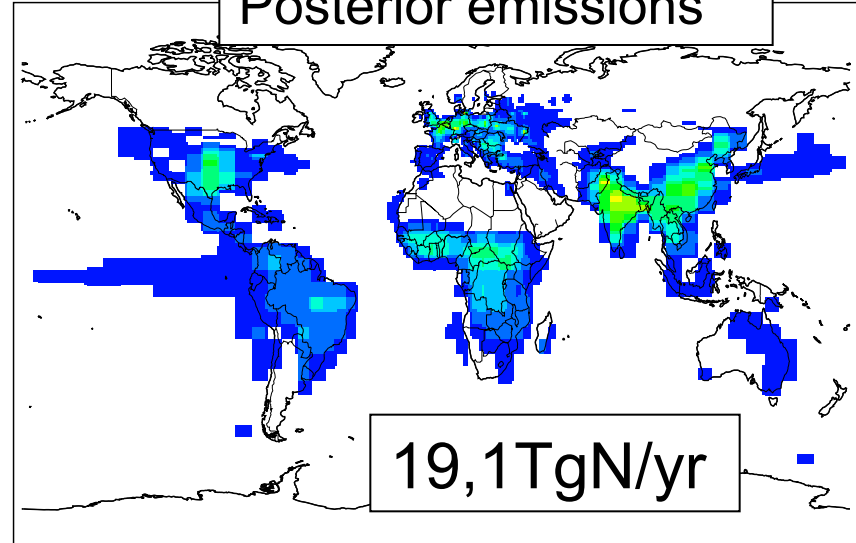


N₂O map fluxes in 2007

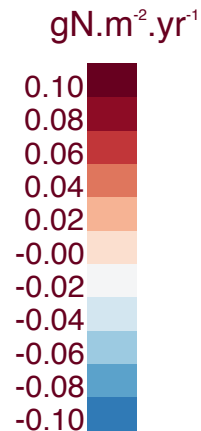
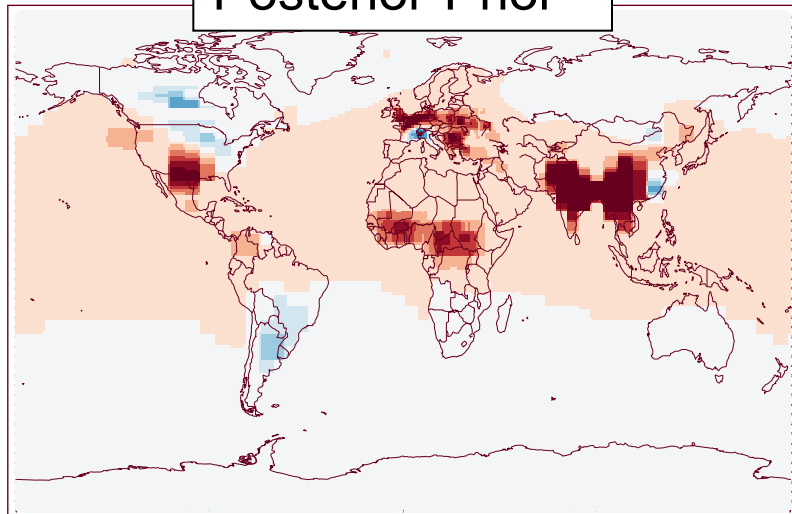
Prior emissions



Posterior emissions



Posterior-Prior



Prior Sink: 12.11 TgN/yr

Posterior Sink: 12.25 TgN/yr