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Supplement of

Comparing national greenhouse gas budgets reported in UNFCCC inventories against atmospheric inversions

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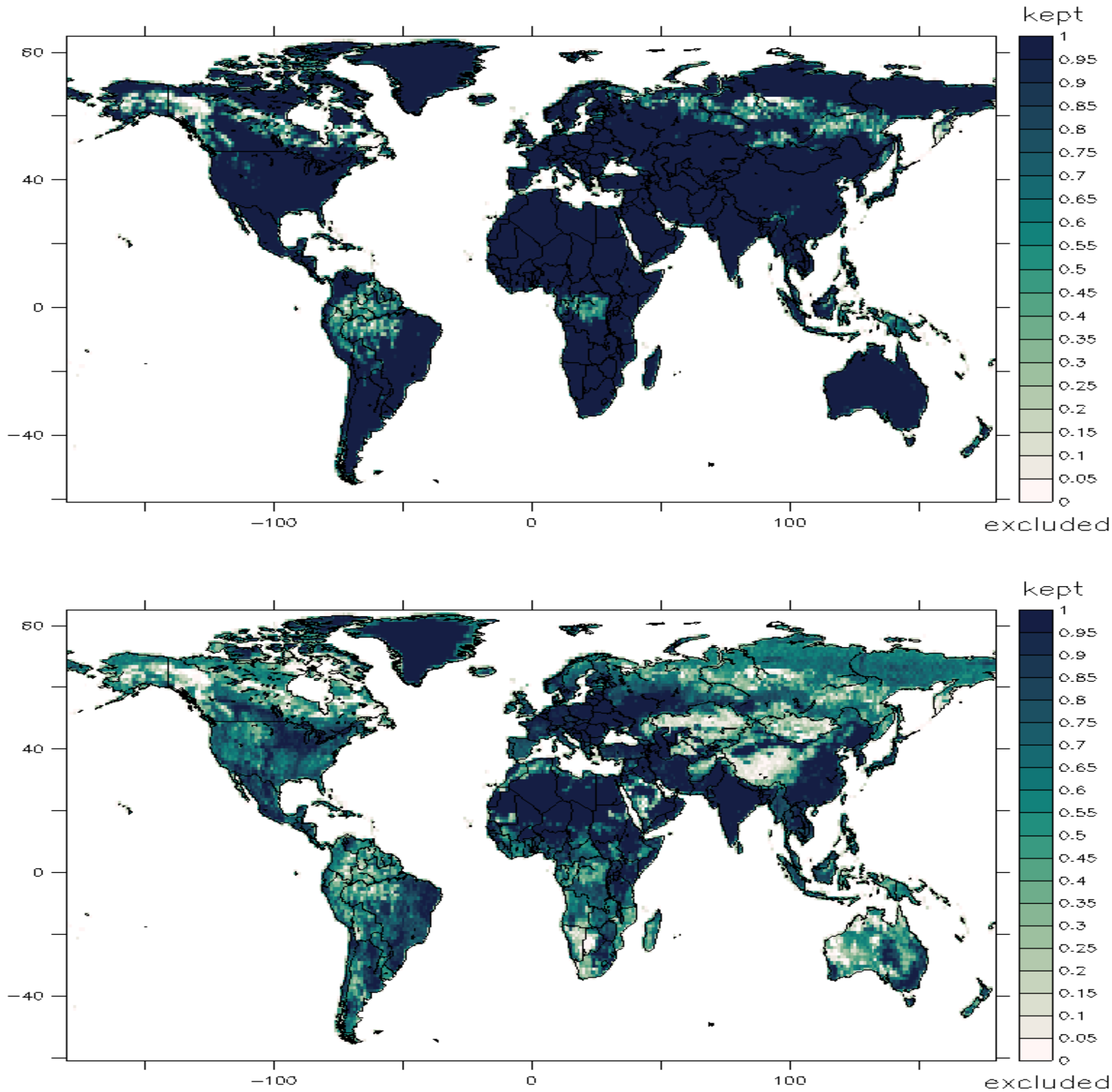
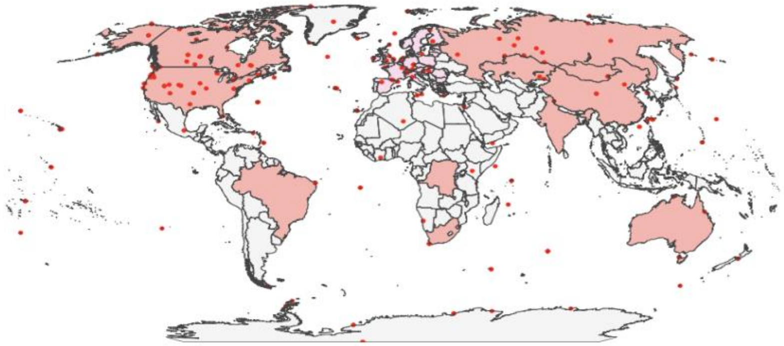


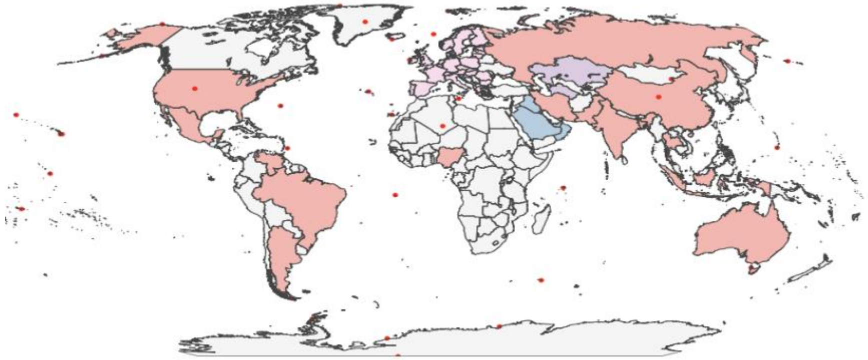
Fig S1. (top) Map of the fraction of managed land (a value of 1 means that 100% of the inversion grid cell, here of 1° resolution, is managed land) after excluding the fraction of intact forest and lightly grazed grasslands, as used to adjust N_2O inversions. (bottom). Map of managed land excluding only intact forests, as used to adjust CO_2 inversions.

(a)

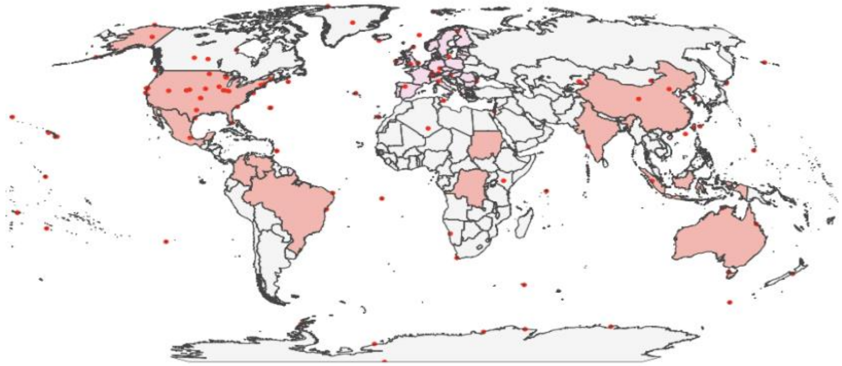
CO_2 network



CH_4 network



N_2O network



(b)

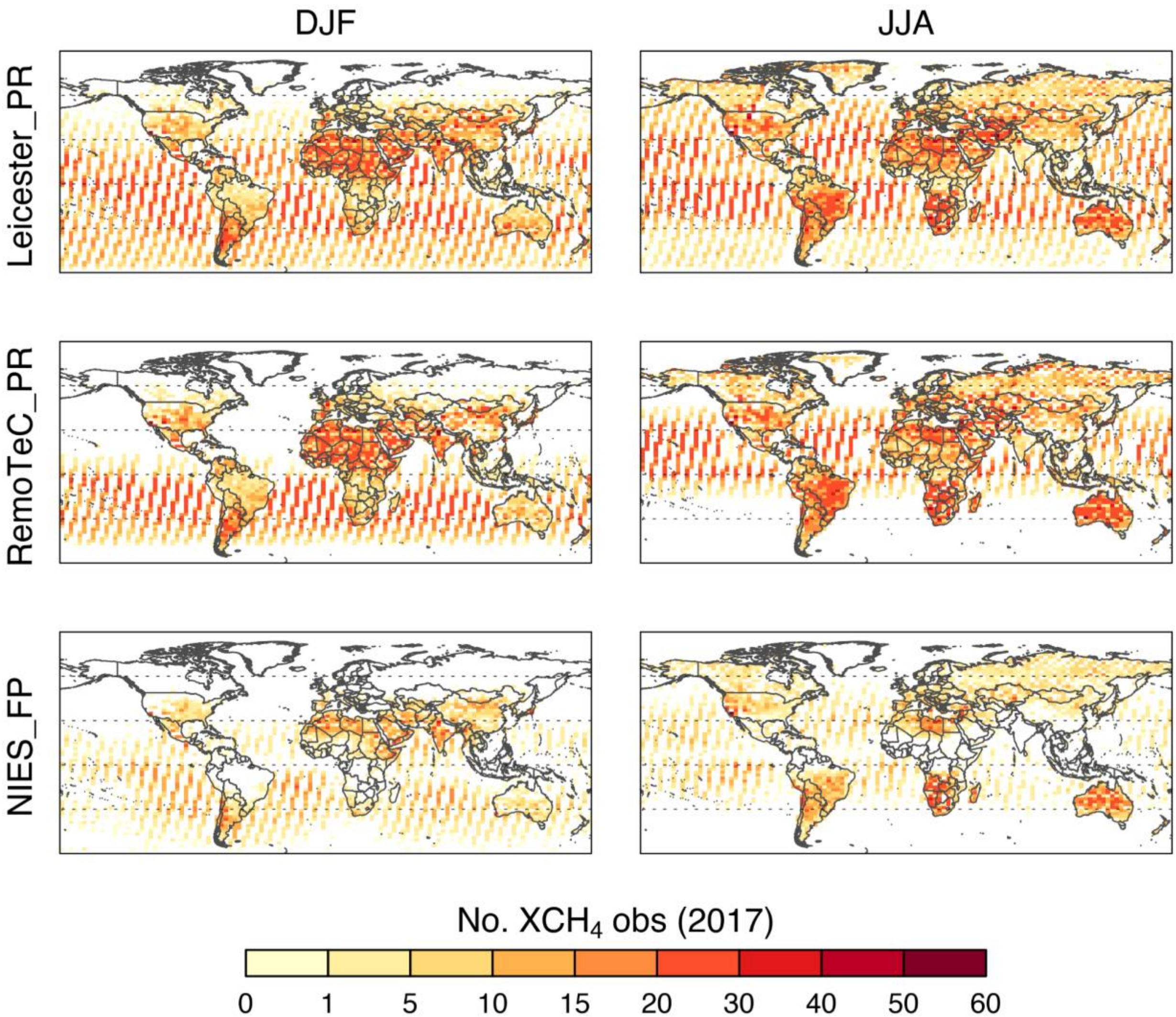


Fig S2. (a) map of the atmospheric in-situ sites whose data have been assimilated in the latest CO_2, CH_4, N_2O CAMS inversions (ship cruises have been removed from the maps). Coloured countries are those analyzed in this study (red when they are studied separately; blue, light pink or light violet when they are studied as part of a group). Note that site selection is inversion-specific: the CAMS selection may be different from any other inversion used in this study. (b) observation density of available GOSAT column CH4 soundings (XCH4) in DJF and JJA respectively for the year 2017. Each panel in (b) shows the number of daily XCH4 observations averaged at the resolution of 2° (in latitude) by 3° (in longitude). Three different GOSAT XCH4 retrievals are presented, i.e. University of Leicester proxy retrievals (v7.2), SRON RemoTeC proxy retrievals (v2.3.8), and NIES full physics retrievals (v2.7.2). See Table 1b for more details about the product used by each inversion.

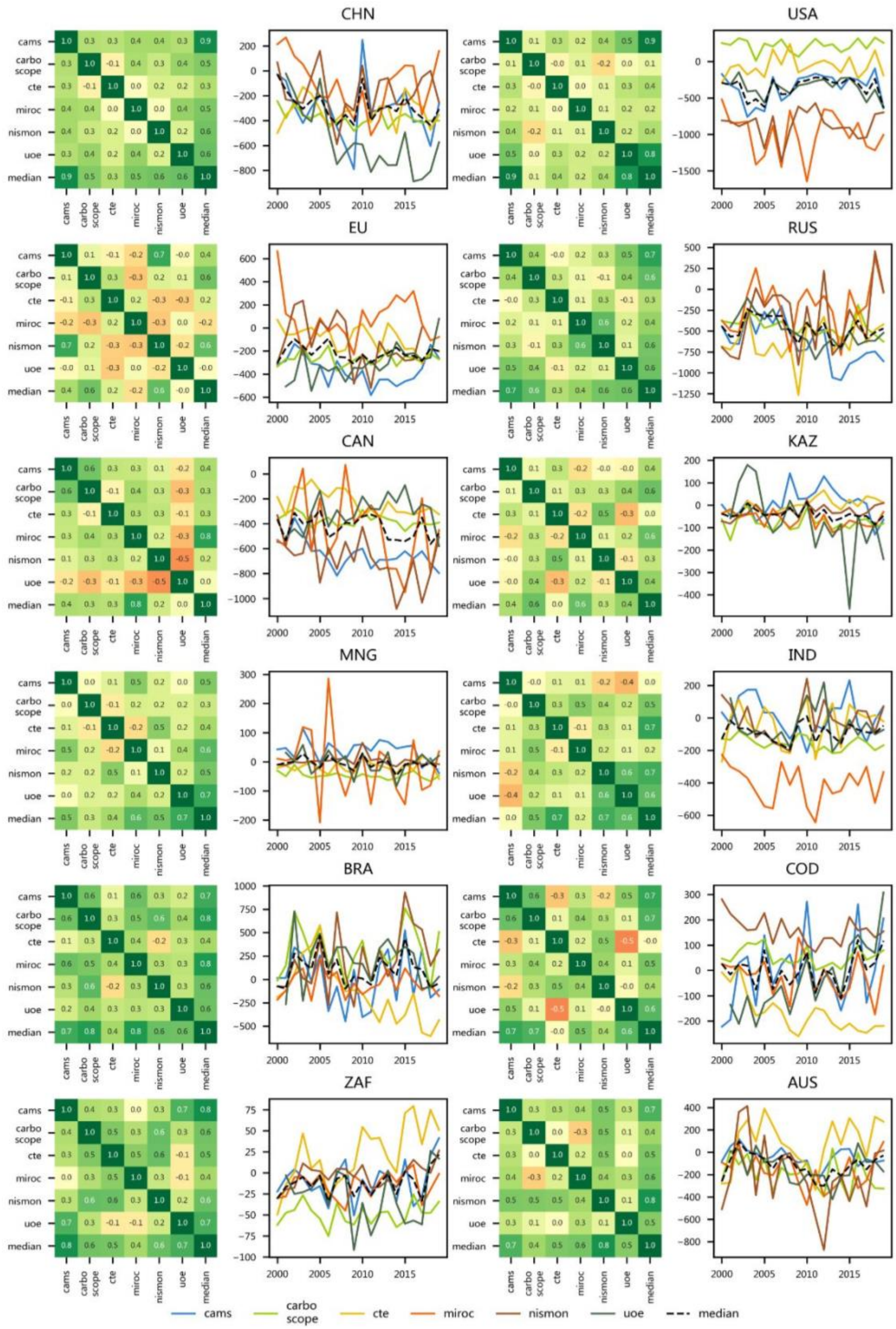


Fig S3. Correlations matrixes of the land CO2 fluxes from the six CO2 inversions for each country among the 12 selected countries shown in Fig 3.

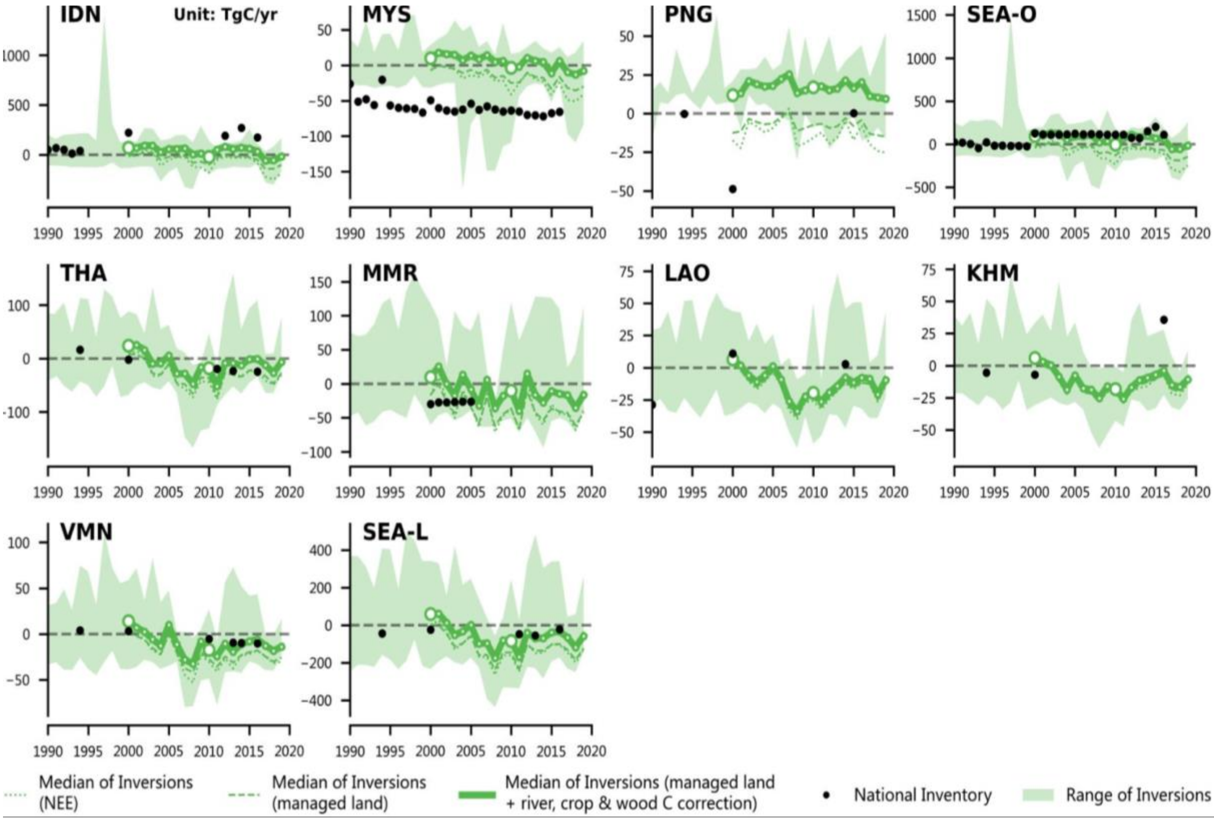


Fig S4. National carbon stock changes from inventories and land CO_2 fluxes from inversion estimates in Southeast Asia maritime continent countries including Malaysia (MYS), Indonesia (IDN), and Papua New Guinea (PNG), grouped into SEA-O, and in Southeast Asia mainland countries, Thailand (THA), Myanmar (MMR), Laos (LAO), Cambodgia (KHM), VNM (Vietnam), grouped into SEA-L.

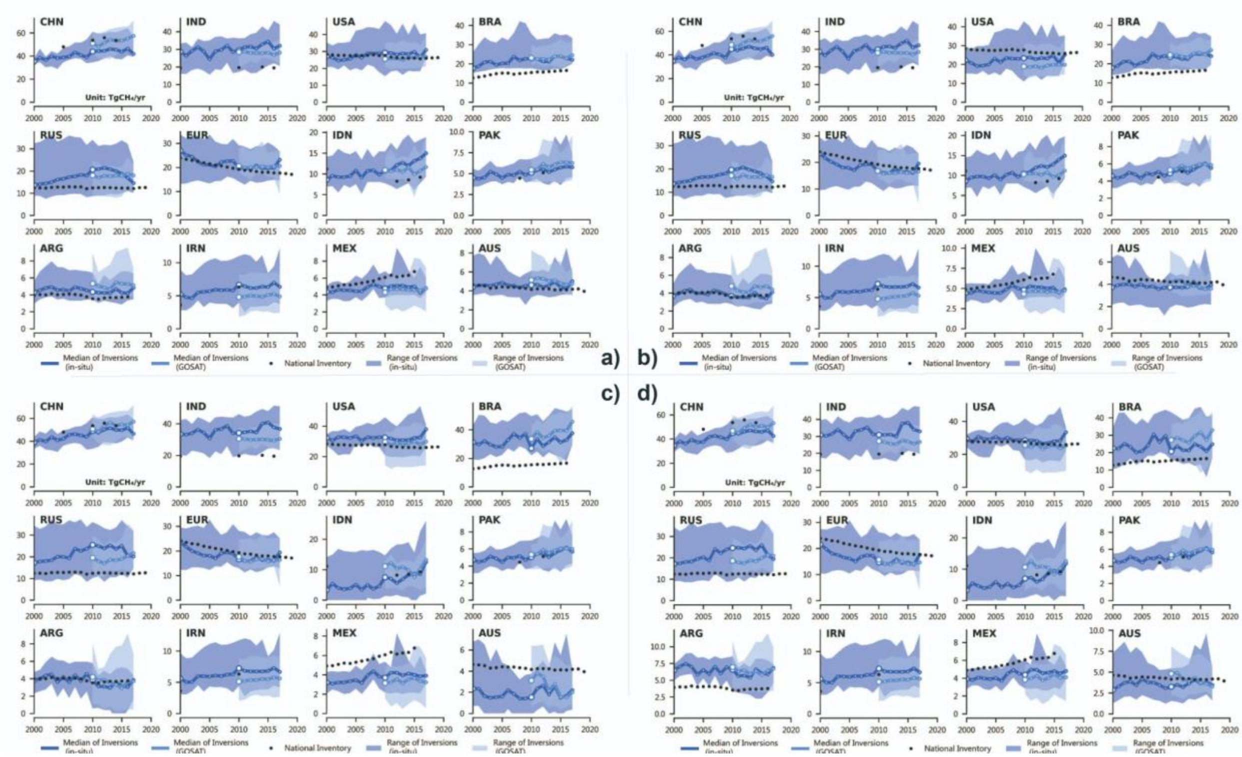


Fig S5. Anthropogenic CH_4 flux calculated from total emissions by three methods (see section 1). a) anthropogenic CH_4 emission is the sum of flux from the fossil sector, the agriculture and waste sector, and the biomass burning sector as reported by each inversion (Method 1). b), c), d) Anthropogenic CH_4 emission is calculated from the total emission of CH4 of each inversion by removing bottom-up estimations of the emissions from termites, freshwaters (lake and reservoirs) and geological, and wetland emission given by the median of inversions (Method 2) (b), or by the median of bottom-up 'diagnostic' wetland emission models prescribed by the the same wetland area (method 3/1) (c) or by the median of 'prognostic' wetland emission models with their own calculated wetland area (Method 3/2) (d).

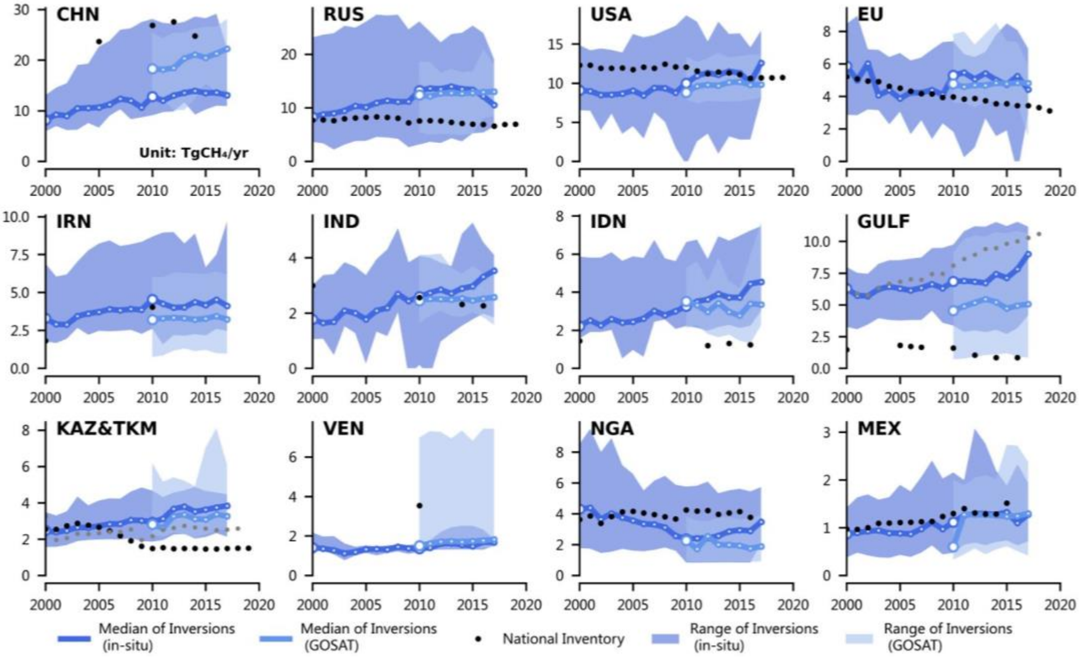


Fig S6. CH_4 emissions from the fossil fuel sector from the top 12 emitters of this sector, with the same labels as Fig 5, except for adding the grey dots for values from the PRIMAP-HIST(Gütschow et al., 2016).

Table S1. (a) List of global inversions used in this study for each greenhouse gas; (b) Global CH4 inversions constrained by GOSAT XCH4. Note that the GOSAT XCH4 retrievals used for assimilation may be different among inversions. Please refer to Table S6 of (Saunois et al., 2020) for more details.

(a)

Gas	Model	Inversion models
CO2	in-situ	CAMS CARBOSCOPE CTE MIROC NISMON UOE
CH4	GOSAT	CTE_GOSAT LMDzPYVAR_GOSAT1 (based on Zheng et al. (2018), prior fluxes based on CEDS mostly) LMDzPYVAR_GOSAT2 (based on Zheng et al. (2018), prior fluxes from GMB protocol) LMDzPYVAR_GOSAT3 (Yin et al. (2021), sim S2_GOSAT_INCA) LMDzPYVAR_GOSAT4 (Yin et al. (2021), sim S2_GOSAT_TR) LMDzPYVAR_GOSAT5 (Yin et al. (2021), sim S3_Multi_INCA)

		LMDzPYVAR_GOSAT6 (Yin et al. (2021), sim S3_Multi_TR) NTF-4DVAR_NIES_GOSAT TM5-JRC_GOSAT1 (using own prior fluxes) TM5-JRC_GOSAT2 (using prior fluxes from GMB protocol) TM5-CAMS_-GOSAT (fom CAMS SRON)
	in-situ	CTE_SURF GELCA_SURF LMDzPYVAR_SURF1 (Yin et al. (2021), sim S1_Surf_INCA) LMDzPYVAR_SURF2 (Yin et al. (2021), sim S1_Surf_TR) MIROCv4_SURF NICAM_SURF NTF-4DVAR_NIES_SURF TM5-4DVAR_SURF1 (using own prior fluxes) TM5-4DVAR_SURF2 (using prior fluxes from GMB protocol) TM5-CAMS_SURF
N2O	in-situ	PyVAR-CAMS INVICAT GEOS-Chem

(b)

CH4 inversion	CTE_CH4	LMDZ-PYVAR	NIES-TM	TM5-CAMS	TM5-JRC
References	Tsuruta et al. (2017)	Zheng et al. (2018a,b) and Yin et al. (2021)	Wang et al. (2019a) Maksyutov et al. (2020)	Segers & Houwelling (20172018, report)	Bergamaschi et al. (2013, 2018)
Resolution	6° x 4° x 25	3.75° x 1.9° x 39	2.5° x 2.5° x 32	3° x 2° x 34	6° x 4° x 25
XCH_4 retrieval	Full physics retrievals GOSAT NIES FP v2.72 (Yoshida et al., 2013)	Proxy retrievals GOSAT Leicester PR v7.2 (Parker et al., 2011)	Full physics retrievals GOSAT NIES FP v2.72 (Yoshida et al., 2013)	Proxy retrievals GOSAT RemoTeC PR v2.3.8 (Detmers & Hasekamp 2016)	Proxy retrievals GOSAT Leicester PR v7.2 (Parker et al., 2011)

Table S2. List of non-Annex I countries for the 20 largest emitters of N_2O for which indirect N_2O emissions from nitrogen leaching and / or atmospheric nitrogen deposition are reported in their UNFCCC communications. * All numbers are rounded and data reported in CO_2 equivalents by some countries were converted to N_2O using a Global Warming Potential of 265. "NOo" means no data reported in thethat national inventoriesy.

Party	NI reported indirect N2O emissions Gg N- N2O *	FAOSTAT indirect N2O emissions
China	154 (1994, NC1) 202 (2005, NC2)	184 (1994) 238 (2005)

	NC1: 154 (1994) NC2: 202 (2005) NC3: No BUR1: No BUR2: No	
Brazil	151 (2005, NC2) 183 (2015, BUR3) 113.8 (2016, NC4) 196 (2016, BUR4) NC1: No NC2: 151 (2005) NC3: No NC4: 113.8 (2016) BUR1: No BUR2: No BUR3: 183 (2015) BUR4: 196 (2016)	85 (2005) 193 (2016)
India	31 (2007, NC2) 45 (2010, BUR1) 43 (2014, BUR2) 42 (2016, BUR3) NC1: No NC2: 31 (2007) BUR1: 45 (2010) BUR2: 43 (2014) BUR3: 42 (2016)	145.8 (2007) 156.5 (2010) 159.8 (2014) 160.8 (2016)
DR Congo	NO NC1: No NC2: No NC3: No	1.2 (2015)
Indonesia	18 (2000, NC2) 37 (2014, NC3) 36 (2012, BUR1) 38 (2016, BUR2) NC1: No NC2: 18 (2000) NC3: 37 (2014) BUR1: 36 (2012) BUR2: 38 (2016)	20.0 (2000) 29.5 (2012) 29.8 (2014) 30.8 (2016)
Mexico	22 (2015, BUR2) NC1: No NC2: No NC3: No NC4: No NC5: No BUR1: No BUR2: 22 (2015)	22.6 (2015)

Colombia	NO NC1: No NC2: No NC3: No BUR1: No BUR2: No	11.2 (2015)
Sudan	NO NC1: No NC2: No	18.7 (2015)
Venezuela	23 (2010, NC2) NC1: No NC2: 23 (2010)	6.7 (2010)
Nigeria	19 (2015, BUR1) 19 (2016, NC3) NC1: No NC2: No NC3: 19 (2016) BUR1: 19 (2015)	20.0 (2015) 21.2 (2016)
Central Africa	NO NC1: No NC2: No	31.2 (2015)
Myanmar	0.8 (2000, NC1) NC1: 0.8 (2000)	5.4 (2000)
Cameroon	NO NC1: No NC2: No	3.2 (2015)
Ethiopia	27 (2013, NC2) NC1: No NC2: 27 (2013)	24.4 (2013)
Peru	10 (1994, NC1) NC1: 10 (1994) NC2: No NC3: No BUR1: No BUR2: No	4.2 (1994)
Thailand	11 (1994, NC1) 12 (2016, BUR3) NC1: 11 (1994) NC2: No NC3: No BUR1: No BUR2: No BUR3: 12 (2016)	8.6 (1994) 11.6 (2016)
Pakistan	0.13 (1993, NC1)	22.0 (1993)

	49 (2015, NC2) NC1: 0.13 (1993) NC2: 49 (2015)	39.9 (2015)

Table S3. IPCC category systems defined by the two IPCC guidelines (IPCC, 1997, 2006)

Non Annex I						**Annex I**		
IPCC 1996			**IPCC 2006**			**CRF (IPCC 2006)**		
1. Energy	**1.A Fuel Combustion - Sectoral Approach**	1.A.1 Energy Industries 1.A.2 Manufacturing Industries and Construction 1.A.3 Transport 1.A.4 Other Sectors 1.A.5 Other (Not elsewhere specified)	**1 ENERGY**	**1A Fuel Combustion Activities**	1A1 Energy Industries 1A2 Manufacturing Industries and Construction 1A3 Transport 1A4 Other Sectors 1A5 Non-Specified	**1. Energy**	**A. Fuel combustion (Reference approach / Sectoral approach)**	1. Energy industries 2. Manufacturing industries and construction 3. Transport 4. Other sectors 5. Other
	1.B Fugitive Emissions from Fuels	1.B.1 Solid Fuels 1.B.2 Oil and Natural Gas		**1B Fugitive Emissions from Fuels**	1B1 Solid Fuels 1B2 Oil and Natural Gas 1B3 Other Emissions from Energy Production		**B. Fugitive emissions from fuels**	1. Solid fuels 2. Oil and natural gas and other emissions from energy production
				1C Carbon Dioxide Transport and Storage	1C1 Transport of CO2 1C2 Injection and Storage		**C. CO2 Transport and storage**	1. Transport of CO2 2. Injection and storage 3. Other
2. Industrial Processes	**2.A Mineral Products**	2.A.1 Cement Production 2.A.2 Lime Production 2.A.3 Limestone and Dolomite Use 2.A.4 Soda Ash 2.A.5 Asphalt	**2 INDUSTRIAL PROCESSES AND PRODUCT USE**	**2A Mineral Industry**	2A1 Cement Production 2A2 Lime Production 2A3 Glass Production 2A4 Other Process Uses of Carbonates	**2. Industrial processes and product use**	**A. Mineral industry**	1. Cement production 2. Lime production 3. Glass production 4. Other process uses of carbonates

		Roofing 2.A.6 Road Paving with Asphalt 2.A.7 Other			2A5 Other (please specify)			
	2.B Chemical Industry	2.B.1 Ammonia Production 2.B.2 Nitric Acid Production 2.B.3 Adipic Acid Production 2.B.4 Carbide Production 2.B.5 Other		**2B Chemical Industry**	2B1 Ammonia Production 2B2 Nitric Acid Production 2B3 Adipic Acid Production 2B4 Caprolactam, Glyoxal and Glyoxylic Acid Production 2B5 Carbide Production 2B6 Titanium Dioxide Production 2B7 Soda Ash Production 2B8 Petrochemical and Carbon Black Production 2B9 Fluorochemical Production 2B10 Other (please specify)		**B. Chemical industry**	1. Ammonia production 2. Nitric acid production 3. Adipic acid production 4. Caprolactam, glyoxal and glyoxylic acid production 5. Carbide production 6. Titanium dioxide production 7. Soda ash production 8. Petrochemical and carbon black production 9. Fluorochemical production 10. Other
	2.C Metal Production	2.C.1 Iron and Steel Production 2.C.2 Ferroalloys Production 2.C.3 Aluminium Production 2.C.4 SF6 Used in Aluminium and Magnesium		**2C Metal Industry**	2C1 Iron and Steel Production 2C2 Ferroalloys Production 2C3 Aluminium Production 2C4 Magnesium Production 2C5 Lead		**C. Metal industry**	1. Iron and steel production 2. Ferroalloys production 3. Aluminium production 4. Magnesium production 5. Lead production

		Foundries			Production 2C6 Zinc Production 2C7 Other (please specify)			6. Zinc production 7. Other
	2.D Other Production	2.D.1 Pulp and Paper 2.D.2 Food and Drink		**2D Non-Energy Products from Fuels and Solvent Use**	2D1 Lubricant Use 2D2 Paraffin Wax Use 2D3 Solvent Use 2D4 Other (please specify)		**D. Non-energy products from fuels and solvent use**	1. Lubricant use 2. Paraffin wax use 3. Other
	2.E Production of Halocarbons and SF_6	2.E.1 By-product emissions		**2E Electronics Industry**	2E1 Integrated Circuit or Semiconductor 2E2 TFT Flat Panel Display 2E3 Photovoltaics 2E4 Heat Transfer Fluid 2E5 Other (please specify)		**E. Electronic industry**	1. Integrated circuit or semiconductor 2. TFT flat panel display 3. Photovoltaics 4. Heat transfer fluid 5. Other
	2.F Consumption of Halocarbons and SF_6			**2F Product Uses as Substitutes for Ozone Depleting Substances**	2F1 Refrigeration and Air Conditioning 2F2 Foam Blowing Agents 2F3 Fire Protection 2F4 Aerosols 2F5 Solvents 2F6 Other Applications		**F. Product uses as substitutes for ODS**	1. Refrigeration and air conditioning 2. Foam blowing agents 3. Fire protection 4. Aerosols 5. Solvents 6. Other applications

	2.G Other			**2G Other Product Manufacture and Use**	2G1 Electrical Equipment 2G2 SF6 and PFCs from Other Product Uses 2G3 N2O from Product Uses 2G4 Other (please specify)		**G. Other product manufacture and use**	1. Electrical equipment 2. SF6 and PFCs from other product use 3. N2O from product uses 4. Other
3. Solvent and Other Product Use				**2H Other (please specify)**	2H1 Pulp and Paper Industry 2H2 Food and Beverages Industry 2H3 Other (please specify)		**H. Other**	
4. Agriculture	**4.A Enteric Fermentation**		**3 AGRICULTURE, FORESTRY AND OTHER LAND USE**	**3A Livestock**	3A1 Enteric Fermentation 3A2 Manure Management	**3. Agriculture**	**A. Enteric fermentation**	1. Cattle 2. Sheep 3. Swine 4. Other livestock
	4.B Manure Management			**3B Land**	3B1 Forest Land 3B2 Cropland 3B3 Grassland 3B4 Wetlands 3B5 Settlements 3B6 Other Land		**B. Manure management**	1. Cattle 2. Sheep 3. Swine 4. Other livestock
	4.C Rice Cultivation	4.C.1 Irrigated 4.C.2 Rainfed 4.C.3 Deep Water		**3C Aggregate Sources and Non-CO2 Emissions Sources on Land**	3C1 Biomass Burning 3C2 Liming 3C3 Urea Application 3C4 Direct N2O Emissions from Managed Soils 3C5 Indirect N2O Emissions from Managed Soils 3C6 Indirect N2O		**C. Rice cultivation**	

					Emissions from Manure Management 3C7 Rice Cultivations 3C8 Other (please specify)			
	4.D Agricultural Soils	4.D.1 Direct Soil Emissions 4.D.2 Pasture, Range and Paddock Manure 4.D.3 Indirect Emissions		**3D Other**	3D1 Harvested Wood Products 3D2 Other (please specify)		**D. Agricultural soils**	
	4.E Prescribed Burning of Savannas						**E. Prescribed burning of savannas**	
	4.F Field Burning of Agricultural Residues	4.F.1 Cereals 4.F.2 Pulses 4.F.3 Tubers and Roots 4.F.4 Sugar Cane					**F. Field burning of agricultural residues**	
	4.G Other						**G. Liming**	
							H. Urea application	
							I. Other carbon-contining fertilizers	
							J. Other	

5. Land-Use Change and Forestry	**5.A Changes in Forest and Other Woody Biomass Stocks**	5.A.1 Tropical Forests 5.A.2 Temperate Forests 5.A.3 Boreal Forests			**4. Land use, land-use change and forestry**	**A. Forest land**	1. Forest land remaining forest land 2. Land converted to forest land
	5.B Forest and Grassland Conversion	5.B.1 Tropical Forests 5.B.2 Tropical Savanna / Grasslands 5.B.3 Temperate Forests 5.B.4 Grasslands 5.B.5 Boreal Forests 5.B.6 Grasslands / Tundra 5.B.7 Other				**B. Cropland**	1. Cropland remaining cropland 2. Land converted to cropland
	5.C Abandonment of Managed Lands	5.C.1 Tropical Forests 5.C.2 Tropical Savanna / Grasslands 5.C.3 Temperate Forests 5.C.4 Grasslands 5.C.5 Boreal Forests 5.C.6 Grasslands / Tundra 5.C.7 Other				**C. Grassland**	1. Grassland remaining grassland 2. Land converted to grassland
	5.D CO_2 Emissions and Removals from Soil	5.D.1 Cultivation of Mineral Soils 5.D.2 Cultivation of Organic Soils 5.D.3 Liming of Agricultural Soils				**D. Wetlands**	1. Wetlands remaining wetlands 2. Land converted to wetlands
	5.E Other					**E. Settlements**	1. Settlements remaining settlements 2. Land converted to settlements

							F. Other land	1. Other land remaining other land 2. Land converted to other land
							G. Harvested wood products	
							H. Other	N2O Emissions from Aquaculture Use CH4 from artificial water bodies
6. Waste	**6.A Solid Waste Disposal on Land**	6.A.1 Managed Waste Disposal on Land 6.A.2 Unmanaged Waste Disposal Sites	**4 WASTE**	**4A Solid Waste Disposal**	4A1 Managed Waste Disposal Sites 4A2 Unmanaged Waste Disposal Sites 4A3 Uncategorised Waste Disposal Sites	**5. Waste**	**A. Solid waste disposal**	1. Managed waste disposal sites 2. Unmanaged waste disposal sites 3. Uncategorized waste disposal sites
	6.B Wastewater Handling	6.B.1 Industrial Wastewater 6.B.2 Domestic and Commercial Wastewater		**4B Biological Treatment of Solid Waste**			**B. Biological treatment of solid waste**	1. Composting 2. Anaerobic digestion at biogas facilities
	6.C Waste Incineration			**4C Incineration and Open Burning of Waste**	4C1 Waste Incineration 4C2 Open Burning of Waste		**C. Incineration and open burning of waste**	1. Waste incineration 2. Open burning of waste
	6.D Other			**4D Wastewater Treatment and Discharge**	4D1 Domestic Wastewater Treatment and Discharge 4D2 Industrial Wastewater Treatment and Discharge		**D. Wastewater treatment and discharge**	1. Domestic wastewater 2. Industrial wastewater 3. Other (as specified in table 5.D)

				4E Other (please specify)			**E. Other**	
7. Other			**5 OTHER**	**5A Indirect N2O Emissions from the Atmospheric Deposition of Nitrogen in NOx and NH3**		**6. Other (please specify)**		
				5B Other (please specify)				
Memo Items	**International Bunkers**	Aviation Marine	**Memo Items**	**International Bunkers**	International Aviation International Water-borne Transport	**Memo Items**	**International Bunkers**	Aviation Marine
	CO_2 Emissions from Biomass			**Multilateral Operations**			**Multilateral operations**	
				CO2 from Biomass Combustion for Energy Production			**CO2 emissions from biomass**	
							CO2 captured	For domestic storage For storage in other countries
							Long-term storage of C in waste disposal sites	
							Indirect N2O	

			Indirect CO2	

CH_4 emissions estimates from ultra- emitters (large point sources) and fossil fuel extraction basins based on S5P TROPOMI satellite data and high resolution inversions

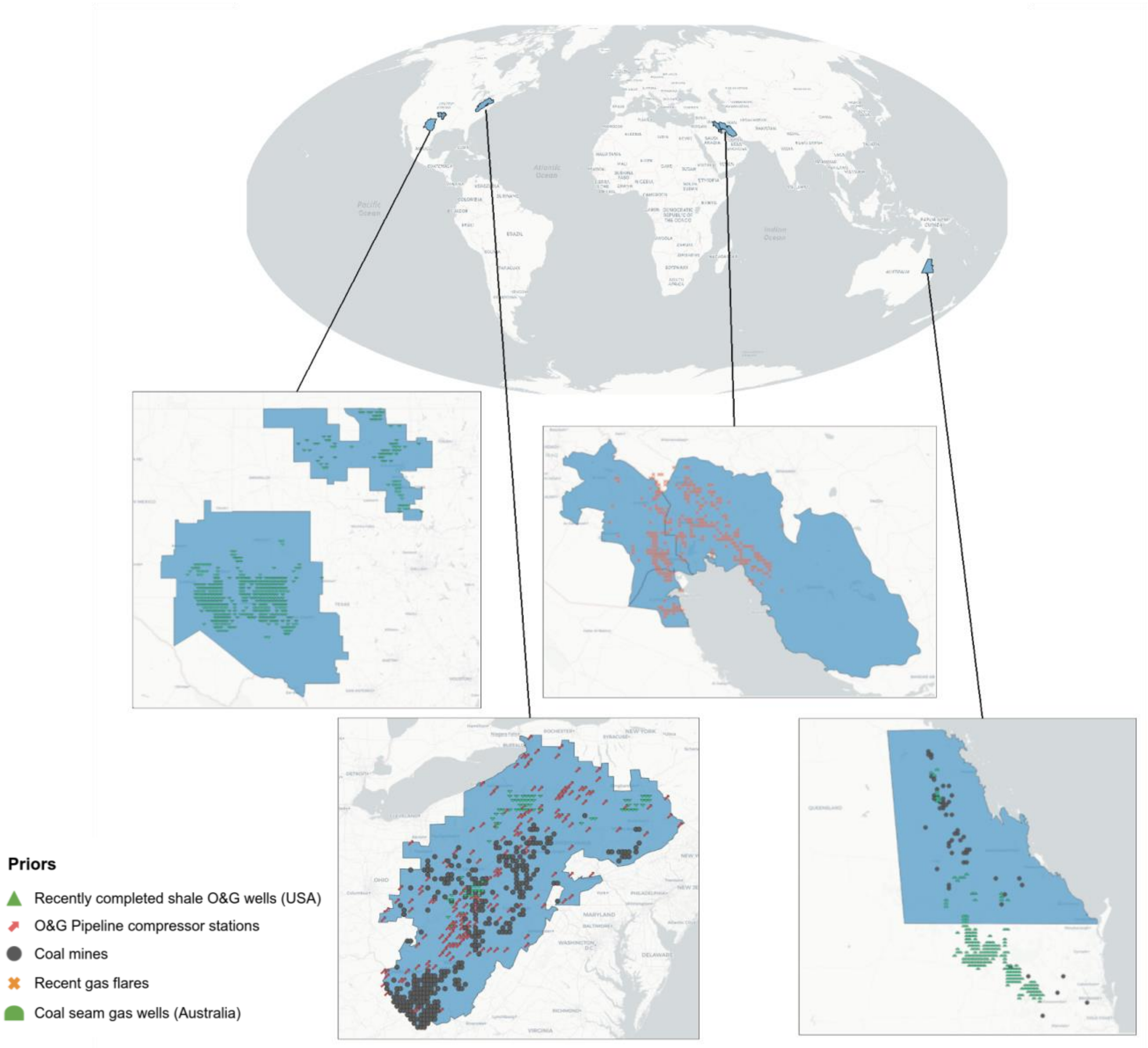


Fig S7. Main oil and gas production basins for which a basin scale inversion was obtained using S5P-TROPOMI data and regional high resolution dispersion models. Some basin inversion priors vary over time (O&G well completions and gas flares); this figure only contains a sample of points for these priors. [Map: ©Mapbox]

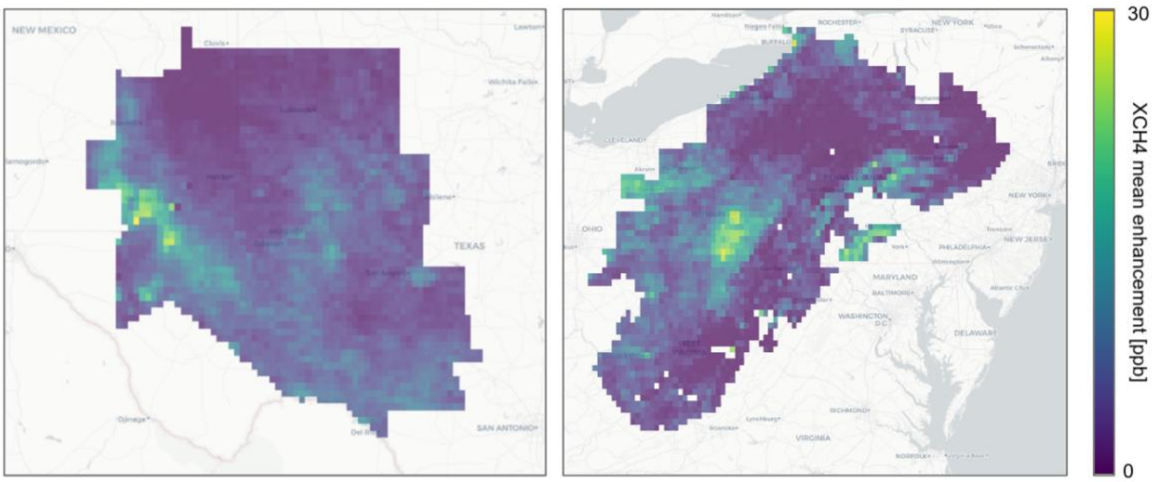


Fig S8. Mean XCH_4 enhancement over the year 2020 for the Permian and Appalachian basins (TROPOMI XCH_4 bias corrected data). [Map: ©Mapbox]

TROPOMI-based methane ultra-emitters detection

Methane ultra-emitters are detected from total atmospheric column XCH_4 images sampled by the TROPOspheric Monitoring Instrument (TROPOMI) over 2019 and 2020. TROPOMI orbits the earth 13 to 14 times per day in a sun-synchronous, near-polar trajectory, and tentatively retrieves XCH_4 measurements for most of the atmosphere on a daily basis at a 7x7 km spatial resolution. We collected and analyzed hundreds of very large point sources located over large O&G production basins and major gas transportation infrastructure. The emission rates of these ultra-emitters is estimated using the Lagrangian particle model HYSPLIT (Stein et al., 2015). Flow rates typically range from a few dozen tons per hour to several hundred tons per hour, and follow a power-law relationship with noticeable variations in emission levels across countries but similar slopes. Compensating for incomplete TROPOMI XCH_4 observations, total methane emissions from O&G ultra-emitters are derived for a sample of countries representing more than 50% of the global onshore natural gas production. The duration of release is estimated by considering that emissions are continuous if visible on two consecutive processable TROPOMI images, and that they lasted for the duration for which the HYSPLIT simulation best fits the image otherwise. A lower bound scenario (in which release durations are taken to be HYSPLIT release durations) and an upper bound scenario (in which all hotspots are supposed to release during 24 hours) are also considered; all scenarii lead to estimates in the same order of magnitude (Lauvaux et al., 2021).

TROPOMI-based methane basin inversions

Inversions of methane emission from O&G and coal basins rely on TROPOMI atmospheric XCH4 measurements. For a set of basins producing fossil fuels (see figure Fig S7), likely sources of methane due to coal or O&G activities are first identified. For shale oil and gas basins, recent well completions from Kayrros proprietary database (derived from the Sentinel 1 and 2 missions) are taken as a prior, whereas gas flares identified using VIIRS are privileged in conventional oil and gas basins. Pipeline compressor stations are added to the prior in the US O&G basin, as well as coal mines in the Appalachian and Bowen basins. In Queensland, coal seam gas wells are also taken into account. In the Appalachian, emissions due to coal are disentangled from those due to O&G by using the relative proportions of the EDGAR v5.0 gridded database. Methane plumes are simulated from the gridded prior using HYSPLIT and fitted to the background-subtracted TROPOMI XCH4 images (Fig S8). The method is similar to (Zhang et al., 2020), although the quadratic optimization program is constrained (methane emissions are non-negative), regularized (oil and gas emissions are supposed to be sparse whereas coal emissions are nearly constant), and thus solved numerically rather than in closed form, without a prior penalty term.

Code	**Country**	**CH_4 Tg yr^{-1} avg (2019-2020)**
ULTRA-EMITTER EVENTS		
GULF	Iraq	0.05
	Kuwait	0.01
KAZ & TKM	Kazakhstan	0.15
	Turkmenistan	1.49
IRN	Iran	0.42
RUS	Russia	1.71
INTENSE-EMITTING OIL AND GAS BASINS		
IRN	Iran	2.34
GULF	Iraq	1.27
	Kuwait	1.05
USA	United States	
	Anardako basin	1.01
	Appalachian basin	1.66
	Permian basin	2.34
INTENSE-EMITTING COAL BASINS		

USA	United States		
		Appalachian basin	1.07
AUS	Australia		
		Bowen Surat basin	1.55

Table S3. Emissions from ultra- emitters and intense-emitting basins of coal and of oil and gas. The uncertainty of the emission estimates have been conducted by Lauvaux et al. (2021).

Reference

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