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A high-resolution satellite-based map of global methane emissions reveals missing wetland, fossil fuel and monsoon sources

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Abstract. We interpret space-borne observations from the TROPOspheric Monitoring Instrument (TROPOMI) in a multi-inversion framework to characterize the 2018–2019 global methane budget. Evaluation of the inverse solutions indicates that methane sources and sinks cannot be separately resolved by methane observations alone—even with the dense TROPOMI sampling coverage. Employing remote carbon monoxide (CO) and hydroxyl radical (OH) observations as additional constraints, we infer from TROPOMI a global methane source of 587 Tg/y and sink of 571 Tg/y for our analysis period. We apply a new downscaling method to map these emissions to 0.1°×0.1° resolution, using the results to uncover key gaps in the prior methane budget. The TROPOMI data point to an underestimate of tropical wetland emissions (+13%; 20 Tg/y), with adjustments following regional hydrology. Some simple wetland parameterizations represent these patterns as accurately as more sophisticated process-based models. Fossil fuel emissions are strongly underestimated over the Middle East (+5 Tg/y), where they have been increasing rapidly over the past decade, and over Venezuela. The TROPOMI observations reveal many fossil fuel emission hotspots missing from the prior inventory, including over Mexico, Oman, Yemen, Turkmenistan, Iran, Iraq, Libya, and Algeria. Agricultural methane sources are underestimated in India, Brazil, the California Central Valley, and Asia. More than 45% of the global upward anthropogenic source adjustment occurs over India and southeast Asia during the summer monsoon (+8.5 Tg in Jul–Oct), likely due to rainfall-enhanced emissions from rice, manure, and landfills/sewers, which increase during this season along with the natural wetland source.

Short Summary. We combine satellite measurements with a novel downscaling method to map global methane emissions at 0.1°×0.1° resolution. These fine-scale emission estimates reveal unreported emission hotspots and shed light on the roles of agriculture, wetlands, and fossil fuels for regional methane budgets. The satellite-derived emissions point in particular to missing fossil fuel emissions in the Middle East and to a large emission underestimate in South Asia that appears to be tied to monsoon rainfall.

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1 Introduction

Methane (CH_4) has a 20-year global warming potential 85 times that of carbon dioxide (CO_2) and is an important driver of decadal climate changes (IPCC, 2021). Global mean methane mole fractions reached 1879 ppb in 2020, 2.6× pre-industrial levels, with a recent growth rate acceleration (+10–15 ppb/y in 2019–2020) whose cause is not well understood (NOAA, 2022; Saunois et al., 2020). Strong heterogeneity in methane emissions and limited observational coverage have historically challenged our ability to explain such trends in terms of underlying sources. However, the recent availability of high-resolution, near-global and daily methane measurements from the TROPOspheric Monitoring Instrument (TROPOMI) provides a transformative advance in this area. Here, we apply these data in a 4D-Var inverse framework to quantify the 2018–2019 global methane budget and determine the importance of missing and underrepresented sources.

The atmospheric methane burden increased by an average of 18 (17–19) Tg/y during 2008–2017 (Saunois et al., 2020), with conflicting explanations proposed. Top-down studies have inferred a ~30 Tg/y emission increase over tropical regions between 2000–2006 and 2017 (Bergamaschi et al., 2013; Lunt et al., 2019; Jackson et al., 2020). However, such inferences can be highly sensitive to even modest uncertainties in the atmospheric hydroxyl radical (OH, the main methane sink)—particularly over the tropics with their sparse observations (Mcnorton et al., 2016; Rigby et al., 2017; Turner et al., 2017). Some top-down studies have approached this problem by co-optimizing methane emissions and sinks (Lu et al., 2021; Maasakkers et al., 2019; Qu et al., 2021; Turner et al., 2017; Zhang et al., 2018). Unfortunately, these terms may be insufficiently resolved for robust inverse analysis when using methane data alone, leading to aliasing between the optimized source and sink terms (Lu et al., 2021). Isotopic analyses invoke increased biogenic sources (Nisbet et al., 2016; Schaefer et al., 2016) to explain the post-2016 ^{13}C depletion, whereas ethane-based constraints indicate a fossil fuel emission underestimate (Franco et al., 2016; Kort et al., 2016; Peischl et al., 2016; Xiao et al., 2008). Unfortunately, the latter approach is limited by the large variability in methane-to-ethane emission ratios.

Bottom-up inventories also point to substantial uncertainties in the spatial distribution of methane sources. For instance, the two most commonly-used anthropogenic inventories for the US (EDGAR v5 (2019) and GEPA (Maasakkers et al., 2016)) are essentially uncorrelated ($R^2 = 0.01$) at 0.1°×0.1° resolution. These spatial biases, when coupled with sparse observations, model transport errors, and source/sink ambiguity, degrade the accuracy of observation-based (top-down) emission estimates—which as a consequence often arrive at inconsistent emission allocations (Alexe et al., 2015; Bruhwiler et al., 2014; Jackson et al., 2020; Lu et al., 2021; Maasakkers et al., 2019; Mcnorton et al., 2018; Monteil et al., 2013; Qu et al., 2021; Yu et al., 2020; Yu et al., 2021a; Yu et al., 2021c; Zhang et al., 2021).

TROPOMI provides an unprecedented observational expansion for addressing these science gaps, offering sub-10 km global monitoring of total column methane concentrations with dense overland coverage (Bousserez et al., 2016; Jacob et al., 2016;

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Turner et al., 2018b). Here, we interpret two years of TROPOMI data in an analysis framework that couples multiple 4D-Var adjoint inversions with a novel spatial downscaling approach to derive emissions at 0.1° × 0.1° horizontal resolution. This yields a suite of candidate solutions for the 2018-2019 methane budget, which we evaluate a posteriori against independent observations of methane, carbon monoxide (CO), and OH. In this way we identify the most robust solution set based on the ensemble of observational constraints, and use this new spatial information to better understand regional and sectoral contributions to the methane budget and the underlying drivers of those emissions.

2 Data and Methods

Figure 1 summarizes our inversion framework. We employ TROPOMI measurements from 01/2018–02/2020 with the GEOS-Chem adjoint model in a suite of 4D-Var inversions to optimize monthly methane emissions at 2° × 2.5° (latitude × longitude) resolution. These derived emissions are then spatially downscaled to 0.1° × 0.1°. We omit the first and final 4 months from interpretation to further minimize initial condition errors and to ensure that all derived fluxes are adequately informed by subsequent observations. Our final analysis timeframe thus spans 18 months from 05/2018 through 10/2019.

2.1 TROPOMI observations

TROPOMI was launched in 10/2017 onboard the Copernicus Sentinel-5 Precursor satellite into a low-Earth polar orbit, and monitors greenhouse gases and air pollutants with daily near-global coverage at ~13:30 LT (equator overpass) on the ascending node (Hu et al., 2018). We use the SRON corrected retrieval described in Lorente et al. (2021), which is based on the S5P-RemoTeC full-physics algorithm with albedo correction and updated regularization scheme, spectroscopic information, and surface treatment. This updated algorithm mitigates the albedo bias that affected earlier versions (Qu et al., 2021). The total column (XCH_4 in ppb) retrievals employ combined solar backscatter measurements in the near-infrared (NIR; 0.8 μm) and shortwave-infrared (SWIR; 2.3 μm), and have 5.5/7 × 7 km^2 nadir resolution on a 2600 km swath. The data have <1% nominal bias, 0.6% instrument noise, and an estimated 0.8% forward model error (Hu et al., 2016). We omit high-latitude (>60°) observations and require quality filter QA > 0.5 to avoid errors associated with high solar or viewing zenith angles, low surface albedo, excessive aerosol loading, clouds, terrain roughness, and measurement noise (Lorente et al., 2021). Figure 2 shows the resulting TROPOMI XCH_4 data for 03/2018–02/2020, gridded to 0.1°×0.1° using the method described by Sun et al. (2018). In total, 91 million retrievals during 05/2018–10/2019 pass quality filtering and are available for analysis, an average of 31,000 per 2°×2.5° GEOS-Chem grid cell (Figure S1).

Figure S2 shows that the TROPOMI measurements agree well with independent TCCON (2014) and GOSAT (2021) observations, with major axis regression slopes of 1.02 and 0.99, respectively. The inter-dataset mean biases are -7.1 ppb (0.4%, TROPOMI - GOSAT) and -5.4 ppb (0.3%, TROPOMI - TCCON; see Text S1). Our initial condition optimization

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further ensures that the model and TROPOMI are unbiased with respect to each other, so that mismatches arising during the simulation timeframe reflect source-sink disparities rather than any systematic observational bias.

2.2 Forward model and initial conditions

We use the GEOS-Chem adjoint model (v35), on a 2° × 2.5° grid with 47 vertical layers, to perform the global 4D-Var inversions. The model uses GEOS-FP meteorological fields from the National Aeronautics and Space Administration (NASA) Global Modeling and Assimilation Office (GMAO, 2013), with 5- and 10-minute timesteps for transport and emissions, respectively. Transport employs fully instantaneous boundary layer mixing (Wu et al., 2007), a relaxed Arakawa-Schubert convection scheme (Moorthi and Suarez, 1992), and a multi-dimensional Flux-Form Semi-Lagrangian (FFSL) treatment for advection (Lin and Rood, 1996). For all model-satellite comparisons the GEOS-Chem output is sampled according to the TROPOMI observation operator at the overpass time and location.

We optimize the model initial conditions for 01/01/2018 in three steps, first starting with a 25-year global spin-up to achieve a globally representative methane field. We then apply a latitude-dependent correction based on the TROPOMI-model difference for 11/2017–01/2018 to speed up the optimization process in the next step. Correction over land employs the median TROPOMI-model difference by latitude; over oceans (which lack TROPOMI XCH_4 data) we use the 0.1 quantile difference. Finally, we optimize the resulting fields in a 4D-Var inversion based on TROPOMI data for 01–02/2018. The optimized global methane burden is 0.99× that in the original 25-year spinup, and the north:south hemispheric (NH:SH) XCH_4 ratio increases from 1.11 to 1.13.

2.3 Prior methane sources and sinks

Global anthropogenic emissions in the prior model use the gridded United Nations Framework Convention on Climate Change (UNFCCC) inventory for fossil fuels (year-2016; (Scarpelli et al., 2020)) and EDGAR v5 for other sources (year-2015; (Crippa et al., 2019; Crippa et al., 2020)). These are superseded by the 2012 GEPA inventory for US anthropogenic emissions (Maasakkers et al., 2016), the CanMex inventory for Canadian (year-2013) and Mexican (year-2010) oil and gas emissions (Sheng et al., 2017), and Sheng et al. (2019) for Chinese coal mine emissions (year-2011). Wetland emissions use the 2018–2019 WetCHARTs ensemble mean flux (Text S2; (Bloom et al., 2017)), scaling the total to 149 Tg/y to match the 2008–2017 global methane budget from Saunois et al. (2020). We apply Fung et al. (1991) and Maasakkers et al. (2019) for termite and geological seep emissions, respectively, and employ biomass burning emissions for 2018–2019 from the Quick Fire Emissions Dataset (QFED (Darmenov and Silva, 2015)). Figure S3 maps these prior emissions, which total 535 Tg/y and include 356 Tg/y from anthropogenic sources (119 Tg/y livestock + 101 Tg/y fossil fuel + 80 Tg/y waste + 37 Tg/y rice + 19 Tg/y other), 165 Tg/y from natural sources (149 Tg/y wetlands + 16 Tg/y geological seeps and termites), and 14 Tg/y from biomass burning. Emissions from EDGAR v5 vary monthly based on national and sub-national sectoral activity levels. GEPA includes monthly emission profiles for US rice and manure management, and we assume aseasonal emissions for

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indoor animal husbandry following Crippa et al. (2020). The WetCHARTs emissions are monthly, reflecting temporal changes in wetland extent, respiration, and temperature. QFED emissions are daily with an hourly diel profile applied.

Atmospheric methane removal by OH (87% of the total sink, 494 Tg/y) in the prior model uses archived monthly oxidant fields from GEOS-Chem (v5) benchmark simulations (Wecht et al., 2014), which have an annual tropospheric air-mass-weighted mean of 1.03×10^6 molecules/cm^3 and 1.04 NH:SH ratio. Other minor sinks include stratospheric oxidation (6%, 33 Tg/y) based on NASA Global Modeling Initiative monthly loss frequencies (Murray et al., 2013), soil uptake (6%, 34 Tg/y) following Fung et al. (1991), and tropospheric oxidation by chlorine (2%, 10 Tg/y) using 3-D monthly Cl fields from Sherwen et al. (2016). The above sinks total 571 Tg/y (Figure 2e) and yield a 9.1-year methane lifetime in our prior simulations.

2.4 Inversion frameworks and sensitivity to OH

Optimizations are performed in the GEOS-Chem adjoint model (Henze et al., 2007) through iterative minimization of the Bayesian cost function $J(\boldsymbol{x})$:

$$J(\boldsymbol{x}) = (\boldsymbol{x} - \boldsymbol{x_a})^{\mathrm{T}}\mathbf{S_a^{-1}}(\boldsymbol{x} - \boldsymbol{x_a}) + \boldsymbol{\gamma}\big(\boldsymbol{y} - F(\boldsymbol{x})\big)^{\mathrm{T}}\mathbf{S_0^{-1}}(\boldsymbol{y} - F(\boldsymbol{x})) \quad (1)$$

The first right-hand term imposes a penalty based on the deviation of $\boldsymbol{x}$ (the state vector to be optimized) from $\boldsymbol{x_a}$ (the prior estimates), weighted by the prior error matrix $\mathbf{S_a}$. The state vector $\boldsymbol{x}$ includes monthly 2° × 2.5° grid-level emissions and (in some cases) the 2-year-mean hemispheric loss to OH. This penalty is counteracted by the second right-hand term, which reflects the mismatch between the observations $\boldsymbol{y}$ and model predictions $F(\boldsymbol{x})$ sampled in the same manner, weighted by the observing system error matrix $\mathbf{S_o}$. The regulation parameter γ is applied to balance the influence of the above two terms in the overall cost function $J(\boldsymbol{x})$.

The prior error covariance matrix $\mathbf{S_a}$ for methane emissions is constructed as follows. We use the Maasakkers et al. (2016) scale-dependent uncertainties for anthropogenic emissions over the GEPA and CanMex domains, and the Sheng et al. (2019) province-level error estimates for Chinese coal mine emissions. For other global anthropogenic emissions we use the gridded fossil fuel uncertainty estimates from Scarpelli et al. (2020) and assume 50% uncertainty in the remaining sources. Uncertainties for wetland emissions are derived as one standard deviation across the WetCHARTs ensemble, averaging 105% at the grid level. Other sources employ a prior error standard deviation of 50%, consistent with earlier studies (Maasakkers et al., 2019; Sheng et al., 2018; Turner et al., 2015; Wecht et al., 2014; Yu et al., 2021c). The diagonal of the prior error matrix combines the above flux-weighted terms in quadrature and averages 66%. We find that the spatial covariance in the total prior emissions decreases by 50% over a mean distance of approximately 300 km, and we populate the exponentially decaying off-diagonal elements of $\mathbf{S_a}$ accordingly. This is comparable to the 200–500 km correlation length scales applied in previous methane studies (Monteil et al., 2013; Wecht et al., 2014; Yu et al., 2021c).

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We test the impacts of OH on our results through three separate inversion treatments: the first uses the prior OH with no optimization, while the second and third optimize methane loss to OH on a hemispheric basis with an assigned uncertainty of either 1% or 10% (Prather et al., 2012; Saunois et al., 2020). We find that both the 1% and 10% uncertainty assumptions for OH give effectively identical inversion solutions: in both cases the optimization has sufficient OH flexibility to correct a global mean budget imbalance on that basis, with the remaining spatial errors resolved through grid-level emission adjustments (Figure S4). We therefore mainly discuss results from the 1% optimization (referred to as optOH), along with those from the fixed OH (fixOH) inversion.

Observing system errors combine measurement errors and forward model errors. Building on Heald et al. (2004), we compute the elements of $\mathbf{S_o}$ (which is diagonal) from the residual standard deviation between the observations and the prior simulations within a 2° × 2° moving window, further imposing a lower bound of 56 ppb^2 (0.25 quantile of the overall results). The resulting observing system errors average 11 ppb, mainly reflecting instrument noise, and are comparable to previous estimates for GOSAT and TROPOMI (11–13 ppb; (Maasakkers et al., 2019; Zhang et al., 2020)).

The regulation parameter γ is defined through sensitivity inversions for 01/2018 with γ varying from 10^{-5} to 10^{3}. The optimal monthly value is selected based on the resulting L-curve and total error reductions (Figure S5), and then scaled to the number of observations for the full 2-year inversion period. The result ($\gamma = 0.03$) is consistent with previous TROPOMI inversions by Qu et al. (2021) ($\gamma = 0.002$) given their optimization of annual rather than monthly emissions.

2.5 Inversion ensemble to explore sensitivity to missing sources

Our previous work using Observing System Simulation Experiments (OSSEs) (Yu et al., 2021a) demonstrated that classical SF-based inversions have limited ability to recover missing sources. Here, we apply multiple inversion formalisms to diagnose and address this issue.

1. Classical scaling factor (SF) inversions, employing the bottom-up inventories described earlier as prior. These inversions solve for scale factors $\boldsymbol{s}$ in $\boldsymbol{x} = \boldsymbol{s} \circ \boldsymbol{x_a}$.
2. Background increment inversions (BI). BI inversions employ a revised prior consisting of the above inventories scaled by 90% plus the remaining 10% as a uniform overland flux. This revised prior is then subjected to SF optimization.
3. Observational guess inversions (OG). OG inversions employ a revised prior informed by long-term TROPOMI data for better recovery of missing sources. Specifically, we find from sensitivity simulations that adding 75 Tg/y to the bottom-up emissions results in a globally unbiased simulation across 2018–2020. We distribute this 75 Tg/y spatially based on the observation-model enhancement mismatches, where the grid-level enhancements are computed as the local XCH_4 value minus the zonal mean (2° bins). Figure S6 shows the resulting grid-level emission increments.

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4. Emission enhancement inversions (EE). EE inversions solve for absolute flux increments rather than scale factors via $\boldsymbol{x} = \boldsymbol{s} \circ \boldsymbol{x_{base}} + \boldsymbol{x_a}$. We define $\boldsymbol{x_{base}}$ as 2600 kg/box/timestep, which is the mean emission for grid cells exceeding 1 kg/box/timestep. We showed previously that this approach has better performance for missing sources than the above three SF inversions (Yu et al., 2021a).

2.6 Emission downscaling

The current GEOS-Chem adjoint model does not have global simulation capability at finer than 2° × 2.5° resolution. Furthermore, each of the 2-year inversions performed here required >12,000 CPU hours (>80 days on multiple processors) to converge, making higher-resolution optimizations computationally impractical. However, the inventories employed as prior, as well as the TROPOMI observations themselves, contain information at much finer scales (e.g., 0.1° × 0.1° and 7 × 7 km^2)—and thus contain additional high-resolution constraints that are neglected by the 2° × 2.5° inversions. We therefore leverage this information to spatially downscale the optimized emissions to 0.1° × 0.1° via:

$$x'_j = \omega_i \beta_{OBS,i\to j} s_i x'_{a,j} + (1-\omega_i)\left(1+\beta_{prior,i\to j}(s_i-1)\right)x'_{a,j} \quad (2)$$

Equation (2) downscales the original optimized emissions from a given 2° × 2.5° parent grid cell i to the subgrid scale (x'_j at 0.1° × 0.1°) by combining spatial information from the observations (first right-hand term) and the prior (second right-hand term). Here, ω_i is a weighting factor to balance these two terms, $\beta_{OBS,i\to j}$ and $\beta_{prior,i\to j}$ are spatial downscaling operators representing the observational and prior information, respectively, $x'_{a,j}$ represents the 0.1° × 0.1° prior emissions for subgrid j, and s_i is the 2° × 2.5° scale factor derived for parent grid cell i.

The observational downscaling operator $\beta_{OBS,i\to j}$ spatially allocates the subgrid-level emissions according to the distribution of column enhancements over the regional background:

$$\beta_{OBS,i\to j} = \left(y_{2y,j} - y_{bg,i}\right) \Big/ \sum_{k\in j}\left(f_k\left(y_{2y,k} - y_{bg,i}\right)\right) \quad (3)$$

where $y_{2y,j}$ is the 2-year mean (03/2018–02/2020) TROPOMI methane column sampled to 0.1° × 0.1° (Sun et al., 2018) and $y_{bg,i}$ is the methane background defined as 95% of the $y_{2y,j}$ 0.1 quantile across the parent 2° × 2.5° grid cell i. This background definition was found via OSSE analysis (described below) to yield the best consistency with the underlying fine-scale emissions. f_k quantifies the prior fraction of total 2° × 2.5° emissions contained in each subgrid $k \in j$.

The prior downscaling operator $\beta_{prior,i\to j}$ spatially allocates the derived flux enhancement $s_i - 1$ based on the prior emission magnitudes and their uncertainties:

$$\beta_{prior,i\to j} = \varepsilon'_{a,j} f_j \Big/ \sum_{k\in j}\left(f_k^2 \varepsilon'_{a,k}\right) \quad (4)$$

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where $\varepsilon'_{a,j}$ is the prior emission error estimate at 0.1° × 0.1°. In this way larger corrections are preferentially assigned to locations with higher prior emissions and uncertainties.

When summed to 2° × 2.5°, both the prior and observational downscaling terms maintain the original adjoint-derived emissions. Weighting between these terms is computed as:

$$\omega_i = \varepsilon_{a,i}\hat{\sigma}_{a,i}/\max_{l\in i}(\varepsilon_{a,l}\hat{\sigma}_{a,l}) \quad (5)$$

where $\varepsilon_{a,i}$ is the prior emission error estimate at 2° × 2.5°, and $\hat{\sigma}_{a,i}$ is the log-transformed standard deviation of all 0.1° × 0.1° prior emissions contained in that parent grid cell (with an imposed zero lower bound). As shown in Figure S7, the resulting downscaling relies most frequently on the prior information, particularly for low-emission areas, but hotspots and locations with higher prior uncertainties are preferentially informed by the observations.

We tested the effectiveness of this downscaling approach in a 1-month OSSE analysis over North America. These synthetic inversions follow Yu et al. (2021a) in prescribing true and prior emissions from distinct inventories (true: Gridded EPA + WetCHARTs ensemble mean; prior: EDGAR v5 + a single WetCHARTs member) that differ by 76 Gg/d domain-wide and have spatial R^2 = 0.26 at 0.25° × 0.3125°. The OSSEs were performed at 2° × 2.5° by sampling the true-state model according to the TROPOMI coverage for 08/2018 (with measurement noise applied), followed by 4D-Var optimization as detailed by Yu et al. (2021a). The 2° × 2.5° adjoint solution was then spatially downscaled to 0.25° × 0.3125° following Eq. (2) and compared to both the true fluxes and to the adjoint solution performed directly on the fine-scale grid. Tests were performed both in the presence and absence of model transport error (Yu et al., 2021a).

Table 1 shows that in the absence of transport error our downscaling approach outperforms the coarse-grid solution and approaches the skill of the native fine-scale inversion in representing the true fluxes. The benefits of the 4D-Var + downscaling approach are even more pronounced when accounting for transport error. Specifically, our previous OSSE analyses showed that high-resolution 4D-Var inversions failed to improve methane emission estimates at 25 km for scenarios with both transport error and spatial source uncertainty (Yu et al., 2021a). Our tests here show that spatial downscaling of the 2° × 2.5° adjoint solution strongly mitigates these effects (Table 1), yielding a larger bias reduction (98% versus just 16%) and more accurate flux distribution (R = 0.70 versus 0.60) than the native fine-grid 4D-Var solution. Given these results and the finer-scale information available here, for the present work we apply Eq. (2) to spatially downscale our inversion solutions to 0.1° × 0.1°.

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3 Derived Global Methane Budget and Sensitivity to OH

3.1 Methane source-sink ambiguity

The set of inversion configurations includes multiple formalisms for emission adjustment and two separate treatments for methane loss: fixOH inversions use the prior OH as a fixed constraint, while optOH inversions optimize both OH concentrations (as a 2-year hemispheric mean) and methane emissions. Figure 2 shows that the fixOH and optOH multi-model means yield similar atmospheric methane distributions, with the strongest enhancements over central Africa, South and East Asia, the Middle East, Amazonia and the southern US, and the lowest column concentrations over high southern latitudes in Australia, South America, and Africa. The two approaches also provide comparable improvement with respect to the TROPOMI observations, with >97% mean bias reduction and >45% root-mean-square-error (RMSE) reduction.

Despite the above patterns of agreement, the fixOH and optOH inversions lead to opposing methane emission changes relative to the prior budget. The fixOH multi-model mean provides a global methane source of 587 Tg/y (10% higher than the prior) and a sink of 571 Tg/y. The optOH multi-model mean yields a 514 Tg/y source (4% below the prior) and a 492 Tg/y sink; the latter is driven by updated OH fields with an air-mass-weighted NH:SH ratio of 0.98. Figure 2e shows that the fixOH and optOH solution sets adhere closely to a linear relationship between global sources and sinks, in all cases with a ~20 Tg/y growth rate in the atmospheric methane burden.

While mutually inconsistent, the fixOH and optOH global methane sink terms are each physically tenable, falling respectively towards the high- and low-end estimates of the Saunois et al. (2020) top-down budget assessment for 2008–2017 (501–574 Tg/y). The OH fields dominating methane removal in these two scenarios are likewise physically viable based on available independent constraints. If we attribute the optOH methane loss correction entirely to OH, we arrive at global-mean [OH] = 8.27×10^5 molecules/cm^3, placing the fixOH (1.03×10^6 molecules/cm^3) and optOH solutions near the middle and lower end of the range indicated by prior assessments ($0.85–1.30 \times 10^6$ molecules/cm^3; (Krol and Lelieveld, 2003; Li et al., 2018; Montzka et al., 2011; Naik et al., 2013; Patra et al., 2021; Prinn et al., 2001; Prinn et al., 2005; Rigby et al., 2017; Zhao et al., 2019)). Our 2018–2019 analysis timeframe also spans an El Niño, which has been tied both to global OH decreases and to methane growth rate acceleration (Anderson et al., 2021; Turner et al., 2018a).

Additional ambiguity arises from the fact that the optOH methane sink adjustments could partly reflect uncertainty in the CH_4 + OH rate coefficient, which here follows Burkholder et al. (2020). While the rate at 298 K has been verified to within 1% across many lab experiments, uncertainties increase for higher and lower temperatures (up to 13% within 273–313 K (Burkholder et al., 2020)). Given all of the above considerations, we turn to independent measurements of methane, CO, and OH to assess the fixOH versus optOH solution fidelity.

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3.2 Independent model assessments to discriminate between conflicting methane budgets

Ground-based methane column (XCH_4) observations from the TCCON network (GGG2014 (2014)) show comparable improvement over the prior for both the fixOH and optOH solutions (and for their individual member inversions), with 71% and 66% mean bias reduction, respectively (Figure 3, Table S1). However, global in-situ measurements from ObsPack (near-real time version v2.0; (2021)) reveal a 93% absolute mean bias improvement for the fixOH framework compared to just 39% for optOH (Figure 3, Table S1). Figure 3 further shows that the optOH solutions overcorrect the prior negative bias with respect to ObsPack, providing a first piece of evidence for a methane sink underestimate in this inversion.

Remote observations of OH and CO (the primary OH sink) from the Atmospheric Tomography (ATom) airborne campaign (Wofsy et al., 2018) also point to an OH underestimate in the optOH solution. Figure S8 compares the ATom observations for these species with predictions from supplemental GEOS-Chem simulations (configured as in Gonzalez et al. (2021)) constrained to the fixOH and optOH oxidant fields. With the exception of ATom 3, the mean model OH biases with respect to ATom observations are ~80% lower for fixOH than for optOH, with the latter exhibiting a consistent OH underestimate. Biases in the simulated background CO levels are likewise lower (by 7–87%) in the fixOH simulations, with a clear CO overestimate for optOH (Figure S8). While the ATom timeframe (2016–2018) is distinct from that of the TROPOMI inversions (2018–2019), the model is sampled at the time of measurement for both comparisons. We therefore expect the OH and CO biases highlighted above to likewise manifest for 2018–2019—an expectation that is supported by the ObsPack comparison.

When co-optimizing methane emissions and loss we thus find that the solutions can achieve a good fit to the TROPOMI data themselves but degrade model agreement with other observations of methane, OH and CO. We conclude that solving for global methane sources and sinks based solely on satellite observations of methane itself remains an under-constrained problem—even with the dense TROPOMI data coverage. In the same way, previous studies using methane data to optimize OH alongside emissions are likely subject to strong error correlations between the derived sources and sinks (Lu et al., 2021; Maasakkers et al., 2019; Qu et al., 2021; Zhang et al., 2018; Zhang et al., 2021). On the other hand, the comparisons here provide robust support for the fixOH inversion solutions based on their fidelity against independent atmospheric observations.

We thus focus the remainder of this paper on the fixOH results, and treat the corresponding multi-model mean as our base-case solution. The fixOH constituent inversions (Scale Factor - SF, Background Increment - BI, Observational Guess - OG, and Emission Enhancement - EE) each employ an alternative framework for spatial emission correction, as described earlier. Figure S9 shows that these individual members converge closely (within 0.5%) in terms of the total derived methane flux, with a high degree of spatial similarity in their derived emissions. Differences in adjustment magnitudes emerge, with the

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OG and EE frameworks generally yielding the largest and smallest emission corrections, respectively. Overall, the derived grid-level emissions agree to within 30% for 42% of the emitting model grid cells, with consistent adjustment direction over areas encompassing 65% of the total optimized emissions. In what follows we focus discussion on these areas of consistency, and use the multi-model spread to draw insights into some of the key disparities.

4. Global Methane Sources and Top Emitting Countries

We infer from the TROPOMI observations an optimized global methane flux of 587 (586–589) Tg/y for 2018–2019, including 375 (367–387) Tg/y from anthropogenic sources, 197 (185–207) Tg/y from natural sources, and 15 (14–15) Tg/y from biomass burning. Values listed reflect the fixOH multi-model mean and range, with emissions partitioned to individual sources according to the prior grid-level sectoral fractions. Our derived natural source falls at the low end of the 2017 Global Methane Project estimates (GMP; 194–489 Tg/y), whereas we infer a larger source from agriculture and waste than does GMP (253–262 versus 198–246 Tg/y) (Jackson et al., 2020). The TROPOMI-derived methane emissions from fossil fuel and industry of 118 (112–127) Tg/y lie between the GMP top-down (91–121 Tg/y) and bottom-up (121–164 Tg/y) estimates for that sector. Jackson et al. (2020) attributed the global methane emission increase between 2000–2006 and 2017 mainly to anthropogenic sources, with similar contributions from agriculture/waste and fossil fuel. Here, we find that emissions from agriculture and waste are even larger than estimated by GMP.

Table 2 lists the top ten national contributors to global anthropogenic methane emissions (see Table S2 for natural and total emissions). Together, we find that these contributors account for 58% of the global anthropogenic flux, and they similarly represent 60% of the global population. However, within this group there are large differences in per capita emissions, which are 72–286% higher than the global average in Brazil, the US, Russia, and Iran, but 17% lower in China and 45% lower in India. Table 2 also includes three of the five countries with the largest natural methane emissions, largely from wetlands: Brazil (39 Tg/y), the US (16 Tg/y), and Russia (12 Tg/y). Together with the Republic of the Congo (22 Tg/y) and Canada (15 Tg/y) these countries account for 53% of natural methane sources globally. This places Brazil (59 Tg/y) and the US (43 Tg/y) as the second and third methane emitters in terms of total flux, after China (60 Tg/y, 94% anthropogenic; Table S2).

Comparing our results to recent year-2019 estimates derived from the GOSAT satellite sensor (Worden et al., 2021), which has effective sampling density two orders of magnitude lower than TROPOMI (Qu et al., 2021), we find that seven countries (China, India, US, Russia, Brazil, Pakistan, Indonesia) are identified in both cases as among the top ten anthropogenic emitters globally. Our inferred fluxes for China, Russia, and Iran are 11–28% higher than the GOSAT-based estimates, mainly reflecting oil, gas, and coal emissions, and 33% lower for Brazil, mainly due to livestock. Emissions for India, the US, the Europe Union, Pakistan, and Indonesia agree to within 10% for the TROPOMI- and GOSAT-based results. However, we find here that anthropogenic emissions from Bangladesh (7 Tg/y versus a prior of 4 Tg/y) are 3× higher than

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the GOSAT estimate (2 Tg/y), while adjacent emissions from Myanmar (4 Tg/y) are half the GOSAT estimate. Worden et al. (2021) conclude that the GOSAT-derived emissions for Myanmar are anonymously high due to impacts from their prior assumptions; we attribute much of that flux to Bangladesh and show later that it mainly arises during the South Asian monsoon.

5. Wetland Sources are Underestimated in the Tropics

The TROPOMI-derived wetland fluxes total 173 (155–182) Tg/y globally, representing 29 (26–31)% of the total methane source and 88 (84–91)% of the natural source. Figure S3 and S10 show that global wetland emissions are lowest during Oct–Feb (12–13 Tg/month) and highest in July (17 Tg/month) due to strong northern-hemisphere seasonality. We find through the inversions that global wetland fluxes are 24 Tg/y higher than the prior estimate, with the increase mainly originating in the tropics (82% within ±23.5° latitude). The tropics thus account for 70 (68–72)% of our optimized wetland emissions. Northern temperate wetlands contribute most of the remainder (46 Tg/y from 23.5°N–66.5°N) with a magnitude that is in-line with the prior bottom-up estimate (44 Tg/y).

Over Amazonia (box 5 in Figure 2b), we obtain wetland fluxes of 51 (44–54) Tg/y, 29% of the global wetland total. These fluxes are underestimated in the WetCHARTs prior inventory by 9 (2–11) Tg/y; the true disparity is likely larger than this since the inversions do not fully mitigate the prior regional XCH_4 bias (Figure 2c). Low observation density due to clouds (Figure S1) leads to some ambiguity in the spatial distribution of these derived fluxes: the SF and BI inversions allocate the upward corrections according to the prior spatial patterns over Amazonia, while the OG inversion identifies broader sources extending to northern Brazil (Figure 4 and S9). Upward corrections occur mainly during the wet season (Dec–Apr) and are temporally correlated with runoff (mean monthly R = 0.85 (ERA5, 2019)). This finding is consistent with the 2010–2018 flux increase (1 Tg/y^2) which has been inferred over Amazonia based on GOSAT and linked to increased flooding with strengthening Walker circulation (Barichivich et al., 2018; Zhang et al., 2021). However, eastern Brazil is also an agricultural frontier with forests transitioning to agricultural lands (Nepstad et al., 2019; Zhang et al., 2021), and our inversions point to a 9 (5–15) % underestimate of Amazonian livestock sources. Bottom-up calculations suggest a 33% increase in this source from 2010–2018 (EDGAR v6, 2021); if such trends are in fact underestimated then our prior-based partitioning would imply an even larger livestock contribution to the derived regional emission corrections.

The inversions also point to a substantial (26 [5–36]%) upward correction for central African wetlands (box 12 in Figure 2b) that is concentrated during Dec–May (Figure S11). The optimized regional emissions are then 33 (28–36) Tg/y, 19% of the global wetland total. However, while these adjustments effectively correct the prior model bias for this latitude band, there is a clear XCH_4 over-correction for the Democratic Republic of the Congo (DRC; Figure 2c). Here, the modeled XCH_4 enhancement over the background (Figure S12) is degraded from a prior model-observation mismatch of +3 ppb to +11 ppb.

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The low data density over central Africa and Amazonia (Figure S1) thus appears to cause some tropical flux mis-attribution—with the DRC overcorrection offset by under-corrections over Amazonia (as discussed above), Nigeria, and the nearby Sudd wetlands (Figure 2c). The latter region is examined further below.

Figure 2a shows that the South Sudd wetlands are a major methane hotspot that is underestimated in the prior model by a column average of 41 (21–65) ppb. Despite this, we derive a regional upward emission correction of just 13% (optimized flux: 1.3 [1.2–1.4] Tg/y for box 13 in Figure 2b). This yields a residual underestimate (Figure 2c) reflecting the aliasing discussed above and showing that the optimized South Sudd fluxes are still too low. Anomalous hydrology may contribute to these elevated Sudd fluxes: wetland extent for this area was ~10% higher in 2019 than the 2010–2019 mean (Jensen and Mcdonald, 2019), and ERA5 reanalysis (2019) points to elevated precipitation over central Africa during Sep–Nov (22% above the 2010–2019 mean). This interpretation is consistent with a previous study by Lunt et al. (2021) linking anomalous East Africa rainfall during the 2018 long rains (Mar–May) and 2019 short rains (Oct–Dec) with 10–40% methane emission increases over South Sudd. Over longer timescales, GOSAT analysis has pointed to a 3 Tg/y emission increase over the broader Sudd region caused by increased inflow from the White Nile and Sobat rivers (Lunt et al., 2019; Maasakkers et al., 2019; Parker et al., 2020).

North American wetlands in Alberta, Saskatchewan, and the US Upper Midwest are revised downward (by -1.6 [-0.7 – -2.6] Tg/y for box 1 in Figure 2b), with most of the adjustment occurring during early summer (-18% for Jun-Jul vs. -1% for Aug–Sep; Figure 4). We thus obtain optimized regional wetland emissions of 16 [15–17] Tg/y, 9% of the global flux from this source. Wetland emissions over this area are highly sensitive to soil temperature and surface water extent ($R > 0.7$ and $R > 0.4$, respectively; Figure 5). The stronger Jun–Jul adjustment derived here suggests that the post-thaw onset of northern wetland fluxes occurs too early in WetCHARTs, which uses surface skin rather than soil temperatures to predict emissions. Similar downward emission corrections have been inferred from GOSAT (Zhang et al., 2021), aircraft, and long-term eddy covariance measurements (Yu et al., 2021c).

Across the wetland regions examined above, Figure S13 shows that our optimized emissions fall towards the middle of the land-surface model estimates from the Global Carbon Project (GCP; (Saunois et al., 2020)). Among the GCP bottom-up estimates, we see in Figure S13 that simple model parameterizations can obtain comparable agreement with the TROPOMI-optimized emissions as more complete process-based models. For example, LPJ-WSL, a parsimonious model that predicts net emissions based on soil characteristics without explicitly representing oxidation, transport, or wetland plant types, achieves similar fidelity (in terms of bias and RMSE versus the optimized values) as JSBACH (Kleinen et al., 2020), which features more sophisticated treatment of soil carbon, roots, and plant-mediated processes. Many simple wetland models rely on reanalysis-based wetland extent estimates such as the Global Lakes and Wetlands Database (GLWD) (Lehner and Döll,

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2004); within the WetCHARTs ensemble we find here that GLWD-based flux predictions overestimate emissions in northern North America while underestimating those in central Africa (Figure S14).

Figure 5c explores the environmental sensitivities of the TROPOMI-derived wetland sources to gauge how future rainfall or temperature changes may alter emission magnitudes. We see that the optimized tropical and subtropical wetland methane emissions exhibit only modest sensitivity to soil temperature (0–7 cm (ERA5, 2019)), but have strong ($R > 0.7$) correlation with surface water extent (SWAMPS (Jensen and Mcdonald, 2019)) for key areas of India, Bangladesh, Brazil, Bolivia, Mexico, and Africa. Conversely, northern temperate wetland emissions in the US Upper Midwest and southeastern China respond more directly to temperature changes, while those in Canada and Russia show strong sensitivity to both hydrology and temperature. Projected precipitation increases for mid-to-high latitudes and decreases for the sub-tropics (IPCC, 2021) may thus increase the importance of temperate and boreal wetland fluxes in coming years.

6. Anthropogenic sources and emission hotspots

We derive global anthropogenic methane sources of 375 Tg/y, with 132 (127–136) Tg/y from livestock, 98 (93–104) Tg/y from fossil fuel, 83 (79–87) Tg/y from waste, 42 (40–45) Tg/y from rice, and 20 (19–23) from other sources. This represents a 19 Tg/y increase over the prior flux that on a global basis is mainly driven by upward corrections for livestock (+11%) and rice (+15%). Since our prior anthropogenic emissions are based on inventories for 2010–2016, these flux corrections could reflect inventory errors, temporal changes between 2010 and 2019, or some combination of the two. Below, we employ the spatially-downscaled TROPOMI-derived emissions to elucidate key anthropogenic sources and to identify missing and underreported flux hotspots.

6.1 The Middle East and North Africa: Missing and underestimated emission hotspots from fossil fuel activities

The largest fossil fuel emission corrections occur over the Middle East, where total fluxes increase throughout the year by 48 (34–60)% over the prior (+12 [9–16] Tg/y, box 9 in Figure 2b). These upward corrections successfully reduce the mean regional model bias from -29 to 0 ppb and are attributed to a combination of fossil fuel (41%), livestock (23%) and waste (20%). The Middle East possesses approximately ~50% of global oil reserves and ~40% of global natural gas reserves, with increasing production over the past three decades (BP, 2021; Schneising et al., 2020; UNFCCC, 2021). Bottom-up information (EDGAR v6, 2021) accordingly points to a significant increase in methane hotspots for this area over the past decade (Figure 5a) and to an overall 26% regional emission increase from 2010 to 2018. The 1.8× larger adjustment revealed here by the TROPOMI observations points to both temporal increases and inventory underestimates for this area.

Middle East methane emissions are highly localized, with just 5% of model grid cells (at 0.1°×0.1°) accounting for 70% of the prior regional emissions and 60% of the derived adjustments. Our multi-model inversion results point to consistent prior

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underestimates for high emitters in Azerbaijan, Turkmenistan and Iran, supporting previous satellite-based analyses (Buchwitz et al., 2017; Lauvaux et al., 2022; Schneising et al., 2020; Varon et al., 2019). However, TROPOMI also reveals a large number of hotspots (over Oman, Yemen, Saudi Arabia, Iraq, Turkmenistan, and Iran; Figures 2a, 5b, and S15) that are entirely missing from the prior inventory. Many of these missing emission hotspots are consistent with facilities on the ground. For example, missing sources in Oman identified through the OG inversion (Figure S9) match the location of the Khazzan gas field—one of the Middle East's largest natural gas fields producing ~1–1.5 billion cubic feet/day of natural gas and ~35,000 barrels/day of light oil (NS Energy, 2022). Other detected hotspots correspond to the Masila Basin in Yemen (EIA, 2022), oil fields in Saudi Arabia (e.g, the Ghawar Field, (Maps Saudi Arabia, 2022)) , and super-emitters in Iraq (Lauvaux et al., 2022).

Over northern Africa, TROPOMI identifies large XCH_4 enhancements extending from the Libyan coast to Algeria (box 8 in Figure 2b). These sources are not well-represented in the prior inventory but correspond to oil fields in Libya and to part of the Greenstream pipeline system. The OG inversion is partially able to identify these sources, but the attribution is limited to a single 2°×2.5° grid cell (Figure S9).

6.2 Western Russia and North China Plain: An overestimate of fossil fuel emissions

We find from the TROPOMI data that emissions are overestimated in northern Asia, mainly reflecting fossil fuel sources. Specifically, fossil fuel methane emissions are overestimated by 27 (11–40)% over western Russia (box 6 in Figure 2b) and by 17 (1–29)% over the North China Plain (box 7), with these downward corrections reducing the regional model bias to <6 ppb. On the other hand, bottom-up information for 2010–2018 points to temporal increases for both areas (+36% over western Russia, +9% over the North China Plain; Figure 5a). The UNFCCC emissions used as prior account for accidental and intentional methane releases not considered in previous inventories (Scarpelli et al., 2020), leading to a >2-fold increase over western Russia compared to EDGAR v5. Our results indicate that these emission pathways may be overestimated in UNFCCC, and that the actual fossil fuel methane source from Russia for 2018-2019 lay between the UNFCCC and EDGAR v5 values.

Methane emissions in the North China Plain are primarily from coal mines; prior work suggests that these facilities have lower emission factors than in South China and that their emissions have been declining since 2012 (Sheng et al., 2019). Satellite-based and in-situ measurements have pointed to EDGAR v4.3.2 and GFEI emission overestimates for this area (Alexe et al., 2015; Lu et al., 2021; Maasakkers et al., 2019; Monteil et al., 2013; Qu et al., 2021; Turner et al., 2015; Zhang et al., 2021). Here, we use a detailed new inventory for China coal emissions (Sheng et al., 2019) that has >50% lower fluxes than EDGAR v4.3.2 over the North China plain—but find that these are still overestimated. By contrast, upward adjustments are derived over other regions in China such as Xinjiang, where TROPOMI XCH_4 enhancements are attributed by the OG and EE inversions to underestimated fossil fuel sources (Figure S9). Recent work by Lorente et al. (2022) points to some

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erroneous emission hotspots for this area associated with surface reflectance; we therefore restrict our interpretation here to the regional scale. Positive corrections are also derived for fossil fuel sources in South China, but their emissions are difficult to isolate from those of nearby rice fields.

6.3 South and Southeast Asia: Major methane emissions during summer monsoon

Methane emissions from South and Southeast Asia are dominated by agriculture (livestock, rice) and waste. The largest emission correction for this area occurs over India, where we find a 23 (10–30)% underestimate (mainly due to those two sources) and derive total national emissions of 61 [55–64] Tg/y. India contains over 35% and 20% of the world's cattle and water buffalo, respectively, with both populations increasing over the past decade (Sonavale et al., 2020). Such changes are reflected in both the EDGAR bottom-up inventory (+0.4 Tg/y annual livestock+waste emission increase for 2010–2018 (EDGAR v6, 2021)) and in analyses based on GOSAT observations (+0.2–0.7 Tg/y inferred annual increase for Indian livestock (Maasakkers et al., 2019; Miller et al., 2019; Zhang et al., 2021)). Our prior estimate employs EDGAR v5 for year-2015; the inferred +23% (11 Tg/y) correction is too large to be explained solely by such trends and thus indicates an emission underestimate for this source.

Over 90% of the world's rice production occurs in India and Southeast Asia, and we find that the associated methane emissions are underestimated by 39 (7–53)% and 17 (7–28)% respectively (boxes 10 and 11 in Figure 2b). Bottom-up statistics from FAOSTAT (2021) indicate an 8% increase in crop production over these areas for 2010–2019 due to a 10% yield increase combined with a 2% cropland area decrease. Such trends cannot fully explain the TROPOMI observations, which therefore indicate that the EDGAR v5 prior emissions for this source are too low. The emission corrections mainly occur during the Jul–Oct rice growing season that coincides with the summer monsoon. The TROPOMI data reveal several other connections between the East Asian summer monsoon and the regional methane budget, which we explore next based on our monthly downscaled top-down emissions.

Approximately 80% of India's annual rain falls during the summer monsoon (IPCC, 2021), and across this Jul-Oct season we find that methane emissions from the India and Southeast Asia boxes in Figure 2b are underestimated by 37 (15–45)%. The resulting seasonal flux increase then accounts for over 68% of the total annual emission correction. While clouds reduce the TROPOMI sampling coverage during this time, we still obtain >370,000 and >9,000 observations per monsoon season over India and Southeast Asia, respectively, after applying the data quality filters described in Section 2.1. The above emission corrections strongly reduce the prior model biases (from -28 to 3 ppb over India and from -41 to -7 ppb over Southeast Asia), with the individual inversions pointing to consistent spatial adjustments. Prior work using GOSAT and in-situ measurements has also identified a peak in Indian emissions during Jul–Oct that was not well-captured by bottom-up predictions (Palmer et al., 2021).

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Figure 6 shows that the TROPOMI-derived South Asian emission corrections are spatially and temporally coherent with the summer monsoon onset and withdrawal. Strong upward adjustments are first derived over Bangladesh and East India following monsoon arrival over these areas (early June 2018; late June 2019). As the monsoon advances, these emission corrections then extend more broadly over northern India during Jul–Sep. In concert with the monsoon, the upward corrections subsequently withdraw south and east to Bangladesh in October. Only minor emission adjustments are derived for this region outside of the summer monsoon.

The above patterns likely reflect hydrologic influences on methane emissions from biotic sources such as wetlands, rice fields, manure, landfills, and sewers. The derived emission corrections exhibit a strong temporal correlation with runoff (R = 0.82 in eastern India; R = 0.62 in western India), supporting the underlying role of hydrology. Baker et al. (2012) similarly concluded on the basis of aircraft measurements that 64% of Indian methane emissions during this season reflect monsoon-driven biogenic sources. Increases in Indian summer monsoon precipitation that are projected over the 21th century (Katzenberger et al., 2021) thus raise the strong possibility of enhanced regional methane emissions in the years to come.

6.4 North and South America: Increases for fossil fuel sources

We turn next to key oil and gas production fields in North and South America. This includes the US Permian, Barnett and Eagle Ford region (box 2 in Figure 5b), previously shown to be the largest US oil and gas-related methane source (Zhang et al., 2020). Here we infer methane fossil fuel emissions for 2018–2019 that are 2% lower than the prior GEPA estimate for year-2016. This is in-line with bottom-up information from EDGAR v6, which finds no significant trend over this area for 2016–2018. Over northern Venezuela (box 4 in Figure 5b), we derive a 28 (2–41)% upward correction for fossil fuel exploration activities that is consistent across the year, in agreement with the +32% bottom-up increase that is estimated to have occurred between the prior inventory and inversion timeframes (2016–2018; (EDGAR v6, 2021)).

Extensive TROPOMI XCH_4 enhancements are seen over southern Mexico (Figure 2b, box 3), and our inversions reveal a 15 (9–25)% emission underestimate for this area (optimized flux 10 [9–10] Tg/y). Trend information from EDGAR v6 (2021) suggests a regional 25% emission increase for 2016–2020, which could in theory fully explain the derived upward adjustments. However, the observed hotspot locations are largely missing in the prior inventory, and as a result the SF inversion falsely attributes the corrections to upwind waste/landfill sources in Sinaloa. The OG inversion solution aligns more closely with the actual locations of these sources based on the oversampled TROPOMI data (Figure S9), and is supported by previous regional-scale TROPOMI inversions (Shen et al., 2021) that point to a >2× emission underestimate of onshore/offshore oil and gas production in the UNFCCC inventory used here. Aircraft measurements in 2018 also revealed substantial (29,000 kg/h) methane emissions in the same general region (Zavala-Araiza et al., 2021).

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7. Conclusions

A suite of two-year 4D-Var inversions using satellite-based data from TROPOMI places new constraints on global methane sources. We obtain in this way optimized global emissions of 587 (586–589) Tg/y for 2018–2019. Compared to the most recent GCP estimates (Jackson et al., 2020), our 2018–2019 results point to a larger role for anthropogenic sources, mainly tied to agriculture and waste. We further develop a framework to map the derived monthly emissions to 0.1° × 0.1° resolution, enabling the identification of key missing and underestimated sources as highlighted below.

We derive a +24 Tg/y increase in wetland emissions over the prior estimate of 149 Tg/y that mainly (82%) occurs over the tropics and appears to be related to positive hydrologic anomalies in Amazonia and the Sudd. Meanwhile, fossil fuel emissions in the Middle East (which have been increasing strongly over the past decade) are underestimated by 47 (23–57)% and reached 15.7 (13.2–16.8) Tg/y during our analysis period. Our inversions further uncover missing emission hotspots over Turkmenistan, Iran, Oman, Yeman, Iraq, Libya, Algeria, and Mexico. We estimate long-standing fossil fuel sources in Venezuela at 4.8 (3.8–5.3) Tg/y, 28 (2–41)% higher than the prior estimate and in-line with trend estimates for the intervening time frame.

Inversions point to underestimated agricultural sources in India (where there has been an increasing trend over the past decade), the Amazon Basin (where forest is transitioning to agriculture), central Africa, the US California Central Valley, and Asia. However, more than 45% (8.5 [3–11] Tg) of the global anthropogenic source adjustment derived here occurs over India and southeast Asia during the summer monsoon (Jul–Oct). We postulate that this reflects the influence of monsoon rainfall on methane emissions from rice, manure, waste, and landfills. Given the projected increase in monsoon precipitation over the coming century (IPCC, 2021), better understanding of these effects is crucially needed.

Finally, our analyses show that even the dense TROPOMI data coverage does not fully resolve variability in methane sources from that in its sinks. We address the issue in this work by employing validated OH fields from a chemical transport model, but future methane inversions can benefit from incorporating additional datasets (e.g., CO, methyl chloroform, formaldehyde) as constraints on the methane sink. Quantitative evaluation of the influence of the 2018–2019 El Niño and the 2021 Hunga Tonga–Hunga Ha‘apai eruption on OH variability will also help to advance the accuracy of contemporary methane budget estimates.

Code and data availability

TROPOMI data is publicly available online at http://www.tropomi.eu/data-products/level-2-products. TCCON data was obtained from the TCCON Data Archive hosted by CaltechDATA at https://tccondata.org. The GEOS-Chem adjoint code is

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available at http://adjoint.colorado.edu:8080/gcadj_std.git. The modified code used here is archived at https://doi.org/10.13020/g5xc-nj81 (Yu et al., 2021b).

Competing interests

The authors declare that they have no conflict of interest.

Author contributions

XY performed the 4D-Var inversions with help from DBM and DKH. XY, DBM, and DKH designed this study. All authors contributed to the analyses. XY and DBM wrote the manuscript, with comments from all authors.

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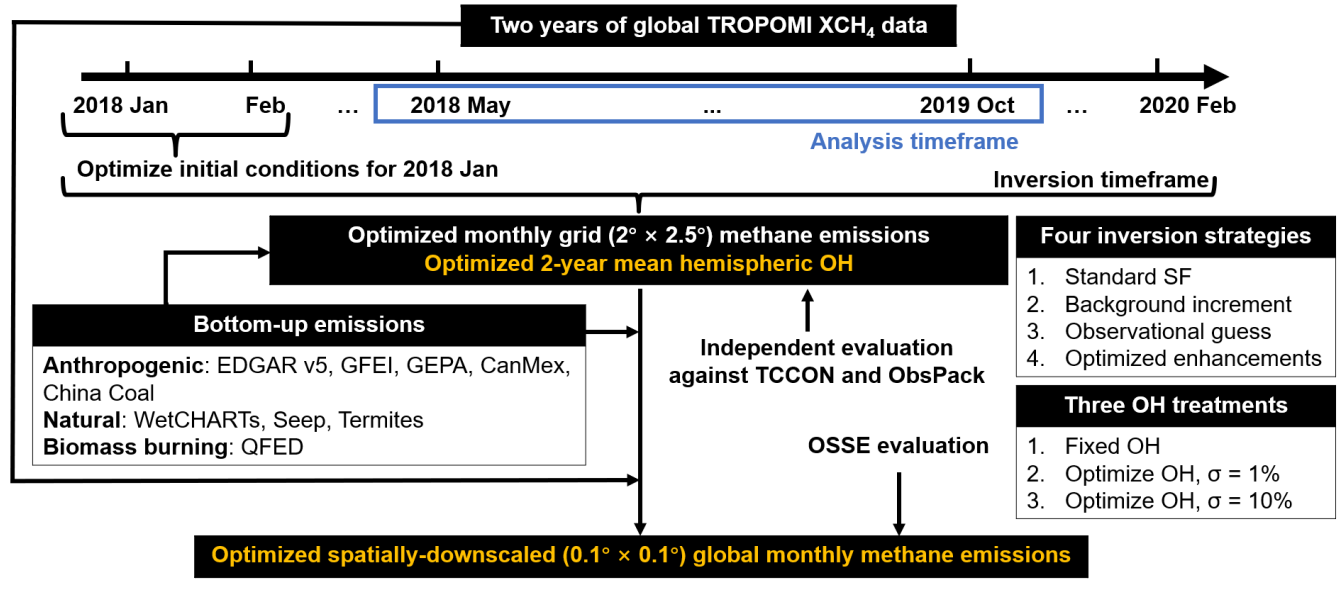

Figure 1. Flow chart showing the TROPOMI methane inversion methodology.

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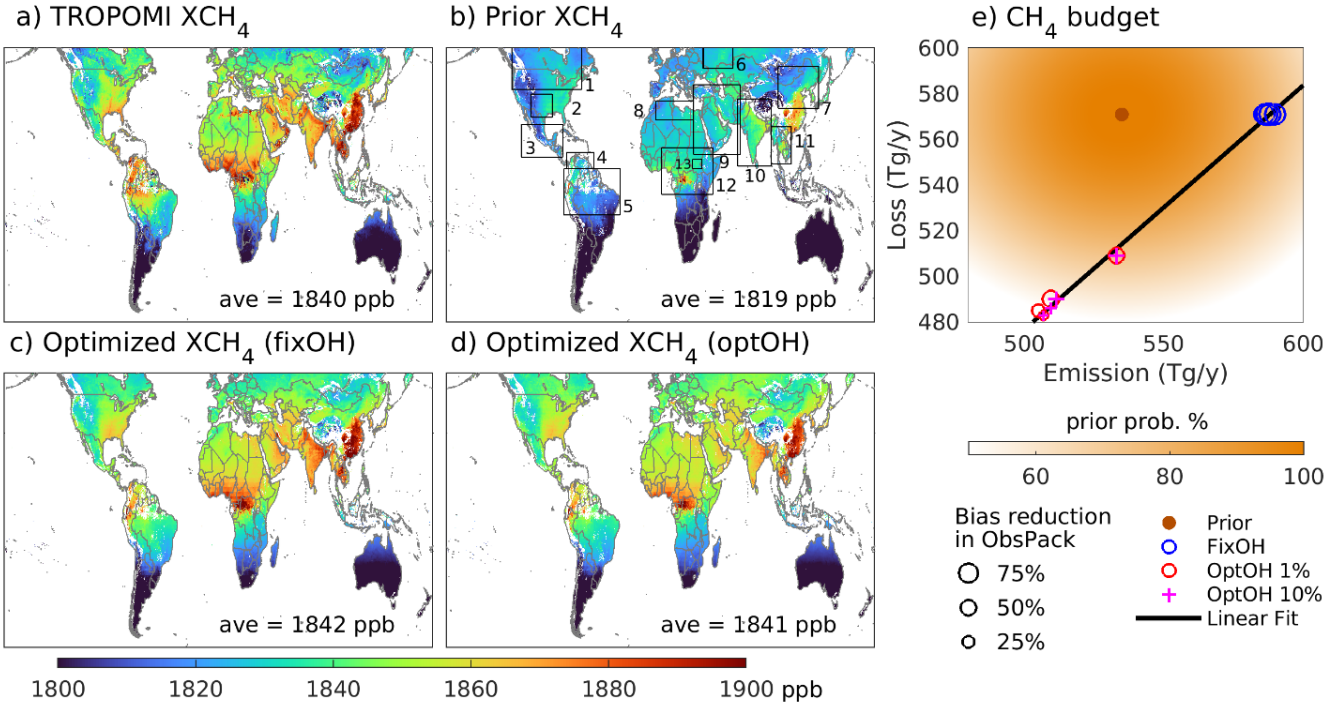

Figure 2. Observed and simulated methane distributions for 03/2018–02/2020. a) TROPOMI methane column (XCH_4) observations oversampled to 0.1° × 0.1°. b) Methane column distribution predicted by the prior model sampled according to the TROPOMI observation operator with numbered regions described in-text. c) Same as panel b but for the fixOH optimized ensemble mean. d) Same as panel b but for the optOH optimized ensemble mean. e) Prior and optimized methane budgets. Prior probabilities (orange) equate one standard deviation to 25% of the bottom-up range from Saunois et al. (2020), following Hozo et al. (2005). Black line shows a linear fit to the solution, emission = 0.93 × loss + 55.26 (units: Tg/y). Symbol sizes indicate the mean bias reduction against ObsPack independent measurements. Annual values are for 11/2018–04/2019 plus the average of 05–10/2018 and 05–10/2019.

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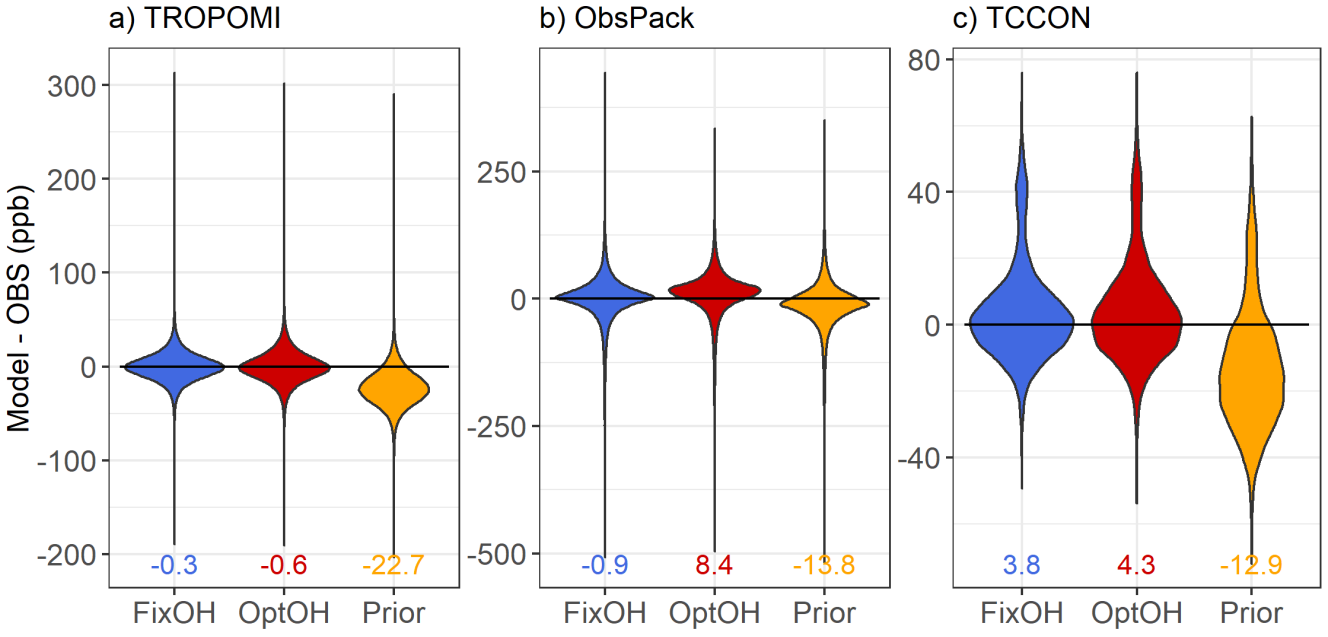

925 **Figure 3. Evaluation of the prior and optimized methane simulations (05/2018–10/2019) against a) TROPOMI XCH_4 observations, b) ObsPack in-situ methane measurements, and c) TCCON ground-based methane column observations. Numbers inset indicate the mean model-observation bias (unit: ppb).**

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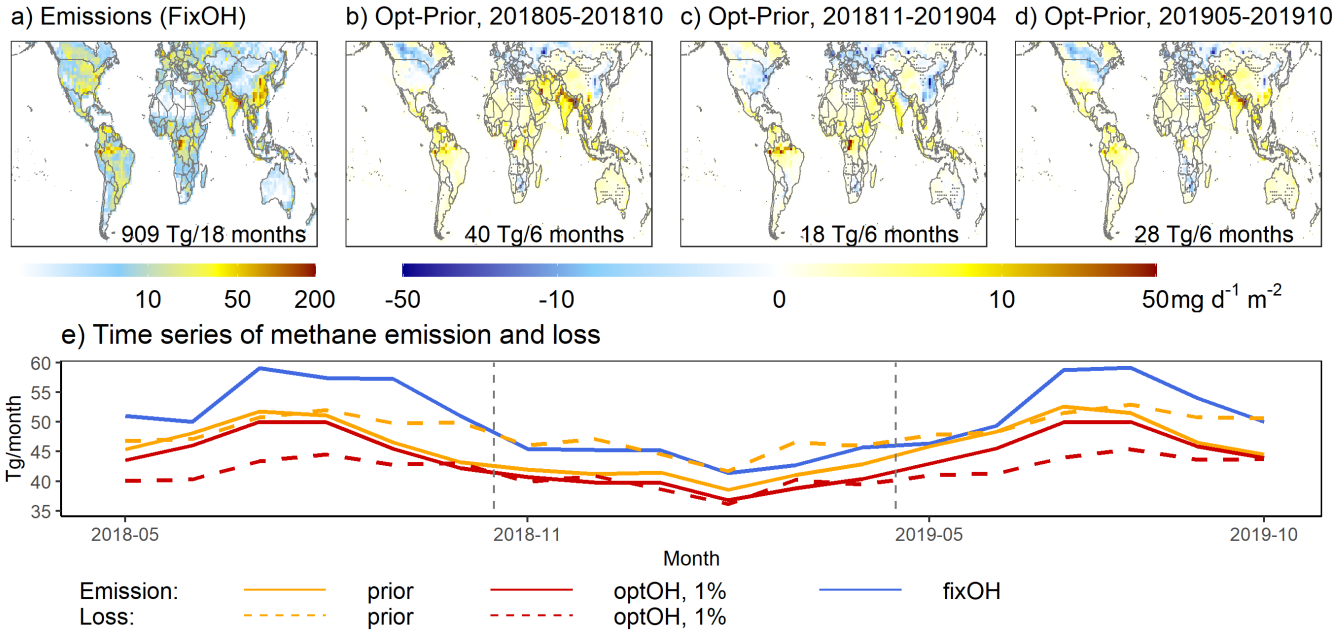

Figure 4. (a) Optimized global methane emissions based on the fixOH inversion ensemble. (b–d) Seasonal fixOH emission corrections. Dots indicate missing sources where the EE and BI inversions point to positive corrections that the SF inversion misses. e) Time series of prior and optimized methane sources and sinks.

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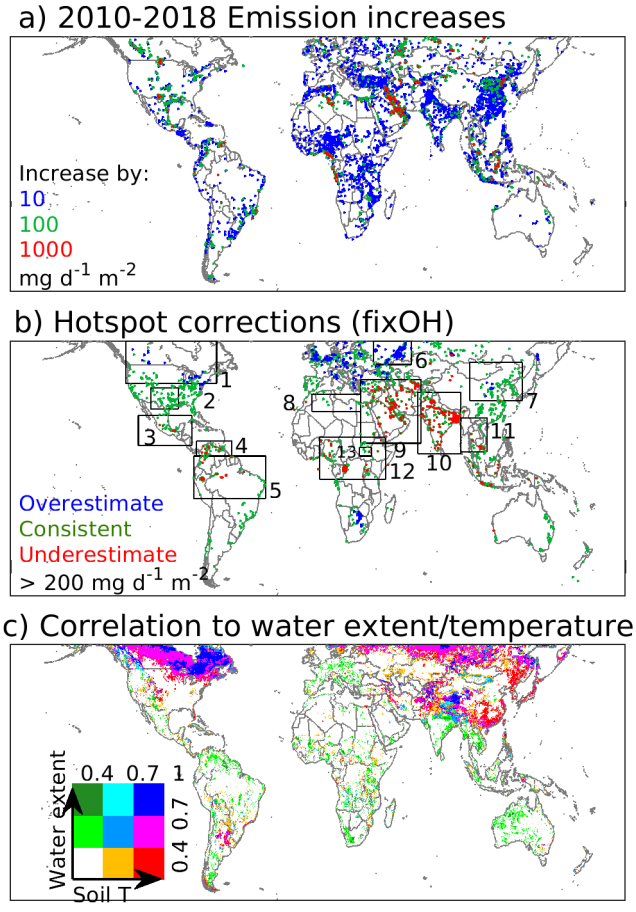

Figure 5. a) Methane emission increases during 2010–2018 estimated from bottom-up information (EDGAR v6, 2022). b) Methane source hotspots (>200 kg d^{-1} km^{-2}) and their TROPOMI-derived emission corrections (blue: >30% overestimate; green: accurate to ±30%; red: >30% underestimate). c) Correlations between the optimized wetland emissions, soil temperature (0-7 cm; ERA5 2019), and surface water extent (Jensen et al., 2019).

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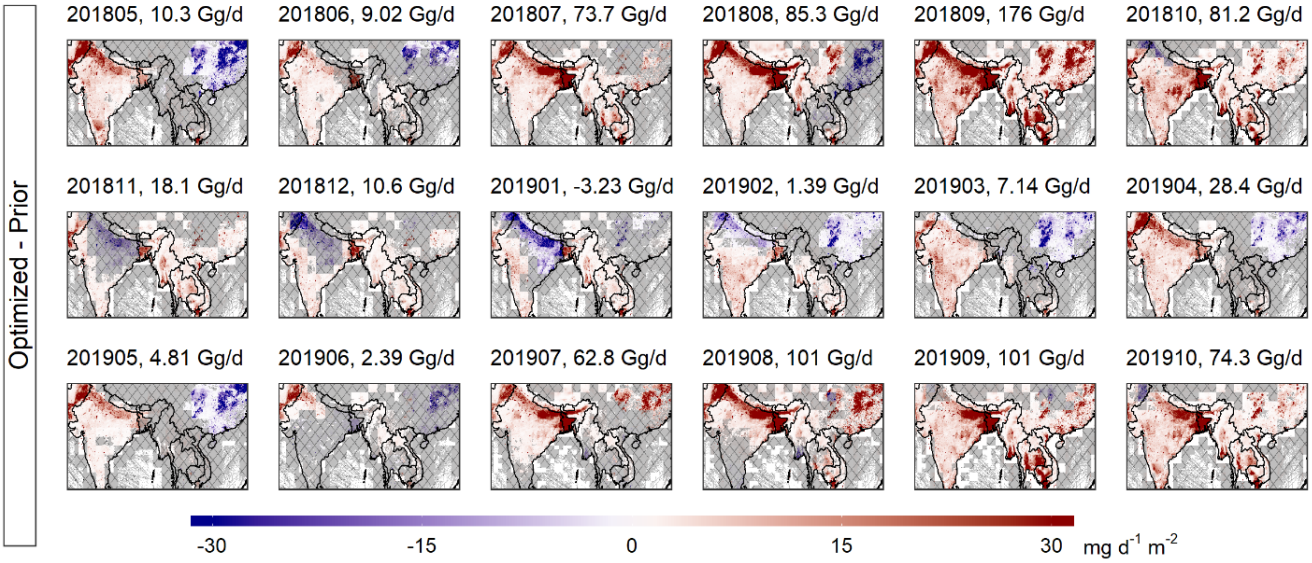

940 **Figure 6. TROPOMI-derived emission corrections over South and East Asia based on the fixOH ensemble mean. Shading indicates areas where corrections are not distinguishable from zero (i.e., inversion ensemble includes both positive and negative adjustments).**

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Table 1. Downscaling performance evaluation via OSSE[1]

	Downscaled solution	Fine-grid prior	Fine-grid adjoint	Coarse-grid prior	Coarse-grid adjoint
Without transport error					
Domain-wide bias reduction	68%		78%		68%
RMSE[2]	15790	13830	11489	13236	12976
R	0.67	0.57	0.74	0.45	0.50
Slope of least squares fit	0.97	0.61	0.84	0.28	0.42
With transport error					
Domain-wide bias reduction	98%		16%		98%
RMSE[2]	15461	13830	19292	13236	12917
R	0.70	0.57	0.60	0.45	0.51
Slope of least squares fit	1.04	0.61	1.01	0.28	0.45

[1]·Observing system simulation experiments performed for one month over North America

[2]·Root-mean-square-error, units: kg d^{-1} box^{-1}

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Table 2. Top 10 contributors to global anthropogenic methane emissions[1]

	Total anthropogenic emissions (Tg/y)	Change from prior (%)	Per capita anthropogenic emissions (kg/y/person)	Sector emissions (Tg/y)			
				Wetland	Agriculture & waste	Fossil fuel	Other
China	57 (52–61)	-4	40	2 (2–2)	34 (32–36)	18 (16–21)	6 (6–6)
India	36 (34–38)	16	26	2 (2–3)	32 (30–33)	2 (2–2)	3 (3–3)
US	27 (26–28)	-5	82	15 (14–15)	16 (15–17)	11 (10–11)	2 (1–2)
Russian	26 (22–29)	-21	184	10 (8–12)	5 (5–6)	21 (17–24)	2 (2–4)
Brazil	20 (20–21)	6	102	35 (31–37)	20 (19–20)	< 0.2	4 (4–4)
European Union	17 (15–17)	-9	38	2 (1–2)	14 (12–14)	2 (2–2)	2 (2–2)
Pakistan	10 (9–10)	28	44	< 0.1	9 (8–9)	1 (1–1)	1 (1–1)
Indonesia	10 (9–10)	15	37	9 (8–10)	8 (8–8)	1 (1–1)	2 (2–2)
Iran	9 (7–9)	70	103	< 0.2	2 (2–3)	6 (4–7)	1 (0–1)
Bangladesh	7 (5–8)	60	43	1 (1–1)	7 (4–8)	< 0.1	<0.4
Others	157 (151–168)	13	50	97 (87–103)	111 (108–118)	37 (36–38)	37 (30–46)

[1]Based on the fixOH inversion ensemble mean and range.