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Abstract

Methane (CH₄), the second most important anthropogenic greenhouse gas, significantly impacts global 19 warming. As the world's largest anthropogenic CH₄ emitter, China faces challenges in accurately 20 21 estimating its emissions. Top-down methods often suffer from coarse resolution, limited data constraints, and result discrepancies. Here, we developed the Regional Methane Assimilation System 22 (RegGCAS-CH₄) based on the WRF-CMAQ model and the EnKF algorithm. By assimilating 23 extensive TROPOMI column-averaged dry CH₄ mixing ratio (XCH₄) retrievals, we conducted high-24 25 resolution nested inversions to quantify daily CH₄ emissions across China and Shanxi Province in 26 2022. Nationally, posterior CH₄ emissions were 45.1 TgCH₄·yr⁻¹, 36.5% lower than the EDGAR estimates, with the largest reductions in the coal and waste sectors. In North China, emissions 27 28 decreased most significantly, mainly attributed to the coal and enteric fermentation sectors. Posterior emissions in coal-reliant Shanxi Province decreased by 46.3%. Sporadic emission increases were 29 30 detected in major coal-producing cities but were missed by the coarse-resolution inversion. Monthly emissions exhibited a winter-low, summer-high pattern, with the rice cultivation and waste sectors 31 showing higher seasonal increases than those in EDGAR. The inversion significantly improved XCH₄ 32 33 and surface CH₄ concentration simulations, reducing emission uncertainty. Compared to other bottom-34 up/top-down estimates, our results were the lowest, primarily because the high-resolution inversion 35 better captured local emission hotspots. Sensitivity tests underscored the importance of nested inversions in reducing the influence of boundary condition uncertainties on emission estimates. This 36 37 study provides robust CH4 emission estimates for China, crucial for understanding the CH4 budget and 38 informing climate mitigation strategies.

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Keywords CH₄ emissions, Emission inversion, Methane assimilation system, China, Data assimilation

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1. Introduction

Methane (CH₄) ranks as the second most significant anthropogenic greenhouse gas and plays a crucial 46 role in global warming. Its global warming potential is 28–29.8 times that of carbon dioxide (CO₂) 47 over a 100-year time scale and contributes approximately 17% to the total radiative forcing of 48 greenhouse gases since the industrial era (Forster et al., 2021; Saunois et al., 2025). With a relatively 49 short steady state atmospheric budget lifetime of slightly over 9 years (Prather, 2007), CH₄ is a key 50 target for rapid climate change mitigation efforts (Tu et al., 2024), as emphasized by the Global 51 52 Methane Pledge, which aims to reduce global anthropogenic CH₄ emissions by 30% below 2020 levels by 2030 (GMP, 2023). China, as the world's largest CH₄ emitter, accounting for around 14%-22% of 53 global anthropogenic CH₄ emissions, faces a complex challenge (Zhang et al., 2022; Janssens-54 55 Maenhout et al., 2019). China's CH₄ emissions originate from diverse sources, with coal mining and rice cultivation being particularly prominent (Nisbet, 2023; Lin et al., 2021). Comprehending the 56 spatiotemporal distribution and trends of China's CH4 emissions is of utmost importance for 57 formulating effective climate policies and fulfilling international climate commitments. 58 The bottom-up approaches, which rely on activity data and emission factors, have been widely used to 59 estimate China's CH4 emissions. However, accurately estimating these emissions remains a significant 60 61 challenge. Existing inventories are often characterized by large uncertainties in both magnitude and sectoral attribution, with differences between various bottom-up estimates reaching up to 40–60% for 62 China (Saunois et al., 2025). Among the various sources, estimates of China's coal mine CH₄ emissions 63 can range from 14–28 Tg·yr⁻¹ (Sheng et al., 2019). This wide disparity leads to much uncertainty in 64 the bottom-up estimates. There are multiple reasons for the uncertainties, especially the lack of 65 66 comprehensive data on emission sources, especially for small-scale and sporadic emitters, and the use of complex and perhaps inappropriate emission factors in different sectors (Stavert et al., 2022). For 67 instance, in coal mining, the emission factors vary significantly depending on the mining method 68 (underground vs. surface mining), geological conditions, and the quality of coal. More generally, due 69 to the use of a spatial proxy approach for the spatial allocation of the total emissions, existing global 70 inventories exhibit significant spatial discrepancies when compared with high-resolution local 71 measurements, resulting in misattribution of emissions (Qin et al., 2024). 72





73 In contrast, top-down inversion methods, that utilize satellite and surface observations in conjunction with atmospheric transport models, have the potential to provide more accurate and comprehensive 74 estimates. Compared with ground-based inversions, satellite-based atmospheric inversions, such as 75 76 those using column-averaged dry CH₄ mixing ratios (XCH₄) data from the Greenhouse Gases Observing Satellite (GOSAT) or the TROPOspheric Monitoring Instrument (TROPOMI), can offer 77 valuable insights into the spatial distribution of emissions (Lu et al., 2023). Studies utilizing satellite 78 79 observations have quantified CH₄ emissions from various sources, including oil, gas, and coal mining sectors at the global, regional, and key source scales (Nesser et al., 2024; Bai et al., 2024; Zhang et al., 80 2021). For instance, Maasakkers et al. (2019) investigated the contribution of different regions to the 81 global CH₄ budget with GOSAT XCH₄ data, identifying areas with significant emissions and sinks. 82 Pandey et al. (2019) used TROPOMI XCH₄ observations to reveal extreme CH₄ leakage from a natural 83 gas well blowout, demonstrating the instrument's ability to detect both large-scale and short-term 84 85 emission events. 86 The Methane Emission Control Action Plan released by China explicitly states the exploration and implementation of research on atmospheric methane emission inversion models, and strengthens the 87 verification of emission inventories by inversion data (MECAP, 2023). A number of inversions 88 relevant to China have been conducted with satellite observations. Using inverse analysis of 2019 89 TROPOMI XCH₄ data, Chen et al. (2022) quantified CH₄ emissions across China and attributed 90 contributions to specific sectors. Zhang et al. (2022) estimated China's CH₄ emissions from 2010 to 91 2017 by combining satellite and surface observations, revealing complex linkages between emission 92 trends and associated policy drivers. However, the trends of China's CH₄ emissions quantified by the 93 top-down approach (Sheng et al., 2021; Miller et al., 2019) are contrary to those estimated by the 94 95 bottom-up approach, which is mainly due to the unclear quantification of emissions from the coal mining sector (Sheng et al., 2019; Liu et al., 2021). 96 Currently, global-scale CH₄ assimilation systems are widely applied, such as CarbonTracker-CH₄ in 97 98 the United States (Bruhwiler et al., 2014), CAMS in Europe (Agustí-Panareda et al., 2023), NTFVAR in Japan (Wang et al., 2019), LMDz-SACS-CIF in France (Thanwerdas et al., 2022), and GONGGA-99 100 CH₄ in China (Zhao et al., 2024). However, significant knowledge gaps remain in accurately estimating 101 CH₄ emissions at the regional scale. There are relatively few existing regional CH₄ assimilation





102 systems, such as the ICON-ART-CTDAS (Steiner et al., 2024) and CarbonTracker Europe-CH₄ (Tsuruta et al., 2017) in Europe, and the IMI in the United States (Varon et al., 2022). Additionally, 103 existing regional inversions often use global atmospheric transport models with relatively coarse 104 105 resolutions on the annual or monthly average scale, lacking high-resolution spatial and temporal coverage to capture local characteristics and short-term variations in emissions (Chen et al., 2022). Qu 106 et al. (2021) highlighted significant challenges in separating rice and coal emissions over southeast 107 China due to coarse grid resolution. High-resolution estimates are crucial for understanding the 108 detailed distribution of emissions, especially in regions with heterogeneous source landscapes. For 109 110 instance, in Shanxi Province, a major coal producing region, the traditional low-resolution models may fail to capture the emissions from numerous small-scale coal mines. Additionally, the inversion results 111 of global and regional systems still show significant differences, mainly due to different observation 112 data, inversion methods, transport models, and resolutions (Kou et al., 2025; Chen et al., 2022; Liang 113 et al., 2023). Another challenge is that regional models need to consider the impact of boundary fields, 114 115 especially for long-lived species. Current studies usually directly derive regional boundary fields from 116 global simulation or analysis fields, which still contain large errors (Zhang et al., 2022;Kou et al., 117 2025). Consequently, China's contribution to the global CH₄ budget remains unclear. In this study, we aimed to address these limitations by developing a Regional Methane Assimilation 118 System (RegGCAS-CH₄) based on the Weather Research and Forecasting-Community Multiscale Air 119 Quality (WRF-CMAQ) model and the Ensemble Kalman Filter (EnKF) algorithm (Evensen, 1994). 120 This system enabled the assimilation of a large volume of TROPOMI XCH4 data to achieve nested 121 inversions of daily CH4 emissions with high spatial-temporal resolution. We conducted a 122 comprehensive analysis of the spatial characteristics and monthly variations of China's CH4 emissions 123 124 in 2022. In particular, we focused on the nested inversion analysis of CH₄ emissions in Shanxi Province, China. 125 The novelty of our study lies in the high spatial resolution emission inversions, which allows for 126 127 capturing the fine-scale features of CH₄ emissions that are often missed by global inversion models. Secondly, by assimilating a larger amount of TROPOMI XCH₄ observations, which feature high 128 129 spatial resolution, wide spatial coverage, and high-frequency retrievals (~100 times more 130 observations than GOSAT) (Qu et al., 2021), we can incorporate the latest and more representative



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information on atmospheric CH₄ concentrations to update the daily emissions. This high temporal resolution is essential for understanding the short-term fluctuations in emissions and their response to 132 various factors. Notably, we have pre-updated the global boundary fields, which effectively mitigates 133 134 the impact of boundary condition errors on regional emission inversions. This is a crucial step that has not been emphasized or implemented in most previous studies. By comparing our results with the 135 currently widely used inventories, we aim to provide a more accurate and detailed understanding of 136 China's CH₄ emissions, which is crucial for formulating effective climate change mitigation strategies.

2. Method and data

2.1 Data assimilation system

The RegGCAS-CH₄ system was extended from RegGCAS (Zhang et al., 2024), which was developed by Feng et al. (2023) based on the Regional multi-Air Pollutant Assimilation System (RAPAS) (Feng et al., 2020) and Global Carbon Assimilation System (GCAS), version 2 (Jiang et al., 2021). RegGCAS-CH4 includes a regional chemical transport model (CTM) and an ensemble square root filter (EnSRF) assimilation module (Whitaker and Hamill, 2002), which are employed to simulate atmospheric compositions and infer anthropogenic emissions, respectively. The basic framework is almost identical to that in Zhang et al. (2024), in which we inferred the fossil fuel CO2 emission using surface CO₂ observations. In this study, the system was extended to high-resolution nested inversion for CH₄ emissions, which can optimize emissions from the outer to inner domain and reduce the influence of inaccurate boundary conditions on the inversion of the inner area (Feng et al., 2022). Moreover, the assimilation framework was updated to optimize CH₄ emissions by assimilating the TROPOMI CH₄ column retrievals. The RegGCAS-CH₄ performed a "two-step" inversion scheme in each data assimilation (DA) window. First, the prior emissions were optimized using the available atmospheric observations. Then, the optimized emissions were input back into the CTM to generate the initial fields for the next assimilation window and boundary conditions for the inner domain. Thus, the uncertainties in boundary conditions for the estimates of emissions in the inner domain were also reduced. Simultaneously, the optimized emissions were transferred to the next window to serve as prior emissions. This "two-step" scheme facilitates error propagation and iterative emission optimization, which can ensure the mass conservation of the system and effectively enhance the stability and



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consistency of the emission updates (Feng et al., 2024).

2.1.1 Atmospheric transport model

The Weather Research and Forecast (WRF v4.0) model (Skamarock and Klemp, 2008) and the 161 Community Multiscale Air Quality Modeling System (CMAQ v5.0.2) (Byun and Schere, 2006) were 162 applied to simulate meteorological conditions and atmospheric chemistry, respectively. In this study, 163 the CMAQ model employed two-nested simulations. The outer domain (D01), which covered the 164 whole mainland of China with a grid of 225 × 165 cells, and the inner domain (D02), which covered 165 the Shanxi Province and surrounding areas with a grid of 195 × 174 cells, had grid spacings of 27 and 166 9 km, respectively (Figure 1). There were 20 levels on the sigma-pressure coordinates extending from 167 the surface to 100 Pa. To account for the rapid expansion of urbanization, we updated the underlying 168 surface information for urban and built-up land using the MODIS Land Cover Type Product 169 (MCD12C1) Version 6.1 of 2022. The detailed configuration of WRF-CMAQ is shown in Table S1. 170



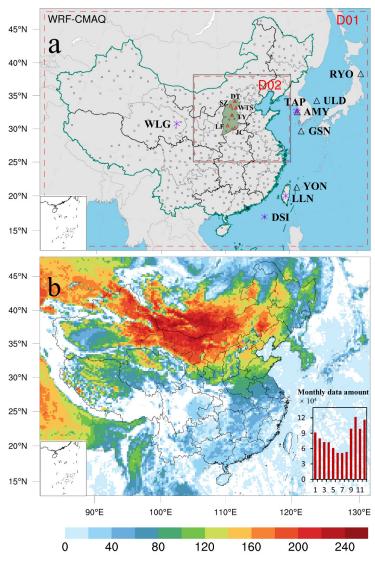


Figure 1 (a) Nested inversion domain and (b) number of TROPOMI XCH₄ retrievals during 2022. The red dashed frame depicts the CMAQ modeling domain; black squares represent the surface meteorological measurement sites; red triangles represent the six *in-situ* CH₄ measurement sites in Shanxi Province; purple asterisks and black triangles represent the flask and *in-situ* CH₄ measurement sites, respectively. The TROPOMI observations that fall within the same model grid are processed and counted as one super-observations. The subfigure in panel (b) shows the total number of super-observations for each month within the study area.





179 The meteorological initial and lateral boundary conditions were obtained from the Final (FNL) Operational Global Analysis data of the National Center for Environmental Prediction (NCEP) with a 180 1° × 1° resolution at 6-h intervals. The chemical lateral boundary conditions for the outer domain were 181 derived from the CAMS global inversion-optimized CH₄ concentrations with a 1° × 1° resolution at 182 6-hour intervals (Bergamaschi et al., 2013), while those for the inner domain were obtained from the 183 forward simulation of the outer domain with optimized CH₄ emissions. In the first DA window, the 184 chemical initial conditions were also extracted from the CAMS, whereas in subsequent windows, they 185 were derived through forward simulation using optimized emissions from the previous window. Since 186 the time required for CH₄ to be transported in the study area is much shorter than its lifetime, to reduce 187 the computational cost to the lowest possible level, we deactivated all the atmospheric chemical 188 reaction processes (Chen et al., 2022; Kou et al., 2025) and incorporated a set of CH₄ tracers variables 189 specifically for conducting ensemble simulations in the CMAQ, enabling all CH₄ concentration sets 190 to be obtained from a single simulation. 191 192 It is of utmost importance to eliminate biases in boundary conditions, as any such biases have the potential to cascade through the system. We found that the boundary conditions extracted from the 193 CAMS global fields still had considerable biases over East Asia (see Section 4). Thus, we calculated a 194 scaling factor for each grid of CAMS fields (50°E-160°E, 0°-70°N) against TROPOMI XCH4 195 retrievals, and then applied these factors to correct the biases in the boundary conditions of CAMS 196 (Figure S1). Based on each pair of CAMS and TROPOMI data, we first calculated the column 197 concentration of the CAMS reanalysis field. Then, the column concentrations of CAMS and 198 TROPOMI were respectively smoothed in the longitudinal and latitudinal directions with a radius of 199 4°, and in terms of time using a 4-day window. Subsequently, the latitudinal average values were 200 201 calculated, along with the average column concentrations biases between CAMS and TROPOMI. After that, linear interpolation was applied to fill in the missing values of the biases in the longitudinal 202 direction. For the biases of the grids where there were missing values at the same latitude, it was 203 assumed that they were consistent with the average biases of the non-missing values at that latitude. 204 Finally, grid-by-grid bias was calculated and correction was carried out for the original CAMS CH₄ 205 concentrations. 206





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the following equations:

2.1.2 EnKF assimilation algorithm

The EnKF is based on the Monte Carlo approach, which uses a stochastic ensemble of model states to approximate the probability distribution of the true state. It has been widely employed for updating the model state by incorporating observational data to minimize the difference between the modelsimulated and observed values (Evensen, 1994). The ensemble square root filter (EnSRF) approach,

introduced by Whitaker and Hamill (2002), was used to constrain the CH₄ emissions in this study.

The EnSRF process commences with the initialization of an ensemble of model states, which are generated by applying a Gaussian perturbation with an average value of zero and the standard deviation of the uncertainty to a state vector X^b . The ensemble-estimated background error covariance matrix P^b is then calculated as:

where N is the ensemble size; X_i^b represents the ith sampling; \overline{X}^b represents the mean of the

ensemble samples. P^b plays a pivotal role in determining how the model state will be adjusted based

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$$\boldsymbol{P^b} = \frac{1}{N-1} \sum_{i=1}^{N} (\boldsymbol{X_i^b} - \overline{\boldsymbol{X}^b}) (\boldsymbol{X_i^b} - \overline{\boldsymbol{X}^b})^T$$
 (1)

221 on new observations.

222 During the forecast step, each ensemble member is advanced in time using the WRF-CMAQ model. As

223 the model runs, uncertainties in emissions can lead to errors in CH₄ concentrations, and thus the

224 response relationships of the concentration ensembles to the emission ensembles are obtained. In the

225 analysis step, observational data y are incorporated to update the analyzed state. The ensemble

226 mean of the analyzed state $\overline{X^a}$ is regarded as the best estimate of emissions, which is obtained through

$$\overline{X^a} = \overline{X^b} + K(y - H\overline{X^b}) \tag{2}$$

$$K = P^b H^T (HP^b H^T + R)^{-1}$$
(3)

where R is an error covariance matrix; K is the Kalman gain matrix, estimated from the ensemble simulations and determining the relative contributions of observation and background to analysis. The state vector was defined as $X^b = (Ea^T, Ep^T)^T$, where Ea and Ep represent the vectors of CH₄ emissions for the area and power plant sources, respectively. Area sources included the daily total





- 234 emissions from the enteric fermentation & manure, landfills & waste, rice cultivation, coal mining, Oil
- 235 & gas, industry, transport sources, etc. The updated emissions are then used as the new initial states
- 236 for the next forecast step, creating a cycle of assimilation that gradually refines the estimate
- 237 of CH₄ emissions.
- The observation operator H maps the model state to the observation space. In the context of CH₄, H is
- 239 configured to first horizontally geo-locate simulated CH₄ concentrations to match the TROPOMI
- 240 XCH₄ retrievals. Subsequently, it remaps the sub-column concentrations from the 20-layer CMAQ
- 241 vertical grid to the 12-layer TROPOMI vertical grid by totally or partially allocating CMAQ layers to
- 242 TROPOMI layers based on pressure edges (Varon et al., 2022). Finally, the column average dry-air
- 243 mixing ratio XCH4_s can be obtained by applying the TROPOMI column averaging kernel of each
- layer a_i to sub-columns:

$$XCH4_{s} = VCH4_{s}/VAIR_{drv,i}$$
 (4)

$$VCH4_s = VCH4_p + \sum_{i=1}^{n} a_i \left(XCH4_{s,i} \Delta VAIR_{dry,i} - \Delta VCH4_{p,i} \right)$$
 (5)

- where n is the number of retrieval layers; $\Delta VCH4_{p,i}$ and $VCH4_p$ represent the prior CH₄ column
- 248 in retrieval layer i and total CH₄ column; XCH4_{s,i} is the simulated dry air mixing ratio of CH₄ in
- retrieval layer i; $\Delta VAIR_{dry,i}$ and $VAIR_{dry,i}$ represent the dry air column in retrieval layer i and
- 250 total dry air column, respectively, provided along with the TROPOMI product.
- 251 In this study, the DA window was set to 1 d, meaning that daily TROPOMI XCH4 retrievals were
- 252 utilized as emission constraints. The ensemble size was set to 50 to fully characterize the system
- 253 uncertainties and inversion accuracy. To address the problem of spurious long-distance correlations in
- 254 the EnKF (Houtekamer and Mitchell, 2001), we applied covariance localization using the Gaspari and
- 255 Cohn function, which is a piecewise continuous fifth-order polynomial approximation of a normal
- distribution (Miyazaki et al., 2017; Gaspari and Cohn, 1999). Taking into account the transmission
- distance of CH₄ within one assimilation window, the localization scale was set to 300 km. By setting
- 258 the covariance to be smaller the farther away from the observations, we could reduce the spurious
- 259 influence of remote observations on the local analysis.

2.2 Prior emissions and uncertainties

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261 The prior anthropogenic CH₄ emissions were taken from the Emission Database for Global





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262 Atmospheric Research version 8.0 (EDGAR v8), which offers detailed monthly gridded CH₄ emissions at 0.1°×0.1° from various anthropogenic sources (Crippa et al., 2024). The daily emissions, 263 obtained by uniformly allocating the aggregated monthly emission inventory, were directly utilized as 264 265 the initial estimate in the RegGCAS-CH₄. For natural CH₄ emissions, we utilized the ensemble average of 18 emissions from the WetCHARTs v1.3.1 inventory (0.5° × 0.5°, monthly) in 2019 for wetland 266 emissions (Bloom et al., 2021), the Global Fire Emissions Database (GFED v4.1s, 0.25° × 0.25°, three-267 268 hourly) in 2022 for biomass burning emissions (van Wees et al., 2022), and CAMS global emissions inventory (CAMS-GLOB-TERM v1.1, 0.5° × 0.5°, monthly) in 2000 for termite emissions (Jamali et 269 al., 2011). Additionally, CH₄ sinks resulting from soil absorption were derived from datasets simulated 270 by Soil Methanotrophy Model (MeMo v1.0, 1∘ × 1∘, yearly) in 2020 (Murguia-Flores et al., 2018). 271 272 Given the model error compensation and the relatively comparable emission uncertainties from one 273 day to the next, we applied an identical uncertainty of 40% to each emission grid at every DA window. 274 Since the RegGCAS-CH₄ adopts a 'two-step' inversion strategy and the daily posterior emissions are iteratively optimized, the emission analysis is generally no longer sensitive to the prior uncertainties 275 of the original emission inventory after several assimilation windows (Zhang et al., 2024). 276 2.3 Assimilation data and errors 277 TROPOMI, onboard the Sentinel-5 Precursor satellite, was launched in 2017. Operating in a sun-278 synchronous orbit with an equator local overpass time of 13:30 h, TROPOMI offers daily global 279 280 continuous monitoring of XCH₄. The RemoTeC full-physics algorithm is used to retrieve XCH₄ from 281 TROPOMI measurements of sunlight backscattered by Earth's surface and atmosphere in the near-282 infrared (NIR) and shortwave-infrared (SWIR) spectral bands (Lorente et al., 2022). After 2019, the 283 spatial resolution of TROPOMI was adjusted to a remarkable 5.5×3.5 km² at nadir, enabling the identification of even relatively small-scale CH₄ emission sources. We used the TROPOMI XCH₄ level 284 2 data product to estimate CH₄ emissions. 285 All TROPOMI individual pixels with a quality assurance value (qa value) smaller than 0.5 were 286 discarded, which corresponds to high-cloud conditions or the presence of snow or ice. However, we 287

still found many unrealistic low values, especially in summer. To further minimize the impact of

outliers, we selected another XCH4 product generated based on the WFMD algorithm for cross-





290 validation (Schneising et al., 2023). Only those pixels that were concurrently available in the TROPOMI/WFMD product and met the quality flag requirements were assimilated. Figure 1b 291 illustrates the observational amount of TROPOMI XCH4 in 2022 at each grid. Although the 292 293 distribution of filtered data exhibits spatial nonuniformity, most grid cells in the central-northern 294 regions with intense anthropogenic emissions have observational coverage for more than 100 days in 2022. Additionally, the monthly variation in the data amount shows fluctuations, with the peak 295 occurring at the beginning and end of the year and the troughs around the middle. According to the 296 latest quarterly validation report, the 1σ spread of the relative difference between the TROPOMI and 297 the TCCON (Wunch et al., 2011) is of the order of 0.7% for bias corrected product, which is 298 recommended to be considered as an upper boundary of the random uncertainty of the satellite data 299 300 (Lambert et al., 2024).

2.4 Evaluation Data

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302 To evaluate the performance of the WRF simulations, we utilized the surface meteorological measurements of 400 stations with 3-hour intervals, including temperature at 2 m (T2), relative 303 humidity at 2 m (RH2), and wind speed at 10 m (WS10). These measurements were obtained from the 304 National Climate 305 Data Center (NCDC) integrated surface database (http://www.ncdc.noaa.gov/oa/ncdc.html, last access: 25 August 2024). Overall, the WRF 306 demonstrated satisfactory performance in reproducing meteorological conditions over China (Figure 307 S2), with minimal biases of -0.4°C for T2, -4.9% for RH2, and 0.5 m/s for WS10, respectively. To 308 309 evaluate the posterior CH₄ emissions, two parallel forward modeling experiments were conducted: the control experiment with prior emissions (CEP) and the validation experiment with posterior emissions 310 (VEP). Both experiments utilized identical meteorological fields, as well as initial and boundary 311 312 conditions. We evaluated: (1) the simulated XCH₄ against TROPOMI XCH₄; (2) the simulated surface CH₄ concentrations against independent ground CH₄ observations from eleven in-situ and five flask 313 monitoring sites (Table S2). The in-situ measurements included five global sites outside China (AMY, 314 315 GSN, RYO, ULD, and YON) with hourly resolution from NOAA's GLOBALVIEWplus CH4 ObsPack v6.0 (Schuldt et al., 2023), as well as six regional sites in Shanxi Province (TY, DT, LF, SZ, JC, and 316 317 WTS) equipped with Picarro G2301 analyzers for high-precision, 5-second CH₄ measurements (data 318 were processed as daily averages to reduce random errors). The Picarro instrument, based on



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- 319 wavelength-scanned cavity ring-down spectroscopy technology, is designated by the World
- 320 Meteorological Organization as the international reference instrument for CH₄ observations in
- international comparisons. Additionally, observations from five flask sampling sites (AMY, DSI, LLN,
- 322 TAP, and WLG) were also obtained from ObsPack dataset, providing weekly measurements.

3. Results and discussion

3.1 Posterior CH₄ emissions

325 Figure 2 shows the spatial distributions of the estimated annual CH₄ emissions and the differences

326 from the prior emissions (i.e., EDGAR) over China in 2022. On a national scale, the high CH₄

emissions were primarily concentrated in the Yangtze River Delta (YRD), Pearl River Delta (PRD),

North China Plain (NCP), and Shanxi Province. For the YRD and PRD characterized by high-density

populations, a large number of industrial plants, as well as extensive agricultural activities such as rice

cultivation, contributed significantly to CH₄ emissions. The NCP, with its heavy reliance on coal-based

331 energy and large-scale livestock farming, was also expected to be a major emission source. Lower

emissions were mainly distributed across Northwest, and Southwest China.

333 Compared with the EDGAR, the posterior emissions were generally smaller over most areas of

mainland China, with total anthropogenic emissions decreasing to 45.1 TgCH₄·yr⁻¹, 36.5% lower than

the EDGAR (71.0 TgCH₄·yr⁻¹). Additionally, previous studies have consistently shown that the

EDGAR inventory overestimates China's CH₄ emissions. For example, through a Bayesian inversion

337 of CH₄ and stable isotope (δ¹³C-CH₄) measurements for East Asia, Thompson et al. (2015) found that

posterior values decreased significantly across eastern and southern China, especially in the

NCP. Overall, EDGAR overestimated China's emissions by 29%. Similarly, Zhang et al. (2021)

340 conducted a global inverse analysis using GOSAT observations and revealed that EDGAR significantly

overestimates anthropogenic emissions in eastern China. The posterior estimate of Chinese

anthropogenic emissions was 30 % lower than that of EDGAR. Turner et al. (2015) also demonstrated

that after assimilating GOSAT observations, China's posterior CH₄ emissions were revised downward

by 50% relative to EDGAR. Using a regional model to assimilate TROPOMI observations, Kou et al.

(2025) detected decreases of varying magnitudes across nearly the entirety of China, with the exception

of Northwest China. Similar results have been reported by Alexe et al. (2015), Pandey et al. (2016),



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and Maasakkers et al. (2019). Our study, using a higher-resolution regional inversion method, provided more detailed emission information. Overall, our inversion results were comparable to the ensemble mean of GCP ground-based inversions (Wang et al., 2025).

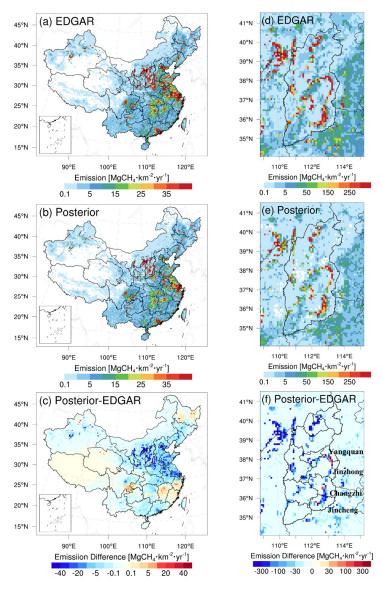


Figure 2 Spatial distribution of the annual total prior emissions (top, EDGAR v8, MgCH₄·km⁻²·yr⁻¹), posterior emissions (middle), and differences (borrom, posterior minus prior) over (a-c) Mainland China and (d-f) Shanxi Province.



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Table 1 shows the comparison of posterior and prior anthropogenic CH₄ emissions of the main emission sectors in China. Consistent with the previous studies, we also found that North China is the 355 region with the most significant reduction nationwide. Almost the entire region has experienced a 356 decrease, with a reduction of 56.9%, indicating that there are indeed substantial systematic biases in the EDGAR inventory. Among them, the coal sector contributed the most to this discrepancy, followed 358 by enteric fermentation, with decreases of 56.2% and 64.0%, respectively (Table 1). 359 For Northeast China, the increase in emissions mainly occurred in Heilongjiang Province, especially 360 in the western part, which is the base of Daqing Oilfield, China's largest oilfield. The oil and gas sectors in the entire Northeast China increased by 27.1% and 23.1%, respectively. However, emissions from 362 other sectors decreased, resulting in an overall 11.1% reduction in emissions. Rice paddy CH₄ 363 emissions serve as the dominant emission source in East China. However, the high spatial 364 heterogeneity and the insufficiency of data on rice cultivation introduce large uncertainties to 365 inventories. Additionally, different fertilization management practices in various regions, such as the 366 367 use of nitrogen fertilizer versus organic fertilizer (Zhang et al., 2022), and mid-season drainage management (Lin et al., 2021), bring about considerable uncertainties in the emission factors related 368 369 to rice cultivation practices. Emissions in Zhejiang, Fujian, and Jiangxi Provinces increased, mainly 370 attributed to emissions from rice paddies. In contrast, emissions in other provinces decreased, dominated by the coal mining, leading to an overall 26.4% reduction in emissions in East China. In 371 372 Central and South China, the dominant sources of emissions remain rice paddies. However, the 373 decreases were mainly attributed to the coal mining and wastewater treatment sectors, which reduced emissions by 51.3% and 12.4%, respectively. Overall, the total emissions in these two regions 374 375 decreased by 21.8% and 7.8%, respectively. In Northwest China, the reduction in emissions was 376 mainly distributed in the hotspot areas of coal mines, which dominated the overall 55.7% decrease in emissions. In Southwest China, the increase in emissions occurred predominantly in the southern part 377 of the Sichuan Basin, a major coal-producing region, while emissions in other regions decreased. 378 Sheng et al. (2019) found that the EDGAR inventory lacks a significant amount of statistics on coal 379 380 mines. Moreover, China has issued an energy policy of "phasing out small coal mines" (approximately 50% of which are located in the southwest with high CH₄ content) to shift production towards lower-381 emission areas and consolidate into large coal mines (Zhang et al., 2022; Sheng et al., 2019). 382





Additionally, the wastewater treatment sector achieved significant emission reductions, driving a 1.7% overall decrease in Southwest China.

Table 1 Comparison of posterior and prior anthropogenic CH₄ emissions (GgCH₄·yr⁻¹) of the main emission sectors in the 7 major regions of China. Waste includes wastewater treatment, solid waste landfills, and solid waste incineration.

		Coal	Gas	Rice	Waste	Livestock	Building	Mature
North	Prior	13841.1	98.4	169.1	2462.1	3882.9	385.5	859.5
	Posterior	6068.4	46.2	116.4	1238.1	1396.4	206.7	279.6
Northeast	Prior	771.9	82.6	879.6	1427.9	251.9	268.4	36.8
	Posterior	665.0	101.8	821.2	1259.6	237.7	243.3	34.1
East	Prior	2257.3	54.4	5100.3	6681.4	907.9	732.9	200.5
	Posterior	1114.5	45.1	4592.4	4750.9	512.7	576.2	114.4
Central	Prior	846.5	32.4	3139.5	1882.9	434.2	399.4	91.2
	Posterior	412.2	25.7	2862.6	1324.4	343.3	314.1	75.2
South	Prior	15.0	20.5	2186.4	1622.5	280.2	254.6	44.0
	Posterior	13.8	18.9	2080.6	1421.3	269.9	235.0	42.2
Northwest	Prior	8188.3	1083.4	143.3	970.5	746.2	226.7	76.4
	Posterior	2899.1	645.8	120.7	593.6	534.3	164.4	39.6
Southwest	Prior	531.3	76.5	2046.4	1124.0	667.0	356.3	107.1
	Posterior	505.6	77.2	2063.0	1060.7	663.7	352.1	107.2

China accounts for as high as 69% of the global mitigation potential in coal mining in 2030 (EPA, 2019). Shanxi Province, where 94% of the emissions come from the coal mining sector, counts for nearly one-third of the country's total CH₄ emissions. Therefore, we conducted a focused analysis on the emissions in Shanxi Province with high-resolution (9 km) inversion, which can better diagnose the spatial characteristics of emissions caused by coal mining (Figure 2d-f). The optimized emissions showed a decrease in the vast majority of areas in Shanxi Province. Compared with the EDGAR inventory (9.0 TgCH₄·yr⁻¹), the posterior emissions decreased by 46.3% to 4.8 TgCH₄·yr⁻¹. The overestimation of coal mining emissions may be due to the fact that the standard IPCC emission factors used by EDGAR are too high for Chinese coal mines (Lin et al., 2021). Additionally, the average





emission factors in the north are lower than those in other regions (Gao et al., 2021). The use of a uniform regional emission factor by EDGAR further exacerbates the overestimation of emissions (Shi et al., 2025). Moreover, in the past decade, the extraction and utilization of coal mine CH4 in China have been largely improved (Lu et al., 2021), but the recovery of coal mine CH₄ is not adequately considered in the EDGAR inventory. Finally, the CH₄ emission intensity of surface mining is ten times lower than that of underground mining. However, surface mining is overlooked in the EDGAR inventory (Gao et al., 2020). In Yangquan, Jincheng, Changzhi, and Jinzhong cities (Figure 2), some increased emission hotspots caused by the coal mining activities could be found. Coincidentally, these cities are the main coal-producing areas in Shanxi Province. Overall, the emissions in these cities have still decreased by 3.3%, 4.2%, 29.5%, and 32.3%, respectively (Figure S3). Tu et al. (2024) utilized TROPOMI observations and implemented a wind-assigned anomaly method to quantify the CH₄ emissions from coal mines in Yangquan, Changzhi, and Jincheng. Compared to EDGAR, emissions decreased by 56.5%, 40.5%, and 65.0%, respectively—a larger reduction than our findings.

3.2 Monthly variations

Figure 3 illustrates the comparison of the monthly variations in prior and posterior emissions both in China and Shanxi province. Nationally, influenced by CH₄ emissions from paddy fields, both the prior and posterior emissions exhibited the characteristic of being low in winter and high in summer. However, the posterior emissions for each month were lower than the prior emissions. The monthly posterior emissions ranged from 2.3 to 7.5 Tg·month⁻¹, with a modification rate ranging from -30.7% to 5.6% compared to the prior emissions (4.8–7.6 Tg·month⁻¹). The month with the smallest difference occurred in August, mainly because the underestimation of CH₄ emissions from paddy fields compensated for the overestimation of emissions from coal mining (Figure 3c). Additionally, due to the weather conditions such as clouds and rain in summer, the amount of TROPOMI data was significantly smaller than that in other seasons (Figure 1b), which might lead to insufficient constraints on emissions. Regarding Shanxi Province, even with an agnostic flat monthly prior (Figure 3b), our estimates generated a monthly variation, and emissions in all months were lower than the prior values. We also detected a decrease and subsequent increase in emissions that correspond to the Spring Festival in February. A noticeable recovery of production capacity in the following two months was also evident. Overall, although relatively large uncertainties were introduced by the amount of





To gain a deeper understanding of the posterior emissions, the assimilated estimations were allocated 427 to different emission sectors, based on the monthly prior proportions at the model grid points. The 428 sectoral patterns offer insights into the underlying factors influencing China's emission changes. 429 430 Consequently, we further concentrated on interpreting the emissions from the coal, gas, rice cultivation, waste, livestock, building, and manure management sectors, which are the most significant sectors in 431 432 China (Table S3). It could be found that the reduction in emissions each month in China was mainly dominated by the 433 coal sector, with an overall annual reduction of 55.9%, indicating a high level of uncertainty in the 434 435 prior emissions. Moreover, the gas, livestock, and manure management sectors also showed varying degrees of reduction in different months, with overall decreases of 34.0%, 45.1%, and 51.5%, 436 respectively. However, for the rice cultivation sector, the posterior emissions were lower than the prior 437 emissions in the first half of the year, while the situation was reversed in the second half of the 438 year. Summer and autumn are the heading and flowering stages for both single-cropping late rice and 439 440 double-cropping late rice across the eastern, central, and southern China. During these periods, the 441 emissions from paddy fields tended to be higher than in other seasons. Overall, the posterior CH₄ emissions from paddy fields have slightly decreased by 4.7%. A similar situation was observed in the 442 waste sector. Previous studies have shown that significant CH₄ production in wastewater is more likely 443 to occur because methanogens become more active as the temperature rises (Hu et al., 2023). Therefore, 444 a significant increase in the posterior emissions compared to the prior emissions could be observed in 445 August. For the building sector, due to winter heating or hot water supply, the emissions in winter were 446 approximately three times higher than those in summer. After optimization, the emissions in winter 447 448 and spring significantly decreased, while in some months of summer and autumn, the decrease was less pronounced, and there was even a slight increase. The inter-monthly difference in emissions 449 decreased, but the monthly variation remained significant. Overall, the posterior emissions have 450 451 decreased by 25.4%. The changes in emissions in Shanxi Province were mainly dominated by the coal 452 mining sector. Compared to EDGAR, the monthly reduction rate ranged from 12.6% in August to 67.3% 453 in February. For other sectors, there was little overall monthly variation.

observations, the seasonal variation of monthly posterior emissions could be roughly captured.



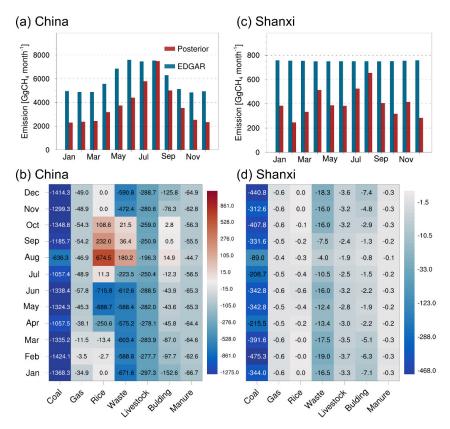


Figure 3 Comparison of the monthly variations and the sectoral differences (GgCH₄·mon⁻¹) in prior and posterior CH₄ emissions over (a, b) China and (c, d) Shanxi Province.

3.3 Evaluation of posterior emission estimates

Figure 4 shows the spatial distribution of XCH₄ in the posterior simulation, as well as the validation of XCH₄ simulated by prior and posterior emissions with TROPOMI observations. It could be observed that relatively large XCH₄ were present over eastern China, which was driven by coal mining in the northern part and rice paddy fields in the southern part (Zhang et al., 2023). The simulation using prior emissions significantly overestimated the XCH₄ concentration in China, especially in the NCP. The maximum overestimation exceeded 100 ppb, which was consistent with the overestimated emissions in these regions (Figure 2). After the inversion optimization, the simulated XCH₄ showed better spatial distribution consistency with the TROPOMI observations. The vast majority of the biases were within 20 ppb, and the national average biases decreased from 14.7 ppb to 7.9 ppb, representing

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a 46.0% reduction. In Shanxi Province, high emissions were mainly concentrated in the coal-rich southeastern region, with an annual average maximum exceeding 1970 ppb, which was difficult to reproduce the characteristics of local high concentrations at a coarse resolution. The prior simulation showed obvious overestimation across the entire region, particularly in northern Shanxi. Due to the high spatial resolution adopted, the model could take into account more practical factors, and thus capture the spatiotemporal variations of emission sources and transport processes more precisely. As a result, the agreement between the posterior simulations and the TROPOMI observations was remarkably enhanced. The average deviation decreased from 37.9 ppb to 11.9 ppb, with a reduction of 68.7%. These changes not only demonstrate the effectiveness of reducing uncertainties in optimizing emission sources but also imply that there are indeed significant uncertainties in the prior emission inventory.



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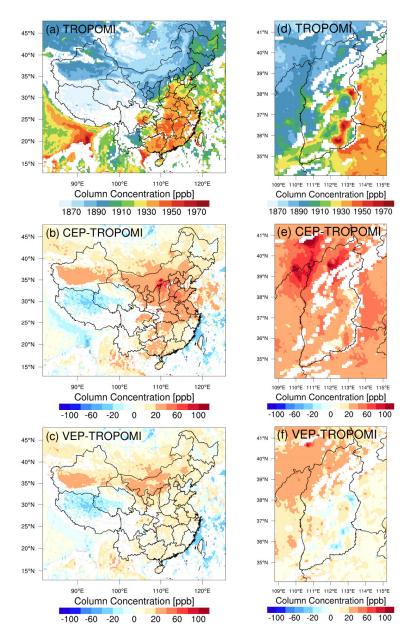


Figure 4 Comparison of simulated XCH₄ (ppb) from prior and posterior emissions with TROPOMI observations over (a-c) China and (d-f) Shanxi Province. (a, d) Spatial distribution of XCH₄ observed by TROPOMI. (b, e) Differences between prior simulations and observations. (c, f) Differences between posterior simulations and observations.



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484 We further conducted an independent evaluation of the posterior estimate by comparing it with 10 insitu and flask monitoring sites from the CH₄ ObsPack v6.0 database. Figure 5 shows the time series 485 comparison of the CEP and VEP experiments with the observations. The evaluation statistics for all 486 sites are presented in Table S4. There was a significant overestimation in the CEP experiment throughout most of the study period. Although most of the sites are located in the eastern regions 488 outside China, influenced by the atmospheric circulation, the westerlies prevail in most parts of 489 China. Thus, the optimized emission information can be effectively reflected in the concentrations 490 observed downwind. The optimized simulation in VEP experiments was more consistent with the observations. Among them, the AMY flask site showed the largest reduction in bias, with the average 492 bias decreasing from 35.3 ppb to 3.3 ppb, a decrease of 90.7%. For WLG, the only ObsPack site 493 available in mainland China, the average bias decreased from 47.4 ppb to 28.3 ppb, a reduction of 494 40.3%. This indicates that the optimized emissions can significantly improve the CH₄ simulation, 495 whether in areas close to the source or downwind of the source. Overall, the average bias of the 10 496 sites decreased by 58.6%, from 28.6 ppb to 11.9 ppb, the root mean square error (RMSE) decreased 497 by 28.9%, from 56.2 ppb to 39.9 ppb, and the correlation coefficient (CORR) increased from 0.72 to 498 0.75. These evaluation results demonstrate that the inversion effectively reduces the uncertainties of 499 500 prior emission inventory.



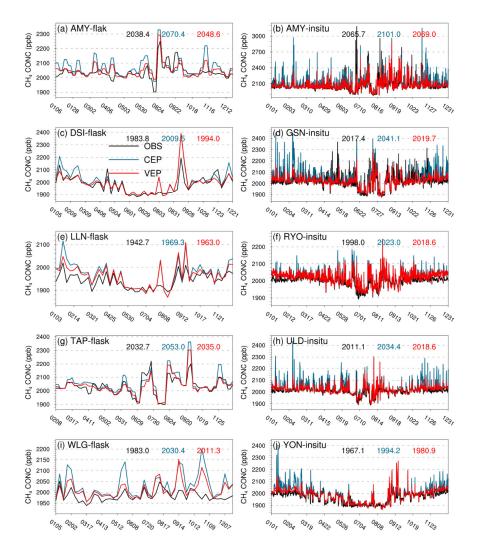


Figure 5 Time series comparison of surface CH₄ concentrations (ppb) from prior (CEP) and posterior (VEP) emission simulations with observations from 5 flask sites and 5 *in-situ* sites within the ObsPack dataset. The black, blue, and red values represent the averaged observations, prior simulations, and posterior simulations, respectively.

We also carried out an independent validation using six high-precision *in-situ* observation sites within Shanxi Province (Figure 1a). Table 2 shows the bias, RMSE, and CORR of the CEP and VEP simulations against these surface observations. Except for the LF site, where the VEP experiment exhibited an underestimation, the VEP experiment demonstrated varying degrees of improvement at





the other five sites. Notably, the bias reduction at the DT and WTS sites exceeded 93.3%. Especially for the WTS site, a high mountain site with an altitude of over 2208 m, the bias was decreased to 8.3 ppb. On average across all sites, the bias significantly decreased by 96.0%, from 535.1 ppb to 21.4 ppb. For the RMSE, the most significant decreases were observed at the DT and SZ sites, both exceeding 73%. Overall, the RMSE reduction ranged from 13.5% to 79.0%. Additionally, the CORR of the VEP experiment increased to 0.48–0.63. In addition, we evaluated the CH₄ concentration simulation for the afternoon, during which the model generally demonstrates better boundary layer simulation performance, with overall lower bias and higher CORR (Table S5). These results further confirm that the RegGCAS-CH₄ system can effectively capture the characteristics of high-resolution CH₄ emission changes and improve the accuracy of concentration simulations.

Table 2 Statistics comparing the daily average CH₄ concentrations (ppb) from the simulations with prior (CEP) and posterior (VEP) emissions against six independent surface *in-situ* observation sites in Shanxi Province, respectively. The numbers under the site names represent the number of valid observations.

Site Name	Mean Obs.	Mean Sim.		BIAS		RMSE		CORR	
		CEP	VEP	CEP	VEP	CEP	VEP	CEP	VEP
TY (351)	2595.5	2771.0	2353.2	175.4	-242.3	408.7	353.6	0.46	0.51
DT (286)	2122.0	2441.1	2139.2	319.1	17.2	489.0	102.6	0.49	0.58
LF (277)	2395.1	2397.2	2197.9	2.0	-197.3	186.8	257.3	0.45	0.35
SZ (359)	2148.8	4560.5	2782.4	2411.7	633.6	2778.1	745.1	0.44	0.48
JC (362)	2446.0	2624.5	2355.2	178.4	-90.9	385.0	356.3	0.52	0.52
WTS (361)	2064.0	2188.0	2072.2	124.1	8.3	162.6	59.1	0.49	0.63

^{*} BIAS, mean bias; RMSE, root mean square error; CORR, correlation coefficient

4. Discussion

To further explore the characteristics of our posterior emissions and offer valuable guidance for the





528 refinement of bottom-up inventory in China, we conducted a comparison analysis with the latest 8 bottom-up inventories and 4 top-down emission estimates (Figure 6). Specifically, apart from EDGAR 529 v8, the bottom-up inventories were sourced from the Copernicus Atmosphere Monitoring Service 530 531 (CAMS v6.2) (Soulie et al., 2024), Community Emissions Data System (CEDS 202407) (Hoesly et al., 2018), Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants inventory 532 (ECLIPSE v6b) from the GAINS model (Stohl et al., 2015), Global Carbon Project (GCP 2024) 533 (Saunois et al., 2025), Peking University (PKU v2) (Liu et al., 2021), United Nations Framework 534 Convention on Climate Change (UNFCCC, 2020), and the United States Environmental Protection 535 Agency (EPA, 2019). In addition, we also obtained the Global Fuel Exploitation Inventory (GFEI 536 v2) (Scarpelli et al., 2022) to assess the emissions from the coal mining sector in Shanxi Province. The 537 top-down emissions were mainly sourced from the CarbonTracker-CH₄ (Oh et al., 2023) and the 538 research results of Chen et al. (2022), Kou et al. (2025), and Peng et al. (2023). 539 540 Typically, the top-down estimates showed lower emissions than the bottom-up inventories. Although 541 our posterior estimates were the lowest across all datasets, they closely matched CEDS, PKU, and CarbonTracker-CH₄ and comparable to the ensemble mean of GCP ground-based inversions (Wang et 542 al., 2025). Overall, our posterior emissions were 22.0% lower than the average of bottom-up 543 inventories and 16.6% lower than the previous top-down estimates. The lower emissions in this study 544 were predominantly driven by the downward revision of coal emissions. Overestimated emission 545 factors and the difficulty in tracking the spatial distribution of coal mines due to mine closures and 546 547 regional transfers remain significant obstacles to the assessment of coal mine emissions (Gao et al., 2021). Our estimate was lower than the previous top-down studies (53-65 Tg yr⁻¹), likely because 548 those previous studies were conducted at much coarser resolutions (0.3°-2.5° versus 27 km) or with 549 550 much sparser observations (in-situ ground measurements and GOSAT versus TROPOMI). For Shanxi Province, compared with the bottom-up inventories, our results were close to those of GAINS and fell 551 within the uncertainty range of Qin et al. (2024), which set multiple groups of emission factors 552 according to coal types, mining methods, and geological structures. There were substantial disparities 553 among the top-down inversions. Notably, for CarbonTracker-CH₄, despite having minimal divergence 554 at the national scale when contrasted with other estimations, the values for Shanxi Province were 555 556 markedly lower than those of other estimates. This could be attributed to the insufficient assimilation





of China's surface observations and systematic biases in the spatial distribution of prior emissions. Overall, the coal-mining emissions in this study were 29.2% lower than the bottom-up inventories and 19.3% lower than the previous top-down inversions.

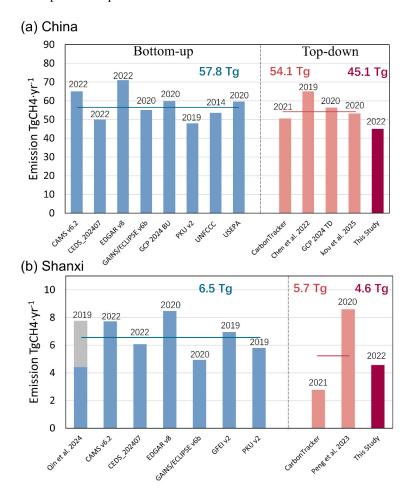


Figure 6 Total anthropogenic CH₄ emissions (TgCH₄·yr⁻¹) in China (a) and CH₄ emissions from the coal mining sector in Shanxi Province (b). GCP 2024 BU and GCP 2024 TD represent the bottom-up and top-down ensemble means included in the dataset, respectively. The grey bars in panel b represent the range of CH₄ emissions estimated by Qin et al. (2024) using four different methods. The latest year in the inventory is marked above the bars.

Uncertainties in boundary conditions constitute a significant source of error in regional inversion.

Despite the optimization of concentration fields in the CAMS, significant biases remained evident

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568 (Figure 7a). To address this issue, we implemented additional constraints on the CAMS concentration fields using TROPOMI XCH4 observations, resulting in a notably improved agreement with the 569 observations. Specifically, the average bias decreased from -3.7 ppb to -1.0 ppb, representing a 570 571 reduction of 73.7%. A sensitivity experiment (SENS) was further conducted, where the unadjusted CAMS global fields were extracted as boundary conditions to invert anthropogenic CH₄ emissions, 572 aiming to evaluate the impact of boundary condition uncertainties on regional emission inversions. 573 574 Compared with the base experiment (BASE), the largest discrepancies in monthly variations were observed during the winter months, indicating significant overestimations. The emission differences 575 across different months closely aligned with the concentration differences, suggesting that 576 underestimations in concentrations prompted more substantial emission adjustments for compensation. 577 Regarding the spatial distribution, due to the relative long lifetime of CH₄, emission changes caused 578 by boundary condition biases were not confined to regional boundaries, such as in Northeast and 579 Northwest China, but were also observable throughout the entire region. In the outer national inversion, 580 581 the average CH₄ emissions increased by 5.1%, while in the inner nested inversion, the increase was 4.3%. This highlights the necessity of expanding the inversion scope to mitigate the influence of 582 583 boundary condition errors on the inversion of central regions. 584 Furthermore, by comparing the difference in CH₄ emissions over the inner domain between two outer national inversions with that between two inner nested regional inversions (BASE and SENS), we 585 586 found that the two inner regional inversions exhibited a 23.1% smaller discrepancy. This indicates that 587 adopting a nested inversion approach is essential, which can further reduce boundary errors during inner inversion through outer optimization, thereby enhancing the robustness of the inversion process. 588

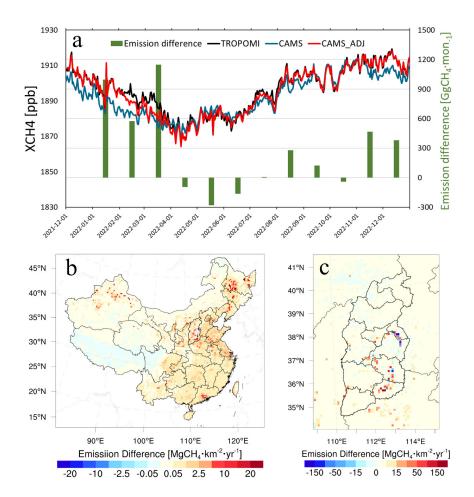


Figure 7 Differences in XCH₄ column concentrations (ppb) between TROPOMI-adjusted and unadjusted CAMS fields, along with the induced monthly posterior CH₄ emission differences (SENS - BASE) over China (a); differences in two posterior emissions (MgCH₄·km⁻²·yr⁻¹) derived from unadjusted and adjusted boundaries over China (b) and Shanxi Province (c)

Our inversion results are generally lower than previous emission estimates. By comparing the differences in emissions from the inner region at different resolutions, we found that in both the BASE and SENS experiments, emissions inverted in the inner region (9 km) were typically 7.6% and 8.4% lower than those in the outer region (27 km), respectively. This discrepancy likely arises because higher-resolution simulations excel at capturing localized emission hotspots that lead to elevated concentration values. Therefore, compared to other coarse-resolution inversions, this study tends to





600 adjust emissions downward to better align with observations. Additionally, China's 2022 COVID-19 restrictions, the most stringent since 2020, may be another factor driving lower emissions due to 601 production shutdowns and home quarantine. Peng et al. (2022) combined bottom-up and top-down 602 approaches to quantify CH₄ source changes, revealing a 1.2 Tg·CH₄ yr⁻¹ reduction in anthropogenic 603 CH₄ emissions in 2020 compared to 2019. Taking oil and gas sector as an example, an IEA report 604 stated that global CH₄ emissions from oil and gas operations decreased by approximately 10% year-605 on-year in 2020 (IEA, 2021), primarily attributed to reduced oil and gas production during the 606 pandemic (Thorpe et al., 2023). 607

5 Summary and conclusions

609 In this study, we developed a Regional Methane Assimilation System (RegGCAS-CH₄) based on the 610 WRF-CMAQ model and EnKF algorithm to perform high-resolution nested inversions of China's CH4 emissions using TROPOMI XCH4 retrievals. Taking the EDGAR v8 as prior emissions, the 611 daily anthropogenic CH₄ emissions over China in 2022 were inferred. 612 Using TROPOMI XCH₄ as constraints, we revealed significant overestimations in the prior emissions. 613 614 Nationally, the posterior anthropogenic CH₄ emissions in 2022 were estimated to be 45.1 TgCH₄·yr⁻¹, 36.5% lower than the EDGAR inventory, mainly driven by the decreases in the coal and waste sectors. 615 North China experienced the most substantial reduction, with a 56.9% decrease, mainly due to 616 downward revisions in the coal and enteric fermentation sectors. In Shanxi Province, influenced by 617 dominant sources of coal mining, the posterior emissions decreased by 46.3% compared to the 618 EDGAR inventory, with value of 4.8 TgCH₄·yr⁻¹. The monthly variation of posterior emissions 619 showed a pattern of being low in winter and high in summer. Except for the rice cultivation and waste 620 621 sectors, which showed some increases in certain months related to seasonal growth stages and temperature-driven methanogen activity, respectively, other sectors exhibited varying degrees of 622 reduction. Observation-constrained emissions significantly improved the performance of simulations 623 regarding XCH₄ columns, as well as surface CH₄ in both the entire region and Shanxi Province, 624 reducing biases by 46.0%, 58.6%, and 96.0%, respectively. This highlights the effectiveness of 625 RegGCAS-CH4 in reducing uncertainty in CH4 emissions. Sensitivity inversions highlight 626 627 the importance of high-resolution satellite-based nested inversions in accurately estimating CH₄ emissions, especially in reducing the impact of boundary condition errors on emission inversions. 628





629 Top-down inversions usually result in lower emissions compared to those constructed by bottom-up methods. When comparing our posterior emissions with other inventories, our estimates were the 630 lowest among both bottom-up and top-down estimations. On average, they were 22.0% lower than 631 632 bottom-up inventories and 16.6% lower than previous top-down estimates. The high-resolution 633 simulations employed in this study are capable of more accurately simulating local high concentration values, and thus, the reduction in emissions is greater than that in previous studies. The overestimation 634 635 of China's CH₄ emissions may have led to an overestimation of the country's climate change mitigation burden. The more accurate emission estimates presented here not only enhance our understanding of 636 637 the CH₄ budget but also contribute to more effective global climate change mitigation efforts. 638 Data availability 639 640 The TROPOMI XCH₄ data is publicly available for download at the Copernicus Data Space Ecosystem (https://browser.dataspace.copernicus.eu/, last access: August 5, 2024). The observations used for 641 642 evaluation and the posterior emissions and simulations produced in this study can be accessed at https://doi.org/10.5281/zenodo.15602944 (Feng, 2025). 643 644 **Author contribution** 645 SF, FJ and YZ conceived and designed the research. SF developed the RegGCAS-CH₄ system, 646 analyzed data, and prepared the paper. SW evaluated the CH₄ simulation in Shanxi Province. HC, HZ, 647 SB, HW, and WJ reviewed and commented on the paper. 648 649 **Competing interests** 650 At least one of the (co-)authors is a member of the editorial board of Atmospheric Chemistry and 651

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References

663

- 665 Agustí-Panareda, A., Barré, J., Massart, S., Inness, A., Aben, I., Ades, M., Baier, B. C., Balsamo, G.,
- Borsdorff, T., Bousserez, N., Boussetta, S., Buchwitz, M., Cantarello, L., Crevoisier, C., Engelen,
- R., Eskes, H., Flemming, J., Garrigues, S., Hasekamp, O., Huijnen, V., Jones, L., Kipling, Z.,
- Langerock, B., McNorton, J., Meilhac, N., Noël, S., Parrington, M., Peuch, V. H., Ramonet, M.,
- Razinger, M., Reuter, M., Ribas, R., Suttie, M., Sweeney, C., Tarniewicz, J., and Wu, L.: Technical
- note: The CAMS greenhouse gas reanalysis from 2003 to 2020, Atmos. Chem. Phys., 23, 3829-
- 671 3859, 10.5194/acp-23-3829-2023, 2023.
- 672 Alexe, M., Bergamaschi, P., Segers, A., Detmers, R., Butz, A., Hasekamp, O., Guerlet, S., Parker, R.,
- Boesch, H., Frankenberg, C., Scheepmaker, R. A., Dlugokencky, E., Sweeney, C., Wofsy, S. C.,
- and Kort, E. A.: Inverse modelling of CH₄ emissions for 2010–2011 using different satellite
- 675 retrieval products from GOSAT and SCIAMACHY, Atmos. Chem. Phys., 15, 113-133,
- 676 10.5194/acp-15-113-2015, 2015.
- 677 Bai, S., Zhang, Y., Li, F., Yan, Y., Chen, H., Feng, S., Jiang, F., Sun, S., Wang, Z., Zhou, C., Zhou, W.,
- and Zhao, S.: High-resolution satellite estimates of coal mine methane emissions from local to
- 679 regional scales in Shanxi, China, Science of The Total Environment, 950, 175446,
- 680 10.1016/j.scitotenv.2024.175446, 2024.
- 681 Bergamaschi, P., Houweling, S., Segers, A., Krol, M., Frankenberg, C., Scheepmaker, R. A.,
- Dlugokencky, E., Wofsy, S. C., Kort, E. A., Sweeney, C., Schuck, T., Brenninkmeijer, C., Chen,
- H., Beck, V., and Gerbig, C.: Atmospheric CH₄ in the first decade of the 21st century: Inverse
- modeling analysis using SCIAMACHY satellite retrievals and NOAA surface measurements,
- Journal of Geophysical Research: Atmospheres, 118, 7350-7369, 10.1002/jgrd.50480, 2013.
- Bloom, A. A., Bowman, K. W., Lee, M., Turner, A. J., Schroeder, R., Worden, J. R., Weidner, R. J.,
- 687 McDonald, K. C., and Jacob, D. J.: CMS: Global 0.5-deg Wetland Methane Emissions and





- Uncertainty (WetCHARTs v1.3.1), in, ORNL Distributed Active Archive Center, 2021.
- 689 Bruhwiler, L., Dlugokencky, E., Masarie, K., Ishizawa, M., Andrews, A., Miller, J., Sweeney, C., Tans,
- 690 P., and Worthy, D.: CarbonTracker-CH₄: an assimilation system for estimating emissions of
- atmospheric methane, Atmos. Chem. Phys., 14, 8269-8293, 10.5194/acp-14-8269-2014, 2014.
- 692 Byun, D., and Schere, K. L.: Review of the governing equations, computational algorithms, and other
- components of the models-3 Community Multiscale Air Quality (CMAQ) modeling system,
- 694 Applied Mechanics Reviews, 59, 51-77, 10.1115/1.2128636, 2006.
- 695 Chen, Z., Jacob, D. J., Nesser, H., Sulprizio, M. P., Lorente, A., Varon, D. J., Lu, X., Shen, L., Qu, Z.,
- Penn, E., and Yu, X.: Methane emissions from China: a high-resolution inversion of TROPOMI
- satellite observations, Atmos. Chem. Phys., 22, 10809-10826, 10.5194/acp-22-10809-2022, 2022.
- 698 Evensen, G.: Sequential data assimilation with a nonlinear quasi-geostrophic model using Monte Carlo
- methods to forecast error statistics, Journal Of Geophysical Research-Oceans, 99, 10143-10162,
- 700 10.1029/94jc00572, 1994.
- 701 Feng, S.: CO Emissions Inferred From Surface CO Observations Over China in December 2013 and
- 702 2017, Journal of Geophysical Research-Atmospheres, 125, 10.1029/2019jd031808, 2020.
- 703 Feng, S.: Anthropogenic CH₄ Emissions over China in 2022 Inverted Using TROPOMI XCH4
- 704 Retrievals [Data set]. Zenodo. 10.5281/zenodo.15602944, 2025.
- Feng, S., Jiang, F., Wu, Z., Wang, H., Ju, W., and Wang, H.: CO Emissions Inferred From Surface CO
- Observations Over China in December 2013 and 2017, Journal of Geophysical Research-
- 707 Atmospheres, 125, 10.1029/2019jd031808, 2020.
- 708 Feng, S., Jiang, F., Wang, H., Shen, Y., Zheng, Y., Zhang, L., Lou, C., and Ju, W.: Anthropogenic
- 709 emissions estimated using surface observations and their impacts on PM2.5 source apportionment
- 710 over the Yangtze River Delta, China, Science of The Total Environment, 828, 154522,
- 711 10.1016/j.scitotenv.2022.154522, 2022.
- 712 Feng, S., Jiang, F., Wu, Z., Wang, H., He, W., Shen, Y., Zhang, L., Zheng, Y., Lou, C., Jiang, Z., and
- 713 Ju, W.: A Regional multi-Air Pollutant Assimilation System (RAPAS v1.0) for emission estimates:
- 714 system development and application, Geosci. Model Dev., 16, 5949-5977, 10.5194/gmd-16-
- 715 5949-2023, 2023.
- 716 Feng, S., Jiang, F., Wang, H., Liu, Y., He, W., Wang, H., Shen, Y., Zhang, L., Jia, M., Ju, W., and Chen,
- 717 J. M.: China's Fossil Fuel CO₂ Emissions Estimated Using Surface Observations of Coemitted
- 718 NO2, Environmental Science & Technology, 58, 8299-8312, 10.1021/acs.est.3c07756, 2024.





- 719 Gao, J., Guan, C., and Zhang, B.: China's CH₄ emissions from coal mining: A review of current bottom-
- 720 up inventories, Science of The Total Environment, 725, 138295, 10.1016/j.scitotenv.2020.138295,
- 721 2020.
- 722 Gao, J., Guan, C., Zhang, B., and Li, K.: Decreasing methane emissions from China's coal mining with
- rebounded coal production, Environmental Research Letters, 16, 124037, 10.1088/1748-
- 724 9326/ac38d8, 2021.
- 725 Gaspari, G., and Cohn, S. E.: Construction of correlation functions in two and three dimensions,
- 726 Quarterly Journal of the Royal Meteorological Society, 125, 723-757, 10.1256/smsqj.55416, 1999.
- 727 GMP: Global methane Pledge, https://www.globalmethanepledge.org/resources/global-methane-
- 728 pledge, last access: 12 Febrary 2025, 2023.
- 729 Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert, J. J.,
- 730 Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N., Kurokawa, J. I., Li,
- 731 M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P. R., and Zhang, Q.: Historical (1750–2014)
- 732 anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data
- 733 System (CEDS), Geosci. Model Dev., 11, 369-408, 10.5194/gmd-11-369-2018, 2018.
- 734 Houtekamer, P. L., and Mitchell, H. L.: A sequential ensemble Kalman filter for atmospheric data
- 735 assimilation, Monthly Weather Review, 129, 123-137, 10.1175/1520-
- 736 0493(2001)129<0123:asekff>2.0.co;2, 2001.
- 737 Hu, C., Zhang, J., Qi, B., Du, R., Xu, X., Xiong, H., Liu, H., Ai, X., Peng, Y., and Xiao, W.: Global
- 738 warming will largely increase waste treatment CH₄ emissions in Chinese megacities: insight from
- 739 the first city-scale CH₄ concentration observation network in Hangzhou, China, Atmos. Chem.
- 740 Phys., 23, 4501-4520, 10.5194/acp-23-4501-2023, 2023.
- 741 IEA: Methane Tracker 2021, https://www.iea.org/reports/methane-tracker-2021, last access: 5 April
- 742 2025, 2021.
- Jamali, H., Livesley, S. J., Dawes, T. Z., Cook, G. D., Hutley, L. B., and Arndt, S. K.: Diurnal and
- seasonal variations in CH₄ flux from termite mounds in tropical savannas of the Northern Territory,
- 745 Australia, Agricultural and Forest Meteorology, 151, 1471-1479,
- 746 10.1016/j.agrformet.2010.06.009, 2011.
- 747 Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Muntean, M., Schaaf, E., Dentener, F.,
- 748 Bergamaschi, P., Pagliari, V., Olivier, J. G. J., Peters, J. A. H. W., van Aardenne, J. A., Monni, S.,
- Doering, U., Petrescu, A. M. R., Solazzo, E., and Oreggioni, G. D.: EDGAR v4.3.2 Global Atlas
- of the three major greenhouse gas emissions for the period 1970–2012, Earth Syst. Sci. Data, 11,





- 751 959-1002, 10.5194/essd-11-959-2019, 2019.
- 752 Jiang, F., Wang, H., Chen, J. M., Ju, W., Tian, X., Feng, S., Li, G., Chen, Z., Zhang, S., Lu, X., Liu, J.,
- Wang, H., Wang, J., He, W., and Wu, M.: Regional CO₂ fluxes from 2010 to 2015 inferred from
- 754 GOSAT XCO₂ retrievals using a new version of the Global Carbon Assimilation System, Atmos.
- 755 Chem. Phys., 21, 1963-1985, 10.5194/acp-21-1963-2021, 2021.
- 756 Kou, X., Peng, Z., Han, X., Li, J., Qin, L., Zhang, M., Parker, R. J., and Boesch, H.: China's methane
- 757 emissions derived from the inversion of GOSAT observations with a CMAQ and EnKS-based
- 758 regional data assimilation system, Atmospheric Pollution Research, 16, 102333,
- 759 10.1016/j.apr.2024.102333, 2025.
- 760 Liang, R., Zhang, Y., Chen, W., Zhang, P., Liu, J., Chen, C., Mao, H., Shen, G., Qu, Z., Chen, Z., Zhou,
- 761 M., Wang, P., Parker, R. J., Boesch, H., Lorente, A., Maasakkers, J. D., and Aben, I.: East Asian
- methane emissions inferred from high-resolution inversions of GOSAT and TROPOMI
- observations: a comparative and evaluative analysis, Atmos. Chem. Phys., 23, 8039-8057,
- 764 10.5194/acp-23-8039-2023, 2023.
- 765 Lin, X., Zhang, W., Crippa, M., Peng, S., Han, P., Zeng, N., Yu, L., and Wang, G.: A comparative study
- of anthropogenic CH₄ emissions over China based on the ensembles of bottom-up inventories,
- 767 Earth Syst. Sci. Data, 13, 1073-1088, 10.5194/essd-13-1073-2021, 2021.
- 768 Liu, G., Peng, S., Lin, X., Ciais, P., Li, X., Xi, Y., Lu, Z., Chang, J., Saunois, M., Wu, Y., Patra, P.,
- 769 Chandra, N., Zeng, H., and Piao, S.: Recent Slowdown of Anthropogenic Methane Emissions in
- 770 China Driven by Stabilized Coal Production, Environmental Science & Technology Letters, 8,
- 771 739-746, 10.1021/acs.estlett.1c00463, 2021.
- 772 Lorente, A., Borsdorff, T., Martinez-Velarte, M. C., Butz, A., Hasekamp, O. P., Wu, L., and Landgraf,
- J.: Evaluation of the methane full-physics retrieval applied to TROPOMI ocean sun glint
- 774 measurements, Atmos. Meas. Tech., 15, 6585-6603, 10.5194/amt-15-6585-2022, 2022.
- 775 Lu, X., Jacob, D. J., Zhang, Y., Shen, L., Sulprizio, M. P., Maasakkers, J. D., Varon, D. J., Qu, Z., Chen,
- 776 Z., Hmiel, B., Parker, R. J., Boesch, H., Wang, H., He, C., and Fan, S.: Observation-derived 2010-
- 777 2019 trends in methane emissions and intensities from US oil and gas fields tied to activity metrics,
- 778 Proceedings of the National Academy of Sciences, 120, e2217900120,
- 779 doi:10.1073/pnas.2217900120, 2023.
- 780 Lu, Y. Y., Zhang, H. D., Zhou, Z., Ge, Z. L., Chen, C. J., Hou, Y. D., and Ye, M. L.: Current Status and
- 781 Effective Suggestions for Efficient Exploitation of Coalbed Methane in China: A Review, Energy
- 782 & Fuels, 35, 9102-9123, 10.1021/acs.energyfuels.1c00460, 2021.





- Maasakkers, J. D., Jacob, D. J., Sulprizio, M. P., Scarpelli, T. R., Nesser, H., Sheng, J. X., Zhang, Y.,
- Hersher, M., Bloom, A. A., Bowman, K. W., Worden, J. R., Janssens-Maenhout, G., and Parker,
- 785 R. J.: Global distribution of methane emissions, emission trends, and OH concentrations and
- trends inferred from an inversion of GOSAT satellite data for 2010–2015, Atmos. Chem. Phys.,
- 787 19, 7859-7881, 10.5194/acp-19-7859-2019, 2019.
- 788 MECAP: Methane Emission Control Action Plan,
- 789 https://www.mee.gov.cn/xxgk2018/xxgk/xxgk03/202311/t20231107_1055437.html, last access:
- 790 12 May 2025, 2023.
- 791 Miller, S. M., Michalak, A. M., Detmers, R. G., Hasekamp, O. P., Bruhwiler, L. M. P., and Schwietzke,
- 792 S.: China's coal mine methane regulations have not curbed growing emissions, Nature
- 793 Communications, 10, 303, 10.1038/s41467-018-07891-7, 2019.
- 794 Miyazaki, K., Eskes, H., Sudo, K., Boersma, K. F., Bowman, K., and Kanaya, Y.: Decadal changes in
- 795 global surface NOx emissions from multi-constituent satellite data assimilation, Atmospheric
- 796 Chemistry and Physics, 17, 807-837, 10.5194/acp-17-807-2017, 2017.
- 797 Murguia-Flores, F., Arndt, S., Ganesan, A. L., Murray-Tortarolo, G., and Hornibrook, E. R. C.: Soil
- Methanotrophy Model (MeMo v1.0): a process-based model to quantify global uptake of
- 799 atmospheric methane by soil, Geosci. Model Dev., 11, 2009-2032, 10.5194/gmd-11-2009-2018,
- 800 2018.
- 801 Nesser, H., Jacob, D. J., Maasakkers, J. D., Lorente, A., Chen, Z., Lu, X., Shen, L., Qu, Z., Sulprizio,
- M. P., Winter, M., Ma, S., Bloom, A. A., Worden, J. R., Stavins, R. N., and Randles, C. A.: High-
- resolution US methane emissions inferred from an inversion of 2019 TROPOMI satellite data:
- contributions from individual states, urban areas, and landfills, Atmos. Chem. Phys., 24, 5069-
- 805 5091, 10.5194/acp-24-5069-2024, 2024.
- Nisbet, E. G.: New hope for methane reduction, Science, 382, 1093-1093,
- doi:10.1126/science.adn0134, 2023.
- 808 Oh, Y., Bruhwiler, L., Lan, X., Basu, S., Schuldt, K., Thoning, K., Michel, S. E., Clark, R., Miller, J.
- B., Andrews, A., Sherwood, O., Etiope, G., Crippa, M., Liu, L., Zhuang, Q., Randerson, J., van
- der Werf, G., Aalto, T., Amendola, S., Andra, S. C., Andrade, M., Nguyen, N. A., Aoki, S.,
- Apadula, F., Arifin, I. B., Arnold, S., Arshinov, M., Baier, B., Bergamaschi, P., Biermann, T.,
- Biraud, S. C., Blanc, P.-E., Brailsford, G., Chen, H., Colomb, A., Couret, C., Cristofanelli, P.,
- Cuevas, E., Chmura, L., Delmotte, M., Emmenegger, L., Esenzhanova, G., Fujita, R., Gatti, L.,
- Guerette, E.-A., Haszpra, L., Heliasz, M., Hermansen, O., Holst, J., Di lorio, T., Jordan, A.,
- Jennifer, M.-W., Karion, A., Kawasaki, T., Kazan, V., Keronen, P., Kim, S.-Y., Kneuer, T.,





- 816 Kominkova, K., Kozlova, E., Krummel, P., Kubistin, D., Labuschagne, C., Langenfelds, R.,
- Laurent, O., Laurila, T., Lee, H., Lehner, I., Leuenberger, M., Lindauer, M., Lopez, M., Mahdi,
- R., Mammarella, I., Manca, G., Marek, M. V., Mazière, M. D., McKain, K., Meinhardt, F., Miller,
- 819 C. E., Mölder, M., Moncrieff, J., Moosen, H., Moreno, C., Morimoto, S., Myhre, C. L., Nahas, A.
- 820 C., Necki, J., Nichol, S., O'Doherty, S., Paramonova, N., Piacentino, S., Pichon, J. M., Plass-
- Dülmer, C., Ramonet, M., Ries, L., di Sarra, A. G., Sasakawa, M., Say, D., Schaefer, H., Scheeren,
- 822 B., Schmidt, M., Schumacher, M., Sha, M. K., Shepson, P., Smale, D., Smith, P. D., Steinbacher,
- M., Sweeney, C., Takatsuji, S., Torres, G., Tørseth, K., Trisolino, P., Turnbull, J., Uhse, K.,
- Umezawa, T., Vermeulen, A., Vimont, I., Vitkova, G., Wang, H.-J., Worthy, D., and Xueref-Remy,
- 825 I.: CarbonTracker CH₄ 2023, NOAA Global Monitoring Laboratory, 2023.
- 826 Pandey, S., Houweling, S., Krol, M., Aben, I., Chevallier, F., Dlugokencky, E. J., Gatti, L. V., Gloor,
- 827 E., Miller, J. B., Detmers, R., Machida, T., and Röckmann, T.: Inverse modeling of GOSAT-
- retrieved ratios of total column CH₄ and CO₂ for 2009 and 2010, Atmos. Chem. Phys., 16, 5043-
- 829 5062, 10.5194/acp-16-5043-2016, 2016.
- 830 Pandey, S., Gautam, R., Houweling, S., van der Gon, H. D., Sadavarte, P., Borsdorff, T., Hasekamp,
- O., Landgraf, J., Tol, P., van Kempen, T., Hoogeveen, R., van Hees, R., Hamburg, S. P.,
- Maasakkers, J. D., and Aben, I.: Satellite observations reveal extreme methane leakage from a
- natural gas well blowout, Proceedings of the National Academy of Sciences, 116, 26376-26381,
- 834 doi:10.1073/pnas.1908712116, 2019.
- 835 Peng, S., Lin, X., Thompson, R. L., Xi, Y., Liu, G., Hauglustaine, D., Lan, X., Poulter, B., Ramonet,
- 836 M., Saunois, M., Yin, Y., Zhang, Z., Zheng, B., and Ciais, P.: Wetland emission and atmospheric
- sink changes explain methane growth in 2020, Nature, 612, 477-482, 10.1038/s41586-022-
- 838 05447-w, 2022.
- 839 Peng, S., Giron, C., Liu, G., d'Aspremont, A., Benoit, A., Lauvaux, T., Lin, X., de Almeida Rodrigues,
- H., Saunois, M., and Ciais, P.: High-resolution assessment of coal mining methane emissions by
- satellite in Shanxi, China, iScience, 26, 108375, 10.1016/j.isci.2023.108375, 2023.
- Prather, M. J.: Lifetimes and time scales in atmospheric chemistry, Philosophical Transactions of the
- Royal Society A: Mathematical, Physical and Engineering Sciences, 365, 1705-1726,
- 844 doi:10.1098/rsta.2007.2040, 2007.
- 845 Qin, K., Hu, W., He, Q., Lu, F., and Cohen, J. B.: Individual coal mine methane emissions constrained
- by eddy covariance measurements: low bias and missing sources, Atmos. Chem. Phys., 24, 3009-
- 3028, 10.5194/acp-24-3009-2024, 2024.
- 848 Qu, Z., Jacob, D. J., Shen, L., Lu, X., Zhang, Y., Scarpelli, T. R., Nesser, H., Sulprizio, M. P.,





- Maasakkers, J. D., Bloom, A. A., Worden, J. R., Parker, R. J., and Delgado, A. L.: Global
- distribution of methane emissions: a comparative inverse analysis of observations from the
- TROPOMI and GOSAT satellite instruments, Atmos. Chem. Phys., 21, 14159-14175,
- 852 10.5194/acp-21-14159-2021, 2021.
- 853 Saunois, M., Martinez, A., Poulter, B., Zhang, Z., Raymond, P. A., Regnier, P., Canadell, J. G., Jackson,
- R. B., Patra, P. K., Bousquet, P., Ciais, P., Dlugokencky, E. J., Lan, X., Allen, G. H., Bastviken,
- D., Beerling, D. J., Belikov, D. A., Blake, D. R., Castaldi, S., Crippa, M., Deemer, B. R., Dennison,
- 856 F., Etiope, G., Gedney, N., Höglund-Isaksson, L., Holgerson, M. A., Hopcroft, P. O., Hugelius,
- 6., Ito, A., Jain, A. K., Janardanan, R., Johnson, M. S., Kleinen, T., Krummel, P. B., Lauerwald,
- 858 R., Li, T., Liu, X., McDonald, K. C., Melton, J. R., Mühle, J., Müller, J., Murguia-Flores, F., Niwa,
- Y., Noce, S., Pan, S., Parker, R. J., Peng, C., Ramonet, M., Riley, W. J., Rocher-Ros, G.,
- Rosentreter, J. A., Sasakawa, M., Segers, A., Smith, S. J., Stanley, E. H., Thanwerdas, J., Tian, H.,
- Tsuruta, A., Tubiello, F. N., Weber, T. S., van der Werf, G. R., Worthy, D. E. J., Xi, Y., Yoshida,
- Y., Zhang, W., Zheng, B., Zhu, Q., Zhu, Q., and Zhuang, Q.: Global Methane Budget 2000–2020,
- 863 Earth Syst. Sci. Data, 17, 1873-1958, 10.5194/essd-17-1873-2025, 2025.
- 864 Scarpelli, T. R., Jacob, D. J., Grossman, S., Lu, X., Qu, Z., Sulprizio, M. P., Zhang, Y., Reuland, F.,
- Gordon, D., and Worden, J. R.: Updated Global Fuel Exploitation Inventory (GFEI) for methane
- emissions from the oil, gas, and coal sectors: evaluation with inversions of atmospheric methane
- observations, Atmos. Chem. Phys., 22, 3235-3249, 10.5194/acp-22-3235-2022, 2022.
- 868 Schneising, O., Buchwitz, M., Hachmeister, J., Vanselow, S., Reuter, M., Buschmann, M.,
- Bovensmann, H., and Burrows, J. P.: Advances in retrieving XCH₄ and XCO from Sentinel-5
- Precursor: improvements in the scientific TROPOMI/WFMD algorithm, Atmos. Meas. Tech., 16,
- 871 669-694, 10.5194/amt-16-669-2023, 2023.
- 872 Schuldt, K. N., Mund, J., Aalto, T., Arlyn, A., Apadula, F., Jgor, A., Arnold, S., Baier, B., Bäni, L.,
- Bartyzel, J., Bergamaschi, P., Biermann, T., Biraud, S. C., Pierre-Eric, B., Boenisch, H., Brailsford,
- 6., Brand, W. A., Brunner, D., Bui, T. P. V., Van Den Bulk, P., Francescopiero, C., Chang, C. S.,
- Huilin, C., Lukasz, C., St. Clair, J. M., Sites, C., Coletta, J. D., Colomb, A., Condori, L., Conen,
- 876 F., Conil, S., Couret, C., Cristofanelli, P., Cuevas, E., Curcoll, R., Daube, B., Davis, K. J., Dean-
- Day, J. M., Delmotte, M., Ankur, D., DiGangi, E., DiGangi, J. P., Van Dinther, D., Elkins, J. W.,
- Elsasser, M., Emmenegger, L., Fischer, M. L., Forster, G., Frumau, A., Fuente-Lastra, M.,
- 6879 Galkowski, M., Gatti, L. V., Gehrlein, T., Gerbig, C., Francois, G., Gloor, E., Goto, D., Hammer,
- 880 S., Hanisco, T. F., Haszpra, L., Hatakka, J., Heimann, M., Heimann, M., Heliasz, M., Heltai, D.,
- Henne, S., Hensen, A., Hermans, C., Hermansen, O., Hintsa, E., Hoheisel, A., Holst, J., Di Iorio,
- T., Iraci, L. T., Ivakhov, V., Jaffe, D. A., Jordan, A., Joubert, W., Kang, H.-Y., Karion, A., Kazan,





V., Keeling, R. F., Keronen, P., Kers, B., Jooil, K., Klausen, J., Kneuer, T., Ko, M.-Y., Kolari, P., 883 Kominkova, K., Kort, E., Kozlova, E., Krummel, P. B., Kubistin, D., Kulawik, S. S., Kumps, N., 884 Labuschagne, C., Lan, X., Langenfelds, R. L., Lanza, A., Laurent, O., Laurila, T., Lauvaux, T., 885 Lavric, J., Choong-Hoon, L., Haeyoung, L., Lee, J., Lehner, I., Lehtinen, K., Leppert, R., 886 887 Leskinen, A., Leuenberger, M., Levin, I., Levula, J., Lindauer, M., Lindroth, A., Mikaell Ottosson, 888 L., Loh, Z. M., Lopez, M., Lowry, D., Lunder, C. R., Machida, T., Mammarella, I., Manca, G., 889 Manning, A., Marek, M. V., Marklund, P., Marrero, J. E., Martin, D., Martin, M. Y., Giordane, A. M., Matsueda, H., De Mazière, M., McKain, K., Meinhardt, F., Menoud, M., Jean-Marc, M., 890 Miles, N. L., Miller, C. E., Miller, J. B., Mölder, M., Monteiro, V., Montzka, S., Moore, F., 891 Moossen, H., Morgan, E., Josep-Anton, M., Morimoto, S., Müller-Williams, J., Munro, D., 892 Mutuku, M., Myhre, C. L., Jaroslaw, N., Nichol, S., Nisbet, E., Niwa, Y., Njiru, D. M., Noe, S. 893 894 M., O'Doherty, S., Obersteiner, F., Parworth, C. L., Peltola, O., Peters, W., Philippon, C., Piacentino, S., Pichon, J. M., Pickers, P., Pitt, J., Pittman, J., Plass-Dülmer, C., Platt, S. M., Popa, 895 M. E., Prinzivalli, S., Ramonet, M., Richardson, S. J., Louis-Jeremy, R., Rivas, P. P., Röckmann, 896 T., Rothe, M., Yves-Alain, R., Ju-Mee, R., Santoni, G., Di Sarra, A. G., Sasakawa, M., Scheeren, 897 B., Schmidt, M., Schuck, T., Schumacher, M., Seifert, T., Sha, M. K., Shepson, P., Sloop, C. D., 898 Smith, P. D., Sørensen, L. L., De Souza, R. A. F., Spain, G., Steger, D., Steinbacher, M., Stephens, 899 B., Sweeney, C., Taipale, R., Takatsuji, S., Thoning, K., Timas, H., Torn, M., Trisolino, P., 900 Turnbull, J., Van Der Veen, C., Vermeulen, A., Vimont, I., Viner, B., Vitkova, G., De Vries, M., 901 902 Watson, A., Weiss, R., Weyrauch, D., Wofsy, S. C., Worsey, J., Worthy, D., Xueref-Remy, I., Yates, 903 E. L., Dickon, Y., Yver-Kwok, C., Zaehle, S., Zahn, A., Zazzeri, G., Zellweger, C., and Miroslaw, 904 Z.: Multi-laboratory compilation of atmospheric carbon dioxide data for the period 1983-2022; 905 obspack ch4 1 GLOBALVIEWplus v6.0 2023-12-01, in, NOAA Global 906 Laboratory, 2023. Sheng, J., Song, S., Zhang, Y., Prinn, R. G., and Janssens-Maenhout, G.: Bottom-Up Estimates of Coal 907 Mine Methane Emissions in China: A Gridded Inventory, Emission Factors, and Trends, 908 Environmental Science & Technology Letters, 6, 473-478, 10.1021/acs.estlett.9b00294, 2019. 909 910 Sheng, J., Tunnicliffe, R., Ganesan, A. L., Maasakkers, J. D., Shen, L., Prinn, R. G., Song, S., Zhang, 911 Y., Scarpelli, T., Anthony Bloom, A., Rigby, M., Manning, A. J., Parker, R. J., Boesch, H., Lan, 912 X., Zhang, B., Zhuang, M., and Lu, X.: Sustained methane emissions from China after 2012 despite declining coal production and rice-cultivated area, Environmental Research Letters, 16, 913 104018, 10.1088/1748-9326/ac24d1, 2021. 914 Shi, X., Peng, Y., Wang, S., Zhang, Y., Zhang, J., Song, H., Cui, Y., Sun, F., Liu, H., Xiao, Q., Hu, N., 915

Xiao, W., Griffis, T. J., and Hu, C.: Revising the coal mining CH₄ emission factor based on





- 917 multiple inventories and atmospheric inversion approach at one of the world's largest coal
- 918 production areas: Shanxi province, China, Science of The Total Environment, 965, 178616,
- 919 10.1016/j.scitotenv.2025.178616, 2025.
- 920 Skamarock, W. C., and Klemp, J. B.: A time-split nonhydrostatic atmospheric model for weather
- 921 research and forecasting applications, Journal Of Computational Physics, 227, 3465-3485,
- 922 10.1016/j.jcp.2007.01.037, 2008.
- 923 Soulie, A., Granier, C., Darras, S., Zilbermann, N., Doumbia, T., Guevara, M., Jalkanen, J. P., Keita,
- 924 S., Liousse, C., Crippa, M., Guizzardi, D., Hoesly, R., and Smith, S. J.: Global anthropogenic
- 925 emissions (CAMS-GLOB-ANT) for the Copernicus Atmosphere Monitoring Service simulations
- of air quality forecasts and reanalyses, Earth Syst. Sci. Data, 16, 2261-2279, 10.5194/essd-16-
- 927 2261-2024, 2024.
- 928 Stavert, A. R., Saunois, M., Canadell, J. G., Poulter, B., Jackson, R. B., Regnier, P., Lauerwald, R.,
- Raymond, P. A., Allen, G. H., Patra, P. K., Bergamaschi, P., Bousquet, P., Chandra, N., Ciais, P.,
- 930 Gustafson, A., Ishizawa, M., Ito, A., Kleinen, T., Maksyutov, S., McNorton, J., Melton, J. R.,
- 931 Müller, J., Niwa, Y., Peng, S., Riley, W. J., Segers, A., Tian, H., Tsuruta, A., Yin, Y., Zhang, Z.,
- 22 Zheng, B., and Zhuang, Q.: Regional trends and drivers of the global methane budget, Global
- 933 Change Biology, 28, 182-200, 10.1111/gcb.15901, 2022.
- 934 Steiner, M., Peters, W., Luijkx, I., Henne, S., Chen, H., Hammer, S., and Brunner, D.: European CH₄
- 935 inversions with ICON-ART coupled to the CarbonTracker Data Assimilation Shell, Atmos. Chem.
- 936 Phys., 24, 2759-2782, 10.5194/acp-24-2759-2024, 2024.
- 937 Stohl, A., Aamaas, B., Amann, M., Baker, L. H., Bellouin, N., Berntsen, T. K., Boucher, O., Cherian,
- 938 R., Collins, W., Daskalakis, N., Dusinska, M., Eckhardt, S., Fuglestvedt, J. S., Harju, M., Heyes,
- 939 C., Hodnebrog, Ø., Hao, J., Im, U., Kanakidou, M., Klimont, Z., Kupiainen, K., Law, K. S., Lund,
- 940 M. T., Maas, R., MacIntosh, C. R., Myhre, G., Myriokefalitakis, S., Olivié, D., Quaas, J.,
- 941 Quennehen, B., Raut, J. C., Rumbold, S. T., Samset, B. H., Schulz, M., Seland, Ø., Shine, K. P.,
- 942 Skeie, R. B., Wang, S., Yttri, K. E., and Zhu, T.: Evaluating the climate and air quality impacts of
- 943 short-lived pollutants, Atmos. Chem. Phys., 15, 10529-10566, 10.5194/acp-15-10529-2015, 2015.
- Thanwerdas, J., Saunois, M., Berchet, A., Pison, I., Vaughn, B. H., Michel, S. E., and Bousquet, P.:
- 945 Variational inverse modeling within the Community Inversion Framework v1.1 to assimilate
- 846 δ13C(CH₄) and CH₄: a case study with model LMDz-SACS, Geosci. Model Dev., 15, 4831-4851,
- 947 10.5194/gmd-15-4831-2022, 2022.
- 948 Thompson, R. L., Stohl, A., Zhou, L. X., Dlugokencky, E., Fukuyama, Y., Tohjima, Y., Kim, S.-Y., Lee,
- 949 H., Nisbet, E. G., Fisher, R. E., Lowry, D., Weiss, R. F., Prinn, R. G., O'Doherty, S., Young, D.,





- and White, J. W. C.: Methane emissions in East Asia for 2000-2011 estimated using an
- atmospheric Bayesian inversion, Journal of Geophysical Research: Atmospheres, 120, 4352-4369,
- 952 10.1002/2014JD022394, 2015.
- 953 Thorpe, A. K., Kort, E. A., Cusworth, D. H., Ayasse, A. K., Bue, B. D., Yadav, V., Thompson, D. R.,
- Frankenberg, C., Herner, J., Falk, M., Green, R. O., Miller, C. E., and Duren, R. M.: Methane
- 955 emissions decline from reduced oil, natural gas, and refinery production during COVID-19,
- 956 Environmental Research Communications, 5, 021006, 10.1088/2515-7620/acb5e5, 2023.
- 957 Tsuruta, A., Aalto, T., Backman, L., Hakkarainen, J., van der Laan-Luijkx, I. T., Krol, M. C., Spahni,
- 958 R., Houweling, S., Laine, M., Dlugokencky, E., Gomez-Pelaez, A. J., van der Schoot, M.,
- Langenfelds, R., Ellul, R., Arduini, J., Apadula, F., Gerbig, C., Feist, D. G., Kivi, R., Yoshida, Y.,
- and Peters, W.: Global methane emission estimates for 2000–2012 from CarbonTracker Europe-
- 961 CH₄ v1.0, Geosci. Model Dev., 10, 1261-1289, 10.5194/gmd-10-1261-2017, 2017.
- Tu, Q., Hase, F., Qin, K., Cohen, J. B., Khosrawi, F., Zou, X., Schneider, M., and Lu, F.: Quantifying
- 963 CH₄ emissions from coal mine aggregation areas in Shanxi, China, using TROPOMI observations
- and the wind-assigned anomaly method, Atmos. Chem. Phys., 24, 4875-4894, 10.5194/acp-24-
- 965 4875-2024, 2024.
- 966 Turner, A. J., Jacob, D. J., Wecht, K. J., Maasakkers, J. D., Lundgren, E., Andrews, A. E., Biraud, S.
- 967 C., Boesch, H., Bowman, K. W., Deutscher, N. M., Dubey, M. K., Griffith, D. W. T., Hase, F.,
- 968 Kuze, A., Notholt, J., Ohyama, H., Parker, R., Payne, V. H., Sussmann, R., Sweeney, C., Velazco,
- 969 V. A., Warneke, T., Wennberg, P. O., and Wunch, D.: Estimating global and North American
- 970 methane emissions with high spatial resolution using GOSAT satellite data, Atmos. Chem. Phys.,
- 971 15, 7049-7069, 10.5194/acp-15-7049-2015, 2015.
- 972 van Wees, D., van der Werf, G. R., Randerson, J. T., Rogers, B. M., Chen, Y., Veraverbeke, S., Giglio,
- 973 L., and Morton, D. C.: Global biomass burning fuel consumption and emissions at 500 m spatial
- 974 resolution based on the Global Fire Emissions Database (GFED), Geosci. Model Dev., 15, 8411-
- 975 8437, 10.5194/gmd-15-8411-2022, 2022.
- 976 Varon, D. J., Jacob, D. J., Sulprizio, M., Estrada, L. A., Downs, W. B., Shen, L., Hancock, S. E., Nesser,
- 977 H., Qu, Z., Penn, E., Chen, Z., Lu, X., Lorente, A., Tewari, A., and Randles, C. A.: Integrated
- 978 Methane Inversion (IMI 1.0): a user-friendly, cloud-based facility for inferring high-resolution
- 979 methane emissions from TROPOMI satellite observations, Geosci. Model Dev., 15, 5787-5805,
- 980 10.5194/gmd-15-5787-2022, 2022.
- 981 Wang, F., Maksyutov, S., Tsuruta, A., Janardanan, R., Ito, A., Sasakawa, M., Machida, T., Morino, I.,
- 982 Yoshida, Y., Kaiser, J. W., Janssens-Maenhout, G., Dlugokencky, E. J., Mammarella, I., Lavric, J.





- 983 V., and Matsunaga, T.: Methane Emission Estimates by the Global High-Resolution Inverse
- 984 Model Using National Inventories, Remote Sensing, 11, 2489, 10.3390/rs11212489, 2019.
- 985 Wang, Y., Zhang, Y., Tian, X., Wang, X., Yuan, W., Ding, J., Jiang, F., Jin, Z., Ju, W., Liang, R., Lu, X.,
- 986 Shen, L., Sun, S., Wang, T., Zhang, H., Zhao, M., and Piao, S.: Towards verifying and improving
- 987 estimations of China's CO₂ and CH₄ budgets using atmospheric inversions, National Science
- 988 Review, 12, 10.1093/nsr/nwaf090, 2025.
- 989 Whitaker, J. S., and Hamill, T. M.: Ensemble data assimilation without perturbed observations,
- 990 Monthly Weather Review, 130, 1913-1924, 10.1175/1520-
- 991 0493(2002)130<1913:Edawpo>2.0.Co;2, 2002.
- 992 Wunch, D., Toon, G. C., Blavier, J.-F. L., Washenfelder, R. A., Notholt, J., Connor, B. J., Griffith, D.
- 993 W. T., Sherlock, V., and Wennberg, P. O.: The Total Carbon Column Observing Network,
- 994 Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering
- 995 Sciences, 369, 2087-2112, doi:10.1098/rsta.2010.0240, 2011.
- 996 Zhang, X., Zhou, C., Zhang, Y., Lu, X., Xiao, X., Wang, F., Song, J., Guo, Y., Leung, K. K. M., Cao,
- 997 J., and Gao, M.: Where to place methane monitoring sites in China to better assist carbon
- 998 management, npj Climate and Atmospheric Science, 6, 32, 10.1038/s41612-023-00359-6, 2023.
- 999 Zhang, Y., Jacob, D. J., Lu, X., Maasakkers, J. D., Scarpelli, T. R., Sheng, J. X., Shen, L., Qu, Z.,
- 1000 Sulprizio, M. P., Chang, J., Bloom, A. A., Ma, S., Worden, J., Parker, R. J., and Boesch, H.:
- 1001 Attribution of the accelerating increase in atmospheric methane during 2010–2018 by inverse
- analysis of GOSAT observations, Atmos. Chem. Phys., 21, 3643-3666, 10.5194/acp-21-3643-
- 1003 2021, 2021.
- Zhang, Y., Fang, S., Chen, J., Lin, Y., Chen, Y., Liang, R., Jiang, K., Parker, R. J., Boesch, H.,
- 1005 Steinbacher, M., Sheng, J.-X., Lu, X., Song, S., and Peng, S.: Observed changes in China's
- methane emissions linked to policy drivers, Proceedings of the National Academy of Sciences,
- 1007 119, e2202742119, doi:10.1073/pnas.2202742119, 2022.
- 2008 Zhang, Z., Feng, S., Chen, Y., Liu, Q., Ju, W., Xiao, W., Huang, C., Wang, Y., Wang, H., Jia, M., Wang,
- 1009 X., and Jiang, F.: Development of a regional carbon assimilation system and its application for
- 1010 estimating fossil fuel carbon emissions in the Yangtze River Delta, China, Science of The Total
- Environment, 957, 177720, 10.1016/j.scitotenv.2024.177720, 2024.
- 1012 Zhao, M., Tian, X., Wang, Y., Wang, X., Ciais, P., Jin, Z., Zhang, H., Wang, T., Ding, J., and Piao, S.:
- Slowdown in China's methane emission growth, National Science Review, 11,
- 1014 10.1093/nsr/nwae223, 2024.