

High resolution assessment of coal mining methane emissions by satellite in Shanxi, China

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52 52	Abstract
00 54	ADSIFACI
55	baselines and assessing the effectiveness of mitigation measures. Such an endeavor is
56	ieopardized however by large uncertainties in current CMM estimates. Here, we
57	assimilated images of methane column atmospheric mixing ratios observed by the
58	TROPOMI space borne instrument in a high-resolution regional inversion to estimate
59	CMM emissions in Shanxi, a province representing 15% of the global coal production.
60	The emissions are estimated to be 8.5 ± 0.6 and 8.6 ± 0.6 Tg CH ₄ yr ⁻¹ in 2019 and 2020
61	respectively, close to upper bound of current bottom-up estimates. The monthly variations
62	of emissions are well reproduced, including the drop and rebound in response to COVID-
63	19 regulation. Data from more than a thousand of individual mines indicate that our
64 65	estimated emission factors increase significantly with coal mining depth at prefecture
00 66	nevel, that is the CH4 emission per volume of extracted coal. This result suggests that
67	for mitigation. Our results show robustness of estimating CMM emissions utilizing
68	TROPOMI images and highlight notential of monitoring methane leakages and emissions
69	from satellites.
70	
71	Teaser

72 Atmospheric methane concentration observed by TROPOMI can be applied to detect and 73

quantify coal mine methane emissions.

75 MAIN TEXT

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76

77 Introduction

78 China is the world's largest anthropogenic methane (CH_4) emitter since the 2000s (1, 2). 79 Coal mining is the largest contributor, accounting for 40-45% of China's anthropogenic CH₄ emissions, and ~5% of global anthropogenic emissions (3-5). During the 2000s, CH₄ 80 emissions from coal mining in China increased by 12 Tg CH₄ yr⁻¹ ([6-18] Tg CH₄ yr⁻¹, 81 82 95% confidence interval) (4), contributing 85% and 32% to the increase of China's and 83 global anthropogenic CH₄ emissions, respectively (2-4). To mitigate climate change, it is 84 urgent for China to curb CH₄ emissions, especially in the coal industry. This requires a 85 solid knowledge of current emissions, including the spatial details (e.g., distribution) about 86 the most emitting areas or sites, in order to target the most effective measures and 87 prioritize mitigation actions (6).

88 Both bottom-up and top-down approaches give evidence for an increase in CH₄ emissions 89 from coal mining in China during the 2000s (3, 4, 7, 8). Yet, they disagree in the 90 magnitude of the trend (2, 8-10), ranging from 0.7 to 1.4 Tg CH₄ yr⁻². Furthermore, more 91 recent bottom-up inventory data shows a stabilization of coal methane emissions in the 92 93 2010s, mainly due to a stabilized coal production (2, 4). In contrast, recent top-down 94 estimates suggested either a significant increase in those emissions after 2010 (0.9–1.1 Tg $CH_4 \text{ yr}^{-2}$ (11, 12) or a small increase (0.1–0.3 Tg $CH_4 \text{ yr}^{-2}$) (13, 14). Differences among 95 inversions could be due to the use of different prior emissions, sparse atmospheric CH₄ 96 97 concentrations measurements from surface stations and previous satellites with coarse 98 resolution (e.g., GOSAT), making it difficult to constrain emission hotspots from coal mines (4). Overall, current top-down inversion estimates of the magnitude and trend in 99 coal methane emissions in China are not consistent enough with each other to evaluate or 00 101 improve bottom-up inventories.

102The Sentinel-5P/TROPOMI (TROPOspheric Monitoring Instrument) mission, launched in 103 2017, collects daily images of the CH₄ column mole fractions (XCH₄) at high spatial 04 resolution (5 km \times 7 km) since 2018. The TROPOMI images have been used to detect and 105 quantify large point sources in the oil and gas production sector, including ultra-emitters 106 107(15-17), and regional extraction basins (18). Coal mine emissions from Australia have been recently examined using TROPOMI (19). Here, we focus on coal mining emissions 108 109from China, the largest coal producer of the world, and more specifically from the Shanxi Province. Shanxi represents about 15% of the global coal production, with more than 239 $\lfloor 10 \rfloor$ mines producing more than 0.6 million tons of coal annually (Fig. 1). We assembled a 111 detailed inventory data for 1,012 coal mines in the Shanxi, including mining depth, coal 112113 production and quality types, reported CH₄ ventilation/leak rates, recovery etc ... (see 114 Methods). The Shanxi province is suitable for TROPOMI's monitoring capabilities to assess coal methane emissions using high-resolution regional inversions. We intend here l15 to support the improvement of bottom-up inventories by not only assessing provincial 116 total emissions, but also by identifying groups of mines with the highest emission rates. 117118

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We assimilated the TROPOMI methane column mixing ratio bias corrected level 2 products in a high-resolution regional inversion to estimate CH₄ emissions from coal

122	production in the Shanxi province. The inversion assumes a prior map of emissions based
123	on an annual (thus flat monthly) bottom-up inventory, with many mines but a uniform
124	emission factor (see Methods; PKU-CH ₄ v2) (4). The XCH ₄ plumes generated by grid
125	cells $(0.1^{\circ} \times 0.1^{\circ})$ containing at least one coal mine are simulated by the Hybrid Single-
126	Particle Lagrangian Integrated Trajectory model (HYSPLIT) (20). Emission rates are
127	optimized by minimizing the distance between the modeled and TROPOMI-observed
128	XCH ₄ enhancements within images for each satellite overpass (see Methods). We perform
129	an ensemble of sensitivity tests with inversions to assess the uncertainty of posterior
130	emissions, from TROPOMI XCH4 measurement errors, background estimation method,
131	meteorological data and other key parameters of the inversion (see Methods and
132	Supplementary Text 1-2). To evaluate the impact of the prior emissions choice on
133	posterior emissions, we also perform an inversion with no prior knowledge
134	(Supplementary Text 3). Then we compare bottom-up inventories including EDGAR v6.0,
135	Global Fuel Exploitation Inventory (GFEI v2) and PKU-CH4 v2, with our independent
136	top-down estimate derived from TROPOMI-based inversions. We further hypothesize that
137	the CH ₄ emission factor of coal mining (EF _{coal}) should be related to the depth of mining
138	and the quality of coal that relates to different methane concentration in coal seams (21-
139	23). Taking advantage of the fact that our inversion has a very high resolution and can
140	distinguish between different groups of mines we finally examine the relationships
141	between the EF_{coal} calculated from the inversion, and mining depth / coal types. This
142	analysis provides direct insights to evaluate the bottom-up inventories emission factors.

143 144 **Results**

145 Methane emissions from large coal mines or clusters of mines produce plumes of high XCH₄, which can be detected by TROPOMI (Fig. 2A). Each large XCH₄ enhancement in 146 147 the Shanxi Province is found to be associated with an ultra-emitter or a cluster of highemitters, systematically assessed from 112 images of TROPOMI from 2019 to 2020, 148 selecting only images with more than 30% of valid pixels (see Methods). As expected, the 149 150 averaged XCH₄ enhancement map during 2019-2020 reconstructed using the optimized posterior emissions shows better agreement with the observed one (r=0.83, p<0.001) 151 152compared to the one based on the prior emissions (r=0.42, p<0.001) from the bottom-up inventory of PKU-CH₄ v2 (Fig. 2A-C). Fig. 2D shows that the (spatial) correlation 153 154 coefficients between observed XCH₄ enhancements from TROPOMI images and those 155 reconstructed from posterior emissions are higher than those modeled from prior 156 emissions, with a mean correlation coefficient of 0.71 for posterior emissions compared to 0.31 for prior emissions across the 112 images. Moreover, the posterior emissions capture 157 77% of the magnitude of observed XCH₄ enhancements from the 112 TROPOMI images 158 on average, much higher than the 51% explained by the prior emissions. In order to test 159 the sensitivity of our inversions to meteorological data, parameters and background 160 161 concentration choices for our inversions, we performed an ensemble of inversions for these uncertainties (see Methods and Supplementary Text 1-2). As a result, we found that 162 163 the TROPOMI images can well constrain the methane emissions from coal mines in 64 Shanxi with an uncertainty less than 10% (Figs. 1 and 2). 165

166Even with an agnostic flat monthly prior, our inversion estimates based on TROPOMI167(INV_{TROPOMI}) produce a seasonal variation of monthly coal production in 2019 and 2020168(r=0.54, p=0.015; Fig. 3A). Interestingly, INV_{TROPOMI} detect also a drop and rise of169emissions that corresponds with the Spring Festival of 2019, and a drop of 0.1 Tg CH4170month⁻¹ (14%) reflecting the impact of the COVID-19 outbreak on coal production, which

171decreased by more than 5% in February 2020. A subsequent recovery of production172capacity in the next two months is also apparent in Fig. 3A. Coal mining in Shanxi emitted173 $8.5\pm0.6 (\pm 1\text{-sigma confidence interval}) Tg CH_4 yr^{-1} in 2019 from INV_{TROPOMI} results, and174<math>8.6\pm0.6 Tg CH_4 yr^{-1} in 2020$ respectively (Fig. 3B). The total emissions from INV_{TROPOMI}175are higher than those from PKU-CH4 v2 ($5.8\pm0.5 Tg CH_4 yr^{-1}$) and GFEI v2 ($7.3\pm2.0 Tg$ 176CH4 year^{-1}), but similar to EDGAR v6.0 ($8.8 Tg CH_4 yr^{-1}$).

178Regarding the spatial distribution of emissions inferred by INV_{TROPOMI} within Shanxi, hot-179 spots are detected in most grids that contain high coal production mines (Figs. 1 and 3C). However, INV_{TROPOMI} retrieves additional emission hotspots that were missing in our 180181 priori emission map (e.g., high-emissions grids in the southwest; Fig. 3). In contrast to the 182 agnostic inversion with no prior, the spatial distribution of emissions from coal mines in EDGAR v6.0, GFEI v2 and PKU-CH₄ v2 depends on activity data such as mine locations 183 184 and production and emission factors. The three bottom-up inventories have different spatial patterns, but they all miss the high-emission hotpots found by the inversion in the 185 southwest, representing 10% of total emissions (Fig. 3D-F). We found that EDGAR v6.0, 186 GFEI v2 and PKU-CH₄ v2 used an outdated coal production map from around 2011 to 187 188 spatialize the total province-level emissions (from the product of an uniform emission factor and total coal production at province-level) into each grid cell of their emission 189 190 maps (2, 24, 25). Some coal mines closed while new coal mines have opened since 2011 (26), which could explain the bias in the spatial distribution of CH₄ emissions from coal 191 mines in the bottom-up inventories, compared to INV_{TROPOMI}. The INV_{TROPOMI} optimized 192 CH₄ emission map thus gives insights on local hotspots and how their emissions change at 193 monthly or sub-monthly scale, although 4 out of 24 months are not covered by the 194 195 inversion due to none or only one image available (fig. S1), which should be further be solved by future satellite missions (16). This method can help improving not only the total 196 197 emission of the basin, but also the spatial distribution map of coal emissions, and provides timely, updated estimates to evaluate bottom-up inventories. 198

Combining INV_{TROPOMI} estimate emissions at $0.1^{\circ} \times 0.1^{\circ}$ with coal production data 200 available at prefecture-level in 2019 and 2020, we deduced an inversion-based average 201 202 emission factor of coal methane emission (EF_{coal}) for each of the ten prefectures of Shanxi 203 (see Methods; Fig. 4). This inversion-based EF_{coal} varies by more than one order of magnitude between prefectures, from $0.9 \text{ m}^3 \text{ t}^{-1}$ in Shouzhou to 17.3 m³ t⁻¹ in Yangquan. 204 Across the ten prefectures, this EF_{coal} shows marginally significant correlation with that 205 206 from the ground inventory (r=0.57, p=0.083; fig. S2). That inventory was conducted in 207 2011 and estimated potential fugitive emissions from underground mining without taking 208 account of coal methane utilization that already has been adopted in Shanxi (the 209 INV_{TROPOMI} assesses actual coal methane emissions including their fractional reduction from methane utilization/recovery), which explains the smaller EF_{coal} in $INV_{TROPOMI}$ (fig. 210 S2). In addition, China's energy reforms in the past decade led to the reorganization and 211212 closure of some Shanxi coal mines with large emission and/or low methane utilization rates (27). The large variation of EF_{coal} between prefectures highlights again that the use of 213 214 the same EF_{coal} for all types of mines in a region, as done in bottom-up inventories (e.g., 215 EDGAR v6.0, PKU-CH₄ v2), could lead to strong biases in the distribution of coal CH₄ 216 emissions. 217

The large spatial variations in EF_{coal} found between prefectures is expected to be related to coal rank and mining depth (21). For example, in the northern Shanxi where the coal seams in Datong, Shuozhou and Xinzhou are shallow, and mostly weakly caking coal and

221 gas coal with low metamorphic degree, we found low EF_{coal} values (Fig. 4A). On the 222 contrary, in Yangquan where the coal is mainly deep anthracite, we found the highest 223 EF_{coal} . As the degree of metamorphism of anthracite increases, methane in the interstices 224 of coal seams could decrease, which may explain the lower EF_{coal} in Shuozhou (mainly 225 gas coal) than in Yangquan (mainly anthracite). 226

227 The gas pressure of coal seam increases with depth, and so does the volume of methane 228 contained in coal. Thus, depth-specific EF_{coal} were suggested by the Intergovernmental Panel on Climate Change (IPCC) methodology (28, 29), with default EF_{coal} of 10 m³ t⁻¹, 229 18 m³ t⁻¹, and 25 m³ t⁻¹ for mines with depth less than 200m, 200-400m, and deeper than 230 231 400 m respectively. Among different coal ranks (coalification), a higher rank coal 232 generally has a higher methane content (23), and thus accounts for a steeper increase in 233 EF_{coal} with depth (21, 22). Almost 99% of the coal is produced from underground mines in 234 Shanxi. Mining depth shows large spatial variations from less than 100 m to more than 235 700 m (fig. S3), leading to large spatial variations in EF_{coal}. We thus regressed the EF_{coal} derived from INV_{TROPOMI} against the average mining depth (from the year 2011) for each 236 237 prefecture, and found a high correlation (r=0.88, p=0.005) when excluding Changzhi and 238 Jinzhong prefectures which are outliers (Fig. 4B). The EF_{coal} in these two prefectures 239 could be explained by substantial subsidence of coal seams in Jinzhong and Changzhi in 240 the tectogenesis after the Carboniferous-Permian when coal was formed (e.g., depth of 241 600 m coal seam in Jinzhong similar as ~400 m coal seam in Changzhi or Linfen) (30, 31). The relationship shown in Fig. 4B suggests that EF_{coal} in these eight prefectures of 242 Shanxi increases by 9.4 m³ t⁻¹ for a 100 m increase in the mining depth. A similar 243 empirical function between coal mining depth and EF_{coal} was included in the inventory of 244 2011 across the eight prefectures (r=0.67, p=0.07; fig. S4) (22). However, at the coal mine 245 scale, this relationship could be weakened by the variability of coal ranks in different coal 246 247 mines (fig. S3B). 248

249

250 **Discussion**

251To analyze the uncertainty of our posterior emissions, we performed an ensemble of tests 252adding perturbations to the parameter values and input data (see Methods and 253 Supplementary Text 1-2). We found that the relative standard deviation of posterior 254 emissions due to errors in XCH₄ retrievals (1.4%), the uncertain release duration as 255 defined by the travel time between source locations and observed XCH₄ enhancements (15%), and the uncertainty of atmospheric transport (12%) are all relatively small 256 257compared to the standard deviation due to the uncertainty in background XCH₄ estimates (38%, fig. S5). This suggests that the choice of background XCH₄ for calculating the 258 259XCH₄ enhancement is the most important parameter to accurately estimate CH₄ emissions. 260 To test whether prior emission input affects the posterior emissions, we ran inversions without any prior knowledge (zero emission for each grid in Shanxi) and used a ridge 261 262 regression to regularize the inverse problem (Supplementary Text 3). This sensitivity test 263 gave similar total emissions as the Bayesian inversion with prior estimates from PKU-CH₄ v2, but with lower emissions in the north and higher emissions in the middle of Shanxi 264 265(figs. S6 and S7). On average, there are two to six images per month (some months have 266 11-12 images) from TROPOMI fulfilling our quality filter with >30% of valid pixels (fig. S1), but for some months no image was available (e.g., June in 2019 and 2020). Future 267 268 satellite missions (e.g., MethaneSAT) (32) complementary with TROPOMI would help 269better constrain the seasonal variations in emissions shown in Fig. 3A. In addition, coal

mines field campaigns for CMM emissions during the days when high-resolution XCH₄
 images from satellites are available can further be used to evaluate top-down inversions in
 the future.

273 Overall, we show that successive TROPOMI images of XCH₄ can constrain monthly CH₄ 274 275 emissions from coal mining in Shanxi well, with annual emission estimates of 8.5±0.6 Tg CH_4 yr⁻¹ and 8.6±0.6 Tg CH_4 yr⁻¹ in 2019 and 2020 respectively. The top-down inversion 276277 with TROPOMI XCH₄ suggests that the use of a weighted average emission factor from 278 the ground inventory dataset established in 2011 underestimates CH₄ emissions from coal mining (4, 25). We further find that deeper mining entails larger emission factors, as 279 280 shown across the prefectures in Shanxi, suggesting that emission factors are likely to 281 increase if mining deepens in the future. In addition, province-level emission factor used 282 in bottom-up inventories may smooth the spatial variation of emissions due to the 283 variation of mining depth and coal types across the mines. As the TROPOMI-based 284 inversion used in our study can capture seasonal variation of coal mining activity 285 (especially the drop in emissions corresponding to the coal production decrease in 286 February, 2020 after the outbreak of COVID-19), a near real-time emission map can be 287 updated on a regular basis. Such a tool could help monitoring and verification of 288 emissions as well as supporting mitigation toward climate neutrality targets.

289

290 Materials and Methods

291 Satellite-based XCH₄ retrievals from TROPOMI

292 We collected bias-corrected methane column mixing ratios derived from the spaceborne 293 instrument TROPOMI on board of the Sentinel 5P satellite (ESA products of S5P L2 CH4 294 OFFLINE) (33). TROPOMI is an imaging spectrometer, orbiting the earth in near-polar, 295 sun-synchronous trajectories with a mean local solar time ascending node at 13:30. 296 TROPOMI's swath is approximately 2600 km wide, achieving near-global coverage on a 297 daily basis. Our study relies on TROPOMI measurements from January 2019 to December 298 2020, with images collected over the Shanxi Province shape, and re-projected on a 299 $0.1^{\circ} \times 0.1^{\circ}$ regular grid using the GDAL library (34). As cloud cover, high solar zenith angle, high viewing zenith angle, large terrain roughness or small surface albedo etc. 300 301 induce substantial bias in XCH_4 retrievals (33), we only used pixels with a quality assurance value (qa value = 1). TROPOMI images were filtered to keep only those with 302 more than 30% of valid pixels (qa value =1). With this filter, 58 and 54 images were 303 304 selected per year in 2019 and 2020 respectively. Uneven albedo and aerosol optical thickness are amongst the parameters influencing the quality and accuracy of TROPOMI 305 XCH₄ retrievals. Although the data product we used in this study includes a correction 306 based on surface albedo in the SWIR domain (35), we also assessed the reliability of 307 308 TROPOMI XCH₄ retrievals by evaluating its correlation with surface shortwave infrared albedo (Albedo) and aerosols optical thickness (AOD) (fig. S8). When using only high-309 quality pixels (qa value = 1), R^2 scores of linear regressions between bias-corrected XCH₄ 310 and Albedo (R²=0.010) and AOD (R²=0.006) are quite low, hence significant bias of 311 XCH₄ from albedo or aerosols can be dismissed. When including medium-quality pixels 312 (qa value > 0.4), the R² scores are estimated respectively at 0.007 and 0.130 for albedo 313 and AOD, confirming that the albedo and aerosols has limited impact on XCH₄ retrievals 314 315 in our region of interest.

316

Bottom-up inventories for coal CH₄ emissions

Three bottom-up inventories for annual methane emissions from coal mining in the Shanxi 318 319 province were used in this study: PKU-CH₄ v2 (4), EDGAR v6.0 (2) and GFEI v2 (24). PKU-CH₄ v2 was updated up to 2020 using annual coal production in 2020 from the latest 320 Statistic Yearbook, following the bottom-up methodology in Liu et al. (2021) (4). The 321 322 annual maps from PKU-CH₄ v2 were used as prior for our top-down inversion. For the 323 EDGAR v6.0, we used the monthly gridded emissions of China in 2018, and then 324 calculated the total emission as the sum of all grids in Shanxi province. We then scaled the 325 emissions in 2018 from EDGAR v6.0 into emissions in 2019 by using the ratio of coal 326 production of Shanxi in 2019 and 2018. This estimate is referred as EDGAR v6.0. The inventory of GFEI v2 gives the annual emissions that incorporate national reports of China 327 328 to the United Nations Framework Convention on Climate Change (UNFCCC) and 329 allocates the total national emissions into infrastructure locations from Sheng et al. (2019) (25) with a $0.1^{\circ} \times 0.1^{\circ}$ spatial resolution (24). We used annual emissions of 2019 from 330 331 GFEI v2 here, updated with IPCC emission factors and yearly activity data from US Energy Information Administration (24, 36). To validate seasonal variation of top-down 332 monthly emissions, the monthly coal production of Shanxi province and yearly coal 333 334 production in prefecture level were collected from Shanxi Statistic Yearbook (37).

Ground inventory from 1012 coal mines in 2011

337 We collected the information of coal mines in Shanxi province publicly available in the ground inventory of the National Coal Mine Methane Level Identification for 2011, by the 338 State Administration of Coal Mine Safety (38). The coal production in Shanxi province 339 increased by $\sim 60\%$ from 2011 to 2020. Emission factors from this inventory have been 340 applied to estimate CH₄ emissions from coal mining in Sheng et al. (2019) (25) and Liu et 341 al. (2021) (4). We collected data from 1012 coal mines, more than 95% coal mines in 342 Shanxi province, including annual coal production (537 coal mines), mining depth (984 343 344 coal mines) and emission factors (727 coal mines) reported in the ground inventory of 2011. The location of these coal mines were derived from Baidu Map Platform 345 (http://api.map.baidu.com/lbsapi/getpoint/index.html). Note that 84 out of the 1012 coal 346 347 mines are excluded in our analysis, as these 84 coal mines have been closed since 2016 348 because of a supply-side structural reform of the coal industry by eliminating low efficiency coal production (27). 349

351 According to the ground inventory and the locations of coal mines, we aggregate the production and average the mining depth, extraction rate and emission factors weighted by 352 the coal production into $0.1^{\circ} \times 0.1^{\circ}$ grid scale in 2011. For the map of coal production in 353 2019 (2020), we scaled the map of 2011 by the ratio of total province-level production in 354 355 2019 (2020) and in 2011. The main types of coal for each prefecture in Shanxi were collected from the dataset in Liu et al. (2015) (39). To compare the EF_{coal} derived from 356 INV_{TROPOMI} in 2019, we adjusted the EF_{coal} derived from ground inventory in 2011 by the 357 composition of coal production mined from low gas mines, high gas mines and outburst 358 359 gas mines in each prefecture between 2019 and 2011 in fig. S3.

TROPOMI-based top-down inversion

362 *Model*

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The estimates are produced using a classical Bayesian regression framework (40, 41). Specifically, we solve the following quadratic programming problem for each useable TROPOMI XCH4 image $min_x \quad J(x) = (y - Kx)^T S_0^{-1} (y - Kx) + \lambda (x - x_P)^T S_p^{-1} (x - x_P)$

s.t. $x \ge 0$

where γ is the methane column mixing ratio enhancement (i.e. methane column mixing 368 369 ratio bias corrected image subtracted from its median value and with negative values 370 clipped to 0); K is the methane dispersion footprints produced using the Hybrid Single-371 Particle Lagrangian Integrated Trajectory model (HYSPLIT) (20); x_P is the prior emission rates vector (all $0.1^{\circ} \times 0.1^{\circ}$ regular grid cells); S₀ is the covariance matrix for 372 observational error; S_p is the covariance matrix for prior error (40, 42). 373 374 If we omit the non-negativity constraint $x \ge 0$, this quadratic program can be solved in 375 closed form by $x = x_{P} + \lambda^{-1}S_{P}K^{T}(\lambda^{-1}K S_{P}K^{T} + S_{O})^{-1}(y - Kx_{P})$ 376 and a condition number of this problem can be computed to assess the stability and 377 robustness of the solution. This condition number is defined as $\kappa(\lambda) = \frac{\sigma_{max}(\tilde{S}_{\lambda})}{\sigma_{min}(\tilde{S}_{\lambda})}$ with 378 σ_{max} (resp. σ_{min}) denoting the highest (resp. smallest) singular value of \tilde{S}_{λ} = 379 $(K^T S_0^{-1} K + \lambda S_P^{-1})^{-1}$. We add the non-negativity constraint for x and solve the 380 quadratic program numerically. This constraint regularizes the solution and limits overfit. 381 382 λ is used to scale the relative weights of the TROPOMI XCH4 image and prior terms. It 383 should ideally be equal to 0 or very small. Yet for several dates, the quadratic program is 384 ill-conditioned due to the sparsity of the TROPOMI XCH4 image and near-colinearity of 385 some HySplit footprints ($\kappa(\lambda) \gg 0$). We ensure that the estimates for each date are 386 produced by a well-conditioned minimization program by incrementing $\lambda \in [10^{-2}, 10^2]$ on a log_{10} scale up to the smallest value such that $\kappa(\lambda) < \tau$. We set $\tau = 10^3$ based on 387 388 the criterions explained in Supplementary Text 1.

Observational and prior error covariance matrices

391 S_{0} is computed using the relative residual error method (42). In particular, we split our work domain into a $2^{\circ} \times 2^{\circ}$ grid and compute the standard deviation matrix of the residual 392 393 error in the bias corrected subtraction of observed methane concentrations by simulated 394 concentrations (derived from HySplit simulations and prior PKU-CH₄ v2 emission rates). 395 S_0 is defined as the normalized, relative standard deviation matrix and, by construction, it 396 accounts for both the sensor error and the model error. S_P is defined as the absolute error 397 between the gridded inventories PKU-CH₄ v2 and EDGAR v6.0, re-projected on the same 398 $0.1^{\circ} \times 0.1^{\circ}$ regular grid as the images. Both S_{O} and S_{P} are normalized so that their relative weight in the objective function J is fully controlled by the parameter λ . In addition, very 399 100 small diagonal values of S_P are set at 1/10 of its maximal value (i.e. 0.1 after minmax 101 normalization) to avoid ill-conditioning that would occur when EDGAR v6.0 and PKU-102 CH4 v2 have very similar values. In more than 97% of the daily inversions, the condition number criterion yields $\lambda \in \{10^{-2}, 10^{-1}, 1\}$, hence giving to the observational term a 103 104 greater of equal weight with respect to the prior penalization term.

HySplit simulations

107 Each grid pixel with a positive methane emissions value in the PKU-CH4 v2 inventory is considered as a potential source from which we simulate methane plumes using the 108109 Lagrangian particle dispersion model HySplit. These simulations are executed in **110** concentration, forward mode on a $0.01^{\circ} \times 0.01^{\circ}$ grid and re-projected on the priors and **1**11 images $0.1^{\circ} \times 0.1^{\circ}$ grid. HySplit parameters are mostly similar to those used in Lauvaux et al. (2022) (16). Particles are released continuously at constant rate (10000 particles per **112 113** hour) from a grid cell; each particle represents a fixed amount of methane spreading 114 horizontally as a gaussian puff with respect to the meteorological fields. The Planetary 115 Boundary Layer in which the particles diffuse vertically is derived from the meteorological data. The release altitude is set at 10 meters, consistent with the fact that 116

389 390

- **117** methane is mainly emitted from coal mines through Ventilation Air Methane (VAM) **118** devices located close to the ground level. Following the analysis of Lauvaux et al. (2022) £19 (16) on the negligible impact of the parameters defining the mixed layer height model and £20 vertical mixing strength, we keep these parameters at default value. These fields come £21 from the Global Data Assimilation System (GDAS) meteorological data produced by the **1**22 National Centers for Environmental Prediction (NCEP) at 1-degree spatial resolution and 123 sampled hourly; they are downloaded from the NOAA FTP server. For sensitivity analysis 124 purposes, we also rely on data from the Global Forecast System (GFS), also produced by 125 NCEP at $0.25^{\circ} \times 0.25^{\circ}$ spatial and hourly temporal resolutions available on the same 126 NOAA FTP server.
- Release is set to start 10 hours before TROPOMI overpass time, assuming that the
 simulated plumes have reached a steady state at sensing time. This hypothesis is discussed
 and supported in Supplementary Text 2. HySplit plumes thereby obtained are normalised
 to produce footprints which play the same role as the Jacobian matrix derived from
 GEOS-FP data in Zhang et al. (2020) (40), Shen et al. (2021) (41).

Uncertainty

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134 Uncertainty in the methane emission estimate stems from several parameters, modelling 135 choices and data-induced constraints. To account for the modelling uncertainty, we 136 perform an ensemble of inversion with perturbations of the parameters and input data. 137 Notably, we evaluate the standard deviation of the methane emission estimates with 138 respect to variations in meteorological data, simulation duration and methane background estimation process, respectively denoted σ_w , σ_d , σ_b . More details on the ensemble 139 140 sensitivity analysis are given in Supplementary Text 2. We also propagate the TROPOMI 141 error, as provided by the precision data product (43), to derive the sensor measurement 142 error σ_m . We also account for the absence of valid readings for some days (namely sampling uncertainty σ_s) by using a poll setting and deriving σ_s from the unbiased 143 Horvitz-Thompson confidence bounds (see paragraph S2) (44). Assuming independence 144 145 of the uncertainty sources, we finally apply the law of propagation of uncertainty to evaluate the total uncertainty $\sigma = \sqrt{\sigma_w + \sigma_d + \sigma_b + \sigma_m + \sigma_s}$. 146 147

The condition numbers and the degrees of freedom for signal (DOFS; defined as $trace(I - \tilde{S}_{\lambda} S_p^{-1})$ where I is the identity matrix) of the quadratic programs solved in the inversions are qualitative indicators for the sensitivity of the results produced. These are discussed in the next section.

Validation

154 To assess the validity and robustness of the results, we compute a series of metrics and 155indicators. This includes the distribution of correlations between observed TROPOMI 156 images and prior and posterior reconstructed images (Fig. 2); the correlation between the 157 mean observed image and the mean reconstructed images (Fig. 2D); and the percentage of 158reconstructed methane with respect to observed images (Fig. 2E). On average, the mean 159 image-per-image correlation increases from 0.31 (prior reconstruction) to 0.71 (posterior £60 reconstruction) after the optimization process. The same metrics on averaged images rises 161 from 0.42 to 0.83, hence validating the ability of the model to explain local methane **1**62 enhancements by emissions originating in coal mining areas. The mean posterior 163 percentage of methane reconstructed is 76%, thus indicating that a minor part of the observed methane enhancements is not explained by coal mining emissions. 164

166 S9 and S10). In particular, we show that this criterion leads to λ parameters mostly comprised in $\{10^{-2}, 10^{-1}, 1\}$, hence efficiently improving the conditioning of the £67 quadratic programs without over-penalizing the objective function (which would 168 artificially lower the emissions estimates). Likewise, the distribution of the DOFS of the 169 system has a lower bound at 20 with a mean value at 75. It shows that the quadratic 170 171programs are well-constrained, namely that output of the optimization problem is largely **1**72 influenced by TROPOMI images and not primarily determined by the prior term (fig. **1**73 S10). Figures 2 and 3 reveal a high stability in the estimates, both monthly and hourly, 174 aggregated and on a gridded basis (standard deviation of the monthly estimates is 0.11Mt). 175 This is expected as ventilation air methane systems are set to continuously ventilate coal 176 £77 mines, and it argues for the robustness of the inversion estimates. 178 We show that the results have a very low sensitivity to the construction of the minimization problem. A pure ridge regression setting $(S_0 = I_n, S_p = I_n \text{ and } x_p = 0)$ 179 produces very similar methane emission estimates and validation metrics (see 180 Supplementary Text 3 and figs. S6 and S7). 181 182 183 184 References 185 National Development and Reform Commission of the People's Republic of China. Second 1. 186 National Communication on Climate Change of The People's Republic of China. (UNFCCC, 187 2012). 188 2. Crippa, M., Guizzardi, D., Solazzo, E., Muntean, M., Schaaf, E., Monforti-Ferrario, F., Banja, 189M., Olivier, J.G.J., Grassi, G., Rossi, S., Vignati, E. GHG emissions of all world countries -190 2021 Report, EUR 30831 EN, Publications Office of the European Union, Luxembourg, 2021, 191 ISBN 978-92-76-41547-3, JRC126363. 192 3. S. Peng, S. Piao, P. Bousquet, P. Ciais, B. Li, X. Lin, S. Tao, Z. Wang, Y. Zhang, F. Zhou, **193** Inventory of anthropogenic methane emissions in Mainland China from 1980 to 2010. 194 Atmospheric Chemistry and Physics Discussions, 1-29 (2016). G. Liu, S. Peng, X. Lin, P. Ciais, X. Li, Y. Xi, Z. Lu, J. Chang, M. Saunois, Y. Wu, P. Patra, 195 4. 196 N. Chandra, H. Zeng, S. Piao, Recent Slowdown of Anthropogenic Methane Emissions in 197 China Driven by Stabilized Coal Production. Environmental Science & Technology Letters 8, 739-746 (2021). 198 199 M. Saunois, A. R. Stavert, B. Poulter, P. Bousquet, J. G. Canadell, R. B. Jackson, P. A. 5. 500 Raymond, E. J. Dlugokencky, S. Houweling, P. K. Patra, P. Ciais, V. K. Arora, D. Bastviken, 501 P. Bergamaschi, D. R. Blake, G. Brailsford, L. Bruhwiler, K. M. Carlson, M. Carrol, S. 502 Castaldi, N. Chandra, C. Crevoisier, P. M. Crill, K. Covey, C. L. Curry, G. Etiope, C. 503 Frankenberg, N. Gedney, M. I. Hegglin, L. Höglund-Isaksson, G. Hugelius, M. Ishizawa, A. Ito, G. Janssens-Maenhout, K. M. Jensen, F. Joos, T. Kleinen, P. B. Krummel, R. L. 504 505 Langenfelds, G. G. Laruelle, L. Liu, T. Machida, S. Maksyutov, K. C. McDonald, J.

The condition number criterion is discussed and supported in Supplementary Text 1 (Figs.

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350	inversion with help of A.A., A.B., T.L. and H.A.R G.L. updated PKU-CH ₄ v2 inventory.
351	S.P., C.G. and G.L. performed the analysis and created all the figures. S.P. drafted the
352	manuscript. All authors contributed to commenting and writing on the draft manuscript.
353	
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355	
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357	the Copernicus Open Access Hub (https://scihub.copernicus.eu/). The meteorological
358	reanalysis data used for the forward HYSPLIT simulations are available from the Global
359	Forecast System (GFS), Environmental Modeling Center, National Centers for
360	Environmental Prediction (National Weather Service, NOAA, U.S. Department of
361	Commerce, NCEI DSI 6182, gov.noaa.ncdc:C00634); and from the Global Data
362	Assimilation System (GDAS), Environmental Modeling Center, National Centers for
363	Environmental Prediction (National Weather Service, NOAA, U.S. Department of
364	Commerce, NCEI DSI 6172, gov.noaa.ncdc:C00379). PKU-CH ₄ v2 is available at
365	https://figshare.com/s/b38a368111749f1412be. EDGAR v6.0 is avaiable at
366	https://edgar.jrc.ec.europa.eu/. GFEI v2 is available at
367	https://doi.org/10.7910/DVN/HH4EUM. The inversion method presented in this article is
368	constructed using the HYSPLIT model (v4.2.0; 2019), which was developed by the Air
369	Resources Laboratory at NOAA and is available from www.arl.noaa.gov/hysplit/.
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374	Figures and Tables
575	8
376	Fig. 1. Distribution of coal production in China. Map of the coal production at
377	province-level in 2019 over China (A) and distribution of gridded coal mines with
378	annual production larger than 0.6 million tons year ⁻¹ with a spatial resolution of
379	$0.1^{\circ} \times 0.1^{\circ}$ in the Shanxi Province (B). Note that the distribution of coal mines in
380	the Shanxi Province in (B) is from the ground survey dataset conducted in 2011.
381	but is scaled by the ratio of total coal production between 2011 and 2019 in the
582	Shanxi Province.



Fig. 2. Spatial patterns of XCH₄ enhancement from TROPOMI, reconstructed XCH₄ using prior and posterior emissions, with a spatial resolution of 0.1°×0.1°. (A), averaged XCH₄ enhancement from 112 images of TROPOMI in 2019 and 2020; (B,C), reconstructed XCH₄ enhancement using prior and posterior emissions corresponding to the 112 images from TROPOMI. (D), frequency distribution of correlation coefficients between TROPOMI observations and prior (yellow) and posterior (blue) XCH₄ enhancement on an image-by-image basis. (E), frequency distribution of percentage of XCH₄ reconstructed by prior (green) and posterior (purple) estimatesIndicate figure parts with bold capital letters: (A), (B), etc.



Fig. 3. CH₄ emissions from coal mining in Shanxi. (A), monthly CH₄ emissions from coal mining from January of 2019 to December of 2020 (green bars) and coal production (red line). The total annual coal mining emissions of Shanxi province estimated from inversion by TROPOMI (INV_{TROPOMI}) and three bottom-up inventories in 2019 (PKU-CH₄ v2, EDGAR v6.0 and GFEI) are summarized in (B). The error bars show standard deviation of the inversion ensemble from INV_{TROPOMI}. Note that the data of INV_{TROPOMI} in Jun 2019, Jun 2020, Jul 2020 and Dec 2020 is missing because valid images are not enough for these months (<2) for the inversion. EDGAR v6.0 only provides the annual emission until 2018, so we scaled the coal mine emissions in 2019 by the ratio of coal production between 2019 and 2018. The (C-F) panel shows the spatial pattern of CH₄ emissions from INV_{TROPOMI} and bottom-up inventories (PKU-CH₄ v2, EDGAR v6.0 and GFEI) for Shanxi in 2019.



Fig. 4. Emission factor (EF) of coal mining, coal quality and depth of mining. (A) The composition of the coal production by coal types for each prefecture in Shanxi. (**B**) The relationship between average maximum mining depth and the emission factor (EF) of coal methane emissions derived from TROPOMI top-down inversion (INV_{TROPOMI}) in Shanxi at prefecture level. Note that the color of dots in (**B**) corresponds to the background color in (**A**), with the blue denoting Jinzhong and Changzhi, the two prefectures having deeper coal seams due to crustal subsidence in three tectogenesis after coal formation in the Carboniferous-Permian (30, 31).

