



*Supplement of*

## **Assessment of methane emissions from European countries using satellite and surface observations**

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To account for spatial and temporal correlations in observing-system errors, we follow the approach of Zhang et al. (2022) and Liang et al. (2023). We first define the full observing-system error covariance matrix as  $\mathbf{S}_0' = \mathbf{\Sigma C \Sigma}$ , where  $\mathbf{\Sigma}$  is a diagonal matrix of standard deviations and  $\mathbf{C}$  is the correlation matrix. We introduce spatiotemporal correlations using a parametric form for  $\mathbf{C}_{ij}$  as a function of the spatial ( $\Delta d$ ) and temporal separation ( $\Delta t$ ) between observations:

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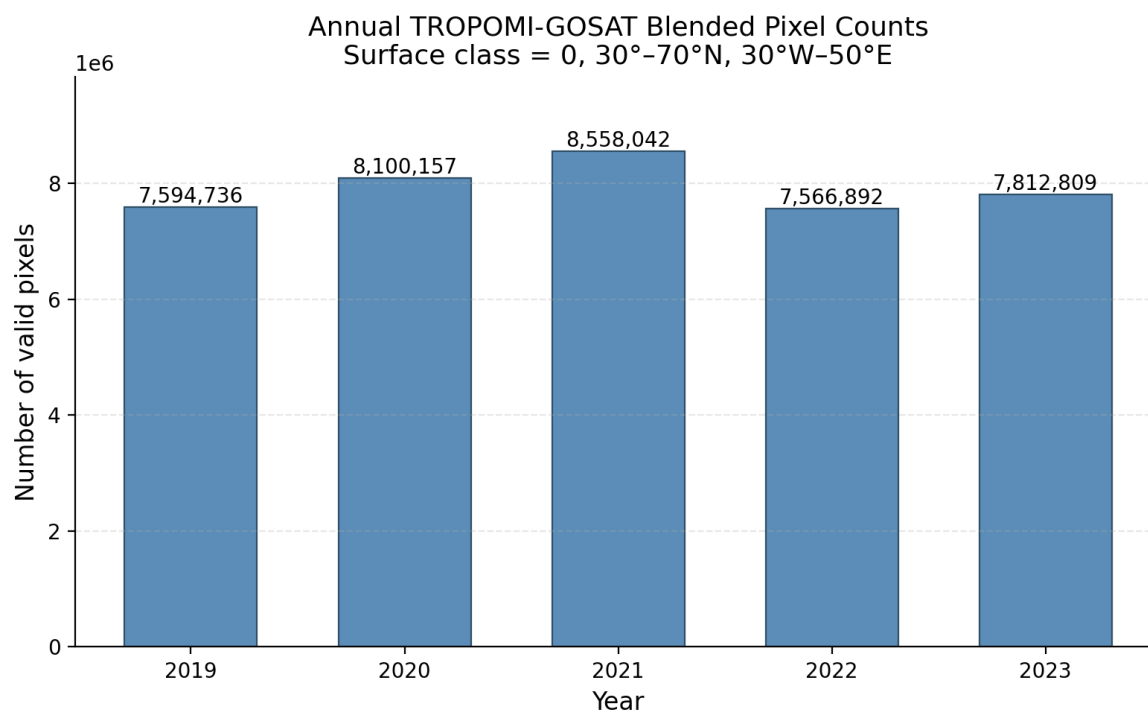
$$\mathbf{C}_{ij} = \exp\left(-\frac{\Delta d}{\rho d}\right) \exp\left(-\frac{\Delta t}{\rho t}\right) \quad \text{Eq. S1}$$

The correlation parameters are estimated from prior model–observation residuals. For TROPOMI observations, both spatial and temporal residual correlations are considered, and the corresponding correlation functions are fitted to the residual statistics shown in Figures S3 and S4. For ICOS observations, spatial correlations between different stations are neglected, whereas temporal correlations are considered following Zhang et al. (2022). The error standard deviations and correlation parameters adopted in the inversion are summarized in Table S4.

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Direct use of the full non-diagonal observing-system error covariance matrix  $\mathbf{S}_0'$  is computationally prohibitive because of the large number of observations. We therefore apply the diagonal approximation of Zhang et al. (2022) and Liang et al. (2023). In this approach, the spatial and temporal correlations between each observation and the surrounding observations are used to derive an observation-specific weight based on the summed squared pairwise correlations. These weights are then incorporated into the effective error variances of individual observations, so that observations with stronger correlations to neighboring observations receive lower effective weights. The resulting observing-system error covariance matrix  $\mathbf{S}_0$  used in the inversion is therefore diagonal, while retaining the effect of spatial and temporal error correlations through the observation-specific weighting. Full mathematical details of the diagonal approximation are provided in Zhang et al. (2022) and Liang et al. (2023).

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45 Figure S1. Annual TROPOMI-GOSAT blended satellite observations count within the inversion during 2019–2023. The counts represent the spatial and temporal coverage of the satellite observations after applying the quality-control procedure described by Balasus et al. (2023).

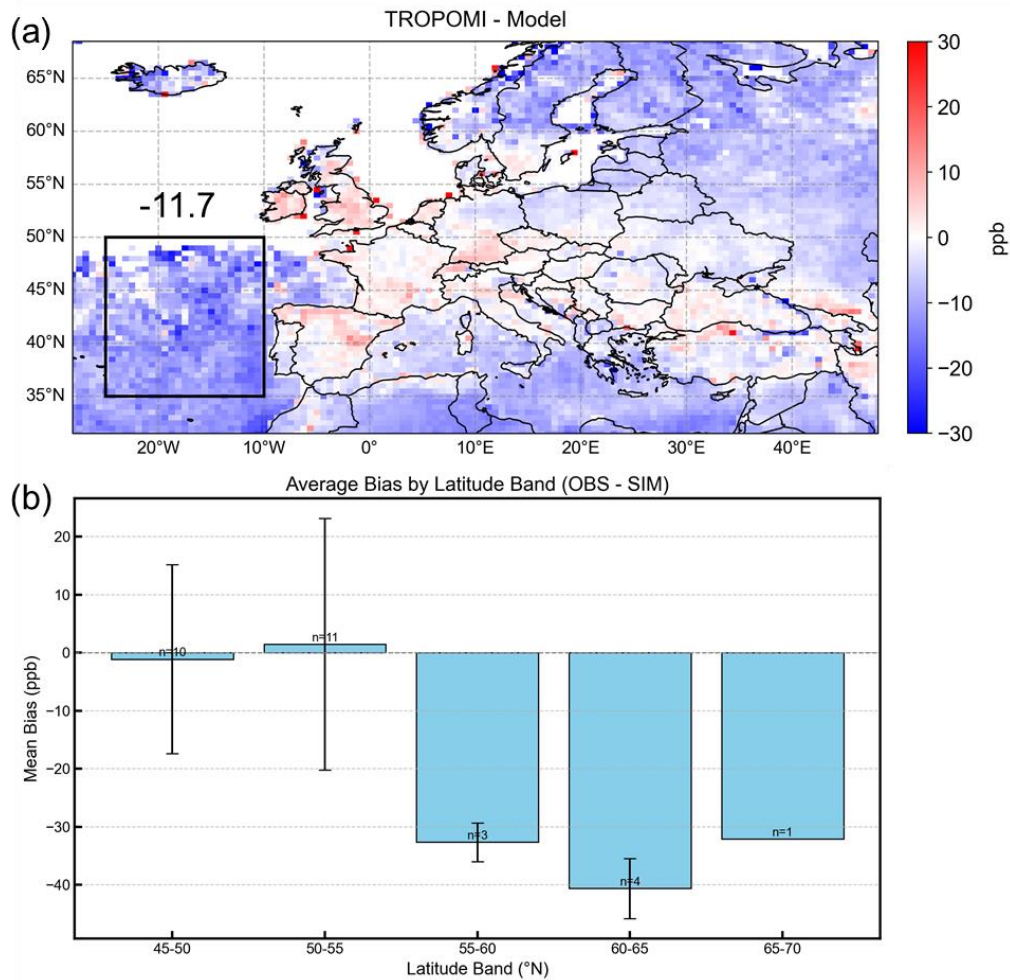
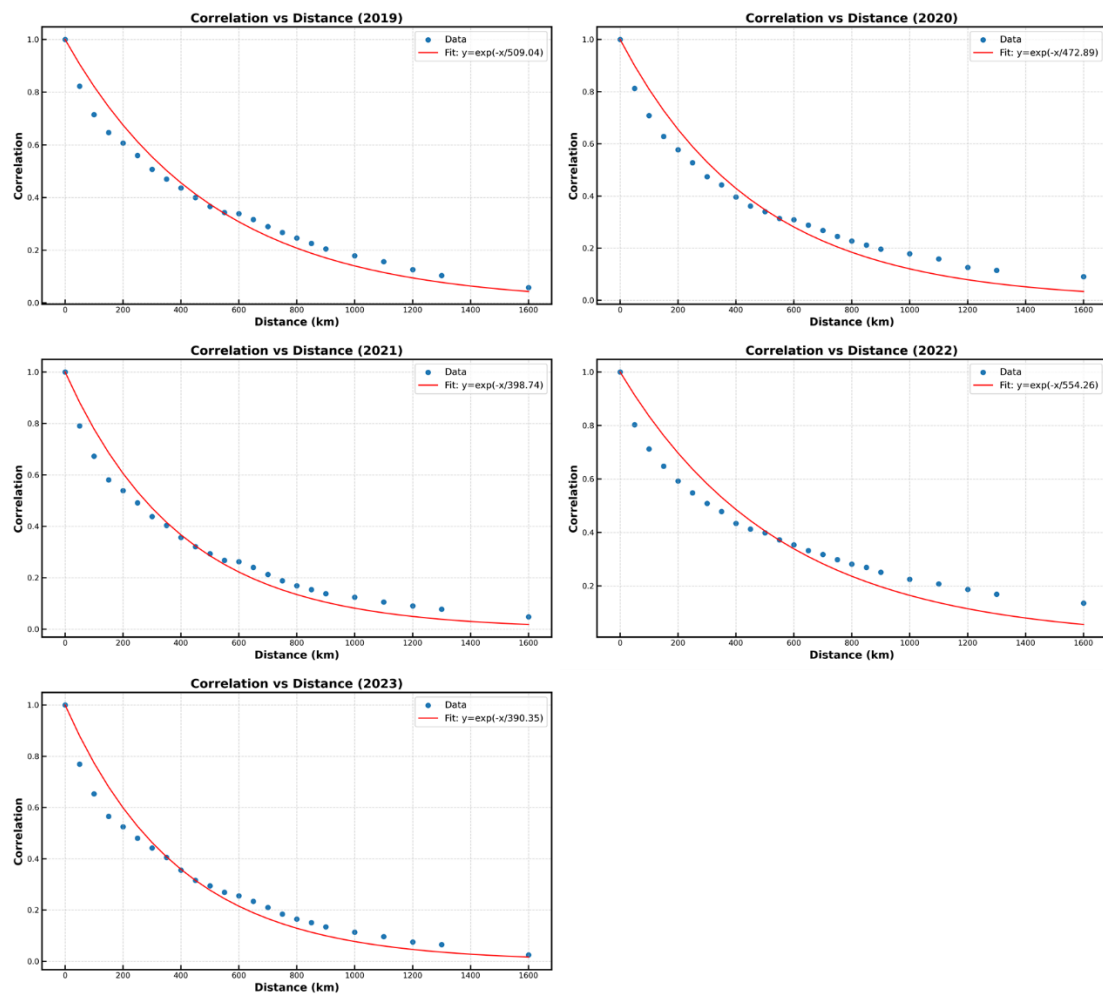


Figure S2. Panel (a) shows the bias (observation – simulation) between TROPOMI satellite retrievals and model simulations, with bias values annotated on the map. Panel (b) presents the same bias binned by latitude, where n denotes the number of ground-based sites within each latitudinal band. The TROPOMI observations over the ocean were used to estimate the correction for the western boundary, while the high-latitude surface observations were used to estimate the correction for the northern boundary. The correction was applied as a fixed offset for each boundary, calculated separately for the western and northern boundary regions. The offset does not vary temporally or horizontally within each boundary region and is applied consistently to the corresponding boundary methane fields.



60 Figure S3. Correlation coefficients of prior residuals are analyzed as functions of spatial distance with results shown as blue dots.

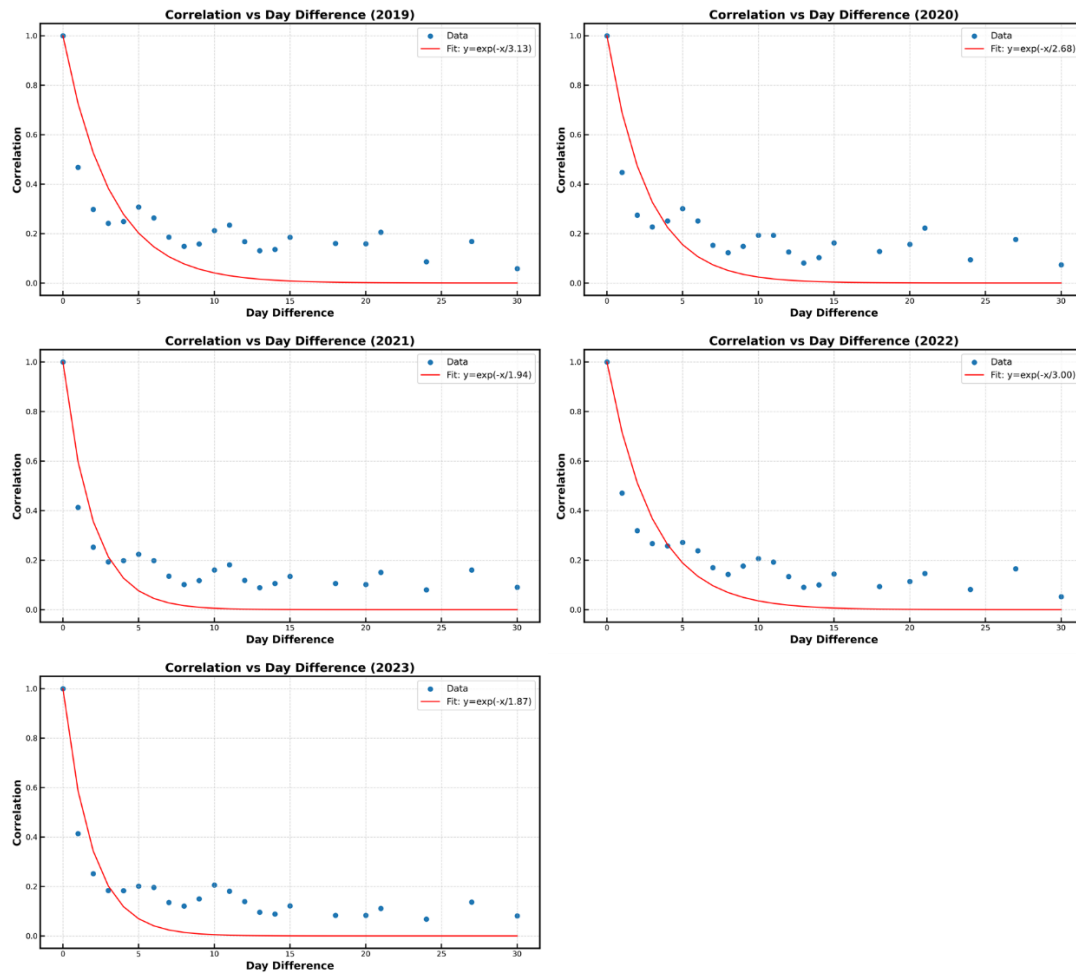


Figure S4. Correlation coefficients of prior residuals are analyzed as functions of temporal separation, with results shown as blue dots.

# Monthly Evolution of XCH<sub>4</sub> Concentration

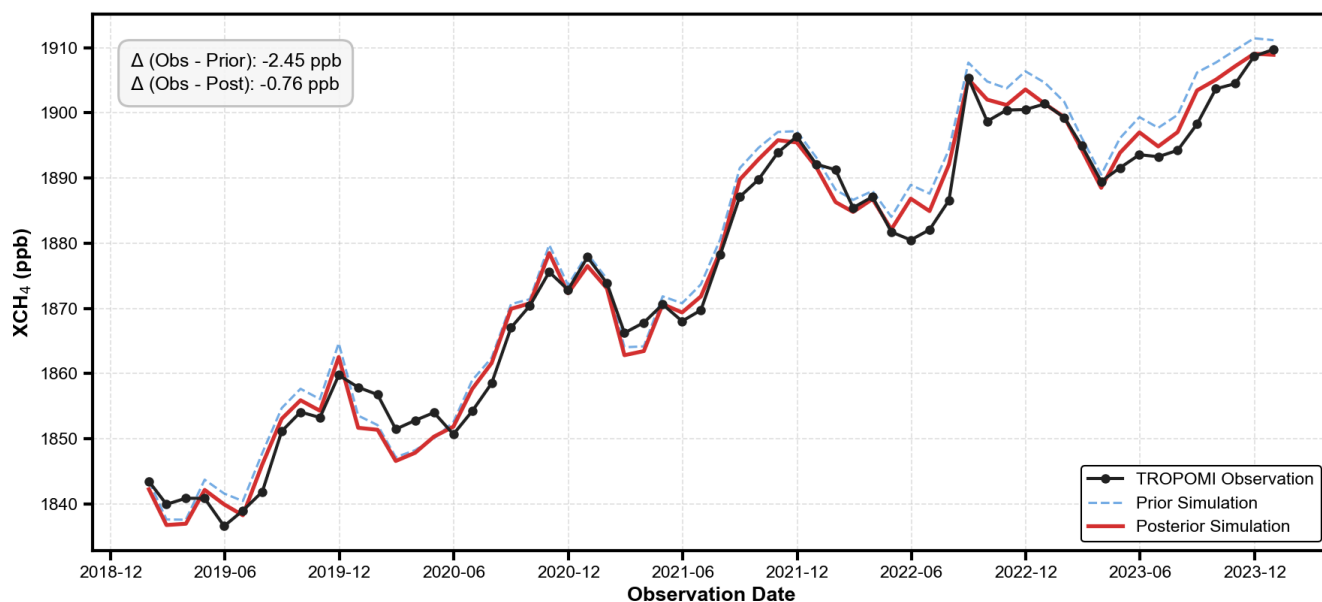
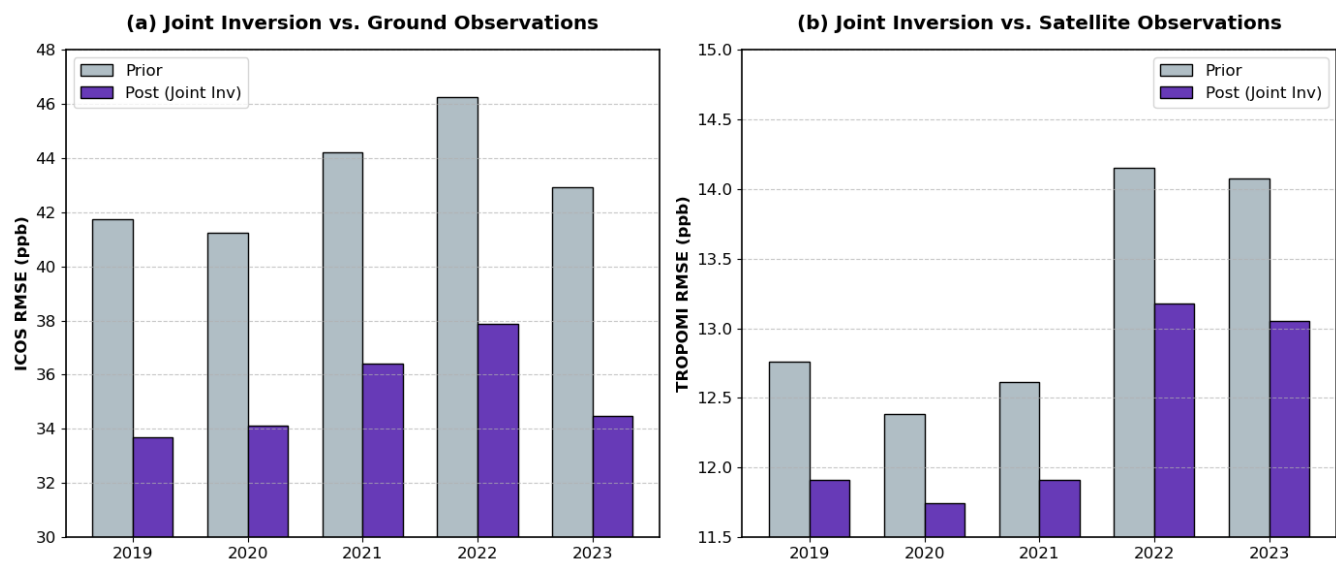


Figure S5. Monthly evolution of XCH<sub>4</sub> concentration. The black solid line represents the observed monthly mean XCH<sub>4</sub>, while the blue dashed and red solid lines represent the corresponding prior and posterior simulations, respectively. The mean differences between observations and simulations are shown in the figure.



75 Figure S6. Panel (a) shows the results of the joint inversion evaluated with ICOS observations, while panel (b) presents the joint inversion assessed using TROPOMI data.

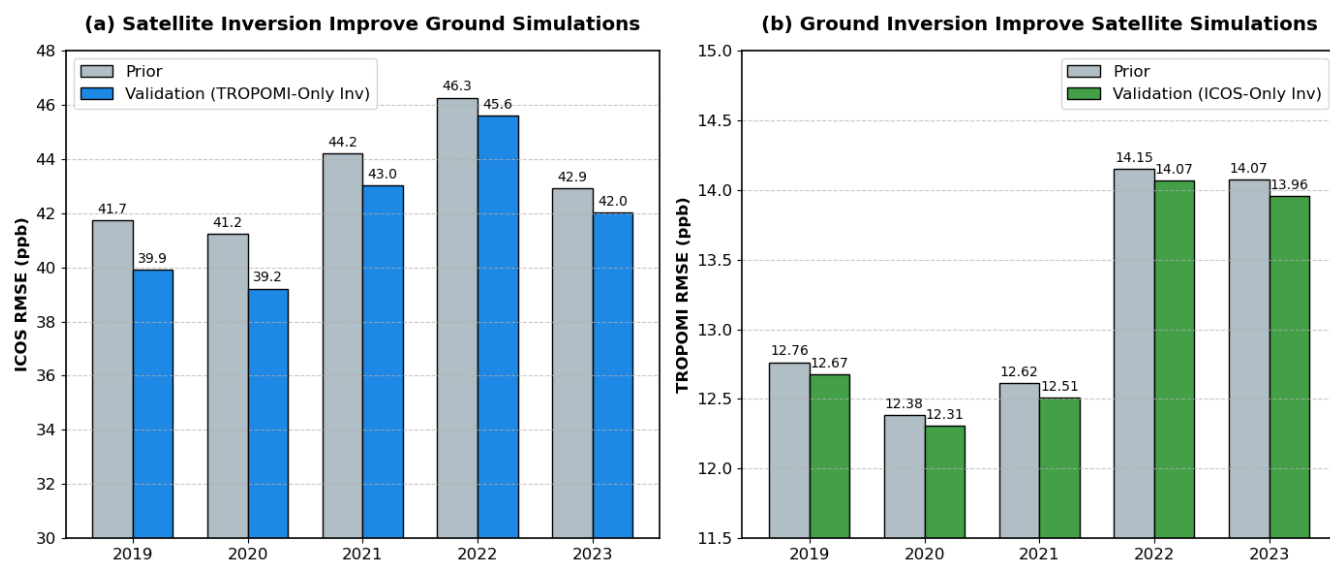


Figure S7. Panel (a) shows the TROPOMI-only inversion evaluated against ICOS surface observations that were not assimilated in the inversion, while panel (b) presents the ICOS-based inversion assessed using TROPOMI data.

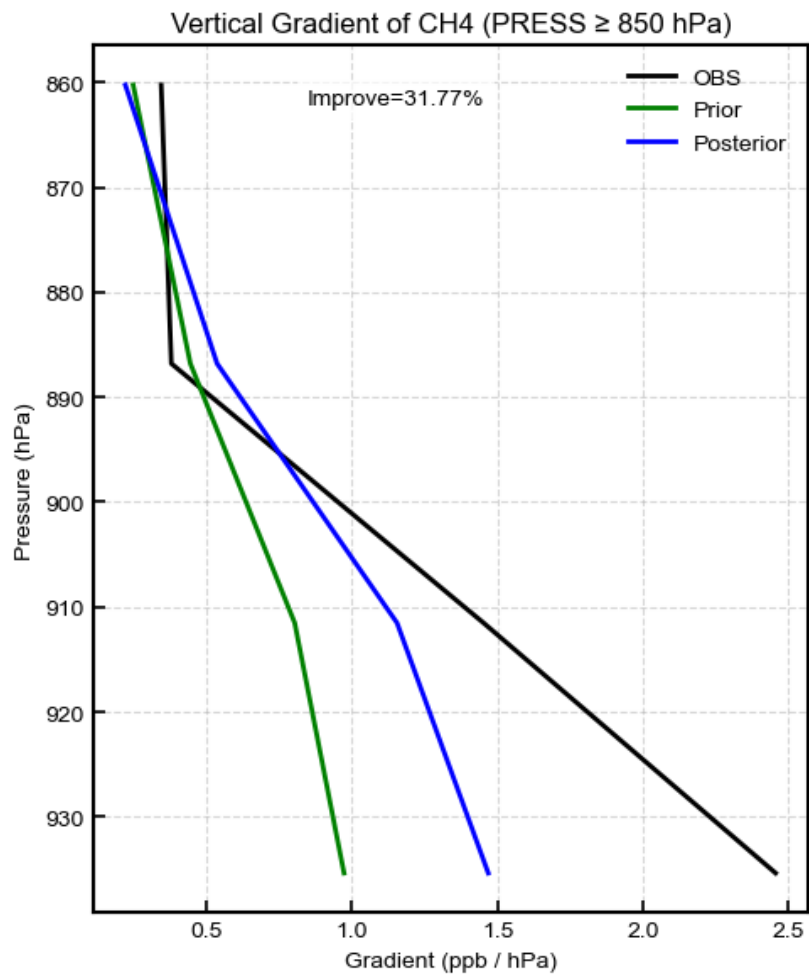


Figure S8. Vertical gradient from IAGOS-CARIBIC aircraft observations, black line is observation, green line is prior simulation, blue line is posterior simulation.

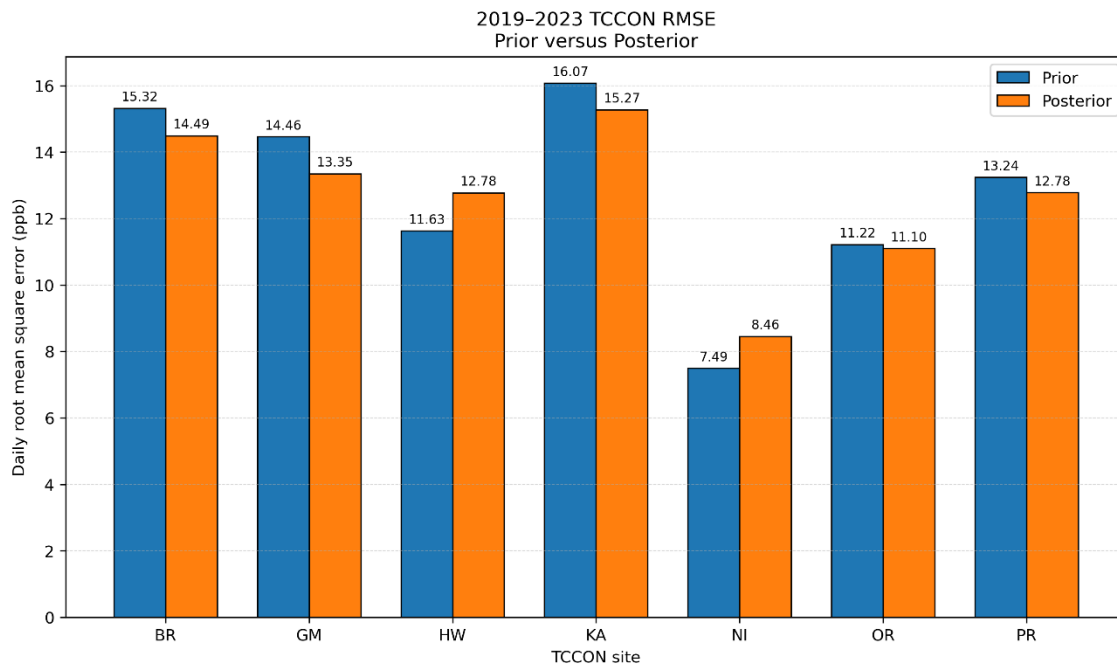


Figure S9. Evaluation of the prior and posterior simulations against independent TCCON column-averaged methane observations at seven European sites. Bars show the root-mean-square error (RMSE) between the observed and simulated XCH<sub>4</sub>, averaged over 2019–2023. Blue bars represent the prior simulation and yellow bars represent the posterior simulation. See Table S3 for site information.

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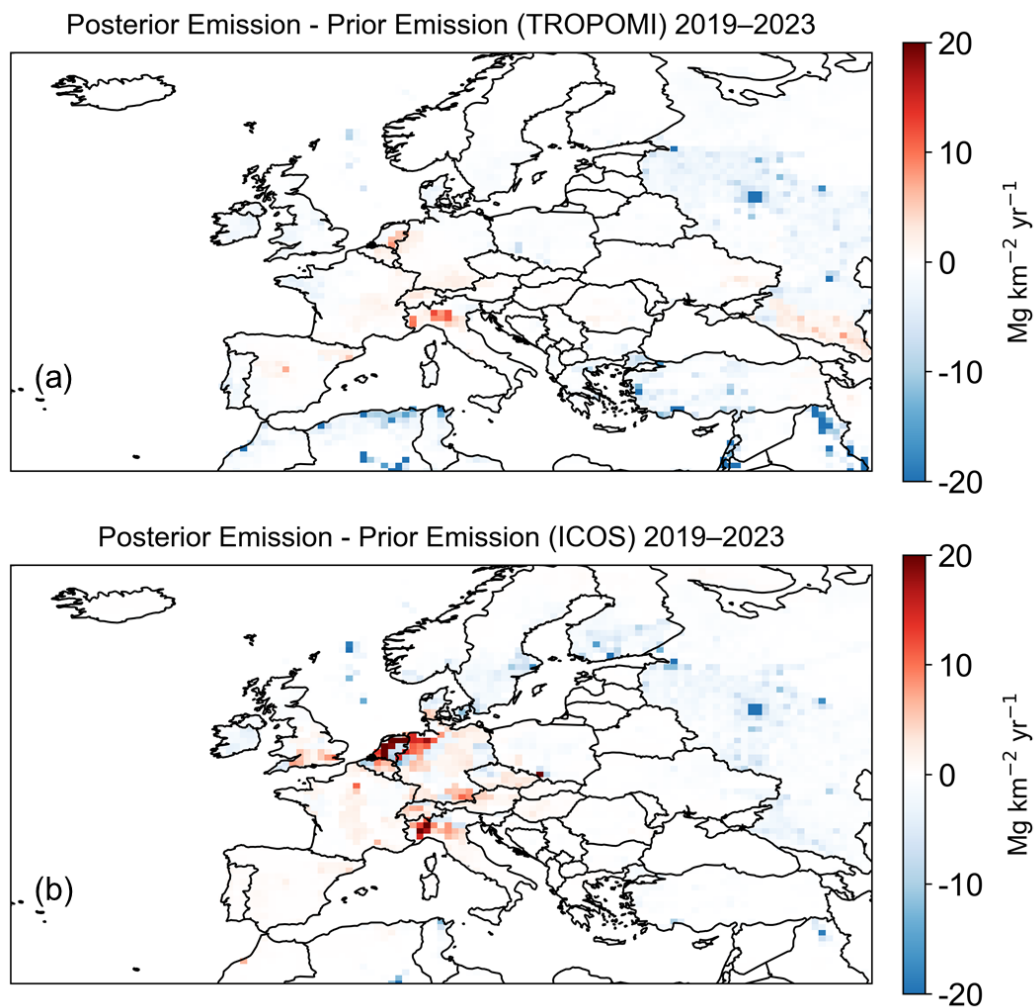


Figure S10. Panel (a) shows the results of the TROPOMI-only inversion, while panel (b) presents the ICOS-only inversion.

Posterior Emission - Prior Emission (Wetland) 2019–2023

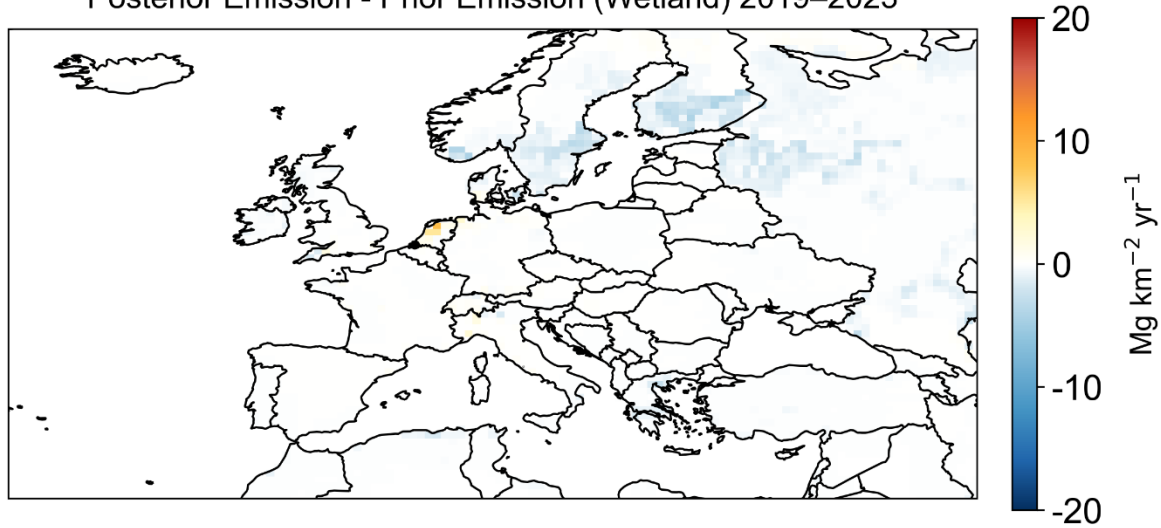


Figure S11. Spatial adjustment of wetland emission.

Table S2. Prior emission inventories used in the inversion.

| Type                                                             | Data source                 | Time period | Spatial resolution | Temporal resolution |
|------------------------------------------------------------------|-----------------------------|-------------|--------------------|---------------------|
| Anthropogenic emissions<br>(agriculture, fossil fuel, and waste) | EDGARv8                     | 2019-2022   | 0.1°×0.1°          | monthly             |
| Wetland emissions                                                | WetCHARTs                   | 2019        | 0.5°×0.5°          | monthly             |
| Biomass burning emissions                                        | QFED2                       | 2015        | 0.1°×0.1°          | daily               |
| Geological emissions                                             | Maasakkers et al.<br>(2019) | 2012        | 0.1°×0.1°          | annually            |
| Termite emissions                                                | Fung et al. (1991)          | 1985        | 4°×5°              | annually            |

Prior methane emission inventories used in the inversion, including their reference years and native spatial and temporal resolutions. EDGAR v8 anthropogenic emissions are available for 2019–2022; the 2022 emission fields were therefore also used as the anthropogenic prior for the 2023 inversion. For the other prior components, the same source-specific temporal fields were repeated for each inversion year. Consequently, these components retain their prescribed subannual variability but do not include prior interannual variability.

Table S3. Observation stations used in this study.

| Network | ID  | Station name                                       | Latitude<br>(deg N) | Longitude<br>(deg E) | Mountain<br>site | Station<br>elevation<br>(m) | Inversion | Validation |
|---------|-----|----------------------------------------------------|---------------------|----------------------|------------------|-----------------------------|-----------|------------|
| ICOS    | BIR | Birkenes                                           | 58.39               | 8.25                 | No               | 219                         | Yes       | No         |
| ICOS    | CBW | Cabauw                                             | 51.97               | 4.93                 | No               | 0                           | Yes       | No         |
| ICOS    | CMN | Monte Cimone                                       | 44.19               | 10.70                | Yes              | 2165                        | No        | No         |
| ICOS    | GAT | Gartow                                             | 53.07               | 11.44                | No               | 70                          | Yes       | No         |
| ICOS    | HEL | Helgoland                                          | 54.18               | 7.88                 | No               | 43                          | Yes       | No         |
| ICOS    | HPB | Hohenpeissenberg                                   | 47.80               | 11.02                | No               | 934                         | Yes       | No         |
| ICOS    | HTM | Hyltemossa                                         | 56.10               | 13.42                | No               | 115                         | Yes       | No         |
| ICOS    | HUN | Hegyhátsál hátté<br>rszenyettőség-mérő á<br>llomás | 46.96               | 16.65                | No               | 248                         | Yes       | No         |
| ICOS    | IPR | Ispra                                              | 45.81               | 8.64                 | No               | 210                         | Yes       | No         |
| ICOS    | JFJ | Jungfraujoch                                       | 46.55               | 7.99                 | Yes              | 3572                        | No        | No         |
| ICOS    | JUE | Jülich                                             | 50.91               | 6.41                 | No               | 98                          | Yes       | No         |
| ICOS    | KIT | Karlsruhe                                          | 49.09               | 8.42                 | No               | 110                         | Yes       | No         |
| ICOS    | KRE | Křešín u Pacova                                    | 49.57               | 15.08                | No               | 534                         | Yes       | No         |
| ICOS    | LIN | Lindenberg                                         | 52.17               | 14.12                | No               | 73                          | Yes       | No         |
| ICOS    | LMP | Lampedusa                                          | 35.52               | 12.63                | No               | 45                          | Yes       | No         |
| ICOS    | LUT | Lutjewad                                           | 53.40               | 6.35                 | No               | 1                           | Yes       | No         |
| ICOS    | NOR | Norunda                                            | 60.09               | 17.48                | No               | 46                          | Yes       | No         |
| ICOS    | OPE | Observatoire pérenne<br>de l'environnement         | 48.56               | 5.50                 | No               | 390                         | Yes       | No         |
| ICOS    | OXK | Ochsenkopf                                         | 50.03               | 11.81                | Yes              | 1022                        | No        | No         |
| ICOS    | PAL | Pallas                                             | 67.97               | 24.12                | No               | 565                         | Yes       | No         |
| ICOS    | PRS | Plateau Rosa                                       | 45.93               | 7.70                 | Yes              | 3480                        | No        | No         |
| ICOS    | PUI | Puijo                                              | 62.91               | 27.65                | No               | 232                         | Yes       | No         |
| ICOS    | PUY | Puy de Dôme                                        | 45.77               | 2.97                 | Yes              | 1465                        | No        | No         |
| ICOS    | RGL | Ridge Hill                                         | 52.00               | -2.54                | No               | 207                         | Yes       | No         |
| ICOS    | SAC | Saclay                                             | 48.72               | 2.14                 | No               | 160                         | Yes       | No         |
| ICOS    | SMR | Hyytiälä                                           | 61.85               | 24.29                | No               | 181                         | Yes       | No         |
| ICOS    | SSL | Schauinsland                                       | 47.92               | 7.92                 | Yes              | 1205                        | No        | No         |
| ICOS    | STE | Steinkimmen                                        | 53.04               | 8.46                 | No               | 29                          | Yes       | No         |
| ICOS    | SVB | Svartberget                                        | 64.26               | 19.78                | No               | 269                         | Yes       | No         |

|       |     |                  |       |       |     |      |     |     |
|-------|-----|------------------|-------|-------|-----|------|-----|-----|
| ICOS  | TOH | Torfhaus         | 51.81 | 10.54 | No  | 801  | Yes | No  |
| ICOS  | TRN | Trainou          | 47.96 | 2.11  | No  | 131  | Yes | No  |
| ICOS  | UTO | Utö - Baltic sea | 59.78 | 21.37 | No  | 8    | Yes | No  |
| ICOS  | WAO | Weybourne        | 52.95 | 1.12  | No  | 17   | Yes | No  |
| ICOS  | WES | Westerland       | 54.92 | 8.31  | No  | 12   | Yes | No  |
| ICOS  | ZSF | Zugspitze        | 47.42 | 10.98 | Yes | 2666 | No  | No  |
| TCCON | BR  | Bremen           | 53.1  | 8.85  | No  | 30   | No  | Yes |
| TCCON | GM  | Garmisch         | 47.48 | 11.06 | No  | 750  | No  | Yes |
| TCCON | HW  | Harwell          | 51.57 | -1.32 | No  | 140  | No  | Yes |
| TCCON | KA  | Karlsruhe        | 49.1  | 8.44  | No  | 110  | No  | Yes |
| TCCON | NI  | Nicosia          | 35.14 | 33.38 | No  | 190  | No  | Yes |
| TCCON | OR  | Orléans          | 47.96 | 2.11  | No  | 130  | No  | Yes |
| TCCON | PR  | Paris            | 48.85 | 2.36  | No  | 60   | No  | Yes |
| TCCON | SO  | Sodankylä        | 67.37 | 26.63 | No  | 190  | No  | No  |

Information on the ICOS surface stations assimilated in the inversion and the TCCON sites used for independent evaluation. Seven high-altitude mountain stations were excluded from the assimilation to avoid potential representativeness issues associated with daytime observations at mountainous sites. For the TCCON evaluation, sites at very high latitudes were excluded because of model-representation limitations.

Table S4. Observing-system error parameters used for the ICOS and blended TROPOMI observations.

| Observations                     | Standard deviation (ppbv) | Temporal correlation scale<br>(day) | Spatial correlation scale<br>(km) |
|----------------------------------|---------------------------|-------------------------------------|-----------------------------------|
| TROPOMI-GOSAT<br>blended product | ~12                       | 2                                   | 500                               |
| ICOS surface observations        | ~40                       | 2                                   | -                                 |

The baseline error standard deviations were estimated separately for the two datasets from the standard deviations of the residuals between the observations and the prior model simulations. Following Zhang et al. (2022), the temporal and spatial correlations of the TROPOMI residuals were estimated and used to derive observation-specific weights. ICOS observations from different sites were assumed to be spatially independent, while a temporal correlation scale of 2 d was prescribed following Zhang et al. (2022). The resulting parameters were used to account for the reduced effective information content of correlated observations.

## References

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