,
Horizon 2020 Societal challenge 5:
Climate action, environment, resource efficiency and raw materials

VERIFY

Observation-based system for monitoring and verification of greenhouse gases

GA number 776810, RIA

Deliverable number (relative in WP)	**D4.13**
Deliverable name:	Methane fluxes using TROPOMI data
WP / WP number:	WP4
Delivery due date:	Month 42 (31/07/2021)
Actual date of submission:	Month 52 (19/05/2022)
Dissemination level:	Public
Lead beneficiary:	CEA-LSCE
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Changes with respect to the DoA
Delivery shifted from M42 to M49.
Dissemination and uptake
The results will be made available via the VERIFY project database and are currently available via a data server to the project partners. **These results, as the first inversion of methane in Europe with TROPOMI data, will be presented in a letter. They are also part of a paper being drafted for ESSD to describe the implementation of satellite data assimilation in the CIF.**
Short Summary of results (<250 words)
This deliverable presents the CH_4 fluxes obtained by atmospheric inversion with the CIF using the chemistry-transport model CHIMERE to assimilate TROPOMI data. **The retrieved CH4 fluxes are provided at a 0.5°latitude ×0.5°longitude and weekly resolution over a European domain (15°W to 35°E and 33°N to 73°N) for the year 2019.**
Evidence of accomplishment **(report, manuscript, web-link, other)**
The retrieved fluxes will be accessible through VERIFY project database. Note that some of these data may be password protected during a consolidation phase and thus only accessible to the VERIFY partners (accessible through the internal share-point platform).

VERIFY is a research project funded by the European Commission under the H2020 program. Grant Agreement number 776810.

Version	Date	Description	Author (Organisation)
V0	14/02/22	Creation/Writing	Isabelle PISON (LSCE)
V0.1	28/03/2022	Internal review	A. Fortems-Cheiney, G. Broquet (LSCE)
V1	02/05/2022	Formatting	Aurélie Paquirissamy (CEA/LSCE)
V1.1	17/05/2022	End of internal review	R. Thompson (NILU)
V2	02/05/2022	Formatting/Delivery on the Participant Portal	Aurélie Paquirissamy and Philippe Peylin (CEA/LSCE)

1. Glossary

Abbreviation / Acronym	Description/meaning
CAMS	Copernicus Atmosphere Monitoring System
CHIMERE	Chemistry-transport area-limited Eulerian model
CIF	Community Inversion Framework
CTM	Chemistry-Transport Model
ECMWF	European Centre for Medium-Range Weather Forecasts
EDGAR	The Emission Database for Global Atmospheric Research
EU	European Union
GCP-CH4	Global Carbon Project – Methane
GFED	Global Fire Emissions Database
GHG	Greenhouse gas
ICOS	Integrated Carbon Observatory System
IFS	Integrated Forecasting System
TROPOMI	TROPOspheric Monitoring Instrument

2. Executive Summary

Inverse modeling of greenhouse gas (GHG) emissions combining atmospheric observations and transport simulations is an important tool with a high potential for mapping and quantifying emissions. Inverse modeling systems have been used for about three decades for retrieving anthropogenic and/or natural GHG fluxes.

One of the challenges today is to use the streams of satellite data, which have tremendously increased in the last five years, in terms of the number of observations as well as spatial resolution for methane (CH_4). TROPOMI now provides CH_4 total columns with a resolution of at first 7 km x 7 km and most recently, 5.5 km x 7 km; over Europe in 2019, after filtering for qualit, more than 645,000 valid retrievals are available. Inversion systems must be able to use as much of the information they contain on the fluxes as possible.

To answer the challenge of numerically treating big data streams, the CIF has been developed in VERIFY (Berchet *et al.*, 2021), based on different inversion systems from various groups.

Here, the CIF is used to invert emissions of CH_4 in Europe by assimilating TROPOMI CH_4 total columns into the area-limited Eulerian CTM CHIMERE. The upper part of the atmosphere (tropopause and stratosphere) not simulated by CHIMERE was taken from the IFS simulation of methane concentrations. A variational inversion is run for the whole year 2019 and fluxes are retrieved.

Technically, the results obtained show that the CIF is able to use satellite data to constrain one-year inversions in Europe at a widely used horizontal resolution (0.5° x 0.5°) in a relatively short computing time (less than one week). This is a key milestone in the development of the CIF, which shows that it can be planned in the near future to set-up inversions with a finer horizontal resolution (e.g. 10 km x 10 km), which will make use of the information provided by the fine resolution of satellite data such as TROPOMI's.

Scientifically, the retrieved fluxes show realistic patterns, such as increased emissions compared to the prior in regions where the contribution from agriculture is large. Moreover, the total fluxes (e.g. in EU27+UK 24,000 ktCH_4 in 2019) are in agreement with other inversions, either regional or global, assimilating either surface or other satellite data (total range in EU27+UK of about 11,900 to 33,500 kt for years previous to 2017).

The next steps are first, the extension of the inversion run here to the whole period of availability of TROPOMI CH4 data (i.e. from May 2018 to the present day); then, a full scientific analysis of the results, with more comparisons to other estimates, either from inventories, biogeochemical models or inversions, and sensitivity studies to the specification of the error statistics.

3. Introduction

Inverse modeling of greenhouse gas (GHG) emissions combining atmospheric observations and transport simulations is an important tool with a high potential for mapping and quantifying emissions. Inverse modeling systems have been used for about three decades for retrieving GHG fluxes, either anthropogenic, natural or total. First, surface measurements were assimilated into global transport models but the sparse distribution of surface stations at the global scale, the lack of continuous measurements and the coarse resolution at which the models were run to keep reasonable computing times lead to retrieving fluxes at a coarse spatial and temporal resolution. Over the last two decades, it has become possible to use continuous measurements, at more and more stations, organized into networks such as ICOS in Europe, and even, in the last decade, satellite data, with a wider coverage albeit larger uncertainties than the surface data. Over the last decade, satellite data have been assimilated into CTMs with optimized codes and providing finer resolutions for shorter computing times. Nevertheless, two challenges remained:
- the inversion systems are complex numerical systems, often relying on various codes in different languages (shell, fortran, python, etc.). It is time consuming to learn how to use them, they are sometimes difficult to maintain and therefore, very heavy to use routinely outside of an academic research environment.
- the streams of satellite date have tremendously increased in the last five years, in number of observations as well as in spatial resolution, for a large number of species. TROPOMI now provides for methane (CH_4) total columns with a resolution of 7 km x 7 km and, more recently, 5.5 km x 7 km; over Europe in 2019, after filtering for quality, more than 645,000 valid retrievals are available. The inversion systems must be able to deal with these data technically and to use as much of the information they contain on the fluxes as possible.

To answer both these challenges, the CIF has been developed in VERIFY (Berchet *et al.* (2021) for a full description and https://community-inversion.eu/index.html for the on-line documentation and tutorials), based on different inversion systems from various groups.
The CIF is used here for the first time to invert emissions of CH_4 in Europe by assimilating TROPOMI CH_4 total columns for the year 2019. The domain covered is described in Section 4.1 and the prior information (data to assimilate, prior fluxes and other input) are described in Section 4.2. The inversion set-up is detailed in Section 4.3. The results are described and a first analysis of them is made in Section 4.4.
Technically, the inversion of methane fluxes described here is a demonstration of how the CIF makes it possible to assimilate large satellite data streams into a CTM which runs at a relatively fine spatial and temporal resolution and to retrieve emission fluxes which are scientifically usable, all this with an inversion system which is easy to install and deploy and well documented.

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4. Methane fluxes retrieved from TROPOMI data assimilation with the CIF

4.1 Targeted domain

The targeted domain covers all EU countries: it spans from 15°W to 35°E and 33°N to 73°N (see Fig. 1). The grid used for discretizing emissions, meteorological data and other inputs and for running the CTM covers this domain at a 0.5 degrees longitude x 0.5 degrees latitude resolution. Vertically, the domain where transport is simulated by CHIMERE extends from the surface to 200 hPa (17 sigma-pressure levels). Above this pressure (approximately the tropopause), the concentration fields are taken from the CAMS simulations (based on the IFS system) for CH_4 concentrations, which go up to 0.1 hPa.
The size of this domain makes it possible to neglect the chemistry of methane (oxidation by hydroxyl radicals OH with a lifetime of CH_4 from 8 to 10 years) compared to the ventilation time of the domain (air masses generally remain in the domain for less than two weeks). Therefore, CH^4 loss within the domain is ignored but it is accounted for in the boundary conditions.

4.2 Input data

4.2.1 Atmospheric data

The assimilated data consist in TROPOMI CH_4 total columns, which are level 2 (L2) products in the version 1.04.00 (available via https://s5phub.copernicus.eu/dhus/#/home). Only data over land and for zenithal angles smaller than 60 degrees are provided. The data is filtered according to the information provided in the associated document entitled *Methane [L2__CH4___] Readme*: the quality flag must be strictly larger than 0.5.
A further filtering of the data is applied after a forward simulation which makes it possible to compare the data to their prior simulated equivalent: data which are associated to a simulated equivalent less than 1650 ppb are filtered out. This second filter eliminates less than 0.04% of the data over the whole year. They correspond to cases when the model simulates very low methane concentrations due to the limitations of the vertical mixing parameterization (note that this requires still further investigation to determine if it can be improved). The distribution of the data to assimilate is illustrated in Fig.1 (top left).

4.2.2 Prior fluxes

The prior fluxes are the same as used in D4.9, their main features are summarized here:
- anthropogenic fluxes include agriculture, fugitive and combustion emissions from oil, gas and coal, biofuel combustion, industry, and waste. They are taken from EDGAR-v6.0 (https://edgar.jrc.ec.europa.eu/dataset_ghg60#intro).
- natural fluxes from peatlands, inundated and mineral soils are from the JSBACH-HIMMELI model (VERIFY product).

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- natural fluxes from inland water bodies are monthly climatological estimates provided by the Université Libre de Bruxelles (VERIFY product)
- the geological emissions are a climatology based on Etiope *et al.* (2019), provided by Marielle Saunois from GCP-CH4 and scaled down to a global total of 15 Tg/y in accordance with the maximum suggested by Petrenko *et al.* (2017).
- the emissions due to termites are a climatology based on the estimate of S. Castaldi (Saunois *et al.*, 2020), provided by Marielle Saunois from GCP-CH4.
- the biomass burning fluxes are taken from GFEDv-4.1s (https://www.globalfiredata.org/index.html)
- the ocean fluxes are a climatology based on Weber *et al.* (2019).

The total fluxes used as the prior are illustrated in Fig.2 (2019 mean, top left).

4.2.3 Boundary and initial conditions

As in D4.9, boundary and initial conditions of methane are taken from the CAMS reanalysis product for CH_4. As indicated in Sect. 4.1, the stratospheric fields are taken from CAMS IFS.

4.2.4 Meteorological input

CHIMERE requires a set of meteorological variables in input. They are taken from the ECMWF's IFS operational forecast (every three hours) retrieved at 0.25 degrees x 0.25 degrees and interpolated onto the model's grid (0.5 degrees x 0.5 degrees).

4.3 Inversion set-up

A variational inversion is run for the whole year 2019 and fluxes, boundary and initial conditions and stratospheric concentration fields are controlled, as described in the following Sections.

4.3.1 Control vector

The control vector contains the increments which are applied to the prior fields to obtain the so-called posterior fields. In this set-up, it consists in:
- CH_4 total fluxes at CHIMERE's horizontal grid resolution for each week of the year: 100 (longitude) x 80 (latitude) x 52 (weeks) = 416,000 components
- CH_4 initial conditions at CHIMERE's spatial resolution: 100 x 80 x 17 = 136,000 components
- CH_4 lateral boundary conditions for each half border (North, East, South, West) at CHIMERE's vertical resolution and every two days: 4x2 x 17 x 365/2 = 24,820 components
- CH_4 top boundary conditions for the whole top of CHIMERE's domain every two days: 1 x 365/2 = 182 components
- CH_4 stratospheric columns at their native resolution (3° longitude x 2° latitude horizontal resolution) every two days: 120 x 90 x 365/2 = 1,971,000 components.

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4.3.2 Observation vector

After filtering (see Sect. 4.2.1), the observation vector has 647,801 components for the whole year 2019.

4.3.3 Error statistics

The error statistics give information on:
-the uncertainties on the prior control variables
-the uncertainties in the difference between the assimilated data and their simulated equivalents.
The larger the uncertainties on the prior fluxes, the larger the increments the inversion can put on them to get simulated equivalents close to the assimilated data. The smaller the uncertainties on the difference between the assimilated data and their simulated equivalent, the closer the inversion tries and brings them (by modifying the control variables).
In the framework of Bayesian inversion used here, the error statistics are described by matrices, with variances on the diagonal and covariances as off-diagonal terms. They are called respectively the prior error (for the error statistics on the prior controlled variables) and the observation error (for the error statistics on the difference between the assimilated data and their simulated equivalents).

In the set-up chosen here, the observation error is not correlated from one observation to another i.e. the observation error is specified as individual errors associated to each difference between data and simulation. These errors are set at 20 ppb (to take into account the retrieval error plus transport errors in CHIMERE).

The prior error is adapted for each type of component in the control vector:
- for CH_4 total fluxes, the standard deviation is set at 100% (for each CHIMERE's grid cell for each week of the year) and covariances are taken into account with correlation lengths of 50 km on land, 100 km on sea and one week through time
- for CH_4 initial conditions, the standard deviation is set at 2% (no covariances)
- for lateral boundary conditions, the standard deviation is set at 2% and covariances are taken into account with correlation lengths of 100 km on land and sea and 5 days through time
- for top boundary conditions, the standard deviation is set at 2% and covariances are taken into account with correlation lengths of 5 days through time
- for stratospheric columns, the standard deviation is set at 2% and covariances are taken into account with correlation lengths of 100 km on land and sea and 5 days through time.

4.4 Inversion results

The reduction in the norm of the gradient and in the value of the cost function in the inversions performed is more than 90%, indicating the convergence of the variational inversion scheme in mathematical terms.

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4.4.1 Analysis: posterior columns

The columns simulated with the posterior fields (emissions but also boundary conditions, etc.) are called "analysis". The performance of the inversion can be illustrated by the decrease of the differences between the simulated equivalent of the data and the data. Over the whole year 2019 (Fig. 1), the prior simulated columns are largely under-estimated, with a mean difference with the data of more than 70 ppb over the whole domain. The mean difference between the analysis and the data is reduced to about 0.75 ppb.

4.4.2 Retrieved fluxes

The mean posterior fluxes for 2019 are shown in Fig. 2 (top right), with the increments compared to the prior (bottom). As expected, since the prior emissions lead to a general under-estimation of the columns, the fluxes retrieved by the inversion are increased compared to the prior in large areas of the domain. Particularly noticeable increases (from 5 to 15%) are found in areas with a large contribution from agricultural sectors according to the prior emissions such as the Po valley (Italy) and Belgium and the Netherlands. The region of Madrid (Spain) and a region close to Silesia in Poland also show increases of more than 2.5% compared to the prior.
From the result file which constitute D4.13, the analysis of the corrections on prior fluxes can be more detailed spatially and temporally. For example, the weekly time scale makes it possible to evaluate the increments on the seasonal cycle (illustration Fig. 3 at the European scale).

To evaluate the fluxes retrieved here, we compare them to the ranges detailed in the recent study over Europe by Petrescu *et al.* (2021) (Fig. 4). The total net methane emissions for the year 2019 in EU27+UK (without Malta in our case) obtained with assimilating TROPOMI total columns is in agreement with the previous regional and global inversions i.e. our total of 24086 $ktCH_4$ is within the ranges of the other regional inversions assimilating surface data (from 22610 to 33483 $ktCH_4$), of global inversions assimilating surface data (from 19702 to 30975 $ktCH_4$) and of global inversions assimilating different satellite data (GOSAT's) (from 11892 to 30842 $ktCH_4$) . The consistency of these results will have to be tested in more details with comparisons per (groups of) countries and, when available (see Section 5), for other years, TROPOMI CH_4 data being available from May 2018.

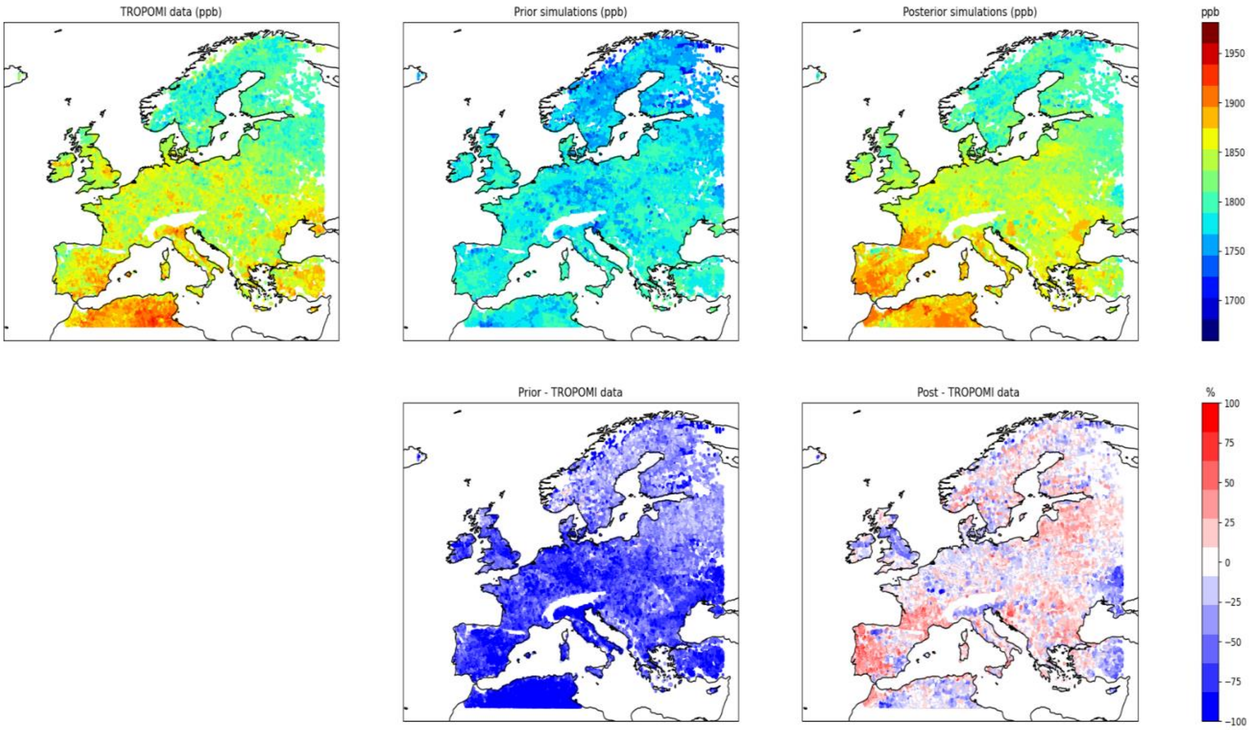

Figure 1: total columns of methane (ppb) as provided by TROPOMI (top left), their equivalents simulated by CHIMERE with the prior inputs (top middle) and their equivalents simulated by CHIMERE with the posterior fields (top right). The differences between the simulated equivalent and the data are shown on the bottom line for the prior (middle) and the posterior (right).

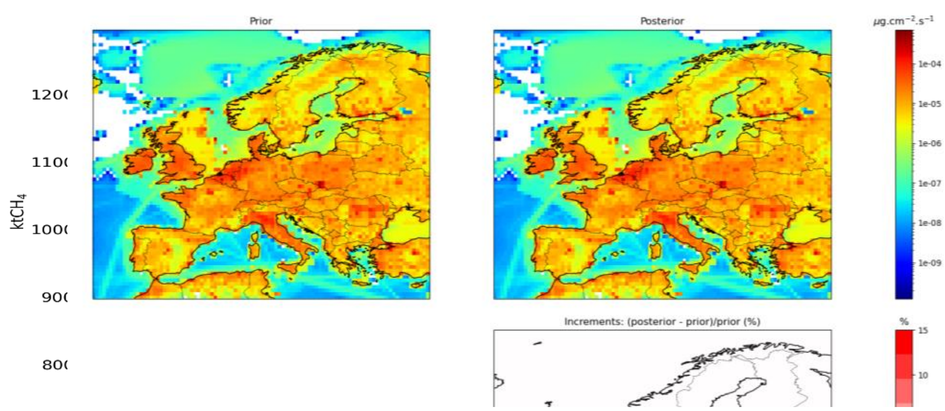

Figure 3: monthly prior and posterior methane emissions (kt) for the year 2019 in EU27+UK (land only, Malta not included).

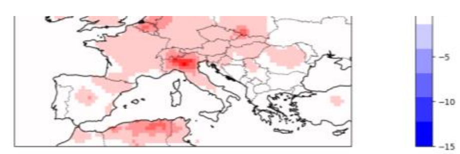

Figure 2: mean total emissions of methane (µgCH4 / cm2/s) in 2019: prior (top middle) and posterior (top right); the mean increments over 2019 are given as a percentage: (post-prior)/prior x 100.

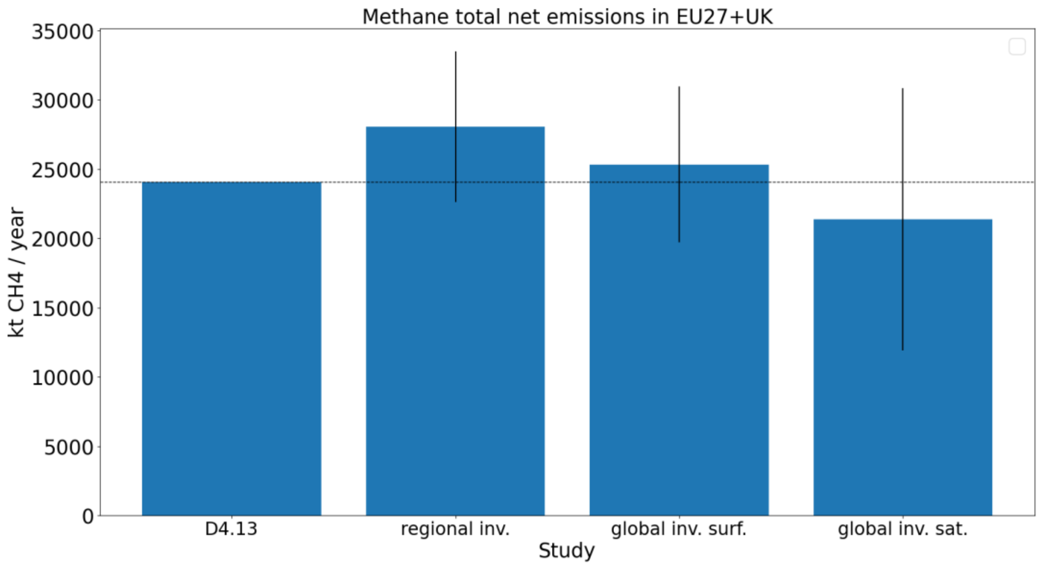

Figure 4: Total net methane emissions (ktCH4/yr) in EU27+UK from inversions. D4.13 is this work. Ranges for other regional inversions and global inversions are taken form Petrescu et al. (2021): see their Fig.4a) and Fig.5a) and Tab.B1, the values reported here are for the last available year (2017 at the latest). Regional inversions assimilate surface stations; some global inversions assimilate surface data, others assimilate GOSAT satellite data. Note: Malta is not included in our domain.

5. Conclusions

The results obtained here have technical as well as scientific interest.
Technically, they show that the CIF is able to use TROPOMI CH_4 data to constrain inversions in Europe at a widely used horizontal resolution (0.5° x 0.5°) and a weekly time resolution for one year in a relatively short computing time (less than one week). This is a key milestone in the development of the CIF. This shows that the CIF could be set-up for inversions with a finer horizontal resolution (if possible, down to 10 km x 10 km), which will be more relevant for national scales for most countries and even regional scales, making use of the information provided by the fine resolution of TROPOMI data.
Scientifically, the retrieved fluxes show patterns which are realistic, such as increased emissions compared to the prior in regions where the contribution from agriculture is large. Moreover, the total fluxes (e.g. in EU27+UK 24,000 ktCH_4 in 2019) are in agreement with other inversions, either regional or global, assimilating either surface or other satellite data (total range in EU27+UK of about 11,900 to 33,500 kt for years previous to 2017).
The next steps are first, the extension of the inversion run here to the whole period of availability of TROPOMI CH4 data (i.e. from May 2018 to the present day); then, a full scientific analysis of the results, with more comparisons to other estimates, either from inventories, biogeochemical models or inversions, and sensitivity studies to the specification of the error statistics.

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