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

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1. Changes with respect to the DoA

None

2. Dissemination and uptake

The model results will be made available via the VERIFY project database and are currently available via a data server (in some cases registration will be necessary). These model results will be used in the synthesis product in WP5.

3. Short Summary of results

This deliverable presents the results of inverse modelling of CH_4 and N_2O. Emissions of CH_4 were estimated using the FLEXPART-ExKF inversion framework, which employs an Extended Kalman Filter (ExKF) and uses the Lagrangian Particle Dispersion Model, FLEXPART to model atmospheric transport (see Section 2.2.2). Emissions of N_2O were estimated using the FlexInvert inversion framework, which finds the maximum posterior probability solution using Bayesian statistics and also employs the FLEXPART transport model (see Section 2.2.1). For both CH_4 and N_2O, the emissions are presented at 0.5°×0.5° resolution and at a minimum of monthly temporal resolution for the period 2005-2017. In the case of CH_4, preliminary results show a significant reduction with respect to the prior emission estimates, whereas for N_2O, the significant increases seen with respect to the prior estimates (see Section 3).

4. Evidence of accomplishment

All the simulation results will be accessible through the dedicated data THREDDS server:

https://verifydb.lsce.ipsl.fr/thredds/catalog/verify/WP3/catalog.html

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).

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1. Introduction

CH_4 and N_2O are the most important long-lived greenhouse gases after CO_2. Due to the nature of CH_4 and N_2O emissions, which primarily originate from bacterial processes, leakages and diffuse sources, uncertainties in these emissions are comparatively large. Top-down emission estimation based on atmospheric observations and transport modeling, therefore, has a great potential to reduce these uncertainties and to identify major issues in current bottom-up inventories.

This deliverable presents top-down estimates of European CH_4 and N_2O emissions for the years 2005-2017 conducted with different transport and inverse modelling systems. State-of-the-art inventories of anthropogenic and natural emissions, which were partly developed within VERIFY, were used as a priori information. Measurements from continuous observations and flask samples were collected from all sites of the gradually expanding network in Europe. To limit the influence of a temporally changing distribution and density of sites on long-term trend estimates, only sites with measurement records of more than 7 years were included in the main inversion.

The study builds on previous top-down estimates of CH_4 and N_2O emissions over Europe (Bergamaschi et al., 2015, 2018; Thompson et al., 2014). It covers a much longer time period than these previous studies and employs more recent a priori information and a larger set of observations. The deliverable will be updated each year by extending the results by another year. Note that the objective is to reach within the project a temporal coverage up to year – 1 at each new release.

2. Set-up

2.1. Transport model description

2.1.1. FLEXPART

FLEXPART is a Lagrangian particle dispersion model that simulates long-range and mesoscale transport, diffusion, as well as dry and wet deposition of tracers released from point, line, area or volume sources (https://www.flexpart.eu). FLEXPART can be used forward in time to simulate the dispersion of tracers from their sources, or backwards in time to determine potential source contributions for given receptors (Stohl et al. 2005; Pisso et al. 2019). FLEXPART is an off-line model and is driven by meteorological fields (either analyses or forecasts).

2.2. Inversion frameworks

2.2.1. FlexInvert

The FlexInvert framework is based on Bayesian statistics and optimizes fluxes using the maximum probability solution (Rodgers 2000). To describe atmospheric transport, FlexInvert uses the Lagrangian model FLEXPART (Stohl et al. 2005; Pisso et al. 2019) run in the backwards time mode. In this mode, virtual particles are released at the times and locations of each

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observation, and their positions are modelled backwards in time following wind fields. From the backwards time simulation, a so-called source-receptor matrix, describing the relationship between the change in a given mole fraction and the fluxes discretized in space and time, can be computed by sampling the near-surface residence times of the virtual particles (Seibert and Frank, 2004). In the simulations used here, the virtual particles were tracked backwards in time for 7 days. A time series of simulated mole fractions can be obtained by integrating the time series of source-receptor matrices with a discretized flux estimate. For use in the inversions, FLEXPART was driven using ECWMF Era Interim wind fields.

The state vector used for D4.7 consists of flux increments (i.e. offsets to the prior fluxes) discretized on an irregular grid based on the source-receptor matrices (Thompson et al. 2014). This grid has finer resolution (in this case the finest is 0.5°×0.5°) where the fluxes have a strong influence on the observations and coarser resolution where the influence is only weak (the coarsest is 2°×2°). The flux increments were solved at 2-weekly temporal resolution. The state vector also includes scalars for the background mole fractions, based on the principle that errors in the *initial fields* of mole fraction will affect the model-observation error in a similar way for different locations (see also paragraph below). FlexInvert can use different methods to solve the inverse problem. For this application, the solution was found using the Conjugate Gradient method (Paige and Saunders, 1975), which enables problems with very large state and observation vectors to be solved efficiently.

The background mole fractions, i.e., the contribution to the modelled mole fractions that is not accounted for in the 7-day backwards time FLEXPART runs, was estimated by coupling the termination points of the virtual particles to *initial fields* of mole fractions from the optimized Eulerian model LMDz (i.e. the CAMS N_2O mole fraction product v18r1) following the method of Thompson et al. 2014.

2.2.2. FLEXPART-ExKF

The FLEXPART-ExKF framework also uses the Lagrangian model FLEXPART (Stohl et al., 2005; Pisso et al., 2019) in the backwards time mode to compute the source-receptor matrix (or footprint) for each observation (Seibert and Frank, 2004). For use in FLEXPART-ExKF, FLEXPART was driven with 3-hourly ERA Interim analyses and forecasts of ECMWF's IFS model. Particles released at observation sites were followed backwards over 10 days but were terminated when they left the domain of Europe (15°E to 35°W, 33°N to 73°N). Potential losses of CH_4 during transport over these 10 days were neglected considering the long lifetime of CH_4 of approximately 10 years.

The estimation of a posteriori emission fluxes for D4.7 is based on the extended Kalman Filter (ExKF) introduced by Brunner et al. (2012) and Brunner et al. (2017). The filter sequentially assimilates all observations of a given day to update the emissions estimated for the previous day to the current day. The filter can simultaneously optimize the emission field (or its logarithm to ensure a log-normal distribution of errors) and the background mole fractions based on the assumption that emissions mainly contribute to peaks in the time series whereas the background contributes to the smoothly varying baseline. Alternatively, the background

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concentrations computed by a global model can be used with (or without) further optimization. The filter includes a forecast step describing the evolution of the state vector from one time step to the next. The simplest assumption is persistence (i.e. no change with time), but to incorporate seasonally varying a priori emissions, a non-zero forecast update was implemented according to the change in a priori emissions from one month to the next. Since the forecast step is associated with an uncertainty, the posterior uncertainty can become larger than the prior uncertainty, which is different from a classical Bayesian inversion where posterior uncertainty is always lower.

The state vector consists of the following components: (i) emissions on a regular grid of 0.5°x0.5° resolution covering the domain (15°E to 35°W, 33°N to 73°N), (ii) background mole fractions at all observation sites, (iii) trends in background mole fractions at all sites, (iv) coefficients of an AR(1) autoregressive process describing temporal correlations in the residuals at all sites. Emission estimates are provided on a monthly basis.

Two separate inversions were conducted, which differed in the way background mole fractions were obtained. In the first inversion, background mole fractions were directly estimated by the filter as described in Brunner et al. (2012). In the second inversion, background mole fractions were taken from a global TM5-4DVAR simulation where emissions over Europe were excluded. In this case, no further optimization of the background was conducted and the state vector was reduced to emissions and autoregression coefficients.

2.3. Atmospheric observations

2.3.1. Methane

Atmospheric observations of CH_4 mole fractions were compiled from a number of sources: 1) the InGOS project harmonized dataset, which approximately covers the period from 2005 to 2014; 2) the World Data Centre for Greenhouse Gases (WDCGG, https://gaw.kishou.go.jp); 3) the NOAA ESRL discrete sampling network (https://www.esrl.noaa.gov/gmd/); 4) the EBAS data base (http://ebas.nilu.no), and 5) personal communications of data from station principle investigators. Based on these, a nearly continuous dataset over 2005-2017 comprising 31 stations was compiled, including 11 discrete sampling sites and 20 in-situ sampling sites. Of these there are 9 tall towers, 11 mountain sites, 5 coastal sites and 6 short or near-surface continental sites.

For tall towers, near-surface and coastal sites, observations between 12 and 15 UTC were assimilated, whereas for the mountain sites observations between 0 and 3 UTC were assimilated, to reduce the impact of errors in the modelled boundary layer heights and up/down slope winds.

To align the measurements with the 3-hour time resolution of the FLEXPART transport simulations, the measurements and their uncertainties were averaged over 3-hour intervals. Reported measurement uncertainties were used whenever provided. Assuming temporally uncorrelated errors, the uncertainty of the 3-hour average was reduced by square_root(*N*), with *N* the number of samples per 3 hours. A minimum uncertainty of 2 ppb corresponding to the

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WMO compatibility goal for CH_4 was assumed (Zellweger et al., 2016), which was also applied when no uncertainty was reported.

2.3.2. Nitrous oxide

Atmospheric observations of N_2O mole fractions were similarly compiled from the sources 1-3 and 5 listed in Section 2.3.1. From these, a nearly continuous dataset over 2005-2017 was compiled consisting of 21 stations, including 7 discrete sampling sites and 14 in-situ sampling sites. Of these there are 6 tall towers, 6 mountain sites, 4 coastal sites, and 5 near-surface continental sites.

Similarly to CH_4, observations from tall towers, near-surface and coastal sites were assimilated during daytime, between 11:00 and 17:00 local time, and for mountain sites during nighttime, between 22:00 and 04:00 local time. For N_2O, observations between these times were assimilated hourly, which was also the temporal resolution of the FLEXPART simulations.

A minimum uncertainty of 0.3 ppbv was used to represent random measurement errors, and where specific uncertainty estimates were available for a given station, and it exceeded this minimum error, it was used instead.

2.4. Prior fluxes

2.4.1. Methane

Anthropogenic emissions

Emission estimates for anthropogenic sources (including agriculture, energy use, transport, and industry) were taken from the EDGAR model version 5 (see D4.1). These were provided globally at 0.1°×0.1° and monthly resolution.

Wetland emissions and soil uptake

Emissions from wetlands for the European domain (34.5°-73.5°N by 10.5°W-33.0°E) were taken from the JSBACH-HIMMELI model (see D4.4). These were provided at 0.1°×0.1° and daily resolution. Outside this domain, emission estimates were taken from the Global Carbon Project (GCP) CH_4 data, which were used as prior information for the 2019 budget (not published yet, see Saunois et al. (2016) for the previous budget). The wetland emissions are the mean climatology of 11 process-based models (described in Poulter et al. 2017). For this work the data were provided by GCP at a 0.5°×0.5° resolution. Methane can also be lost by oxidation in dry soils, this negative emission was estimated by JS-BACH over the European domain at 0.1°×0.1° and daily resolution. Outside this domain, the soil sink was based on the climatology of Ridgwell et al. (1999) with developments by GCP for the 2019 budget (not published yet, see Saunois et al. (2016) for the previous budget) and provided at a 1°×1° resolution.

Inland water bodies

Emissions from inland water bodies (such as lakes and reservoirs) were taken from an empirical model that scales-up measurements at field scale to the European domain using proxy data (see

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D4.4). This estimate is an annual climatology and covers the domain 26°W-55°E by 34°N-78°N at 0.1°×0.1° resolution. Since no other spatially resolved estimate for this source type could be found, emissions outside of this domain were set to zero.

Biomass burning

Estimates of the biomass burning source of CH_4 were taken from the Global Fire Emissions Database (GFED-v4.1s, https://www.globalfiredata.org). These estimates are monthly and are provided at 0.25°×0.25° resolution.

Ocean

Emissions from the ocean were based on the climatology by Weber et al. (2019), which is also the prior used by GCP for the 2019 budget (not published yet). The emissions were provided at a 1°×1° resolution.

Geological

The geological emissions were taken from the climatology by Etiope et al. (2019), which was also used by GCP for the 2019 budget (not published yet, see Saunois et al. (2016) for the previous one) and were provided at a 1°×1° resolution. They include geothermal manifestations, microseepage, as well as onshore and offshore seeps.

Termites

Emissions from termites were taken from a climatological estimate determined by GCP for the 2019 budget (not published yet, see Saunois et al. (2016) for the previous one) and were provided at 1°×1° resolution.

All emissions were averaged/interpolated to 0.5°×0.5° to match the spatial resolution used by the inversions. In addition, the emissions were averaged/interpolated in time to monthly resolution.

2.4.2. Nitrous oxide

Anthropogenic

Anthropogenic emissions of N_2O (including direct and indirect soil emissions, waste water, manure management, industry and energy) were taken from the EDGAR model version 5 (see D4.1). These were provided globally at 0.1°×0.1° and monthly resolution.

Natural soils

Natural soil emissions of N_2O (or the so-called *background* emission) was assumed to be equivalent to the pre-industrial soil emission of N_2O. This was estimated using the land-surface model, OCN (Zaehle et al. 2011). The emissions were provided as a climatology at 1°×1° resolution and monthly resolution.

Biomass burning

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Estimates of the biomass burning source of N_2O were taken from the Global Fire Emissions Database (GFED-v4.1s, https://www.globalfiredata.org). These estimates are monthly and are provided at 0.25°×0.25° resolution.

Ocean

Ocean emissions were taken from a prognostic model of N_2O production, which is embedded in the ocean biogeochemistry model, PlankTom10, (Buitenhuis et al. 2018). This model uses the observed correlation between apparent oxygen utilization (AOU) and excess N_2O (ΔN_2O) in oxic waters to estimate the open ocean source of N_2O production and the increased yield of N_2O in suboxic waters. These emissions were provided as a monthly climatology at 1°×1° resolution.

All emissions were averaged/interpolated to 0.5°×0.5° to match the spatial resolution used by the inversions. In addition, the emissions were averaged/interpolated in time to monthly resolution.

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3. Results

3.1. CH_4 emissions for the period 2005-2017

Inversions of CH_4 have been performed using the FLEXPART-ExKF framework for Europe (15°W-35°E by 33°N-73°N) for the period 2005-2017.

Results from the two inversions with (1) optimized baseline and (2) TM5-4DVAR baseline, respectively, and using the prior fluxes and observations described in Sections 2.3 and 2.4, are presented in Figures 1-2 below. Figure 1 shows the generally good performance of the transport model FLEXPART with respect to reproducing the observed CH_4 time series. Results are shown here for inversion (1) but are similar for inversion (2). The a posteriori simulated values tend to be much closer to the observations than the a priori with reduced root-mean-square-errors (RMSE) and improved correlations (R^2). Biases in the a priori are mostly corrected well except for Pallas where the relatively sparse observations provide insufficient constraints on both the emissions and the baseline. At the northernmost sites MHD and PAL, the a priori is largely overestimated suggesting too high a priori emissions in these regions.

A posteriori emissions are compared with a priori emissions in Figure 2. Overall, the model estimates a significant reduction compared to the a priori except for the area of the Benelux countries and parts of southwestern Germany. Large enhancements are also estimated for the easternmost parts of the model domain which are poorly constrained by observations. These enhancements are likely an artefact of the approach, which estimates background concentration levels irrespective of the origin of the air mass. The largest reductions are estimated for regions with large a priori natural fluxes such as Ireland and UK (large wetland emissions) or northern Italy (large geological emissions).

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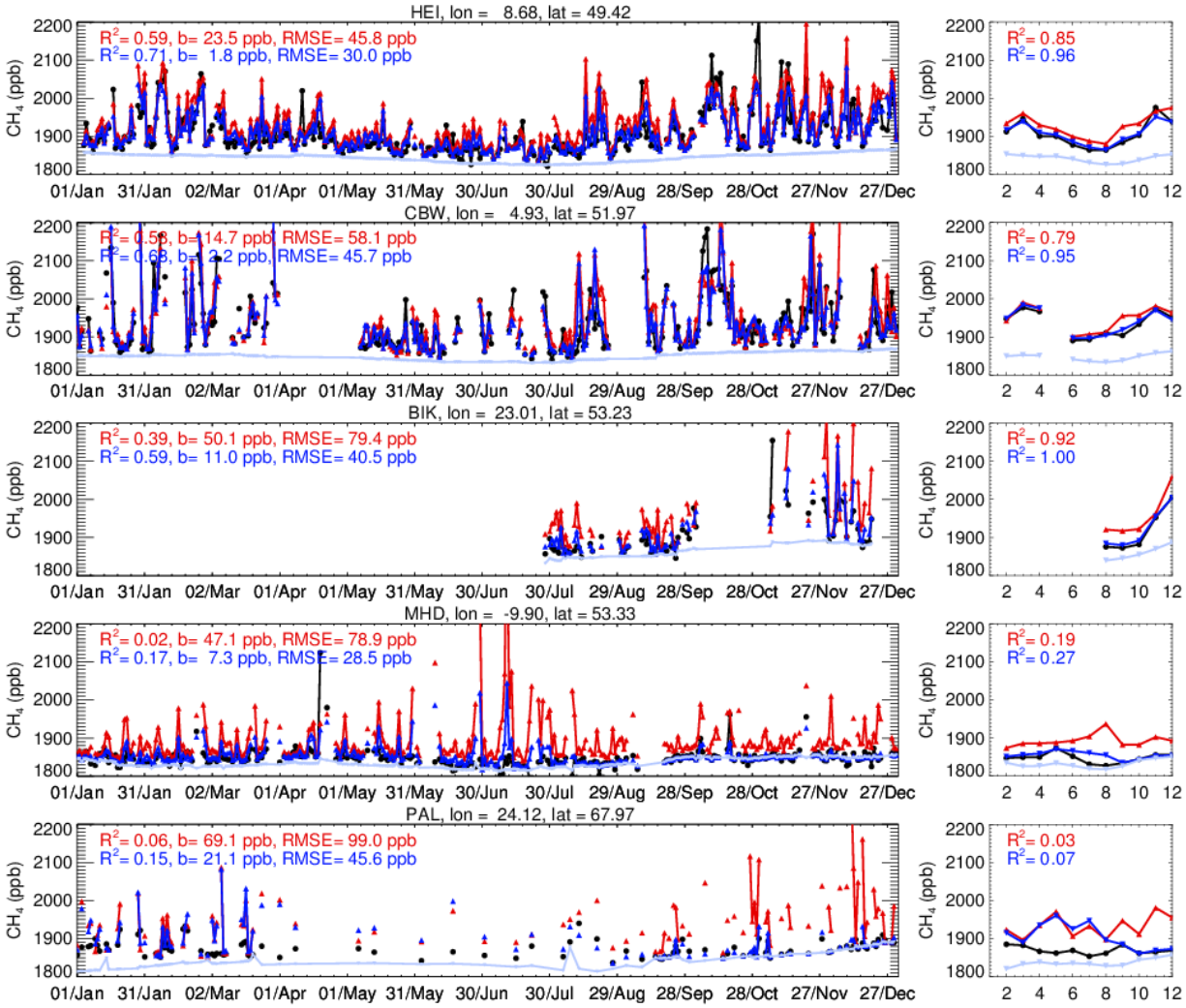

Figure 1: Simulated (red: a priori, blue: a posteriori) versus observed CH_4 mixing ratios at a few sites in 2005. The light blue line is the baseline, which is optimized together with the emissions. The observation sites are Heidelberg (HEI), Cabauw (CBW), Bialystok (BIK), Mace Head (MHD), and Pallas (PAL). The righthand panels show the seasonal cycle of monthly mean values

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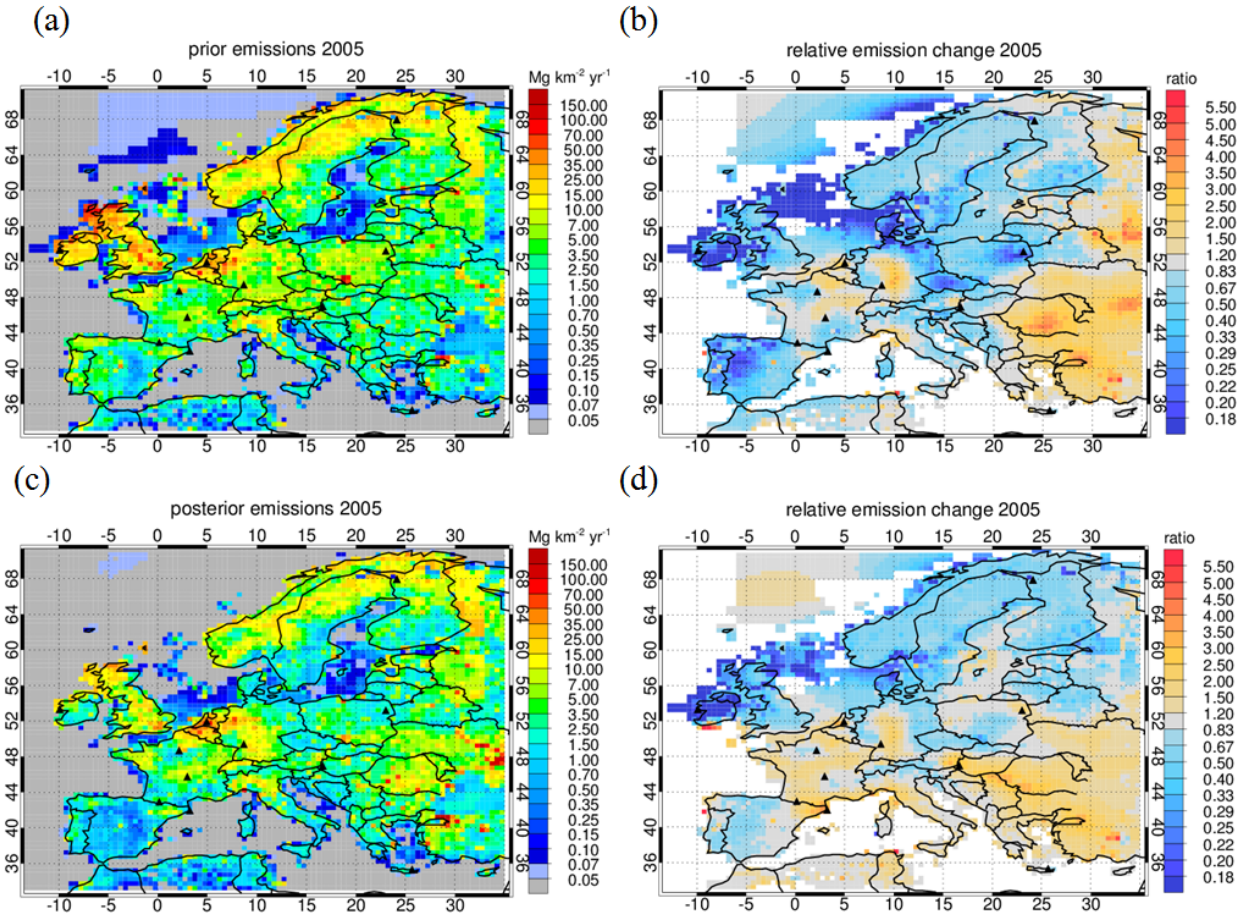

Figure 2: Comparison of prior (top left) and posterior emissions (top right) in 2005. The lower panels show the absolute (bottom left) and relative (bottom right) emission changes. Locations of stations assimilated in the model are shown as black triangles

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3.2. N_2O emissions for the period 2005-2017

Emissions of N_2O were estimated using the FlexInvert inversion framework for Europe (15°W-35°E by 33°N-73°N) for the period 2005-2017. The inversion used the prior fluxes and observations as described in Sections 2.3 and 2.4 as described above. The inversions were separated by year and run from 1-Jan to 31-Jan of the proceeding year to include the constraint of observations on fluxes up to several days afterwards.

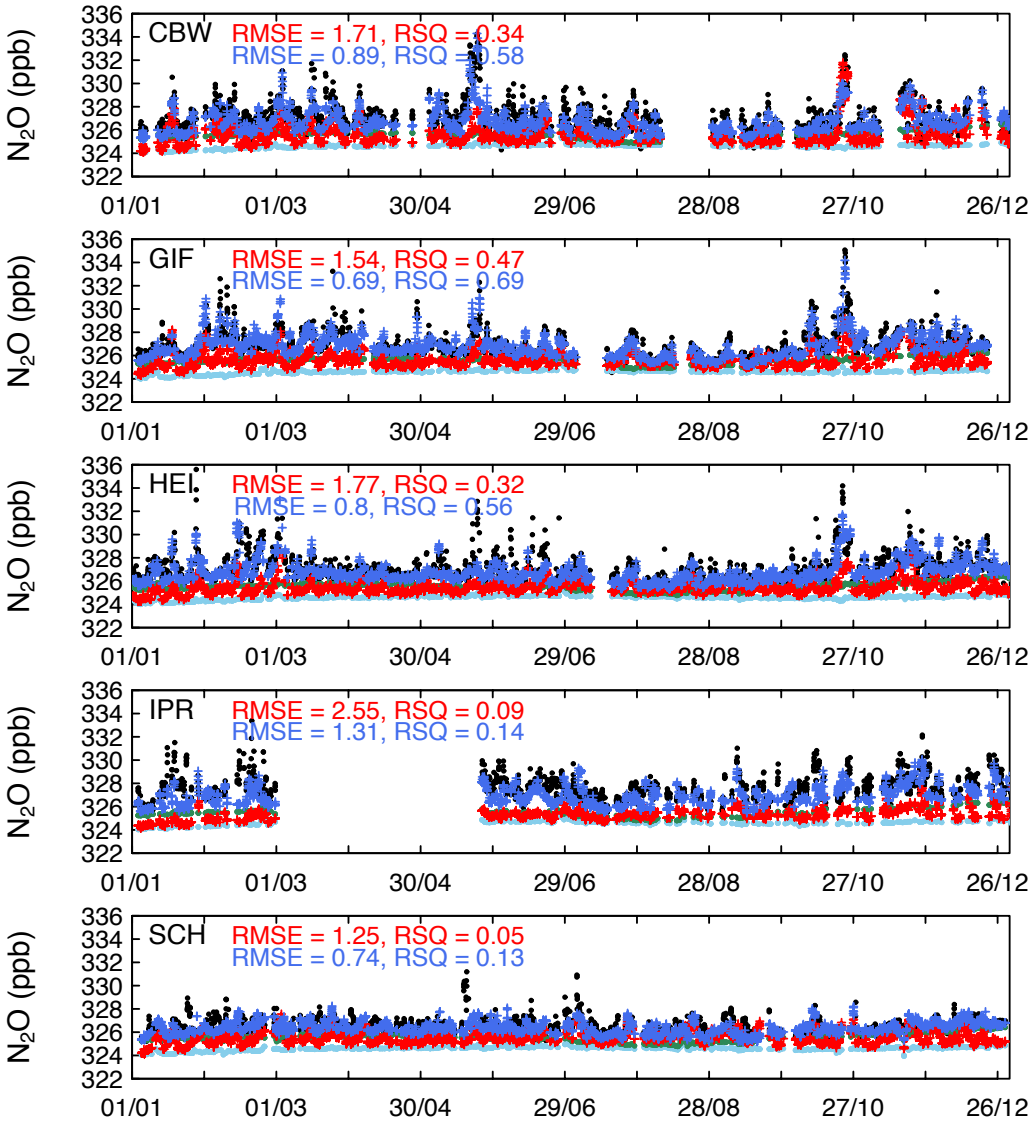

Figure 3: Comparison of modelled (red: a priori, blue: a posteriori) versus observed (black) N_2O mole fractions (ppb) at continental sites for 2012. The light blue and green points indicate the prior and posterior modelled background mole fractions, respectively. The sites are Cabauw (CBW), Gif sur Yvette (GIF), Heidelberg (HEI), Ispra (IPR) and Schauinsland (SCH).

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Figure 3 shows the comparison of the prior and posterior modelled N_2O mole fractions with observations at a number of continental sites for the example year 2012. At all sites, the optimized background mole fraction is higher by approximately 1 ppbv than that a priori, indicating that the initial mole fraction fields from the global Eulerian model used in CAMS have a low bias. As expected, the posterior modelled mole fractions are closer to the observations compared to those a priori, with lower RMSE and higher R^2 statistics.

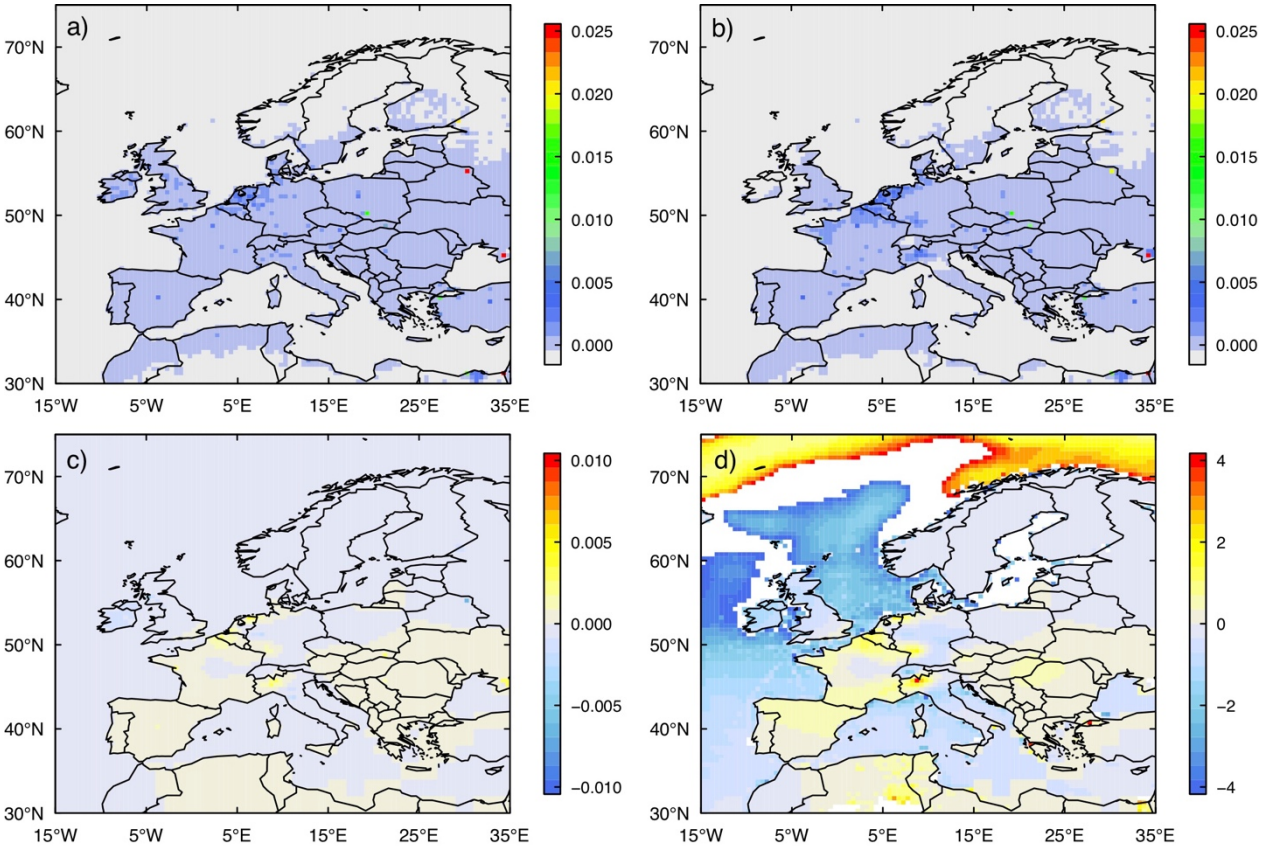

Figure 4. a) prior flux estimates (gN m-2 day-1), b) posterior flux estimates (gN m-2 day-1), c) flux difference posterior-prior (gN m-2 day-1) and d) flux difference relative to the prior estimate.

The prior and posterior emissions are compared in Figure 4. The posterior emissions are higher in most of France, Spain, and the Benelux countries, as well as in southeastern Europe. There are also some increases around the southern boundary of the domain, which may be due to the fact that the emissions are poorly constrained in this region, and as a partial compensation for the low background mole fractions.

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4. Planned developments

4.1. FlexInvert

Planned developments of FlexInvert include improvements to the computational efficiency and parallelization of the most time consuming processes. This should significantly improve the run time of large inversion problems. In addition, the method for calculating and optimizing the background mole fractions will be further studied, to see where improvements may be made.

4.2. Flexpart-ExKF

The set-up of Flexpart-ExtKF will be further refined, for example by testing different spatial correlations of the prior uncertainties or testing different magnitudes of the uncertainties of the emission and background predictions. Different settings will be checked for consistency with χ^2-statistics. Furthermore, the time resolution of the output will be increased from monthly to two-weekly.

The influence of the treatment of background concentrations will be further investigated by systematically comparing the results of three different approaches. In addition to the two approaches presented above, a third inversion will make use of a background that is computed by the TM5-4VAR system, and a third inversion where the background is computed by interpolating the 3D CH_4 fields of TM5-4DVAR to the end positions of the particles computed by FLEXPART. This inversion is expected to yield similar results as the inversion using the TM5-4DVAR background.

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