MEMO²: MEthane goes MObile – MEasurements and MOdelling

Top-down estimates of EU-scale CH_4 emissions

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Deliverable 3.4

Delivery month Annex I	42		
Actual delivery month	48		
Lead participant: Empa	Work package: 3	Nature: Report	Dissemination level: PU
Version: 01			

This project has received funding from the European Union’s Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No 722479.

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1. Executive Summary

Backward Lagrangian atmospheric transport simulations have been performed and used in combination with ground-based CH_4 dry air mole fraction measurements to quantify European CH_4 emissions for the year 2018. The simulations were conducted with the Lagrangian particle dispersion model FLEXPART-COSMO, which has also been used for simulations of stable CH_4 isotopes at the station Lutjewad, the Netherlands, presented in deliverable D3.3.

Emissions were estimated "top-down" using an extended Kalman Filter by assimilating the measurements from 13 stations of the European Integrated Carbon Observation System (ICOS) network and weekly flask samples from 5 sites of NOAA's global monitoring network. The Extended Kalman filter improves upon an *a priori* emission estimate by sequentially assimilating the observations day by day. The estimated *a posteriori* emissions minimize the difference between simulated and observed CH_4 while considering the uncertainties in the transport model, the observations, and the a priori emissions. A priori anthropogenic and natural emissions were taken from state-of-the-art inventories including TNO's high-resolution inventory of anthropogenic CH_4 emissions in Europe.

As a first check of the quality of the model system, we compared the simulated CH_4 mole fractions with the observations. At most stations we found an excellent agreement, which demonstrates the high quality of the atmospheric transport model and justifies its application for top-down emission quantification.

A posteriori emissions were estimated on a grid covering large parts of Europe (except for Scandinavia and the easternmost parts of Europe). The difference between a posteriori and a priori emissions suggests that some countries like France or Belgium likely underestimate their emissions. The results also suggest that emissions from coal mining activities are likely smaller than previously assumed.

2. Introduction

2.1 Background

CH_4 is the second most important long-lived greenhouse gas after CO_2. The amount of CH_4 in the atmosphere has strongly increased since pre-industrial times (Saunois et al., 2019), and in recent years the rate of increase has accelerated. In the last decade, about 60 % of the global CH_4 source has been due to human activities, namely, agriculture, waste, fossil fuels and biomass burning. Natural sources of CH_4 include wetlands, wildfires, termites and geological sources.

A large portion of the CH_4 emissions is due to microbial processes sensitive to environmental conditions or due to leakages and diffuse release processes. Bottom-up inventories of CH_4 emissions therefore have large uncertainties. Top-down emission estimation based on atmospheric observations and transport modelling has a great potential to reduce these uncertainties and to identify major issues in current inventories.

2.2 Scope of the deliverable

This deliverable presents a top-down estimate of European CH_4 emissions for the year 2018 produced with the Lagrangian atmospheric transport model FLEXPART-COSMO and using TNO's latest anthropogenic CH_4 emission inventory and state-of-the-art inventories of natural emissions produced in the EU project VERIFY as a priori information.

Measurements from continuous observations of the European ICOS network and weekly flask samples from NOAA were assimilated to compute optimized a posteriori emission flux. The study builds on previous top-down estimates of CH_4 emissions over Europe (Bergamaschi et al., 2015, 2018) but provides output on a much higher spatial (up to 7 km x 7 km) and temporal (monthly) resolution.

3. Methodology

3.1 Atmospheric transport model simulations with FLEXPART-COSMO

Atmospheric transport simulations were conducted with the Lagrangian particle dispersion model FLEXPART (Stohl et al., 2005; Pisso et al., 2019) in a special high-resolution version called FLEXPART-COSMO (Henne et al., 2016). FLEXPART-COSMO was driven by hourly meteorological analysis fields from the numerical weather prediction model COSMO produced operationally by the Swiss weather service MeteoSwiss for the domain of Europe at a resolution of approximately 7 km x 7 km.

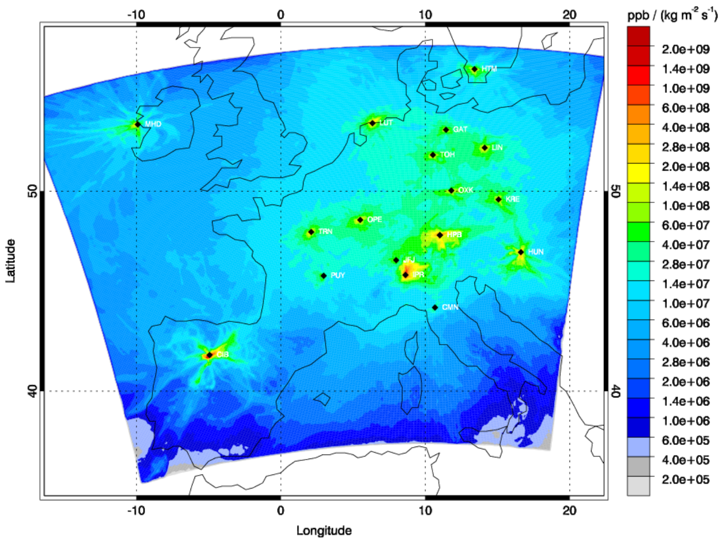

Fig. 1: Mean footprint in January 2018 of all sites (black diamonds) assimilated in FLEXPART-COSMO.

For all observation sites (see Sect. 3.3) 50000 air parcels (so-called *particles*) were released every 3 hours during the year 2018 and traced backward in time over four days to compute the source-receptor matrix (*footprint*) for each observation (Seibert and Frank, 2004). Potential losses of CH_4 during transport over these 4 days were neglected considering the long lifetime of CH_4 of approximately 10 years.

An example of the total footprint (average of all individual footprints) for all sites for a single month is presented in Fig. 1. The figure shows the model domain covering central Europe and the location of the sites (black diamonds). The color contours represent the sensitivity of the measurements to CH_4 emissions over Europe. The sensitivity is low in the southern parts of Europe due to a lack of observation sites.

3.2 Inversion framework FLExKF

The inversion framework is based on the extended Kalman Filter (ExKF) described 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 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 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.

In the setup used here, the logarithm of emissions was optimized to ensure a positive solution. Background mole fractions were not optimized but were taken from a global CH_4 assimilation of the TM5-

4DVAR model conducted by Peter Bergamaschi (JRC, Ispra) for the VERIFY project. Following the two-step approach for nested global and regional inversions proposed by Rödenbeck et al. (2009), the TM5-4DVAR simulation was conducted in a way that excludes contributions from emissions over the European domain covered by FLEXPART-COSMO. A priori anthropogenic and natural emissions were taken from various data sources as listed in Table 1. Anthropogenic emissions were taken from TNO's latest high-resolution emission inventory TNOGHGco. A posteriori emissions were estimated on an irregular grid with higher resolution near the measurement sites and lower resolution with increasing distance as described in Brunner et al. (2012). The grid accounts for the higher sensitivity in the near field of the measurement sites. In total, emissions were estimated for 3371 individual grid cells.

Table 1: A priori emissions used in the inversion

Type	Time resolution	Inventory	Reference
Anthropogenic	annual (2015)	TNOGHGco	Kuenen et al. 2014, EU project CHE
Wetlands	monthly (climatology)	Poulter et al. (2017)	GCP-CH4
Geological	annual (climatology)	Etiope (2008) scaled to total of 15.7 Tg/yr (Petrenko 2017)	GCP-CH4
Ocean	annual (climatology)	Lambert et al. (1993)	GCP-CH4
Termites	annual (climatology)	Castaldi et al. (2006)	GCP-CH4

3.3 Observations

Observations of 13 sites of the ICOS network with continuous CH_4 measurements and of 5 NOAA sites with discreet weekly or biweekly flask samples have been used. The location of the sites is shown in Fig. 1. In order to avoid systematic errors due to the inability of the model to reproduce shallow planetary boundary layers during nighttime, only daily mean afternoon values (12-15 UTC) were assimilated for low elevation sites. For mountain sites located above the nighttime planetary boundary layer, on the other hand, mean values (0-3 UTC) during nighttime were assimilated.

4. Results

Fig. 2 presents examples of the comparison between simulated CH_4 at two stations of the ICOS monitoring network. A priori simulated values (i.e. FLEXPART-COSMO simulations using the a priori emission inventories) agree very well with the observations. Correlations (r^2) are as high as 0.65 for the site KRE and 0.8 for the site LIN, suggesting that the simulations capture 65 % and 80 % of the observed variance, respectively. As expected, the a posteriori simulation shows even better correlations and lower root mean square errors. The figure demonstrates the high quality of the atmospheric transport simulations and the successful assimilation of the measurements.

A map of the a posteriori emissions estimated by the FLExKF inversion system for the year 2018 is presented in Fig. 3.b. The emissions are compared to the a priori emissions (panel a), which are dominated by anthropogenic emissions taken from the TNOGHGco inventory for the year 2015. Total anthropogenic emissions per country in the TNO inventory are consistent with the values officially reported in National Inventory Reports (NIR) to the United Nations Framework Convention for Climate Change (UNFCCC). The difference between a posteriori and a priori emissions (panel c) suggests e.g. a significant underreporting by France, Belgium and Portugal. The results for Portugal, however, should be interpreted with care, since they are constrained by only one measurement site in Spain. In other countries like United Kingdom, Germany or Italy, there is a strong spatial rearrangement of emissions. In UK, for example, the inversion suggests higher emissions in the south but lower emissions in the north as compared to the a priori.

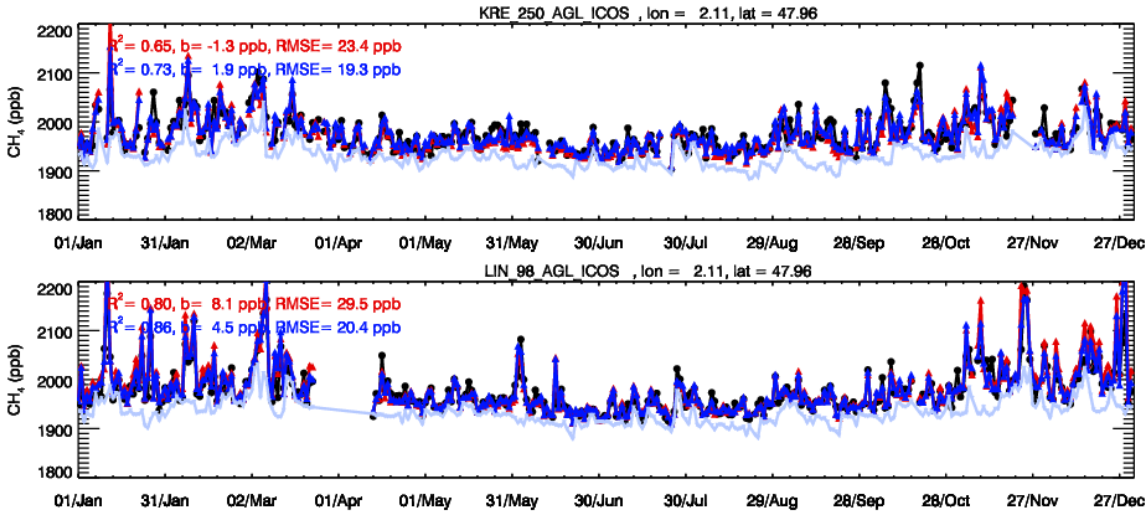

Fig. 2: Comparison of simulated and measured CH_4 at the two ICOS sites Křešín u Pacova, Czech Republic (KRE) and Lindenberg, Germany (LIN) in 2018. Background mole fractions from TM5-4DVAR are shown in light blue. A priori simulated values are shown in red, a posteriori values in blue. Measured CH_4 mole fractions are shown in black. Correlations and root mean square errors (RMSE) between model and observations are also given.

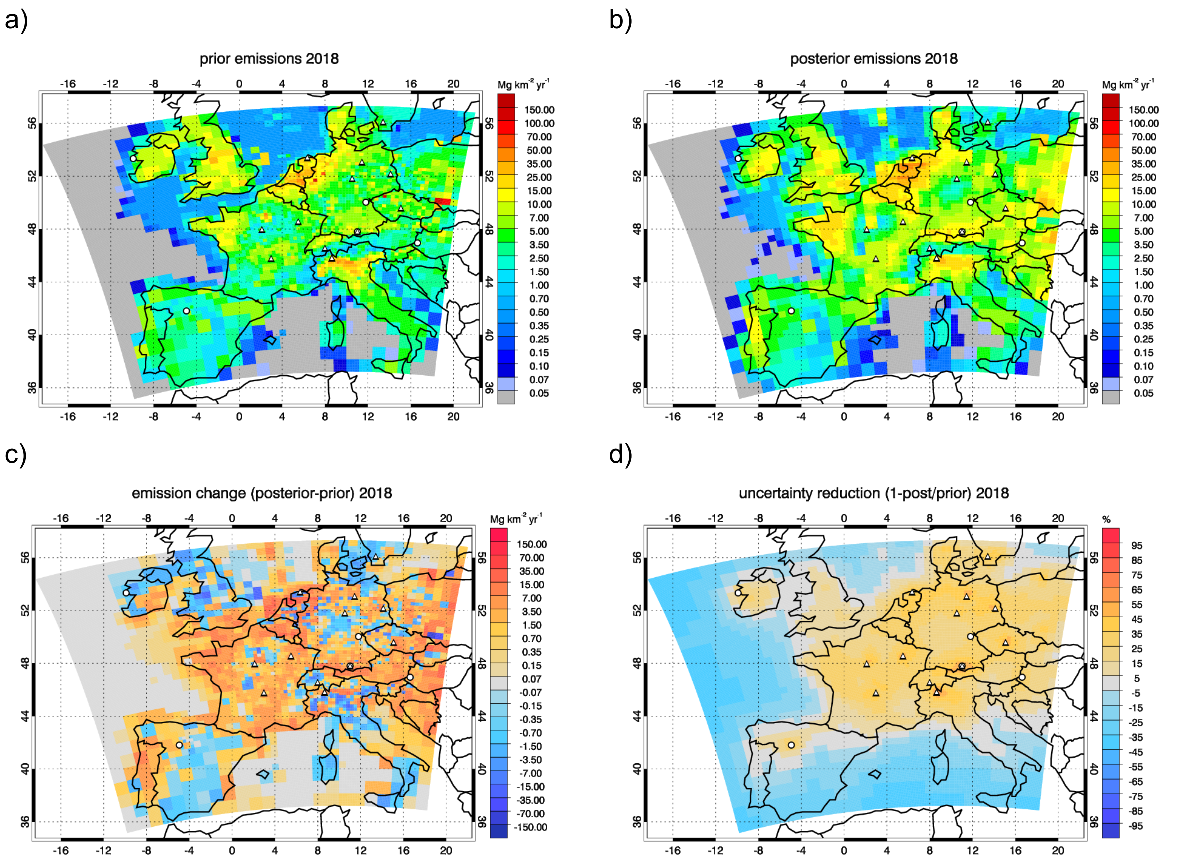

Fig. 3: (a) A priori emissions of anthropogenic and natural sources. (b) A posteriori emissions after assimilation of the observations. Sites with continuous measurements are shown as triangles, flask sampling sites as circles. (c) Difference between a posteriori and a priori. (d) Uncertainty reduction in percent.

Interestingly, many of the emission hot spots seen as red pixels in the a priori, which are associated with cites (e.g. Paris) or with coal mining activities in the German Ruhr area, the Czech Republic and in the Upper Silesia region in Poland, largely disappear in the a posteriori, suggesting that this type of emissions is overestimated. Emissions due to fossil fuel exploration activities in the North Sea, on the other hand, tend to be higher in the a posteriori than in the a priori. The reduction of the uncertainty in the emissions (panel d) shows that the current observation network is insufficient to constrain emissions in southern Europe.

4. Conclusion and possible impact

The MEMO^2 project has produced invaluable data on CH_4 emissions from individual sources using mobile measurements in close vicinity of the sources. Ultimately, this information will be used to improve bottom-up emission inventories. A further step to verify and improve bottom-up inventories is to use CH_4 measurements from regionally representative stations such as those of the ICOS network in combination with regional atmospheric transport models. The application of such an inverse modelling framework was demonstrated in this Deliverable.

Local CH_4 observations downwind of individual sources and regionally representative measurements sensitive to the integrated signal of a large number of sources thus provide highly complementary information to reduce uncertainties in current CH_4 emission inventories.

5. Dissemination & Exploitation

This report will be available via the MEMO2 website to all project partners. Similar simulations though at lower resolution contributed to the CH_4 synthesis publication of Petrescu et al. (2020). The high-resolution model setup developed here will be compared to another high-resolution inversion framework developed in the VERIFY project and is planned to be published by Bergamaschi et al. later in 2021.

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