Atmospheric monitoring of methane emissions at the European scale

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Introduction

CH_4 emissions are a major contributor to Europe's global warming impact but they are not well quantified yet. To quantify sources with adequate uncertainties, we would like to use the information from atmospheric measurements on top of the information from bottom-up inventories. This is made possible with data assimilation methods, from which we want to retrieve top-down emissions.
The objective of this project is to derive a new CH_4 emission map, including isotopic information, across Europe and to evaluate its accuracy.

In this project, forward simulations of CH_4 have been performed with the CHIMERE chemistry-transport model using two emission inventories. The simulation outputs have been compared to surface measurements of the World Data Center for Greenhouse Gases (WDCGG) dataset.

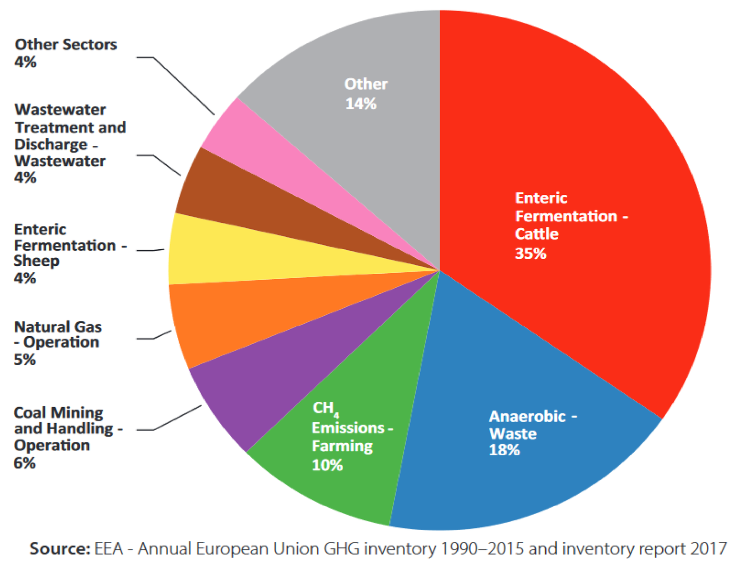

Figure 1: Source categories of CH_4 emissions for EU-28 and Iceland in 2015 (EEA, 2017).

Inventories

For the simulations, the following emission inventories have been used (Kuenen et al., 2014 and Janssens-Maenhout et al, 2017):

Table 1: Description of the inventories

Inventory	EDGAR version 4.3.2	TNO-MACC_III
Coverage	Global	Europe
Spatial resolution	0.1° x 0.1°	0.125° x 0.0625°
Resolution	Monthly and yearly	Yearly
Available years	1970-2012	2000-2011
Available specifications	Sector- and country-specific (NFR code)	Sector- and country-specific (SNAP code)

Model description and setup

CHIMERE is a Eulerian chemistry-transport model designed for regional atmospheric composition (Mailler et al., 2017). The grid resolution ranges from 4 km for urban-scale to about 50 km for regional-scale. It calculates and provides the atmospheric concentrations of gas-phase and aerosol species.
CHIMERE is driven by the PYVAR-CHIMERE system, which makes it possible to run forward simulations and variational data assimilation for emission fluxes.

Table 2: Simulation setup

Species	Total CH_4
Period simulated	2011-2015
Type of emissions	Anthropogenic
Horizontal resolution	50 km
Nr. of vertical levels	29
Top pressure in the vertical	300 hPa
Appr. thickness of vertical layers	From about 10 m at the surface level up to about 1700 m at the top of the domain
Bundary and initial concentrations	LMDz

First results

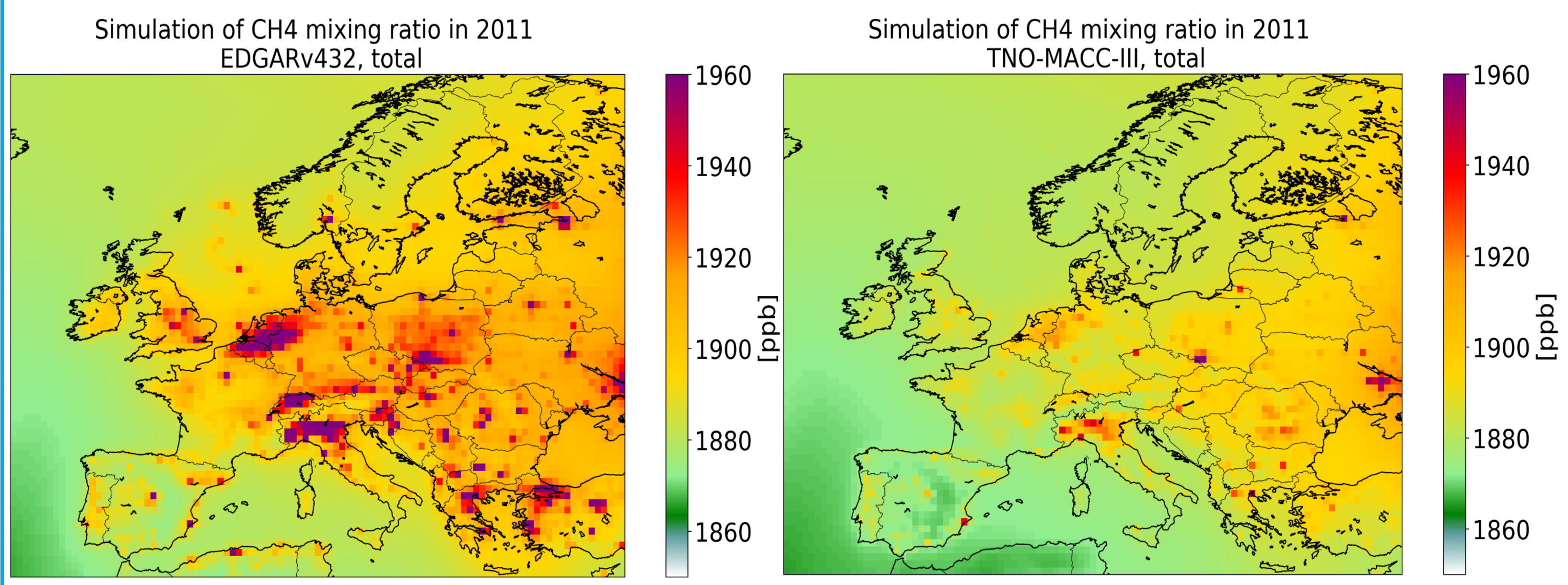

Figure 2: Simulated annual mean CH_4 mixing ratios at the surface level with CHIMERE using the EDGARv4.3.2 (*left*) and the TNO-MACC_III (*right*) emission inventories.

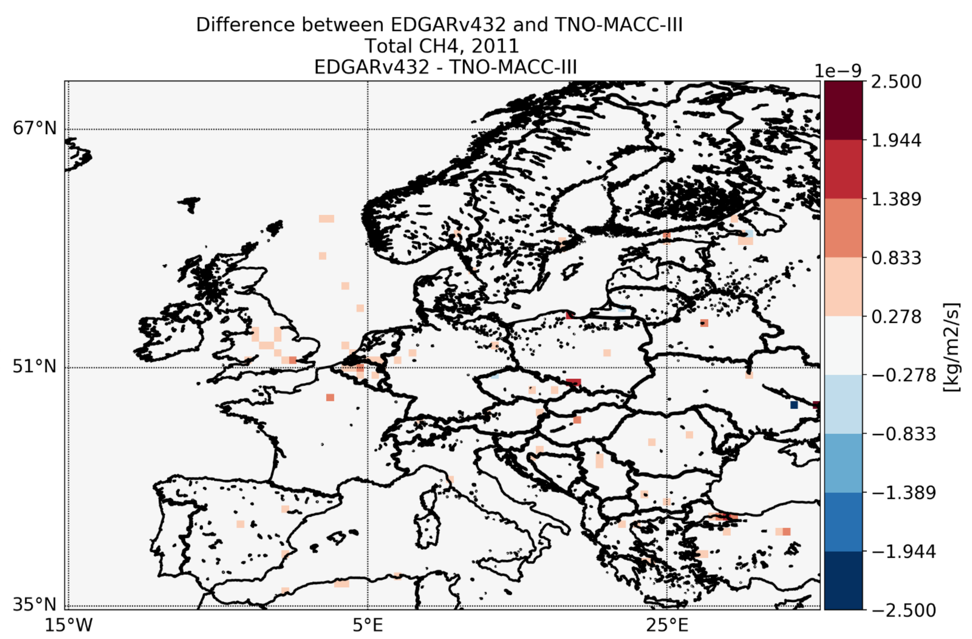

Figure 3: Differences between total anthropogenic CH_4 emissions.

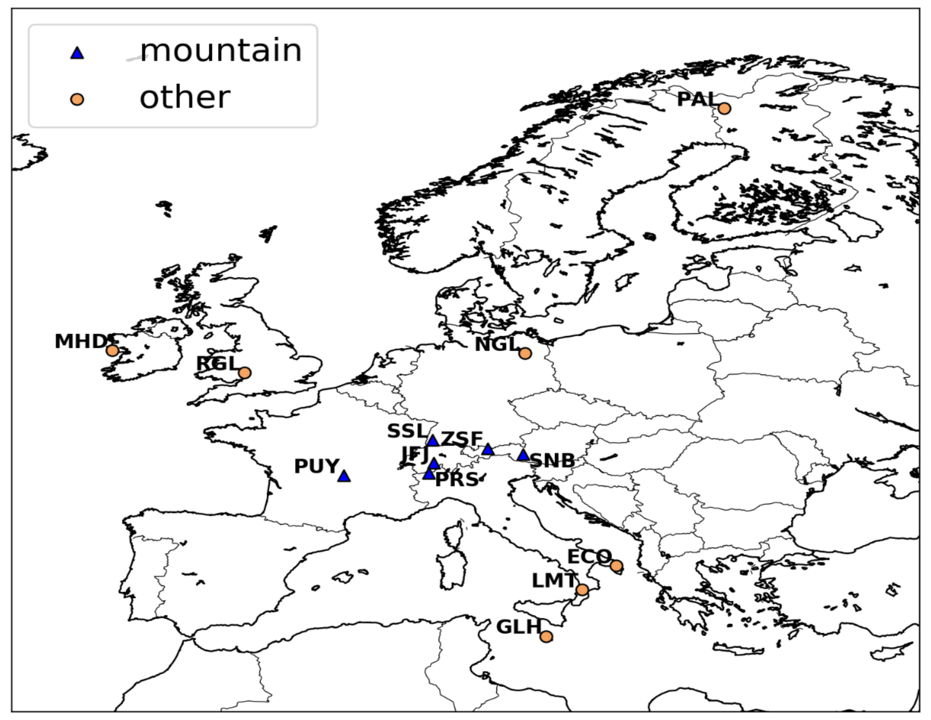

Figure 4: Locations of the measurement sites used for analysis.

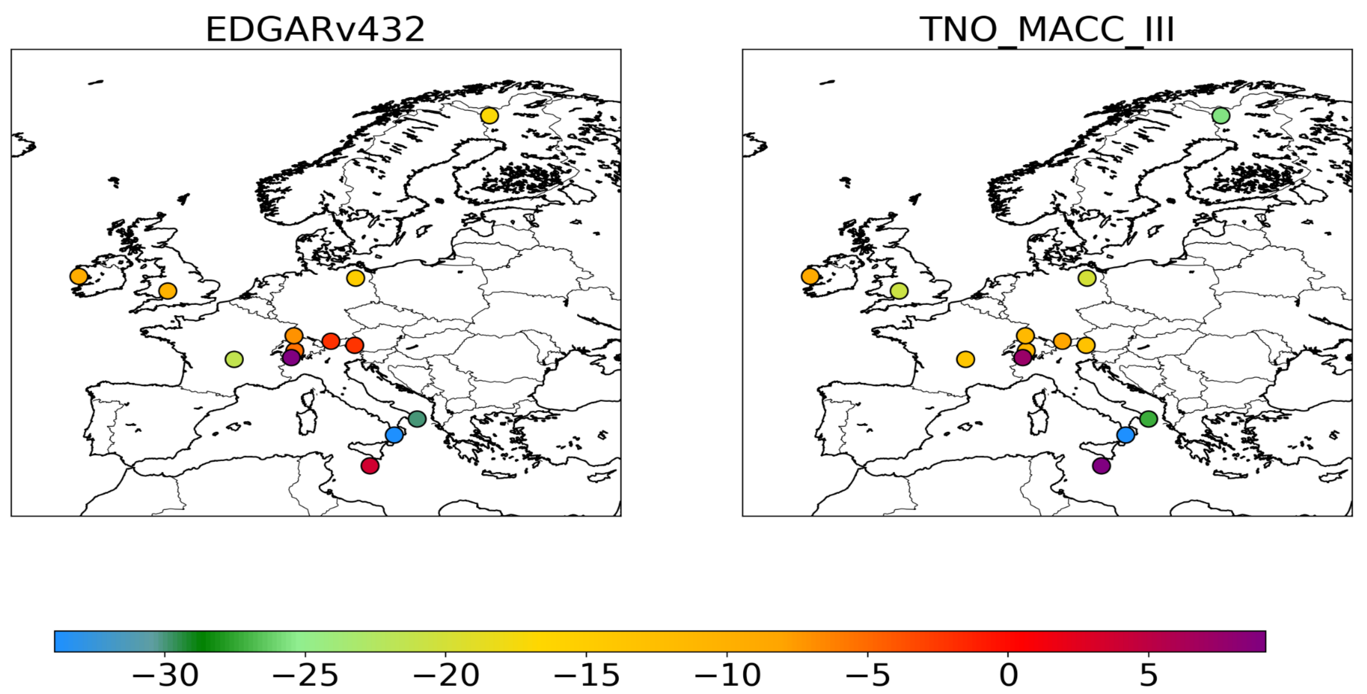

Figure 5: Bias between 2011-2015 averaged hourly afternoon (2–6 pm) values of measured and simulated CH_4 using the two inventories.

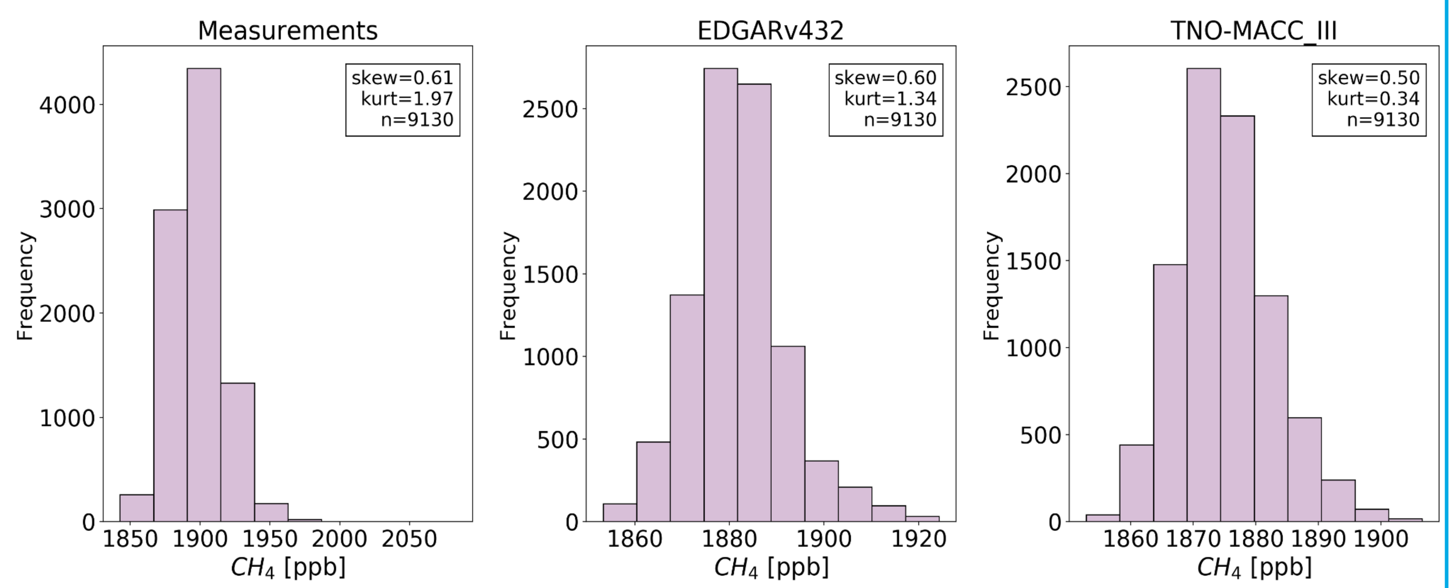

Figure 6: Histograms of 2011-2015 averaged hourly afternoon (2–6 pm) values of measured and simulated CH_4 mixing ratios using the two inventories.

Inverse modelling

The atmospheric inversion approach uses optimization methods to estimate surface fluxes from atmospheric measurements of concentrations. Therefore, we work in a Bayesian framework with Gaussian assumptions. The inversion consists of minimizing:

$$J(x) = (x - x_b)^T B^{-1}(x - xb) + (y - Hx)^T R^{-1}(y - Hx)$$

with x being the state vector, x_b the prior knowledge, B the prior uncertainty covariance, y the observation vector, R the error covariance and *(y-Hx)* the innovation vector that contains the additional knowledge brought in by the observation compared to the model. (Jacob, 2007)

As the inversion requires that the errors of the prior emission inventories and the errors of the difference between modelled and measured concentrations are specified, it is necessary to compare measured and modelled CH_4 concentrations.

Conclusion / Outlook

The mixing ratios simulated with EDGARv4.3.2 are up to 100 ppb higher than those with TNO-MACC_III, with larger differences at the hotspots, e.g. over the Po Valley or the area over Belgium, the Netherlands and North-western Germany.
The comparison to the measurements resulted in the measured values being mostly underestimated by the simulations over the simulated period.
The agreement between the simulated and measured mixing ratios is highest for mountain stations as the measured values are lower at these sites.

Further forward simulations will include CH_4 mixing ratio data for sectorial and national budgets of emissions, and biogenic emissions. Simulation runs will be completed until present time.
The simulation results of CH_4 will be compared to atmospheric measurements from ICOS and INGOS and will be evaluated against higher resolution configurations of CHIMERE and optimized emissions.
We will determine the representation error of the vertical mixing of CH_4. In order to do so, MicroHH (van Heerwaarden et al., 2017), a fluid dynamics code for simulating turbulent flows in the atmosphere, will be used.
The use of methane isotopes (e.g. ^{13}C) can help bringing additional constraints on the top of ^{12}C data. For that, available data on isotopes will be included in the model simulations.

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