Attributing and Verifying European and National Greenhouse Gas and Aerosol Emissions and Reconciliation with Statistical Bottom-up Estimates

Deliverable 1.3
TOPAS CH_4 source apportionment service

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Executive summary

Methane (CH_4) is the second-largest contributor to the anthropogenic greenhouse effect and has been the focus of recent climate summits and global commitments towards emission reductions. To mitigate and study the impact of anthropogenic Greenhouse gases including CH_4 emissions, the European Union funded AVENGERS, a research and innovation project with 13 Partners from 6 countries, whose objective is to reconcile reported greenhouse gasses (GHGs) emissions with independent information from atmospheric observations using top-down methods and process-based models, and thereby reduce the most important uncertainties of national emission inventories. As part of the AVENGERS project, TOPAS-CH4 was developed following the example of a previous TNO source apportionment tool for particulate matter. This online public service provides users with the relevant daily information on CH_4 concentrations over the EU countries, and how much different sources contribute to these observed concentrations. TOPAS CH4 is based on the chemical transport model, LOTOS-EUROS, that runs daily to simulate source-labelled CH_4 concentrations over Europe. The predicted CH_4 concentrations are compared with the Integrated Carbon Observation System (ICOS) observations on a daily basis, showing that LOTOS-EUROS is able to realistically simulate the CH_4 volume mixing ratios. The simulated CH_4 concentrations are labelled per source sector and country of origin, providing the users more insights about which European sectors and countries are mostly responsible for the additional CH_4 concentrations above the background for a specific region. This information can be used to identify mitigation targets and, on the longer term, monitor changes in source contributions following successful abatement.

1 Deliverable objective

The Attributing and Verifying European and National Greenhouse gas and aerosol Emissions and Reconciliation with Statistical bottom-up estimates – AVENGERS project aims to establish and disseminate knowledge and tools facilitating the use of top-down techniques to support the verification of national greenhouse gas (GHG) inventories. In a broader sense this implies the use of atmospheric observations to confirm, support and/or improve bottom-up emission inventories. The work described in here is part of WP1 "Requirements, Reporting and Reconciliation" with the overall goal to reconciliate reported GHG emissions and removals in national inventories as currently reported in the National Inventory Reporting (NIR) and corresponding Common Reporting Format (CRF)-tables to the UNFCCC and EU (bottom-up) with measured atmospheric signals based on the top-down approach. Complementary to so-called "full inversions" as foreseen in WP2 "Top-down GHG emissions estimation & source attribution", the objective is to develop a prototype methane (CH_4) source apportionment service, which enables the countries to receive sector-specific results for inventory verification at observational sites.

2 Methodology

The prototype source apportionment service for methane is based on a similar service developed by TNO for particulate matter (topas.tno.nl) to track the contributions of predefined source categories to atmospheric concentrations at selected locations. The calculations are done with the chemical transport model LOTOS-EUROS (LE), which is equipped with a special labelling technique to track the contributions. For the methane application, the model uses the official reported UNFCCC CH_4 emissions gridded by TNO as model input. Source contributions are available as maps and at selected locations (such as ICOS stations) where observational CH_4 data are available for validation. The different components are briefly discussed below.

2.1 LOTOS-EUROS model

LOTOS-EUROS is an open-source chemical transport model (CTM) maintained by TNO (airqualitymodeling.tno.nl/lotos-euros) that is used for a wide range of applications supporting scientific research, regulatory programs, and air quality forecasts. With an origin in ozone and sulfur modelling LOTOS-EUROS' application areas were gradually extended with persistent organic pollutants, reactive nitrogen, and primary and secondary particulate matter. The model is described in detail by Manders et al. (2017). LOTOS–EUROS has been a front-runner in data assimilation of ground-based and satellite observations and has participated in many model intercomparison studies. The model is no longer confined to applications over Europe but is also applied to other regions of the world, e.g. China. The increasing interaction with emission experts has also contributed to the improvement of the model's performance. LOTOS–EUROS is one of the European models contributing to the regional air quality production of the Copernicus Atmosphere Monitoring Service (CAMS) which is based on an ensemble of 11 state-of-the-art numerical air quality models developed in Europe (atmosphere.copernicus.eu/european-air-quality-forecast-plots).

2.1.1 LOTOS-EUROS for methane

For the source apportionment service within this project, the model has been configured to simulate concentrations of methane; in addition, also ethane is simulated as this could serve as tracer for fossil fuel extraction. The only processes required are emissions (described below), transport (advection and vertical mixing), and the chemical sink. For the sinks the model reads concentration fields of the hydroxyl radical (OH) that have been saved from a source apportionment service that uses full chemistry. An example of the use of LOTOS_EUROS for methane can be found in the CoCO2 project deliverable report D4.6 Inter-Comparison of National-Scale Inversions. (coco2-project.eu/node/412).

2.1.2 Emission input

For the prototype service, anthropogenic gridded emissions from the CAMS-REG v5.1 emission inventory were used as input for the simulations. This version is an update of the data set described in (Kuenen et al., 2022). The latest available year in this set is 2018, and these were used also for recent years. In the near future the emission data set will be updated based on the dataset prepared by TNO under CAMS in 2025 (CAMS-REGv8). The gridded emissions have a resolution of 0.05° × 0.1° in latitude and longitude, which is approximately 6 × 6 km over central Europe. Emissions are classified per GNFR (Gridding Nomenclature For Reporting) sector and per country, and this information is used to track the origin of concentrations in the source apportionment. The reported emission data are annual totals by detailed source sectors. The annual data are converted to hourly emission data using sector-specific time profiles that provide the monthly fraction, day-of-the-week fraction and diurnal distribution. A more detailed description of the TNO dataset for Greenhouse gases is also given in AVENGER deliverable report 2.2 on *Prior Emissions*.

A dedicated emission set developed by TNO is used for anthropogenic emissions of ethane. This set has the same resolution as CAMS-REG-v5.1 but has a lower number of contributing GNFR sectors as not all sectors emit ethane.

An important natural source of methane is emission from wetlands, which is also one of the focus points of the AVENGERS projects. For the prototype service, wetland emissions are obtained from a global simulation with the LPJ-GUESS model at 0.5°x0.5° resolution. It is foreseen that this will be updated to the emissions sets produced in the AVENGERS project towards the end of the project. Wild fires are a small natural source of methane and ethane; these are obtained from the operational daily CAMS GFAS service.

2.1.3 Boundary conditions

Boundary conditions for methane and ethane are obtained from the CAMS daily near-real-time global chemistry simulations (Peuch et al., 2022). In the various source apportionment services these simulations are used as boundary conditions of regional full-chemistry air quality simulations. For methane the global simulations could be strongly biased, as they are not based on assimilation of observations. In the methane source-apportionment service, the contribution of these boundary conditions on concentrations is therefore adjusted based on the observations as described in section 2.2. Data post processing.

2.1.4 Labelling

The LOTOS-EUROS model is equipped with a labelling technique to keep track of various sources of concentrations during the simulation (Kranenburg et al., 2013). The labelling implementation for methane follows the existing method for chemically active tracers containing a carbon atom, as this is conserved and traceable.

Concentrations are labelled when introduced in the simulation based on the GNFR classification of the anthropogenic emissions or the type of natural emissions, and for the anthropogenic emissions also the country where emission takes place. In total 666 labels are defined: 39 countries with 17 GNFR sectors each, plus 3 extra labels for global boundaries, wetlands, and fires. In the post-processing an aggregation is applied to limit the output to 6 selected AVENGER countries (countries with an inventory agency as partner of the project) and main source categories; the aggregated sectors and/or regions are presented in *Annex I: Sector and country allocation labels*.

2.2 Data post processing

When the model simulations are finished, the post processing of the data for the source apportionment service will first aggregate countries and labels as described above. This will limit the output to make results easier to interpret.

The core part of the data post-processing is to perform the daily comparison between the LOTOS-EUROS 3D labelled simulated CH_4 concentrations with the ICOS tall tower measurements. Every day the post-processing tool downloads the latest CH_4 concentration measurements from all the available ICOS towers from the ICOS Data Portal (see also Annex 2). After the ICOS data is downloaded, the post-processing tool extracts the labelled concentrations for the different ICOS tower locations. Since many towers have more than one detector at different heights, the post-processing tool interpolates the model concentrations among the layers in the vertical direction and returns the values corresponding to the sampling height of each ICOS tower detector. This guarantees that the simulated concentrations from a specific ICOS tower with multiple sampling heights are different and best approximate real world concentration differences even if the detector heights are within the same model layer.

Finally, the post-processing code produces the comparisons between the labelled simulated concentrations and the ICOS observations for the current day and the previous 14 days. In the comparison part, for each ICOS tower the code computes a local bias correction factor that needs to be applied to the labelled background concentrations to better match the simulated data and ICOS data. This local bias is computed as the time average difference over this 14-days period between the ICOS and the simulated data. For stations with more than one detector, the local bias is only computed for the detector at the highest height (where the mixing layer is more developed), and it is then applied to the time series of the other detectors.

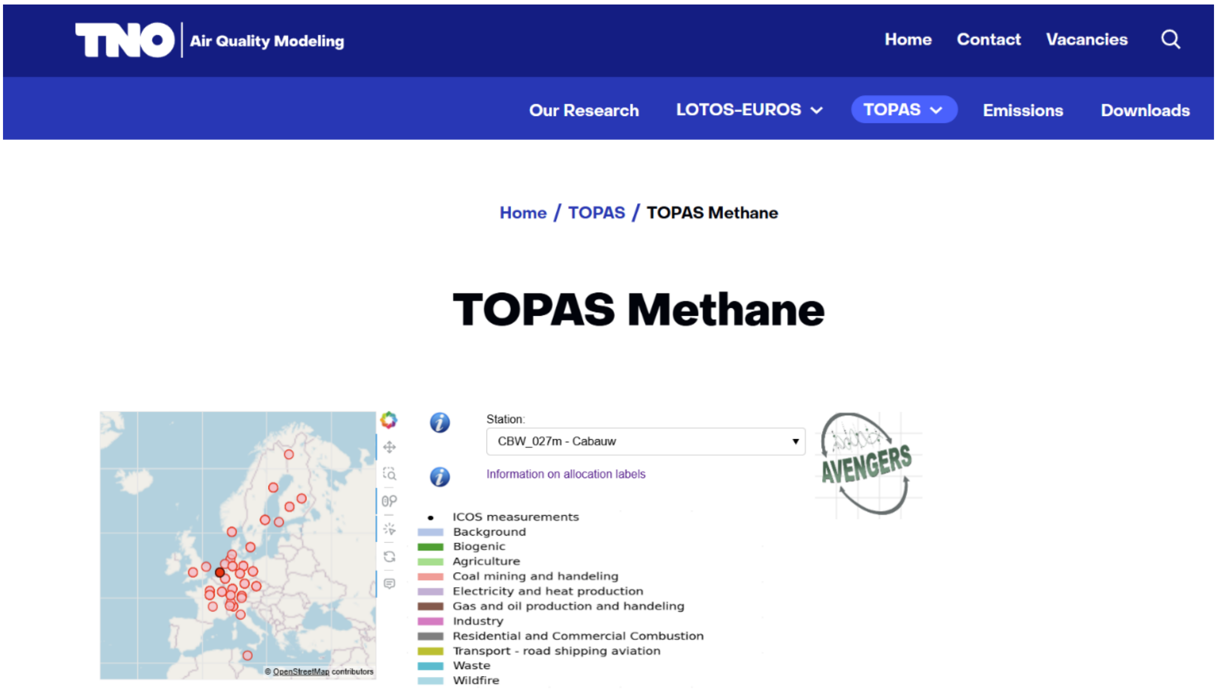
For each ICOS tower, the post-processing code produces the following plots:

- Hourly comparison.
- concentrations for low-altitude stations (below 700m), or night (00:00-03:00 local time) concentrations for high-altitude stations.
- Noon or night label fraction contribution pie chart (but without including the background sector)

The average noon (12:00-15:00 local time) or night (00:00-03:00 local time) plots facilitate a more robust interpretation because at these times the mixing in the atmosphere is more complete for the low and high latitude stations, respectively. All plots are then updated and uploaded on the TOPAS website as described in Chapter 3.

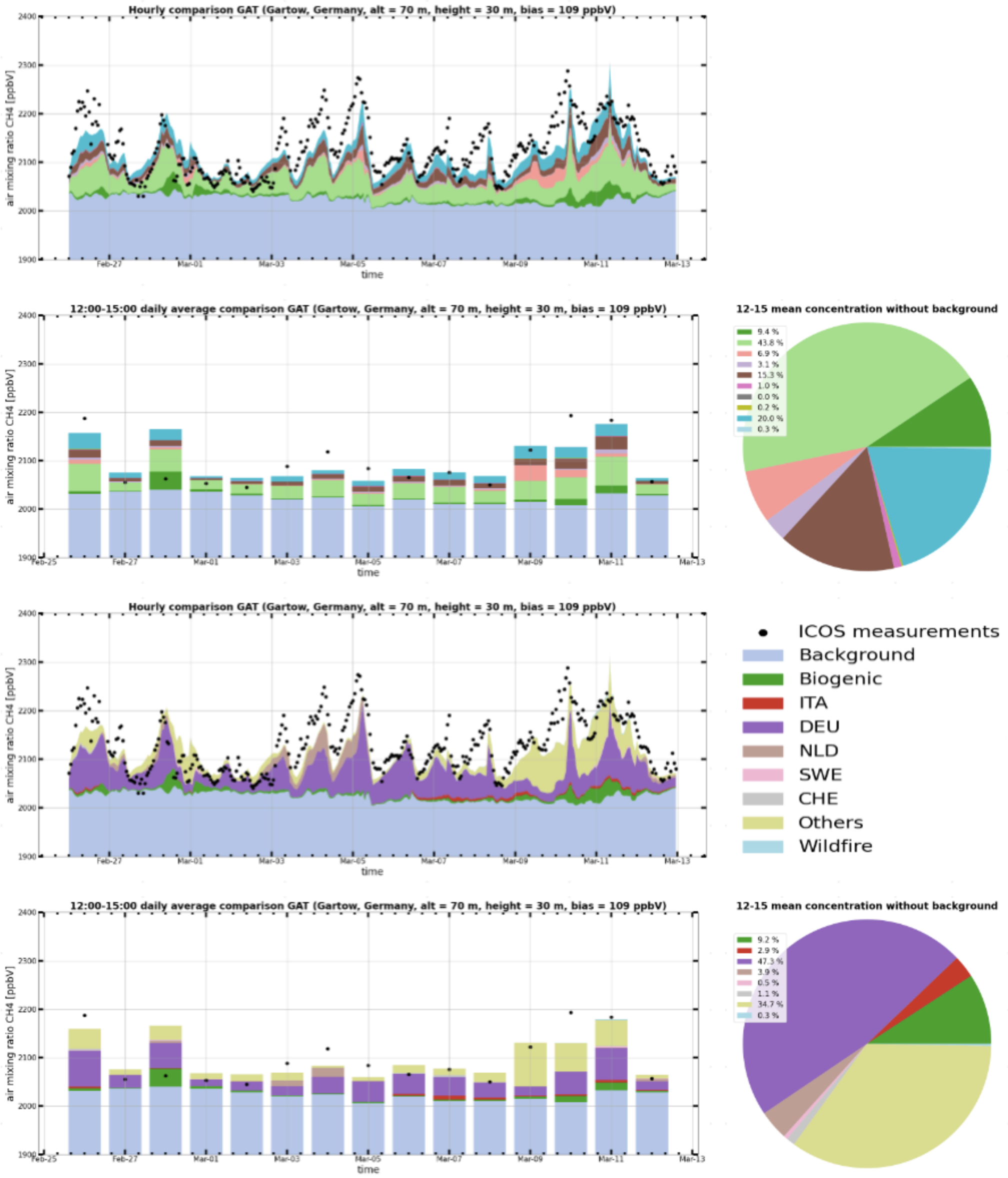
3 TOPAS website

The TOPAS-CH4 website is the location where all the LE-ICOS data comparison plots are displayed (https://airqualitymodeling.tno.nl/topas/topas-methane/). The website is open access to the public and it updates every day around 12:00 pm UTC. Below a selection of screenshots of the website are provided as well as some instructions on how to use the TOPAS CH_4 service.



The figure above shows the top part of the TOPAS-CH4 website. On the left side there is a map of Europe with red dots representing the different ICOS stations that can be selected to show the comparison plots. In the central part the user can also use the drop-down menu to select the ICOS stations together with the sampling height of the observational data. Below the drop-down menu, there is the legend of the source sector labelling. For clarity purposes, the post-processing tools groups together the original 22 LOTOS-EUROS labels into 11 ones.

The next figure shows the second part of the website: the comparison plots. These comparison plots show the LOTOS-EUROS result and ICOS data comparison for the last 15 days. . All the plots have a title describing the show the name of the ICOS station, as well as the location country, the altitude above sea level, the height of the selected detector, and the local bias that has been applied to the LE background to match the ICOS data.



The top panel shows the hourly comparison for the sector labelling. The second panel shows the sector average noon (12-15 hr) or night (00-03 hr) comparison depending on if the station is a low or high altitude station; noon comparison for low altitude (< 700m above sea level), and night comparison for high altitude (>700m asl) locations. On its right, the noon or night sector fraction contribution pie chart is displayed. This pie chart shows what is the average total contribution from the different sectors, if the background sector is excluded.

The third panel shows the hourly comparison for the country labelling, while the country legend it is displayed on its right. The fourth panel again shows the country average at noon (12-15 hr) or at night (00-03 hr) comparison depending on ICOS tower location altitude: noon comparison for low altitude (< 700m), and night comparison for high altitude (>700m) areas. On its right, the noon or night country fraction contribution pie chart is displayed. This pie chart shows what is the average total contribution from the different countries, if the background sector is excluded

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Annex I: Sector and country allocation labels

Currently LE simulates 668 labelled CH4 concentrations:

- 39 European countries with 17 emission sectors (663 labels in total).
- Five general labels that are independent from the regional location: wildfire, Sahara dust, sea salt, biogenic, boundary.

Not all the 668 are shown individually on the TOPAS-CH4 website. Many labels are grouped by country or sector, while Sahara dust and sea salt labels are not displayed as they do not contribute to CH_4 emission. The lists below show the aggregated label for sector and country.

The following labels are used to denote a source type:

- **Agriculture**: animal husbandry, manure and fertilizer application, agricultural stationary combustion.
- **Biogenic**: emission from wetlands due to anaerobic archaeal methanogens.
- **Coal mining and handling**: Coal mining extraction and distribution.
- **Electricity and heat production**: power plants, refineries and other combustion in the energy industry.
- **Gas and oil production and handling**: Oil and gas production; gas distribution and flaring.
- **Industry**: iron and steel industry, chemical industry, paper and pulp industry, cement production, Storage and handling, etc.
- **Residential and commercial combustion**: heating and cooking from residential and commercial buildings.
- **Transport - road and shipping aviation**: aviation, rail, inland shipping, mobile machinery, cars, motorcycles, etc.
- **Waste**: waste incineration, landfills, BBQ and tobacco.
- **Wildfire**: natural origin, e.g. grassland and forest fires.
- **Background:** emitted outside model domain.

The following labels are used to denote the European countries that contribute to the AVENGER project.

- **Background**: origin outside the model domain (i.e. other continents), in particular from Southern Europe.
- **Biogenic**: emission from wetlands due to anaerobic archaeal methanogens.
- **CHE**: Switzerland.
- **DEU**: Germany.
- **ITA**: Italy.
- **NLD**: Netherlands.
- **SWE**: Sweden.
- **Others**: all the other European countries.
- **Wildfire**: natural origin, e.g. grassland and forest fires

Annex II: Observation sites

Station	Country	Sample Heights (m)	Altitude (m)
Birkenes	Norway	10, 50, 75	219
Cabauw	The Netherlands	27, 67, 127, 207	27
Monte Cimone	Italy	8	2165
Gartow	Germany	20, 60, 132, 216, 341	70
Helgoland	Germany	110	43
Hohenpeissenberg	Germany	50, 93, 131	934
Hyltemossa	Sweden	30, 70, 150	115
Ispra	Italy	40, 60 ,100	115
Jungfraujoch	Switzerland	5	3571
Julich	Germany	50, 80, 100	98
Karlsruhe	Germany	30, 60, 100, 200	110
Kresin u Pacova	Czechia	10, 50, 125, 250	534
Lindenberg	Germany	2, 10, 40, 98	73
Lampedusa	Italy	8	45
Lutjewad	The Netherlands	60	1
Nordua	Sweden	32, 58, 100	46
Observatorie Perenne	France	10, 50, 120	390
Ochsenkopf	Germany	23, 90, 163	1022
Pallas	Finland	12	565
Plateau Rosa	Italy	10	3480
Pujo	Finland	47, 84	232
Puy de Dome	France	10	1465
Ridge Hill	UK	45, 90	207
Saclay	France	15, 60, 100	160
Hyytiala	Finland	16, 67, 125	181
Schauinsland	Germany	12, 35	108
Steinkimmen	Germany	32, 82, 127, 187	29
Svaterbergt	Sweden	35, 85, 150	269
Torfhaus	Germany	10, 76, 110, 147	801
Trainou	France	5, 50, 100, 180	131
Uto	Finland	57	8
Weybourne	UK	10	17
Westerland	Germany	14	12
Zugspitze	Germany	3	2666

The table above lists the ICOS stations that are displayed on the TOPAS website with the information about their location country, their altitude above sea level and the heights of their different detectors. More information on the ICOS station network can be found here: https://www.icos-cp.eu/observations/station-network .