Modeling of non-CO_2 greenhouse gas concentrations at the high-mountain station using a Lagrangian emissions model COMET.

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1. PURPOSE OF THE VISIT

The author's PhD thesis (to be finished in 2015) will be focused on the description of the nitrous oxide (N_2O) budget in the region of southern Poland. An important part of the workplan involves using regional transport modelling tools to verify strengths of the regional sources of N_2O, with the measurements from at least two ground stations as a validation dataset. These stations are: an urban station in Kraków city-centre and the regional background station at Kasprowy Wierch (1987 m a.m.s.l.) in Tatra Mountains (see e.g. [1] for detailed site descriptions). However, because N_2O long-term records for these stations were not available at the start of the visit, it has been decided that, although the sources and chemistry of these two components are different, modelling of CH_4 transport will be a good alternative. Methodology of modelling using the COMET model is almost identical, and quality – controlled records of CH_4 mixing ratios at both of the stations span several years[1].

In this framework, the goals of the exchange visit has been set as follows:

a) To familiarize with the model setup, including the input data pre-processing, setting the model parameters *etc*.,
b) For chosen time periods – to calculate backward trajectories for Cabauw, Kasprowy Wierch, Kraków and Mace Head stations using the FLEXTRA transport model, with different meteorological datasets as input (as described in table 1),
c) Use the calculated trajectories as input for the COMET model, run in the forward mode with different emission databases (table 2) as input, to predict the concentrations at the sites, and then afterwards to compare the results for with the measured concentrations,
d) If possible, use the results obtained to gain insight into the validity of these databases for the sources located in Southern Poland.

[1] As generally accepted, the hourly averages were used rather than raw measurements data.

2. DESCRIPTION OF THE WORK CARRIED OUT

2.1. Model input preparation.

The first task to be performed at the Energy Research Centre of the Netherlands (ECN) was to prepare the input data for the subsequent COMET model runs. 144 hour long backward trajectories have been calculated for a variety of locations (which are described in detail in Box 1) using the FLEXTRA model (v. 4.0) [2]. The input meteorological data that was used is described in table 1.

table 1. The meteorological fields used for model calculations.

	Normal runs	Sensitivity test	
Meteo data source	ERA-Interim	ERA-Interim	ERA-Interim
Grid span	Global	Global	Nested (Europe)
Horizontal resolution	1.0° x 1.0° lat - long	3.0° x 2.0° lat - long	0.2° x 0.2° lat - long
Vertical resolution	37 levels	37 levels	37 levels
Time interval	3-hourly	3-hourly	3-hourly
Time coverage of available data	01-01-1996 to 30-10-2011	18-04-2008 to 10-07-2008	18-04-2008 to 10-07-2008

Box 1. Modelled sites information.

4 sites have been selected to perform model runs: Cabauw, Mace Head, Kasprowy Wierch and Kraków. Next to the name and the country of the site, coordinates of the measurement stations are given.

Cabauw, Netherlands (51.971° N, 4.927° E): model calculations has been performed for 4 points of different heights, which correspond to 4 measurement levels present at the Cabauw tall tower. These are at 20 m, 60 m, 120 m and 200 m above ground level. In this report, only the results from 20 m level are presented.

Mace Head, Ireland (53.333° N, 9.090° W): calculations performed for only 1 level, 10 m above ground level, at which height the station inlet is located.

Kasprowy Wierch, Poland (49.233° N, 19.983° E): trajectories and model results were calculated for 10 different model levels to determine the point that best represents the station's location on a model grid. This is caused by the specific topography for Kasprowy Wierch, which lies in the middle of the Tatra Mountains. One of the characteristic features of this mountain chain is its relatively small spatial coverage (approx. 50 km E–W x 20 km N–S), smaller than the grid sizes used in the calculations. As a result, the surface height for e.g. the 1.0° x 1.0° lat – long ERA-Interim data is much lower than actual ground level (approx. 630 m). Therefore in a model a point 2 m above ground level does not represent the station inlet appropriately.

Overall, results were obtained for 10 different heights, above ground level: 2 m, 100 m, 250 m, 500 m, 750 m, 1000 m, 1250m, 1336 m (corresponding to the station's inlet level), 1500 m, 1750 m. Please note that not all of the results them are presented in this report.

Kraków, Poland (50.067° N, 19.916° E): although Kraków topography is not as complicated as in case of Kasprowy Wierch, it was decided that trajectories and model results will be calculated for 4 different model levels, as this was the first time that such detailed analysis was performed with this model. Their height above ground level were: 8 m (corresponding to the stations's inlet level), 20 m, 50 m and 100 m. Model performance test has shown that the results from 8 m level correspond best with the observed results, therefore the rest are omitted from this report.

The second task was to calculate the mixing heights on the chosen grids, using the routine that uses the bulk Critical Richardson number approach (with R_i = 0.25), kindly provided by A. Vermeulen. Together with the backward trajectories and methane emission databases, these formed the input for the COMET model. The technical details are closely described in a paper by Vermeulen et al. [3]. For the purpose of this visit, three different databases were used. Detailed information on these databases can be found via their respective web pages (see table 2).

table 2. Methane emission databases used in the calculations.

Name	IER	EDGAR	NEU
Source	University of Stuttgart	Joint Research Centre	NitroEurope Project
Version	2008	4.1.	n/a
Grid resolution	1.0° x 1.0° lat - long	0.1° x 0.1° lat - long	1.0° x 1.0° lat - long
Details	www.ier.uni-stuttgart.de	edgar.jrc.ec.europa.eu	www.nitroeurope.eu

2.2. Background calculations.

The model simulates the additions of the surface-based sources to the air column, represented on a tetragonal latitude – longitude grid. This approach does not take into account the information about the background concentrations of the air masses moving above the emissions sources, therefore this had to be provided as an additional input. COMET has a provision to read background mixing ratio data from global transport models such as TM5 in the *netcdf* or *hdf* format, but currently no complete data record for methane nor nitrous oxide is available for the full study period.

The simplest way to provide an alternative background mixing ratio input to the model is to use values measured in the conditions that may be regarded as representative for the background conditions. Mace Head station is generally accepted as the appropriate background station for the European continent, therefore it has been decided that the averaged CH_4 record from this station will be used as a background for every station for which simulations have been performed. Clean air data from the Mace Head station was downloaded from the AGAGE website [4]. Afterwards, monthly means were calculated for the period from January 1996 to October 2011. To this data, an interpolation curve was fitted using the method of Forsythe, Malcolm and Moler. All the COMET model simulation results were added to this curve during the post – processing.

2.3. Grid sensitivity test.

In order to compare the backward trajectory model sensitivity to the resolution of the meteorological fields, the model results calculated for all the locations were performed for the period between April 18th and July 10th 2008. As described in table 1, the "normal runs" (NR) were performed using a 1.0° x 1.0° lat – long grid of the meteorological data. The "sensitivity test runs" (SR) results were calculated using a nested high-resolution 0.2° x 0.2° lat. – long. grid data superimposed over the 3.0° x 2.0° lat. – long. global grid. All of the meteorological data were obtained from the ECMWF ERA-Interim databases [5] and were accordingly pre-processed in order to be FLEXTRA-readable.

2.4. Model performance for Kasprowy Wierch high mountain station.

Using the measurements data from the years 2007-2011, model performance was estimated for Kasprowy Wierch mountain station using the EDGAR methane emission database (table 2). Because of the specifics of the Tatra Mountains topography (see Box 1), simulations were performed for 10 different heights in order to find the spot that best represents the station's location in the given meteorology grid. It is known from the literature that capturing mixing ratios of air pollutions and long-lived tracers like greenhouse gases at mountain stations with the current atmospheric transport model is a challenging task. In the driving meteorological models the surface heterogeneity, roughness and terrain elevation are represented by a smoothed and averaged field that does not allow for a local flow to be modelled well. This means that the subgrid phenomena that occur in mountainous regions are not represented in the current models. Therefore, the topography in the model is usually very different from the actual one, and the height from which the mixing ratio should be sampled in the model is generally expected to be lower than its real height (ASL), and should always be chosen carefully.

2.5. Model performance for Kraków urban station.

Due to the low correlation between the model results and the observed values at the Kasprowy Wierch mountain station, it has been decided to check the model performance only for the Kraków urban station. Using the nested ERA-Interim meteorology data (same as in the sensitivity test), the predicted values for 4 different heights were calculated and then compared with the available observational data from the period between April 18th – July 10th 2008.

2.6. Local methane emissions from specific sources in southern Poland: event-based assessment in Kraków area.

For a one chosen event of Kraków station a detailed analysis has been performed as example of how to assess the validity of the EDGAR emissions database values specific to the sources in southern Poland.

3. Description of the main results obtained

3.1. Sensitivity test results.

3.1.1. Cabauw.

The COMET model was developed by the team responsible for the measurements at Cabauw station, therefore the calculations for this station were performed as a training and quality-assurance. Detailed description of the previous comparisons between Cabauw measurement data and model predictions can be found in a paper published by Vermeulen et al. [3]. It has been decided that the sensitivity test will be performed using the data available only at one of the four tower levels – namely 20m – for the period between April 18th and July 10th 2008.

The comparison of the modelled results and the observations can be seen in fig. 1, the scatter plots in - fig. 2 and the most important model parameters are summarized in table 3. As can be seen, usage of the finer resolution meteo-data does not significantly improve the model performance. In case of the results obtained with EDGAR and NitroEurope emission databases, model parameters seem to point at a slight loss in prediction quality. Therefore the usage of 1.0° x 1.0° grid meteorological data with the COMET model with these emission databases seems to be justified. The difference between the results using the different emission databases is remarkable, especially in the light of the fact that the NEU emission database has been derived from the EDGAR database, by averaging it to 1 degree resolution.

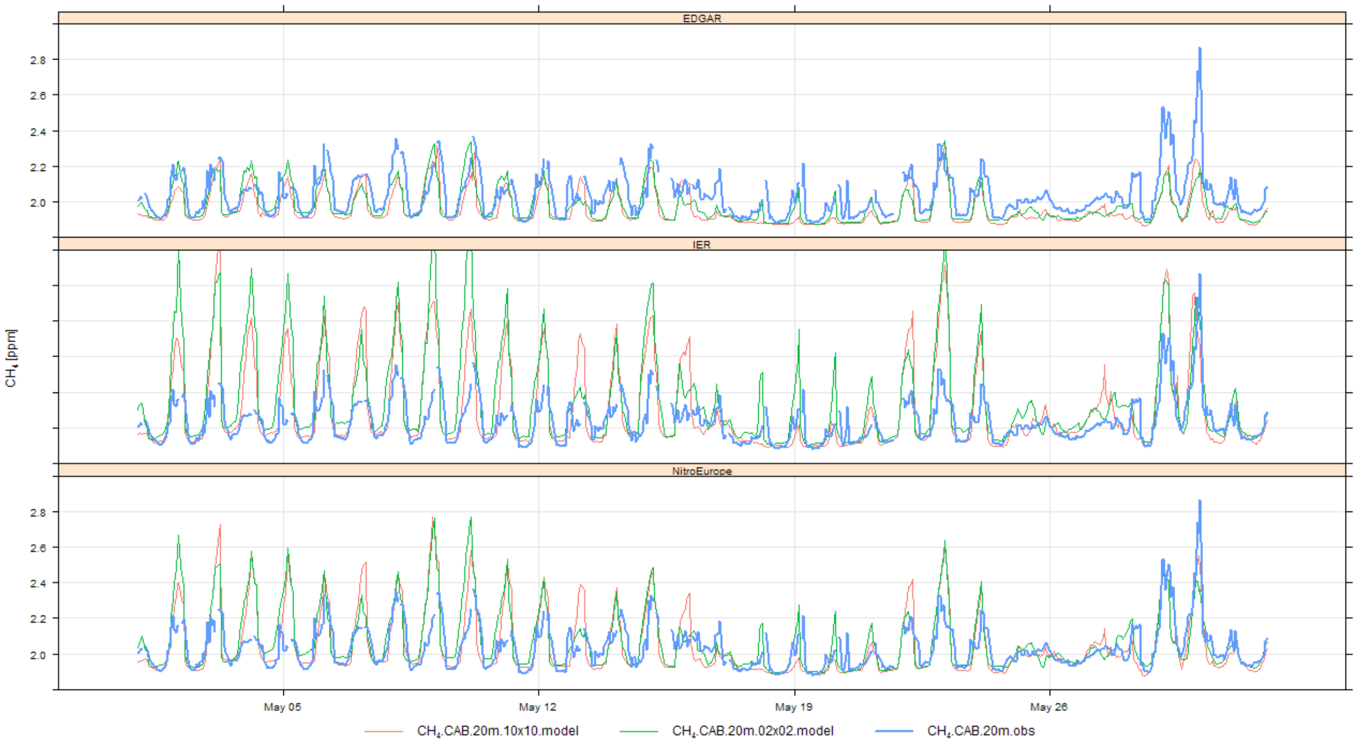

fig. 1. A comparison between the output modeled with two different meteorological grid resolutions for three emission databases – Cabauw tall tower. For clarity, only one month of the results is presented.

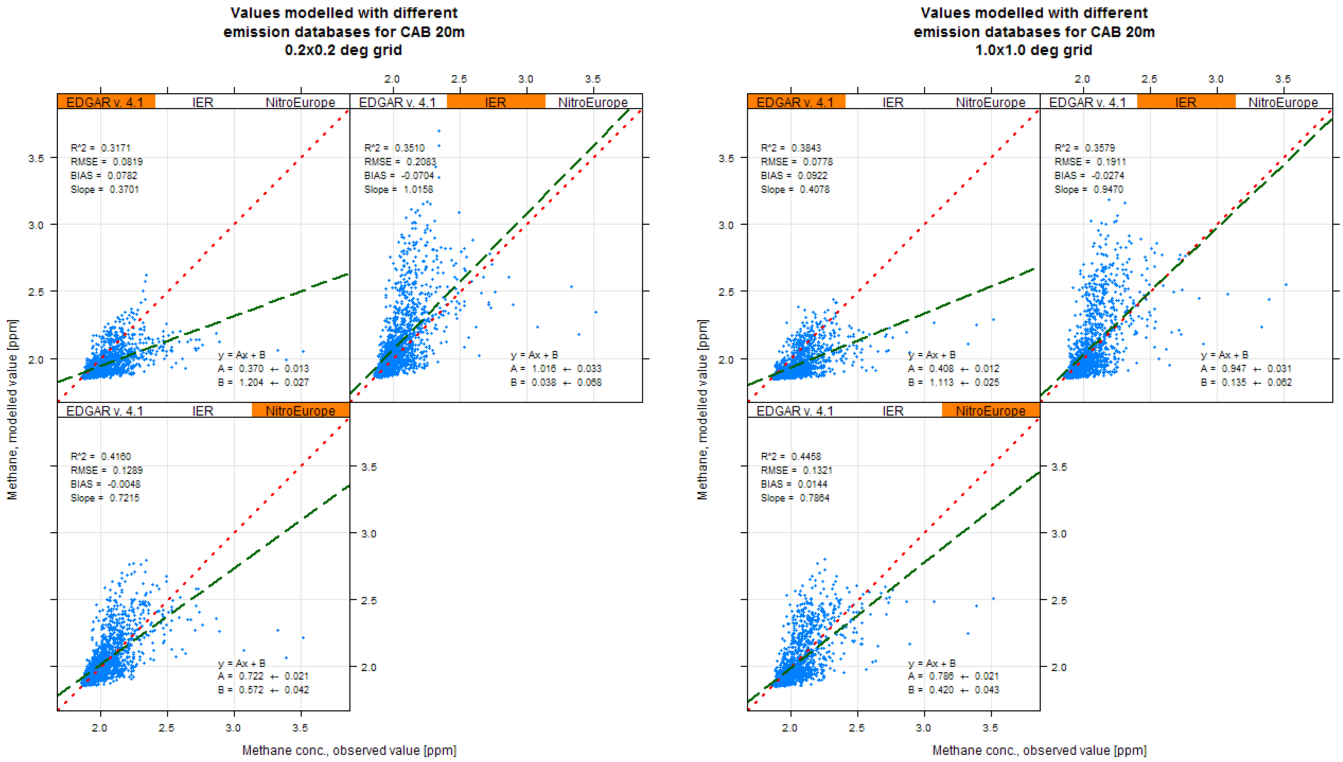

fig. 2. Model results vs. observations for Cabauw station, 20m level. Red dotted lines show a perfect agreement. Green line shows calculated linear regression lines. Plots were made for the whole sensitivity test time period. Missing data has been excluded from the analysis.

table 3. Summary of the model results for Cabauw station, 20m level.

Grid resolution	Database	R^2	RMSE	Bias	Slope
0.2° x 0.2° lat-long	EDGAR	0.3171	0.0819	0.0782	0.3701
	IER	0.3510	0.2083	-0.0704	1.0158
	NitroEurope	0.4160	0.1289	-0.0048	0.7215
1.0° x 1.0° lat-long	EDGAR	0.3843	0.0778	0.0922	0.4078
	IER	0.3579	0.1911	-0.0274	0.9470
	NitroEurope	0.4458	0.1321	0.0144	0.7804

3.1.2. Kraków.

Analogous to the previous section, results of the sensitivity test for Kraków urban station can be seen in fig. 3, fig. 4 and table 4. Again, there is no significant improvement visible in the modelled results when using finer grid resolution. Differences between different databases are even more distinct in this case, probably due to larger discrepancies in the descriptions of the methane sources in the Central-Eastern Europe area. High excursions of the modelled values compared to the observations may point to overestimation of the sources strength in the Kraków area by specific databases – e.g. events from May 13 and May 14 in the EDGAR database. A similar event will be described in more detail in section 0.

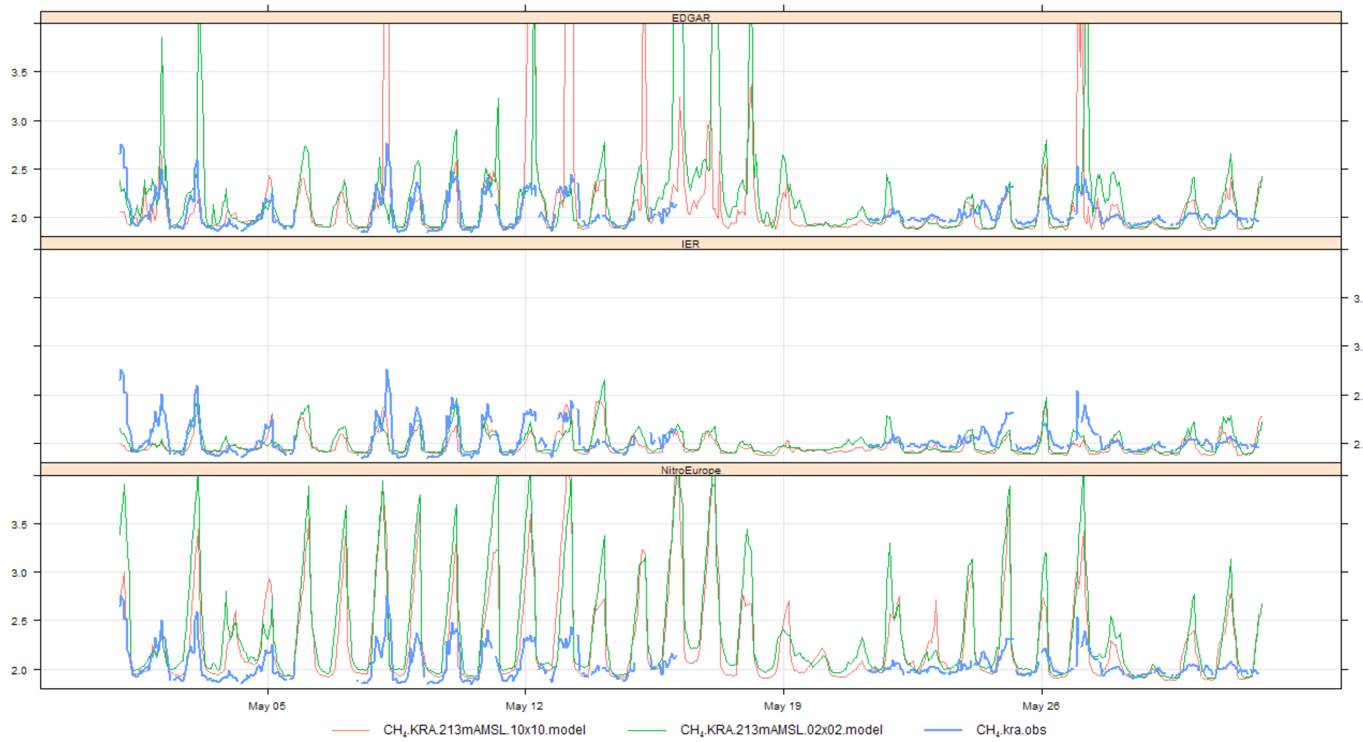

fig. 3. A comparison between the output modeled with two different meteorological grid resolutions for three emission databases – Kraków urban station. For clarity, only one month of the results is presented.

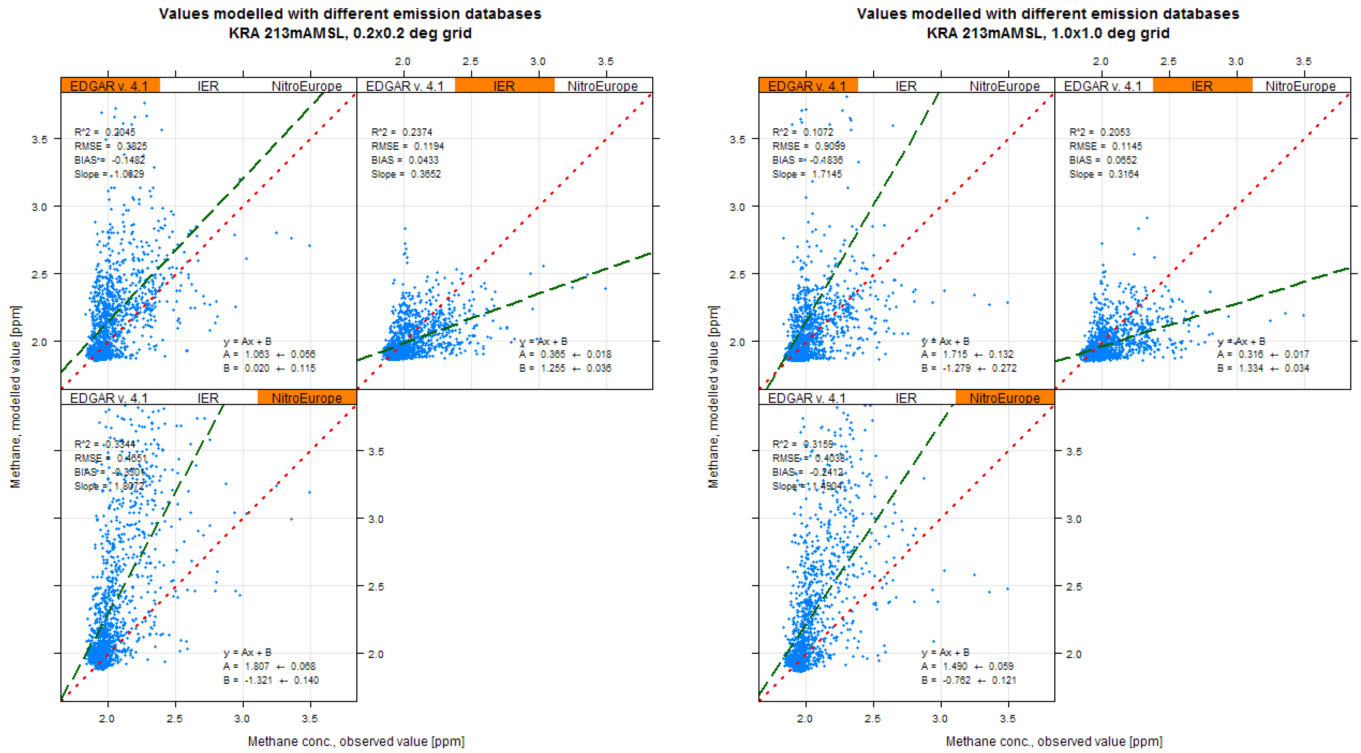

fig. 4. Model results vs. observations for Kraków station. Red dotted line shows theoretical perfect agreement. Green line shows the linear regression line. Plots were made for the whole sensitivity test time period. Missing data has been excluded from the analysis.

table 4. Summary of the model results for Kraków station, 8m level.

Grid resolution	Database	R^2	RMSE	Bias	Slope
0.2 x 0.2	EDGAR	0.2045	0.3825	-0.1482	1.0629
	IER	0.2374	0.1194	0.0433	0.3652
	NitroEurope	0.3344	0.4651	-0.3301	1.8072
1.0 x 1.0	EDGAR	0.1072	0.9099	-0.1836	1.7145
	IER	0.2053	0.1145	0.0652	0.3164
	NitroEurope	0.3159	0.4038	-0.2412	1.4904

3.1.3. Kasprowy Wierch.

Unfortunately, Kasprowy Wierch station data could not be used for the sensitivity test due to the lack of the observations data in the period of April – July 2008. Measurement devices at the site were not operating optimally during that time (results not shown).

3.2. Model performance for Kasprowy Wierch high-mountain station.

As an example for estimating the model performance for Kasprowy Wierch station, a period of the 5 months (March – June 2010) of methane measurement data and model results was selected. Model calculations were performed using the EDGAR database and 1.0° x 1.0° lat-long grid meteorological ERA-Interim data. The results can be seen in fig. 5, from which certain features can be easily distinguished:

- first, the baseline of the model results does not always correspond with that of measurements. This probably results from the simplifications made during the background calculations (chapter 2.2, page 4) While Mace Head data can be a very good approximation of the continental background conditions for Cabauw station, where marine air masses prevail, in case of Kasprowy Wierch the events of the easterly flow (with different background conditions) are more frequent, thus reducing the goodness of fit. A solution to that would be to use background conditions calculated from a global model (e.g. TM5) instead.
- second, peaks that occur in the modelled results rarely correspond to peaks in the observation records without a significant time shift. This may be caused by the fact that as an elevated station, Kasprowy Wierch's footprint is much larger than that of Cabauw station, and while this improves the ability to detect peaks coming from distant emission sources, it also means that the sensitivity for errors in the trajectory calculations is relatively high. When considering the stations inside the Planetary Boundary Layer (PBL), results are influenced by the very local sources most of the time. In such case, the misplacement of the Area of Influence (AOL; several tens of km in size) which follows the path of the trajectory will not be large enough to procure large differences in the predicted model values. However, with the growth of the trajectory length, this effect might take place, which may generate significant differences between the observations and model predictions that are present in the record shown here.
- third, possible problems with observations. Although the data has passed quality control procedures, there is a possibility of error occurrences, e.g. low values in the record observed around mid June.

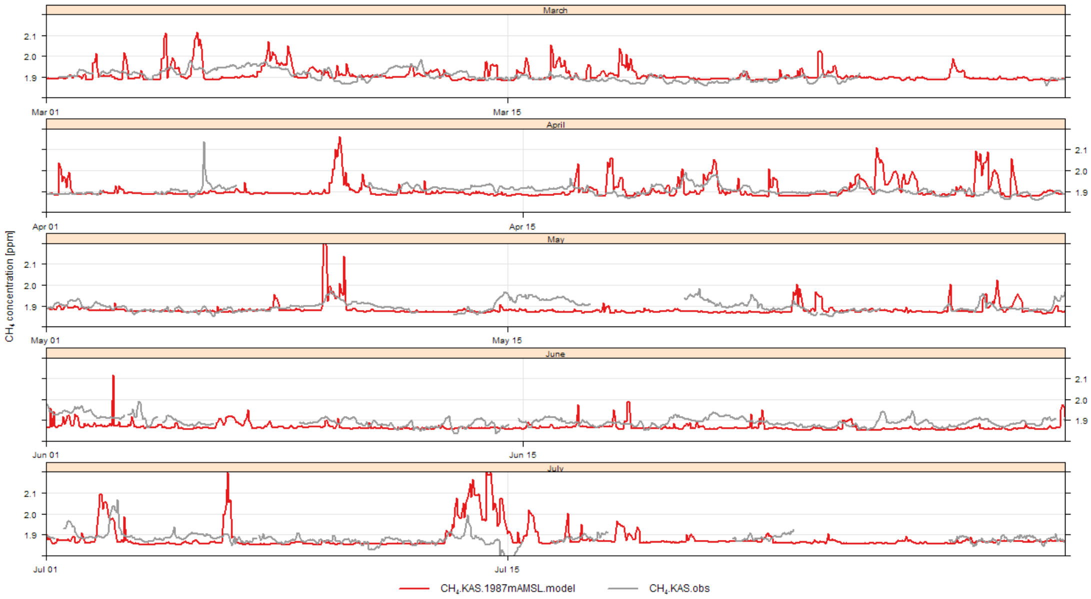

fig. 5. 5 months of modeled results (1987mAMSL level, 1.0° x 1.0° ERA-Interim meteo, EDGAR v. 4.1 emissions database) against measurements at Kasprowy Wierch.

To quantify the model performance, a scatter plot of the measured values vs. modelled values is shown in fig. 6 for seven different heights of the station (see section 2.4 for explanation). As can be seen, none of the levels gives satisfying results in comparison to the observations, with overall explained variability smaller than 5%. RMSE and BIAS are lowest for the highest model sampling level, but even at these levels the observed variability of the methane mixing ratios at the stations is low, only ~100ppb, compared to the model error RMSE of 47 ppb.

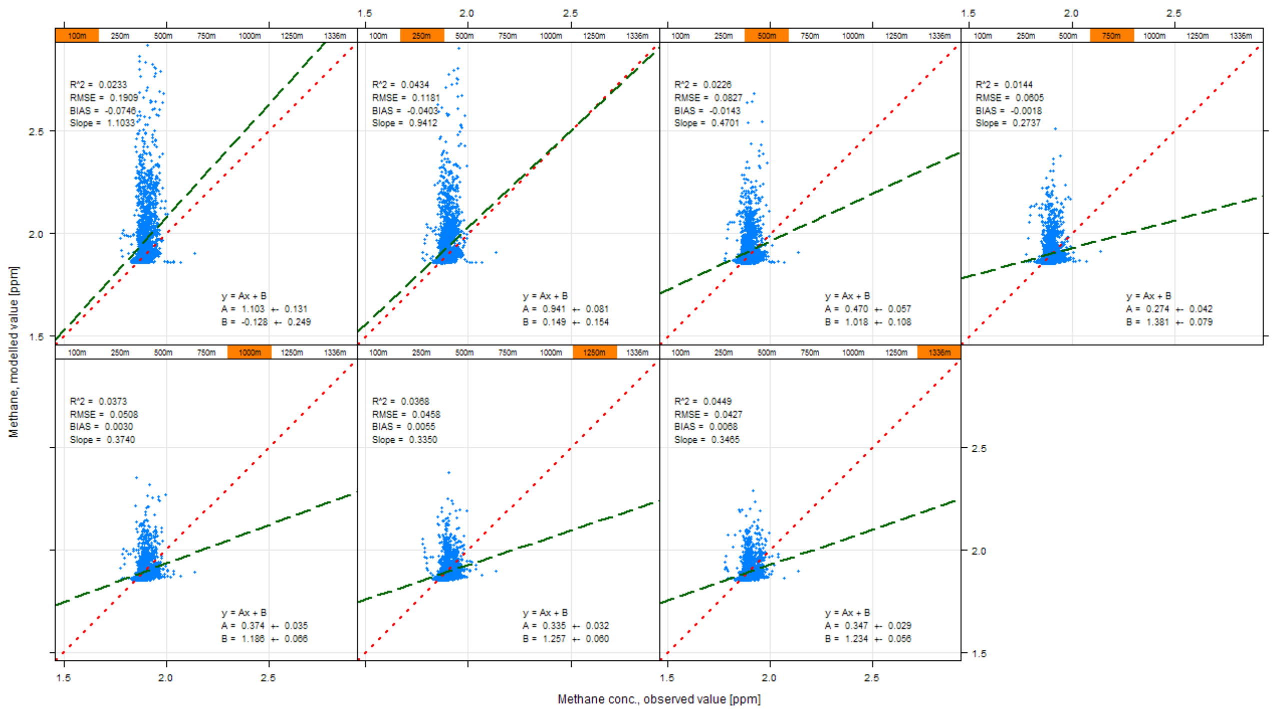

fig. 6. Scatter plots of the observed values vs. model results for 7 different levels at Kasprowy Wierch location. Model runs were performed using EDGAR v. 4.1 emissions database and 1.0° x 1.0° ERA-Interim meteo data. Note that 1336m level is the precise point of the station in space (equal to 1987m AMSL).

Model runs for Kasprowy Wierch site were also performed for other emission databases, yielding similar results, not shown here. It is therefore necessary to conclude that the simulations at this high mountain site are not well represented with the COMET model. One cause of the low correlation may be the poor representation of the terrain topography on the chosen grid scale, which is always a problem when modelling in the mountain areas. Also, the emission databases may not be as precise (in values) in the Central Europe region as in the North-Western Europe – owing to the necessity to rely on statistics rather than detailed in-situ measurements in the used emission databases.

3.3. Model performance for Kraków urban station.

Inability to successfully predict Kasprowy Wierch concentrations with the COMET model means that the verification of the methane emission databases in the region of Southern Poland is impossible in this way. Therefore observations from the more locally influenced station located in Kraków were investigated, for which a methane mixing ratios record has been kindly made available by colleagues at the AGH University.

To test the COMET model performance for Kraków site, the data was used for the period between April 18th and July 10th, for which meteorological data on a fine grid of 0.2° x 0.2° lat – long was available. All available methane emission databases were used for this part of the exercise. The time-series of the results are plotted in fig. 7 and scatter plots can be seen in fig. 4 (page 8, left side).

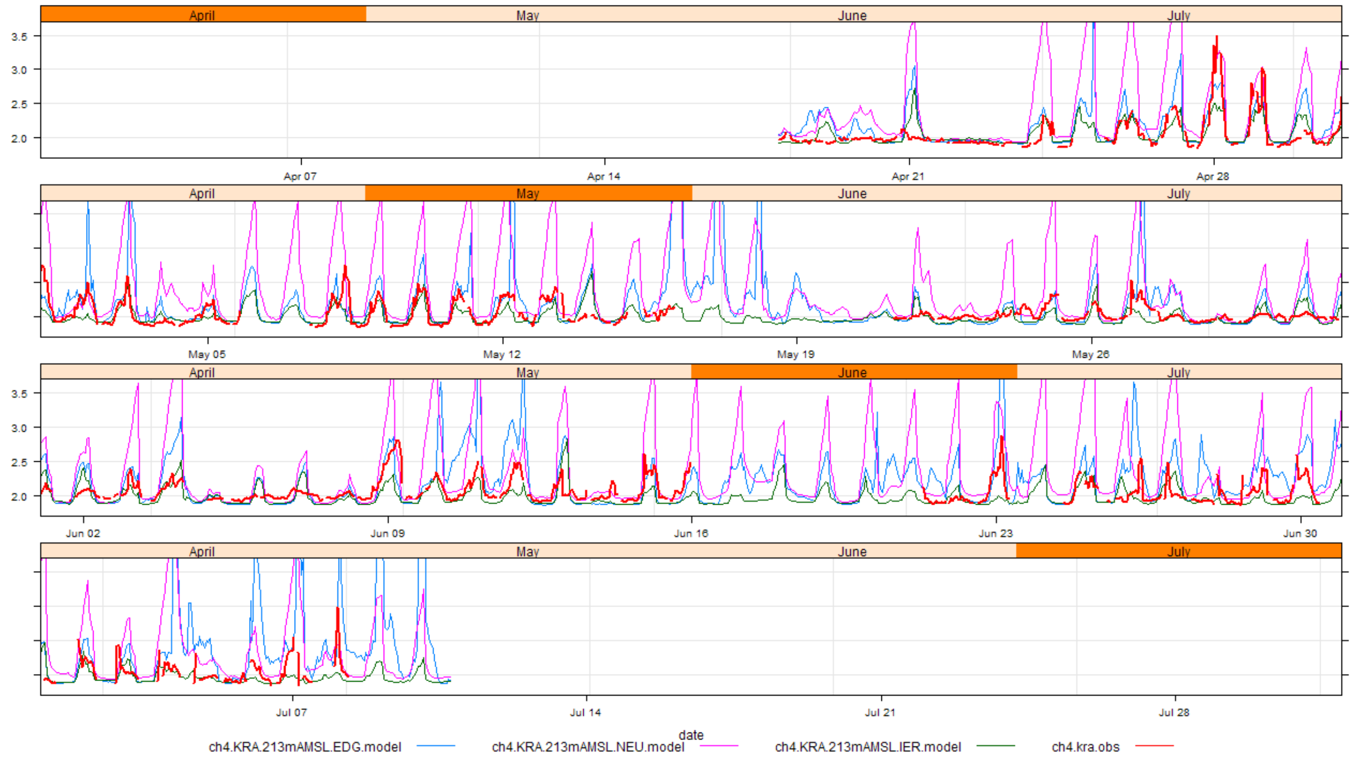

fig. 7. Model results vs. observations. 213mAMSL level, 0.2° x 0.2° ERA-Interim meteo data, all available emissions databases.

The analysis of time-series shows quite some similarity between the model predictions and measured concentrations. This is also confirmed by the scatter plot analysis, which point at the NitroEurope database as the one that gives the best results. Although the parameters of the fit are not very good even in the best of cases (R^2 = 0,33 with the slope of 1.8), the results may include some information on the validity of the values of the emissions contained in the used databases. For example, it is clear from the fig. 7 that values calculated using the IER database are always higher than the observations, which would point to a general overestimation of sources strength in the Kraków area. However, to quantify the size of these overestimations, a detailed event-based analysis is required, with a closer analysis of the meteorological conditions, the modelled trajectory path, and the emissions grid cells values. An example of such an analysis is shown in the next section.

3.4. Event-based validation of EDGAR emission database.

Using the methane concentration values measured at the surface station, it is possible to crudely evaluate the quality of the used emission database using a simple event-analysis. In this section an example of such an analysis will be performed using the Kraków station measurements and values calculated with a COMET model coupled with EDGAR emissions database.

A map extracted from EDGAR database provides an overview of the total surface methane emissions in the Southern Poland region (fig. 8). As can be seen, there are several strong sources in the area of Kraków, including the Upper Silesia, a region with a multitude of coal mines and coal-based industry. In the EDGAR database, the values of the emissions can be very large here, sometimes above 200 000 tons / grid cell / year which is approximately equal to 80 g km^{-2} s^{-1}. As can be seen in fig. 9, this generates peaks in methane concentrations much higher than the observed values.

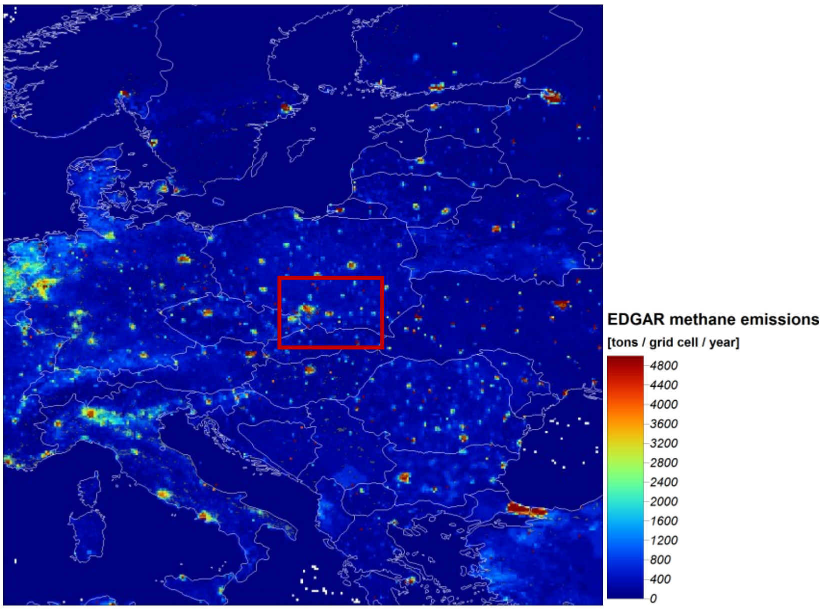

fig. 8. EDGAR methane emissions in Central-Eastern Europe. Total annual emissions shown. The red rectangle represents the study area.

From the modelled values generated as described in section 3.3, one period of interest has been chosen for the analysis. Spanning one week of measurements and corresponding modelled values, it includes an initial period of good correlation between the two (events A-D), after which the model starts to overestimate the concentrations at the station (events E-H).

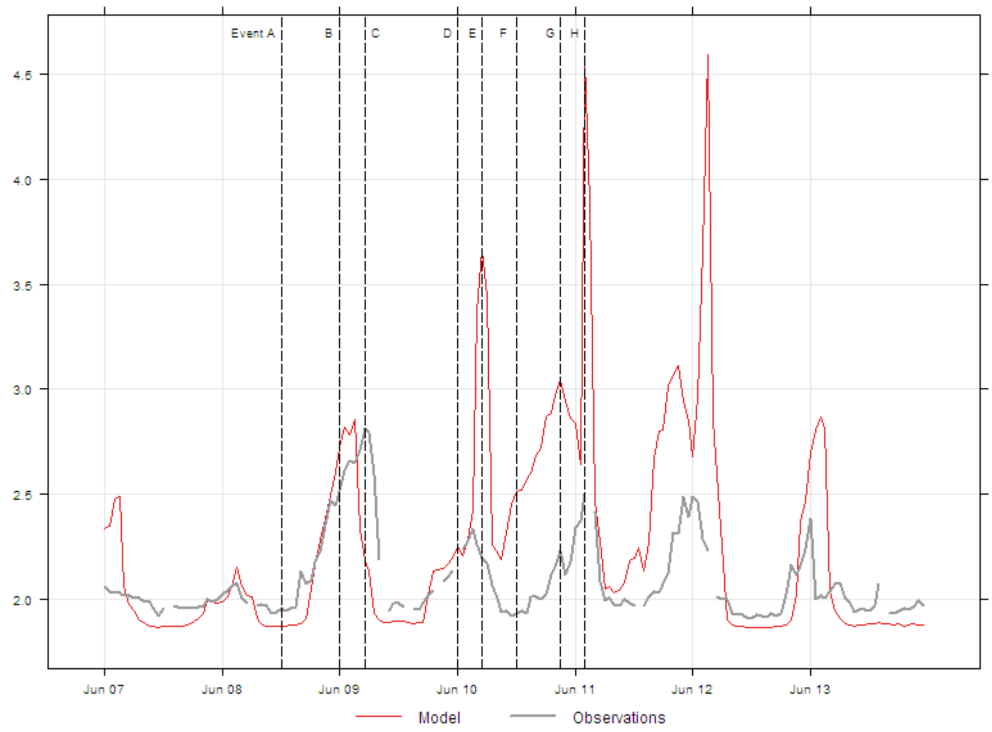

fig. 9. The period chosen for the event analysis. Observation data from Krakow station (2008). Modelled data calculated as described in section 3.3.

Analysis of the reasons behind these events shows that during the chosen period there has been a change of wind direction over the measurement station. While during the first few days the prevailing wind direction was easterly (E), it changed to westerly (W) on June 10th. This can be seen by the trajectory analyses plotted on following pages (fig. 10 - fig. 12). Each circle represents one of the positions of the trajectory one hour after the previous position. The size of circles diminishes as the trajectory goes forward in time, also representing (although not accurately[2]) a reduction in area of influence (AOL) used in model calculations.

[2] Sizes of circles are smaller than respective AOL's, which would span 2-3 grid cells on the 0.1 x 0.1 grid. They are not shown here in a proper scale for clarity of other information.

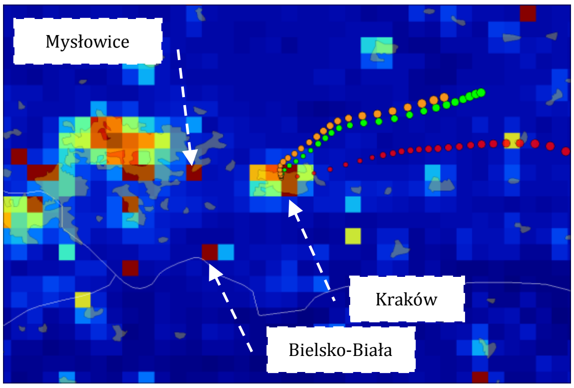

fig. 10. Trajectories for the events: A – red. B – green. C – Orange. Main urban areas are shown in pale grey. Important locations are indicated by names.

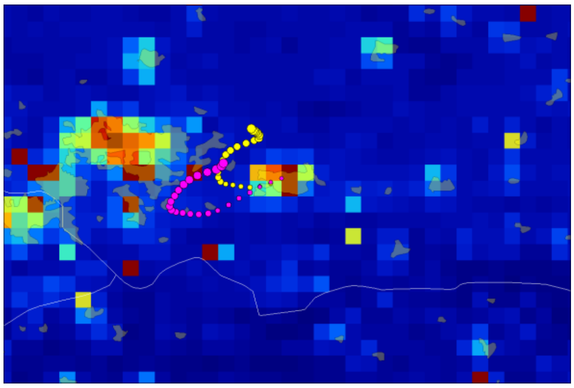

fig. 11. Trajectories for the events: D – yellow. E – purple. Main urban areas are shown in pale grey

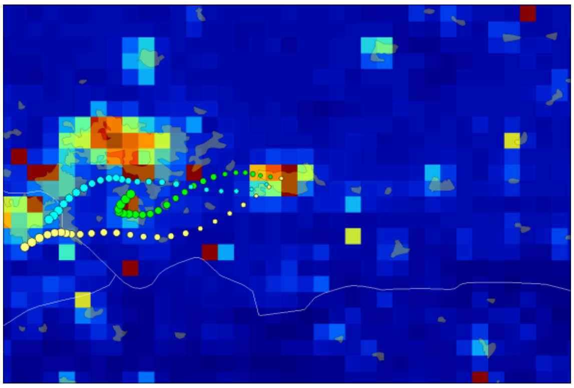

fig. 12. Trajectories for the events: F – green. G – blue. H – yellow. Main urban areas are shown in pale grey

Further analysis requires the information on the height of the PBL for each of the trajectory points. It can be seen that the final values of the mixing height for the events A and F are both high. This corresponds to the well-mixed atmosphere during the daytime. Because of the mixing, the information on the sources of the methane is consequently lost in the model, and predicted (as well as measured) values of methane mixing ratios are low.

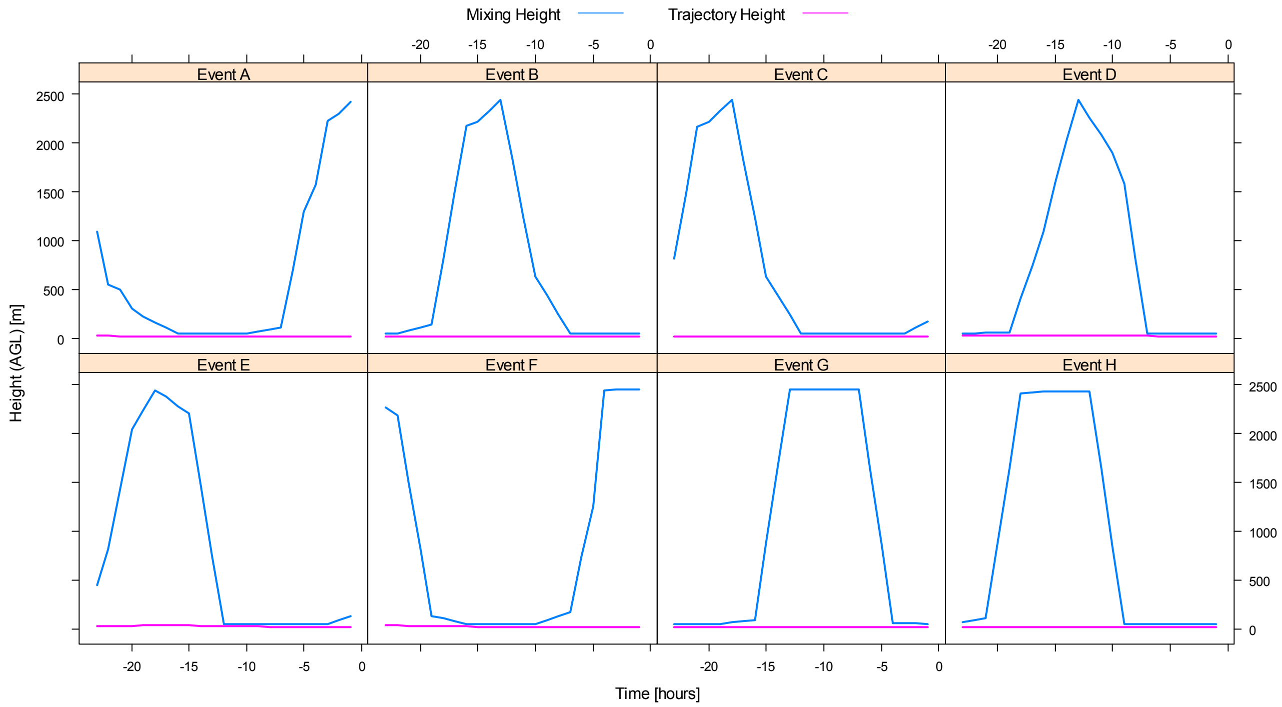

fig. 13. Modeled mixing height and trajectory height time plots. Please note that 0 on the x-axis marks the time of a trajectory arrival at the measurement site.

Events B, C and D show a different behaviour of the mixing height values. The measurements follow a several hours long periods when the air column was characterised by low mixing heights, representing night-time inversions. In this case, large amounts of methane emitted from the surface sources are distributed in a relatively low volume of air, resulting in its high concentrations. Since this inversion state can last only some 10 hours, only the closest sources can influence the concentrations. In the case of events B and C there are no other significant sources of methane beside the low emission of the northern part of the city. The correlation between the modelled values and measurements is good, therefore the values reported by EDGAR seem to be confirmed.

This is not the case when we consider the events E, G, H. Although the mechanism that causes high methane concentration is the same, the values predicted by the model deviate strongly from the values measured at the site. Especially interesting are the E and H events, where the predicted methane mixing ratios are particularly high. Although each of these events is influenced by the local city emissions, it is unlikely that the source of error lies in the overestimation of methane emissions in the western part of the city, because in that case similar overestimations would be seen during the D event, which is not the case. Therefore the only plausible explanation is that the overestimated values of emissions are farther away from the city, most probably in Silesia.

In the case of events F and G, the only possible source of error is the overestimation of the emissions in the Silesia area, since the trajectory is passing in the close vicinity of its location. Since this region is spatially a large source, quantification of the error in the emission grid values may prove to be difficult and is not attempted in this report.

3.5. Conclusions.

The main goals of this Exchange Grant were to allow the author to learn in practice how to use an atmospheric transport modelling tools to describe methane circulation in the atmosphere while trying to adopt the COMET model for the conditions of Kasprowy Wierch mountain station.

While it is safe to assume that the first task was fully accomplished, the results from the second are less than encouraging. Due to the complicated terrain topography, that is not well described in the mechanics of the current COMET version and the underlying transport model and ECMWF meteorology, the model predictions cannot be viewed as reliable for such a specific site and that other solutions will be better suited for the task.

However, this is not the case when a station inside PBL is considered. For Kraków city station, model results correspond to the observations much better, although there is still a significant space for improvement. Possible courses of further action should include an improvement of the background calculations method, and also modifications in the emissions database to more recent versions.

4. Projected publications to result from the grant

One publication is planned as a result of this exchange grant. It will describe a detailed comparison between the several model estimations (COMET / FLEXTRA, COMET / FLEXPART and WRF-Chem) and observations from sites located in Poland (Kasprowy Wierch, Kraków and possibly Białystok). Similarly to this report, an analysis regarding the quality of the emissions database is also planned, although with more sophisticated methods.

5. Other comments

The author wishes to sincerely thank A. Vermeulen for his invitation, and for all the support and help provided during the author's stay in the Netherlands. The hospitality of the Host Insitution – Energy Centre of the Netherlands is also kindly acknowledged.

6. References

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