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Retrieval of methane source strengths in Europe using a simple modeling approach to assess the potential of space-borne lidar observations

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Abstract

We investigate the sensitivity of future space-borne lidar measurements to changes in surface methane emissions. We use surface methane observations from nine European ground stations, and a Lagrangian transport model to obtain surface methane emissions for 2010. Our inversion shows the strongest emissions from the Netherlands, the coalmines in Upper Silesia Poland, and wetlands in southern Finland. Our simulated methane surface concentration captures at least half of the daily variability in the observations, suggesting that the transport model is correctly simulating the regional transport pathways over Europe. With this tool we can perturb the surface fluxes and see the resulting changes in the simulated column methane measurements. For example, we show that future lidar instruments can detect a 50 % reduction in methane emissions from the Netherlands and Germany, but only after averaging measurements on a monthly time scale.

1 Introduction

Although methane (CH_4) is the second most important anthropogenic greenhouse gas, it is arguably just as important as carbon dioxide (CO_2) from a policy perspective. Several studies conclude that in order to reduce net anthropogenic radiative forcing, it costs less to cut CH_4 emissions compared with CO_2 emissions (Shindell et al., 2012; Delhotel et al., 2006). To monitor future CH_4 emissions, policy makers and government officials will desire estimates of surface fluxes at a fine temporal and spatial resolution. Scientists also need this information to understand global and regional CH_4 budgets and the physical processes that control them.

Because observed emission rates of CH_4 are highly variable over small temporal and spatial scales, scientists have often resorted to a top-down approach to determine emissions. This method uses total column CH_4 observations from space in an inversion algorithm to estimate surface emission fluxes (Bergamaschi et al., 2009; Meirink

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et al., 2008). Currently, the space-borne sources for near-surface CH_4 information are the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) instrument onboard the late ENVISAT satellite platform (Bergamaschi et al., 2007; Frankenberg et al., 2011), and more recently the Greenhouse gases Observing SATellite (GOSAT) operated by the Japan Aerospace Exploration Agency (Schepers et al., 2012). These instruments measure changes in CH_4 spectral absorption from reflected near-infrared solar radiation, so they are susceptible to contamination from undetected clouds and aerosols. Too often global maps of CH_4 retrievals from today's passive satellite instruments have data voids over persistent cloudy regions, even if the clouds are optically thin. They also cannot make measurements in darkness or low sunlight conditions.

There are several planned space and aircraft based instruments that will use laser technology to measure the total CH_4 column (Riris et al., 2013; Ehret et al., 2008) and total CO_2 column (Abshire et al., 2010). This approach should remedy some of the current issues concerning high-latitude coverage and scattering from clouds and aerosols. Indeed, a recent aircraft demonstration campaign to test the feasibility of lasers to measure CO_2 has successfully retrieved CO_2 concentrations in thin cloud conditions (Anand Ramanathan, personal communication, 2013; Abshire et al., 2013). In the frame of a German–French climate monitoring initiative, DLR (Deutsches Zentrum fuer Luft- und Raumfahrt) and CNES (Centre National d'Etudes Spatiales) proposed a MEthane Remote LIdar MissioN (MERLIN) on a small polar orbiting satellite. The DLR Institute of Atmospheric Physics is also developing an airborne lidar system for demonstration and satellite validation purposes. Performance simulations have shown the basic ability of such active remote sensing systems in improving the accuracy of global methane observations (Kiemle et al., 2011; Stephan et al., 2011).

The question remains whether these new instruments will be able to detect changes in surface CH_4 emissions that occur at the state, nation or continent spatial scale. These changes may arise from policies that hopefully reduce anthropogenic emissions or from natural processes. These include climate feedback effects on permafrost soils,

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ocean hydrate sediments, and wetlands, in regions that are often difficult to access. This study's approach is to first build a retrieval algorithm that estimates CH_4 emissions using the FLEXPART Lagrangian transport model constrained by hourly CH_4 surface observations. Then we evaluate our emission estimates with existing emission inventories to insure that our emissions are realistic. Finally, we perform sensitivity studies by changing the CH_4 emissions and comparing the predicted changes in atmospheric CH_4 column amounts with the precision of future space-borne instruments.

2 Inverse method and results

We have developed a retrieval algorithm that estimates CH_4 emissions using the FLEXPART Lagrangian transport model along with CH_4 data sampled hourly at nine European ground stations in 2010. As shown below, the surface CH_4 concentrations simulated by the transport model reproduce much of the short time scale (hourly) perturbations in the CH_4 concentrations suggesting that these fast fluctuations, rather than the CH_4 absolute value, provide information about the source strength. The approach is to divide northern Europe into 262 tiles (Fig. 1) and initially assume a constant emission flux from each tile.

The CH_4 is transported by a 3-D Particle Dispersion Model (FLEXPART), developed at the Norwegian Institute for Air Research (Stohl et al., 2005). The FLEXPART model is run independently for each tile using NOAA Global Forecast System (GFS) meteorology fields at 3 h time and 0.5° resolution. The GFS uses 64 vertical sigma-pressure hybrid layers. The FLEXPART model actually transports the CH_4 as particles that have a lifetime of 20 days. We understand that CH_4's actual lifetime is on the order of twelve years, but uncertainties of the FLEXPART model grow so that after 20 days the results are unreliable. We are implicitly assuming that after 20 days the CH_4 particles become part of a background concentration term.

For each tile we release the equivalent of 1 kg day^{-1} of CH_4 at 150 m above ground level, assume a climatological OH field and an OH reaction rate that varies with temper-

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ature (3.5×10^{-15} cm^3 s^{-1} at 25 °C). We have set the FLEXPART model vertical domain to simulate concentrations from the earth surface to about 400 hPa. We do not attempt to simulate stratospheric intrusions. Our forward model (shown in Fig. 2) calculates perturbations in the surface CH_4 concentration at any geographical location by adding up the contribution ($S_i \cdot C_i$) from each of the 262 tiles. A background concentration (B) is also added to the perturbations. S_i is the source strength for the ith tile and C_i is the surface concentration simulated by FLEXPART using a 1 kg day^{-1} source. The background value is a retrieved quantity from the retrieval algorithm.

Once the individual trajectory calculations are run for each tile, we perform a retrieval algorithm. As shown in Fig. 3, we retrieve a source strength (S_i) for each tile and a background value (B) for each station. The retrieval algorithm adjusts the source strengths of each individual tile, until the simulated (analyzed) observations best match those observed. The background value at each station is also adjusted. A standard linear inverse method (chapt. 3 of Rodgers, 2000) is used and iterated until the source strengths and background values have converged. Note that the retrieved source strengths are constant over a 45 day period and the retrieved background values vary linearly with time over each 45 day period.

The observations for this retrieval algorithm are sampled at nine European stations that continually measure surface CH_4, preferably every hour. These are listed in Table 1 and further described in the Appendix. There are additional European stations that only sample weekly or monthly (labeled by "Event"), but we could see no significant change in the retrieved emissions when these datasets were included. The high temporal resolution hourly observations captures information on CH_4 filaments passing over a ground station, which are then deconvolved by our retrieval algorithm to yield the surface emission strengths. This interesting finding is due to the complexity of transport in association with a strong spatial heterogeneity of emissions, as Fig. 5 will demonstrate.

Figure 4 shows the observed hourly CH_4 concentrations at the nine European ground stations used for this study (black trace). The FLEXPART simulated (analyzed) concen-

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trations that best match the observations are also shown (red trace). The FLEXPART model is often able to reproduce the weekly variability and sometimes captures hourly spikes in the observations. The model performs best at the Mace Head Ireland site and the Pallas-Sammaltunturi site in northern Finland. These are remote sites situated far away from any pollution sources and Pallas is usually above the convective boundary layer. So the variability at these sites is largely influenced by regional transport from European sources.

The model is unable to capture the minimum values at mountain stations: the Jungfraujoch site (elevation of 3580 m), the Plateau Rosa site (3480 m) and Monte Cimeone (2165 m), all located in the Alps; and the Kasprowy Wierch (1989 m) station in the Carpathian mountains. One explanation is that the 0.5° resolution NOAA GFS winds are not able to capture the actual wind patterns driven by the local complex topography. Conditions of strong upslope and down slope winds increase the influence of local CH_4 sources. Indeed, at these sites the surface elevation a.s.l. reported by the GFS meteorological fields is much lower than the actual elevation of the station.

Also note that while the model is able to reproduce the timing of the spikes at the Kollumerwaald site, it only simulates half the amplitude. Issues with the GFS fields and FLEXPART's simulation of boundary layer height, and mixing may explain why the model captures the timing of spikes but not the amplitude. If a local source is present, a shallower nighttime boundary layer capped by an inversion will foster higher CH_4 concentrations. However, a time-series plot of hourly CH_4 concentrations at Kollumerwaald sorted by nighttime and daily measurements (not shown) shows that spikes are not limited to nighttime conditions. Understanding these spikes requires further research.

The squared correlation coefficients between the observed and simulated concentrations range from 0.48 to 0.77. This suggests that at least half of the variability in the observed surface concentrations can be explained by the regional transport simulated by the FLEXPART model. The remaining unexplained variability in the surface concentrations is from short temporal scale variability in the emissions strengths (we assume

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a constant emission over a 45 day time period), or weaknesses in the trajectory model, or weaknesses in the GFS meteorological fields.

In a similar study Vermulen et al. (2006) used the FLEXPART trajectory model to simulate a time series of hourly CH_4 surface concentrations at Mace Head and at the Cabauw tower in the Netherlands during 2002. They used prescribed CH_4 surface fluxes from the METDAT (METhane DATabase) and the EDGAR databases. They were able to simulate ~ 75 % of the variability of the Cabauw observations and their simulation for Mace Head looks very similar to ours.

Another similar study used CO_2 concentration observations at three ground-based mountain stations: Plateau Rosa, Monte Cimone and Zugspitze along with the FLEXPART trajectory model to determine CO_2 source and sink regions (Apadula et al., 2003).

Figure 5 shows our retrieved source strengths for each tile. Note that our forward model assumes that these emissions are unchanged over a 45 day period. The strength values and uncertainties for tiles with significant emission are shown in black and white respectively. The uncertainties are derived from the averaging kernels (see Rodgers, 2000, chapt. 4) which are a diagnostic quantity from the retrieval algorithm.

Ideally, each of the nine ground stations (Table 1) would have back-trajectories transporting CH_4 from each one of the 262 source tiles during a 45 day period. Instead, there are times when no trajectories from a source tile pass over one of the nine observing ground stations and the strength uncertainty (as determined by the averaging kernel) will be high. This was too often the case when we attempted to resolve the strengths at temporal resolutions less than 45 days – a significant number of tiles had strength uncertainties that were larger than the actual values.

3 Discussion

We compare our retrieved emission strengths with reanalysis fluxes from the European Monitoring Atmospheric Composition and Climate (MACC) Collaborative Project

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for 2010 (Fig. 6a). These surface fluxes are inverted from total column CH_4 amounts from SCIAMACHY (Bergamaschi, 2009) and should include both natural and anthropogenic sources. Our retrieved surface fluxes are qualitatively consistent with the much smoother MACC reanalysis over the UK and Central Europe. (Their inversion grid has a much coarser spatial resolution than ours.) However the MACC reanalysis does not show the strong Scandinavian emissions that we retrieve in July.

We can also compare our surface fluxes with the Emission Database for Global Atmospheric Research (EDGAR, 2011) and understand that this data set does not include natural emissions. Figure 6b shows the total anthropogenic emissions based on government and commercial statistics for 2008. The fine 0.1° by 0.1° spatial scale of this dataset is able to resolve the strong emissions from the major metropolitan, industrial, mining and agricultural areas. To facilitate comparison we show our average retrieved emissions for May–December 2010 in Fig. 6c. The fine resolution EDGAR emissions are integrated over our 262 surface tiles in Fig. 6d.

Almost all of Poland's coal mining activities are concentrated in Upper Silesia (shown by the dark pentagon symbol in Fig. 6). It is one of the largest in Europe and produces almost all of Poland's coal. During the extraction process significant CH_4 is released from the coal and surrounding rock. This CH_4 must be quickly removed from underground mines through ventilation systems. Although some of the mines recover the CH_4, a significant amount is still emitted directly to the atmosphere. The EDGAR database reports that "fugitive emissions from solid fuels" (i.e. coal production) constitute half of Poland's CH_4 emissions. So the strong emission in southern Poland reported by EDGAR ($113\,mg\,day^{-1}\,m^{-2}$, Fig. 6d) is largely from venting coalmines. Our retrieved surface fluxes also show a strong source near Upper Silesia of $57\,mg\,day^{-1}\,m^{-2}$ shown in Fig. 6c.

The EDGAR database also shows the Netherlands as strong source of CH_4 (Fig. 6b and d). This is not associated with the tulip industry, instead the EDGAR database lists: "enteric fermentation and manure management" as the largest contributors to the Netherlands. Our retrievals consistently show strong emissions in this location.

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The strong summer surface fluxes we retrieve over Scandinavia (Fig. 5) are from natural sources (wetlands) so they are not reported in the EDGAR database. In Scandinavia the natural wetlands are dormant during the period 15 May to 30 June. Snow is melting and temperatures are cold. The active tiles to the East of Pallas are probably leaks from CH_4 gas production fields located just outside the tile domain in the Russian Federation. From 1 July to 14 August many Scandinavian tiles, especially over Finland, become active. The strongest source location (468 mg day^{-1} m^{-2}) is in Southern Finland (tile #201) and is entirely dominated by lakes and wetlands. We show negligible CH_4 sources during October over Scandinavia – consistent with the colder temperatures. The active tiles over Finland in December are probably anthropogenic sources but they are not understood.

To study whether space based instruments will be sensitive to changes in surface fluxes, we need a model that generally simulates the spatial distribution of emissions and uses source strengths that are at least in qualitative agreement with accepted values. We feel that our model meets these criteria so we can proceed with some sensitivity experiments.

For all of our FLEXPART model runs the simulated CH_4 is able to move vertically in the atmosphere due to diabatic and convective processes captured by the GFS meteorology. We output the CH_4 at 20 vertical levels in the atmosphere (surface to $\sim$ 400 hPa). While the model does not calculate an absolute column amount, it provides perturbations from an unknown total column value. This should be adequate for our purposes. Figure 7 (left hand panels) shows the average column mean CH_4 above an unknown background level for two typical summer days. The plumes of CH_4 over Europe are at least 30 ppb above background and sometimes more.

Remote sensing laser instruments designed to measure CH_4 from air and spaceborne platforms are being developed in Europe and the United States. The spaceborne and aircraft version of the MERLIN (DLR/CNES) instrument is expected to have a precision of $\sim$ 18 ppb over 50 km spatial averaging (Kiemle et al., 2011). The Methane Sounder, being developed at NASA Goddard Space Flight Center, was tested on the

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NASA DC-8 in the summer of 2011 (Riris et al., 2012). While this is a breadboard instrument, only designed to demonstrate the use of laser technology, its precision is ∼ 50 ppb. The expected precision from the mature instrument design is ∼ 14 ppb. At maturity, both of these technologies would be able to detect the CH_4 plumes shown on the left hand panels of Fig. 7. This will definitely be an improvement over the current passive satellite instruments.

But will these new instruments (MERLIN and NASA Methane Sounder) be able to detect changes in the current emission rates? The right hand panels of Fig. 7 show the change in the total column CH_4 when surface emissions from Germany and the Netherlands are decreased 50 %. These perturbations are on the order of 3 ppb, which is below the single sample detection limit of the new instruments. So we would need an instrument with a precision of 3 ppb to detect *daily* changes in CH_4 flux at the national level. If we relax our requirements to *monthly* (N = 30) averages in CH_4 fluxes then we will need an instrument with $3\,\text{ppb} \cdot \sqrt{30} = 16.4$ ppb which is close to the specifications of the proposed instruments.

4 Conclusions

We have developed a simple forward model using the FLEXPART trajectory model that can simulate space-borne (total column) and aircraft (partial column) measurements from proposed CH_4 lidar instruments. This forward model is used in conjunction with a retrieval algorithm to obtain estimates of surface emissions over Europe that are constrained by 3 h surface observations sampled at nine European ground stations. The model is often able to simulate the daily variability in surface CH_4 concentrations observed at the ground stations. This suggests the model correctly simulates the filaments that transport CH_4 from their sources to the ground-based stations.

This model can then be used to determine if the detection limits and measurement precision of the proposed instruments are low enough to detect significant changes in CH_4 surface emissions. We have applied our model to a future scenario where the

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emissions from Germany and the Netherlands are reduced by 50 %. In order to detect this perturbation, at least one month of column CH_4 amounts would need to be sampled using the specifications of the proposed instruments.

We plan to modify our analysis package to retrieve surface fluxes using simulated space-borne and aircraft column measurements (and their uncertainties) as the observed quantity. This tool will enable us to more conclusively assess if the new instruments can detect changes in the surface emissions.

Appendix

The Pallas-Sammaltunturi station (560 m) is located within the northern boreal forest zone and is free of large local and regional pollution sources (Aalto et al., 2006). Analysis of radon-222 concentration indicates that the station is very rarely inside the surface inversion layer (Paatero et al., 1999). This is consistent with its location atop a 560 m hill.

The Schauinsland monitoring station (1205 m) is situated on a mountain ridge in the Black Forest, southwest Germany, above the polluted Rhine valley. During night the station is usually above the boundary layer, while at daytime, particularly in summer, Schauinsland station mostly lies within the convective boundary layer.

The Neuglobsow sampling site (65 m) is surrounded by lakes and forested areas in all directions, and therefore is minimally influenced by local sources. Measurements are representative for the background in North East Germany.

The Jungfraujoch station (3580 m) is a High-Alpine station located in the centre of Western Europe. During extended periods, the Jungfraujoch is decoupled from the tropospheric boundary layer below. On the other hand, transport of polluted boundary layer air to the height of the Jungfraujoch occurs periodically because of meteorological transport connected with the passage of fronts, foehn winds, or thermally driven convection during anticyclonic periods in summer (e.g. Reimann et al., 2008).

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Kasprowy Wierch (1989 m) located on a Peak in Tatra Mountains vertically situated within the transition zone between the free troposphere and the boundary layer (Necki et al., 2003).

The Mace Head station (5 m) is located on the west coast of Ireland, offering westerly exposure to the North Atlantic Ocean (clean sector, 180° through west to 300°) and the opportunity to study atmospheric composition under Northern Hemispheric background conditions as well as European continental emissions. The meteorological records show that on average, over 60 % of the air masses arrive at the station via the clean sector. These air masses are ideal for carrying out background aerosol and trace gas measurements. Significant pollution events also occur at the site when European continental air masses, generally originating from an easterly direction, reach Mace Head.

The Kollumerwaard station (0 m) is located in a coastal agricultural area.

The Monte Cimone station (2165 m) is situated in the Italian northern Alpennines. There are no local sources of contamination and no access by road.

Plateau Rosa (3480 m) is situated in the Western Italian Alps. Due to its high altitude and location, i.e. in the free atmosphere upon a large snow-clad bare mountain plateau and far from urban and polluted zones, it is suitable for the background measurements of greenhouse gases.

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Table 1. Ground stations sampling hourly methane concentrations used in this study. Elevation is in meters. CRDS is cavity ring down spectrometry and GC-FID is gas chromatography with flame ionization detector.

Station	Lat, Lon	Elev	Method/Scale	Institution	PI
Pallas-Sammaltunturi Finland	67.97, 24.12	560	CRDS/NOAA04	FMI	Juha Hatakka, Tuula Aalto
Mace Head, Ireland	53.33, −9.9	5	GCFID/NOAA04	AGAGE	Ray Wang Simon O'Doherty
Kollumerwaard, Netherlands	53.33, 6.28	0	GC-FID/NIST	RIVM	Hans Berkhout
Neuglobsow, Germany	53.17, 13.03	65	GCFID/NOAA04	UBA	Karin Uhse
Kasprowy, Poland	49.23, 19.98	1989	GCFID/NOAA04	AGH-UST	Jaroslaw Necki
Schauinsland, Germany	47.92, 7.92	1205	GC-FID/NOAA04	EMPA	Karin Uhse
Jungfraujoch, Switzerland	46.54, 7.99	3580	CRDS/NOAA04	EMPA	Martin Steinbacher
Plateau Rosa	45.93, 7.71	3480	GCFID/NOAA04	RSE	Francesco Apadula Daniela Heltai Andrea Lanza
Monte Cimone, Italy	44.18, 10.7	2165	GCFID/NOAA04	ISAC	Jgor Arduini

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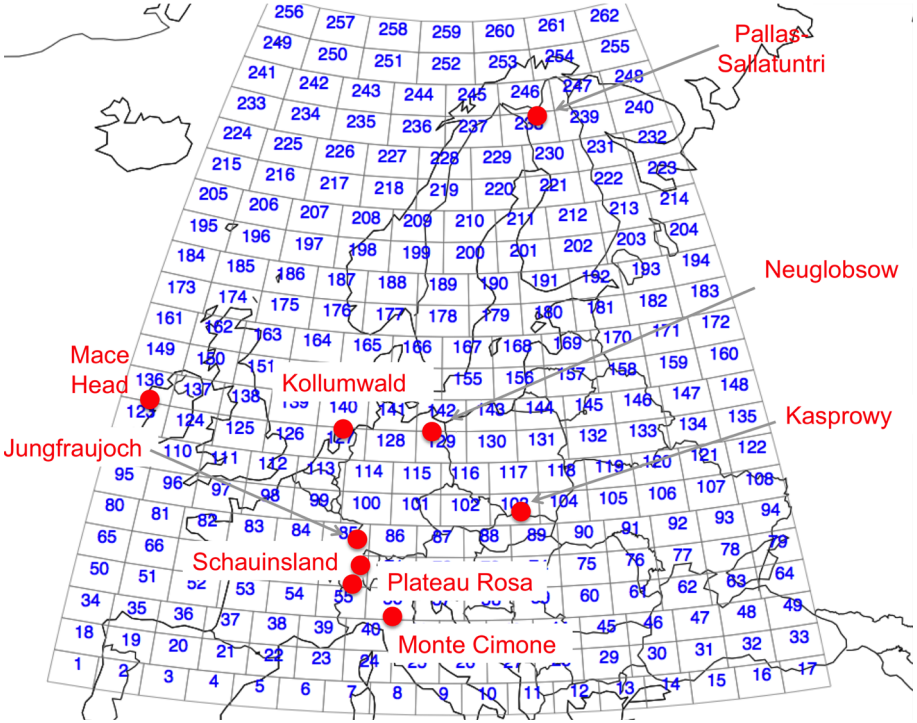

Fig. 1. The 262 tiles used in this study are 1.5° latitude, and 1.5 to 7° longitude, from S to N, in order to obtain tiles at a constant area of approximately 43 000 km^2.

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13, 19559–19582, 2013

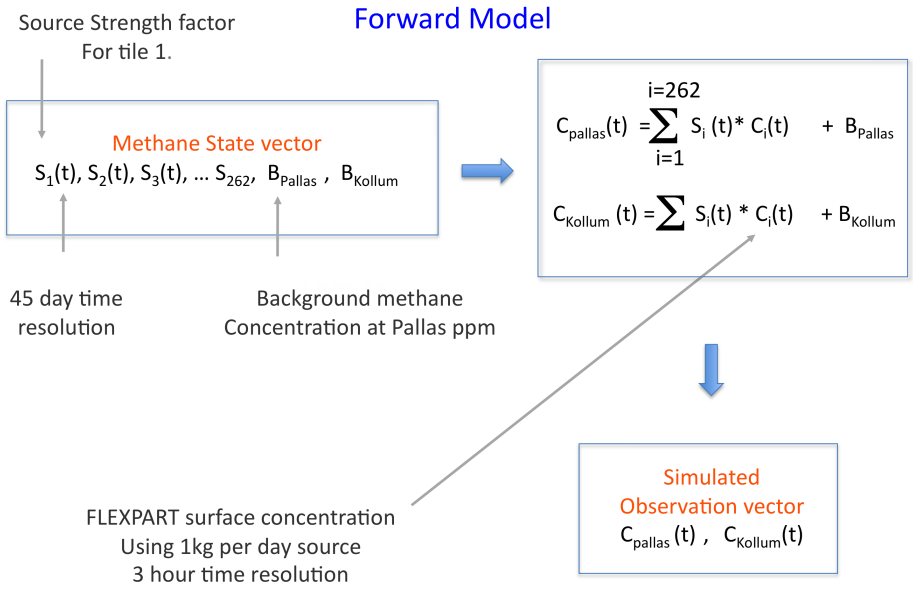

Fig. 2. The forward model used to calculate surface methane concentrations. For simplicity this schematic only shows equations for two geographic locations.

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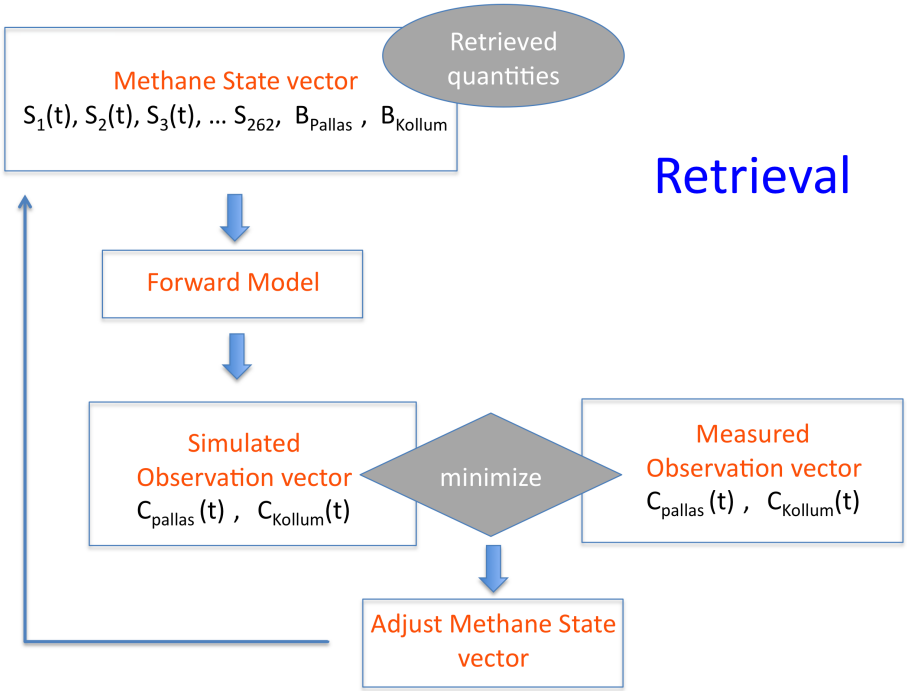

Fig. 3. Schematic of the retrieval algorithm used to obtain source strengths.

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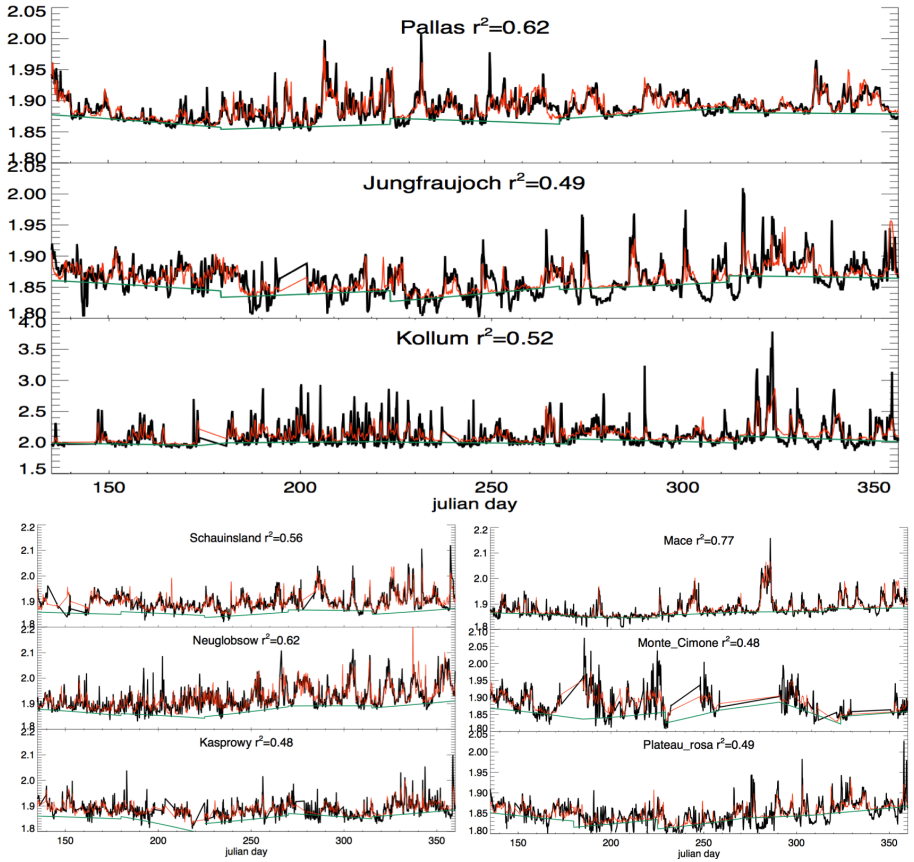

Fig. 4. Observed (black) and analyzed (red) hourly methane concentrations (ppm) at nine European locations for the year 2010. Contribution from the retrieved background concentration is shown in green.

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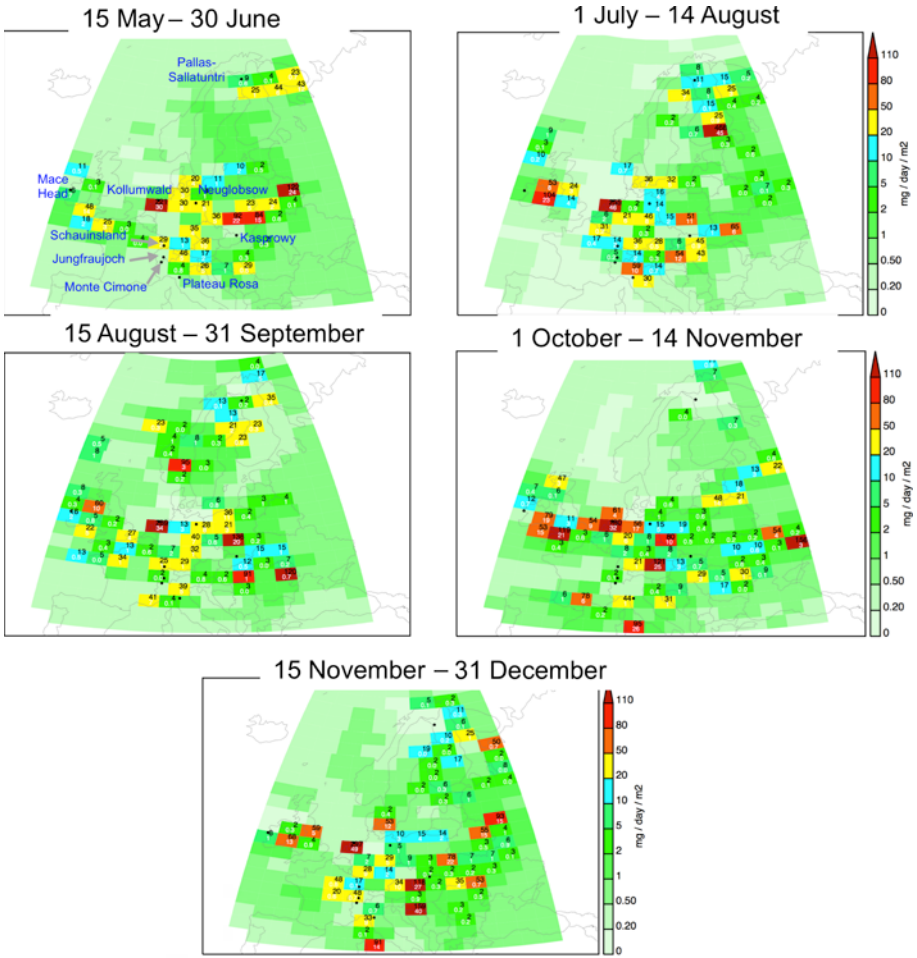

Fig. 5. Retrieved surface source strengths (mg day^{-1} m^{-2}) for five 45 day periods in 2010. Each tile with significant emission has value shown in black and estimated uncertainty in white.

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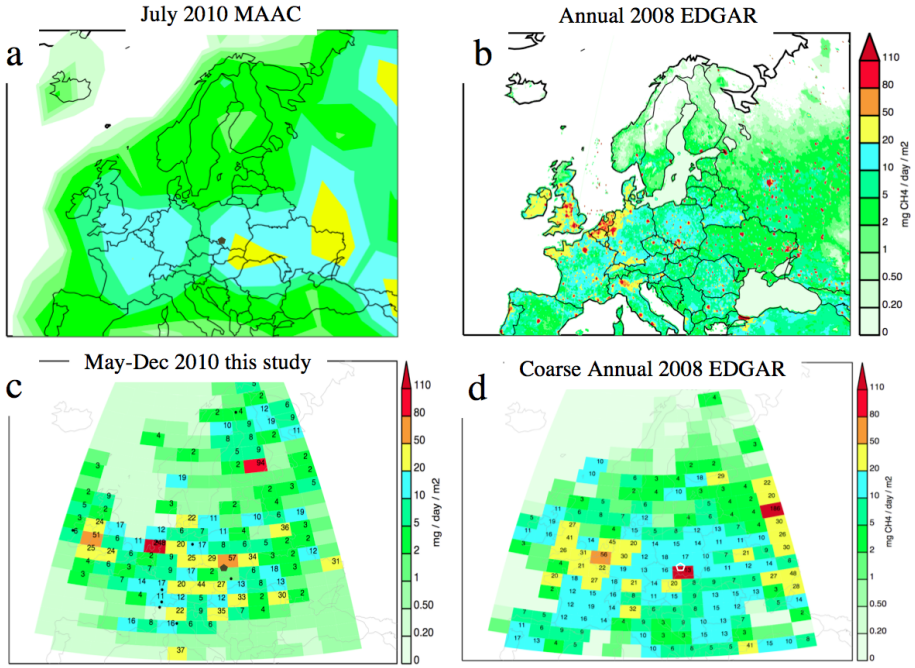

Fig. 6. (a) Monthly retrieved emission strengths from the European Monitoring Atmospheric Composition and Climate (MACC) Collaborative Project for July 2010. **(b)** Annual anthropogenic surface emissions from the Emission Database for Global Atmospheric Research (EDGAR) for 2008. **(c)** Average retrieved emission strengths from this study May–December 2010. Each tile with significant emission shows value in black. **(d)** EDGAR 2008 integrated over our 262 surface tiles. **(a)**, **(c)** and **(d)** show the upper Silesia mining district with a dark pentagon.

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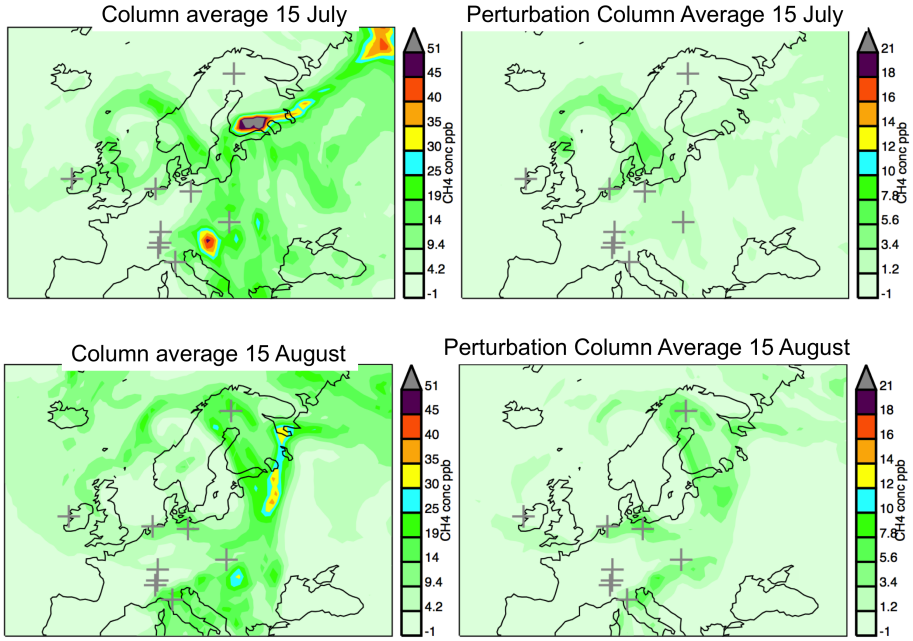

Fig. 7. Average Column methane concentration (left) and the perturbation in average column methane (right) from a 50 % decrease in emissions from Germany and the Netherlands during the summer of 2010.

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