

## Response to Anonymous Referee #1

We thank the referee for the thoughtful comments. We've responded to the major comments interspersed below. Referee comments are in red italics, and our responses are in black.

*By design of the method, the CH<sub>4</sub> emissions estimate cannot be more accurate than the CO inventory used for scaling. A priori, I would argue that CH<sub>4</sub> inventories are per se better than CO inventories since there is less variability in CH<sub>4</sub> sources and less changes in source patterns. Figure 3 also indicates that the EDGAR and the TNO inventories show much larger discrepancies for CO than for CH<sub>4</sub> (likewise text p.6, l.21 ff). Then, the authors need to extrapolate the CO inventories under changing background conditions (change of source type) inducing further uncertainties. What is the total CO-induced error on your CH<sub>4</sub> emission estimates? Are the conclusions robust against this error? The conclusion needs to be quantitative in that respect.*

We agree completely that this method hinges on having an accurate CO inventory, and we have stressed this point further in the updated manuscript, and added a more quantitative discussion regarding the impact the CO inventory has on the methane emissions inferred.

In other regions of the world, in particular the US, other research has suggested that fugitive natural gas and agriculture emissions are significantly underestimated, and CO inventories are reasonably accurate (e.g., Maasackers *et al.*, 2016; Wunch *et al.*, 2009; Kort *et al.*, 2016; Frankenberg *et al.*, 2016). We were surprised by our results from this region that seem to indicate that the methane emissions are overestimated.

*The above concerns a touched on by the manuscript but I would argue that they are so critical that they require quantitative answers not just handwaving arguments. In general, my concerns 2 and 3 could be addressed by a comprehensive simulation study based on CH<sub>4</sub> and CO emissions inventories, a chemical transport model and an observation operator that mimicks the sampling by the ground-based spectrometers. I would recommend implementing such an approach to invalidate my concerns and to verify the assumptions or to reconsider the methodology.*

We have run GEOS-Chem v12.1.0 with EMEP CO and EDGAR CH<sub>4</sub> emissions for the year 2010. From this global 3D run, we have integrated the profiles of CO and CH<sub>4</sub>, and computed dry-air mole fractions. The dry-air mole fractions were interpolated to the values at the locations of the 5 TCCON stations (Fig 1).

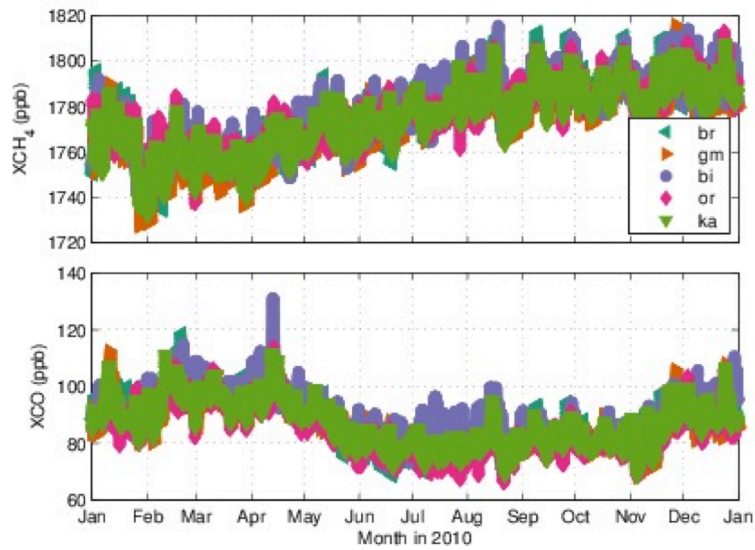


Figure 1:  $XCH_4$  (top panel) and  $XCO$  (bottom panel) integrated from GEOS-Chem and interpolated to the TCCON station locations.

Pairwise differences at each hour were computed between the 5 stations (Fig 2), and the changes, or anomalies in CO were related to the anomalies in  $CH_4$  (Fig 3).

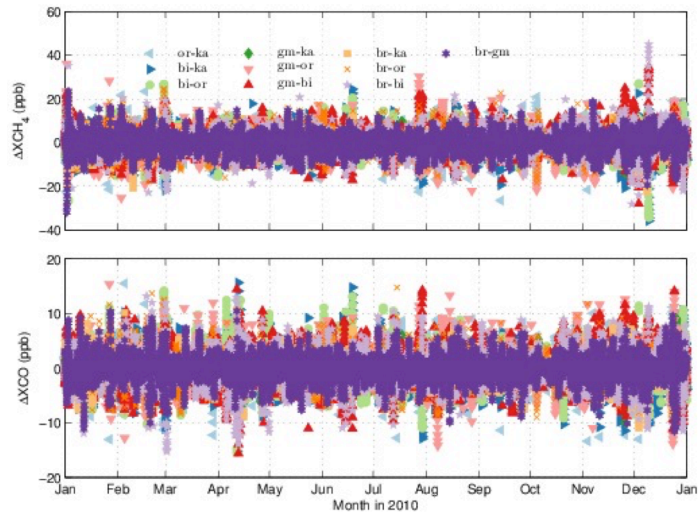


Figure 2: Pairwise differences are computed for  $XCH_4$  (top panel) and  $XCO$  (bottom panel) between each pair of stations. These are the "anomalies."

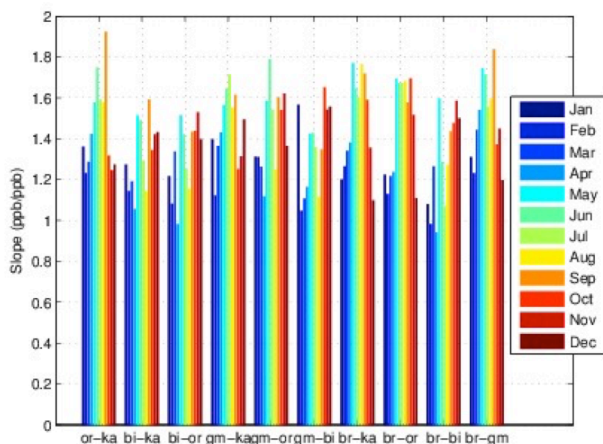


Figure 3: These are the anomaly slopes per station pair, coloured by month of the year.

Using these tracer-tracer anomaly slopes, we multiply the slope by the emission of CO from the model within the study region. We were able to return the annual CH<sub>4</sub> emissions in the model to within 2% of the true value (Fig 4).

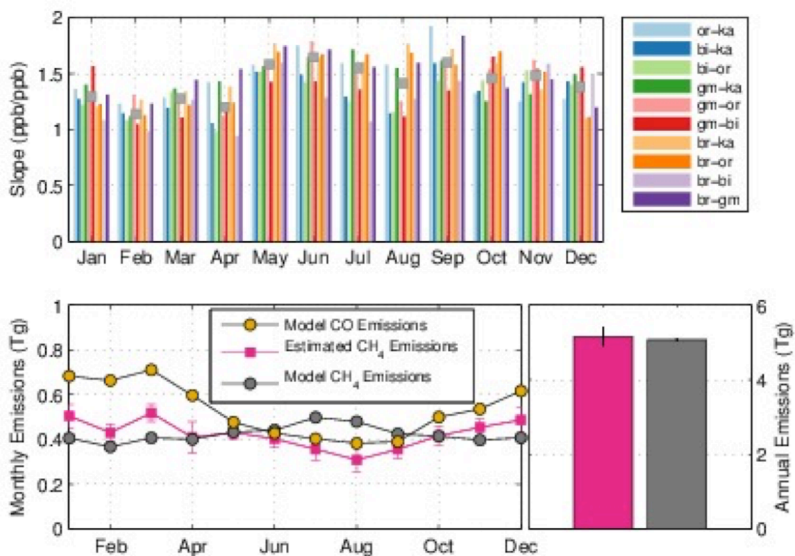


Figure 4: The top panel shows the slopes as a function of the month. The bottom left panel shows the monthly emissions from the model (CO and CH<sub>4</sub>) and the inferred monthly methane emissions from the anomaly analysis. The bottom right panel shows the annual inferred methane emissions (pink) and the model emissions (grey).

These results appear to show that our method is reasonably sound. The description and results from this

model experiment have been added in a new section to the paper.

### References:

Frankenberg, C. et al. (2016), Airborne methane remote measurements reveal heavy-tail flux distribution in Four Corners region, *Proc. Natl. Acad. Sci.*, 201605617, doi:10.1073/pnas.1605617113.

Kort, E. A., M. L. Smith, L. T. Murray, A. Gvakharia, A. R. Brandt, J. Peischl, T. B. Ryerson, C. Sweeney, and K. Travis (2016), Fugitive emissions from the Bakken shale illustrate role of shale production in global ethane shift, *Geophys. Res. Lett.*, 43(9), 4617–4623, doi:10.1002/2016GL068703.

Maasackers, J. D. et al. (2016), Gridded National Inventory of U.S. Methane Emissions, *Environ. Sci. Technol.*, 50(23), 13123–13133, doi:10.1021/acs.est.6b02878.

Wunch, D., P. O. Wennberg, G. C. Toon, G. Keppel-Aleks, and Y. G. Yavin (2009), Emissions of greenhouse gases from a North American megacity, *Geophys. Res. Lett.*, 36(15), 1–5, doi:10.1029/2009GL039825.

## Response to Anonymous Referee #2

We thank the referee for the thoughtful comments. We've responded to the major comments interspersed below. Referee comments are in red italics, and our responses are in black.

*The paper relies on a set of five stations which are distributed far apart from each other and assumes that the CO and CH<sub>4</sub> emitted between the stations are completely measured. – Can the authors show some evidence that for the majority of the days when the airmass was sampled by an upwind station then it was a few days later also sampled by the downwind station?*

The footprint of a TCCON measurement is large – typically hundreds to thousands of kilometers. Thus, on average, measurements from each TCCON station will be sensitive to the entire region. To prove this more quantitatively, we have run the GEOS-Chem model, sampling the model at the TCCON stations and applying the same analysis. The results show that our anomaly slope method returns the model methane emissions to within 2%. We have added a new section to the paper describing this.

*It also assumes that the typical emissions are consistent over time periods longer than a few days so that the CO and CH<sub>4</sub> are advected together. – This can only be true considering that no CO is lost, is that true especially as the major source of CO in Europe is coming from the urban area?*

We agree that our assumption is that the emissions are consistent over periods longer than a few days. The atmospheric lifetime of CO is about a month or so – much longer than the time it would take for the CO to advect from one end of Europe to the other. Although sources of CO and CH<sub>4</sub> are not necessarily (fully) co-located, with CO more in urban areas and CH<sub>4</sub> more in rural areas, in the European landscape these are relatively close to each other, and at scales of 10 x 10 or 25 x 25 km would appear mixed. We're unclear as to the referee's point regarding the urban source of CO – perhaps this is referring to co-emissions of NO<sub>x</sub> and O<sub>3</sub> in urban centres increasing OH? If so, we expect that a resulting shortened CO lifetime will still be long enough to advect across the study area

*As the author points out the TNO-MACC\_III CO emissions are on average 15% higher and CH<sub>4</sub> emissions are about 2% lower than the EDGAR v4.3.1 emissions in the study area. This indicates that the variability of CO is much higher for the selected study area and its surroundings. How does this variability propagate into the emission estimates of CH<sub>4</sub>?*

The relationship between CO and CH<sub>4</sub> should only rest on whether they are advected together, not as much on whether they are emitted in exactly the same place. Of course, in general, CO and CH<sub>4</sub> are **not** emitted from the same stack, and so it should be expected that the spatial distribution of CO and CH<sub>4</sub> are not identical. However, the emissions inventory maps show that typically the emissions are broadly co-located (i.e., within a few hundred km of each other).

*The author partly correlates the dip in CH<sub>4</sub> emissions in 2013 shown in Fig. 4 to the small up-tick in CO in the same year. However, there is a similar CO up-tick in 2010 but no CH<sub>4</sub> dip can be seen in the measurement. – Any explanation?*

The reviewer is correct that although the dip in 2013 is remarkable, there is also a CO uptick in 2010 without an impact on CH<sub>4</sub>. Hence, we have removed the explanation for 2013 from the manuscript since we cannot also explain the lack of response for 2010.

*Minor comments:*

*Page 4 line 6-7: but each slope for each season has*

Fixed.

*Page 4 line 13: is the reference to the fig 4 correct?*

Indeed, no. That should be Fig. A3.

*Page 5 line 33: check your argument – if winter is cold it should rather result in increased heating needs*

Thank you for pointing this out. 2013 was cold, which would indicate that more CO would be produced due to heating needs (not less CH<sub>4</sub> consumption). This discussion was removed from the manuscript as mentioned earlier.

*Page 8 line 29: reference of the European Environment Agency National Database (?) missing*

Fixed.

# Emissions of methane in Europe inferred by total column measurements

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**Abstract.** Using five long-running ground-based atmospheric observatories in Europe, we demonstrate the utility of long-term, stationary, ground-based measurements of atmospheric total columns for verifying annual methane emission inventories. Our results indicate that the methane emissions for the region in Europe between Orléans, Bremen, Białystok, and Garmisch are overestimated by the state-of-the-art inventories [EDGAR Emission Database for Global Atmospheric Research \(EDGAR\) v4.2 FT2010](#) and TNO-MACC\_III, possibly due to the disaggregation of emissions onto a [spatial](#) grid. Uncertainties in the carbon monoxide inventories used to compute the methane emissions **may** contribute to the discrepancy between our inferred emissions and those from the inventories.

## 1 Introduction

Recent global policy agreements have led to renewed efforts to reduce greenhouse gas emissions to cap global temperature rise (e.g., COP-21 (UNFCCC, 2015) and the Covenant of Mayors (European Commission, 2016)). This, in turn, has motivated countries to seek methods of reducing their greenhouse gas emissions. In Europe, methane emissions account for a significant fraction (about 11% by mass of CO<sub>2</sub> equivalent) of the total greenhouse gas emissions (UNFCCC, 2017). The lifetime of atmospheric methane is significantly shorter than for carbon dioxide, its 100-year global warming potential is significantly larger, and it is in near steady-state in the atmosphere, therefore significant reductions in methane emissions are an effective **short-term** [short-term](#) strategy for reducing greenhouse gas emissions (Dlugokencky et al., 2011). Emission reduction strategies that include both methane emission reductions and carbon dioxide reductions are thought to be among the most effective at slowing the increase in global temperatures (Shoemaker et al., 2013). Thus, it is important to know exactly how much methane is being emitted, and the geographic and temporal source of the emissions. This requires an approach that combines state-of-

the-art emissions inventories that contain information about the specific point and area sources of the known emissions, and timely and long-term measurements of greenhouse gases in the atmosphere to verify that the emissions reduction targets are met.

Because atmospheric methane is well-mixed and has a lifetime of about 9-12 years (Stocker et al., 2013), it is transported far from its emission source, making source attribution efforts challenging from atmospheric measurements alone. Atmospheric measurements are often assimilated into “flux inversion” models to locate the sources of the emissions (e.g., Houweling et al., 2014) but rely on model wind fields to drive transport, and tend to have spatial resolutions that do not resolve sub-regional scales. Methane measurement schemes that constrain emissions on local and regional scales are thus important to help identify the sources of the emissions and to verify inventory analyses. Regional or country-scale emissions are important to public policy as those emissions are reported ~~to the UNFCCC annually~~ annually to the United Nations Framework Convention on Climate Change (UNFCCC).

The atmospheric measurement techniques that are used to estimate methane emissions include measurements made in situ, either on the ground, from tall towers, or from aircraft. Remote sensing techniques are also used, either from space or from the ground. The spatial scale of the sensitivity to emissions differs by the measurement technique: surface in situ measurements provide information about local emissions on urban scales (e.g., McKain et al., 2015; Hopkins et al., 2016), aircraft in situ measurements can provide information about regional and synoptic-scale fluxes (e.g., Karion et al., 2016; Kort et al., 2008, 2010; Frankenberg et al., 2011). Satellite remote sensing techniques provide information useful for extracting emissions information on larger scales (regional to global) (e.g., Turner et al., 2015; Schneising et al., 2014; Alexe et al., 2015; Silva et al., 2013) (e.g., Silva et al., 2013; Schneising et al., 2014) and for large point or urban sources (e.g., Kort et al., 2012, 2014; Nassar et al., 2017). Several studies (e.g., Aydin et al., 2011; Simpson et al., 2012) have shown the importance of simultaneous measurements of co-emitted species (e.g., ~~and; and~~) (e.g.,  $C_2H_6$  and  $CH_4$ ;  $CO$  and  $CO_2$  Aydin et al., 2011) and co-located measurements (e.g., Wunch et al., 2009, 2016) showing the added analytical power of the combination of atmospheric tracer information. Ground-based remote sensing instruments have been used to estimate methane emissions on urban (e.g., Wunch et al., 2009, 2016; Hase et al., 2015) (e.g., Wunch et al., 2009; Hase et al., 2015; Wunch et al., 2016) and sub-urban (e.g., Viatte et al., 2017; Chen et al., 2016) (e.g., Chen et al., 2016; Viatte et al., 2017) scales. In Hase et al. (2015), Viatte et al. (2017), and Chen et al. (2016), the authors have placed mobile ground-based remote sensing instruments around a particular emitter of interest (e.g., a city, dairy, or neighbourhood) and have designed short-term campaigns to measure the difference between upwind and downwind atmospheric methane abundances. From these differences, ~~they~~ the authors have computed emission fluxes. However, there is a network of non-mobile ground-based remote sensing instruments that have been ~~taking~~ collecting long-term measurements of atmospheric greenhouse ~~gases~~ gas abundances. These instruments were not placed intentionally around an emitter of interest, but collectively, they ought to contain information about nearby emissions. To date, there have been no studies that have attempted to extract regional methane emissions information from these existing ground-based remote sensing ~~data~~ observatories.

In this paper, we will describe our methods for computing the emissions of methane using five stationary ground-based remote sensing instruments located in Europe in §2. Our results, and comparisons to the state-of-the-art inventories are shown in §3, and we summarize our results in §4.



## 2 Methods

Our study area is the region between five long-running atmospheric observatories situated in Europe. Three of the stations are in Germany: Bremen (Notholt et al., 2014), Karlsruhe (Hase et al., 2014), and Garmisch (Sussmann and Rettinger, 2014). The other two are in Poland (Bialystok, Deutscher et al., 2014), and France (Orléans, Warneke et al., 2014). Each station measures the vertical column-averaged dry-air mole fraction of carbon dioxide ( $X_{CO_2}$ ), carbon monoxide ( $X_{CO}$ ), methane ( $X_{CH_4}$ ), and ~~a variety of~~ other trace gas species. The locations are shown in Figure 1, overlaid on a night lights image from NASA to provide a sense of the population density of the area. These observatories are part of the Total Carbon Column Observing Network (~~TCCON, ?~~)[\(TCCON, Wunch et al., 2011\)](#), and have been tied to the World Meteorological Organization trace-gas scale through comparisons with vertically integrated, ~~coincident aircraft and AirCore measurements calibrated in situ profiles over the observatories~~ (Wunch et al., 2010; Messerschmidt et al., 2011; Geibel et al., 2012; Wunch et al., 2015).

Following a similar method to ~~Wunch et al. (2009)~~[Wunch et al. \(2009, 2016\)](#), we estimate emissions of methane from the data recorded from the TCCON observatories, coupled with gridded inventories of carbon monoxide within the region. We compute changes (or “anomalies”) in  $X_{CH_4}$  and  $X_{CO}$  that we will refer to as  $\Delta X_{CH_4}$  and  $\Delta X_{CO}$ , and then compute the slopes relating  $\Delta X_{CH_4}$  to  $\Delta X_{CO}$ . From the computed slopes ( $\alpha$ ), we can infer emissions of methane ( $E_{CH_4}$ ) if emissions of carbon monoxide ( $E_{CO}$ , in mass per unit time) are known, using the following relationship:

$$E_{CH_4} = \alpha \frac{m_{CH_4}}{m_{CO}} E_{CO} \quad (1)$$

where  $\frac{m_{CH_4}}{m_{CO}}$  is the ratio of the molecular masses of  $CH_4$  and  $CO$ .

~~In the method described by Wunch et al. (2009), the authors compute anomalies from~~

~~In the Wunch et al. (2009, 2016) papers, measurements from a single atmospheric observatory were used to infer emissions, because the unique dynamics of the region advected the polluted air mass into and out of the study area diurnally. In this paper, we rely on several stations to provide measurements of the boundary of the study region to measure  $CO$  and  $CH_4$  emitted between the stations. This analysis relies on a few assumptions about the nature of the difference between afternoon and morning dry-air mole fractions at the same solar zenith angle from a single TCCON station. This method eliminates the impact of any air mass-dependent biases in the retrieved abundances. The vertical sensitivity of the TCCON measurements is explicitly taken into account by dividing the anomalies by the surface layer column averaging kernel value, as emissions. First, that the lifetimes of the gases of interest are longer than the transport time within the region. This is the case both for methane, which has an atmospheric lifetime of 12 years, and for carbon monoxide, which has an atmospheric lifetime of a few weeks. Second, we assume that the anomalies are due to emissions near the surface. Furthermore, in the Wunch et al. (2009) paper, typical emissions are consistent over time periods longer than a few days so that they are advected together. The nature of the emissions in this region (mostly residential and industrial energy needs) supports this assumption. Third, we assume that the spatial distribution of the authors use measurements of to derive the slopes and emissions (i.e., replacing with in equation 1), but in Wunch et al. (2016), they argue that emissions is similar for  $CH_4$  and  $CO$  is a better choice because its emissions inventory (provided for the region by the California Air Resources Board) is more accurate, as confirmed by the inventory maps~~

(Fig. A3). This method does not require that carbon monoxide and methane are co-emitted (as they generally do not have the same emissions sources).

~~Whereas Wunch et al. (2009) calculate anomalies from the afternoon-morning difference at a single site, in this paper, we compute anomalies between stations. First~~To compute anomalies and slopes, we first filter the data to minimize the impacts of data sparsity and air mass differences between stations (Appendix A). Then, for each station, the daily median value is subtracted from each measurement. This reduces the impact of the station altitude and any background seasonal cycle from aliasing into the results. ~~Second~~Subsequently, we compute the differences in the  $X_{\text{CH}_4}$  and  $X_{\text{CO}}$  abundances measured at the same solar zenith and solar azimuth angles on the same day at two TCCON stations. ~~This analysis is repeated for all combinations of pairs of stations within the study area. The slopes computed for each year for each pair of stations is shown in Figure 2. As in Wunch et al. (2009), this method also minimizes~~By computing anomalies at the same solar zenith angles, we minimize any impact that airmass-dependent biases could have on the calculated anomalies. ~~We also choose to compute emissions using measurements of instead of in this work because the natural fluxes in the region are large compared with the anthropogenic emissions, and they have a strong diurnal and seasonal cycle. The distance between the stations is large enough that local (sub-daily) uptake of differs from station to station, significantly impacting the anomaly slopes, especially in the summer months. While the emissions inventory of may be more accurate than the inventory in the region, This analysis is repeated for all combinations of pairs of stations within the study area. The vertical sensitivity of the presence of these large natural fluxes of precludes its use in the anomaly slope calculation.~~

~~A data filtering scheme is implemented to minimize the impacts of data sparsity and air mass differences between stations, and is described in detail in Appendix A. Computing separate slopes for each season shows little systematic variability in the anomaly slopes between seasons. The summer (June, July, August) slopes are lower than the other seasons by ~15%, but each the slopes for each season have uncertainties of ~15%.~~

~~In the Wunch et al. (2009) and Wunch et al. (2016) papers, measurements from a single atmospheric observatory were used to infer emissions, because of the diurnal onshore-offshore advection in the well-contained air mass in the South Coast Air Basin. In this paper, we rely on several stations to measure and emitted between the stations. This analysis relies on a few assumptions about the nature of the emissions. First, that typical emissions are consistent over time periods longer than a few days so that they are advected together. Second, that the spatial distribution of the emissions is similar for and, as confirmed by the inventory maps (Fig. 5). This method does not require that carbon monoxide and methane are co-emitted (as they generally do not have the same emissions sources)~~TCCON measurements is explicitly taken into account by dividing the anomalies by the surface layer column averaging kernel value, as we assume that the anomalies are due to emissions near the surface. The slopes computed for each year for each pair of stations are shown in Figure 2.

The farthest distance between the European TCCON stations included in this study is between Orléans and Białystok (1580 km). Climatological annual mean surface wind speeds from the NCEP/NCAR reanalysis (Kalnay et al., 1996) within the study area are about  $6 \text{ km} \cdot \text{h}^{-1}$  (Fig. A1). The air from Orléans will quickly mix vertically from the surface where the winds aloft are more rapid than at the surface (see Appendix B). Thus, air from Orléans would normally reach Białystok in a few days. ~~We binned the data into  $4^\circ$  solar zenith angle averages, and analysed anomalies from the same day between sites within~~

~~the same bin. We also analysed~~ To determine whether these anomalies are consistent throughout the transport time through the study area, we compute anomalies between sites ~~within the same bin, but~~ lagged by up to 14 days. The slopes of the anomalies ~~did do~~ not change significantly or systematically with the lag time (Appendix B; Fig. A2), presumably because ~~either~~ the atmospheric composition within the study area is relatively well-mixed or because the emissions are relatively consistent from day to day within the study area.

Previous papers have used carbon dioxide instead of carbon monoxide to infer methane emissions. We choose to compute emissions using measurements of  $X_{\text{CO}}$  instead of  $X_{\text{CO}_2}$  in this work because the natural  $\text{CO}_2$  fluxes in the region are large compared with the anthropogenic emissions, and they have a strong diurnal and seasonal cycle. The distance between the stations is large enough that local (sub-daily) uptake of  $\text{CO}_2$  differs from station to station, significantly obscuring the relationships between methane and carbon dioxide, and thus the anomaly slopes, especially in the summer months. While the emissions inventory of anthropogenic  $\text{CO}_2$  may be more accurate than the  $\text{CO}$  inventory in the region, the presence of these large natural fluxes of  $\text{CO}_2$  precludes its use in the anomaly slope calculation. The accuracy of our method, then, is limited by the accuracy of the carbon monoxide emission inventory. Fires could provide a large flux of  $\text{CO}$  without a large  $\text{CH}_4$  flux, and this should also be taken into consideration in these types of analyses. In our study area, fluxes from fires are small.

## 2.1 Inventories

To obtain an estimate of carbon monoxide emissions ( $E_{\text{CO}}$ ) within the study area, we use gridded inventories, and sum the emissions within the study area to compare with our emissions inferred from the TCCON measurements (see Appendix C and Fig. A3 for details). The two inventories employed here are ~~EDGAR and TNO-MACC. The~~ the Emission Database for Global Atmospheric Research (EDGAR) and TNO-MACC\_III. The EDGAR version v4.3.1\_v2 of January 2016 annual gridded inventory (Olivier et al., 1994; EC-JRC and PBL, 2016) is available at  $0.1^\circ \times 0.1^\circ$  spatial resolution and reports global emissions from the year 2000 to ~~2010. 2010~~ (Olivier et al., 1994; EC-JRC and PBL, 2016). The TNO-MACC\_III (Kuenen et al., 2014) inventory is a Europe-specific air quality emissions inventory, available on a  $0.125^\circ \times 0.0625^\circ$  grid, and reports emissions for 2000–2011 (Kuenen et al., 2014). Both EDGAR and TNO-MACC\_III provide spatially and temporally coincident methane inventories which we use to compare with our inferred emissions. We use the EDGAR version v4.2 FT2010 and the TNO-MACC\_III methane inventories.

Using country-level emissions reported through 2015 from the European Environment Agency (EEA, 2015), we extrapolate the EDGAR and TNO-MACC gridded inventory  $\text{CO}$  emissions for the study area through 2015. This facilitates more direct comparisons with the TCCON measurements, which begin with sufficient data for our study in 2009. We extrapolate the emissions by scaling the total emissions from the countries that are intersected by the area of interest (Germany, Poland, Belgium, France, Luxembourg, Czech Republic) to the last reported year of emissions from the inventory. We then assume that the same scaling factor applies for each subsequent year. The details of the extrapolation method are in Appendix D and Figs. A4 and A5.

The time series of the reported emissions from 2000–2015 are shown in Fig. 3. The inventories and scaled country-level reported emissions for this region suggest that emissions of CO and CH<sub>4</sub> have decreased by about 40% and 20%, respectively, between 2000 and 2015.

~~The TNO-MACC\_III carbon monoxide emissions are on average 15% higher than the EDGAR v4.3.1 emissions in the study area. The total TNO-MAC\_III and EDGAR methane emissions agree to within 2% in the study area.~~

~~An earlier version of the~~ EDGAR carbon monoxide inventory was evaluated by Stavrou and Müller (2006) and Fortems-Cheiney et al. (2009), who assimilated satellite measurements of CO using the EDGAR v3.3FT2000 CO emissions inventory as the a priori. ~~Stavrou and Müller find~~ ~~Stavrou and Müller (2006) found~~ that over Europe, the a posteriori emissions increase by less than 15% when assimilating carbon monoxide from the Measurements of Pollution in the Troposphere (MOPITT) satellite instrument (Emmons et al., 2004). ~~Fortems-Cheiney et al. assimilate~~ ~~Fortems-Cheiney et al. (2009) assimilated~~ Infrared Atmospheric Sounding Interferometer (IASI) CO (Clerbaux et al., 2009) and MOPITT CO, and ~~find~~ ~~found~~ that the a posteriori emissions increase by 16% and 45%, respectively. ~~The-~~

~~The more recent~~ EDGAR v4.3.1 CO emissions in ~~the study area~~ ~~our study~~ are 24% lower than the EDGAR v3.3FT2000 CO emissions for the year 2000, so it may be that the EDGAR v4.3.1 CO emissions are significantly underestimated. However, assimilations of CO are known to be very sensitive to the chemistry described in the model: most notably the OH chemistry (Yin et al., 2015; Protonotariou et al., 2010). ~~The TNO-MACC\_III carbon monoxide emissions are on average 15% higher than the EDGAR v4.3.1 emissions in the study area~~ (Protonotariou et al., 2010; Yin et al., 2015). ~~Therefore it is difficult to determine how much of the discrepancy between versions of the model is from the inventory or the model chemistry.~~

The EDGAR methane inventory has been evaluated in several previous studies. It has been shown to overestimate regional CH<sub>4</sub> emissions (e.g., Wunch et al., 2009; Wecht et al., 2014), but to underestimate oil and gas emissions (e.g., Miller et al., 2013; Buchwitz et al., 2017). However, recent methane isotope analysis by Röckmann et al. (2016) has suggested that the EDGAR inventory overestimates fossil fuel-related emissions. The study area of interest here has little oil and gas production, except for some test sites in Poland (USEIA, 2015), no commercial shale gas industry, and few pipelines.

## 2.2 Model Experiment

~~To test whether the anomaly method described in §2 can accurately infer methane emissions, we conducted a modeling experiment using version v12.1.0 of the GEOS-Chem model (www.geos-chem.org) to simulate methane and carbon monoxide for the year 2010. The model is driven by MERRA-2 meteorology from the NASA Global Modeling and Assimilation Office. The native resolution of the meteorological fields is 0.25°x 0.3125°, with 72 vertical levels from the surface to 0.01 hPa, which we degraded to 2°x 2.5° and 47 vertical levels. We use the linear CO-only and CH<sub>4</sub>-only simulations of GEOS-Chem, with prescribed monthly mean OH fields. In the CO-only simulation, global anthropogenic emissions are from EDGAR v4.3.1, which are overwritten regionally with the following emissions: the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP), the U.S. Environmental Protection Agency National Emission Inventory for 2011 (NEI2011), the MIX inventory for Asia, the Visibility Observational (BRAVO) Study Emissions Inventory for Mexico, and the criteria air contaminants (CAC) inventory for Canada. The sources of CO from the oxidation~~

of CH<sub>4</sub> and volatile organic compounds (VOCs) are prescribed following Fisher et al. (2017). For the CH<sub>4</sub>-only simulation, the emissions are as described in Maasakkers et al. (2019). Global anthropogenic emissions are from EDGAR v4.3.2, but the U.S. emissions were replaced with those from Maasakkers et al. (2016), and emissions from wetlands are from WetCHARTs version 1.0 (Bloom et al., 2017). For both CO and CH<sub>4</sub> simulations, emissions from biomass burning are from the Quick Fire Emissions Dataset (QFED) (Darmanov and Silva, 2015). The biomass burning in the study area produces less than 2% of the total anthropogenic emissions of CO.

We used identical OH fields (from version v7-02-03 of GEOS-Chem) for the CO and CH<sub>4</sub> simulations so that the chemical losses of methane and carbon monoxide are consistent, and ran tagged CO experiments so that we could identify the source of the emissions. The model atmospheric carbon monoxide and methane profiles were integrated to compute simulated X<sub>CO</sub> and X<sub>CH<sub>4</sub></sub>. These were interpolated to the locations of the TCCON stations in this study, and anomalies and slopes were computed following the method applied to the observed atmospheric data. We then applied Equation 1 to our anomaly slopes to compute methane emissions from the known CO emissions, accounting for only the CO emissions from anthropogenic, biomass burning, and biofuel sources. We neglect sources of CO emissions from the oxidation of CH<sub>4</sub> and VOCs because the column enhancements for those emissions are relatively spatially uniform across Europe and thus should not contribute significantly to the anomalies. The resulting annual CH<sub>4</sub> emissions agree well with the model emissions: the inferred emissions from the anomaly analysis are higher than the model emissions by less than 2% percent (Fig. 4).

While the inferred annual emissions agree well with the modeled annual emissions, the seasonal pattern of the emissions inferred from the anomaly analysis differs from that of the model. The anomaly analysis overestimates emissions in the winter and underestimates emissions in the summer. This may be due to small spatial inhomogeneities in the column enhancements from VOC (biogenic) emissions that influence the anomaly analysis most in summertime when VOC emissions are largest. Including the VOC emissions in the total carbon monoxide emissions leads us to infer annual methane emissions that are overestimated by 15%, increasing the inferred summertime emissions without significantly changing the inferred wintertime emissions.

This modeling experiment shows that for this region of Europe, where VOC and methane oxidation emissions lead to relatively spatially uniform column enhancements, and fire emissions are small, we can successfully use the anomaly method described in §2 to infer annual methane emissions.

### 3 Results and Discussion

Using To compute methane emissions, we apply equation 1 to our anomaly slopes and the inventory-reported carbon monoxide emissions in the study region (Fig. 5). If we choose the mean of the reported CO emissions from EDGAR v4.3.1 and TNO-MACC\_III, the methane emissions we compute within the study area based on the TCCON measurements are  $1.7 \pm 0.3 \text{ Tg} \cdot \text{yr}^{-1}$  in 2009, with a non-monotonic decrease to  $1.2 \pm 0.3 \text{ Tg} \cdot \text{yr}^{-1}$  in 2015 (Fig. 5). The uncertainties quoted here are from the standard errors on the data slope fitting only; we have not included uncertainties from the inventories themselves, which would shift the inferred emissions but would not change the trend. The magnitude of methane emissions

we compute from the TCCON data are, on average, about 2.3 times lower than the methane emissions reported by EDGAR, and about 2 times lower than the methane emissions reported by TNO-MACC\_III.

~~The year 2013 stands out in Fig. 5 as it appears to have lower emissions than previous and subsequent years. The emissions from both inventories show a small (~1%) up-tick in 2013, partially explaining the decrease in emissions. The temperature anomalies from the GISS Surface Temperature Analysis (Hansen et al., 2010; GISTEMP Team, 2016) indicate that 2013 was colder in winter and spring, possibly resulting in decreased heating needs and thus further reducing 2013 consumption. Our method of inferring methane emissions depends critically on the carbon monoxide inventory. The carbon monoxide emissions for 2010 in the study area from our GEOS-Chem model run, derived from EMEP emissions, were 6.4 Tg, about 35% higher than the average of the EDGAR and TNO-MACC\_III emissions for that year. This magnitude underestimate has also been suggested by Stavrou and Müller (2006) and Fortems-Cheiney et al. (2009) using independent data. Using the GEOS-Chem carbon monoxide emissions increases the methane emissions inferred by the anomaly analysis to  $2.4 \pm 0.3$  Tg in 2010. This value remains lower than the EDGAR and TNO-MACC\_III methane emissions estimates for 2010, which are 3 Tg, but by only 20%. Therefore, we find that the inventories likely overestimate methane emissions, but the accuracy of our results relies on the accuracy of the carbon monoxide inventory.~~

~~The~~

~~Although the EDGAR and TNO-MACC\_III inventory has inventories agree to within 15% larger in carbon monoxide emissions in this region than EDGAR v4.3.1 and about and 2% lower methane emissions than EDGAR since 2010. in methane emissions in the study region, they spatially distribute these emissions differently.~~ Maps of the spatial differences between the TNO-MACC\_III and EDGAR emissions are shown in Fig. 6 for carbon monoxide and Fig. 7 for methane. EDGAR estimates ~~relatively~~ larger emissions of carbon monoxide from the main cities in the study region and the surrounding areas. This is clearly visible from the difference map (Fig. 6), where cities such as Hamburg, Berlin, Prague, Wrocław, Warsaw, Munich, Paris, and Vienna appear in blue. However, the overall carbon monoxide emissions from TNO-MACC\_III in the study area are higher than EDGAR, and this comes from regions between the main cities, particularly in Poland and eastern France.

The differences between EDGAR and TNO-MACC\_III methane emissions also show that the EDGAR emissions estimates near large cities are significantly larger (Fig. 7). In contrast to the carbon monoxide spatial distribution, the TNO-MACC\_III methane emissions are generally smaller everywhere, except for discrete point sources.

~~Comparing 2010 reported country-level carbon monoxide emissions with the inventories shows reasonable agreement, which is expected since the inventories use country-level reports as input.~~ The sum of the carbon monoxide emissions within the entire countries of Germany, Poland, France, Luxembourg, Belgium, and Czech Republic differ between EDGAR and TNO-MACC\_III by 18% ~~for 2010~~, with EDGAR estimates lower than those from TNO-MACC\_III. Emissions from Germany, most of which are included in the study area, differ by only 6% between EDGAR and TNO-MACC\_III, again with EDGAR estimates lower than TNO-MACC\_III. The ~~differences between national carbon monoxide emissions reported to the Convention on Long-range Transboundary Air Pollution (LRTAP Convention, [https://www.eea.europa.eu/ds\\_resolveuid/0156b7a0ca47485593e7754c52c](https://www.eea.europa.eu/ds_resolveuid/0156b7a0ca47485593e7754c52c) to within a few percent of the TNO-MACC\_III country-level emissions (e.g., 5.5% for Germany in 2010).~~

The differences between 2010 country-level emissions estimates are larger for methane: EDGAR estimates are larger than TNO-MACC\_III estimates by 36% when summing all countries intersected by the study area, and 8% when considering only German emissions. The TNO-MACC\_III 2010 country-level emissions estimates agree to within a few percent of the United Nations Framework on Climate Change (UNFCCC, UNFCCC ([http://di.unfccc.int/time\\_series](http://di.unfccc.int/time_series)) country-level reported methane emissions for 2010, and the national emissions reported to the Convention on Long-range Transboundary Air Pollution (LRTAP Convention, https://www.eea.europa.eu/ds\_resolveuid/0156b7a0ea47485593e7754e52e24afd, EEA, 2015) carbon monoxide emissions for 2010. (e.g., 8% for Germany in 2010).

The differences between the EDGAR and TNO-MACC\_III inventories suggest that the spatial distribution of emissions is less certain than the larger-scale emissions, since the total carbon monoxide and methane emissions between the inventories agree to within 15% and 2% respectively in the study area, but these estimates can disagree by a factor of two on city scales. ~~It should be noted that Stavrakou and Müller (2006) and Fortems-Cheiney et al. (2009) suggest that the EDGAR v4.3.1 inventory could be underestimated by up to ~40% in Europe. Since our method of inferring methane emissions depends heavily on the carbon monoxide inventory, this would increase our methane emissions to  $2.4 \pm 0.4$  in 2009, decreasing to  $1.7 \pm 0.4$  in 2015, representing an overestimation of the EDGAR v4.3.1 FT2010 methane emissions by 1.25. However, extrapolation of EDGAR v3.3 2000 emissions to 2015 is not straightforward, as the dominant source of carbon monoxide in Europe has changed from on-road transport to residential combustion.~~

If we assume that the ~~larger-scale carbon monoxide emissions for the most recent EDGAR and TNO-MACC inventories are approximately correct, and that the~~ country-scale methane emissions are correctly reported in EDGAR and TNO-MACC\_III, our results indicate that the methane emissions in the region are incorrectly spatially distributed in the inventories. It could be that point or urban sources outside the study area, but within the countries intersected by the study area, emit a larger proportion of the country-level emissions ~~, or that large cities not contained within the study area are contributing more~~ than previously thought.

## 4 Conclusions

Using co-located measurements of methane and carbon monoxide from five long-running ground-based atmospheric observing stations, we have shown that in the area of Europe between Orléans, Bremen, Białystok, and Garmisch, the inventories significantly likely overestimate methane emissions, and point to a large uncertainty in the spatial distribution (i.e., the spatial disaggregation) of country-level emissions. However, the magnitude of our inferred methane emissions relies heavily on the EDGAR v4.3.1 and TNO-MACC\_III carbon monoxide inventories, and thus there is a need for rigorous validation of the carbon monoxide inventories.

This study demonstrates the potential of clusters of long term ground-based stationary monitoring of total columns of atmospheric greenhouse and tracer gases. It also shows the potential of having co-located measurements of multiple pollutants to derive better estimates of emissions. These types of observing systems can help policymakers verify that greenhouse gas emissions are reducing at a rate necessary to meet regulatory obligations. The atmospheric measurements are agnostic to the

source (and country of origin) of the methane, measuring only what is emitted into the atmosphere in a given area. Thus they can help validate and reveal inadequacies in the current inventories, and in particular, how country-wide emission reports are disaggregated on a grid. To enhance these results, simultaneous measurements of complementary atmospheric trace gases, such as ethane, acetylene, nitrous oxide, nitrogen dioxide, ammonia, and isotopes would help distinguish between sources of methane. This would provide additional, valuable information that would likely improve inventory disaggregation.

*Data availability.* TCCON data are available from the TCCON archive, hosted by the California Institute of Technology at <https://tccondata.org>. The Emission Database for Global Atmospheric Research (EDGAR) inventory is available from the European Commission, Joint Research Centre (JRC) / Netherlands Environmental Assessment Agency (PBL), <http://edgar.jrc.ec.europa.eu>. The GEOS-Chem v12.1.0 model is available from <https://doi.org/10.5281/zenodo.1553349>.

~~TCCON data were obtained from the TCCON archive, hosted by the California Institute of Technology at . The Emission Database for Global Atmospheric Research (EDGAR) inventory was obtained from the European Commission, Joint Research Centre (JRC)/Netherlands Environmental Assessment Agency (PBL), <http://edgar.jrc.ec.europa.eu>.~~

## Appendix A: Filtering

The filtering method was designed to remove days of data for which the atmospheric air mass was inconsistent between sites (e.g., a front was passing through or there were significant stratospheric incursions into the troposphere), and for years in which there were too few simultaneous measurements at a pair of TCCON stations to compute robust annually-representative anomalies.

To address the consistency of the air mass between sites, we retained days on which the retrievals of  $X_{\text{HF}}$  were between 50 ppt and 100 ppt, and deviated by less than 10 ppt of the median  $X_{\text{HF}}$  value for all sites on that day. HF is a trace gas that exists only in the stratosphere, and thus serves as a tracer of tropopause height (Saad et al., 2014; Washenfelder et al., 2003) (Washenfelder et al., 2003). Since the concentration of  $\text{CH}_4$  decreases significantly above the tropopause in the mid-latitudes, its total column dry-air mole fraction ( $X_{\text{CH}_4}$ ) is sensitive to the tropopause height. Filtering out days on which  $X_{\text{HF}}$  varies significantly between sites also ensures that the anomalies (and thus the slopes) are minimally impacted by stratospheric variability. This filter removed less than 5% of the data.

To ensure that the anomalies are representative of the full year, we require that each year has 400 coincident measurements across at least three seasons.

## Appendix B: Transport time between stations

Figure A1 shows the annual change in monthly mean climatological wind speeds from the NCEP/NCAR reanalysis (Kalnay et al., 1996). These are interpolated to surface pressure and 850 hPa pressures (~1500 m geopotential height) from model



(sigma) surfaces and cover January 1948 through March 2017. Vertical mixing into the boundary layer occurs on the time scale of a day or two (Jacob, 1999), and thus the relevant wind speed is between the surface and 850 hPa. The annual mean surface wind speed is  $6 \text{ km} \cdot \text{hr}^{-1}$ , which gives a mean transit time between Orléans and Białystok of 11 days. The annual mean 850 hPa winds are  $17 \text{ km} \cdot \text{hr}^{-1}$ , which give a shorter mean transit time between Orléans and Białystok of 4 days.

To test whether the transport time impacts the anomalies, we computed the slopes for time lags between sites of 0 – 14 days. Figure A2 shows a small change in anomaly slope as a function of the lag used to calculate the anomalies. This figure shows that the transport time between TCCON stations is of negligible importance to the slopes and lends weight to the decision to compute anomalies from data recorded at two TCCON stations on the same day.

### **Appendix C: Computing study area emissions from the inventories**

The study area emissions for 2010 are shown in Figure A3. We define the study area as the area bounded by the TCCON stations at (clockwise from the West) Orléans, Bremen, Białystok, and Garmisch, which is marked by the black lines in the figure. To compute the emissions from the study area, the grid points intersected by and contained within the solid black lines are summed for each year. The EDGAR v4.3.1\_v2 emissions inventory for CO and FT2010 inventory for CH<sub>4</sub> provide estimates for years 2000–2010. The TNO-MACC\_III inventory provides emissions estimates for both CO and CH<sub>4</sub> for years 2000–2011.

### **Appendix D: Projecting inventory emissions beyond 2010**

Using data from the European Environment Agency National Database ([European Environment Agency, 2016](#)), we extrapolate the inventory CO and CH<sub>4</sub> emissions for the study area through 2015. This is done by summing the total emissions for the five countries that are intersected by the study area (France, Belgium, Germany, Poland, Luxembourg, Czech Republic), and normalizing the emissions to the last year of the inventory (2010 for EDGAR, 2011 for TNO-MACC\_III). Figures A4 and A5 show the process for the EDGAR and TNO-MACC\_III CO and CH<sub>4</sub> emissions, respectively.

The top panel of Fig. A4 shows the reported country-level emissions for the years 1990–2015, their sum (black stars), and the sum of the inventory emissions for the years available (2000–2010 for EDGAR; 2000–2011 for TNO-MACC\_III) in squares. The second and third panels show the ratio of the country-level emissions to the area emissions, normalized to 1 for the last year available in the inventory. These panels show that the ratio of the summed country total emissions to the emissions from the area of interest is less variable from year to year than the emissions reported for individual countries. Thus, we choose to extrapolate the area emissions using the country total emissions, scaled to the last year of the inventory for the study area.

The bottom panel shows the results of using a single scaling factor to estimate the study area emissions from the country-level emissions for each year. We use the summed study area emissions for the years available, and the extrapolated emissions through 2015 for subsequent analysis (e.g., Figs. 3 and 5).

These boxplots show the NCEP/NCAR reanalysis long-term climatological monthly mean wind speeds at the surface (filled black boxes) and at 850 hPa (open red boxes) in the study area. The solid black and dashed red horizontal lines indicate the annual mean wind speed at the surface and 850 hPa (~1.5 km), respectively. Wind speeds aloft are significantly swifter than those at the surface.

These are the anomaly slopes ( $f$ ) in for each station pair, for the entire time series. The anomalies are computed by subtracting data within the same SZA bin between two TCCON stations. The x-axis indicates the number of days separating the measurements. The legend identifiers are as follows: br – Bremen, gm – Garmisch, bi – Białystok, or – Orléans, ka – Karlsruhe.

These maps show the inventory emissions for the year 2010 in the study area (delineated by the solid straight lines) and the surrounding region. The TCCON stations are marked with black “x” symbols and labeled in black bold font. The countries intersected by, or contained within, the study area are labeled in grey. The top left panel shows the EDGAR v4.3.1 emissions inventory for carbon monoxide. The top left map shows the EDGAR FT2010 emissions inventory for methane. The lower left panel shows the TNO-MACC\_III emissions inventory for carbon monoxide. The lower right panel shows the TNO-MACC\_III emissions inventory for methane.

This four-panel plot shows the methodology for scaling the country-level reported emissions of to extrapolate the gridded inventory emissions to 2015. The top panel shows the emissions reported by the European Environment Agency (EEA) for the countries contained within the study area (Germany, France, Czech Republic, Belgium, Luxembourg, and Poland). The black stars with a joining line represent the summed total from the five countries. The EDGAR (green) and TNO-MACC\_III (orange) inventories summed within the study area are plotted with squares joined by solid lines. The second panel shows the ratio between the individual country totals and the EDGAR area total, normalized to produce an emission ratio of 1 in 2010. The quantity with the least interannual variability in the ratio is from the country total (black stars with line). The third panel shows the ratio between the individual country totals and the TNO-MACC\_III area total, normalized to produce an emission ratio of 1 in 2011. The quantity with the least interannual variability in the ratio is, again, from the country total. The bottom panel shows the scaled country total, normalized to produce the EDGAR emissions for 2010 and the TNO-MACC\_III emissions for 2011. This permits us to compute a sensible emission for the study area through to 2015.

This four-panel plot shows the methodology for scaling the country-level emissions of reported to the UNFCCC to extrapolate the gridded inventory emissions to 2015. The panels and symbols follow the same description as in Fig. A4.

*Competing interests.* The authors declare no competing interests.

*Acknowledgements.* Source: European Commission, Joint Research Centre (JRC)/Netherlands Environmental Assessment Agency (PBL): Emission Database for Global Atmospheric Research (EDGAR) , [data were obtained from http://edgar.jrc.ec.europa.eu](http://edgar.jrc.ec.europa.eu) , [TCCON data were downloaded from the CaltechDATA archive: https://tcondata.org](https://tcondata.org) . The NASA Earth Observatory images were prepared by Joshua Stevens, using Suomi NPP VIIRS data from Miguel Román, NASA's Goddard Space Flight Center. TCCON data were obtained from the TCCON

[archive, hosted by the California Institute of Technology at https://tccondata.org](https://tccondata.org). [The GEOS-Chem v12.1.0 model was obtained from https://doi.org/10.5281/zenodo.1553349](https://doi.org/10.5281/zenodo.1553349). [The authors would like to thank two anonymous reviewers for thoughtful comments and suggestions that significantly strengthened the paper.](#)

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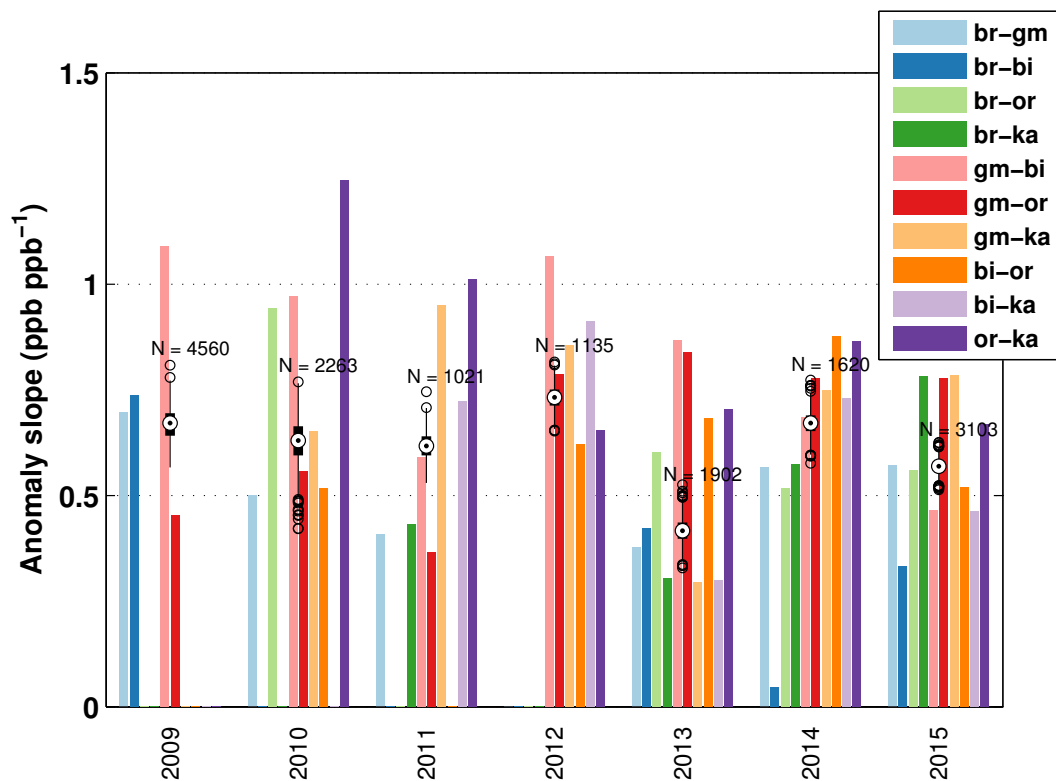
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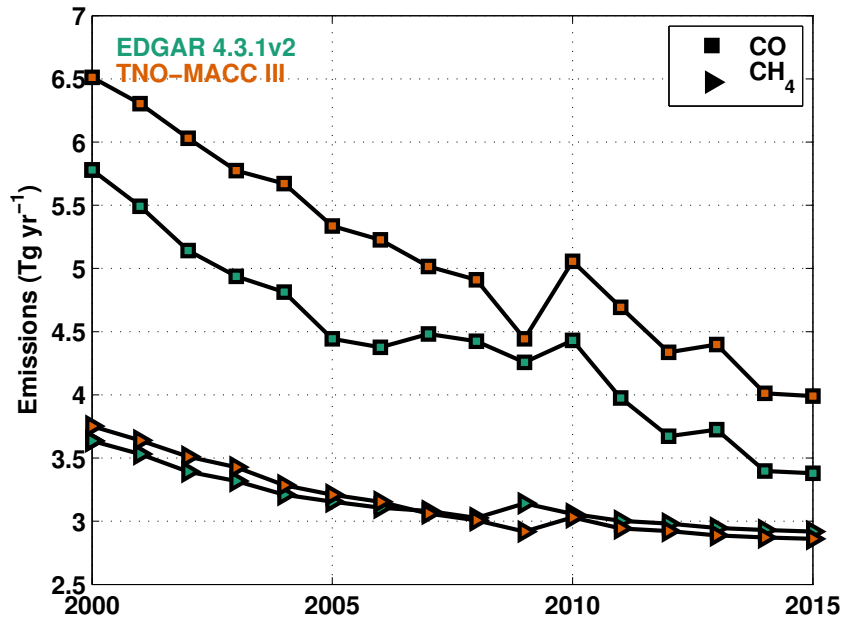
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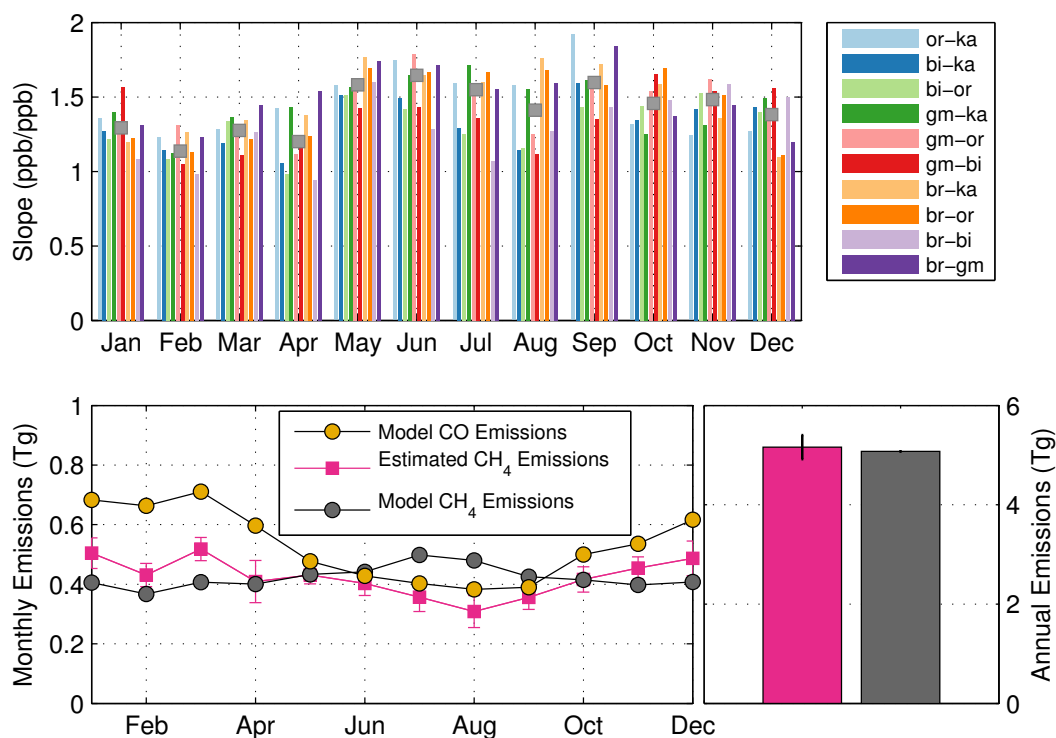
**Figure 1.** The locations of the TCCON observatories overlaid on a NASA night lights image. [NASA Earth Observatory images by Joshua Stevens, using Suomi NPP VIIRS data from Miguel Román, NASA's Goddard Space Flight Center.](#) From west to east, the stations are: Orléans (or, pink), Karlsruhe (ka, green), Bremen (br, blue-green), Garmisch (gm, orange), Białystok (bi, purple).



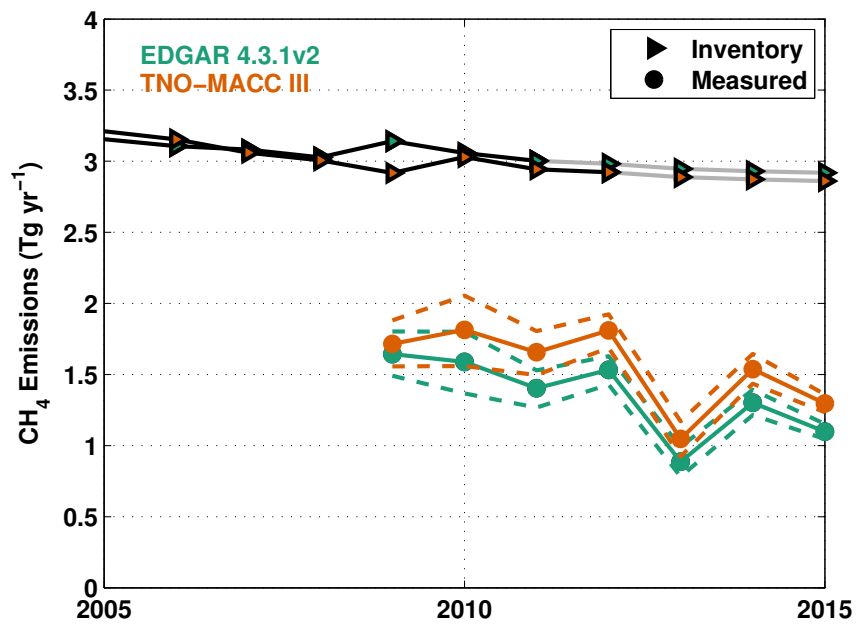
**Figure 2.** The bars show the methane to carbon monoxide anomaly slopes for each site pair. The [method of computing these anomaly slopes is detailed in §2 of the main text.](#) The black targets indicate the median value of the slope for that year, when all site pairs are considered simultaneously, and the 25<sup>th</sup> and 75<sup>th</sup> quintiles about that median value are indicated by the vertical black bars. Outliers are indicated by open black circles.



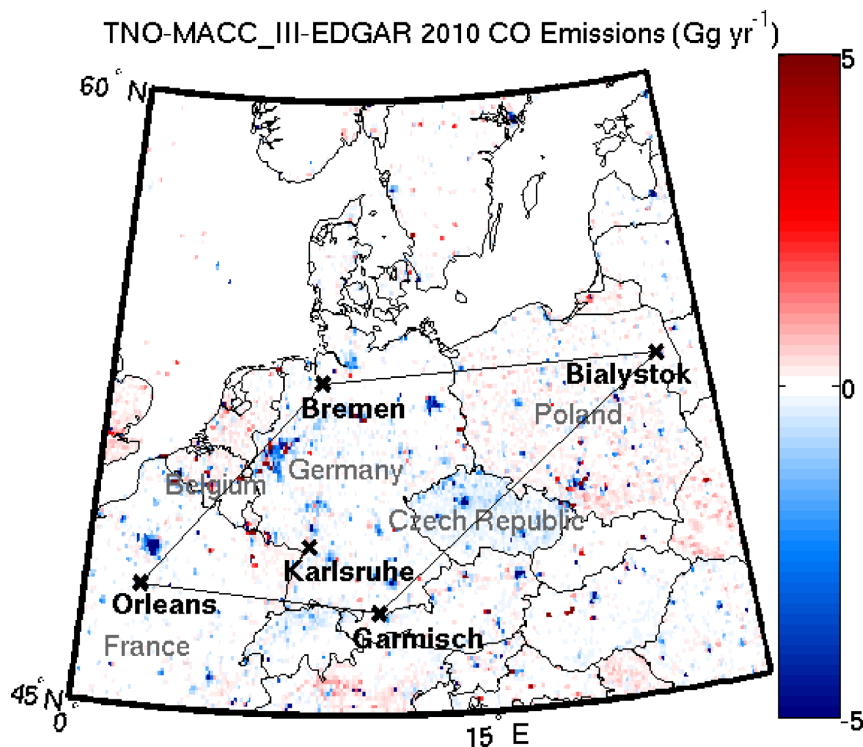
**Figure 3.** This figure shows the summed EDGAR (green) and TNO-MACC\_III (orange) emissions within the study area for CO (squares), and CH<sub>4</sub> (triangles). [The study area is defined in Figure 1.](#) All emissions are shown in units of Tg · yr<sup>-1</sup>. Extrapolation begins after 2010 for EDGAR and 2011 for TNO-MACC\_III.



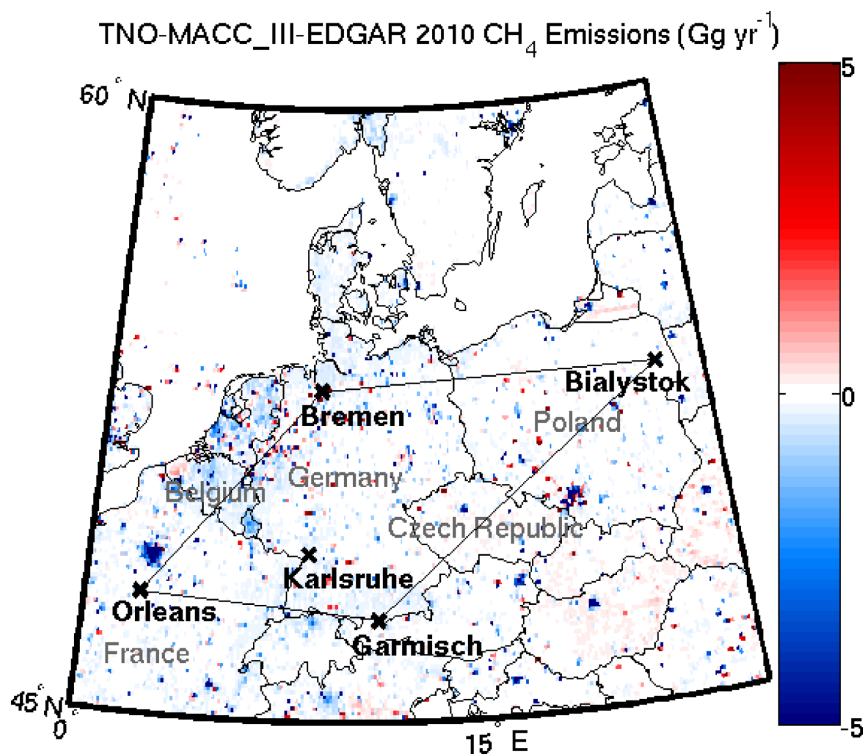
**Figure 4.** [This figure shows the results from the modeling experiment using GEOS-Chem. The top panel shows the model  \$\Delta X\_{\text{CH}\_4} - \Delta X\_{\text{CO}}\$  slopes for each month and pair of stations \(indicated by the colours\). The median slopes for each month are overlaid in grey squares. The bottom left panel shows the model carbon monoxide emissions \(excluding VOC and methane oxidation\) and the model methane emissions. The inferred methane emissions from our tracer-tracer slope method are plotted in pink squares. The bottom right panel shows the annual methane emissions from the tracer-tracer slope method and the model.](#)



**Figure 5.** The black line is the summed EDGAR (green) and TNO-MACC\_III (orange) methane emissions within the study area [shown in Figure 1](#). The grey lines indicate the projected emissions based on scaling the country-level emissions reported by the UNFCCC (UNFCCC, 2017) to the area emissions in 2010 for EDGAR and 2011 for TNO-MACC\_III. The lower solid lines show the emissions inferred from the TCCON anomaly analysis using CO emissions from the two models, and the dashed lines indicate the 5<sup>th</sup> and 95<sup>th</sup> percentiles.

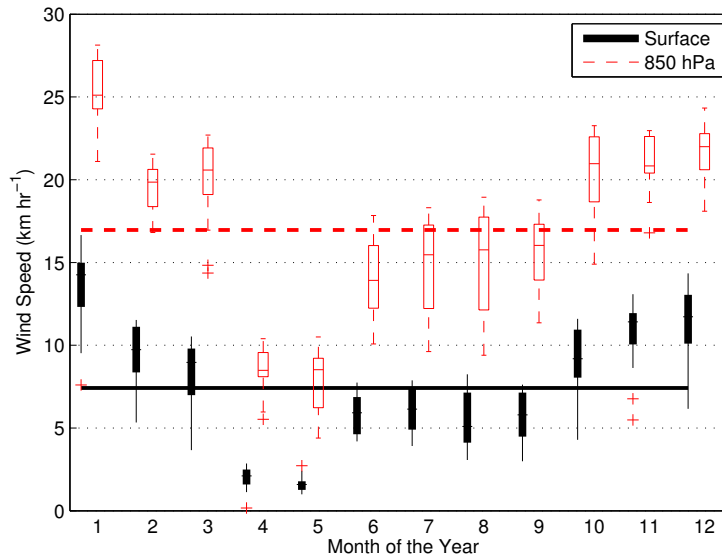


**Figure 6.** This map shows the difference between the TNO-MACC\_III carbon monoxide emissions and the EDGAR emissions for the year 2010. The black straight lines delineate the study area from the surrounding region. The TCCON stations included in this study are marked with black “x” symbols and labeled in black bold font. The countries intersected by or contained within the study area are labeled in grey. Warm (red) colours indicate that the TNO-MACC\_III inventory is larger than the EDGAR inventory; cool (blue) colours indicate that the EDGAR inventory is larger than TNO-MACC\_III.

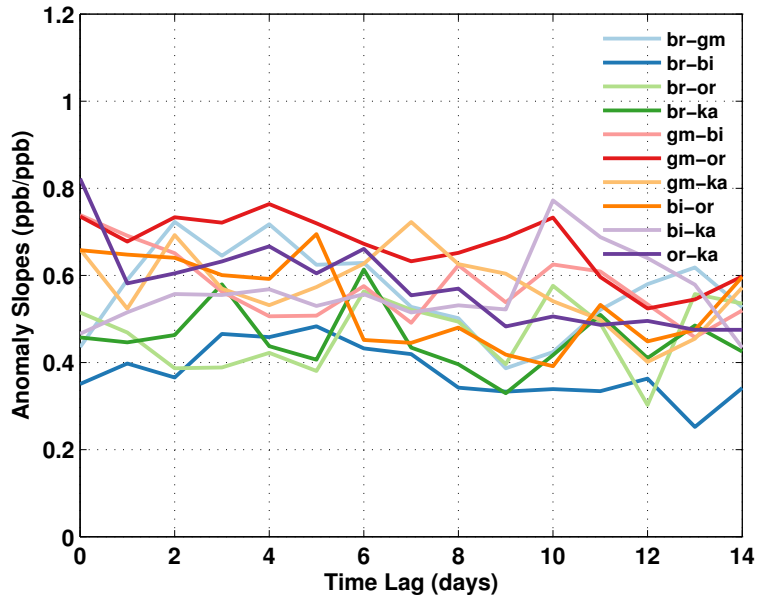


**Figure 7.** This map shows the difference between the TNO-MACC\_III methane emissions and the EDGAR emissions for the year 2010. The labeling [and colouring](#) follows that in Fig. 6.

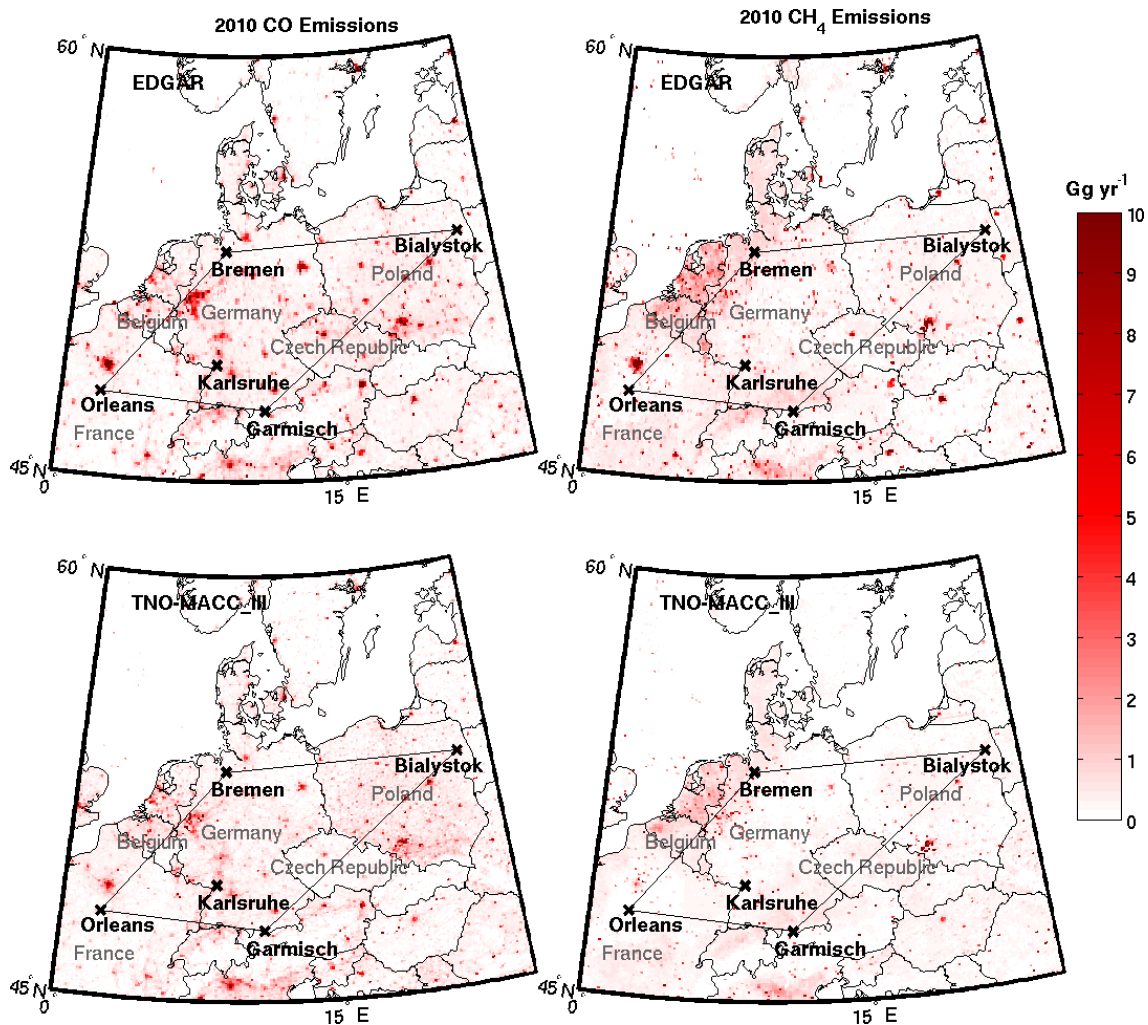




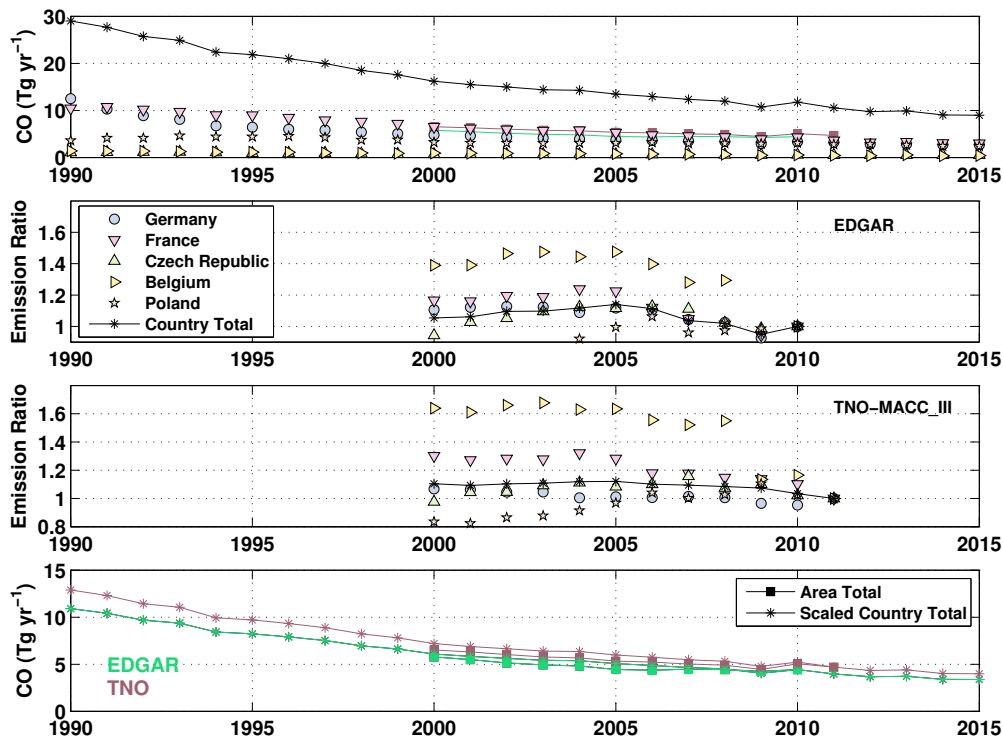
**Figure A1.** These boxplots show the NCEP/NCAR reanalysis long-term climatological monthly mean wind speeds at the surface (filled black boxes) and at 850 hPa (open red boxes) in the study area (see Figure 1, 6, or 7 for study area maps). The solid black and dashed red horizontal lines indicate the annual mean wind speed at the surface and 850 hPa (~1.5 km), respectively. Wind speeds aloft (on average  $17 \text{ km} \cdot \text{hr}^{-1}$ ) are significantly swifter than those at the surface (on average  $7.5 \text{ km} \cdot \text{hr}^{-1}$ ).



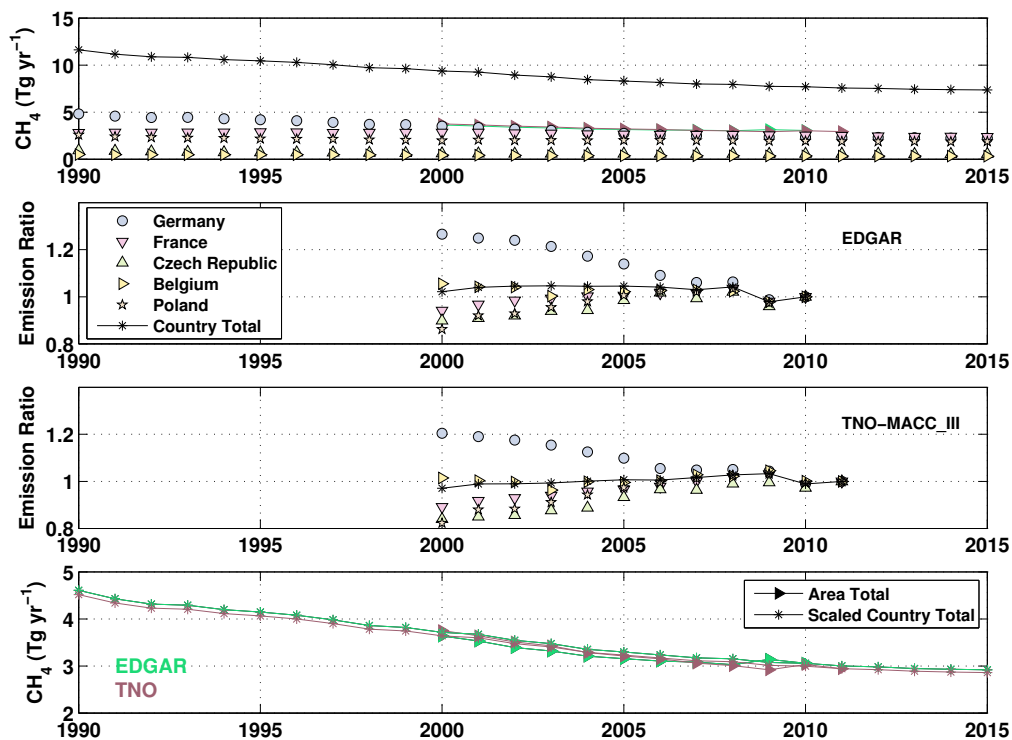
**Figure A2.** These are the anomaly slopes ( $\Delta\text{CH}_4/\Delta\text{CO}$ ) in  $\text{ppb ppb}^{-1}$  for each station pair, for the entire time series. The anomalies are computed by subtracting data within the same SZA bin between two TCCON stations. For more detail, see §2 in the main text. The x-axis indicates the number of days separating the measurements. The legend identifiers are as follows: br - Bremen, gm - Garmisch, bi - Bialystok, or - Orléans, ka - Karlsruhe.



**Figure A3.** These maps show the inventory emissions for the year 2010 in the study area (delineated by the solid straight lines) and the surrounding region. The TCCON stations are marked with black "x" symbols and labeled in black bold font. The countries intersected by, or contained within, the study area are labeled in grey. The top left map shows the EDGAR v4.3.1 emissions inventory for carbon monoxide. The top right map shows the EDGAR FT2010 emissions inventory for methane. The lower left map shows the TNO-MACC\_III emissions inventory for carbon monoxide. The lower right map shows the TNO-MACC\_III emissions inventory for methane.



**Figure A4.** This four-panel plot shows the methodology for scaling the country-level reported emissions of CO to extrapolate the gridded inventory emissions to 2015. The top panel shows the CO emissions reported by the European Environment Agency (EEA) for the countries contained within the study area (Germany, France, Czech Republic, Belgium, Luxembourg, and Poland). The black stars with a joining line represent the summed total from the five countries. The EDGAR (green) and TNO-MACC\_III (orange) inventories summed within the study area are plotted with squares joined by solid lines. The second panel shows the ratio between the individual country totals and the EDGAR area total, normalized to produce an emission ratio of 1 in 2010. The quantity with the least interannual variability in the ratio is from the country total (black stars with line). The third panel shows the ratio between the individual country totals and the TNO-MACC\_III area total, normalized to produce an emission ratio of 1 in 2011. The quantity with the least interannual variability in the ratio is, again, from the country total. The bottom panel shows the scaled country total, normalized to produce the EDGAR CO emissions for 2010 and the TNO-MACC\_III CO emissions for 2011. This permits us to compute a sensible emission for the study area through to 2015.



**Figure A5.** This four-panel plot shows the methodology for scaling the country-level emissions of CH<sub>4</sub> reported to the UNFCCC to extrapolate the gridded inventory emissions to 2015. The panels and symbols follow the same description as in Fig. A4.

# Emissions of methane in Europe inferred by total column measurements

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**Abstract.** Using five long-running ground-based atmospheric observatories in Europe, we demonstrate the utility of long-term, stationary, ground-based measurements of atmospheric total columns for verifying annual methane emission inventories. Our results indicate that the methane emissions for the region in Europe between Orléans, Bremen, Białystok, and Garmisch are overestimated by the state-of-the-art inventories Emission Database for Global Atmospheric Research (EDGAR) v4.2 FT2010 and TNO-MACC\_III, possibly due to the disaggregation of emissions onto a spatial grid. Uncertainties in the carbon monoxide inventories used to compute the methane emissions contribute to the discrepancy between our inferred emissions and those from the inventories.

## 1 Introduction

Recent global policy agreements have led to renewed efforts to reduce greenhouse gas emissions to cap global temperature rise (e.g., COP-21 (UNFCCC, 2015) and the Covenant of Mayors (European Commission, 2016)). This, in turn, has motivated countries to seek methods of reducing their greenhouse gas emissions. In Europe, methane emissions account for a significant fraction (about 11% by mass of CO<sub>2</sub> equivalent) of the total greenhouse gas emissions (UNFCCC, 2017). The lifetime of atmospheric methane is significantly shorter than for carbon dioxide, its 100-year global warming potential is significantly larger, and it is in near steady-state in the atmosphere, therefore significant reductions in methane emissions are an effective *short-term* strategy for reducing greenhouse gas emissions (Dlugokencky et al., 2011). Emission reduction strategies that include both methane emission reductions and carbon dioxide reductions are thought to be among the most effective at slowing the increase in global temperatures (Shoemaker et al., 2013). Thus, it is important to know exactly how much methane is being emitted, and the geographic and temporal source of the emissions. This requires an approach that combines state-of-the-art

emissions inventories that contain information about the specific point and area sources of the known emissions, and timely and long-term measurements of greenhouse gases in the atmosphere to verify that the emissions reduction targets are met.

Because atmospheric methane is well-mixed and has a lifetime of about 12 years (Stocker et al., 2013), it is transported far from its emission source, making source attribution efforts challenging from atmospheric measurements alone. Atmospheric measurements are often assimilated into “flux inversion” models to locate the sources of the emissions (e.g., Houweling et al., 2014) but rely on model wind fields to drive transport, and tend to have spatial resolutions that do not resolve sub-regional scales. Methane measurement schemes that constrain emissions on local and regional scales are thus important to help identify the sources of the emissions and to verify inventory analyses. Regional or country-scale emissions are important to public policy as those emissions are reported annually to the United Nations Framework Convention on Climate Change (UNFCCC).

The atmospheric measurement techniques that are used to estimate methane emissions include measurements made in situ, either on the ground, from tall towers, or from aircraft. Remote sensing techniques are also used, either from space or from the ground. The spatial scale of the sensitivity to emissions differs by the measurement technique: surface in situ measurements provide information about local emissions on urban scales (e.g., McKain et al., 2015; Hopkins et al., 2016), aircraft in situ measurements can provide information about regional and synoptic-scale fluxes (e.g., Jacob et al., 2003; Kort et al., 2008, 2010; Wofsy, 2011; Baker et al., 2012; Frankenberg et al., 2016; Karion et al., 2016). Satellite remote sensing techniques provide information useful for extracting emissions information on larger scales (regional to global) (e.g., Silva et al., 2013; Schneising et al., 2014; Alexe et al., 2015; Turner et al., 2015), and for large point or urban sources (e.g., Kort et al., 2012, 2014; Nassar et al., 2017). Several studies have shown the importance of simultaneous measurements of co-emitted species (e.g.,  $C_2H_6$  and  $CH_4$ ;  $CO$  and  $CO_2$  Aydin et al., 2011; Simpson et al., 2012; Peischl et al., 2013; Silva et al., 2013; Hausmann et al., 2016; Wunch et al., 2016; Jeong et al., 2017) or co-located measurements (e.g., Wunch et al., 2009, 2016) showing the added analytical power of the combination of atmospheric tracer information. Ground-based remote sensing instruments have been used to estimate methane emissions on urban (e.g., Wunch et al., 2009; Hase et al., 2015; Wunch et al., 2016) and sub-urban (e.g., Chen et al., 2016; Viatte et al., 2017) scales. In Hase et al. (2015), Viatte et al. (2017), and Chen et al. (2016), the authors have placed mobile ground-based remote sensing instruments around a particular emitter of interest (e.g., a city, dairy, or neighbourhood) and have designed short-term campaigns to measure the difference between upwind and downwind atmospheric methane abundances. From these differences, the authors have computed emission fluxes. However, there is a network of non-mobile ground-based remote sensing instruments that have been collecting long-term measurements of atmospheric greenhouse gas abundances. These instruments were not placed intentionally around an emitter of interest, but collectively, they ought to contain information about nearby emissions. To date, there have been no studies that have attempted to extract regional methane emissions information from these existing ground-based remote sensing observatories.

In this paper, we will describe our methods for computing the emissions of methane using five stationary ground-based remote sensing instruments located in Europe in §2. Our results, and comparisons to the state-of-the-art inventories are shown in §3, and we summarize our results in §4.

## 2 Methods

Our study area is the region between five long-running atmospheric observatories situated in Europe. Three of the stations are in Germany: Bremen (Notholt et al., 2014), Karlsruhe (Hase et al., 2014), and Garmisch (Sussmann and Rettinger, 2014). The other two are in Poland (Bialystok, Deutscher et al., 2014), and France (Orléans, Warneke et al., 2014). Each station measures the vertical column-averaged dry-air mole fraction of carbon dioxide ( $X_{CO_2}$ ), carbon monoxide ( $X_{CO}$ ), methane ( $X_{CH_4}$ ), and other trace gas species. The locations are shown in Figure 1, overlaid on a night lights image from NASA to provide a sense of the population density of the area. These observatories are part of the Total Carbon Column Observing Network (TCCON, Wunch et al., 2011), and have been tied to the World Meteorological Organization trace-gas scale through comparisons with vertically integrated, calibrated in situ profiles over the observatories (Wunch et al., 2010; Messerschmidt et al., 2011; Geibel et al., 2012; Wunch et al., 2015).

Following a similar method to Wunch et al. (2009, 2016), we estimate emissions of methane from the data recorded from the TCCON observatories, coupled with gridded inventories of carbon monoxide within the region. We compute changes (or “anomalies”) in  $X_{CH_4}$  and  $X_{CO}$  that we will refer to as  $\Delta X_{CH_4}$  and  $\Delta X_{CO}$ , and then compute the slopes relating  $\Delta X_{CH_4}$  to  $\Delta X_{CO}$ . From the computed slopes ( $\alpha$ ), we can infer emissions of methane ( $E_{CH_4}$ ) if emissions of carbon monoxide ( $E_{CO}$ , in mass per unit time) are known, using the following relationship:

$$E_{CH_4} = \alpha \frac{m_{CH_4}}{m_{CO}} E_{CO} \quad (1)$$

where  $\frac{m_{CH_4}}{m_{CO}}$  is the ratio of the molecular masses of  $CH_4$  and  $CO$ .

In the Wunch et al. (2009, 2016) papers, measurements from a single atmospheric observatory were used to infer emissions, because the unique dynamics of the region advected the polluted air mass into and out of the study area diurnally. In this paper, we rely on several stations to provide measurements of the boundary of the study region to measure  $CO$  and  $CH_4$  emitted between the stations. This analysis relies on a few assumptions about the nature of the emissions. First, that the lifetimes of the gases of interest are longer than the transport time within the region. This is the case both for methane, which has an atmospheric lifetime of 12 years, and for carbon monoxide, which has an atmospheric lifetime of a few weeks. Second, we assume that typical emissions are consistent over time periods longer than a few days so that they are advected together. The nature of the emissions in this region (mostly residential and industrial energy needs) supports this assumption. Third, we assume that the spatial distribution of the emissions is similar for  $CH_4$  and  $CO$ , as confirmed by the inventory maps (Fig. A3). This method does not require that carbon monoxide and methane are co-emitted (as they generally do not have the same emissions sources).

To compute anomalies and slopes, we first filter the data to minimize the impacts of data sparsity and air mass differences between stations (Appendix A). Then, for each station, the daily median value is subtracted from each measurement. This reduces the impact of the station altitude and any background seasonal cycle from aliasing into the results. Subsequently, we compute the differences in the  $X_{CH_4}$  and  $X_{CO}$  abundances measured at the same solar zenith and solar azimuth angles on the same day at two TCCON stations. By computing anomalies at the same solar zenith angles, we minimize any impact that air mass-dependent biases could have on the calculated anomalies. This analysis is repeated for all combinations of pairs of



stations within the study area. The vertical sensitivity of the TCCON measurements is explicitly taken into account by dividing the anomalies by the surface layer column averaging kernel value, as we assume that the anomalies are due to emissions near the surface. The slopes computed for each year for each pair of stations are shown in Figure 2.

The farthest distance between the European TCCON stations included in this study is between Orléans and Białystok (1580 km). Climatological annual mean surface wind speeds from the NCEP/NCAR reanalysis (Kalnay et al., 1996) within the study area are about  $6 \text{ km} \cdot \text{h}^{-1}$  (Fig. A1). The air from Orléans will quickly mix vertically from the surface where the winds aloft are more rapid than at the surface (see Appendix B). Thus, air from Orléans would normally reach Białystok in a few days. To determine whether these anomalies are consistent throughout the transport time through the study area, we compute anomalies between sites lagged by up to 14 days. The slopes of the anomalies do not change significantly or systematically with the lag time (Appendix B; Fig. A2), presumably because the atmospheric composition within the study area is relatively well-mixed or because the emissions are relatively consistent from day to day within the study area.

Previous papers have used carbon dioxide instead of carbon monoxide to infer methane emissions. We choose to compute emissions using measurements of  $X_{\text{CO}}$  instead of  $X_{\text{CO}_2}$  in this work because the natural  $\text{CO}_2$  fluxes in the region are large compared with the anthropogenic emissions, and they have a strong diurnal and seasonal cycle. The distance between the stations is large enough that local (sub-daily) uptake of  $\text{CO}_2$  differs from station to station, significantly obscuring the relationships between methane and carbon dioxide, and thus the anomaly slopes, especially in the summer months. While the emissions inventory of anthropogenic  $\text{CO}_2$  may be more accurate than the CO inventory in the region, the presence of these large natural fluxes of  $\text{CO}_2$  precludes its use in the anomaly slope calculation. The accuracy of our method, then, is limited by the accuracy of the carbon monoxide emission inventory. Fires could provide a large flux of CO without a large  $\text{CH}_4$  flux, and this should also be taken into consideration in these types of analyses. In our study area, fluxes from fires are small.

## 2.1 Inventories

To obtain an estimate of carbon monoxide emissions ( $E_{\text{CO}}$ ) within the study area, we use gridded inventories, and sum the emissions within the study area to compare with our emissions inferred from the TCCON measurements (see Appendix C and Fig. A3 for details). The two inventories employed here are the Emission Database for Global Atmospheric Research (EDGAR) and TNO-MACC\_III. The EDGAR version v4.3.1\_v2 of January 2016 annual gridded inventory is available at  $0.1^\circ \times 0.1^\circ$  spatial resolution and reports global emissions from the year 2000 to 2010 (Olivier et al., 1994; EC-JRC and PBL, 2016). The TNO-MACC\_III inventory is a Europe-specific air quality emissions inventory, available on a  $0.125^\circ \times 0.0625^\circ$  grid, and reports emissions for 2000–2011 (Kuenen et al., 2014). Both EDGAR and TNO-MACC\_III provide spatially and temporally coincident methane inventories which we use to compare with our inferred emissions. We use the EDGAR version v4.2 FT2010 and the TNO-MACC\_III methane inventories.

Using country-level emissions reported through 2015 from the European Environment Agency (EEA, 2015), we extrapolate the EDGAR and TNO-MACC gridded inventory CO emissions for the study area through 2015. This facilitates more direct comparisons with the TCCON measurements, which begin with sufficient data for our study in 2009. We extrapolate the emissions by scaling the total emissions from the countries that are intersected by the area of interest (Germany, Poland,

Belgium, France, Luxembourg, Czech Republic) to the last reported year of emissions from the inventory. We then assume that the same scaling factor applies for each subsequent year. The details of the extrapolation method are in Appendix D and Figs. A4 and A5.

The time series of the reported emissions from 2000–2015 are shown in Fig. 3. The inventories and scaled country-level reported emissions for this region suggest that emissions of CO and CH<sub>4</sub> have decreased by about 40% and 20%, respectively, between 2000 and 2015. The TNO-MACC\_III carbon monoxide emissions are on average 15% higher than the EDGAR v4.3.1 emissions in the study area. The total TNO-MACC\_III and EDGAR methane emissions agree to within 2% in the study area.

An earlier version of the EDGAR carbon monoxide inventory was evaluated by Stavrakou and Müller (2006) and Fortems-Cheiney et al. (2009), who assimilated satellite measurements of CO using the EDGAR v3.3FT2000 CO emissions inventory as the a priori. Stavrakou and Müller (2006) found that over Europe, the a posteriori emissions increase by less than 15% when assimilating carbon monoxide from the Measurements of Pollution in the Troposphere (MOPITT) satellite instrument (Emmons et al., 2004). Fortems-Cheiney et al. (2009) assimilated Infrared Atmospheric Sounding Interferometer (IASI) CO (Clerbaux et al., 2009) and MOPITT CO, and found that the a posteriori emissions increase by 16% and 45%, respectively.

The more recent EDGAR v4.3.1 CO emissions in our study are 24% lower than the EDGAR v3.3FT2000 CO emissions for the year 2000, so it may be that the EDGAR v4.3.1 CO emissions are significantly underestimated. However, assimilations of CO are known to be very sensitive to the chemistry described in the model: most notably the OH chemistry (Protonotariou et al., 2010; Yin et al., 2015). Therefore it is difficult to determine how much of the discrepancy between versions of the model is from the inventory or the model chemistry.

The EDGAR methane inventory has been evaluated in several previous studies. It has been shown to overestimate regional CH<sub>4</sub> emissions (e.g., Wunch et al., 2009; Wecht et al., 2014), but to underestimate oil and gas emissions (e.g., Miller et al., 2013; Buchwitz et al., 2017). However, recent methane isotope analysis by Röckmann et al. (2016) has suggested that the EDGAR inventory overestimates fossil fuel-related emissions. The study area of interest here has little oil and gas production, except for some test sites in Poland (USEIA, 2015), no commercial shale gas industry, and few pipelines.

## 2.2 Model Experiment

To test whether the anomaly method described in §2 can accurately infer methane emissions, we conducted a modeling experiment using version v12.1.0 of the GEOS-Chem model ([www.geos-chem.org](http://www.geos-chem.org)) to simulate methane and carbon monoxide for the year 2010. The model is driven by MERRA-2 meteorology from the NASA Global Modeling and Assimilation Office. The native resolution of the meteorological fields is 0.25°x 0.3125°, with 72 vertical levels from the surface to 0.01 hPa, which we degraded to 2°x 2.5° and 47 vertical levels. We use the linear CO-only and CH<sub>4</sub>-only simulations of GEOS-Chem, with prescribed monthly mean OH fields. In the CO-only simulation, global anthropogenic emissions are from EDGAR v4.3.1, which are overwritten regionally with the following emissions: the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP), the U.S. Environmental Protection Agency National Emission Inventory for 2011 (NEI2011), the MIX inventory for Asia, the Visibility Observational (BRAVO) Study Emissions Inventory for Mexico, and the criteria air contaminants (CAC) inventory for Canada. The sources of CO from the oxidation

of CH<sub>4</sub> and volatile organic compounds (VOCs) are prescribed following Fisher et al. (2017). For the CH<sub>4</sub>-only simulation, the emissions are as described in Maasakkers et al. (2019). Global anthropogenic emissions are from EDGAR v4.3.2, but the U.S. emissions were replaced with those from Maasakkers et al. (2016), and emissions from wetlands are from WetCHARTs version 1.0 (Bloom et al., 2017). For both CO and CH<sub>4</sub> simulations, emissions from biomass burning are from the Quick Fire Emissions Dataset (QFED) (Darmanov and Silva, 2015). The biomass burning in the study area produces less than 2% of the total anthropogenic emissions of CO.

We used identical OH fields (from version v7-02-03 of GEOS-Chem) for the CO and CH<sub>4</sub> simulations so that the chemical losses of methane and carbon monoxide are consistent, and ran tagged CO experiments so that we could identify the source of the emissions. The model atmospheric carbon monoxide and methane profiles were integrated to compute simulated X<sub>CO</sub> and X<sub>CH<sub>4</sub></sub>. These were interpolated to the locations of the TCCON stations in this study, and anomalies and slopes were computed following the method applied to the observed atmospheric data. We then applied Equation 1 to our anomaly slopes to compute methane emissions from the known CO emissions, accounting for only the CO emissions from anthropogenic, biomass burning, and biofuel sources. We neglect sources of CO emissions from the oxidation of CH<sub>4</sub> and VOCs because the column enhancements for those emissions are relatively spatially uniform across Europe and thus should not contribute significantly to the anomalies. The resulting annual CH<sub>4</sub> emissions agree well with the model emissions: the inferred emissions from the anomaly analysis are higher than the model emissions by less than 2% percent (Fig. 4).

While the inferred annual emissions agree well with the modeled annual emissions, the seasonal pattern of the emissions inferred from the anomaly analysis differs from that of the model. The anomaly analysis overestimates emissions in the winter and underestimates emissions in the summer. This may be due to small spatial inhomogeneities in the column enhancements from VOC (biogenic) emissions that influence the anomaly analysis most in summertime when VOC emissions are largest. Including the VOC emissions in the total carbon monoxide emissions leads us to infer annual methane emissions that are overestimated by 15%, increasing the inferred summertime emissions without significantly changing the inferred wintertime emissions.

This modeling experiment shows that for this region of Europe, where VOC and methane oxidation emissions lead to relatively spatially uniform column enhancements, and fire emissions are small, we can successfully use the anomaly method described in §2 to infer annual methane emissions.

### 3 Results and Discussion

To compute methane emissions, we apply equation 1 to our anomaly slopes and the inventory-reported carbon monoxide emissions in the study region (Fig. 5). If we choose the mean of the reported CO emissions from EDGAR v4.3.1 and TNO-MACC\_III, the methane emissions we compute within the study area based on the TCCON measurements are  $1.7 \pm 0.3 \text{ Tg} \cdot \text{yr}^{-1}$  in 2009, with a non-monotonic decrease to  $1.2 \pm 0.3 \text{ Tg} \cdot \text{yr}^{-1}$  in 2015. The uncertainties quoted here are from the standard errors on the data slope fitting only; we have not included uncertainties from the inventories. The magnitude

of methane emissions we compute from the TCCON data are, on average, about 2.3 times lower than the methane emissions reported by EDGAR, and about 2 times lower than the methane emissions reported by TNO-MACC\_III.

Our method of inferring methane emissions depends critically on the carbon monoxide inventory. The carbon monoxide emissions for 2010 in the study area from our GEOS-Chem model run, derived from EMEP emissions, were 6.4 Tg, about 5 35% higher than the average of the EDGAR and TNO-MACC\_III emissions for that year. This magnitude underestimate has also been suggested by Stavrakou and Müller (2006) and Fortems-Cheiney et al. (2009) using independent data. Using the GEOS-Chem carbon monoxide emissions increases the methane emissions inferred by the anomaly analysis to  $2.4 \pm 0.3$  Tg in 2010. This value remains lower than the EDGAR and TNO-MACC\_III methane emissions estimates for 2010, which are 3 Tg, but by only 20%. Therefore, we find that the inventories likely overestimate methane emissions, but the accuracy of our 10 results relies on the accuracy of the carbon monoxide inventory.

Although the EDGAR and TNO-MACC\_III inventories agree to within 15% in carbon monoxide emissions and 2% in methane emissions in the study region, they spatially distribute these emissions differently. Maps of the spatial differences between the TNO-MACC\_III and EDGAR emissions are shown in Fig. 6 for carbon monoxide and Fig. 7 for methane. EDGAR estimates larger emissions of carbon monoxide from the main cities in the study region and the surrounding areas. This is clearly 15 visible from the difference map (Fig. 6), where cities such as Hamburg, Berlin, Prague, Wrocław, Warsaw, Munich, Paris, and Vienna appear in blue. However, the overall carbon monoxide emissions from TNO-MACC\_III in the study area are higher than EDGAR, and this comes from regions between the main cities, particularly in Poland and eastern France.

The differences between EDGAR and TNO-MACC\_III methane emissions also show that the EDGAR emissions estimates near large cities are significantly larger (Fig. 7). In contrast to the carbon monoxide spatial distribution, the TNO-MACC\_III 20 methane emissions are generally smaller everywhere, except for discrete point sources.

Comparing 2010 reported country-level carbon monoxide emissions with the inventories shows reasonable agreement, which is expected since the inventories use country-level reports as input. The sum of the carbon monoxide emissions within the entire countries of Germany, Poland, France, Luxembourg, Belgium, and Czech Republic differ between EDGAR and TNO-MACC\_III by 18%, with EDGAR estimates lower than those from TNO-MACC\_III. Emissions from Germany, most of which 25 are included in the study area, differ by only 6% between EDGAR and TNO-MACC\_III, again with EDGAR estimates lower than TNO-MACC\_III. The national carbon monoxide emissions reported to the Convention on Long-range Transboundary Air Pollution (LRTAP Convention, [https://www.eea.europa.eu/ds\\_resolveuid/0156b7a0ca47485593e7754c52c24afd](https://www.eea.europa.eu/ds_resolveuid/0156b7a0ca47485593e7754c52c24afd), EEA, 2015) agree to within a few percent of the TNO-MACC\_III country-level emissions (e.g., 5.5% for Germany in 2010).

The differences between 2010 country-level emissions estimates are larger for methane: EDGAR estimates are larger than 30 TNO-MACC\_III estimates by 36% when summing all countries intersected by the study area, and 8% when considering only German emissions. The TNO-MACC\_III country-level emissions estimates agree to within a few percent of the UNFCCC ([http://di.unfccc.int/time\\_series](http://di.unfccc.int/time_series)) country-level reported methane emissions (e.g., 8% for Germany in 2010).

The differences between the EDGAR and TNO-MACC\_III inventories suggest that the spatial distribution of emissions is less certain than the larger-scale emissions, since the total carbon monoxide and methane emissions between the inventories 35 agree to within 15% and 2% respectively in the study area, but these estimates can disagree by a factor of two on city scales.

If we assume that the country-scale methane emissions are correctly reported in EDGAR and TNO-MACC\_III, our results indicate that the methane emissions in the region are incorrectly spatially distributed in the inventories. It could be that point or urban sources outside the study area, but within the countries intersected by the study area, emit a larger proportion of the country-level emissions than previously thought.

## 5 4 Conclusions

Using co-located measurements of methane and carbon monoxide from five long-running ground-based atmospheric observing stations, we have shown that in the area of Europe between Orléans, Bremen, Białystok, and Garmisch, the inventories likely overestimate methane emissions, and point to a large uncertainty in the spatial distribution (i.e., the spatial disaggregation) of country-level emissions. However, the magnitude of our inferred methane emissions relies heavily on the EDGAR v4.3.1  
10 and TNO-MACC\_III carbon monoxide inventories, and thus there is a need for rigorous validation of the carbon monoxide inventories.

This study demonstrates the potential of clusters of long term ground-based stationary monitoring of total columns of atmospheric greenhouse and tracer gases. It also shows the potential of having co-located measurements of multiple pollutants to derive better estimates of emissions. These types of observing systems can help policymakers verify that greenhouse gas  
15 emissions are reducing at a rate necessary to meet regulatory obligations. The atmospheric measurements are agnostic to the source (and country of origin) of the methane, measuring only what is emitted into the atmosphere in a given area. Thus they can help validate and reveal inadequacies in the current inventories, and in particular, how country-wide emission reports are disaggregated on a grid. To enhance these results, simultaneous measurements of complementary atmospheric trace gases, such as ethane, acetylene, nitrous oxide, nitrogen dioxide, ammonia, and isotopes would help distinguish between sources of  
20 methane. This would provide additional, valuable information that would likely improve inventory disaggregation.

*Data availability.* TCCON data are available from the TCCON archive, hosted by the California Institute of Technology at <https://tccodata.org>. The Emission Database for Global Atmospheric Research (EDGAR) inventory is available from the European Commission, Joint Research Centre (JRC) / Netherlands Environmental Assessment Agency (PBL), <http://edgar.jrc.ec.europa.eu>. The GEOS-Chem v12.1.0 model is available from <https://doi.org/10.5281/zenodo.1553349>.

## 25 Appendix A: Filtering

The filtering method was designed to remove days of data for which the atmospheric air mass was inconsistent between sites (e.g., a front was passing through or there were significant stratospheric incursions into the troposphere), and for years in which there were too few simultaneous measurements at a pair of TCCON stations to compute robust annually-representative anomalies.

To address the consistency of the air mass between sites, we retained days on which the retrievals of  $X_{\text{HF}}$  were between 50 ppt and 100 ppt, and deviated by less than 10 ppt of the median  $X_{\text{HF}}$  value for all sites on that day. HF is a trace gas that exists only in the stratosphere, and thus serves as a tracer of tropopause height (Washenfelder et al., 2003; Saad et al., 2014). Since the concentration of  $\text{CH}_4$  decreases significantly above the tropopause in the mid-latitudes, its total column dry-air mole fraction ( $X_{\text{CH}_4}$ ) is sensitive to the tropopause height. Filtering out days on which  $X_{\text{HF}}$  varies significantly between sites also ensures that the anomalies (and thus the slopes) are minimally impacted by stratospheric variability. This filter removed less than 5% of the data.

To ensure that the anomalies are representative of the full year, we require that each year has 400 coincident measurements across at least three seasons.

## 10 Appendix B: Transport time between stations

Figure A1 shows the annual change in monthly mean climatological wind speeds from the NCEP/NCAR reanalysis (Kalnay et al., 1996). These are interpolated to surface pressure and 850 hPa pressures (~1500 m geopotential height) from model (sigma) surfaces and cover January 1948 through March 2017. Vertical mixing into the boundary layer occurs on the time scale of a day or two (Jacob, 1999), and thus the relevant wind speed is between the surface and 850 hPa. The annual mean surface wind speed is  $6 \text{ km} \cdot \text{hr}^{-1}$ , which gives a mean transit time between Orléans and Białystok of 11 days. The annual mean 850 hPa winds are  $17 \text{ km} \cdot \text{hr}^{-1}$ , which give a shorter mean transit time between Orléans and Białystok of 4 days.

To test whether the transport time impacts the anomalies, we computed the slopes for time lags between sites of 0 – 14 days. Figure A2 shows a small change in anomaly slope as a function of the lag used to calculate the anomalies. This figure shows that the transport time between TCCON stations is of negligible importance to the slopes and lends weight to the decision to compute anomalies from data recorded at two TCCON stations on the same day.

## Appendix C: Computing study area emissions from the inventories

The study area emissions for 2010 are shown in Figure A3. We define the study area as the area bounded by the TCCON stations at (clockwise from the West) Orléans, Bremen, Białystok, and Garmisch, which is marked by the black lines in the figure. To compute the emissions from the study area, the grid points intersected by and contained within the solid black lines are summed for each year. The EDGAR v4.3.1\_v2 emissions inventory for CO and FT2010 inventory for  $\text{CH}_4$  provide estimates for years 2000–2010. The TNO-MACC\_III inventory provides emissions estimates for both CO and  $\text{CH}_4$  for years 2000–2011.

## Appendix D: Projecting inventory emissions beyond 2010

Using data from the European Environment Agency National Database (European Environment Agency, 2016), we extrapolate the inventory CO and  $\text{CH}_4$  emissions for the study area through 2015. This is done by summing the total emissions for the

five countries that are intersected by the study area (France, Belgium, Germany, Poland, Luxembourg, Czech Republic), and normalizing the emissions to the last year of the inventory (2010 for EDGAR, 2011 for TNO-MACC\_III). Figures A4 and A5 show the process for the EDGAR and TNO-MACC\_III CO and CH<sub>4</sub> emissions, respectively.

5 The top panel of Fig. A4 shows the reported country-level emissions for the years 1990–2015, their sum (black stars), and the sum of the inventory emissions for the years available (2000–2010 for EDGAR; 2000–2011 for TNO-MACC\_III) in squares. The second and third panels show the ratio of the country-level emissions to the area emissions, normalized to 1 for the last year available in the inventory. These panels show that the ratio of the summed country total emissions to the emissions from the area of interest is less variable from year to year than the emissions reported for individual countries. Thus, we choose to extrapolate the area emissions using the country total emissions, scaled to the last year of the inventory for the study area.

10 The bottom panel shows the results of using a single scaling factor to estimate the study area emissions from the country-level emissions for each year. We use the summed study area emissions for the years available, and the extrapolated emissions through 2015 for subsequent analysis (e.g., Figs. 3 and 5).

*Competing interests.* The authors declare no competing interests.

*Acknowledgements.* Emission Database for Global Atmospheric Research (EDGAR) data were obtained from <http://edgar.jrc.ec.europa.eu>.  
15 TCCON data were downloaded from the CaltechDATA archive: <https://tccodata.org>. The NASA Earth Observatory images were prepared by Joshua Stevens, using Suomi NPP VIIRS data from Miguel Román, NASA's Goddard Space Flight Center. TCCON data were obtained from the TCCON archive, hosted by the California Institute of Technology at <https://tccodata.org>. The GEOS-Chem v12.1.0 model was obtained from <https://doi.org/10.5281/zenodo.1553349>. The authors would like to thank two anonymous reviewers for thoughtful comments and suggestions that significantly strengthened the paper.

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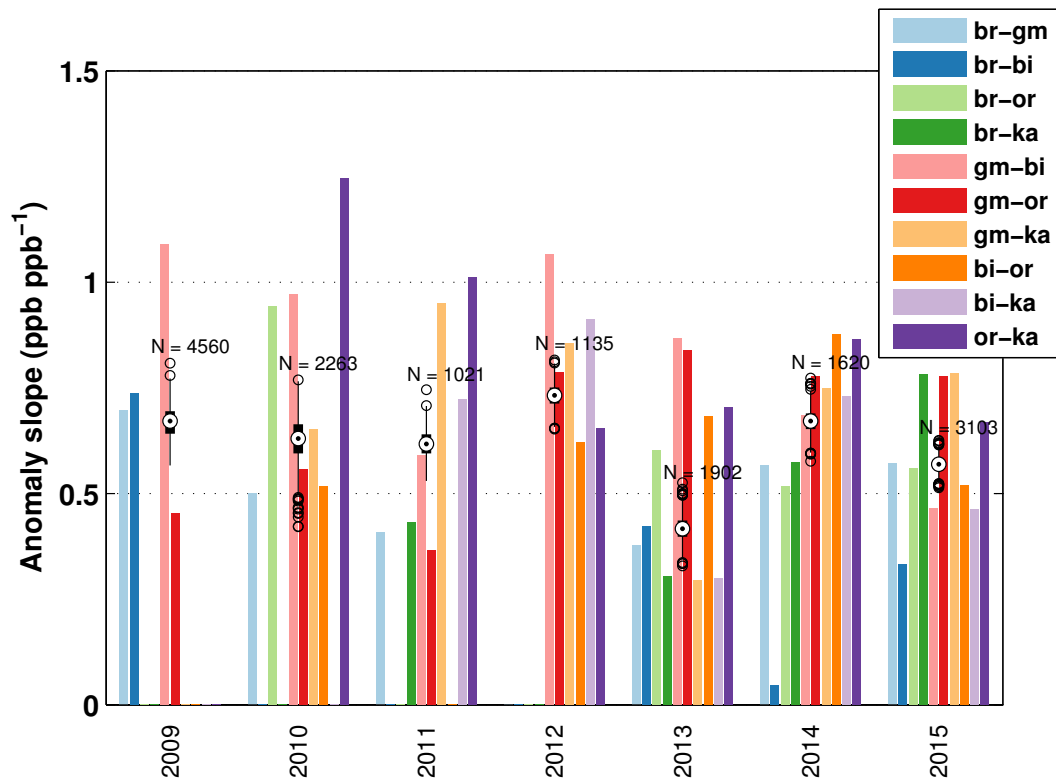
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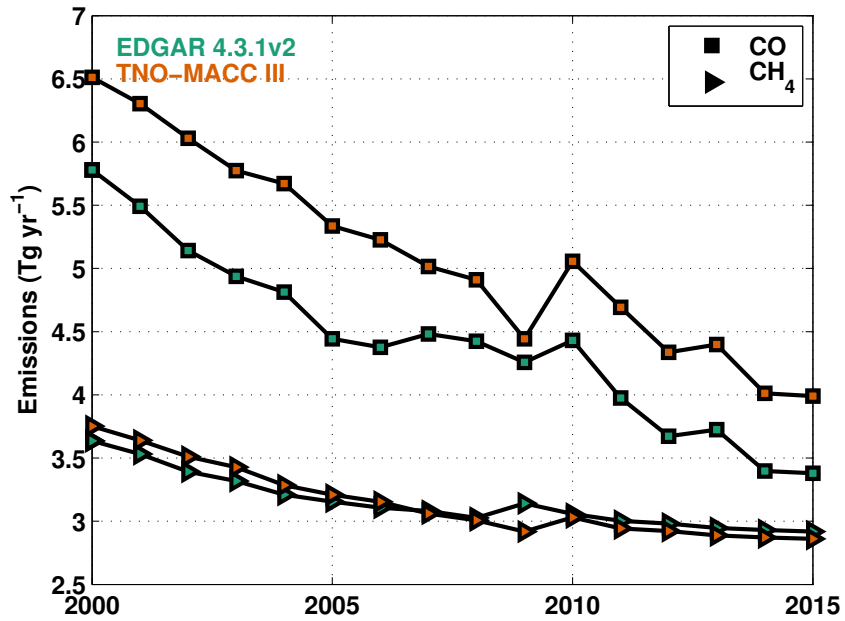
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**Figure 1.** The locations of the TCCON observatories overlaid on a NASA night lights image. From west to east, the stations are: Orléans (or, pink), Karlsruhe (ka, green), Bremen (br, blue-green), Garmisch (gm, orange), Białystok (bi, purple).

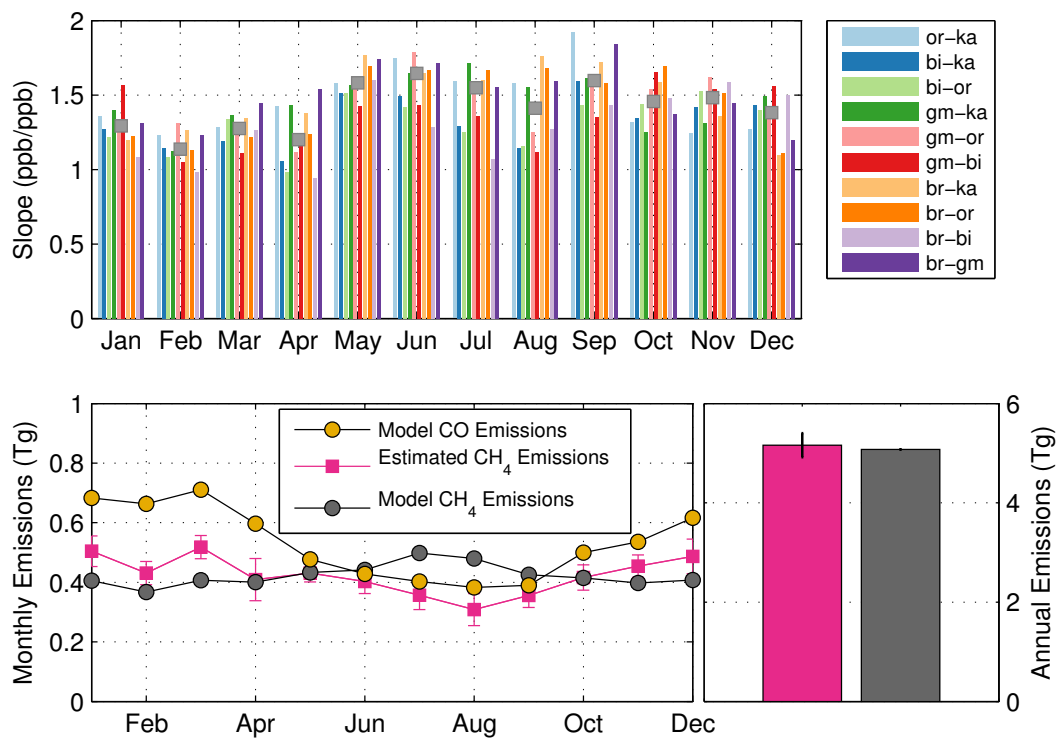


**Figure 2.** The bars show the methane to carbon monoxide anomaly slopes for each site pair. The method of computing these anomaly slopes is detailed in §2 of the main text. The black targets indicate the median value of the slope for that year, when all site pairs are considered simultaneously, and the 25<sup>th</sup> and 75<sup>th</sup> quartiles about that median value are indicated by the vertical black bars. Outliers are indicated by open black circles.

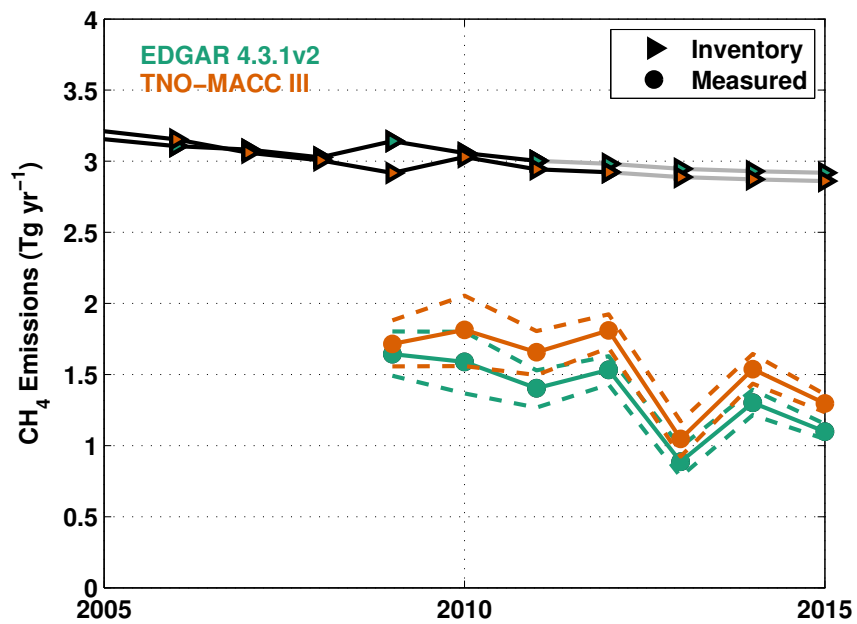


**Figure 3.** This figure shows the summed EDGAR (green) and TNO-MACC\_III (orange) emissions within the study area for CO (squares), and CH<sub>4</sub> (triangles). The study area is defined in Figure 1. All emissions are shown in units of Tg · yr<sup>-1</sup>. Extrapolation begins after 2010 for EDGAR and 2011 for TNO-MACC\_III.

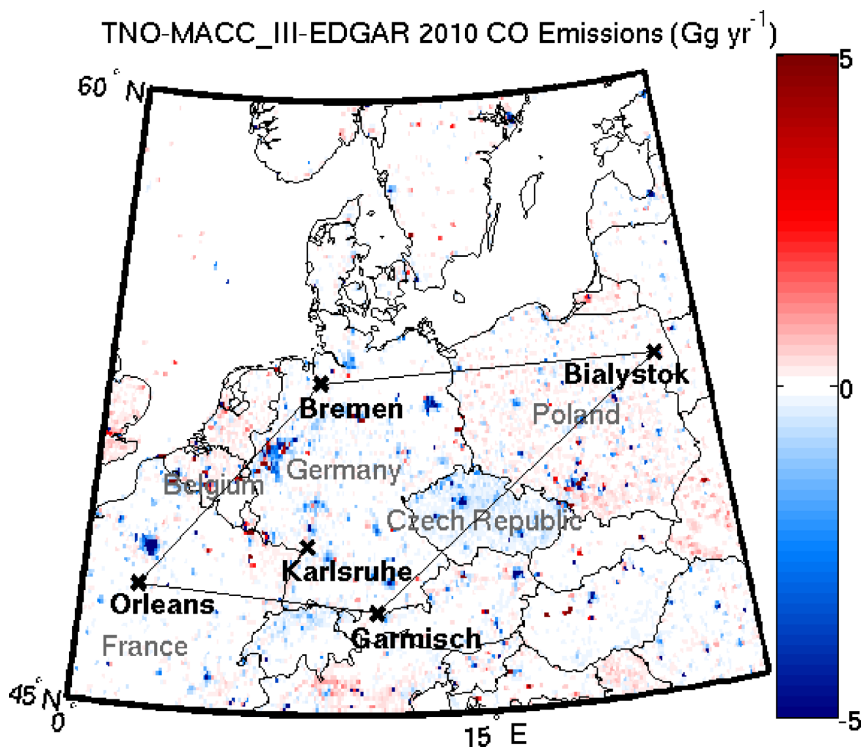




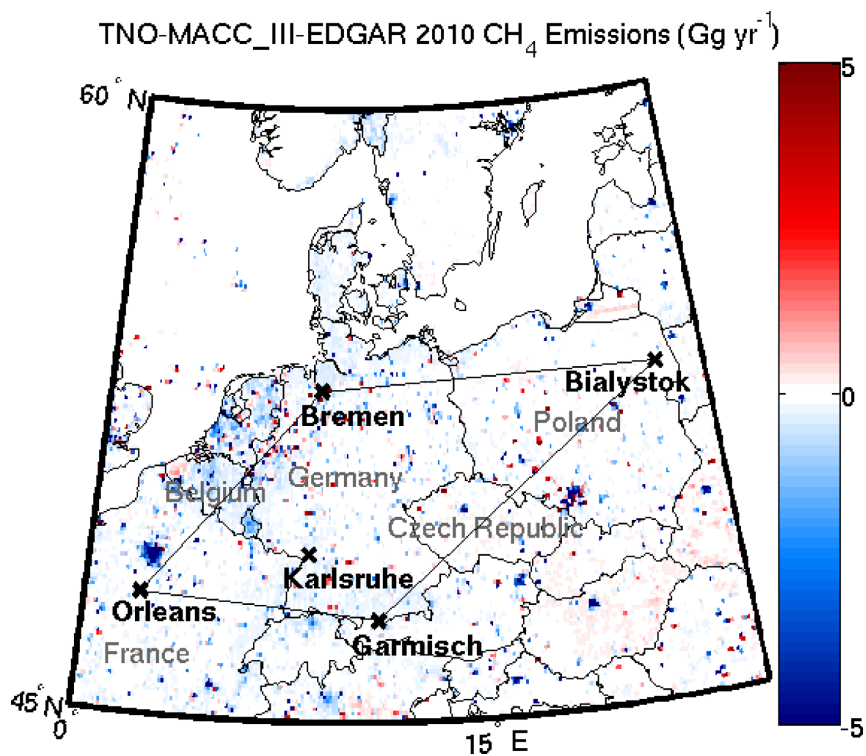
**Figure 4.** This figure shows the results from the modeling experiment using GEOS-Chem. The top panel shows the model  $\Delta X_{\text{CH}_4} - \Delta X_{\text{CO}}$  slopes for each month and pair of stations (indicated by the colours). The median slopes for each month are overlaid in grey squares. The bottom left panel shows the model carbon monoxide emissions (excluding VOC and methane oxidation) and the model methane emissions. The inferred methane emissions from our tracer-tracer slope method are plotted in pink squares. The bottom right panel shows the annual methane emissions from the tracer-tracer slope method and the model.



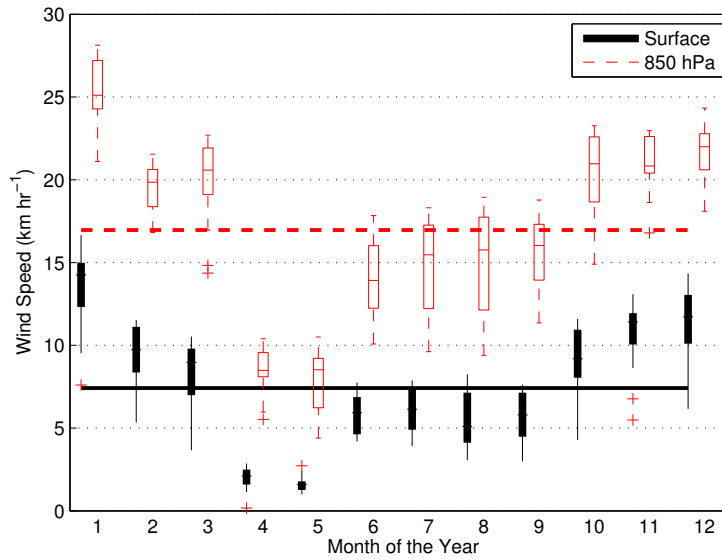
**Figure 5.** The black line is the summed EDGAR (green) and TNO-MACC\_III (orange) methane emissions within the study area shown in Figure 1. The grey lines indicate the projected emissions based on scaling the country-level emissions reported by the UNFCCC (UNFCCC, 2017) to the area emissions in 2010 for EDGAR and 2011 for TNO-MACC\_III. The lower solid lines show the emissions inferred from the TCCON anomaly analysis using CO emissions from the two models, and the dashed lines indicate the 5<sup>th</sup> and 95<sup>th</sup> percentiles.



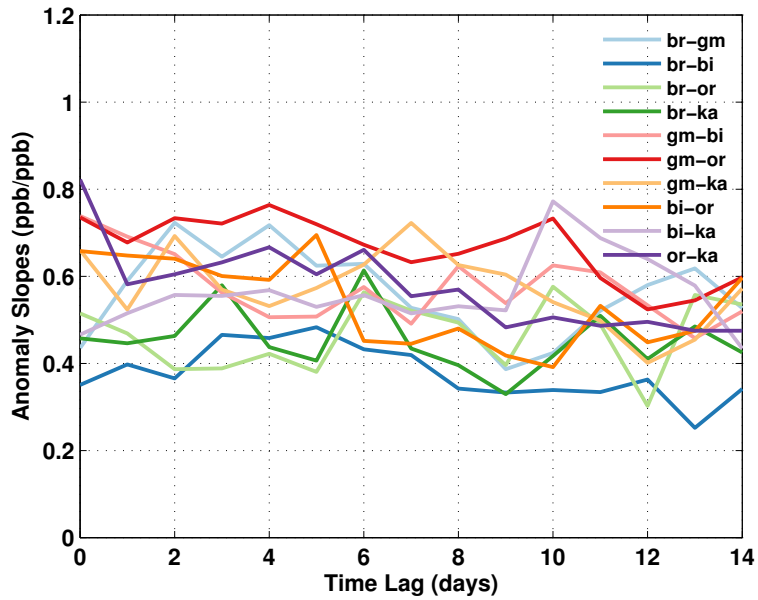
**Figure 6.** This map shows the difference between the TNO-MACC\_III carbon monoxide emissions and the EDGAR emissions for the year 2010. The black straight lines delineate the study area from the surrounding region. The TCCON stations included in this study are marked with black “x” symbols and labeled in black bold font. The countries intersected by or contained within the study area are labeled in grey. Warm (red) colours indicate that the TNO-MACC\_III inventory is larger than the EDGAR inventory; cool (blue) colours indicate that the EDGAR inventory is larger than TNO-MACC\_III.



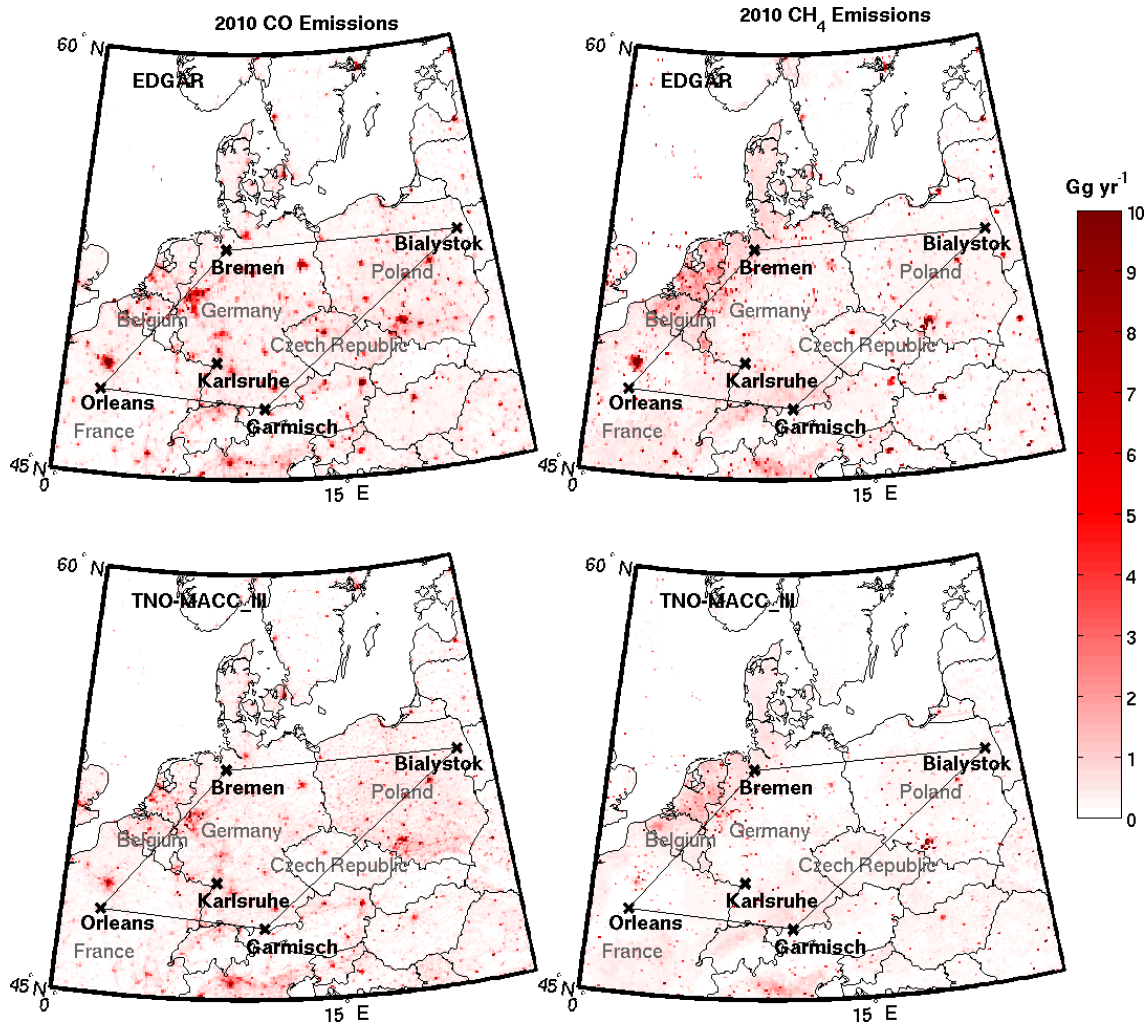
**Figure 7.** This map shows the difference between the TNO-MACC\_III methane emissions and the EDGAR emissions for the year 2010. The labeling and colouring follows that in Fig. 6.



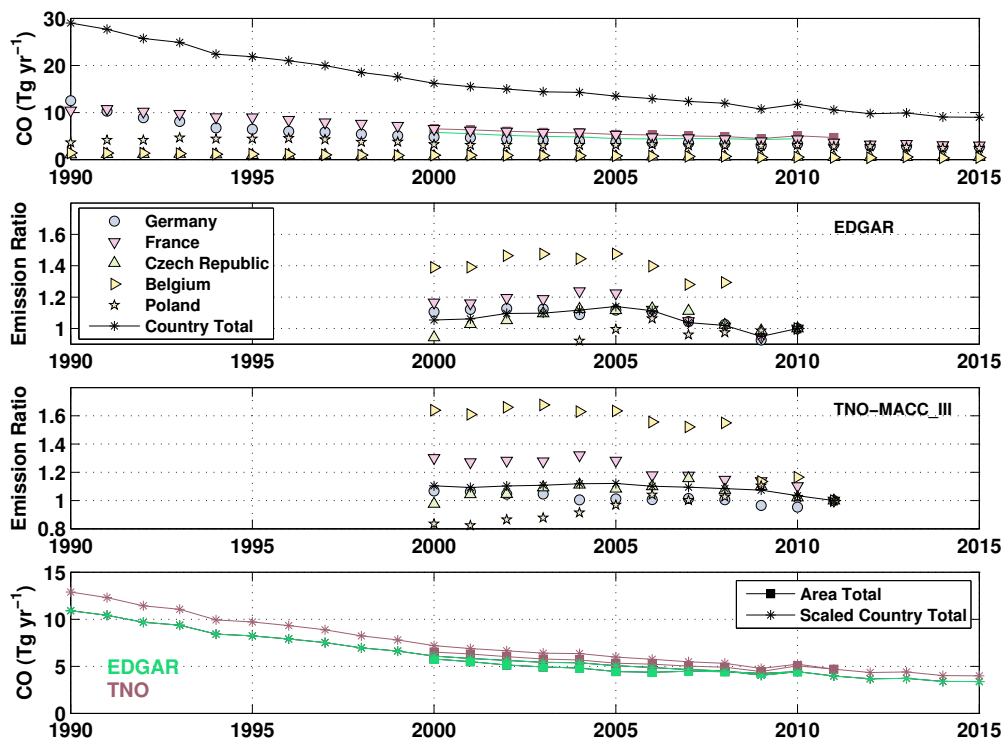
**Figure A1.** These boxplots show the NCEP/NCAR reanalysis long-term climatological monthly mean wind speeds at the surface (filled black boxes) and at 850 hPa (open red boxes) in the study area (see Figure 1, 6, or 7 for study area maps). The solid black and dashed red horizontal lines indicate the annual mean wind speed at the surface and 850 hPa ( $\sim 1.5$  km), respectively. Wind speeds aloft (on average  $17 \text{ km} \cdot \text{hr}^{-1}$ ) are significantly swifter than those at the surface (on average  $7.5 \text{ km} \cdot \text{hr}^{-1}$ ).



**Figure A2.** These are the anomaly slopes ( $\Delta\text{CH}_4/\Delta\text{CO}$ ) in  $\text{ppb ppb}^{-1}$  for each station pair, for the entire time series. The anomalies are computed by subtracting data within the same SZA bin between two TCCON stations. For more detail, see §2 in the main text. The x-axis indicates the number of days separating the measurements. The legend identifiers are as follows: br - Bremen, gm - Garmisch, bi - Białystok, or - Orléans, ka - Karlsruhe.

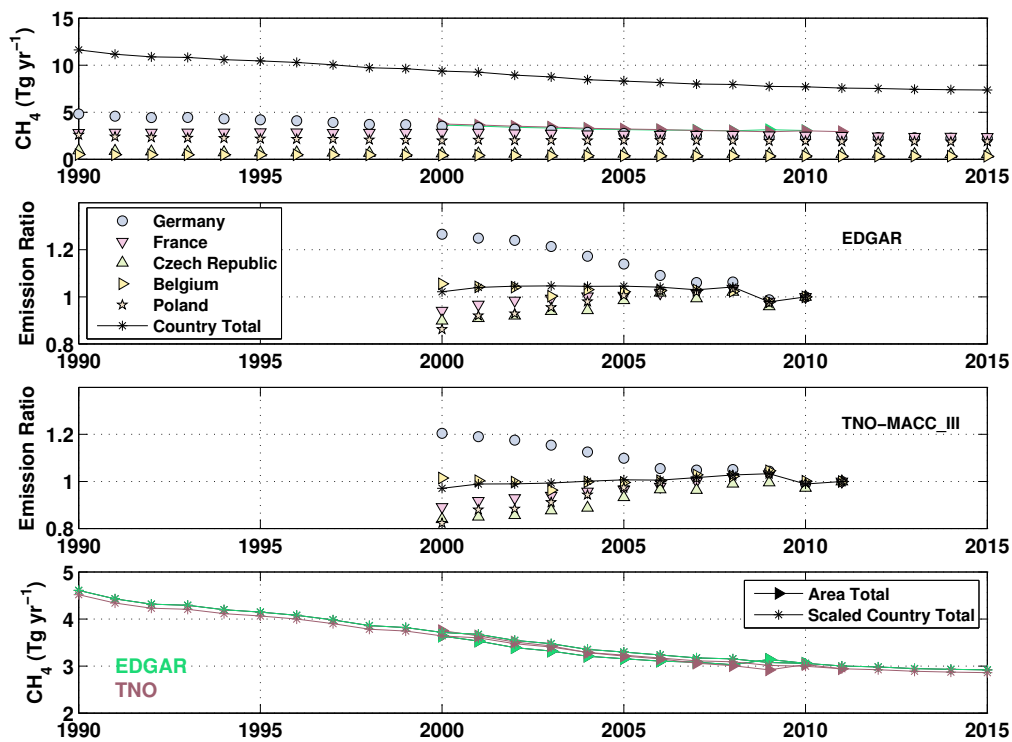


**Figure A3.** These maps show the inventory emissions for the year 2010 in the study area (delineated by the solid straight lines) and the surrounding region. The TCCON stations are marked with black “x” symbols and labeled in black bold font. The countries intersected by, or contained within, the study area are labeled in grey. The top left map shows the EDGAR v4.3.1 emissions inventory for carbon monoxide. The top right map shows the EDGAR FT2010 emissions inventory for methane. The lower left map shows the TNO-MACC\_III emissions inventory for carbon monoxide. The lower right map shows the TNO-MACC\_III emissions inventory for methane.



**Figure A4.** This four-panel plot shows the methodology for scaling the country-level reported emissions of CO to extrapolate the gridded inventory emissions to 2015. The top panel shows the CO emissions reported by the European Environment Agency (EEA) for the countries contained within the study area (Germany, France, Czech Republic, Belgium, Luxembourg, and Poland). The black stars with a joining line represent the summed total from the five countries. The EDGAR (green) and TNO-MACC\_III (orange) inventories summed within the study area are plotted with squares joined by solid lines. The second panel shows the ratio between the individual country totals and the EDGAR area total, normalized to produce an emission ratio of 1 in 2010. The quantity with the least interannual variability in the ratio is from the country total (black stars with line). The third panel shows the ratio between the individual country totals and the TNO-MACC\_III area total, normalized to produce an emission ratio of 1 in 2011. The quantity with the least interannual variability in the ratio is, again, from the country total. The bottom panel shows the scaled country total, normalized to produce the EDGAR CO emissions for 2010 and the TNO-MACC\_III CO emissions for 2011. This permits us to compute a sensible emission for the study area through to 2015.





**Figure A5.** This four-panel plot shows the methodology for scaling the country-level emissions of CH<sub>4</sub> reported to the UNFCCC to extrapolate the gridded inventory emissions to 2015. The panels and symbols follow the same description as in Fig. A4.