Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-266
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 7 May 2018

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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 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_2 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

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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 years (Stocker et al., 2013), it is transported far from its emission source, making source attribution efforts challenging. 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.

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 (e.g., 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) have shown the importance of simultaneous measurements of co-emitted species (e.g., C_2H_6 and CH_4; CO and CO_2) 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, they have computed emission fluxes. However, there is a network of non-mobile ground-based remote sensing instruments that have been taking long-term measurements of atmospheric greenhouse gases. These instruments were not placed 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.

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.

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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 (Białystok, 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, 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 (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), 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 ΔX_{CH_4} and ΔX_{CO}, and then compute the slopes relating ΔX_{CH_4} to ΔX_{CO}. From the computed slopes (α), 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:

$$E_{CH_4} = \alpha \frac{m_{CH_4}}{m_{CO}} E_{CO} \tag{1}$$

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 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 airmass-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 we assume that the anomalies are due to emissions near the surface. Furthermore, in the Wunch et al. (2009) paper, the authors use measurements of X_{CO_2} to derive the slopes and emissions (i.e., replacing CO with CO_2 in equation 1), but in Wunch et al. (2016), they argue that CO is a better choice because its emissions inventory (provided for the region by the California Air Resources Board) is more accurate.

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, 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, 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. 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 any impact that airmass-dependent biases could have on the calculated anomalies. We also choose to compute emissions using measurements of X_{CO} instead of X_{CO_2} in this work because the natural 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

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that local (sub-daily) uptake of CO_2 differs from station to station, significantly impacting the anomaly slopes, especially in the summer months. While the emissions inventory of CO_2 may be more accurate than the CO inventory in the region, the presence of these large natural fluxes of CO_2 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 CO and CH_4 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 CH_4 and CO, as confirmed by the inventory maps (Fig. 4). This method does not require that carbon monoxide and methane are co-emitted (as they generally do not have the same emissions sources).

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 $\mathrm{km \cdot h^{-1}}$ (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° solar zenith angle averages, and analysed anomalies from the same day between sites within the same bin. We also analysed anomalies between sites within the same bin, but lagged by up to 14 days. The slopes of the anomalies did 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.

To obtain an estimate of carbon monoxide emissions (E_{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 Emission Database for Global Atmospheric Research (EDGAR) version v4.3.1_v2 of January 2016 annual gridded inventory (Olivier et al., 1994; EC-JRC and PBL, 2016) is available at $0.1° \times 0.1°$ spatial resolution and reports global emissions from the year 2000 to 2010. The TNO-MACC_III (Kuenen et al., 2014) inventory is a Europe-specific air quality emissions inventory, available on a $0.125° \times 0.0625°$ grid, and reports emissions for 2000–2011. Both EDGAR and TNO-MACC_III provide spatially and temporally coincident methane inventories. 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,

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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_4 have decreased by about 40% and 20%, respectively, between 2000 and 2015.

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) find 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) assimilate Infrared Atmospheric Sounding Interferometer (IASI) CO (Clerbaux et al., 2009) and MOPITT CO, and find that the a posteriori emissions increase by 16% and 45%, respectively. The EDGAR v4.3.1 CO emissions in the study area 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). The TNO-MACC_III carbon monoxide emissions are on average 15% higher than the EDGAR v.4.3.1 emissions in the study area.

The EDGAR methane inventory has been evaluated in several previous studies. It has been shown to overestimate regional CH_4 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.

3 Results and Discussion

Using 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 $1.7 \pm 0.3\ \mathrm{Tg \cdot yr^{-1}}$ in 2009, with a non-monotonic decrease to $1.2 \pm 0.3\ \mathrm{Tg \cdot yr^{-1}}$ in 2015 (Fig. 4). The uncertainties quoted here are from the standard errors on the data 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. 4 as it appears to have lower CH_4 emissions than previous and subsequent years. The CO emissions from both inventories show a small (~1%) increase in 2013, partially explaining the decrease in CH_4 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 CH_4 consumption.

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The TNO-MACC_III inventory has 15% larger carbon monoxide emissions in this region than EDGAR v4.3.1 and about 2% lower methane emissions than EDGAR since 2010. Maps of the spatial differences between the TNO-MACC_III and EDGAR emissions are shown in Fig. 5 for carbon monoxide and Fig. 6 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. 5), 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. 6). In contrast to the carbon monoxide spatial distribution, the TNO-MACC_III methane emissions are generally smaller everywhere, except for discrete point sources.

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 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, 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/0156b7a0ca47485593e7754c52c24afd, EEA, 2015) carbon monoxide emissions for 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 CO 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 $2.4 \pm 0.4\ \mathrm{Tg \cdot yr^{-1}}$ in 2009, decreasing to $1.7 \pm 0.4\ \mathrm{Tg \cdot yr^{-1}}$ in 2015, representing an overestimation of the EDGAR v4.3.1FT2010 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, our results indicate that the methane emissions in the region are incorrectly spatially distributed in the inventories. It could be that point sources outside 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.

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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 overestimate methane emissions, and point to a large uncertainty in the spatial distribution (i.e., the 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 were obtained from the TCCON archive, hosted by the California Institute of Technology at http://tccondata.org. 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.europe.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_{HF} were between 50 ppt and 100 ppt, and deviated by less than 10 ppt of the median X_{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 CH_4 decreases significantly above the tropopause in the mid-latitudes, its total column dry-air mole fraction (X_{CH_4}) is sensitive to the tropopause height. Filtering out days on which X_{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.

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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 $6\ \mathrm{km \cdot hr^{-1}}$, which gives a mean transit time between Orléans and Białystok of 11 days. The annual mean 850 hPa winds are $17\ \mathrm{km \cdot hr^{-1}}$, 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 $\mathrm{CH_4}$ provide estimates for years 2000–2010. The TNO-MACC_III inventory provides emissions estimates for both CO and $\mathrm{CH_4}$ 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 $\mathrm{CH_4}$ 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 $\mathrm{CH_4}$ 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 4).

Competing interests. The authors declare no competing interests.

Acknowledgements. Emission Database for Global Atmospheric Research (EDGAR) data were obtained from http://edgar.jrc.ec.europe.eu. TCCON data were downloaded from the CaltechDATA archive: https://tccondata.org.

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Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-266
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 7 May 2018

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).

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-266
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 7 May 2018

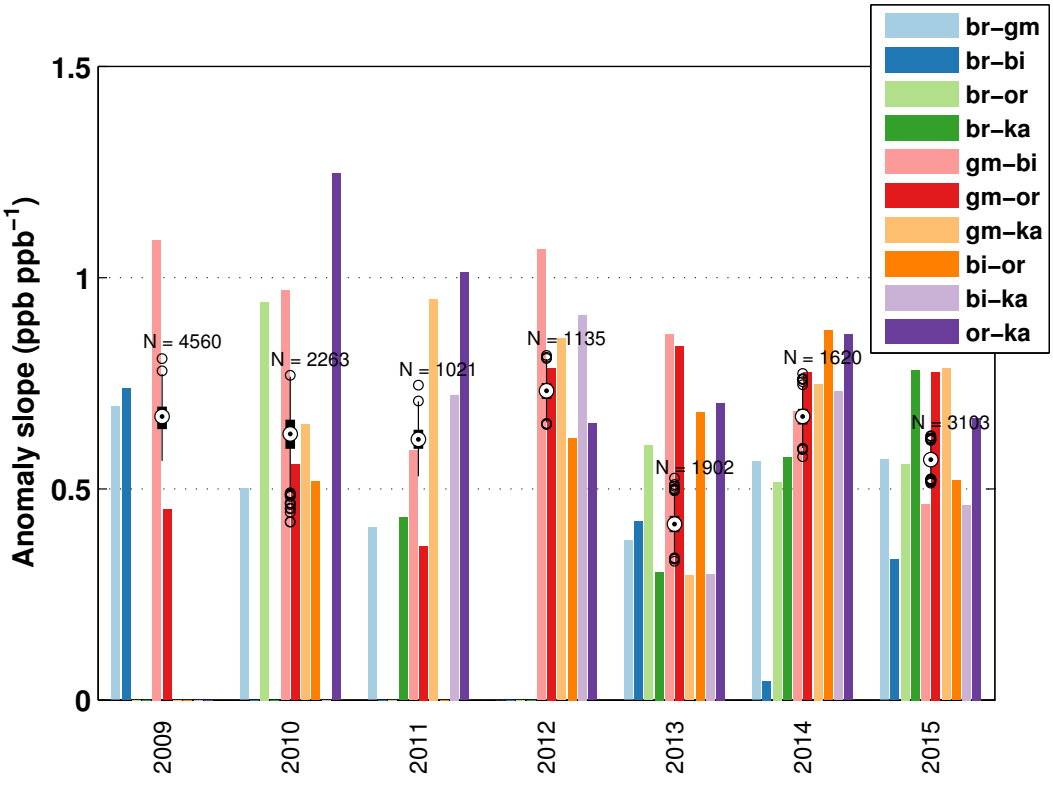

Figure 2. The bars show the methane to carbon monoxide anomaly slopes for each site pair. The method of computing these anomaly slopes is detailed in §2 of the main text. The black targets indicate the median value of the slope for that year, when all site pairs are considered simultaneously, and the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

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-266
Manuscript under review for journal Atmos. Chem. Phys.
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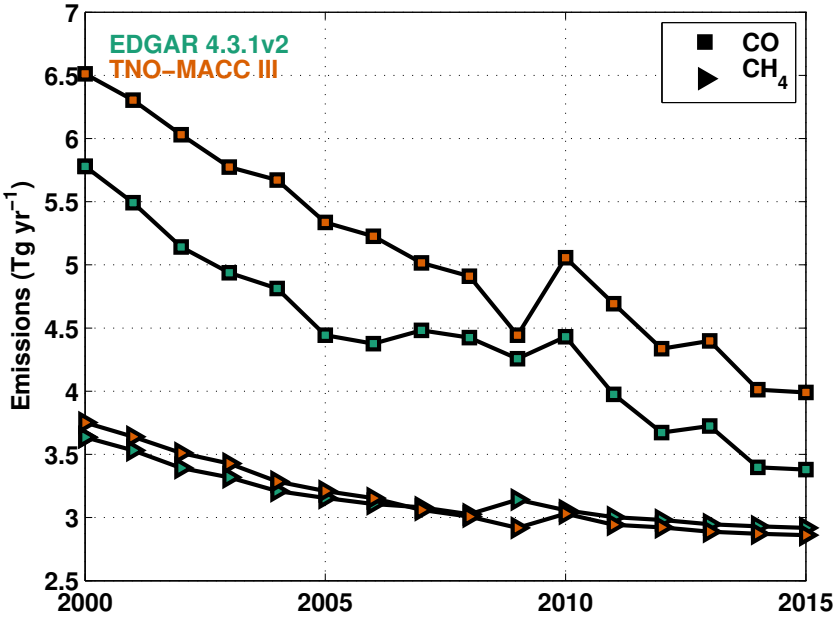

Figure 3. This figure shows the summed EDGAR (green) and TNO-MACC_III (orange) emissions within the study area for CO (squares), and CH_4 (triangles). The study area is defined in Figure 1. All emissions are shown in units of $\mathrm{Tg} \cdot \mathrm{yr}^{-1}$. Extrapolation begins after 2010 for EDGAR and 2011 for TNO-MACC_III.

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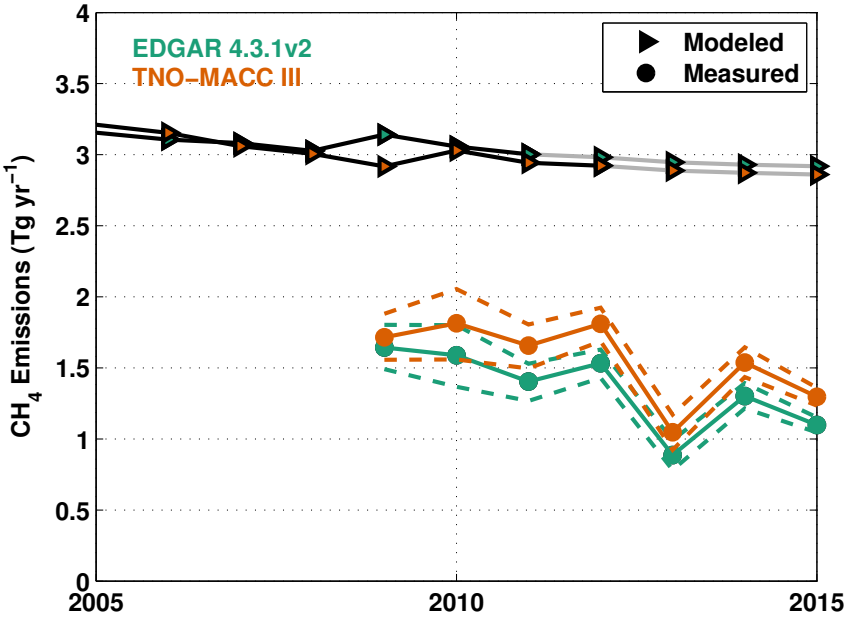

Figure 4. The black line is the summed EDGAR (green) and TNO-MACC_III (orange) methane emissions within the study area shown in Figure 1. The grey lines indicate the projected emissions based on scaling the country-level emissions reported by the UNFCCC (UNFCCC, 2017) to the area emissions in 2010 for 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^{th} and 95^{th} percentiles.

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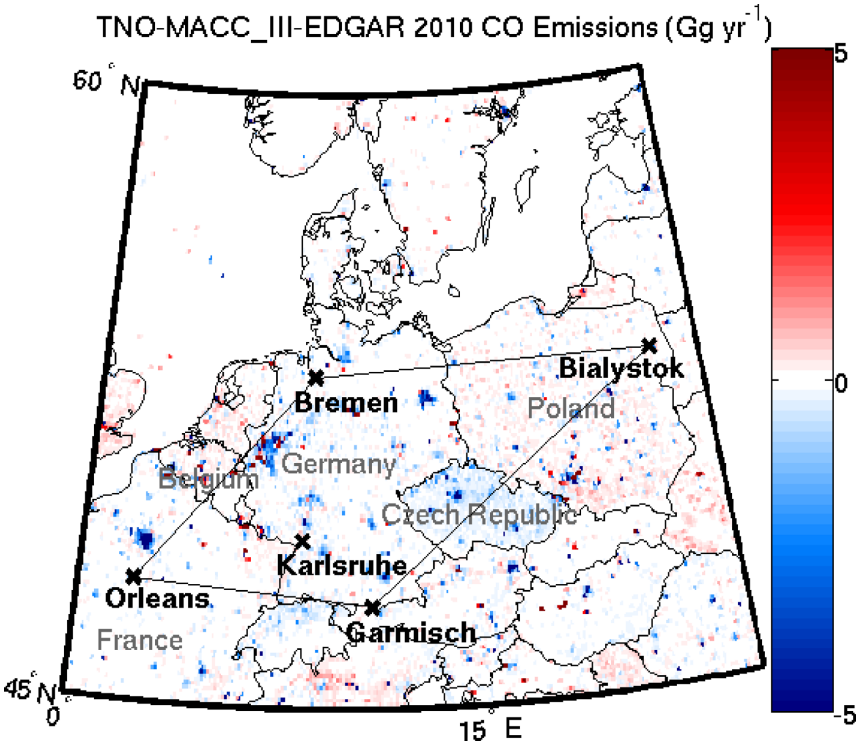

Figure 5. This map shows the difference between the TNO-MACC_III carbon monoxide emissions and the EDGAR emissions for the year 2010. The black straight lines delineate the study area from the surrounding region. The TCCON stations included in this study are marked with black "**x**" symbols and labeled 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.

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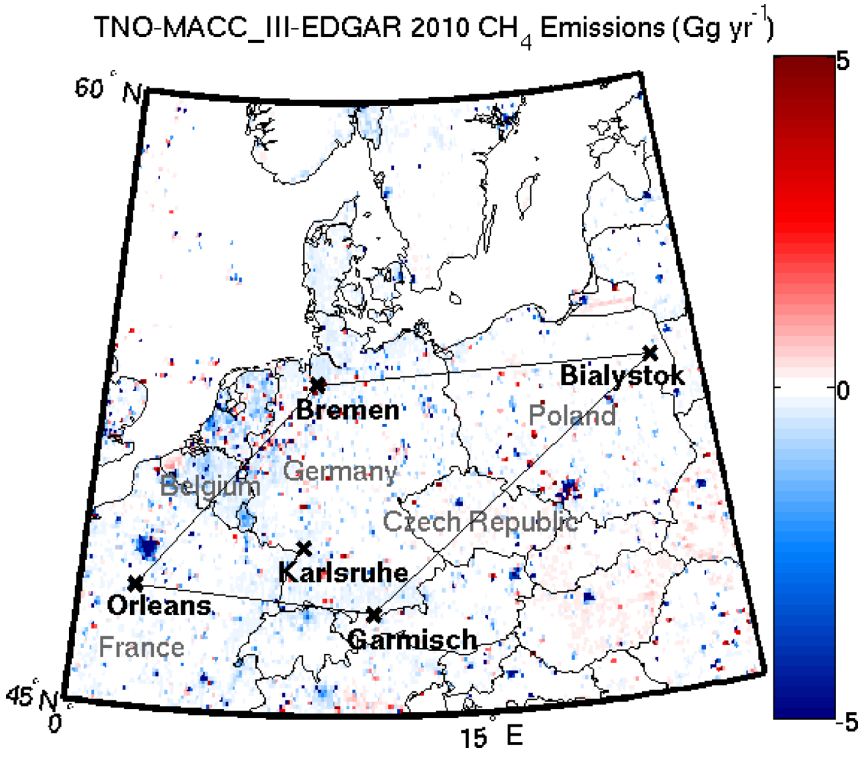

Figure 6. This map shows the difference between the TNO-MACC_III methane emissions and the EDGAR emissions for the year 2010. The labeling and colouring follows that in Fig. 5.

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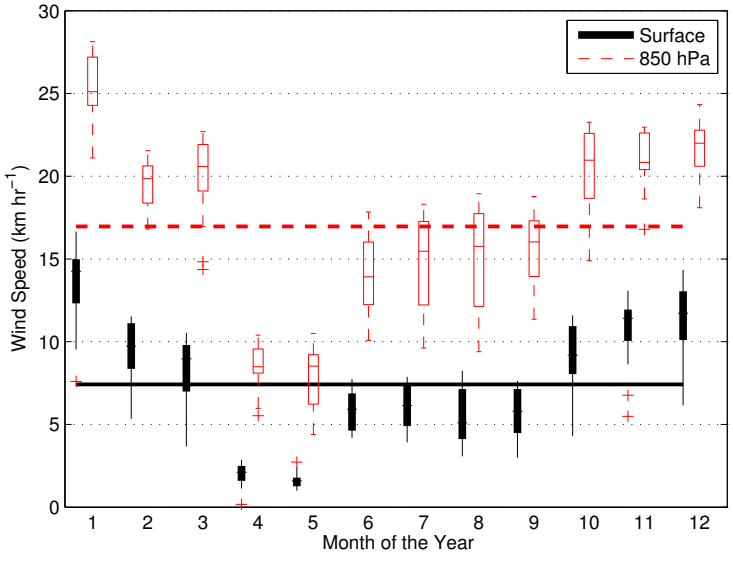

Figure A1. These boxplots show the NCEP/NCAR reanalysis long-term climatological monthly mean wind speeds at the surface (filled black boxes) and at 850 hPa (open red boxes) in the study area (see Figure 1, 5, or 6 for study area maps). The solid black and dashed red horizontal lines indicate the annual mean wind speed at the surface and 850 hPa (~1.5 km), respectively. Wind speeds aloft (on average 17 $\mathrm{km} \cdot \mathrm{hr}^{-1}$) are significantly swifter than those at the surface (on average 7.5 $\mathrm{km} \cdot \mathrm{hr}^{-1}$).

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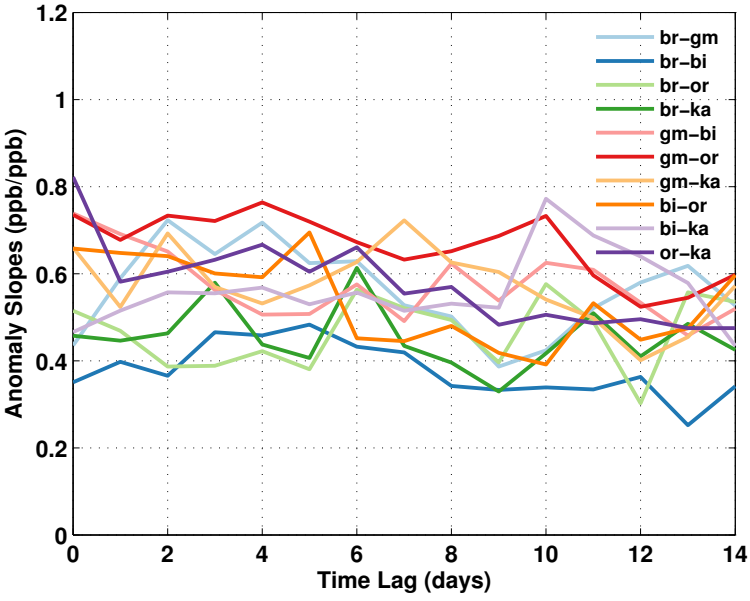

Figure A2. These are the anomaly slopes ($\Delta CH_4/\Delta CO$) in $\mathrm{ppb\ ppb^{-1}}$ for each station pair, for the entire time series. The anomalies are computed by subtracting data within the same SZA bin between two TCCON stations. For more detail, see §2 in the main text. The x-axis indicates the number of days separating the measurements. The legend identifiers are as follows: br - Bremen, gm - Garmisch, bi - Białystok, or - Orléans, ka - Karlsruhe.

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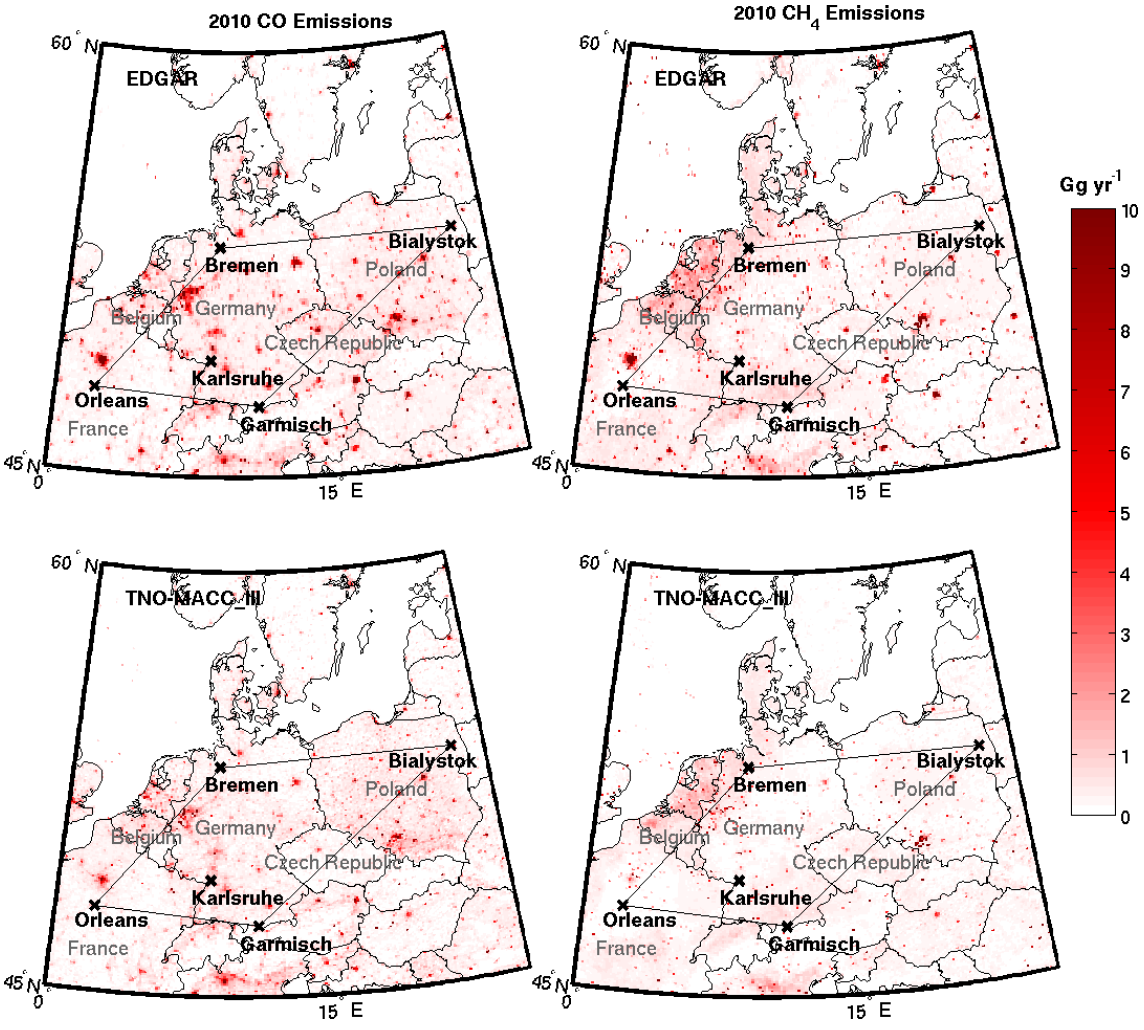

Figure A3. These maps show the inventory emissions for the year 2010 in the study area (delineated by the solid straight lines) and the surrounding region. The TCCON stations are marked with black "**x**" symbols and labeled in black bold font. The countries intersected by, or contained within, the study 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.

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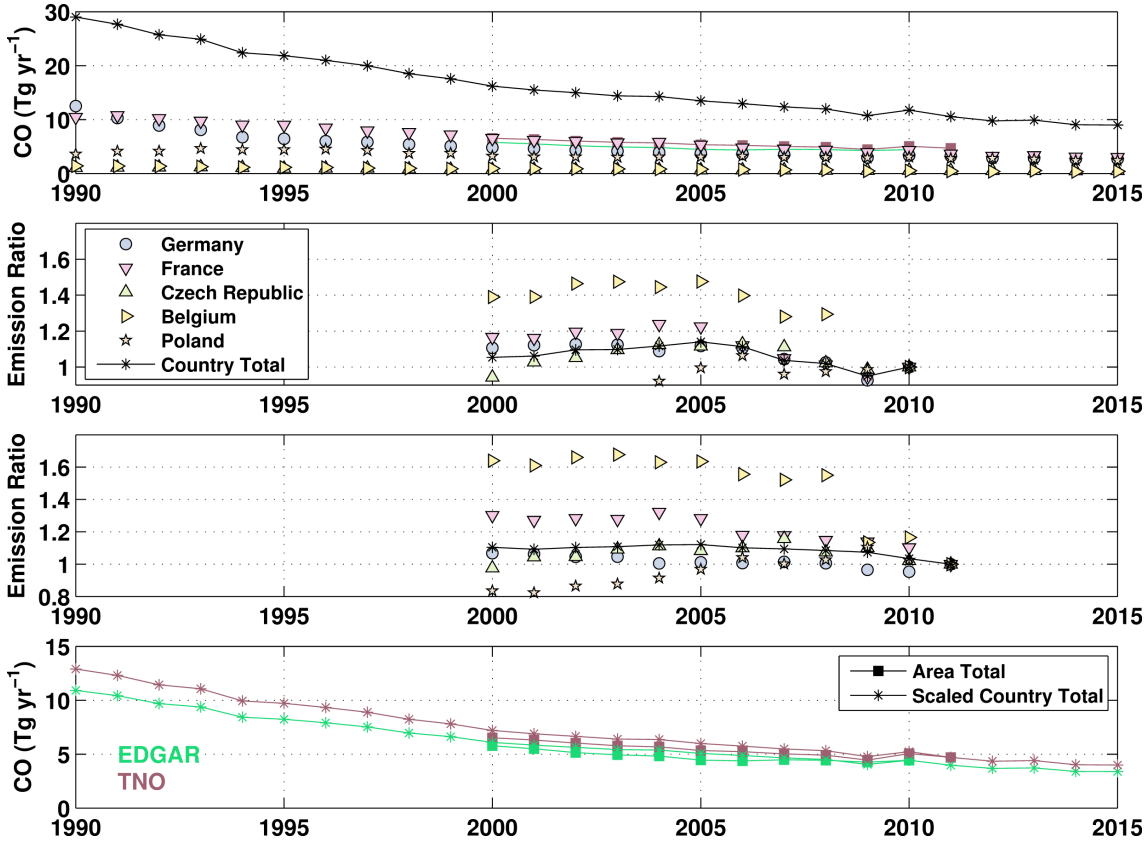

Figure A4. This four-panel plot shows the methodology for scaling the country-level reported emissions of CO to extrapolate the gridded inventory emissions to 2015. The top panel shows the CO emissions reported by the European Environment Agency (EEA) for the countries contained within the study area (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.

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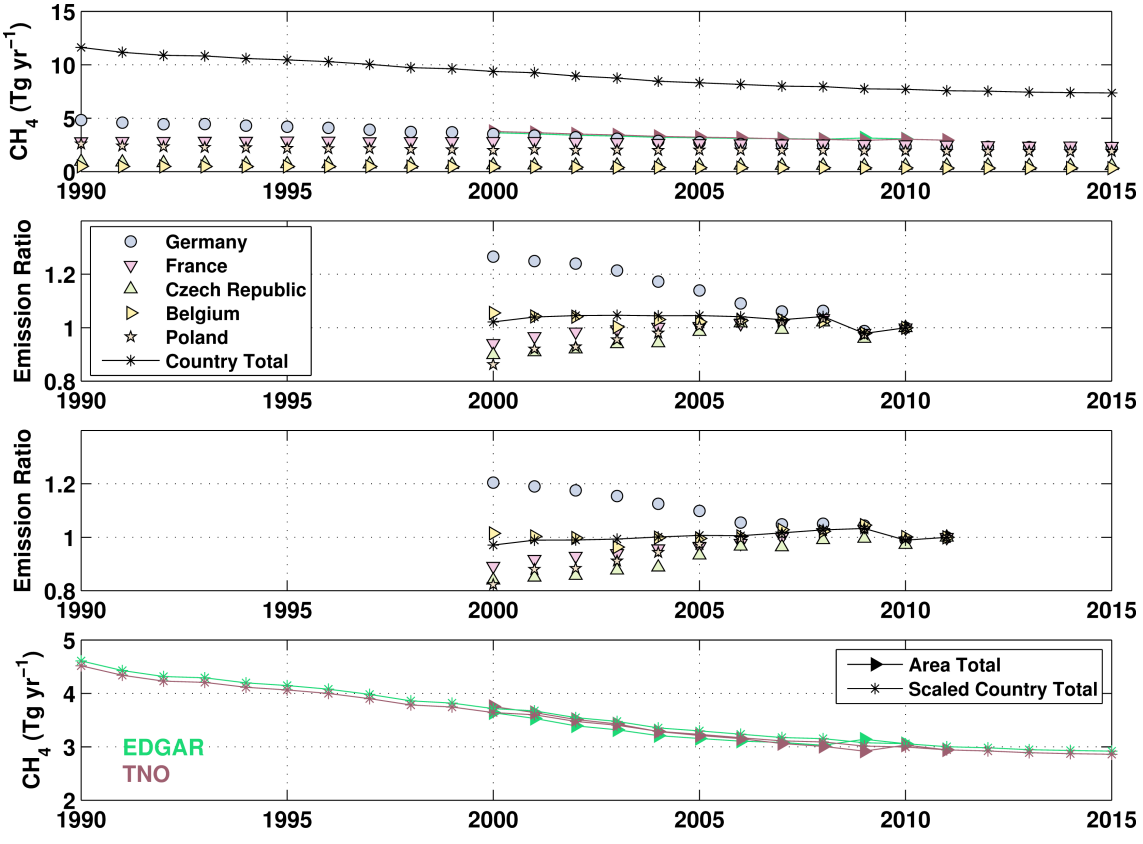

Figure A5. This four-panel plot shows the methodology for scaling the country-level emissions of CH_4 reported to the UNFCCC to extrapolate the gridded inventory emissions to 2015. The panels and symbols follow the same description as in Fig. A4.