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Co Emmisions in Mine

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Abstract.

A severe reduction of greenhouse gas emissions is necessary to reach the objectives of the Paris Agreement. The implementation and continuous evaluation of mitigation measures requires regular independent information on emissions of the two main anthropogenic greenhouse gases, carbon dioxide (CO2) and methane (CH4). Our aim is to employ an observation-based method to determine regionalscale greenhouse gas emission estimates with high accuracy. We use aircraft- and ground-based in situ observations of CH4, CO2, carbon monoxide (CO), and wind speed from two research flights over the Upper Silesian Coal Basin (USCB), Poland, in summer 2018. The flights were performed as a part of the Carbon Dioxide and Methane (CoMet) mission above this European CH4 emission hot-spot region. A kriging algorithm interpolates the observed concentrations between the downwind transects of the trace gas plume, and then the mass flux through this plane is calculated. Finally, statistic and systematic uncertainties are calculated from measurement uncertainties and through several sensitivity tests, respectively. For the two selected flights, the in-situ-derived annual CH4 emission estimates are 13.8±4.3 and 15.1±4.0 kg s−1 , which are well within the range of emission inventories. The regional emission estimates of CO2, which were determined to be 1.21 ± 0.75 and 1.12 ± 0.38 t s−1 , are in the lower range of emission inventories. CO mass balance emissions of 10.1±3.6 and 10.7±4.4 kg s−1 for the USCB are slightly higher than the emission inventory values. The CH4 emission estimate has a relative error of 26 %–31 %, the CO2 estimate of 37 %–62 %, and the CO estimate of 36 %–41 %. These errors mainly result from the uncertainty of atmospheric background mole fractions and the changing planetary boundary layer height during the morning flight. In the case of CO2, biospheric fluxes also add to the uncertainty and hamper the assessment of emission inventories. These emission estimates characterize the USCB and help to verify emission inventories and develop climate mitigation strategies.

Introduction

One of the main objectives of the Paris Agreement is to keep the global temperature rise well below 2 ◦C compared to preindustrial levels (UNFCCC, 2015).

This ambitious goal can only be reached by a severe reduction of greenhouse gas emissions. The development of efficient mitigation strategies and the implementation and management of long-term policies requires consistent, reliable, and timely information on emissions of the two main anthropogenic greenhouse gases, carbon dioxide (CO2) and methane (CH4). Carbon monoxide (CO) can be used as an additional tracer for comparison with emission inventories and as a proxy for CO2 from fossil fuel combustion. It is produced from the incomplete combustion of fossil fuels and biomass and reacts with the hydroxyl radical (OH), thus affecting the main sink of CH4. The globally averaged atmospheric abundances of CO2 and CH4 have increased by 47 % to 407.8 ± 0.1 ppm and by 159 % to 1869±2 ppb, respectively, in the period from 1750 to 2018 (WMO, 2019).

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The relative contribution of individual sources and sinks to atmospheric CH4 is still highly uncertain and the factors that affect these sources and sinks are not fully understood (Saunois et al., 2020). After a period of stable mole fractions since 2000, the atmospheric abundance of CH4 has started to increase again in 2007, and after 2014 the increase intensified yet again (Nisbet et al., 2014, 2016). The reason for this increased growth is currently investigated in several studies, which partly contradict each other by discussing biogenic sources, fossil fuel emissions and/or a decrease in the OH sink (Hausmann et al., 2016; Schaefer et al., 2016; Saunois et al., 2017; Turner et al., 2017; Worden et al., 2017; Nisbet et al., 2019). Atmospheric emission inventories for trace species are usually based on bottom-up data-based approaches. Here, emissions for individual facilities, sectors, or sources are compiled into a comprehensive database. If direct emission data are not available, they are often calculated using activity data, like the mass of coal extracted, together with emission factors.

For Annex I countries, sector-specific emissions of greenhouse gases have to be reported annually under the United Nations Framework Convention on Climate Change (UNFCC). Other countries are encouraged to report national totals of emissions. Bottom-up inventories can thus include single-source emissions or national totals, or they can be disaggregated on different spatial scales. These gridded emission inventories commonly use national emission totals and distribute them across each country using proxy data like population density or single facility locations. This method is used to compile emission inventories, which are used in climate projections, for example. The neglect of regional differences and the uncertainties in the proxy data and emission factors introduce high uncertainties into the emission inventories at grid cell level (Janssens-Maenhout et al., 2019). Without accurate emission estimates it is challenging to create reliable future climate projections and develop efficient mitigation strategies.

Therefore, there is a strong need for an independent and objective verification of emissions from individual sources or source regions based on atmospheric observations, usually referred to as top-down approaches. Top-down studies based on satellite data provide information on global and regional scales. For methane, emission quantification of individual sources has recently been demonstrated on very large point sources (Pandey et al., 2019; Varon et al., 2019), but quantification of smaller sources is still difficult. Here, airborne measurements reveal more detailed insights on smaller scales, because in situ measurements allow the study of emission sources with high spatial resolution and accuracy. Highprecision measurements of atmospheric concentration can be used for the top-down estimation of emissions from specific regions or sectors using atmospheric inversion models (Gurney et al., 2002; Thompson et al., 2014; Bergamaschi et al., 2018) and for the validation of numerical models used to calculate atmospheric abundances based on bottom-up emission inventories (Krinner et al., 2005; O'Shea et al., 2014). Airborne measurements provide highly valuable data for an independent assessment of anthropogenic CH4, CO2 and CO emissions, because the majority of these emissions originate from a small fraction of the globe, namely fossil fuel exploitation facilities, cities and power plants. Airborne measurements have shown to be useful in emission assessment of anthropogenic emissions from several sectors, including landfills (Cambaliza, 2015; Krautwurst et al., 2017) and oil and gas production regions (Karion et al., 2015; Yuan et al., 2015; Alvarez et al., 2018; Barkley et al., 2019). Plant et al. (2019) and Ren et al. (2018) showed that North American cities emit more CH4 than suspected, because of underestimation of natural gas leakage or lack of inclusion of end use emissions. Aircraft top-down approaches can be used in several ways to obtain greenhouse

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Vol. 9 Issue 02
February2022

gas flux estimates. One way is the mass balance approach, where the emissions are estimated from observed in situ mole fractions and wind speeds in the target region. Different flight patterns are used for mass balance studies: a single downwind flight transect in the approximate vertical center of the boundary layer (Karion et al., 2013) or several transects of the plume at the same height but different distances from the source (Turnbull et al., 2011) are sufficient in the case of a well-mixed planetary boundary layer (PBL). A better understanding of vertical trace gas distribution is achieved by several transects at different heights but the same distance (Cambaliza, 2015; Karion et al., 2015; Pitt et al., 2019). Single point sources or small areas can be assessed by circular flight paths at different heights (Conley et al., 2017; Tadic et al., 2017; Ryoo et al., 2019). The airborne ´ eddy covariance technique can directly infer vertical fluxes (Hiller et al., 2014; Yuan et al., 2015). Further techniques for airborne emission estimation include active and passive remote sensing instruments (Amediek et al., 2017; Krautwurst Observational data During the CoMet 1.0 campaign several aircraft- and groundbased instruments were used to extensively sample greenhouse gas emissions of the USCB in early summer 2018.

Here we present measurements taken aboard the DLR Cessna Grand Caravan 208B (Caravan). The Caravan was based in Katowice, Poland, from 29 May to 13 June 2018. Ten research flights were conducted in the USCB targeting different parts of the USCB. The flight paths were planned using a CH4 plume forecast provided by the online-coupled, 3 times nested global and regional MECO(n) model (Nickl et al., 2020). For our estimation of entire USCB emissions, we use airborne in situ observations from two flights on June 6, 2018, one in the morning (09:22–11:45 UTC, 11:22– 13:45 CEST) and one in the afternoon (13:01–15:28 UTC, 15:01–17:28 CEST), in the following referred to as flights A and B, respectively. Figure 1 shows the flight track of flight B on a map with the CH4 emission sources. Both flights were designed in a box pattern with an upwind leg in the northeast approximately in the middle of the PBL and the downwind wall in the southwest with flight transects at several heights. CH4, CO2 and CO enhancements were clearly observed in the downwind wall. The flights were conducted in coordination with ground-based teams, which drove the instrumented vans below the upwind and downwind legs. Their tracks and sampled CH4 mole fractions for the afternoon flight are shown in Fig. 1. For the emission estimation, we selected ground-based data according to closeness in time. Sampling times for flight and ground-based data are listed in Table S1 in the Supplement. Additionally, three Doppler wind lidar Leosphere Windcube 200S instruments were stationed at Rybnik, Wisła Mala and Krzykawka to measure vertical profiles of wind speed, wind direction and turbulence parameters (Fig. 1). Details on the CoMet lidar wind measurement setup and the planetary boundary layer height (PBLH) determination are given in Wildmann et al. (2020) and Luther et al. (2019). A sophisticated suite of instruments aboard the Caravan gathered both meteorological parameters and trace gas concentrations. A five-hole probe, connected to a pressure transducer, is mounted on a nose boom under the left wing of the aircraft and measured the three-dimensional wind vectors. The temperature, pressure and humidity sensors and the calibration of the wind measurement system are described in detail by Mallaun et al. (2015). A flight-ready cavity ring-down spectroscopy (CRDS) analyzer (G1301-m, Picarro) was installed in the cabin of the aircraft. It measured CH4, CO2 and water vapor at a frequency of 0.5 Hz with cavity ringdown spectroscopy. Trace gas concentrations for water vapor were corrected according to Rella et al. (2013). The calibration and uncertainty assessment were conducted analogously to Klausner et al. (2020), who used the same instrument.

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Downwind and upwind background determination methods For the mass balance approach, the background mole fraction m0 of the trace gases needs to be determined. Here we compare two methods: (i) background estimated from the downwind wall's edges and (ii) background estimated from the upwind leg. The downwind background method assumes that the boundary layer height remains constant for the time of sampling within the wall, while the upwind method requires the boundary layer to stay at the same height for the whole flight time and ideally a quasi-Lagrangian sampling of the same air mass in the upwind and downwind transects. Thus, the less strict criteria of the downwind background method are more likely to be met in real conditions, and we will use this method in our best estimate and the upwind background as a sensitivity test. The downwind method also requires that there are no sources upwind of the area of interest which would create a complex concentration pattern flowing into the domain. To show this we used our upwind flight transect similar to previous studies (Karion et al., 2013; Heimburger et al., 2017). In order to determine the downwind background mole fraction from the wall's edges, we evaluate the variability of the CH4 observations within the PBL. The background is separated from the plume using the standard deviation within a 2 min interval for airborne and 10 min interval for groundbased data. Starting at the edges of the wall, the interval is moved towards the center. We define the boundary between CH4 atmospheric background and plume where the standard deviation surpasses 3.4 ppb CH4. The average CH4 background standard deviation is 2.9 ppb. The CO2 background section is adopted from the CH4 background, because the variability in the background is too high for this approach to be applicable. The CO background threshold for the 2 min interval is 4.5 ppb with an average background standard deviation of 3.5 ppb. We average all background mole fraction observations within the PBL to the south and north of the plume separately. The mean of these two values is considered to be the average background for the downwind method. Thus, we assume a linear spatial gradient in the trace gas background. The second way of determining the atmospheric background mole fraction uses the observations within the bound

Conclusions

In times of rising atmospheric concentrations of greenhouse gases and countries trying to reduce their associated emissions, it is important to develop an independent and objective emission monitoring system. During the CoMet campaign the European CH4 emission hot spot of the Upper Silesian Coal Basin (USCB) was sampled by in situ techniques as well as passive and active remote sensing on ground and from aircraft. From two flights A and B around the USCB, conducted on 6 June 2018, combined with vehicle-based ground measurements, we determined a regional emission estimate of CH4, CO2 and CO for the entire USCB using in situ data and a mass balance approach. The plumes of all three trace gases could be observed and separated from the atmospheric background in all downwind transects. For the morning flight

A, a trace gas enhancement correction was employed to account for the temporal change of PBLH during the sampling. We employed a kriging algorithm for the interpolation of observed CH4, CO2, CO and wind speed between the flight transects and towards the ground. CH4 ground-based observations confirmed the existence of a well-mixed PBL with similar trace gas enhancements at the ground and in the aircraft transects. From the kriged fields we calculated the USCB emission estimate as the mass flux through the downwind wall for each flight. Using error propagation and several sensitivity tests, we carefully determined the total error of our mass balance approach. The CH4 emission estimate has a total relative error of 26 %–31 %, the CO2 estimate of 37 %–62 % and the CO estimate of 36 %–41 %.

These uncertainties are mainly caused by the background determination, wind speed variability and missing knowledge of mole fractions below the lowest flight track for CO2 and CO. The higher uncertainty values apply to the morning flight estimate, because the temporal variation in the PBLH introduced a large error. Thus, we highlight the importance of a constant PBLH over time, knowledge of trace gas mole fractions at the ground and the exact knowledge of background mole fractions. The large uncertainties in the CO2 estimate are dominated by the uncertainties in biospheric CO2 fluxes. These estimates could be improved by performing flights in wintertime, when the biospheric fluxes are negligible. Flights during different seasons would also better constrain the annual cycle in CO2 emissions from the residential sector. The calculation of emission estimates from single flight transects is not advisable, because the single-transect estimates showed deviations from their mean and the kriging method of more than 40 % in both directions.

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