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EXTENDED ABSTRACT

Quantifying Greenhouse Gas Emissions from Point Sources

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Abstract

This work presents a unique set of various ground-based remote sensing measurements. These measurements can provide a solid basis for assessing the capabilities and uncertainties involved in quantifying GHG emissions from large point sources, by combining remote sensing with atmospheric simulations. The facility under study is the steel factory complex of Bremen, Germany. Consisting of two blast furnaces and an associated blast-furnace-gas-fired power plant, it contributes approximately 5 Mt/yr to the city of Bremen's CO_2 emissions, representing nearly half of the total. Campaign measurements, conducted between April and May 2025, target the plumes of the steelworks and the surrounding industries. Two portable Bruker EM27/SUN FTIR spectrometers provide accurate observations of the column-averaged abundances of CO_2, CO and CH_4. Mobile zenith-sky measurements using a DOAS instrument retrieve vertical column densities of emitted NO_2, enabling real-time plume tracking. Information on the wind profile is collected using a wind lidar. Field measurements are supported by the Bruker 125HR FTIR spectrometer at the University of Bremen, which provides background values for atmospheric GHG concentrations. The CO/CO_2 emission ratio (3.46 ± 0.85%) agrees well with inventory data, though satellite estimates were higher. Plume observations were compared with a Gaussian dispersion model. While the model alone could not reproduce the observed plume behaviour due to atmospheric variability, integrating real-time plume measurements allowed preliminary emission estimates ranging from 40% to 179% of inventory values, with a mean of 79% ± 49%. This study demonstrates that combining multiple ground-based instruments provides valuable constraints on emissions from complex industrial facilities, highlighting both the potential and current limitations of facility-level GHG monitoring.

Introduction

Power plants and large industrial facilities are responsible for over half of global anthropogenic CO_2 emissions. As a result, accurate and independent monitoring of these sources is essential to refine global carbon estimates and support international climate commitments.

The simulation of plume trajectories using atmospheric models is employed to estimate source emissions by comparing with forward modelling or utilising inversions. This approach is often complementary to satellite-based GHG measurements. One model that is employed in many studies to date is the simple Gaussian plume model (e.g. Guo et al. 2023). However, atmospheric instability and turbulence introduce high uncertainty into these estimates, and the model requires a large dataset to be statistically meaningful. Various other dispersion models utilising high-resolution meteorological data can be used to achieve greater accuracy, but uncertainties remain (Brunner et al. 2023). High background atmospheric CO_2 levels further complicate emission quantification. This issue can be overcome by using different co-emitters as trace gases for CO_2 in emission plumes, such as CO for steel factories (Schneising et al. 2024).

The limited temporal and spatial resolution of satellite-based greenhouse gas concentration measurements necessitates the use of ground-based measurement techniques to support emission quantification at the facility level. Ground-based remote sensing instruments, such as Fourier transform infrared spectrometers, are commonly used to validate satellite measurements (Buchwitz et al. 2018). The portable EM27/SUN FTIR spectrometer allows field measurements to be taken near emitters to enable direct emission estimates. These can be made either in a stationary configuration in combination with dispersion models (Ohyama et al. 2021), or in a mobile configuration using flux estimation methods (e.g. Luther et al. 2019). Further contributions from ground-based remote sensing techniques to improving emission estimates from point sources are investigated.

Methodology

Precise and accurate measurements of column-averaged abundances of CO_2, CH_4, and CO are conducted with two stationary EM27/SUN FTIR spectrometers, providing a spectral resolution of 0.5 cm^{-1} (Gisi et al. 2012). They are provided by the Karlsruhe Institute of Technology. The measurement location is chosen day-wise to be downwind of the emission stacks. Changes in wind conditions shift the plume in and out of the observed atmospheric column. From simultaneous changes in XCO and XCO_2 an emission ratio was derived.

The wind field is observed with a Wind-LIDAR ZX300. It records 15 sec mean values of wind direction and wind speed in 10 different heights between 10 m and 300 m with a precision of 0.1 m/s for speed and 0.5° for direction. The lidar is located approx. 1.5 km south-east of the steelworks complex.

The behaviour of the plume in the changing wind field is monitored with mobile zenith-sky DOAS measurements taken from a car (car DOAS, Schreier et al. 2019). These measurements are used to determine the vertical column density of NO_2 in the atmospheric column above the measurement point. NO_2 is co-emitted from the steelworks stack and can therefore be used as a tracer for the emission plume. The rapid retrieval enables real-time plume tracking and helps determine its location and dynamics.

Emission dispersion is simulated with the Gaussian dispersion model, including calculations of the effective stack height with source specific stack parameters and usage of the dispersion coefficients according to the atmospheric stability determined by wind speed and solar insolation (Hanna et al. 1982). The horizontal dispersion resulting from this modelling approach was compared to the actual measured plume width from the DOAS measurements. For intervals with DOAS data, measured plume locations were used to constrain the Gaussian model for preliminary emission estimates.

Results

Measurements from both the DOAS and the EM27/SUN instruments clearly show the emission plume. When the plume intersects with the observed air column, the EM27/SUN records increases in column-averaged abundances of up to 5 ppm in XCO_2 and up to 200 ppb in XCO at a distance of 2 km from the source. The DOAS measurements reveal plume cross sections of gaussian shape in the vertical column values of NO_2. Examples can be seen in Figures 1a, 1b and 1c.

The CO/CO_2 emission ratio obtained from the changes in XCO_2 and XCO when the plume moves in or out of the observed columns gives a mean value of
3.46 ± 0.85 %. Satellite-based studies (Schneising et al. 2024) report a ratio of 6.28 ± 4.09%, while inventory records indicate a value of 3.33 % (Umweltbundesamt 2022).

Comparisons of the measured GHG abundance excess values and Gaussian model predictions, which are driven by atmospheric stability, wind direction and wind speed measurements reveal large discrepancies, preventing reliable direct emission estimates. Too many uncertainties regarding the plume behaviour persist. Figure 1d shows differences of up to 150% between modelled and measured plume widths. Using intervals where DOAS data are available, the model can be partially constrained. However, plume height corrections remain unaccounted for. Filtering scenes that are insensitive to plume height, given the viewing geometry of the instrument, allows for preliminary emission estimates ranging from 40% to 179% of inventory values, with an average of 79% ± 49%.

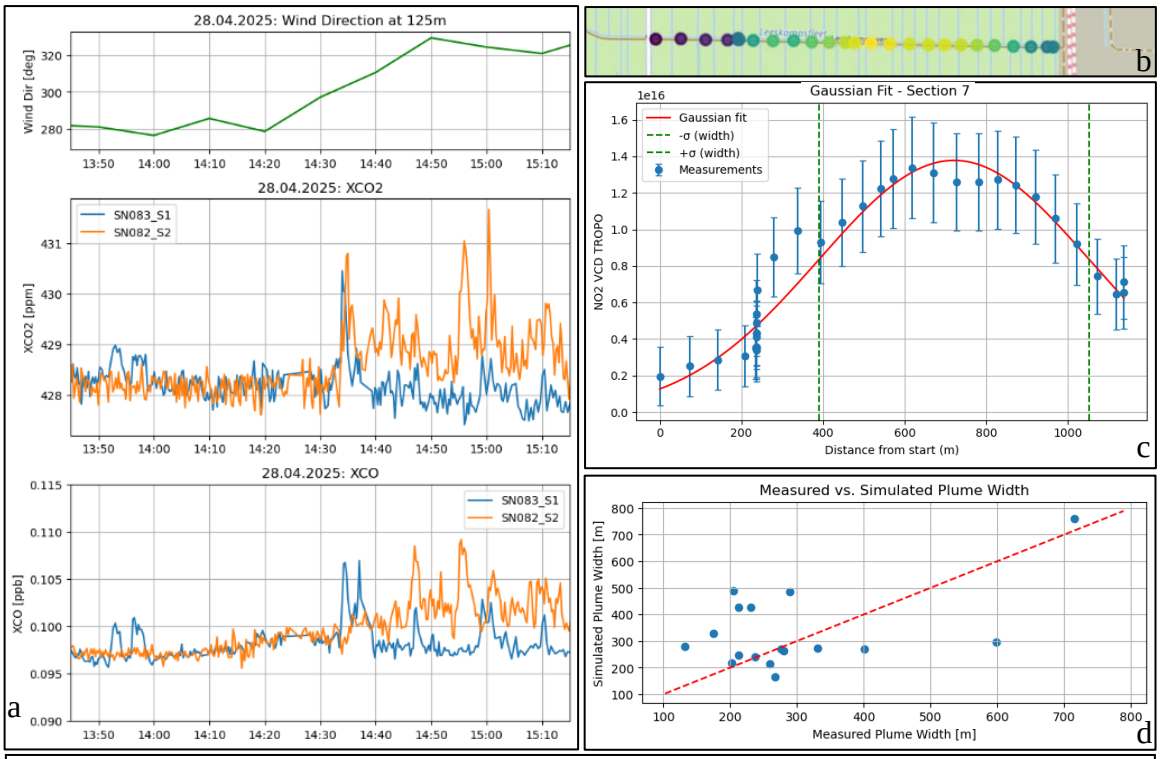

Figure 1. a) 10-min averaged wind direction from the lidar system and XCO_2 and XCO as measured by the two EM27/SUN spectrometers for the 28th April 2025. It shows the correlation between the wind direction and the measured excess values in XCO_2 and XCO, which results from the emission plume crossing the observed air column. b) Plume cross-section shown in NO_2 vertical columns as measured by the DOAS instrument (maximum in yellow). c) Gaussian fit of the plume cross-section in b) and determined plume width. d) Comparison of the plume width as determined by the measurements and the model.

Conclusions

This study demonstrates the potential of combining multiple ground-based remote sensing techniques - EM27/SUN FTIR, mobile DOAS, and Wind-LIDAR – to monitor GHG emissions from large industrial sources. The CO/CO_2 emission ratio obtained from the changes in XCO_2 and XCO when the plume moves in or out of the observed columns results in a value of 3.46 ± 0.85 %, which is markedly lower than the ratio determined by satellites (6.28 ± 4.09%) but in good agreement with the theoretical value from the emission inventory of the thru.de portal (3.33 %). Plume observations were compared with a Gaussian dispersion model. Constraining the model by integrating real-time plume measurements yielded preliminary emission estimates ranging from 40% to 179% of inventory values, with an average of 79% ± 49%. These results highlight both the promise and current limitations of ground-based remote sensing in reducing uncertainties of facility-level emission quantification. Future work should extend measurement coverage, include plume height observations, and utilise high-resolution atmospheric dispersion models to improve emission accuracy.

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