Title: Monitoring and quantifying CO2 emissions of isolated power plants from space

MS No.: egusphere-2022-1490

Below we reply to the reviewer comments point by point. The reviewer comments are shown in *italic*, and corresponding modifications and citations of the manuscript are quoted.

Referee #2

(1) The study estimates CO2 emission of power plant using OCO-2 and OCO-3 observations using the Gaussian plume inversion and cross-sectional flux method with different input parameters. The methods are tested for U.S. power plant for which bottom-up reports of hourly emissions are available and afterwards applied globally. The paper well written but some aspects on the method are unclear. I would recommend publication following a revision based on the general and specific comments below:

Response: We thank Referee #2 for the encouraging comments. All comments and suggestions have been considered carefully and addressed below.

(2) Background: For Gaussian plume model method please describe already in L153ff how you calculate the background. In L200, you write that the 90th percentile was used, which seems to be based on tests with different percentiles with the aim to minimize the difference between estimated and reported emissions (L220, Figure S2). The choice of the percentile (60-99th) will mostly result in a bias in the estimated emissions, which might be caused by the background, but can also be the result of other systematic errors in other parameters (e.g. effective wind speed). Therefore, how does this choice of the background agree with the background that you compute with the cross sectional flux method (L175)? Would a different background affect your conclusions on the best approach for computing the effective wind speed?

Response: The different backgrounds do not affect the conclusions of this study regarding the comparison of three wind fields, because the difference in background obtained by these two methods is very small (Fig. S11a), with a maximum of 0.86 ppm and a minimum of 0.004 ppm (Fig. S11b). Under the two background calculation methods, the GPM method has good consistency in the estimation results driven by three wind field (Fig. S11c-e). With the background computing by Eq. (3), the conclusion that estimated emissions have better accuracy using the WPBL is still valid (Fig. S12).

For all 50 cases, the difference between the background calculated by the 99 percentile and the background calculated by the 60 percentile ranges from 0.23 to 0.77 ppm. The standard deviation of the background calculated for the 9 percentile bins for each case ranges from 0.08 to 0.26 ppm.

In this study, the background for the Cross-sectional flux method (CFM) is determined by fitting of Eq. (3), while the background for the Gaussian plume model method (GPM) is determined by the 90th percentile. However, more importantly, the Gaussian fitting in Eq. (3) may fail for some cases (e.g., Fig.S8a, S9a), but these cases can be estimated by the GPM method. The GPM method can simulate the enhancements of XCO2 at any location downwind of the emission source in the two-dimensional plane (Fig.1d), which cannot be well described by the Gaussian fitting. Therefore, in this study, the two methods use two different background calculation methods.

In response to the sensitivity regarding the background information mentioned above, we have made changes to the manuscript in L280-286: "In this study, the background for the cross-sectional flux method is determined by fitting of Eq. (3), while the background for the Gaussian plume model method (GPM) is determined by the 90th percentile. The difference in background obtained by these two methods is very small (Fig. S11a), with a maximum difference of 0.86 ppm and a minimum of 0.004 ppm (Fig. S11b). Under the two background calculation methods, the GPM method has good consistency in the estimation results driven by three wind field (Fig. S11c-e). With the background computing by Eq. (3), the conclusion that estimated emissions have better accuracy using the WPBL is still valid (Fig. S12)." and the supplement in L25-27 and L74-85. We have also added the following Figure S11, S12 to the supplement:

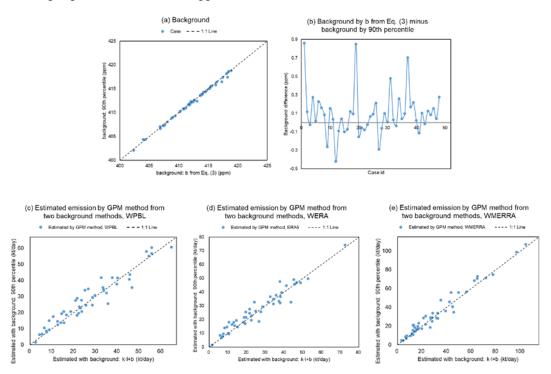
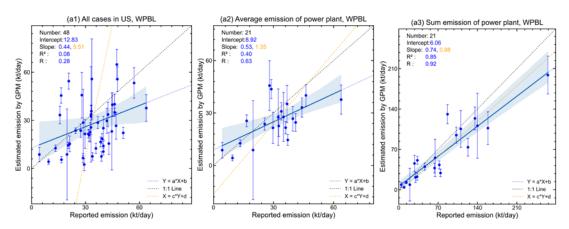


Figure S11. Comparison of two methods of computing the background. The background constant b from Eq. (3) is in good agreement with the background calculated by using the 90th percentile (a), and their difference is small (b). Under the two background calculation methods, the GPM method has good consistency in the estimation results driven by WPBL (c), WERA (d), and WMERRS (e) wind fields, respectively.



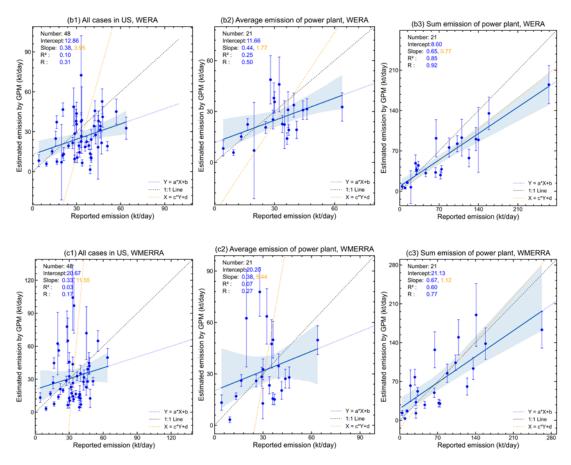


Figure S12. The conclusion that estimated emissions have better accuracy under the WPBL is still valid when using the background calculated by the method of Eq. (3). These three panels are based on WPBL (a1-a3), WERA(b1-b3), and WMERRA(c1-c3).

(3) Wind: The evaluation of the different wind products in your study is inconsistent. You use winds from ERA-5 (0.25°), MERRA-2 (0.5°x0.625°) and high-resolution ECWMF forecast (resolution not mentioned). You use the wind speed at the center of the PBL for the ECMWF forecast. However, for ERA-5 and MERRA-2, you take 10-m winds multiplied by the empirical scaling factor of 1.4 from Varon et al. (2018). When you compare the impact of the different wind products on the estimated emissions, it is not possible to identify if the different performances are caused by differences between the products or the different computation of the final product (scaling factor or wind at half PBL height). To analyse this better, I suggest comparing all datasets using both the 1.4-factor and the wind at half the height of the PBL. Note that the scaling factor of 1.4 is derived for CH4 plumes measured by high-resolution satellites, which are inherently different to CO2 plumes from power plants measured by OCO-2. Therefore, using the value might be not the best approach, even it is true that it has been used in previous studies "for convenience" (Reuter et al. 2019).

Response: The resolution of ECMWF (WPBL) is $0.25^{\circ} \times 0.25^{\circ}$ (added in L140).

In this study, we compared the effective wind computed from ERA5 and MERRA2 10 m wind speed with the wind at half the height of the PBL. To some extent, the wind at half the height of the PBL is also a representation of the average state of the plume and an effective wind. Finally, we found that the results using the wind at half the height of the PBL were the best (Fig. 2), which suggests that it represents the spreading of CO₂ plumes in the vertical direction more accurately (Fig. 2, Fig.

S3, Fig. S4). The reason why the results using MERRA2 were the worst (Fig. S4) is due to its low resolution, which cannot provide precise wind information for emission sources. Here, ERA-5 and operational forecast of ECMWF are very similar and assumed to perform in the same way. We didn't use the MERRA2 wind at half the height of PBL because of its lower resolution.

Varon *et al.* (2018) calculated an effective wind speed $U_{\rm eff}$ (Eq. S1-2) representing the average state of the vertical variability of the wind, based on the known Q and C, and found a multiple relationship (1.3 ~ 1.5) between $U_{\rm eff}$ and the 10 m wind speed. Hence, subsequent studies used a scaling factor of 1.4 applied to the 10 m wind speed to represent the average state of the plume in the vertical direction (Reuter et al., 2019; Hakkarainen et al., 2021). Although this factor was derived for methane (lighter than CO2) plumes, it is a commonly used method.

$$Q = \int_{-\infty}^{+\infty} U(x, y) \Delta\Omega(x, y) dy,$$
(S1)

$$Q = CU_{\text{eff}}$$
, with $C = \int_{-\infty}^{+\infty} \Delta\Omega(x, y) dy$. (S2)

We changed the manuscript in L267~272:

"The correlation coefficient R of the estimated emission and time-corrected reported US EPA emission of the 50 cases of isolated power plants are 0.35, 0.28, and 0.14, for WPBL, WERA and WMERRA respectively (Figure 2a, Figure S3a, Figure S4a). The results show that the emission estimate obtained using WPBL give better results than the other two wind options, which suggests that it represents the spreading of CO2 plumes in the vertical direction more accurately (Fig. 2, Fig. S3, Fig. S4). The reason why the results using MERRA2 were worse (Fig. S4) is due to its low resolution, which cannot provide precise wind information for emission sources."

(4) Uncertainties: You seem to compute the uncertainties using an ensemble approach with a rather small number of members (3 for wind and 4 for background) for computing reasonable statistics (see also previous comment on the wind). How large are the uncertainties of wind speed in m/s and background in ppm? How do these uncertainty estimates compare to estimated uncertainties in previous studies? How large is the uncertainty of the fitting parameters for the background in Equation 3? Finally, how large would be the uncertainties from the assumption and simplification of your methods such as the assumption of steady state conditions?

Response:

How do these uncertainty estimates compare to estimated uncertainties in previous studies?

We added the uncertainty comparison in the revised manuscript L316-317: "The total uncertainty is comparable to the uncertainty of power plant emissions in previous studies, which ranged from 3.42 to 19.2 (Nassar et al., 2017; 2022)"

How large is the uncertainty of the fitting parameters for the background in Equation 3?

For all cases, the uncertainty of the background was only considered for the four percentile values (75th, 80th, 85th, 90th percentile). As mentioned in our response to question (2), the difference in background values calculated by the two methods is small (Fig. S11). The manually selected cases ensured the reliability of the isolated power plant observation signal, i.e., the background uncertainty is low.

how large would be the uncertainties from the assumption and simplification of your methods such as

the assumption of steady state conditions?

We think this is out of scope of our current paper and to be analysed in future research. We mentioned this in the revised manuscript of discussion L399-398, as follows:

"This study has only considered three sources of uncertainty. Future research may investigate additional sources, such as the assumption of steady state conditions and the plume rise, to better understand their impact on the results."

How large are the uncertainties of wind speed in m/s and background in ppm?

We added the uncertainty in the revised manuscript L317-318: "The uncertainty of wind speed is between 0.08 and 1.4 m s⁻¹, and the uncertainty of background varies between 0.03 and 0.1 ppm (Table S1)."

Table S1. The uncertainties of wind speed in m/s and background in ppm for each plant

Name	Uncertainty of background (ppm)	Uncertainty of wind (m/s)
James H Miller Jr (AL)	0.098	0.087
Apache Station (AZ)	0.039	0.235
Arlington, Mesquite, Redhawk Facility (AZ)	0.043	0.246
Prairie State Generating Station (IL)	0.063	0.238
Gibson (IN)	0.067	0.307
Jeffrey Energy Center (KS)	0.062	0.220
Iatan (MO)	0.076	0.355
Labadie (MO)	0.069	0.315
Colstrip (MT)	0.067	0.421
Gerald Gentleman Station (NE)	0.110	0.548
Four Corners Steam Elec Station (NM)	0.052	0.727
Cardinal (OH)	0.066	0.230
Conemaugh, Seward (PA)	0.054	1.491
Cumberland (TN)	0.066	0.940
Harrington, Nichols station (TX)	0.078	0.172
Oak Grove (TX)	0.077	0.366
Parish, Carbon-Capture, Brazos Energy (TX)	0.069	0.528
Sam Seymour (TX)	0.070	0.583
Hunter (UT)	0.070	0.219
Intermountain (UT)	0.051	0.209
Dry Fork Station (WY)	0.042	0.960
Laramie River (WY)	0.062	1.052

Specific comments

(5) L171ff: You write here that you fit equation 3 to obtain parameters, k, b, A and sigma. Then, you write that you subtract the background to compute the line density. However, your parameter A is already the line density, so I don't understand why you need to calculate it again.

Response: We use the Eq. 3 to fit the discrete observations along-track distance into a continuous curve, and further obtain the integral of the cross section (C in Eq. S2). The parameter A is one of the parameters to be fitted and not the line density. The line density (ppm·m) is the shaded area of Fig. R4 (the cross-plume integral), and then converted to g m^{-1} through Eq. 2. Note that the definition of the parameter A

here is different from Kuhlmann et al. (2021)

$$V(x,y) = \frac{F}{\sqrt{2\pi \cdot \beta \cdot (\frac{x}{1000})^{0.894} \cdot u}} e^{-\frac{1}{2} (\frac{y}{\beta \cdot (x/1000)^{0.894}})^2} , \qquad (1)$$

$$XCO_2 = V \cdot \frac{m_{air}}{m_{CO_2}} \cdot \frac{g}{P_{surf} - \omega \cdot g} \cdot 1000 , \qquad (2)$$

$$f(l) = k \cdot l + b + \frac{A}{\sigma\sqrt{2\pi}} e^{[-(l)^2/2\sigma^2]}$$
, (3)

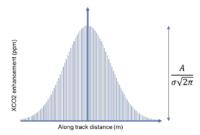


Figure R4. The blue shaded area is the line density. The peak is A/(sigma*sqrt (2*pi)).

(6) L182: It is not clear to me how you compute the wind here. Do you rotate the wind vector so that it points from the source location to the maximum in the OCO swath?

Response: For the CFM, we use the same fitting of the wind direction as the GPM described in the previous paragraph: "The wind direction is allowed to rotate within a range of \pm 60° to account for errors in the wind data. The optimal wind direction is derived by maximizing the correlation coefficient between the simulated and the observed XCO2 enhancement". The end result is that the wind direction will roughly point towards the plume points (red points in Fig. 1b), possibly deviating from the maximum, because it is determined by all the red points together.

(7) L235: You write that the peak is well described by a Gaussian [curve] in Figure 1b. However, no curve is shown in the figure.

Response: Thanks. We have added the Gaussian curve in Figure 1b of the revised manuscript, as follows:

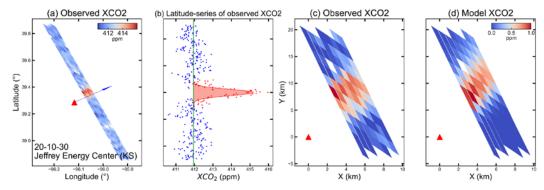


Figure 1. Estimation process of power plant emissions (a-d).

(8) L240ff: You partly repeat the description of your method here, which seems unnecessary.

Response: We agree that this part repeats the description of method. However, to better help readers understand the cases in Figure 1 and Figure S10, we explained how to distinguish and select clear plume cases and reject noise cases, so we prefer to keep it.

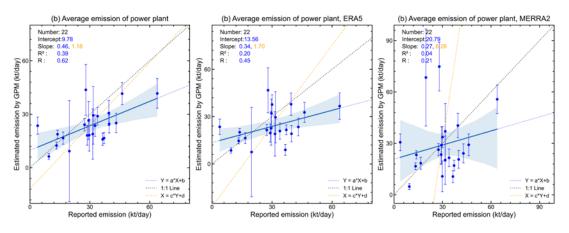
(9) L261: Please discuss why WPBL provides better results than the other two options.

Response: We have added why WPBL provides better results in the revised manuscript L267-272: "The correlation coefficient R of the estimated emission and time-corrected reported US EPA emission of the 50 cases of isolated power plants are 0.35, 0.28, and 0.14, for WPBL, WERA and WMERRA respectively (Figure 2a, Figure S3a, Figure S4a). The results show that the emission estimate obtained using WPBL gives better results than the other two wind options, which suggests that it represents the spreading of CO₂ plumes in the vertical direction more accurately (Fig. 2, Fig. S3, Fig. S4). The reason why the results using MERRA2 were worse (Fig. S4) is due to its low resolution, which cannot provide precise wind information for emission sources."

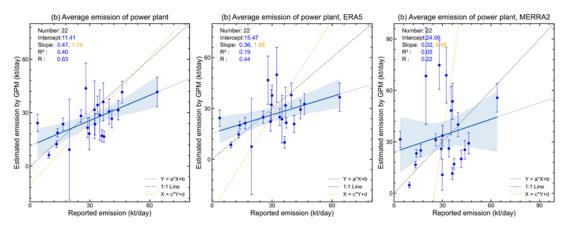
(10) L265: Do you use the arithmetic average or the weighted average considering the uncertainty of the estimates?

Response: We used the arithmetic average. We also tested the weighted average considering the uncertainty of the estimates and get the same conclusion that WPBL has the best performance shown in the following figures. We added an additional statement in the manuscript L275-277: "We also tested the weighted average considering the uncertainty of the estimates and reached the same conclusion that WPBL shows the best performance."

The weighted average considering the uncertainty of the estimates:



The arithmetic average (used):



(11) L268/Figure 2: It surprises me that r² is so much higher for summing compared to averaging? Can

you explain why this is the case?

Response:

When summing emissions for each plant, the data range is much higher than averaging. When a power plant has multiple observations, the difference between the sum of estimated and reported emissions for these observations is very small. Using the sum of the emissions is a non-standard approach. Generally one wants to compare similar characteristics of a process, in this case the typical (average) emissions per power plant.

However, with significant variations in number of collocations per powerplant, errors in average emissions vary considerably between power plants. Furthermore, emissions themselves vary considerably between power plants. As a result, more accurate and less accurate emission estimates are mixed in figure 2, which complicates reading and interpreting the plot.

Using the sum of emissions helps to visually discriminate between locations with more or fewer collocations. For the emission estimates themselves this does not matter: if the method is accurate the results should always align regardless of whether comparing averages or sums.

By displaying results from both approaches in one plot the reader can always compare the results from both methods.

(12) L279: This relates back to my previous comment how you do calculate the normal wind for both methods. Are the difference between estimates and wind speed used in both method correlated? Another reason for deviations can be the method for computing the background.

Response: The wind data finally used is the same for the GPM and CFM and is WPBL. The background fitted using Eq. (3) and the background calculated from percentiles differ very little (Fig. S11). More details about the error of the CFM method are shown in our response to question (16).

(13) Figure 5: The red line is somewhat misleading, because without reading the caption one would assume that you could estimate emissions for 8% of all tracks near power plant, while in truths it is only 0.05%. I would strongly suggest removing it to avoid confusion.

Response: We agree that the red line is confusing. We have removed the red line of Figure 5 in the revised manuscript.

(14) L340ff: Does this number of 1522 Mt/a correctly accounts for observing the same power plant in different years or does this never happens? I am asking because the percentage numbers for the individual years add up exactly to 17, which would not happen if you estimate for the same power plant more than once.

Response: For the same power plant, when there are multiple observation cases spanning multiple years, we directly take the estimated average of these cases. The 106 cases involved 78 unique power plants. Then we extrapolated the average emission (kt/day) of 78 power plants to the annual emission (Mt/year), and finally it was concluded that it accounted for 17% of the power sector emissions in 2018.

(15) Figure 7b: It is difficult to see the bars for most countries. Maybe the figure can use a logarithmic scale on the y-axis.

Response: We thank you for the suggestions. We have changed the Figure 7b with a logarithmic scale on the y-axis in the revised manuscript, as follows:

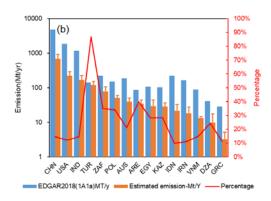
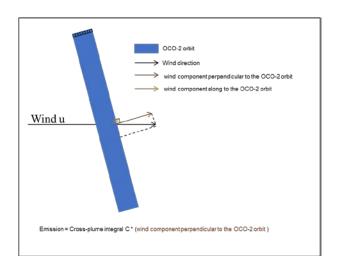


Figure 7. (b) The red curve in the right figure shows the percentage of estimated emissions in comparison to the country total power plant emissions according to the inventory.

(16) L367f: The conclusion on the difference between cross-sectional flux and Gaussian plume method needs more explanations (see previous comment).

Response: For the CFM method, by mass balance, the integral of the cross-section (Eq. S1) perpendicular to the downwind direction of the emission source multiplied by the wind speed of the emission source is the emission rate of the source. However, for the OCO-2 satellite with a narrow-width scanning mode, its orbit has a certain angle with the cross-section perpendicular to the plume, that is, the satellite orbit is not perpendicular to the wind direction of the emission source. Note although OCO-3 obtained plume data over a larger area in urban regions through multiple scans, we only selected one scan orbit as our analysis target. Therefore, an alternative method is to estimate that the emission rate is equal to the cross-sectional flux area multiplied by the component of wind perpendicular to the orbit. The maximum error in the instability of this method comes from the component of wind perpendicular to the orbit. If the satellite's trajectory and wind direction are completely perpendicular, the CFM method is feasible. This is the fundamental reason for the large error in estimating point source CO2 emissions using the CFM method applied to OCO-2 satellite observations. On the other hand, GPM directly simulates XCO2 enhancement at any downwind position using the wind direction of the emission source, avoiding this issue and obtaining more stable results.

We added explanations in the revised manuscript L378-382: "We found that the cross-sectional flux method has a larger variability than the GPM method. This is because the emission rate from the cross-sectional flux method is equal to the cross-sectional flux area multiplied by the component of wind perpendicular to the orbit. The maximum error in the instability of this method comes from the component of wind perpendicular to the orbit. However, the GPM method directly simulates XCO2 enhancements at any downwind position using the wind direction of the emission source, avoiding this issue and obtaining more stable results."



(17) L374f: Unfortunately, GeoCarb was recently canceled. CO2M is developed by ESA and EU with involvement from EUMETSAT and ECMWF. It is probably easiest to call it a "European mission". The Japanese GOSAT-GW should be mention here, as well.

Response: Thanks. We have corrected it in the revised manuscript, as follows:

"such as the planned European Carbon Dioxide Monitoring Mission (CO2M) and the Japanese Global Observing Satellite for Greenhouse gases and Water cycle (GOSAT-GW)".

(18) Supplement: The resolution of the figures in the supplement is very low making it difficult to read the labels. In some cases, labels and units are missing (e.g., S9).

Response: We thank you for the suggestions. We have changed the figures with higher resolution in the revised manuscript, as follows:

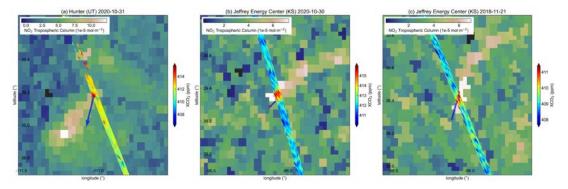
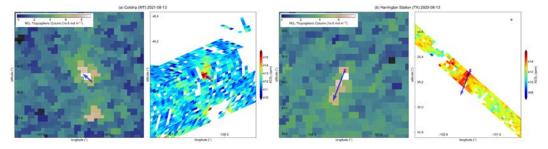


Figure S8. XCO2 and same-day NO2 concentration $(0.025^{\circ} \times 0.025^{\circ})$ for three OCO2 cases.



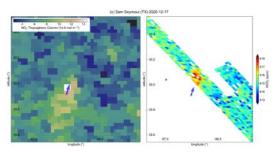


Figure S9. XCO2 and same-day NO2 concentration $(0.025^{\circ} \times 0.025^{\circ})$ for three OCO3 cases.

Hakkarainen, J., Szeląg, M. E., Ialongo, I., Retscher, C., Oda, T. and Crisp, D.: Analyzing nitrogen oxides to carbon dioxide emission ratios from space: A case study of Matimba Power Station in South Africa, Atmos. Environ.: X, 10, 100110, https://doi.org/10.1016/j.aeaoa.2021.100110, 2021.

Kuhlmann, G., Henne, S., Meijer, Y. and Brunner, D.: Quantifying CO2 Emissions of Power Plants With CO2 and NO2 Imaging Satellites, Frontiers in Remote Sensing, 2, https://doi.org/10.3389/frsen.2021.689838, 2021.

Nassar, R., Hill, T. G., McLinden, C. A., Wunch, D., Jones, D. B. A. and Crisp, D.: Quantifying CO2 Emissions From Individual Power Plants From Space, Geophys. Res. Lett., 44, 10045-10053, https://doi.org/10.1002/2017gl074702, 2017.

Nassar, R., Moeini, O., Mastrogiacomo, J.-P., O'Dell, C. W., Nelson, R. R., Kiel, M., Chatterjee, A., Eldering, A. and Crisp, D.: Tracking CO2 emission reductions from space: A case study at Europe's largest fossil fuel power plant, Frontiers in Remote Sensing, 3, https://doi.org/10.3389/frsen.2022.1028240, 2022.

Reuter, M., Buchwitz, M., Schneising, O., Krautwurst, S., O'Dell, C., Richter, A., Bovensmann, H. and Burrows, J. P.: Towards monitoring localized CO2 emissions from space: co-located regional CO2 and NO2 enhancements observed by the OCO-2 and S5P satellites, Atmos. Chem. Phys., 19, 9371-9383, https://doi.org/10.5194/acp-19-9371-2019, 2019.

Varon, D. J., Jacob, D. J., McKeever, J., Jervis, D., Durak, B. O. A., Xia, Y. and Huang, Y.: Quantifying methane point sources from fine-scale satellite observations of atmospheric methane plumes, Atmos. Meas. Tech., 11, 5673-5686, https://doi.org/10.5194/amt-11-5673-2018, 2018.