Cover Letter

Dear editor,

We have carefully addressed the thoughtful comments of the reviewers and believe that our manuscript is now much improved and ready for publication in ACP. In our paper, we present a methodology to infer CO_2 emissions from power plants using satellite observations of co-emitted NO_2. Reliable estimates of emissions of this important climate gas are necessary for predicting climate change and developing effective mitigation strategies.

Our work is timely as 1) it is currently not feasible to infer CO_2 emissions directly from satellite observations of CO_2 with current sensors, and 2) CO_2 emissions are not reliably measured (at stack) and reported for most power plants around the world. We demonstrated our methodology on eight US power plants. Though we were limited by current (OMI) sensor capabilities, we fully expect that our methodology will be more broadly applied to global power plants using improved NO_2 data from new and upcoming sensors (TROPOMI, TEMPO), which have improved signal-to-noise, finer spatial resolutions, etc.

I am looking forward to hearing from you.

Best regards,
Fei

Anonymous Referee #1
General comments
The manuscript presents a methodology to derive CO_2 emissions using satellite-based NO_2 retrievals from OMI instrument. The topic is very interesting as not many studies have successfully attempted space-based CO_2 emission estimation (while much more common is the top-down emission estimation for short-lived gases such as NO_2) and most of the previous studies only derive emissions for a few sites in the world. The results could be a good addition to the existing literature on the subject but I feel this work still does not dramatically improve what was achieved in previous studies in terms of emission estimation from CO_2 point sources. The methodology is reasonable but more effort should be put in proving how this approach could be extended to more than the 8 point sources analysed in the manuscript.
Therefore I would suggest to provide some sort of recommendations (or criteria) on how to apply the same approach to other point sources depending on the characteristics of the power plants. One possibility could be to test the approach on a few other cases outside US in addition to Matimba in order to illustrate the potential differences.
The manuscript can be published after addressing this issue and the following comments.

Response: We thank Referee #1 for the thoughtful comments. We have added Figure 8 to compare the US ratios derived in this study with the ratios for other countries from a bottom-up emission database. We have added Figure 9 to further clarify how to apply our approach to power plants outside the US. We have added a new subsection 3.3.1 to discuss the addition.

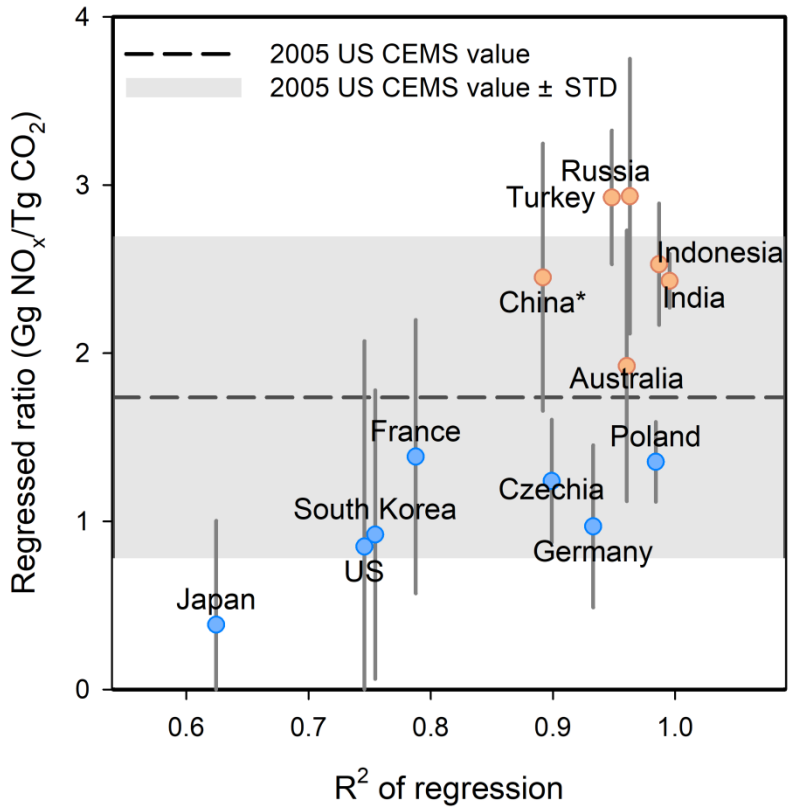

Figure 8 Comparison of the regressed NO_x to CO_2 emission ratios derived from the global power emissions database (GPED) for different regions versus the correlation coefficient of the regression. The blue and red circles denote regions that are subject to more strict standard for NO_x emissions from power plants (i.e., a NO_x ELV of 200 mg/m^3 or less) and other regions, respectively. Y axis: the slope of the regression of the NO_x to CO_2 emissions with an assumed y-intercept of zero. Error bars show the standard deviations for the NO_x to CO_2 emission ratios for individual power plants. X axis: correlation coefficient of the regression. The dashed line represents 2005 US $ratio_{regressed}^{CEMS}$ for bituminous coal derived in this study. The grey shadow represents 2005 US $ratio_{regressed}^{CEMS}$ $\pm$ standard deviation.

*China switched from being a less strict country to a more strict country in 2014, when most coal-fired power plants in China were required to comply with its new emission standards (GB13223-2011).

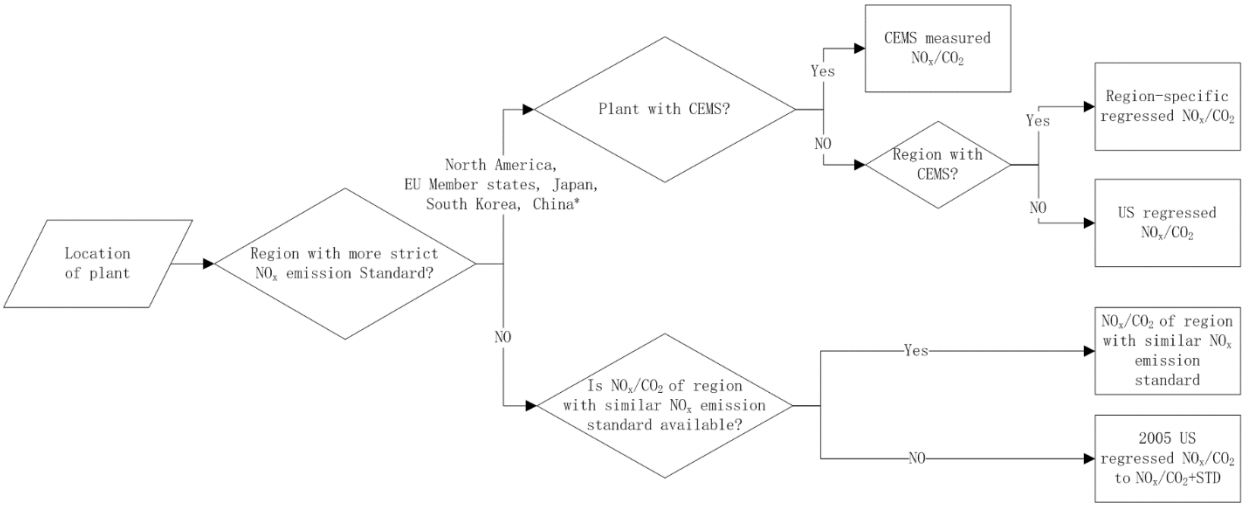

Figure 9 Schematic of our methodology to estimate the NO_x to CO_2 emission ratios for power plants outside the US.

*China switched from being a less strict country to a more strict country in 2014, when most coal-fired power plants in China were required to comply with its new emission standards (GB13223-2011).

To better place the importance of our work into context, we added the following paragraph to the conclusions:

"We found that it is feasible to infer CO_2 emissions from satellite NO_2 observations, but limitations of the current satellite data (e.g., spatio-temporal resolution, signal-to-noise) only allow us to apply our method to eight large and isolated U.S. power plants. Looking forward, we anticipate that these limitations will diminish for the recently launched (October 2017) European Union Copernicus Sentinel 5 precursor TROPOMI, and three upcoming (launches expected in the early 2020s) geostationary instruments (NASA TEMPO; European Space Agency and Copernicus Programme Sentinel-4; Korea Meteorological Administration Geostationary Environment Monitoring Spectrometer, GEMS), which are designed to have superior capabilities to OMI. For example, higher spatial and temporal resolutions will likely improve the estimation

of NO_x emissions as well as allow for the separation of more power plant plumes from nearby sources, thus increasing the number of power plants available for analysis. Therefore, future work will be to apply our method to these new datasets, especially after several years of vetted data become available. Additional future work will include applying our ratio-regression method to other regions of the world with reliable CEMS information, such as Europe, Canada and, more recently, China, to develop a more reliable and complete database with region-specific ratios."

Specific comments

1. P2 L33 -> There is a recent update to this paper where the anomalies are calculated on global scale and also TROPOMI data are used for comparison on local scale. You might want to add this as well in your intro: Hakkarainen, J.; Ialongo, I.; Maksyutov, S.; Crisp, D. Analysis of Four Years of Global XCO2 Anomalies as Seen by Orbiting Carbon Observatory-2. Remote Sens. 2019, 11, 850.

Here also another work it might be worth mentioning: Wang, S., Zhang, Y., Hakkarainen, J., Ju, W., Liu, Y., Jiang, F., & He, W. (2018). Distinguishing anthropogenic CO_2 emissions from different energy intensive industrial sources using OCO-2 observations: A case study in northern China. Journal of Geophysical Research: Atmospheres, 123, 9462–9473. https://doi.org/10.1029/2018JD029005

Response: We thank for the comments. We have added both references in the introduction of the revised manuscript.

2. P7 L19-20 "We assume the NO_x to CO_2 emission ratio of Matimba is on the upper end of the US values, considering that it is not equipped with any NO_x control devices, even low-NO_x burners which are widely installed in US power plants" This step is quite critical if you think about extending the method to other sources. You are basically saying that you have to know already something on the source before applying the method. . . how do you expect to make this choice for other sources? Please comment.

Response: The aim of the method developed by this study is to simplify the information needed to derive the NO_x to CO_2 ratio. The basic information needed for the method is generally available. We have clarified this in Section 3.3.1, as follows:

"The application of the method contributes to simplifying the information needed to derive a reasonable NO_x to CO_2 emission ratio. In a bottom-up approach, we often need many details including coal type, coal quality, boiler firing type, NO_x emission control device type, and operating condition of boiler and emission control device when calculating NO_x and CO_2 emissions. As demonstrated in Section 2.2, the method developed in this study can derive a reasonable estimate of the ratio for power plants without post-combustion NO_x control device by merely given coal type. Even for regions without reliable emission information, the information on coal type, particularly for large power plants, are very likely publicly available. For power plants installing post-combustion NO_x control technology, we additionally require the removal efficiency of the device to derive the ratio. The removal efficiency of post-combustion NO_x control devices is usually directly reported, as the operation of such devices is very expensive and is expected to be subject to strict quality control and assurance standards. "

3. Fig. 8 How do your emission estimates for Matimba compare with Reuter 2019 estimate?

Response: Our estimate for Matimba (including the nearby Medupi which has operated since 2015) is comparable to Reuter 2019 estimate. Our estimate is 1.9–3.0 Gg/h for 2016* (i.e., the period from 2015 to 2017); Reuter 2019 estimate is 3.5±0.8 Gg/h for 2018. It should be noted that the Medupi power plant started operation in 2015 with limited capacity and that it still has not reached its nominal capacity. Therefore, it is no surprise that our estimate is lower than Reuter 2019 estimate. We have added Reuter 2019 estimate in Figure 11 and the related discussion in Section 3.3.2, as follows:

"Figure 11 shows $E_{CO_2}^{Sat}$ derived in this study and other independent estimates reported in the literature, including two top-down (Nassar et al., 2017; Reuter et al., 2019) and three bottom-up estimates (Wheeler and Ummel, 2008; Tong et al., 2018; Oda et al., 2018). Despite the uncertainties associated with each of these methods, the CO_2 emissions estimates agree reasonably well."

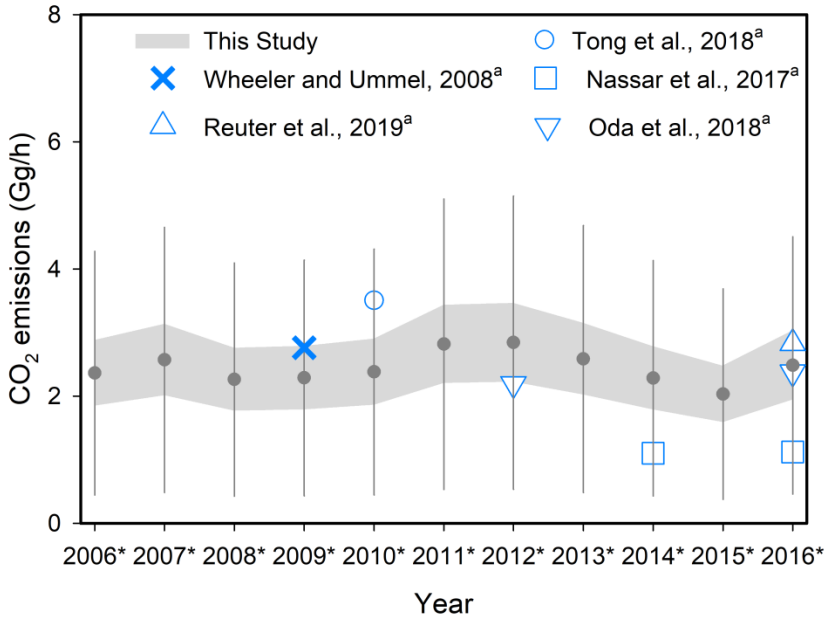

Figure 11 Comparison of $\mathbf{E_{CO_2}^{Sat}}$ (Gg/h) derived in this study with existing estimates for the Matimba power plant during 2005 to 2017. $\mathbf{E_{CO_2}^{Sat}}$ is inferred based on the NO_x to CO_2 emissions ratio ranging from $\boldsymbol{ratio_{regressed}^{CEMS}}$ to $\boldsymbol{ratio_{regressed}^{CEMS}}$ + standard deviation of ratio. The upper and lower grey bands denote the emissions inferred from $\boldsymbol{ratio_{regressed}^{CEMS}}$ and $\boldsymbol{ratio_{regressed}^{CEMS}}$+ standard deviation of ratio, respectively. The grey dots and error bars show the mean of the upper and lower grey bands and their uncertainties, respectively.

[a]Emissions are estimated for 2009 by Wheeler and Ummel (2008); for 2010 by Tong et al. (2018); for 2014 and 2016 by Nassar at al. (2017); for 2016 by Reuter et al. (2019); and for 2012 and 2016 by Oda at al. (2018).

4. P11 L25 This paper in now published: Reuter, M., Buchwitz, M., Schneising, O., Krautwurst, S., O'Dell, C. W., Richter, A., Bovensmann, H., and Burrows, J. P.: Towards monitoring localized CO2 emissions from space: co-located regional CO_2 and NO_2 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.

Response: Thanks for pointing out this. We have updated the reference in the revised manuscript.

5. Sect. 2.2 Is there any other dataset in addition to EPA's CEMS you could verify these ratio with?

Response: EPA's CEMS has been widely used to develop emission inventories. To the best of our knowledge, all the widely-used regional and global bottom-up emission inventories adopt EPA's CEMS to estimate NO_x and CO_2 emissions for US power plants. To the best of our knowledge, there is no independent dataset available to verify the derived ratios for the US power plants.

Technical comments
P3 L20 ". . .plants.As discussed" there is a space missing here

Response: Thanks for pointing out this. We have updated it in the revised manuscript.

P7 L11 I would change the title with "Application to Matimba power plant" or something like that more specific

Response: Thanks. We have changed the title for section 3.3.2 accordingly.

Anonymous Referee #2

Liu et al. describe a method to estimate CO_2 emissions from power plants using satellite observations of tropospheric NO_2 columns. The method involves the estimation of NO_x emissions using a top-down approach previously developed by the authors and estimation of CO_2 emissions by applying a NO_x/CO_2 emission ratio derived from direct stack emission measurements of both gases. The topic of the manuscript is important and relevant in the context of the ongoing development of the global emission monitoring system intended to support the elaboration of climate control and mitigation strategies. Although the idea to use satellite NO_2 measurements to constrain CO_2 emissions from fossil fuel burning is not new, application of this approach to specifically power plant emissions is a step forward. Another new point of the study is the analysis of the relationship between NO_x and CO_2 emissions from different types of coal-fired power plants in the US. That said, I keep wondering whether and how the method proposed in this manuscript can be proven useful in any scientific or practical applications. The weak points of the manuscript and my suggestions to the authors are outlined in my comments below.

Response: We thank Referee #2 for the thoughtful comments, which we address carefully below.

Major comment

I find that the manuscript lacks clear logic in presenting the ideas and results of the authors. Specifically, while the main focus in Section 2 ("Method") is given to the analysis of the CEMS stack measurements in the US in the period from 2005 to 2017, it is not explained and justified how the outcome of this analysis can be used for applications outside of the US. Such possible applications are illustrated in the manuscript (in Sect. 3.3) by the example of only one power plant (Matimba), for which the authors use the NO_x/CO_2 emission ratio estimated only for 2005 and even argue that this estimate (based on the US data) is not directly applicable to the Matimba plant. Furthermore, if the "regressed" estimates of the NO_x/CO_2 emission ratio are not directly applicable to power plants outside of the US, the application of these approximate estimates to the selected 8 power plants inside of the US (presumably to test the method) seems to be pointless, as the CEMS measurements provide accurate direct estimates of the NO_x/CO_2 emission ratio for any power plant in the US. As for the Matimba power plant, a reasonable alternative to using the CEMS measurements would be to get a corresponding estimate of the NO_x/CO_2 emission ratio from the ODIAC inventory. Therefore, in the present form, the discussion and evaluation of the method is very confusing and, to some extent, misleading. In this respect, I recommend that the authors illustrate the potential of their method and the usefulness of the analysis of the US CEMS data by considering a few more power plants outside of the US (e.g., in China), paying special attention to the accuracy of the estimates of the NO_x/CO_2 emission ratio based on the US CEMS data versus the accuracy of corresponding estimates that can be obtained directly from available data of global and regional emission inventories.

Response: We address the major comment as below.

- The significance of the method validation for US power plants:

In our study, we investigate the feasibility of using satellite data of NO_2 to infer CO_2 emissions, which could serve as a complementary verification of bottom-up inventories or be used to supplement these inventories.

We first apply our methodology to U.S. power plants, which have accurate CEMS emissions. We systematically identify sources of variation (i.e., coal type and type of NO_x control device). The

high degree of accuracy of the U.S. CEMS data allows us to verify whether our methodology is feasible or not. In short, we found that it is feasible, but limitations of the current satellite data (e.g., spatio-temporal resolution, signal-to-noise) only allow us to apply our methodology to eight power plants.

Looking forward, we anticipate that current (e.g., TROPOMI) and future sensors (e.g., TEMPO, Sentinel-4, GEMS) will reduce the limitations of the satellite data, especially after their time records have lengthened, allowing us to apply our methodology to more the US and world power plants.

We have clarified this in the revised abstract, introduction and conclusion.

- The potential application of the method and the US ratio derived in this study:

CEMS measurements are available for some power plants in the US, Canada, European Union (EU) member states, Japan, South Korea, and, more recently, China. However, there is still a significant number of power plants in those countries without CEMS technology, particularly for CO_2 measurements. For example, EU member states do not require power plants to use CEMS for CO_2 reporting and the majority of plants in the EU therefore reports CO_2 emissions based on emission factors (Sloss, 2011). Therefore, we recommend applying our method described in Section 2.2 to infer region-specific ratios for those power plants. The method developed in this study provides a simplified but reliable method to determine the ratios for those power plants.

Many or most power plants in South America, Africa, and Asia (minus China) do not report CEMS measurements at all or their observations are of questionable quality. Therefore, bottom-up emission inventories for NO_x and CO_2 from these countries are highly uncertain, confounding national and international efforts to design effective climate mitigation strategies. We have added a new subsection 3.3.1 to discuss how to apply the ratios derived in this study to other regions.

As suggested, we added the comparison of the ratios derived in this study with those in the global coal-fired power plant emissions database (GPED) in Section 3.3.1. GPED is the only publicly available bottom-up emission database reporting both NO_x and CO_2 emissions for individual power plants all over the world. The US values show reasonable agreement with other countries' values identified by GPED. The details are as follows:

"Figure 8 shows the NO_x to CO_2 emission ratios for 2010 from the global power emissions database (GPED; Tong et al., 2018), which is the only publicly-available bottom-up emission database that reports both NO_x and CO_2 emissions for individual power plants for every country. All countries with over 30 coal-fired power plants in GPED are shown in Figure 8. Not surprisingly, countries with more strict standards in place for NO_x emissions from power plants (i.e., NO_x emission limit value (ELV) < 200 mg/m^3; hereafter referred to as "more strict countries") have smaller NO_x to CO_2 ratios (i.e., 1.0 versus 2.5 on average) than countries with less strict standard (i.e., NO_x ELV > 200 mg/m^3; hereafter referred to as "less strict countries"). Additionally, the correlation coefficients are smaller for more strict countries (i.e., 0.82 on average) as compared to less strict countries (i.e., 0.96 on average), because power plants in more strict countries are more likely to have installed post-combustion NO_x control systems, which likely lowered $ratio_y^{CEMS}$, similar to what occurred in the US over our analysis period (Section 2.2.2).

We further compare the 2005 US $ratio_{regressed}^{CEMS}$ in Table 1 with the GPED NO_x to CO_2 emission ratios for less strict countries. We chose the 2005 value for comparison based on the following considerations. In 2005, the US EPA issued the Clean Air Interstate Rule (CAIR) to address the interstate transport of ozone and fine particulate matter pollution for eastern US states, which reduced NO_x emissions and, thus, NO_x to CO_2 ratios ($ratio_y^{CEMS}$). However, similar comprehensive control strategies have not been adopted

in less strict countries. In this way, the 2005 values are expected to show better consistency with the NO_x to CO_2 ratios of less strict countries than values for more recent years. Note that the GPED database does not give information on ratios by coal type. Therefore, we use $ratio_{regressed}^{CEMS}$ for bituminous coal, which is the most widely used coal type in coal-fired power plants in most countries.

The ratios for individual power plants in less strict countries tend to be larger than the US $ratio_{regressed}^{CEMS}$ for 2005, considering that power plants in those countries may not be equipped with any NO_x control devices or even low-NO_x burners, a technology which is widely installed in US power plants with and without post-combustion NO_x control devices. Most ratios range from US 2005 $ratio_{regressed}^{CEMS}$ to 2005 $ratio_{regressed}^{CEMS}$ + standard deviation (Figure 8). It is no surprise that some less strict countries have ratios higher than this range, which also occurs for some US power plants without post-combustion emission controls (Figure 4). However, there are considerable uncertainties in the GPED database given the scarcity of reliable emissions information in less strict countries. For example, the GPED NO_x and CO_2 emissions estimates for Turkey and Russia, which are outliers in Figure 8, are subject to more assumptions and, thus, larger uncertainties than countries with high-quality country-specific emission data, such as China, which has a high-resolution emissions database (CPED; Liu et al., 2015), and India, which has a database developed by Argonne National Laboratory (Lu et al., 2011).

Figure 9 shows a schematic of our methodology to estimate the NO_x to CO_2 emission ratios for power plants outside the US. We adopt different approaches for more and less strict countries. More strict countries, including Canada, European Union (EU) member states, Japan, South Korea, and, more recently, China, usually use CEMS to monitor emissions, particularly from the largest emitters. For power plants with CEMS measurements for both NO_x and CO_2 emissions, it is straightforward to use the measured ratios. However, there is still a significant number of power plants in those countries without CEMS technology, particularly for CO_2 measurements. For example, EU member states do not required power plants to use CEMS for CO_2 reporting and the majority of plants in the EU therefore reports CO_2 emissions based on emission factors (Sloss, 2011). Therefore, we recommend applying our method described in Section 2.2 to infer region-specific ratios for those power plants. The US $ratio_{regressed}^{CEMS}$ could be a less accurate, but reasonable approximation when no CEMS data are available, considering those countries share similar NO_x ELVs for power plants as the US. For less strict countries, we recommend using the 2005 US values by coal type when ratios from countries with similar NO_x emission standard are not available. We also recommend assigning a range from 2005 $ratio_{regressed}^{CEMS}$ to 2005 $ratio_{regressed}^{CEMS}$ + standard deviation, instead of a fixed value, to the ratio for inferring CO_2 emissions, considering the knowledge on ratios from those regions are too few to narrow the constraint.

As demonstrated in Section 2.2, our method presented in this study provides a reasonable estimate of the ratio for power plants without post-combustion NO_x control devices with only knowing coal type. Even for regions without reliable emission information, the information on coal type, particularly for large power plants, are very likely publicly-available. For power plants that install post-combustion NO_x control technology, we additionally require the removal efficiency of the device to derive the ratio. The removal efficiency of post-combustion NO_x control devices is usually directly reported, as the operation of such devices is very expensive and is expected to be subject to strict quality control and assurance standards. In contrast to bottom-up approaches, many details are required, including coal type, coal quality, boiler firing type, NO_x emission control device type, and operating condition of boiler and

emission control device, when calculating NO_x and CO_2 emissions."

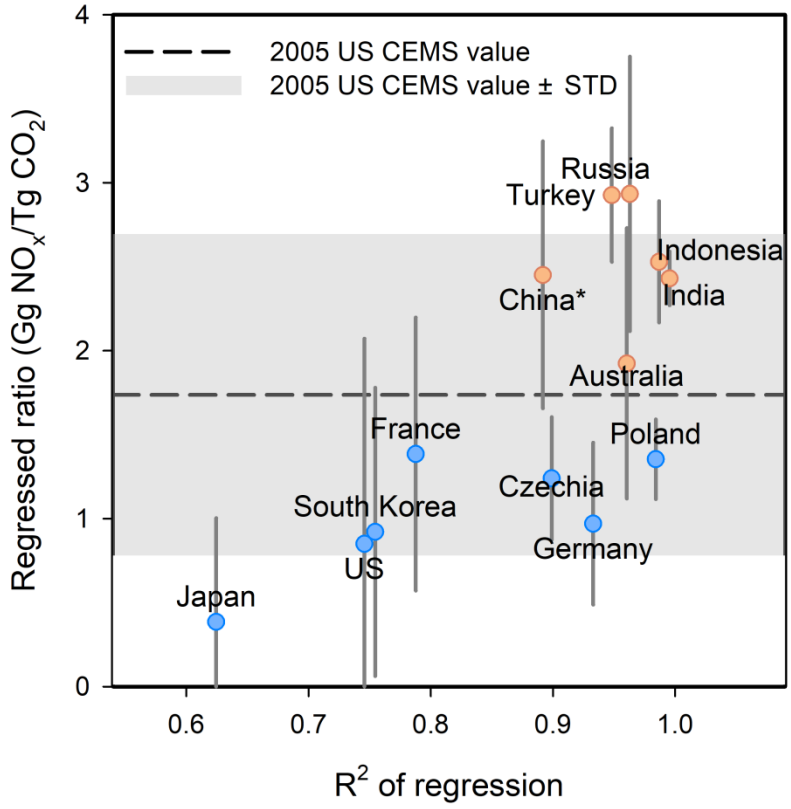

Figure 8 Comparison of the regressed NO_x to CO_2 emission ratios derived from the global power emissions database (GPED) for different regions versus the correlation coefficient of the regression. The blue and red circles denote regions that are subject to more strict standard for NO_x emissions from power plants (i.e., a NO_x ELV of 200 mg/m^3 or less) and other regions, respectively. Y axis: the slope of the regression of the NO_x to CO_2 emissions with an assumed y-intercept of zero. Error bars show the standard deviations for the NO_x to CO_2 emission ratios for individual power plants. X axis: correlation coefficient of the regression. The dashed line represents 2005 US $ratio_{regressed}^{CEMS}$ for bituminous coal derived in this study. The grey shadow represents 2005 US $ratio_{regressed}^{CEMS}$ ± standard deviation.

*China switched from being a less strict country to a more strict country in 2014, when most coal-fired power plants in China were required to comply with its new emission standards (GB13223-2011).

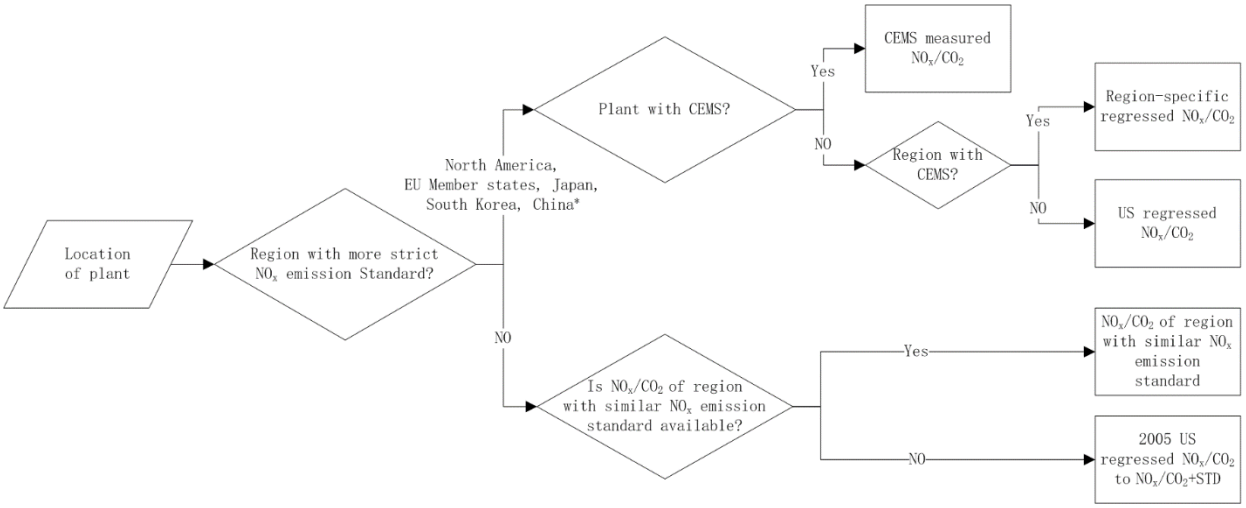

Figure 9 Schematic of our methodology to estimate the NO_x to CO_2 emission ratios for power plants outside the US.

*China switched from being a less strict country to a more strict country in 2014, when most coal-fired power plants in China were required to comply with its new emission standards (GB13223-2011).

- The recommendation of using the ODIAC inventory to derive the ratios.

We agree that ODIAC is a great source for CO_2 emissions. However, it does not provide NO_x emissions. It is not practical to calculate the ratios based on ODIAC.

Specific comments

p.2, l.16-18: I believe that the narrow swath of the OCO-2 sensor is not the main reason for the limitations of the novel and promising method proposed by Reuter et al. (2019). I suggest that the authors provide a more extensive and accurate discussion (not necessarily in Introduction) of the advantages and disadvantages of their approach with respect to that of Reuter et al. (2019).

Response: We have added the discussion in the revised introduction, as follows:

"More recently, the co-located regional enhancements of CO_2 observed by OCO-2 and NO_2 observed by TROPOMI were analysed to infer localized CO_2 emissions for six hotspots including one power plant globally (Reuter et al., 2019). As emissions plumes are significantly longer than the swath width of OCO-2 (10km), OCO-2 sees only cross sections of plumes, which may not be sufficient to infer emission strengths. Because power plant emissions can have substantial temporal variations (Velazco et al., 2011) and the cross-sectional CO_2 fluxes are valid only for OCO-2 overpass times, the cross-sectional fluxes may not adequately represent the annual or monthly averages, which are required for the development of climate mitigation strategies. In addition, the cross-sectional fluxes may not be a good approximation for emission strengths if meteorological conditions are not taken into account (Varon et al., 2018). As compared to the method proposed in this study, Reuter's method has the advantage of not requiring a priori emission information. However, there are currently no satellite instruments with a wide enough swath to allow wider application of Reuter's method. "

p.2, l.37: I recommend that the authors avoid boasting about the "novel" method here and elsewhere. Actually, the only significant new point of their method is that it is focused on a

particular source of CO_2 emissions (as noted above). A very similar method to constrain CO_2 emissions is described in previous papers (cited in this manuscript) focused on estimating fossil fuel burning CO_2 emissions in China and in Europe. Certainly, there are differences concerning the ways to estimate the NO_x emissions and NO_x/CO_2 emission ratio in the different studies, but these differences are mostly of technical nature. Furthermore, the method which was used to estimate NO_x emissions in this study is identical to that presented by the same authors in their previous papers.

Response: We have deleted the term of novel in the revised manuscript.

p.3, l.7-12: It would be useful to explain briefly why a special approximation procedure is needed to estimate a NO_x/CO_2 emission ratio while using the CMES data (i.e. why the NO_x/CO_2 emission ratio for any given power plant in the US could not be directly evaluated using the corresponding CMES measurements).

Response: CEMS measurements are available for some power plants in the US, Europe, Canada and, more recently, China. For those power plants with CEMS measurements, we agree that it is more straightforward and accurate to use the measured values. However, there is still a significant number of power plants in those countries without CEMS technology, particularly for CO_2 measurements. The method developed by this study provides a more reliable method to determine the ratios for those power plants without CEMS based on CEMS data for other plants. We have clarified this in the revised Section 3.3.1, as follows:

"More strict countries, including Canada, European Union (EU) member states, Japan, South Korea, and, more recently, China, usually use CEMS to monitor emissions, particularly from the largest emitters. For power plants with CEMS measurements for both NO_x and CO_2 emissions, it is straightforward to use the measured ratios. However, there is still a significant number of power plants in those countries without CEMS technology, particularly for CO_2 measurements. For example, EU member states do not require power plants to use CEMS for CO_2 reporting and the majority of plants in the EU therefore reports CO_2 emissions based on emission factors (Sloss, 2011). Therefore, we recommend applying our method described in Section 2.2 to infer region-specific ratios for those power plants. The US $ratio_{regressed}^{CEMS}$ could be a less accurate, but reasonable approximation when no CEMS data are available, considering those countries share similar NO_x ELVs for power plants as the US."

P3. l.21. It is quite unusual and inconvenient that the first figure ever mentioned in the manuscript is Figure 5 (instead of Figure 1). The order of the figures should be corrected.

Response: Thanks. We have reordered the figures in the revised manuscript.

p.3, l.29-32: The authors should explain the origin and significance of the value "1.32". Would their estimates be less accurate if they assumed that the NO_x/NO_2 ratio equals, say, to 1.3? Further, do the authors imply that if one had a way to measure the NO/NO_2 ratio around any power plant anywhere in the world with a spatial resolution of 13 km×24 km, then the measured NO_x/NO_2 ratio would be exactly 1.32? Wouldn't the NO_x/NO_2 ratio actually strongly vary from

site to site and would depend on the ozone level (which is frequently not determined by local pollution sources) and the age of the plume? Doesn't the fact that the estimates of the NO_x lifetime inferred from satellite measurements vary across the 8 power plants within almost a factor of 2 (according to Table 2) mean that OH (and therefore O_3) levels are quite different in plumes from different power plants? Overall, I believe that the uncertainty associated with the estimation of the NO_x/NO_2 ratio should be carefully discussed and evaluated (perhaps, using a chemistry transport model). A brief and superficial discussion of this important point in Liu et al. (2016) is certainly insufficient.

Response: The number of 1.32 used for scaling up the NO_2 to NO_x is based on the typical assumptions made in the section 6.5.1 of Seinfeld and Pandis (2006) for "typical urban conditions and noontime sun" following the recommendation by Beirle et al. (2011). We agree that the NO/NO_2 ratio might vary locally. But these local variations are not expected to be significant over spatial scales of ~100−200 km and annual temporal averaging. We included increased uncertainty of the NO_x/NO_2 ratio from 10% to 20% when calculating the overall uncertainties. We recognize that uncertainties resulting from the NO_x/NO_2 ratio may be better understood when more direct measurements are available in the future. We have clarified this in the Section 3.2 of the revised manuscript, as follows:
"The number of 1.32 used for scaling the NO_2 to NO_x ratio is based on assumptions presented in section 6.5.1 of Seinfeld and Pandis (2006) for "typical urban conditions and noontime sun". Note that conditions are quite similar in this study because of the overpass time of OMI close to noon, the selection of cloud-free observations, the focus on the ozone season, and the focus on polluted regions. A case study of CTM simulations shows an identical value of 1.32 for Paris in summer (Shaiganfar et al., 2017). The simulated NO_x/NO_2 ratio at the OMI overpass time within the boundary layer (up to 2 km) in a chemistry–climate model, EMAC (Jöckel et al., 2016), was 1.28 ± 0.08 for polluted ($NO_x > 1\times10^{15}$ molec cm^{-2}) regions for the July 1, 2005, and 1.32 ± 0.06 on average for the ozone season. However, the coarse grid of EMAC ($2.8^\circ \times 2.8^\circ$ in latitude and longitude) may not capture the true range of variation of the NO_x/NO_2 ratio. Therefore, we assumed an uncertainty of 20% arising from the NO_x/NO_2 ratio, double than the standard deviation of the EMAC ratio. "

Table S1: The authors provided some useful supplementary information for Sect. 2.1 in Table S1, but this table is not mentioned and discussed anywhere in the manuscript.

Response: Thanks for pointing out this. We have introduced the table in the revised manuscript, as follows:
"The locations of the 8 plants are shown in Figure 1 and given in Table S1."
"The fitted lifetimes and other fitting parameters for all power plants are given in Table S1."

p.4, l.3-33: I suggest the authors provide an additional figure illustrating the NO_2 plume from the Rockport power plant along with a corresponding Gaussian fit.

Response: Thanks. We have added it as Figure 2 in the revised manuscript.

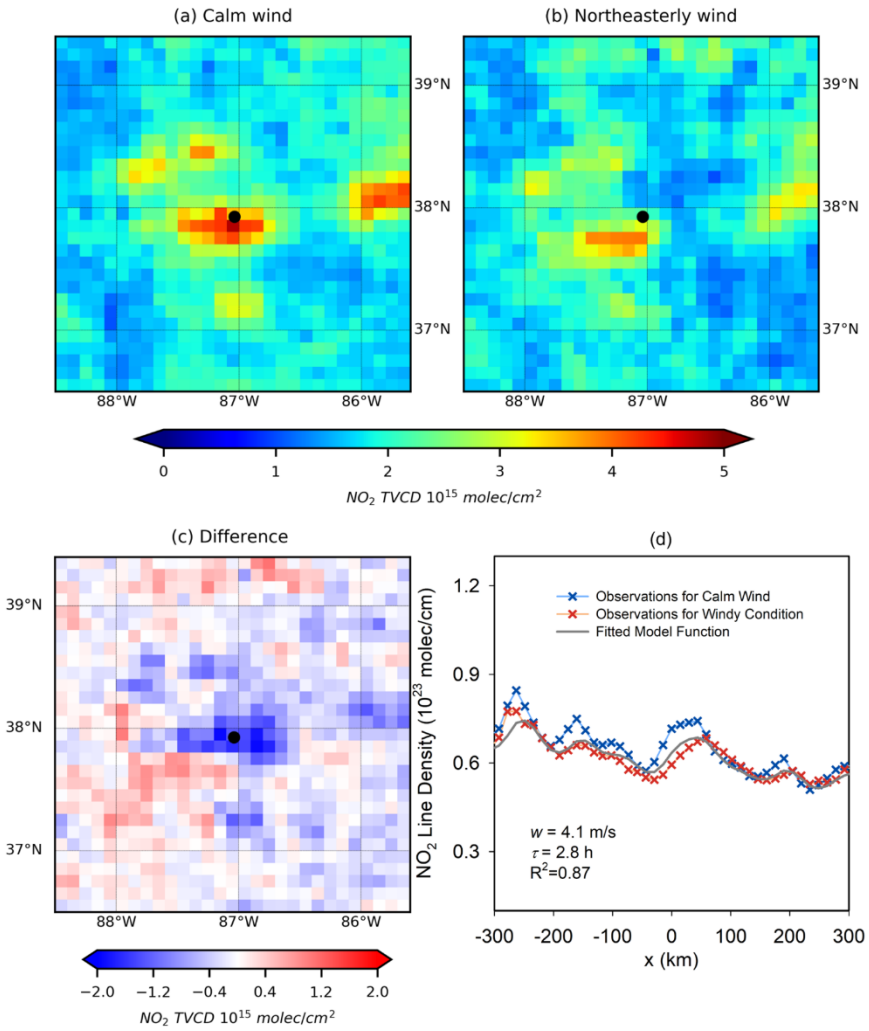

Figure 2 Mean OMI NO_2 tropospheric VCDs around the Rockport power plant (Indiana, USA) for (a) calm conditions, (b) northeasterly winds and (c) their difference (northeasterly − calm) for the period of 2005 – 2017. The location of Rockport is labelled by a black dot. (d) NO_2 line densities around Rockport. Crosses: NO_2 line densities for calm (blue) and northeasterly winds (red) as function of the distance x to Rockport center. Grey line: the fit result. The numbers indicate the net mean wind velocities (windy − calm) from MERRA-2 (w) and the fitted lifetime τ.

p.5, l.5: It would be helpful if the authors explained here what is the purpose of creating "continuous and consistent records of ratio_CEMS...". Are these records supposed to be helpful for estimating CO_2 emissions inside of the US (although accurate estimates of the NO_x/CO_2 ration are already provided by CEMS for each power plant) or outside of the US (although the applicability of the CEMS data outside of the US is very questionable)?

Response: The sentence indicates that $ratio^{CEMS}$ for plants prior to and after installing post-combustion NO_x control systems is continuous and consistent, because the estimation is based on

$ratio_{regressed}^{CEMS}$ for plants without post-combustion control systems in operation. We have deleted the terms of continuous and consistent in the revised manuscript to prevent misunderstanding.

Sect. 3.2: In my opinion, the uncertainties of the emission estimates inferred from the OMI measurements are well characterized by the standard deviations reported in Table 3. However, these "data-based" uncertainty estimates are not discussed in the manuscript. The present discussion of the uncertainties, however, looks very superficial. I suggest the authors provide a separate table (e.g., in the Supporting information) reporting the uncertainties associated with each power plant and with each individual factor contributing to the total uncertainty. Also, I wonder how a reader acquainted with the basic knowledge of the mathematical statistics is supposed to interpret the values of the uncertainty reported in this section. Do these values represent the standard deviation (that is, the confidence interval corresponding to the 68.3 percentile)? If so, does the fact that the uncertainty estimates range from 62%–96% mean that there is a significant chance that a true value of the emissions can be below zero (assuming that the error distribution is Gaussian)? My suggestion is to consider reporting the so huge uncertainties in terms of the geometric standard deviation (thus assuming that the error distribution is log-normal).

Response: We agree that the standard deviation reported in Table 3 is a good indicator of the uncertainty. We also calculate the geometric standard deviation of the difference between $E_{CO_2}^{CEMS}$ and $E_{CO_2}^{Sat}$ from 2006* to 2016* for individual power plants in Table S2 as an alternative measure to reflect the uncertainty following the suggestion of the reviewer. In the revised manuscript, we have added the discussion on this "data-based" uncertainty analysis, as follows:

"The mean and the standard deviation of the relative differences between $E_{NO_x}^{CEMS}$ and $E_{NO_x}^{Sat}$, and $E_{CO_2}^{CEMS}$ and $E_{CO_2}^{Sat}$ for all eight power plants provide a good alternative measure of uncertainties (Table 3). The relative differences are rather small, which are 0% ±33% and 8% ±41% (mean ± standard deviation) for NO_x and CO_2, respectively. We additionally calculate the geometric standard deviations (GSDs) of the difference between $E_{CO_2}^{CEMS}$ and $E_{CO_2}^{Sat}$ from 2006* to 2016* for individual power plants in Table S2. The small values of GSDs ranging from 1.07 to 1.31 further improve our confidence in the accuracy of the derived emissions in this study."

We have added a separate Table S2 to list the contributors to the overall uncertainties as suggested by the reviewer. We report the derived uncertainties as a 95% confidence interval (CI). Note that we adjust our uncertainty estimates for some contributors. We increased the uncertainty of the NO_x/NO_2 ratio from 10% to 20% (see response to the comments on the NO_x/NO_2 ratio). We decreased the uncertainty arising from the variations of fitted lifetimes by wind direction from 40% to 20%, because the average of the standard deviation of lifetimes for all wind directions decreased from 40% in Liu et al. (2016) to 20% in this study. The details are given in section 1 of the supplement, as follows:
"The uncertainty analysis is similar to the procedure described in our previous work (Liu et al., 2016), based on the fit performance and the dependencies on the a priori settings as determined in sensitivity studies. We report the derived uncertainties as a 95% confidence interval (CI). Here we briefly list the sources of uncertainties and how they are quantified. Further details are provided in Section 3 of the Supplement of Liu et al. (2016). In summary, we conclude that:

- *Choice of integration and fit intervals:* Uncertainties arising from the choice of integration and fit intervals are about 10% for the lifetime and 20% for the total NO_2 mass, respectively, based on our sensitivity analysis by changing integration and fit intervals.
- *Fit errors:* The fit errors expressed as 95% confidence interval (CI) are derived from the least-squares fit routine directly for individual sources. They are typically on the order of 10% for both lifetime and total NO_2 mass, both of which are propagated into the uncertainty of $E_{NO_x}^{Sat}$. In addition, the standard deviation of fitted lifetimes for all wind direction sectors is regarded as a measure of uncertainty to reflect the reliability of lifetimes, which is 20% on average for all power plants.
- *Wind fields:* The uncertainty associated with the wind data is 30%. The choice of wind layer height and the uncertainties of wind fields themselves contribute to the overall uncertainty.
- The derived NO_x emissions are affected by the uncertainty of the NO_2 tropospheric VCDs (~30%) and the NO_x/NO_2 ratio (~20%).
- Effects of a possible systematic change of NO_2 tropospheric VCDs from calm to windy conditions result in an uncertainty of ~10%.
- $ratio_{regressed}^{CEMS}$ contributes to an uncertainty of 15%.
- For power plants with post-combustion NO_x control devices, an additional uncertainty of 20% comes from the predicted NO_x removal efficiency of the devices.

The uncertainties of each contributor for individual power plants are listed in Table S2. We assume that their contributions to the total uncertainty are independent and define the total uncertainty as the root of the quadratic sum of the aforementioned contributions."

p.7, l.19,20: If the authors believe that the NO_x/CO_2 emission ratio at Matimba is on the upper end of the US values, then perhaps they should have used a maximum value of the NO_x/CO_2 emission ratios among all of the US power plants without NO_x emission control. Anyway, it is not clear how the standard deviation of ratio_regressed was evaluated? Is it the standard deviation of the slope of a linear fit or the standard deviation of the original NO_x/CO_2 emission ratios from the CMES data?

Response: We assume the NO_x to CO_2 emission ratio of Matimba is on the upper end of the US values, considering South Africa has not implemented improvements in boiler operations to decrease the ratio, such as optimizing furnace design and operating conditions, as in the US. We thus use the ratio for year 2005, instead those for more recent years to infer CO_2 emissions for the entire period. The standard deviation is that of the NO_x/CO_2 emission ratios for individual power plants from CEMS. We believe the ratio of Matimba is more likely to range from 2005 $ratio_{regressed}^{CEMS}$ to 2005 $ratio_{regressed}^{CEMS}$ + standard deviation, instead of being 2005 $ratio_{regressed}^{CEMS}$, considering that it is not equipped with any NO_x control devices, even low-NO_x burners which are widely installed in US power plants. But we don't use the maximum value of the ratio in this study, because it may be related with some plant-specific operating conditions, which is not applicable to other plants. We have clarified how to apply the ratios derived in this study to other regions in Section 3.3.1.

p.7, l.29-31: According to Reuter et al. (2019), the CO_2 emission estimates for the Matiba power plant are available also from the ODIAC inventory. The authors could consider using the corresponding estimates for comparison.

Response: Thanks. We have added the estimates from ODIAC in Figure 11 of the revised manuscript. $E_{CO_2}^{Sat}$ derived in this study shows reasonable agreement with the ODIAC.

Conclusions: This section looks unusually short for ACP. Furthermore, instead of providing a clear and logical summary of the major findings of the study, the authors preferred to speculate about possible future developments of their method. Accordingly, I believe this section needs to be re-written and significantly extended.

Response: We have extended the conclusion substantially to provide a summary of the major findings, as follows:

"In our study, we investigated the feasibility of using satellite data of NO_2 from power plants to infer co-emitted CO_2 emissions, which could serve as complementary verification of bottom-up inventories or be used to supplement these inventories that are highly uncertain in many regions of the world. For example, our estimates will serve as an independent check of CO_2 emissions that will be inferred from satellite retrievals of future CO_2 sensors (Bovensmann et al., 2010). Currently, uncertainties in CO_2 emissions from power plants confound national and international efforts to design effective climate mitigation strategies.

We estimate NO_2 and CO_2 emissions during the "ozone season" from individual power plants from satellite observations of NO_2 and demonstrate its utility for US power plants, which have accurate CEMS with which to evaluate our method. We systematically identify the sources of variation, such as types of coal, boiler, and NO_x emission control device, and change in operating conditions, which affect the NO_x to CO_2 emissions ratio. Understanding the causes of these variations will allow for better informed assumptions when applying our method to power plants that have no or uncertain information on the factors that affect their emissions ratios. For example, we estimated CO_2 emissions from the large and isolated Matimba power plant in South Africa, finding that our emissions estimate shows reasonable agreement with other independent estimates.

We found that it is feasible to infer CO_2 emissions from satellite NO_2 observations, but limitations of the current satellite data (e.g., spatio-temporal resolution, signal-to-noise) only allow us to apply our method to eight large and isolated U.S. power plants. Looking forward, we anticipate that these limitations will diminish for the recently launched (October 2017) TROPOMI, and three upcoming (launches expected in the early 2020s) geostationary instruments (NASA TEMPO; European Space Agency and Copernicus Programme Sentinel-4; Korea Meteorological Administration Geostationary Environment Monitoring Spectrometer, GEMS), which are designed to have superior capabilities to OMI. As demonstrated in Ialongo et al. (2019), high resolution TROPOMI observations are capable of describing the spatio-temporal variability of NO_2, even in a relatively small city like Helsinki. Higher spatial and temporal resolutions will likely reduce uncertainties in estimates of NO_x emissions as well as allow for the separation of more power plant plumes from nearby sources, thus increasing the number of power plants available for analysis. Therefore, future work will be to apply our method to these

new datasets, especially after several years of vetted data become available. Additional future work will include applying our method to other regions of the world with reliable CEMS information, such as Europe, Canada and, more recently, China, to develop a more reliable and complete database with region-specific ratios. ”

Figure 2: Do the emissions shown in this figure correspond to the ozone season only? If so, this should be indicated in the figure caption. The regression coefficients could be reported only with one or two digits after the point. Is there a reason for showing a linear regression with the intercept term in the panel (c) and without the intercept in other panels?

Response: For comparison to $E_{NO_x}^{Sat}$ and $E_{CO_2}^{Sat}$, we use emissions averaged over the ozone season derived from Air Markets Program Data (available at https://ampd.epa.gov/ampd/). However, Air Markets Program Data do not provide information about each plant's boiler firing types (e.g., tangential or wall-fired boiler), NO_x control device type, fossil fuel type (with categories of coal, oil, gas and other), and coal type (with categories of bituminous, lignite, subbituminous, refined and waste coal), which are required to get reasonable ratio. Thus, we choose eGRID as the data source for Figure2. We use eGRID annual emissions in Figure 2, because eGRID does not provide CO_2 emissions specifically for the ozone season.

We have changed the regression coefficients to two digits after the point. We intent to show the linear regression without intercept in panels (a) and (b), because the regression slope was calculated requiring zero intercept for deriving $\boldsymbol{ratio}_{\boldsymbol{regressed}}^{\boldsymbol{CEMS}}$.

Figure 8: The meaning of a shaded band should be clearly explained in the figure caption. I suggest also to supply the emission estimates inferred from the OMI observations with the error bars corresponding to the mean of the standard deviations reported in Table 3.

Response: We have added the explanation for the shaded band in the revised caption, as follows: “The upper and lower grey bands denote the emissions inferred from $\boldsymbol{ratio}_{\boldsymbol{regressed}}^{\boldsymbol{CEMS}}$ and $\boldsymbol{ratio}_{\boldsymbol{regressed}}^{\boldsymbol{CEMS}}$+ standard deviation of ratio, respectively.”
We have added error bars in the revised figure.

A methodology to constrain carbon dioxide emissions from coal-fired power plants using satellite observations of co-emitted nitrogen dioxide

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Abstract. We present a ~~novel~~ method to infer CO_2 emissions from individual power plants based on satellite observations of co-emitted nitrogen dioxide (NO_2~~) and~~), which could serve as complementary verification of bottom-up inventories or be used to supplement these inventories. We demonstrate its utility on eight large and isolated US power plants, where accurate stack emission estimates of both gases are available for comparison. In the first step of our methodology, we infer nitrogen oxides (NO_x) emissions from ~~isolated~~US power plants using Ozone Monitoring Instrument (OMI) NO_2 tropospheric vertical column densities (VCDs) averaged over the ozone season (May-September) and a "top-down" approach that we previously developed. Second, we determine the relationship between NO_x and CO_2 emissions based on the direct stack emissions measurements reported by continuous emissions monitoring system (CEMS) programs, accounting for coal ~~type~~quality, boiler firing ~~type~~technology, NO_x emission control device type, and ~~changes~~any change in operating conditions. Third, we estimate CO_2 emissions ~~of the ozone season~~ for ~~a plant~~power plants using the OMI-estimated NO_x emissions and the CEMS NO_x/CO_2 emission ratio. We find that the CO_2 emissions estimated by our satellite-based method during 2005–2017 are in reasonable agreement with the US CEMS measurements, with a relative difference of 8% ± 41% (mean ± standard deviation~~) for the selected US power plants in our analysis. Total uncertainty in the inferred CO_2 estimates is partly associated with the uncertainty associated with the OMI NO_2 VCD data, so we expect that it will decrease when our method is applied to OMI-like sensors with improved capabilities, such as TROPOspheric Monitoring Instrument (TROPOMI) and geostationary Tropospheric Emissions: Monitoring Pollution (TEMPO~~). The broader implication of our methodology is that it has the potential to provide an additional constraint on CO_2 emissions from power plants in regions of the world without reliable emissions accounting. We explore the feasibility by comparing the derived NO_x/CO_2 emission ratios for the US with those from a bottom-up emission inventory for other countries and applying our methodology to a power plant in South Africa, where the satellite-based emission estimates show reasonable consistency with other ~~estimates~~independent estimates. Though our analysis is limited to a few power plants, we expect to be able to apply our method to more US (and world) power plants when multi-year data records become available from new OMI-like sensors with improved capabilities, such as the TROPOspheric Monitoring Instrument (TROPOMI) and upcoming geostationary satellites, such as the Tropospheric Emissions: Monitoring Pollution (TEMPO) instrument.

1 Introduction

Thermal power plants, particularly coal-fired power plants, are among the largest anthropogenic CO_2 emitters, contributing ~40% of energy-related CO_2 emissions globally in 2010 (Janssens-Maenhout et al., 2017). Coal-fired power

plants are expected to be one of the primary contributors of CO_2 emissions in the coming decades because of abundant world coal reserves (Shindell and Faluvegi, 2010). Therefore, it is important to accurately monitor global CO_2 emissions from power production in order to better predict climate change (Shindell and Faluvegi, 2010) and to support the development of effective climate mitigation strategies.

CO_2 emissions from power plants are typically quantified based on bottom-up approaches using fuel consumption and fuel quality, though fuel properties are not always well known, resulting in uncertainties in the estimated CO_2 emissions for individual plants (Wheeler and Ummel, 2008). Even for US power plants that are considered to have the most accurate information on fuel usage among world nations, the difference between emissions estimated based on fuel usage and those reported as part of continuous emissions monitoring systems (CEMS) programs is typically about 20% (Ackermann and Sundquist, 2008). Thus, emission estimates based on independent data sources, such as satellite observations, are a desirable complement to validate and improve the current CO_2 emissions inventories, especially in countries without CEMS data, which is the case in most of the world.

Anthropogenic CO_2 emissions have been estimated from space-based CO_2 observations, but the existing satellite CO_2 sensors are designed to provide constraints on natural CO_2 sources and sinks (Basu et al., 2013; Houweling et al, 2015), and thus their capability for monitoring anthropogenic point sources is limited (Nassar et al., 2017). Observations from sensors, including the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY; Burrows et al., 1995), Greenhouse gases Observing SATellite (GOSAT; Yokota et al., 2009), and Orbiting Carbon Observatory-2 (OCO-2; Crisp et al., 2015), show statistically significant enhancements over metropolitan regions (Kort et al., 2012; Schneising et al., 2013; Janardanan et al., 2016; Buchwitz et al., 2018; Reuter et al., 2019~~).~~; Wang et al., 2018). However, very few studies have focused on individual point sources. Bovensmann et al. (2010) and Velazco et al. (2011) presented a promising satellite remote sensing concept to infer CO_2 emissions for power plants based on the atmospheric CO_2 column distribution. Nassar et al. (2017) ~~and Reuter et al. (2019)~~ presented the ~~only~~first quantification of CO_2 emissions from individual power plants using OCO-2 observations. However, ~~due to~~because of the narrow swath (~10 km at nadir) and 16-day repeat cycle of the OCO-2 sensor, ~~their method cannot be currently applied to generate~~the number of clear-day overpasses is too small to allow for the development of a global CO_2 emissions database.

In contrast to CO_2, inferring NO_x emissions from individual power plants using satellite NO_2 column retrievals has been done with a higher degree of confidence (e.g., Duncan et al., 2013; de Foy et al., 2015). The Dutch-Finnish Ozone Monitoring Instrument (OMI) on NASA's Earth Observing System Aura spacecraft (Schoeberl et al., 2006) provides near daily, global NO_2 tropospheric ~~vertical column densities (~~VCDs~~)~~ at a spatial resolution of 13×24 km^2 (at nadir) (Levelt et al., 2006; 2018; Krotkov et al., 2017), which allows emission signals from individual power plants to be resolved. Beirle et al. (2011) first analyzed isolated large sources (i.e., megacities and the US Four Corners power plant) by averaging OMI NO_2 tropospheric VCDs separately for different wind directions, which allows ~~to determine~~for the estimation of NO_x emissions and lifetimes by fitting an exponentially modified Gaussian function. Several follow-up studies (e.g., de Foy et al., 2015; Lu et al., 2015 and Goldberg et al., 2019a) further developed this approach and inferred NO_x emissions from isolated power plants and cities. More recently, we advanced this approach for sources located in polluted areas to infer NO_x emissions for 17 power plants and 53 cities across China and the US (Liu et al., 2016; 2017).

Since NO_x is co-emitted with CO_2, NO_x emissions inferred from satellite data may be used to estimate CO_2 emissions from thermal power plants. Previous analyses estimated regional CO_2 emissions based on satellite-derived NO_x emissions and the NO_x to CO_2 emission ratios from bottom-up emission inventories (Berezin et al., 2013; Konovalov et al., 2016; Goldberg et al., 2019b) or co-located satellite retrievals of CO_2 and NO_2 (Reuter et al., 2014). Hakkarainen et al. (2016) confirmed the spatial correlation between CO_2 spatial anomalies and OMI NO_2 VCD enhancements at the regional scale using satellite observations at higher resolution. ~~More recently, the co-located regional enhancements of CO_2 observed by~~

~~OCO-2 and NO_2 observed by TROPOMI were analysed to monitor localized CO_2 emissions (Reuter et al., 2019).~~Hakkarainen et al. (2019) also showed how overlapping OCO-2 CO_2 data and data of NO_2 from the recently launched (October 2017) European Union Copernicus Sentinel 5 precursor TROPOspheric Monitoring Instrument (TROPOMI) can be used to identify small scale anthropogenic CO_2 signatures.

More recently, the co-located regional enhancements of CO_2 observed by OCO-2 and NO_2 observed by TROPOMI were analysed to infer localized CO_2 emissions for six hotspots including one power plant globally (Reuter et al., 2019). As emissions plumes are significantly longer than the swath width of OCO-2 (10 km), OCO-2 sees only cross sections of plumes, which may not be sufficient to infer emission strengths. Because power plant emissions can have substantial temporal variations (Velazco et al., 2011) and the cross-sectional CO_2 fluxes are valid only for OCO-2 overpass times, the cross-sectional fluxes may not adequately represent the annual or monthly averages, which are required for the development of climate mitigation strategies. In addition, the cross-sectional fluxes may not be a good approximation for emission strengths if meteorological conditions are not taken into account (Varon et al., 2018). As compared to the method proposed in this study, Reuter's method has the advantage of not requiring a priori emission information. However, there are currently no satellite instruments with a wide enough swath to allow wider application of Reuter's method.

In this study, we present a ~~novel~~ method to estimate CO_2 emissions from individual power plants using OMI NO_2 observations and auxiliary CEMS information ~~on~~ necessary to estimate NO_x to CO_2 emission ratios. Such estimates could serve as complementary verification of bottom-up CO_2 inventories or be used to supplement these inventories. For instance, Liu et al. (2018) used satellite data of SO_2 to identify large SO_2 sources that were missing from a bottom-up emissions inventory and created a merged bottom-up/top-down SO_2 emissions inventory. We apply our approach to US power plants, which have an exceptionally detailed CEMS database of NO_x and CO_2 emissions, in order to validate our method. Using auxiliary CEMS information, we explore the relationship between NO_x and CO_2 emissions for individual power plants, assessing variations in the ratio associated with coal quality, boiler firing type, NO_x emission control device technology, and changes in operating conditions. Understanding the causes of these variations will allow for better informed assumptions when applying our method to power plants that have no or uncertain information on the factors that affect their emissions ratios. We discuss the uncertainties and ~~limitations~~applications of our approach~~. Finally, we present the application of our method to power plants in South Africa. We discuss other~~, and the potential ~~applications in conclusion, including to other~~of NO_2 datasets from new and upcoming satellite instruments, which will improve the utility of our method for inferring CO_2 emissions from power plants around the world. Finally, we discuss future research directions.

2 Method

In this section, we present ~~a novel~~our method to infer CO_2 emissions ($E_{CO_2}^{Sat}$) from satellite-derived NO_x emissions ($E_{NO_x}^{Sat}$) for individual coal-fired power plants using the following equation:

$$E_{CO_2,y}^{Sat} = \frac{E_{NO_x,y}^{Sat}}{ratio_{i,y}^{CEMS}}, \quad (1)$$

where *i* represents coal type and *y* represents the target year. We demonstrate our method on US power plants since there are accurate CEMS stack measurements of NO_x and CO_2 emissions with which to validate $E_{CO_2}^{Sat}$. In Section 2.1, we describe how ~~to~~we estimate $E_{NO_x}^{Sat}$ from OMI NO_2 tropospheric VCD observations. In Section 2.2, we discuss how ~~to~~we estimate the ratio of NO_x to CO_2 emissions ($ratio_y^{CEMS} = E_{NO_x,y}^{CEMS} / E_{CO_2,y}^{CEMS}$) from CEMS stack measurements in the US Emissions & Generation Resource Integrated Database (eGRID; USEPA, 2018). Since post-combustion NO_x control systems, including selective noncatalytic reduction (SNCR) and selective catalytic reduction (SCR), change the ~~correlation~~relationship between

$E_{NO_x}^{CEMS}$ and $E_{CO_2}^{CEMS}$, we present separate methods to determine $ratio_y^{CEMS}$ for power plants without and with post-combustion NO_x control systems in Section 2.2.1 and Section 2.2.2, respectively. We discuss the validation of the estimated $E_{CO_2}^{Sat}$ in Section 3.

2.1 Estimating satellite-derived NO_x emissions ($E_{NO_x}^{Sat}$)

From all US coal-fired power plants, we selected 21 power plants for estimating $E_{NO_x}^{Sat}$. We chose these plants based on the magnitude of their annual emissions (i.e., $E_{NO_x}^{CEMS}$ ~~(i.e.,~~ > 10 Gg/yr in 2005) and relative isolation from other large sources ~~and relative isolation from other large sources~~ to avoid "contamination" of a power plant's NO_x ~~plumes by NO_x from other sources.~~plume. Power plants located in urban areas (i.e., within a radius of 100 km from a city ~~centers~~center), or clustered in close proximity (i.e., 50 km) with other large industrial plants ~~are~~were excluded by visual inspection using satellite imagery from Google Earth. ~~The~~We used the top 200 largest US cities (~~rank~~ranked by 2018 population as estimated by the United States Census Bureau, available at https://en.wikipedia.org/wiki/List_of_United_States_cities_by_population) ~~are used~~ to select power plants. As discussed below, we ~~are~~were able to estimate $E_{NO_x}^{Sat}$ for 8 of the 21 plants. The locations of the 8 plants are shown in Figure ~~5~~1 and given in Table S1.

We ~~follow~~followed the method of Liu et al. (2016; 2017) to estimate $E_{NO_x}^{Sat}$ for 2005 to 2017. In our analysis, we ~~use~~used OMI NO_2 tropospheric VCDs from the NASA OMI standard product, version 3.1 (Krotkov et al., 2017) together with meteorological wind information from the Modern-Era Retrospective Analysis for Research and Applications, version 2 (MERRA-2; Gelaro et al., 2017). We only ~~analyze~~analysed data for the ozone season (May-September), in order to exclude winter data, which have larger uncertainties and NO_x lifetimes are longer ~~NO_x lifetime.~~. As in our previous study (Liu et al., 2017), we calculated 1-dimensional NO_2 "line densities", i.e. NO_2 per cm, as function of distance for each wind directions separately by integration of the mean NO_2 VCDs (i.e. NO_2 per cm^2) perpendicular to the wind direction. We then~~use~~ used the changes of NO_2 line densities~~VCDs~~ under calm wind conditions (wind speed < 2 m/s below 500 m) and windy conditions (wind speed > 2 m/s) to fit the effective NO_x lifetime. We then ~~estimate~~estimated the average NO_2 total mass integrated around a power plant on the basis of the 3-year mean VCDs, in agreement with previous studies (Fioletov et al., 2011; Lu et al., 2015). The NO_2 total mass ~~is~~was scaled by a factor of 1.32 in order to derive total NO_x mass following Beirle et al. ~~(2011). The NO/NO_2 ratio might differ locally in plumes, but the influence is not expected to be dramatic on the scales of the OMI footprint (at least 13 km×24 km), considering the overpass time of OMI close to noon, the selection of cloud free observations, and the focus on the ozone season and polluted regions with generally high tropospheric ozone.~~(2011). The uncertainty associated with the NO/NO_2 ratio has been discussed in detail in Section 3 of the supplement in Liu et al. (2016). The 3-year average $E_{NO_x}^{Sat}$ ~~is~~was derived from the corresponding 3-year average NO_x mass divided by the average NO_x lifetime of the entire study period (Liu et al., 2017). Fitting results of insufficient quality (e.g., correlation coefficient of the fitted and observed NO_2 distributions <0.9) ~~are~~were excluded from this analysis, consistent with the criteria in Section 2.2 of Liu et al. (2016). This final filtering ~~leaves~~left 18 power plants, of which 8 ~~have~~had valid results for all consecutive 3-year periods between 2005 and 2017. More details of the approach are documented in Liu et al. (2017). The fitted lifetimes and other fitting parameters for all power plants are given in Table S1.

We use the Rockport power plant (37.9 °N, 87.0 °W) in Indiana to demonstrate our approach. This power plant is particularly well suited for estimating $E_{NO_x}^{Sat}$, because it is a large and isolated NO_x point source. Figure ~~1~~2 presents the NO_2 VCD map around Rockport and the fitted results. Figure 3 displays $E_{NO_x}^{Sat}$ based on 3-year mean VCDs. ~~For simplicity, the~~Each 3-year period is represented by the middle year with an asterisk (e.g., 2006* denotes the period from 2005 to 2007). For comparison to $E_{NO_x}^{Sat}$, $E_{NO_x}^{CEMS}$ ~~averaged over the period of May to September is derived~~is from Air Markets Program Data (available at https://ampd.epa.gov/ampd~~).~~/) and averaged over the period of May to September. For this particular plant,

$E_{NO_x}^{Sat}$ ~~and~~is always higher than $E_{NO_x}^{CEMS}$ ~~show positive biases (of varying magnitude)~~ during the entire period, except the last two years. The coefficient of determination for the entire period is R^2=0.68. The relative differences for individual 3-year means (defined as $(E_{NO_x}^{Sat} - E_{NO_x}^{CEMS})/E_{NO_x}^{CEMS}$) range from -20% to 41%, because of the uncertainties of $E_{NO_x}^{Sat}$ as discussed in Section 3.2. Both datasets present a declining trend from 2012*. The total declines of 45% and 26% since 2012* in $E_{NO_x}^{Sat}$ and $E_{NO_x}^{CEMS}$ are attributed to the 25% decrease in net electricity generation for the plant. The average relative difference of $E_{NO_x}^{Sat}$ and $E_{NO_x}^{CEMS}$ for the 8 plants in this study is 0% ±33%, ranging from -58% to 72% for individual 3-year periods (Figure ~~5~~1).

2.2 Estimating NO_x to CO_2 emission ratios using CEMS data ($ratio^{CEMS}$)

We ~~determine~~determined the ~~observed~~ relationship between $E_{NO_x}^{CEMS}$ and $E_{CO_2}^{CEMS}$ for coal-fired power plants using eGRID information about each plant's net electric generation, boiler firing ~~types~~technology (e.g., tangential or wall-fired boiler), NO_x control device type, fossil fuel ~~type (with categories of~~category (i.e., coal, oil, gas and other), and coal ~~type (with categories of~~quality (i.e., bituminous, lignite, subbituminous, refined and waste coal). We ~~only use~~used data of power plants with more than 99% of the fuel burned being coal as reported in eGRID. We ~~analyze~~analyzed the relationship between $E_{NO_x}^{CEMS}$ and $E_{CO_2}^{CEMS}$ by coal type, as emission characteristics vary widely by coal type.

eGRID includes two ~~sets~~datasets of ~~emission data~~emissions for NO_x and CO_2: 1) calculated from fuel consumption data and 2) observed by stack monitoring (i.e., $E_{NO_x}^{CEMS}$ and $E_{CO_2}^{CEMS}$). Here we focus on eGRID CEMS data as $E_{NO_x}^{CEMS}$ are reported to be highly accurate with an error of less than 5% (e.g., Glenn et al., 2003). $E_{CO_2}^{CEMS}$ may have larger uncertainties than fuel-based emissions estimates ~~due to~~because of uncertainties in the calculation of flue gas flow (Majanne et al., 2015). Nevertheless, we ~~use~~used $E_{CO_2}^{CEMS}$ to relate NO_x emissions to CO_2 emissions, since the primary uncertainty of $E_{NO_x}^{CEMS}$ and $E_{CO_2}^{CEMS}$ arises from the calculation of the flue gas flow, which will cancel in $ratio^{CEMS}$.

2.2.1 Coal-fired power plants without post-combustion NO_x control systems

We ~~first~~initially limited our analysis to $E_{NO_x}^{CEMS}$ and $E_{CO_2}^{CEMS}$ from coal-fired power plants without post-combustion NO_x control systems in operation in a given year (Table 1). We find that $E_{NO_x}^{CEMS}$ and $E_{CO_2}^{CEMS}$ have a strong linear relationship (Figure ~~2~~4). In Figure ~~2a~~4a, we compare $E_{NO_x}^{CEMS}$ and $E_{CO_2}^{CEMS}$ from power plants (using bituminous coal) by boiler firing type in 2005. We use bituminous coal-fired plants for illustration, as bituminous coal is the most widely used coal in US power plants. We ~~analyze~~analyzed power plants that use cyclone or cell burner boilers separately and exclude them in Figure ~~2~~4 because they typically produce higher NO_x emissions than other boiler types (USEPA, 2009; available at https://www3.epa.gov/ttn/chief/ap42/ch01/index.html). A strong linear relationship between $E_{NO_x}^{CEMS}$ and $E_{CO_2}^{CEMS}$ is evident with excellent correlation (R^2= 0.93, N = 278), regardless of boiler firing ~~types~~type. Similar linear relationships exist for other years (e.g., year 2016 in Figure ~~2b~~4b) and other types of coal (Table 1). The slope of the regression of $E_{NO_x}^{CEMS}$ and $E_{CO_2}^{CEMS}$, $ratio_{regressed}^{CEMS}$, is assumed by setting the intercept to zero. Table 1 shows $ratio_{regressed,i,y}^{CEMS}$ by coal type and year. In Section 3.1, $ratio_{regressed,i,y}^{CEMS}$ will be applied to approximate $ratio_{i,y}^{CEMS}$ when estimating $E_{CO_2}^{Sat}$ from $E_{NO_x}^{Sat}$ for the 8 plants ~~in Section 2.1~~(Figure 1) for years before post-combustion control systems were in operation.

$ratio_{regressed}^{CEMS}$ for power plants using bituminous coal decreased from 2005 (Figure ~~2a~~4a) to 2016 (Figure ~~2b~~4b) by 31% on average because of reductions in NO_x emission factors associated with improvements in boiler operations, such as by optimizing furnace design and operating conditions. The NO_x emissions factors, defined as NO_x emission rates per net electricity generation (Gg/TW h~~)~~), declined by 33% from 2005 to 2016 (Figure ~~2c~~4c). We ~~interpolate~~interpolated

$ratio^{CEMS}_{regressed}$ to get year-specific ratios by coal type for the entire study period, as eGRID data are only available for some years (i.e., 2005, 2007, 2009, 2010, 2012, 2014 and 2016).

In addition, $ratio^{CEMS}_{regressed}$ shows significant variation by coal type and year (Figure ~~3~~5). $ratio^{CEMS}_{regressed}$ is 1.7, 1.3 and 0.91 Gg NO_x/Tg CO_2 for bituminous, subbituminous and lignite coal types in 2005, respectively. A reduction over time in $ratio^{CEMS}_{regressed}$ is observed for all coal types (Figure ~~3~~5). $ratio^{CEMS}_{regressed}$ displays a large decrease of 31%, 36% and 20% from 2005 to 2016 for bituminous, subbituminous, and lignite coal types, respectively.

2.2.2 Coal-fired power plants with post-combustion NO_x control systems

Here, we describe how we ~~create continuous and consistent records of~~estimated $ratio^{CEMS}$ for the entire study period for plants that had post-combustion NO_x control systems installed at some time during our study period, 2005–2017. The estimation is based on $ratio^{CEMS}_{regressed}$ derived in Section 2.2.1 for plants without post-combustion control systems in operation. We introduce a NO_x removal efficiency parameter, *f*, to adjust $ratio^{CEMS}_{regressed}$ for years after the installation of post-combustion control systems, $ratio^{CEMS-Estimated}$:

$$ratio^{CEMS-Estimated}_{i,y} = ratio^{CEMS}_{regressed,i,y} \times (1 - f_y) , \quad (2)$$

f is commonly measured for individual power plants to describe the performance of their post-combustion NO_x control systems. It is directly reported by some power plant databases, such as the China coal-fired Power plant Emissions Database (CPED; Liu et al., 2015). For databases that do not report *f*, like eGRID used in this study, one can estimate it for an individual power plant by first estimating the unabated emissions per electricity generation, $e_{unabated}$, which is the emission factor before the flue gas enters the post-combustion control system:

$$f_y = \frac{e_{unabated,y} - e_{CEMS,y}}{e_{unabated,y}} , \quad (3)$$

where e_{CEMS} denotes the actual emission factor in terms of CEMS NO_x emissions per net electricity generation (Gg/TW h). $e_{unabated}$ for a given year, $e_{unabated,y}$, is estimated based on the emission per electricity generation for years prior ~~to~~, *p*, to the installation of the post-combustion control system, $e_{unabated,p}$:

$$e_{unabated,y} = k_y \times e_{unabated,p} , \quad (4)$$

where the scaling factor, k_y, is used to account for the change over time in $e_{unabated}$ associated with improvements in boiler operations discussed in Section 2.2.1. k_y is calculated as the ratio of the averaged $e_{unabated}$ (i.e., the slope of the regression of NO_x emissions on electricity generation) in year, *t*, to that in year, *p*.

To assess the reliability of $ratio^{CEMS-Estimated}$, we ~~select~~selected all power plants which had post-combustion devices installed between 2005 and 2016. Figure ~~4~~6 shows a scatterplot of $ratio^{CEMS}$ (i.e., the ratio of $E^{CEMS}_{NO_x}$ to $E^{CEMS}_{CO_2}$ for individual ~~plant~~plants) and $ratio^{CEMS-Estimated}$ ~~for these power plants.~~. We ~~use~~used the NO_x emissions factor in 2005, $e_{unabated,2005}$, to predict the unabated emission factor in 2016, $e_{unabated,2016}$, following Equations (3) and (4) in order to quantify the removal efficiencies for 2016, f_{2016}. $ratio^{CEMS-Estimated}_{2016}$ is based on the estimated f_{2016} and $ratio^{CEMS}_{regressed,2016}$ from Section 2.2.1. $ratio^{CEMS}$ and $ratio^{CEMS-Estimated}$ show good correlation (R^2 = 0.64), which increases our confidence that the estimated removal efficiencies approximate the actual efficiencies ~~well~~. The slight underestimation suggested by the slope of 0.85 arises from ~~the~~ uncertainties in estimating unabated NO_x emission factors ~~(~~$e_{unabated,y}$~~)~~ using Equation (4) and thus removal efficiencies~~, (*f*),~~ which is a major source of error of $E^{Sat}_{CO_2}$ for power plants that install post-combustion NO_x control systems (see details in Section 3.2~~) contributing to the overall uncertainties of $E^{Sat}_{CO_2}$~~).

3 Results and Discussion

In Section 3.1, we present $E_{CO_2}^{Sat}$ for our eight selected power plants and, in Section 3.2, we discuss the uncertainties associated with $E_{CO_2}^{Sat}$. ~~We~~In Section 3.3, we compare the US ratios derived in this study with those from a bottom-up inventory for other regions to explore the potential of applying our method to regions outside the US. We finally apply ~~the~~our approach to one power ~~plants~~plant in South Africa, which has several independent estimates for its CO_2 emissions as presented in the scientific literature. Table 2 shows three-year means of $E_{NO_2}^{Sat}$, $E_{NO_2}^{CEMS}$, $E_{CO_2}^{Sat}$ and ~~in Section 3.3~~$E_{CO_2}^{CEMS}$ for eight power plants (Figure 1). Table 3 lists the mean and the standard deviation of the relative differences between $E_{NO_x}^{CEMS}$ and $E_{NO_x}^{Sat}$, and $E_{CO_2}^{CEMS}$ and $E_{CO_2}^{Sat}$ for all eight power plants.

3.1 Satellite-derived CO_2 emissions ($E_{CO_2}^{Sat}$)

Figure ~~6a~~7a is a scatterplot of $E_{CO_2}^{Sat}$ and $E_{CO_2}^{CEMS}$ for the ~~8~~eight power plants (Figure ~~5), 7~~1), seven of which did not have post-combustion NO_x control systems installed during the study period, 2005–2017. The comparison shows a good correlation, R^2, of 0.66. ~~$E_{CO_2}^{Sat}$ and $E_{CO_2}^{CEMS}$ for individual power plants are tabulated in Table 2 and their relative difference with CEMS measurements are listed in Table 3. The average $E_{CO_2}^{CEMS}$ for all power plants is 2.0 Gg/h and the average $E_{CO_2}^{Sat}$ is 1.8 Gg/h. The relative difference for individual 3~~three year means ~~(defined as $(E_{CO_2}^{Sat} - E_{CO_2}^{CEMS})/E_{CO_2}^{CEMS}$) is 8% ± 41% (mean ± standard deviation). For example, Figure 13 shows $E_{CO_2}^{Sat}$ for the Rockport power plant, which typically has a positive bias as compared to $E_{CO_2}^{CEMS}$ because of a positive bias in $E_{NO_x}^{Sat}$. The average $E_{CO_2}^{CEMS}$ for all power plants is 2.0 Gg/h and the average $E_{CO_2}^{Sat}$ is 1.8 Gg/h. The relative difference for individual three year means (defined as $(E_{CO_2}^{Sat} - E_{CO_2}^{CEMS})/E_{CO_2}^{CEMS}$) is 8% ± 41% (mean ± standard deviation). For example, Figure 3 shows $E_{CO_2}^{Sat}$ for the Rockport power plant, which typically has a positive bias as compared to $E_{CO_2}^{CEMS}$ because of a positive bias in $E_{NO_x}^{Sat}$:~~

The time series between $E_{CO_2}^{Sat}$ and $E_{CO_2}^{CEMS}$ are generally consistent, with their annual averages for the ~~8~~eight power plants exhibiting a declining trend of 5%/yr and 3%/yr from 2006* to 2016* for $E_{CO_2}^{Sat}$ and $E_{CO_2}^{CEMS}$, respectively. The reduction in net electricity generation is the driving force underlying the emission changes, which ~~has~~ decreased by 37% for the ~~8~~eight power plants from 2005 to 2016, as power producers shut down coal-fired units in favor of cheaper and more flexible natural gas as well as solar and wind (USEIA, 2018). It is interesting to note that the temporal variations in $E_{CO_2}^{Sat}$ are not as "smooth" as those in $E_{CO_2}^{CEMS}$, which results from fluctuations in $E_{NO_x}^{Sat}$. Such fluctuations are caused by uncertainties associated with $E_{NO_x}^{Sat}$ as discussed in Section 3.2. For example, changes in VCDs do not necessarily relate linearly with NO_x emissions (e.g., Figure 2 in Duncan et al., 2013) ~~due to~~because of temporal variations in meteorology, and nonlinear NO_x chemistry (Valin et al, 2013) and transport. Averaging VCDs for a long-term period (3 years in this study) helps reduce those influences, but small fluctuations may still exist.

3.2 Uncertainties

We ~~estimate~~estimated the uncertainty of $E_{CO_2}^{Sat}$ based on the fit performance of $E_{NO_x}^{Sat}$ and comparison with $E_{CO_2}^{CEMS}$. The major sources of uncertainty ~~include (a) the fitted NO_x lifetimes and~~are (a) $E_{NO_x}^{Sat}$ (Liu et al., 2016); (b) $ratio_{regressed}^{CEMS}$; and (c) *f*. ~~We assume that their contributions to the total uncertainty are independent and define the total uncertainty as their root mean square~~We give the estimated uncertainties of each source for individual power plants in Table S2.

~~(a) The uncertainty of the fitted NO_x lifetimes and $E_{NO_x}^{Sat}$ are~~ : The uncertainty of $E_{NO_x}^{Sat}$ is quantified following the method described in Liu et al. (2017), accounting for errors arising from ~~both~~ the fit procedure, the NO_x/NO_2 ratio and OMI NO_2 VCD observations (Liu et al., 2016). ~~Particularly~~The number of 1.32 used for scaling the NO_2 to NO_x ratio is based on assumptions presented in section 6.5.1 of Seinfeld and Pandis (2006) for "typical urban conditions and noontime sun". Note

that conditions are quite similar in this study because of the overpass time of OMI close to noon, the selection of cloud-free observations, the focus on the ozone season, and the focus on polluted regions. A case study of CTM simulations shows an identical value of 1.32 for Paris in summer (Shaiganfar et al., 2017). The simulated NO_x/NO_2 ratio at the OMI overpass time within the boundary layer (up to 2 km) in a chemistry–climate model, EMAC (Jöckel et al., 2016), was 1.28 + 0.08 for polluted (NO_x>1×10^{15} molec cm^{-2}) regions for the July 1, 2005, and 1.32 + 0.06 on average for the ozone season. However, the coarse grid of EMAC (2.8° × 2.8° in latitude and longitude) may not capture the true range of variation of the NO_x/NO_2 ratio. Therefore, we assumed an uncertainty of 20% arising from the NO_x/NO_2 ratio, double than the standard deviation of the EMAC ratio.

Additionally, the tropospheric air mass factors (AMF) used in NO_2 retrievals are based on relatively coarsely--resolved surface albedo data and a priori NO_2 vertical profile shapes, likely causing low-biased VCDs over strong emission sources (e.g., Russell et al., 2011; McLinden et al., 2014; Griffin et al., 2019). The average AMF uncertainty of ~30% (see Table 2 in Boersma et al., 2007) likely contributes to the underestimation of emissions from some power plants in this study. Both random and systematic (bias) uncertainties in VCDs directly propagates into the uncertainty of $E_{NO_x}^{Sat}$ (see details in the supplement of Liu et al. (2016) and Section 3.4 of Liu et al. (2017)).

The overall uncertainties of $E_{NO_x}^{Sat}$ range from 6057% to 9564% for all power plants in our analysis, which is comparable with the level of differences between $E_{NO_x}^{Sat}$ and $E_{NO_x}^{CEMS}$. We expect this uncertainty to be less for new (e.g., TROPOMI) and upcoming (e.g., NASA Tropospheric Emissions: Monitoring Pollution, TEMPO) OMI-like sensors, which have enhanced capabilities relative to OMI. Further details are provided in Text S1 of the Supplement.

(b)$ratio_{regressed}^{CEMS}$: For power plants without post-combustion devices, $ratio_{regressed}^{CEMS}$ derived from the regression (Figure 2a4a & b) and the plant-specific CEMS measurements are found to be within 15%, which is assumed as the uncertainty of the ratio to be applied tofor all power plants.

(c)*f*: For power plants with post-combustion devices, an additional uncertainty of 20% is applied to reflect the difference between the predicted and the true removal efficiency as suggested by Figure 46.

We assume that their contributions to the overall uncertainty are independent. We then define the total uncertainty, expressed as a 95% confidence interval, as the sum of the root of the quadratic sum of the aforementioned contribution. The overall uncertainties of $E_{CO_2}^{Sat}$ range from 62% 96are ~60% for theall power plants in our analysis.

However, it is worth noting that this uncertainty estimate is rather conservative. For power plants,The mean and the standard deviation of the relative differences between $E_{NO_x}^{CEMS}$ and $E_{NO_x}^{Sat}$, and $E_{CO_2}^{CEMS}$ and $E_{CO_2}^{Sat}$ $E_{CO_2}^{Sat}$are for all eight power plants provide a good alternative measure of uncertainties (Table 3). The relative differences are rather small, which are 0% ± 33% and 8% ± 41% (mean ± standard deviation) (Figure 6a).for NO_x and CO_2, respectively. We additionally calculate the geometric standard deviations (GSDs) of the difference between $E_{CO_2}^{CEMS}$ and $E_{CO_2}^{Sat}$ from 2006* to 2016* for individual power plants in Table S2. The small values of GSDs ranging from 1.07 to 1.31 further improve our confidence in the accuracy of the derived emissions in this study.

3.3 Application

We apply the approach proposed in this study to estimate CO_2 emissions from a power plant in South Africa, aiming to In this section, we assess the capabilityfeasibility of the approachapplying our method to provide constraint oninfer CO_2 emissions ($E_{CO_2}^{Sat}$) for regionspower plants outside the US. We chose South Africa, a country without reliable emissions accounting, as We first compare the NO_x to CO_2 emission ratios derived from this study with those from a bottom-up emission database in Section 3.3.1. We then apply the US ratio to a power plant in South Africa in Section 3.3.2.

3.3.1 Comparison with bottom-up ratios

Figure 8 shows the ~~area of interest, because we found~~ NO_x to CO_2 emission ratios for 2010 from the global power emissions database (GPED; Tong et al., 2018), which is the only publicly-available ~~information on coal type~~bottom-up emission database that reports both NO_x and CO_2 emissions for individual power plants for every country. All countries with over 30 coal-fired power plants in GPED are shown in Figure 8. Not surprisingly, countries with more strict standards in place for NO_x emissions from power plants (i.e., NO_x emission limit value (ELV) < 200 mg/m^3; hereafter referred to as "more strict countries") have smaller NO_x to CO_2 ratios (i.e., 1.0 versus 2.5 on average) than countries with less strict standard (i.e., NO_x ELV > 200 mg/m^3; hereafter referred to as "less strict countries"). Additionally, the correlation coefficients are smaller for more strict countries (i.e., 0.82 on average) as compared to less strict countries (i.e., 0.96 on average), because power plants in more strict countries are more likely to have installed post-combustion NO_x control ~~status for its power plants.~~ systems, which likely lowered $ratio_y^{CEMS}$, similar to what occurred in the US over our analysis period (Section 2.2.2).

We further compare the 2005 US $ratio_{regressed}^{CEMS}$ in Table 1 with the GPED NO_x to CO_2 emission ratios for less strict countries. We chose the 2005 value for comparison based on the following considerations. In 2005, the US EPA issued the Clean Air Interstate Rule (CAIR) to address the interstate transport of ozone and fine particulate matter pollution for eastern US states, which reduced NO_x emissions and, thus, NO_x to CO_2 ratios ($ratio_y^{CEMS}$). However, similar comprehensive control strategies have not been adopted in less strict countries. In this way, the 2005 values are expected to show better consistency with the NO_x to CO_2 ratios of less strict countries than values for more recent years. Note that the GPED database does not give information on ratios by coal type. Therefore, we use $ratio_{regressed}^{CEMS}$ for bituminous coal, which is the most widely used coal type in coal-fired power plants in most countries.

The ~~power plant of Matimba (including the nearby Medupi~~ratios for individual power plants in less strict countries tend to be larger than the US $ratio_{regressed}^{CEMS}$ for 2005, considering that power plants in those countries may not be equipped with any NO_x control devices or even low-NO_x burners, a technology which is widely installed in US power plants with and without post-combustion NO_x control devices. Most ratios range from US 2005 $ratio_{regressed}^{CEMS}$ to 2005 $ratio_{regressed}^{CEMS}$ + standard deviation (Figure 8). It is no surprise that some less strict countries have ratios higher than this range, which also occurs for some US power plants without post-combustion emission controls (Figure 4). However, there are considerable uncertainties in the GPED database given the scarcity of reliable emissions information in less strict countries. For example, the GPED NO_x and CO_2 emissions estimates for Turkey and Russia, which are outliers in Figure 8, are subject to more assumptions and, thus, larger uncertainties than countries with high-quality country-specific emission data, such as China, which has ~~operated since~~a high-resolution emissions database (CPED; Liu et al., 2015~~) in South Africa are~~), and India, which has a database developed by Argonne National Laboratory (Lu et al., 2011).

Figure 9 shows a schematic of our methodology to estimate the NO_x to CO_2 emission ratios for power plants outside the US. We adopt different approaches for more and less strict countries. More strict countries, including Canada, European Union (EU) member states, Japan, South Korea, and, more recently, China, usually use CEMS to monitor emissions, particularly ~~suitable for application of our method, because~~from the largest emitters. For power plants with CEMS measurements for both NO_x and CO_2 emissions, it is straightforward to use the measured ratios. However, there is still a significant number of power plants in those countries without CEMS technology, particularly for CO_2 measurements. For example, EU member states do not require power plants to use CEMS for CO_2 reporting and the majority of plants in the EU therefore reports CO_2 emissions based on emission factors (Sloss, 2011). Therefore, we recommend applying our method described in Section 2.2 to infer region-specific ratios for those power plants. The US $ratio_{regressed}^{CEMS}$ could be a less accurate, but reasonable approximation when no CEMS data are available, considering those countries share similar NO_x ELVs for power plants as the US. For less strict countries, we recommend using the 2005 US values by coal type when ratios from

countries with similar NO_x emission standard are not available. We also recommend assigning a range from 2005 $ratio^{CEMS}_{regressed}$ to 2005 $ratio^{CEMS}_{regressed}$ + standard deviation, instead of a fixed value, to the ratio for inferring CO_2 emissions, considering the knowledge on ratios from those regions are too few to narrow the constraint.

As demonstrated in Section 2.2, our method presented in this study provides a reasonable estimate of the ratio for power plants without post-combustion NO_x control devices with only knowing coal type. Even for regions without reliable emission information, the information on coal type, particularly for large power plants, are very likely publicly-available. For power plants that install post-combustion NO_x control technology, we additionally require the removal efficiency of the device to derive the ratio. The removal efficiency of post-combustion NO_x control devices is usually directly reported, as the operation of such devices is very expensive and is expected to be subject to strict quality control and assurance standards. In contrast to bottom-up approaches, many details are required, including coal type, coal quality, boiler firing type, NO_x emission control device type, and operating condition of boiler and emission control device, when calculating NO_x and CO_2 emissions.

3.3.2 Application to Matimba power plant in South Africa

We apply the methodology shown in Figure 9 to estimate CO_2 emissions from a South African power plant, Matimba, which is a strong isolated NO_x point source (Figure ~~7).~~10). It is a well-studied power plant, having had its emissions estimated using several different methods as reported in the literature. We estimate $E^{Sat}_{NO_x}$ for Matimba from 2005 to 2017 based on OMI NO_2 observations following the approach in Section 2.1. Matimba ~~use~~uses subbituminous coal with ~~the~~a calorific value of ~ 20 MJ/kg (Makgato and Chirwa, 2017). ~~We assume the NO_x-to-CO_2 emission ratio of Matimba is on the upper end of the US values, considering that it is not equipped with any NO_x control devices, even low NO_x burners which are widely installed in US power plants~~ We apply~~(Pretorius et al., 2015). We thus use~~ the ratio ranging from 2005 $ratio^{CEMS}_{regressed}$ to 2005 $ratio^{CEMS}_{regressed}$ + standard deviation to Matimba, following the methodology in Figure 9, considering that South Africa is a less strict country without any post-combustion NO_x control devices (Pretorius et al., 2015). ~~for subbituminous coal listed in Table 1 to infer $E^{Sat}_{CO_2}$ based on $E^{Sat}_{NO_x}$. The~~Our derived $E^{Sat}_{CO_2}$ is shown in Figure ~~8~~11 and fluctuates over time. ~~Note that the range of $E^{Sat}_{CO_2}$ in Figure 8 represents the emissions based on a range of NO_x-to-CO_2 ratios, not the uncertainty. The overall uncertainty of $E^{Sat}_{CO_2}$ is ~70% for the Matimba power plant.~~ The growth after 2008* is most likely caused by the increased unit operating hours driven by the desire to meet fully the demand for electricity in South Africa after a period of rolling blackouts (2007–2008) (Duncan et al., 2016). The decline afterwards may be associated with the tripping of generating units at the Matimba ~~due to overload and the shortage of coal supply. The newly-built power plant of Medupi contributes to the increase from 2015*.~~because of overload and shortage of coal as reported by South African government news agency (available at https://www.sanews.gov.za/south-africa/eskom-alone-cannot-solve-our-energy-challenges). The increase in 2016* may be associated with a newly-built power plant, Medupi, which began limited operations in 2015. Note that the range of $E^{Sat}_{CO_2}$ (grey band) in Figure 11 represents the emissions based on a range of NO_x-to-CO_2 ratios, not the uncertainty. We calculate the uncertainty of $E^{Sat}_{CO_2}$ for Matimba following Section 3.2 with an additional uncertainty of ~50% to reflect the fact that the ratio may range from $ratio^{CEMS}_{regressed}$ to $ratio^{CEMS}_{regressed}$ + standard deviation. The overall uncertainty of $E^{Sat}_{CO_2}$ for Matimba is 81%, as shown by the error bars in Figure 11.

Figure ~~8 compares~~11 shows $E^{Sat}_{CO_2}$ derived in this study ~~with~~and other ~~publicly available~~independent estimates ~~and shows reasonable agreement.~~ reported in $E^{Sat}_{CO_2}$ ~~falls between~~ the ~~estimates based on OCO-2 CO_2 observations~~literature, including two top-down (Nassar et al., 2017; Reuter et al., 2019) and ~~two~~three bottom-up estimates ~~including~~ (Wheeler and Ummel ~~(~~, 2008~~) and~~; Tong et al. ~~(~~., 2018~~). We make a further comparison~~; Oda et al., 2018). Despite the uncertainties associated with each of ~~NO_x~~ these methods, the CO_2 emissions ~~esttimates~~estimates agree reasonably well, but we do not have sufficient information to understand the differences between these estimates. However, Tong et al. (2018~~), the only~~) present in their

CPED database ~~that reports~~ both CO_2 and NO_x emissions, ~~in order~~which allows us to ~~shed light on the reason for~~determine that the difference~~. The differences~~ between $E_{NO_x}^{Sat}$ and the CPED bottom-up ~~estimates contribute~~estimate contributes significantly to the ~~differences of~~difference in CO_2 estimates from the two methods. $E_{NO_x}^{Sat}$ for Matimba is 3.8 Mg/h for 2010*, which is 65% smaller than the estimate by Tong et al. (2018) for 2010. It is not surprising to see such differences considering the uncertainties of satellite-derived NO_x emissions and bottom-up estimates for power plants without reliable CEMS measurements. ~~On one hand~~For instance, $E_{NO_x}^{Sat}$ is potentially underestimated~~, due to~~ because of the bias in the OMI NO_2 standard product (version 3.1) associated with a low-resolution static climatology of surface Lambert-Equivalent Reflectivity (OMLER) (Kleipool et al., 2008). We perform a sensitivity analysis by using the preliminary new version of the OMI NO_2 product, which uses new geometry dependent Moderate Resolution Imaging Spectroradiometer (MODIS~~-~~)-based surface reflectivity. The inferred $E_{NO_x}^{Sat}$ based on the new product ~~increases by~~is over 10~~%. On the other hand, the~~% higher than version 3.1. The bottom-up estimates for Matimba are subject to significant uncertainties as well. For example, Tong et al. (2018) used national total fuel consumption of the power sector for South Africa as reported by the International Energy Agency ~~is used~~ to estimate fuel consumption at the plant level as detailed fuel consumption for each plant is not currently available. Additionally, ~~due to the absence of country specific measurement data,~~they used default NO_x emission factors obtained from the literature ~~are applied (Tong et al., 2018).~~because of the absence of country-specific measurement data.

4 Conclusions

~~We present a method to estimate CO_2 emissions of ozone season from individual power plants from satellite observations of co-emitted~~In our study, we investigated the feasibility of using satellite data of NO_2 from power plants to infer co-emitted CO_2 emissions, which could serve as complementary verification of bottom-up inventories or be used to supplement these inventories that are highly uncertain in many regions of the world. For example, our estimates will serve as an independent check of CO_2 emissions that will be inferred from satellite retrievals of future CO_2 sensors (Bovensmann et al., 2010). Currently, uncertainties in CO_2 emissions from power plants confound national and international efforts to design effective climate mitigation strategies.

We estimate NO_2 and CO_2 emissions during the "ozone season" from individual power plants from satellite observations of NO_2 and demonstrate its utility for US power plants, which have accurate CEMS with which to evaluate our method. ~~While~~We systematically identify the ~~uncertainty associated with~~ sources of variation, such as types of coal, boiler, and NO_x emission control device, and change in operating conditions, which affect the NO_x to CO_2 emissions ratio. Understanding the causes of these variations will allow for better informed assumptions when applying our method to power plants that have no or uncertain information on the factors that affect their emissions ratios. For example, we estimated CO_2 emissions from the large and isolated Matimba power plant in South Africa, finding that our emissions estimate shows reasonable agreement with other independent estimates.

We found that it is ~~relatively high when applied to OMI~~feasible to infer CO_2 emissions from satellite NO_2 observations, but limitations of the current satellite data~~, we expect that the uncertainty will be less~~ (e.g., spatio-temporal resolution, signal-to-noise) only allow us to apply our method to eight large and isolated U.S. power plants. Looking forward, we anticipate that these limitations will diminish for the recently launched ~~European Union Copernicus Sentinel 5 precursor (TROPOMI, launch~~ (October 2017~~),~~) TROPOMI, and ~~the~~three upcoming ~~NASA geostationary Earth Venture one (TEMPO, launch~~(launches expected in the early 2020s~~), both of which~~) geostationary instruments (NASA TEMPO; European Space Agency and Copernicus Programme Sentinel-4; Korea Meteorological Administration Geostationary Environment Monitoring Spectrometer, GEMS), which are designed to have superior capabilities~~. For example, higher spatiotemporal~~ to OMI. High resolution TROPOMI observations are capable of describing the spatio-temporal variability of NO_2, even in a

relatively small city like Helsinki. (Ialongo et al., 2019) and allow estimates of NO_x emissions to be calculated for shorter timeframes (Goldberg et al., 2019c). Higher spatial and temporal resolutions will likely ~~improve the estimation~~reduce uncertainties in estimates of NO_x emissions as well as allow for the separation of more power plant plumes from nearby sources, thus increasing the number of power plants available for analysis. Therefore, future work will be to apply our method to these new datasets, especially after several years of vetted ~~TROPOMI~~ data become available.

~~We explore the feasibility of estimating CO_2 emissions from power plants in regions of the world without reliable emissions accounting by applying our method to a South African plant. The emissions estimates for the power plant of Matimba show reasonable agreement with existing estimates. The ratios derived in this study have the potential to be applied to power plants located in other regions by carefully investigating their coal type and NO_x control devices, in order to provide an additional constraint on CO_2 emissions. Future~~Additional future work will include applying our method to other regions of the world with reliable CEMS information, such as Europe, Canada and, more recently, China, to develop a more reliable and complete database with region-specific ratios. ~~This method will also serve as an independent approach to check CO_2 emissions based on satellite retrievals of CO_2 average mixing ratio from future CO_2 sensors with improved accuracy and extended the spatial coverage (Bovensmann et al., 2010).~~

Data availability

The OMI NO_2 and MERRA-2 data can be downloaded from the Goddard Earth Sciences Data and Information Services Center (GES DISC; available at https://disc.gsfc.nasa.gov/datasets).

Author contribution

Fei Liu, Bryan N. Duncan, and Nickolay A. Krotkov designed the framework. Fei Liu, Steffen Beirle, Lok N. Lamsal, Debora Griffin, Chris A. McLinden, and Daniel L. Goldberg developed the NO_x emission fitting algorithm and Fei Liu carried it out. Fei Liu and Zifeng Lu analysed the NO_x/CO_2 emission ratio. Fei Liu and Bryan N. Duncan prepared the manuscript with contributions from all co-authors.

Competing interests

The authors declare that they have no conflict of interest.

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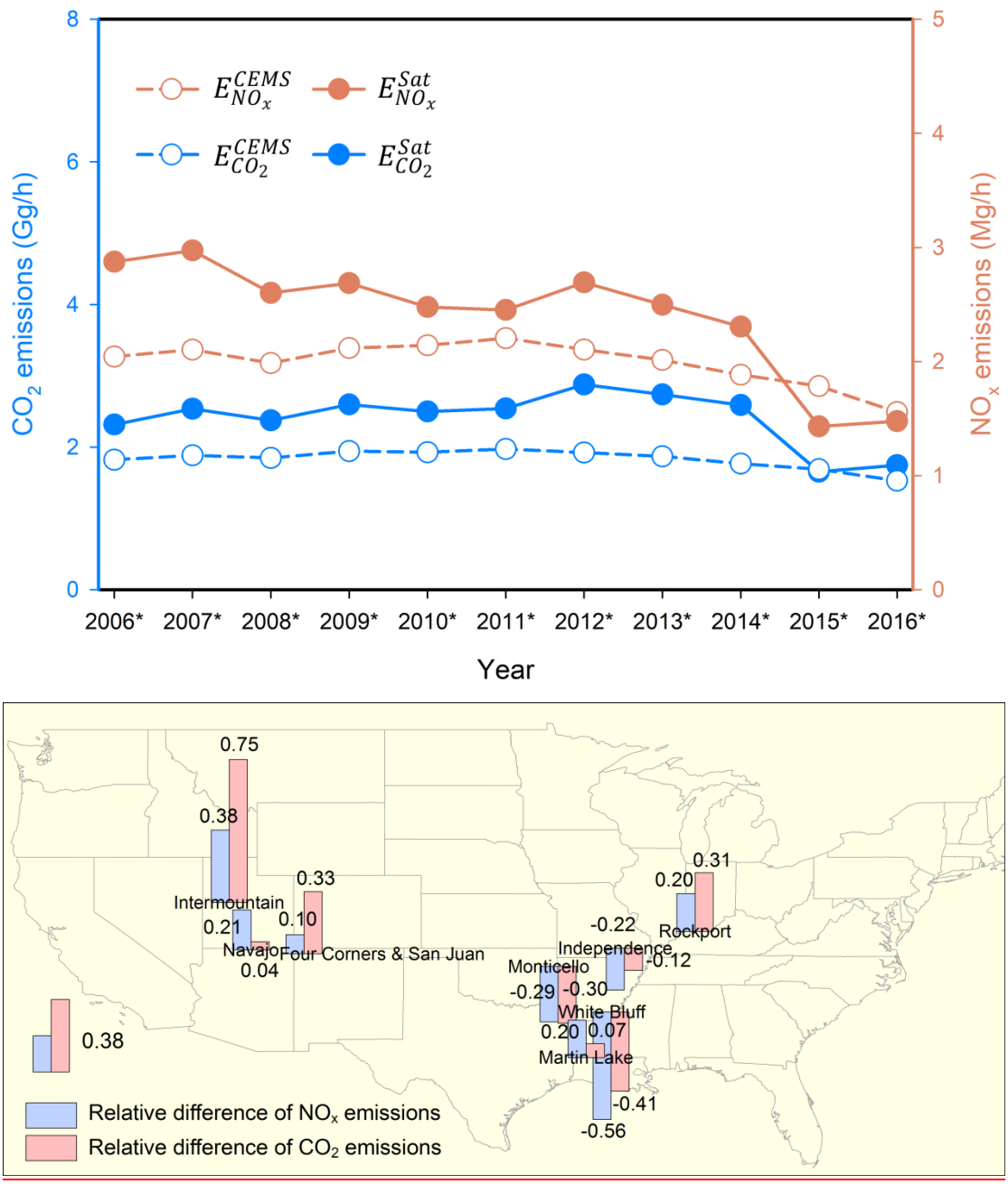


Figure 1 Locations of the power plants investigated in this study. The bar charts denote the relative differences, defined as $(E^{Sat}-E^{CEMS})/E^{CEMS}$, averaged over 2005–2017, for NO_x (blue) and CO_2 (red) emissions. The upward and downward bars represent positive and negative differences, respectively. The Monticello power plant installed SNCR to control NO_x emissions in 2008. The other ~~Other~~ power plants are not equipped with post-combustion NO_x control devices.

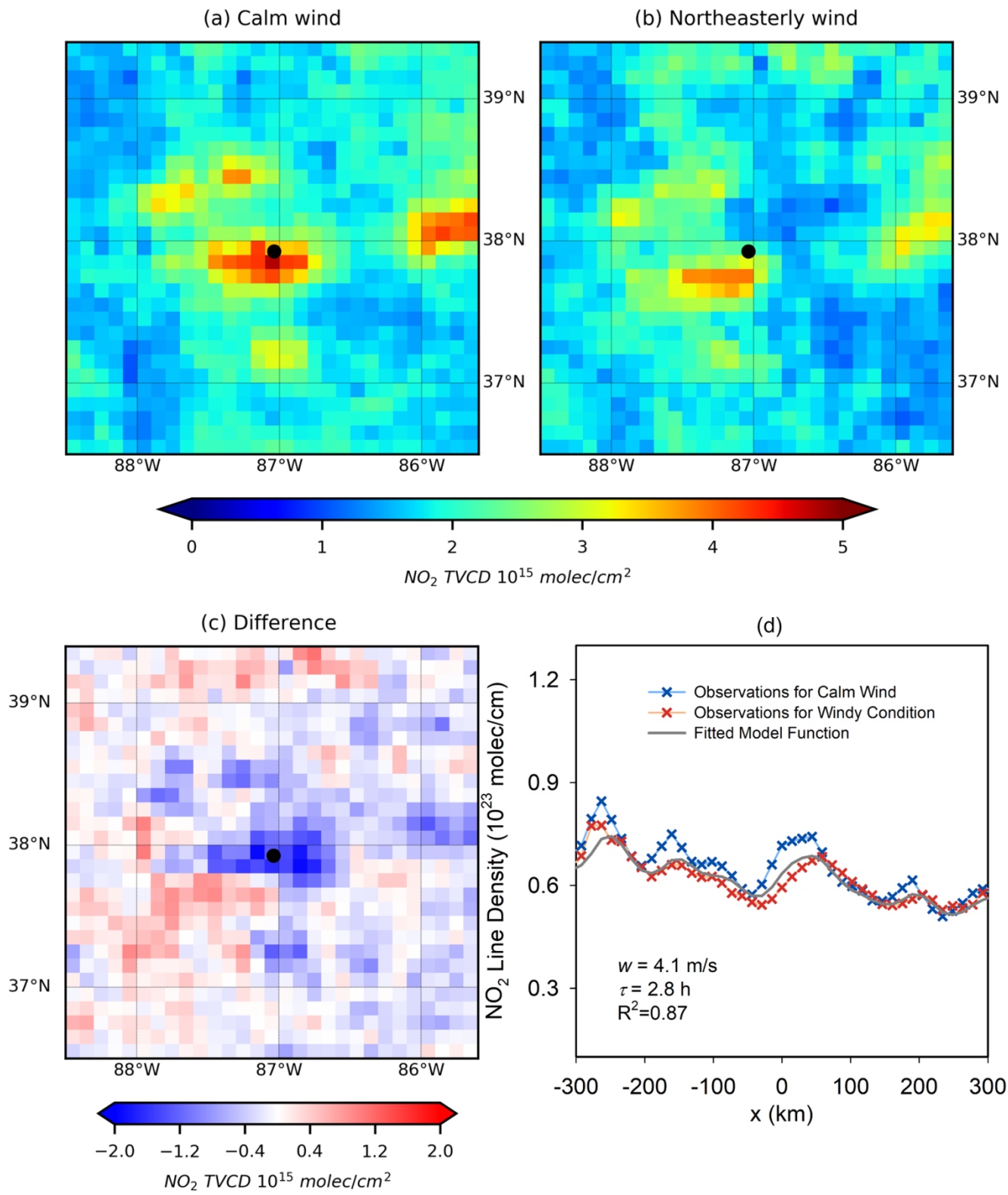


Figure 2 Mean OMI NO_2 tropospheric VCDs around the Rockport power plant (Indiana, USA) for (a) calm conditions, (b) northeasterly wind and (c) their difference (northeasterly − calm) for the period of 2005 – 2017. The location of Rockport is labelled by a black dot. (d) NO_2 line densities around Rockport. Crosses: NO_2 line densities for calm (blue) and northeasterly winds (red) as function of the distance x to Rockport center. Grey line: the fitted results for NO_2 line densities for northeasterly winds. The numbers indicate the net mean wind velocities (windy − calm) from MERRA-2 (w), the fitted lifetime (τ), and the coefficient of determination (R^2) of the fit.

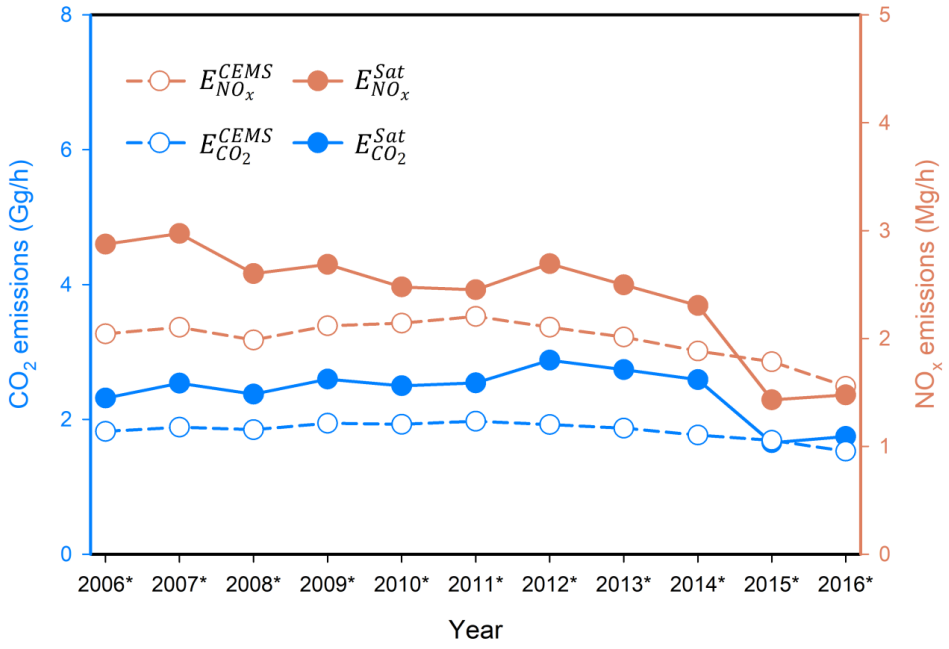


Figure 3 $\boldsymbol{E_{NO_x}^{Sat}}$ (Mg/h; orange solid lines – right axis) and $\boldsymbol{E_{CO_2}^{Sat}}$ (Gg/h; blue solid line – left axis) for the Rockport power plant ~~during~~ from 2005 to 2017. $\boldsymbol{E_{NO_x}^{CEMS}}$ and $\boldsymbol{E_{CO_2}^{CEMS}}$ (dashed lines) are also shown. The 3-year periods are represented by the middle year with an asterisk (e.g., 2006* denotes the period from 2005 to 2007).

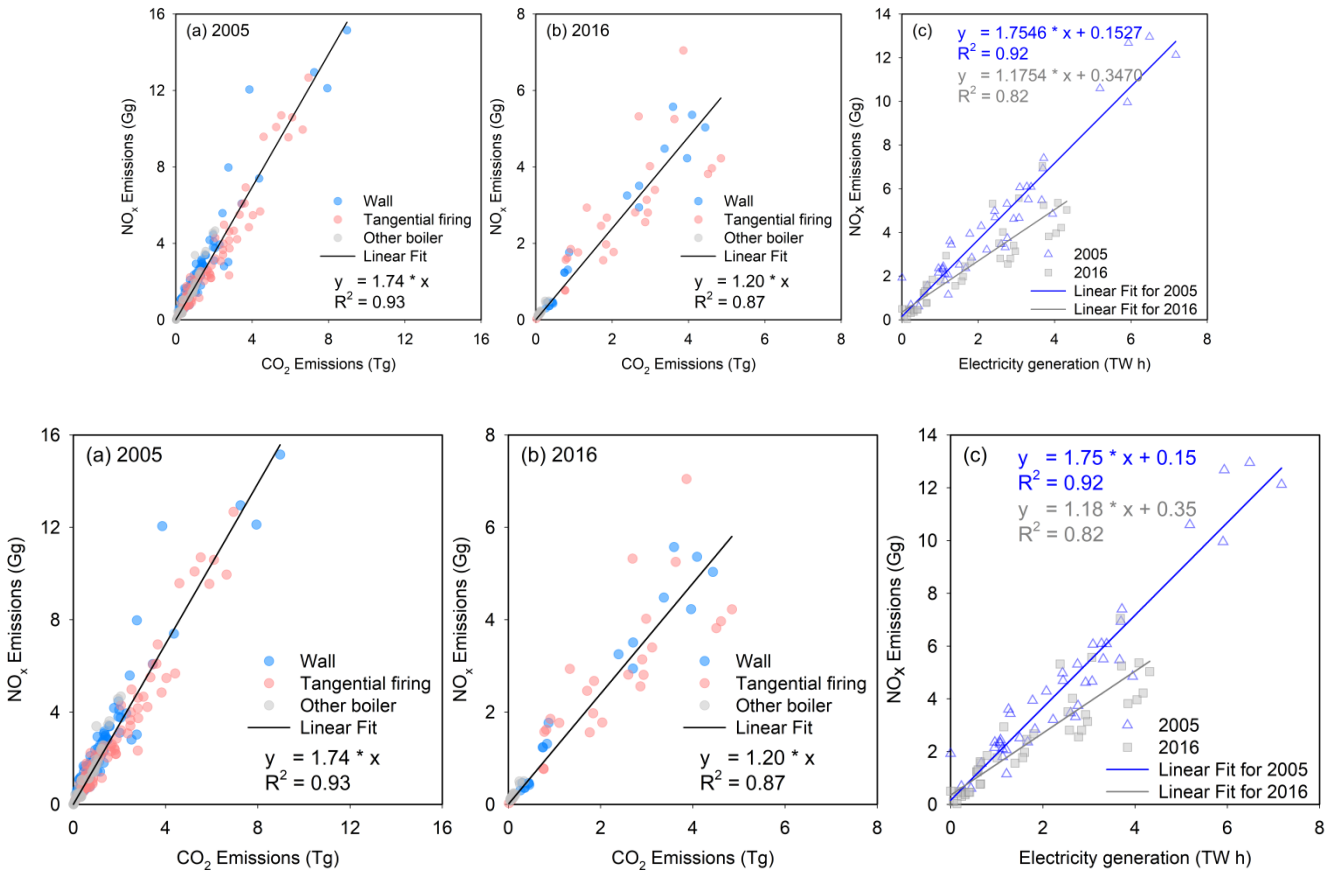


Figure ~~2~~4 Scatter plots of $E_{NO_x}^{CEMS}$ versus $E_{CO_2}^{CEMS}$ for all the US bituminous coal-fired electric generating units for (a) 2005 and (b) 2016. Values are color coded by firing type. (c) Scatter plot of $E_{NO_x}^{CEMS}$ versus electricity generation of the same units for years 2005 (triangle) and 2016 (square). Only plants without post-combustion NO_x control devices within a given year are used. The electricity generation data are also from eGRID. The lines in all three panels represent the computed linear regressions.

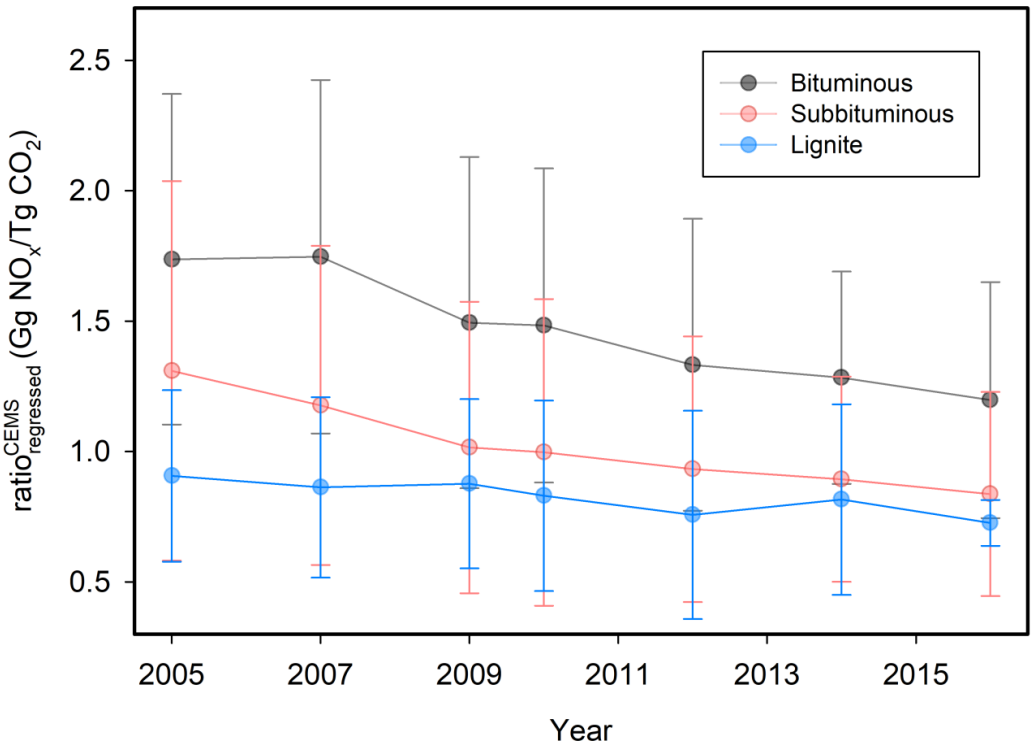


Figure ~~35~~ 35 Interannual trends of $ratio^{CEMS}_{regressed}$ for power plants using bituminous, subbituminous and lignite coal types and without post-combustion NO_x control devices in a given year. Error bars show the standard deviations for ratios of $E^{CEMS}_{NO_x}$ to $E^{CEMS}_{CO_2}$ for individual power plants.

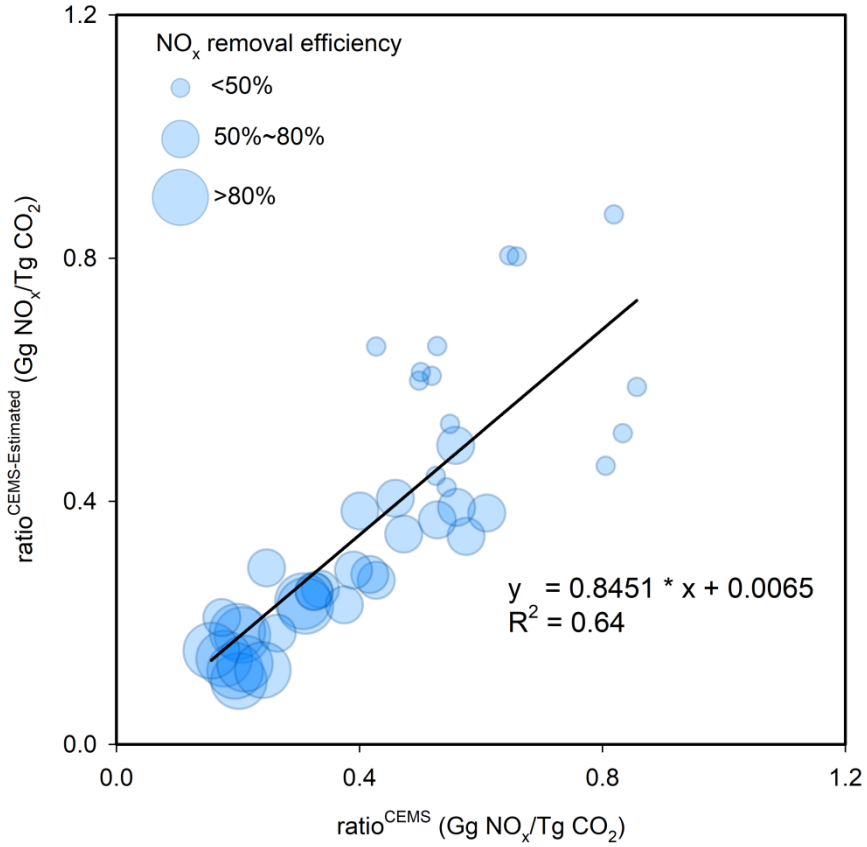
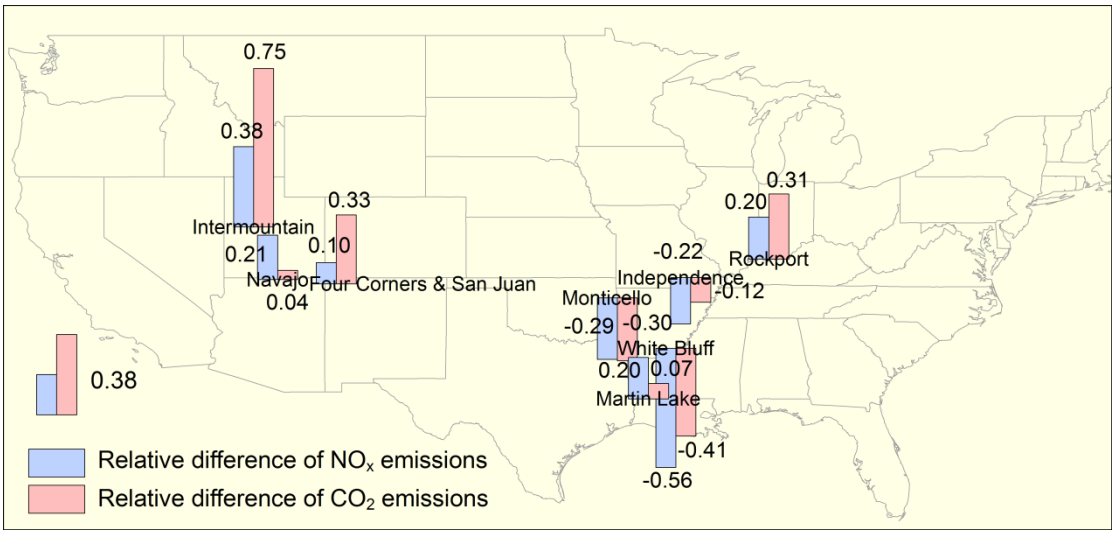
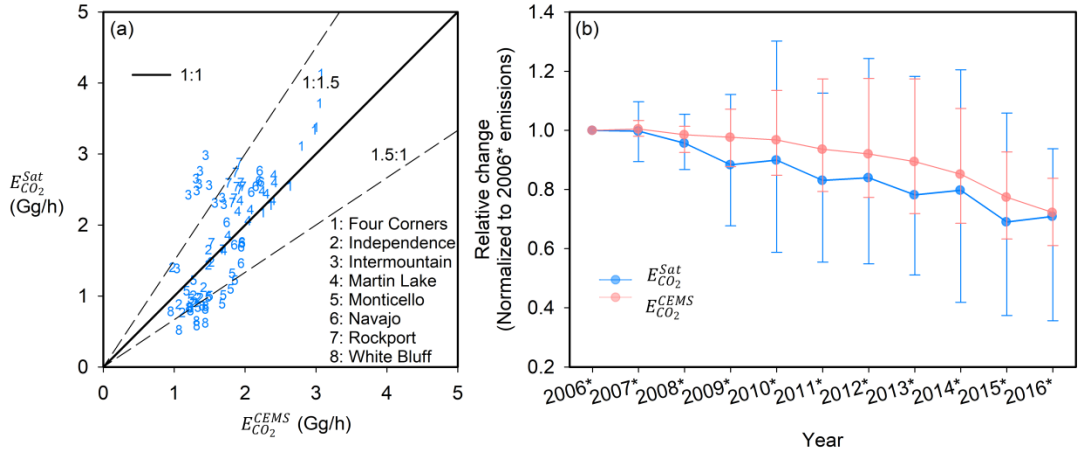


Figure ~~46~~ 46 Scatterplot of ~~the ratio of~~ $ratio^{CEMS-Estimated}$ as compared to $ratio^{CEMS}$ for 2016. All 44 coal-fired power plants that operated post-combustion ~~techniques~~devices after 2005 and before 2016 (including 2016) are used in the plot. The sizes of the circles denote the magnitude of the NO_x reduction efficiency of post-combustion control devices estimated in this study. The line represents the linear regression of $ratio^{CEMS}$ to $ratio^{CEMS-Estimated}$.





~~**Figure 5** The bar charts denote the relative differences, defined as $(E^{Sat} - E^{CEMS})/E^{CEMS}$, averaged over 2005–2017, for NO_x (blue) and CO_2 (red) emissions. The upward and downward bars represent positive and negative differences, respectively. The Monticello power plant installed SNCR to control NO_x emissions in 2008. Other power plants are not equipped with post-combustion NO_x control devices.~~

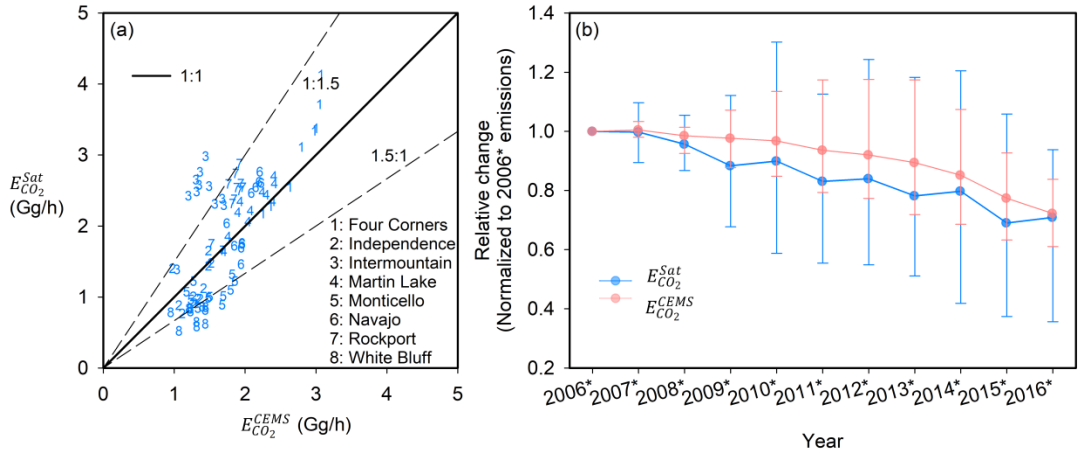


Figure ~~6~~7 (a) Scatterplot of $E_{CO_2}^{Sat}$ for ~~8~~eight power plants as compared to $E_{CO_2}^{CEMS}$ from 2006* to 2016*. The ~~straight~~ solid ~~and~~lines represent the ratio of 1:1. The dashed lines represent the ratio of 1:1.5 and ~~1:~~1.5:1, respectively. (b) Interannual trends of the averaged $E_{CO_2}^{Sat}$ (blue lines) and $E_{CO_2}^{CEMS}$ (pink lines) are~~is~~ for all power plants analyzed in this study from 2006*–2016*, as normalized to the 2006* value. The whiskers denote the maximum and minimum values.

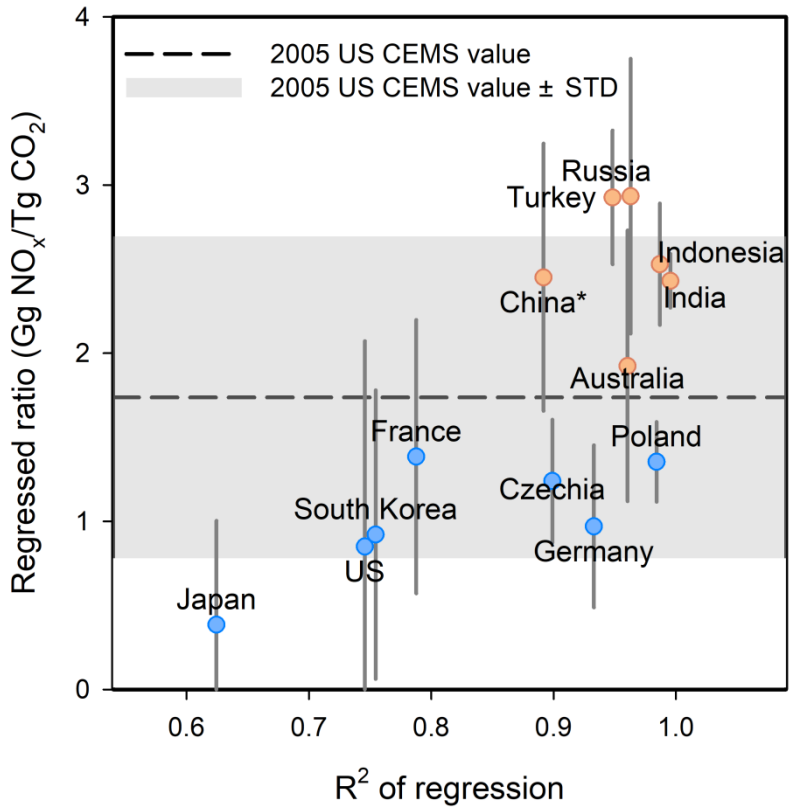


Figure 8 Comparison of the regressed NO_x to CO_2 emission ratios derived from the global power emissions database (GPED) for different regions versus the correlation coefficient of the regression. The blue and red circles denote regions that are subject to more strict standard for NO_x emissions from power plants (i.e., a NO_x ELV of 200 mg/m^3 or less) and other regions, respectively. Y axis: the slope of the regression of the NO_x to CO_2 emissions with an assumed y-intercept of zero. Error bars show the standard deviations for the NO_x to CO_2 emission ratios for individual power plants. X axis: correlation coefficient of the regression. The dashed line represents 2005 US $ratio_{regressed}^{CEMS}$ for bituminous coal derived in this study. The grey shadow represents 2005 US $ratio_{regressed}^{CEMS}$ ± standard deviation.
*China switched from being a less strict country to a more strict country in 2014, when most coal-fired power plants in China were required to comply with its new emission standards (GB13223-2011).

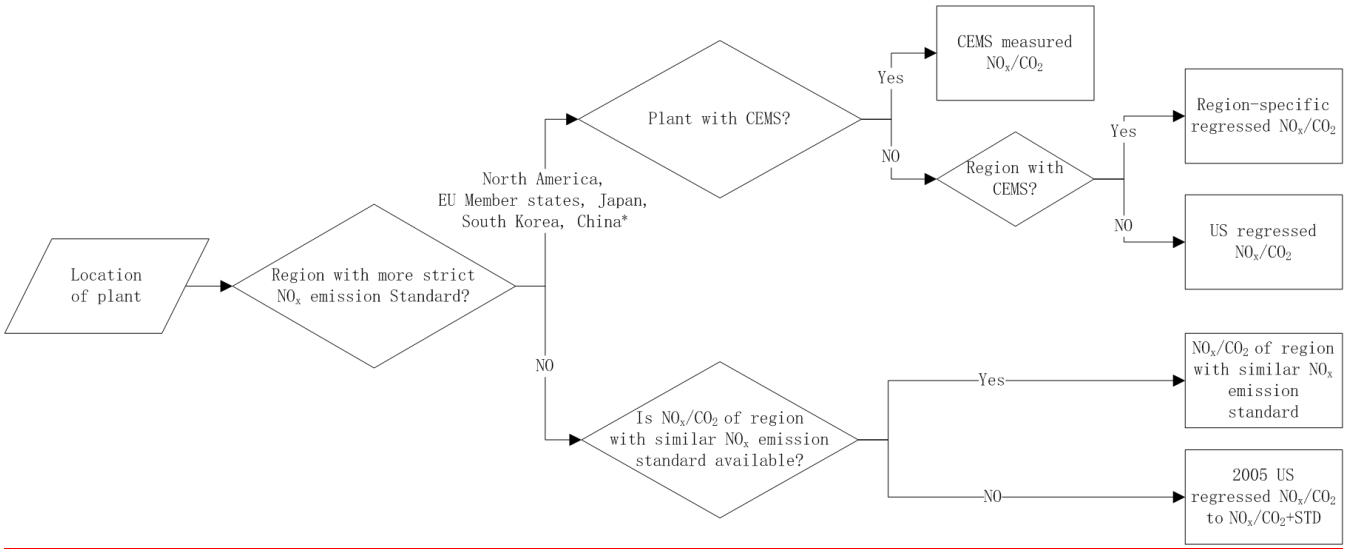


Figure 9 Schematic of our methodology to estimate the NO_x to CO_2 emission ratios for power plants outside the US.

*China switched from being a less strict country to a more strict country in 2014, when most coal-fired power plants in China were required to comply with its new emission standards (GB13223-2011).

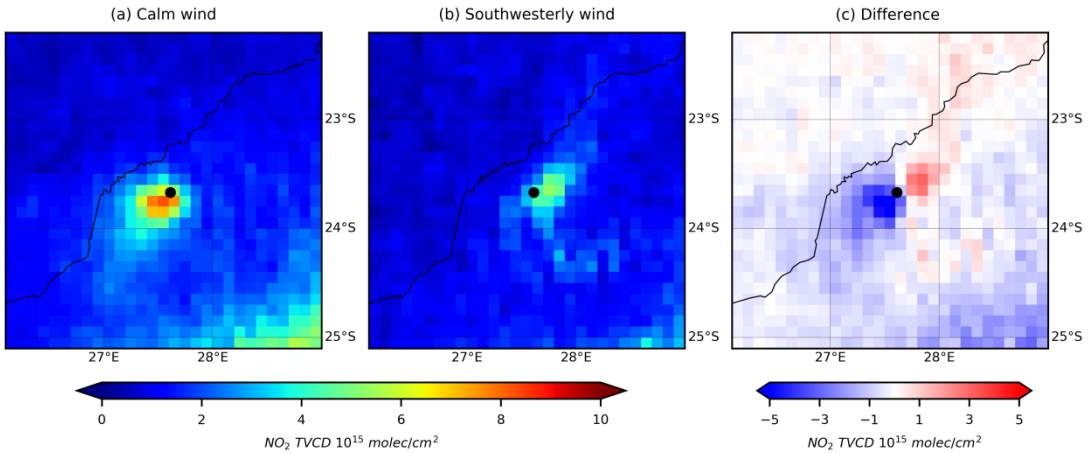
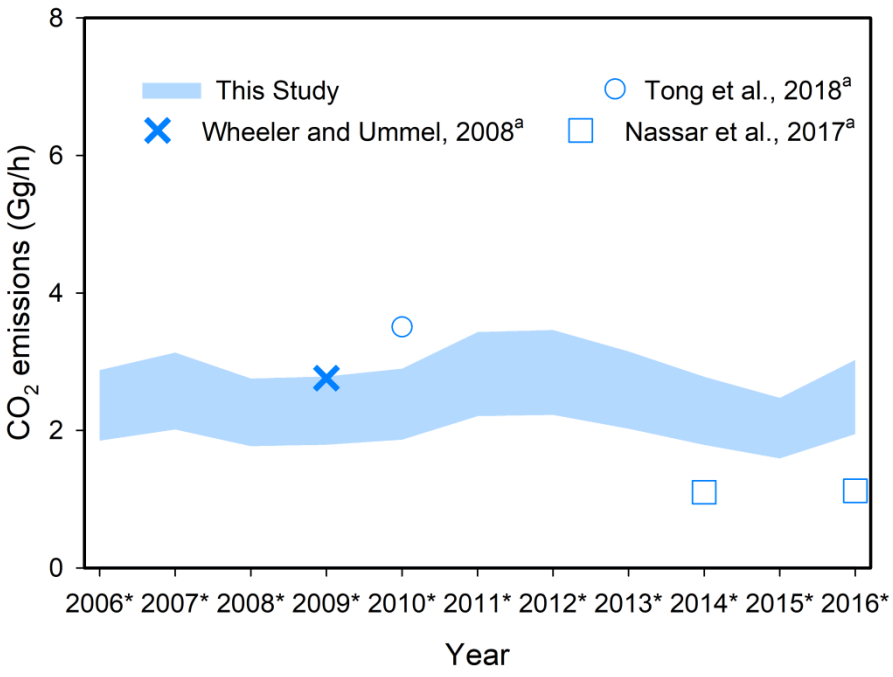


Figure 10 Mean OMI NO_2 tropospheric VCDs around the Matimba power plant (Lephalale, South Africa) for (a) calm, (b) southwesterly wind conditions and (c) their difference (southwesterly − calm) for the period of 2005 – 2017. The location of Matimba is ~~**labelled**~~represented by a black dot.



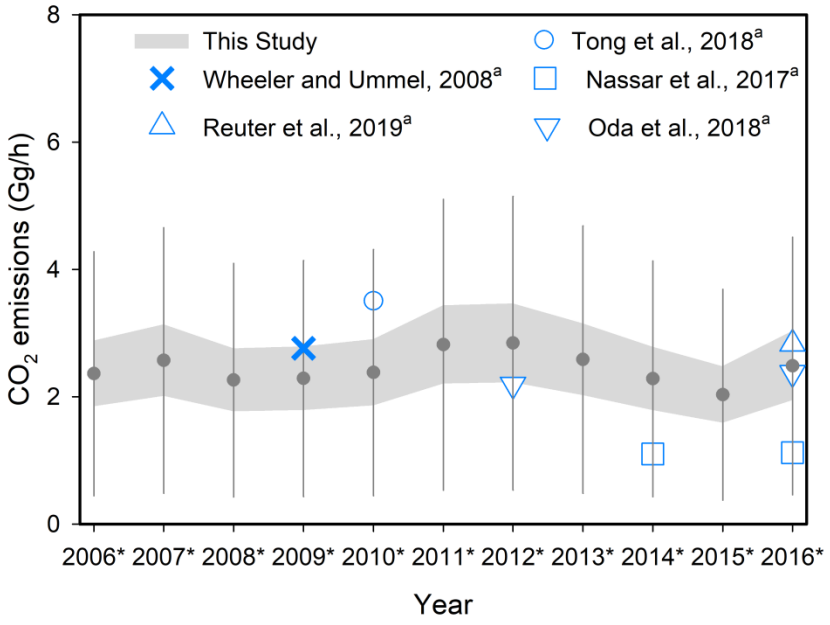


Figure ~~8~~11 Comparison of $\mathbf{E}_{\mathbf{CO_2}}^{\mathbf{Sat}}$ (Gg/h) derived in this study with existing estimates for the Matimba power plant during 2005 to 2017. $\mathbf{E}_{\mathbf{CO_2}}^{\mathbf{Sat}}$ is inferred based on the NO_x to CO_2 emissions ratio ranging from $\boldsymbol{ratio}_{\boldsymbol{regressed}}^{\boldsymbol{CEMS}}$ to $\boldsymbol{ratio}_{\boldsymbol{regressed}}^{\boldsymbol{CEMS}}$ + standard deviation of ratio. The upper and lower grey bands denote the emissions inferred from $\boldsymbol{ratio}_{\boldsymbol{regressed}}^{\boldsymbol{CEMS}}$ and $\boldsymbol{ratio}_{\boldsymbol{regressed}}^{\boldsymbol{CEMS}}$ + standard deviation of ratio, respectively. The grey dots and error bars show the mean of the upper and lower grey bands and their uncertainties, respectively.

[a]Emissions are estimated for 2009 by Wheeler and Ummel (2008); for 2010 by Tong et al. (2018); ~~and~~ for 2014 and 2016 by Nassar at al. (2017); for 2016 by Reuter et al. (2019); and for 2012 and 2016 by Oda at al. (2018).

Table 1 The slope ($ratio_{regressed}^{CEMS}$), coefficient of determination, standard deviation and sample number of the linear regression of $E_{NO_x}^{CEMS}$ and $E_{CO_2}^{CEMS}$ by year for all US power plants without post-combustion NO_x control devices from 2005 to 2016.

Coal type	Year	$ratio_{regressed}^{CEMS}$	R^2	Standard deviation	Sample number[a]
Bituminous	2005	1.74	0.93	0.63	278
	2007	1.75	0.91	0.68	286
	2009	1.49	0.88	0.64	241
	2010	1.48	0.86	0.60	235
	2012	1.33	0.87	0.56	190
	2014	1.28	0.87	0.41	136
	2016	1.20	0.87	0.45	66
Subbituminous	2005	1.31	0.65	0.73	226
	2007	1.18	0.61	0.61	221
	2009	1.02	0.66	0.56	230
	2010	1.00	0.67	0.59	216
	2012	0.93	0.74	0.51	200
	2014	0.89	0.74	0.39	165
	2016	0.84	0.70	0.39	111
Lignite	2005	0.91	0.74	0.33	20
	2007	0.86	0.82	0.35	22
	2009	0.88	0.91	0.32	16
	2010	0.83	0.94	0.37	18
	2012	0.76	0.91	0.40	15
	2014	0.82	0.92	0.37	12
	2016	0.73	0.78	0.09	9

[a]The sample number generally decreases from 2005 to 2016 as power plants installed post-combustion NO_x control devices over time.

Table 2 Summary of effective NO_x lifetimes, satellite-derived NO_x emissions ($E_{NO_x}^{Sat}$), CO_2 emissions ($E_{CO_2}^{Sat}$) and bottom-up NO_x emissions ($E_{NO_x}^{CEMS}$), CO_2 emissions ($E_{CO_2}^{CEMS}$) for 8 US power plants during May to September from 2005 to 2017. The 3-year periods are represented by the middle year with an asterisk.

Category	Year	Four Corners & San Juan	Independence	Intermountain	Martin Lake	Monticello	Navajo	Rockport	White Bluff
NO_x lifetime	2005-2017	2.7	2.5	2.2	2.3	3.2	2.3	2.4	4.3
$E_{NO_x}^{Sat}$ (Mg/h)	2006*	10.5	2.0	4.0	2.4	1.1	4.6	2.9	1.0
	2007*	10.0	1.7	4.1	2.3	1.1	4.4	3.0	0.9
	2008*	9.4	1.6	3.7	2.0	0.8	4.5	2.6	0.9
	2009*	7.2	1.2	3.9	2.1	0.7	3.9	2.7	0.7
	2010*	6.8	1.0	4.4	2.1	0.6	3.6	2.5	0.9
	2011*	6.5	0.9	3.6	1.8	0.7	2.5	2.5	0.8
	2012*	6.3	0.9	3.4	1.6	0.6	2.3	2.7	0.8
	2013*	5.6	0.8	3.5	1.8	0.5	1.9	2.5	0.6
	2014*	4.4	0.7	3.5	1.7	0.8	2.2	2.3	0.5
	2015*	3.8	0.8	3.0	1.4	0.7	2.1	1.4	0.4
	2016*	3.5	1.2	1.7	1.2	0.6	2.5	1.5	0.7
$E_{NO_x}^{CEMS}$ (Mg/h)	2006*	7.4	1.8	3.0	1.8	1.5	3.8	2.0	1.7
	2007*	7.3	1.8	3.1	1.8	1.4	3.9	2.1	1.6
	2008*	6.8	1.8	2.9	1.8	1.3	3.8	2.0	1.6
	2009*	6.5	1.6	2.9	1.8	1.2	3.4	2.1	1.8
	2010*	6.2	1.6	2.8	1.7	1.1	2.8	2.1	1.8
	2011*	6.2	1.4	2.5	1.5	1.0	2.2	2.2	1.9
	2012*	6.1	1.3	2.4	1.4	0.9	1.9	2.1	1.9
	2013*	5.6	1.3	2.4	1.3	0.9	1.9	2.0	2.0
	2014*	5.2	1.2	2.5	1.3	0.8	1.9	1.9	1.9
	2015*	4.3	1.2	2.0	1.3	0.8	1.7	1.8	1.5
	2016*	3.9	1.1	1.5	1.2	0.8	1.6	1.6	1.2
$(E_{NO_x}^{Sat} - E_{NO_x}^{CEMS}) / E_{NO_x}^{CEMS}$	2005-2017	10%	-22%	38%	20%	-29%	21%	20%	-56%
$E_{CO_2}^{Sat}$ (Gg/h)	2006*	6.1	1.6	2.3	2.7	1.2	2.6	2.3	0.8
	2007*	5.9	1.5	2.4	2.6	1.3	2.6	2.5	0.8
	2008*	5.6	1.4	2.3	2.3	1.1	2.8	2.4	0.8
	2009*	4.1	1.1	2.6	2.4	1.0	2.5	2.6	0.6
	2010*	3.7	1.0	3.0	2.5	0.9	2.5	2.5	0.9
	2011*	3.4	1.0	2.6	2.2	1.0	1.7	2.5	0.8
	2012*	3.3	1.0	2.5	2.1	1.0	1.7	2.9	0.9
	2013*	3.1	0.9	2.6	2.3	0.8	1.5	2.7	0.6
	2014*	2.5	0.8	2.8	2.2	1.2	1.8	2.6	0.6
	2015*	2.3	0.9	2.4	1.8	1.1	1.7	1.7	0.5
	2016*	2.2	1.4	1.4	1.6	1.0	2.0	1.7	0.8
$E_{CO_2}^{CEMS}$ (Gg/h)	2006*	3.1	1.5	1.7	2.4	1.9	2.2	1.8	1.2
	2007*	3.1	1.5	1.7	2.4	1.8	2.2	1.9	1.2
	2008*	3.0	1.5	1.6	2.4	1.8	2.2	1.8	1.2
	2009*	3.1	1.4	1.5	2.3	1.7	2.1	1.9	1.3
	2010*	3.0	1.4	1.4	2.2	1.7	2.1	1.9	1.4
	2011*	3.0	1.3	1.3	2.1	1.5	2.0	2.0	1.4

	2012*	3.0	1.3	1.3	2.0	1.5	1.9	1.9	1.4
	2013*	2.8	1.3	1.3	1.9	1.3	1.9	1.9	1.4
	2014*	2.6	1.1	1.4	1.9	1.3	2.0	1.8	1.3
	2015*	2.4	1.1	1.2	1.8	1.2	1.8	1.7	1.1
	2016*	2.2	1.0	1.0	1.7	1.2	1.7	1.5	0.9
$(E_{CO_2}^{Sat} - E_{CO_2}^{CEMS}) / E_{CO_2}^{CEMS}$	2005-2017	33%	-12%	75%	7%	-30%	4%	31%	-41%

Table 3 Summary of relative difference between satellite-derived NO_x emissions ($E_{NO_x}^{Sat}$) and bottom-up NO_x emissions ($E_{NO_x}^{CEMS}$), satellite-derived CO_2 emissions ($E_{CO_2}^{Sat}$) and bottom-up CO_2 emissions ($E_{CO_2}^{CEMS}$) for 8 US power plants during May to September from 2005 to 2017. The 3-year periods are represented by the middle year with an asterisk.

Year	Relative Difference for NO_x		Relative Difference for CO_2	
	Mean	Standard Deviation	Mean	Standard Deviation
2006*	15%	29%	17%	39%
2007*	10%	29%	16%	38%
2008*	5%	30%	14%	39%
2009*	-3%	34%	6%	39%
2010*	-1%	38%	9%	46%
2011*	-5%	31%	3%	40%
2012*	-3%	31%	5%	41%
2013*	-4%	38%	4%	49%
2014*	-3%	36%	7%	46%
2015*	-8%	35%	2%	41%
2016*	-2%	29%	8%	22%