#Reveiwer 2: Quantifying burning efficiency in Megacities using NO2 /CO ratio from the Tropospheric Monitoring Instrument (TROPOMI)"

The paper presents NOx/CO emission ratios as derived from satellite observations of NO2 and CO. It demonstrates the high potential of TROPOMI for atmospheric research. The paper is generally well written. However, the details of the method lack some details, and the robustness of the results is hard to evaluate with the given information. Thus, before publication, major revisions are necessary. The method description has to be extended. In particular, the two approaches have to be illustrated for real data rather than just for a schematic plot. In addition, the definitions of background and upwind areas have to be made consistent for all cities. If this is not possible add a discussion of uncertainties due to the a-priori settings. Error bars only account for statistical day-to-day variations, but ignore the impact of a-priori settings and the CER procedure.

Author Response:

Thank you for your time and suggestions, particularly concerning details of the method that we used, which helped to improve the manuscript.

Detailed comments:

- Line 117: Is this bias in NO2 accounted for in your study? How would the results change if NO2 would be scaled up accordingly?

Author Response:

The bias in S5P NO2 retrieval has been assessed for European cities. However, since we don't know yet how representative this estimate is for the cities that we study, it was decided to account for the impact of the bias as an additional the source of uncertainty of 25% of the TROPOMI inferred NO2/CO ratio (see Table S3).

In case, if 25 % bias would apply for all the cities, TROPOMI derived emission ratio increase by factor of 1.25. TROPOMI derived emission ratio for Mexico City remains close to MACCITY (within 5%) and EDGAR (within 25%). For Tehran, Cairo, Lahore and Riyadh MACCity emission ratio is lower by 50 % in contrast to TROPOMI derived ratio. EDGAR emission ratio is close to TROPOMI derived ratio for Riyadh and Lahore (within 25 %). For Los Angeles, TROPOMI derived emission ratio is lower by factor 2 and 3 than EDGAR and MACCity ratio.

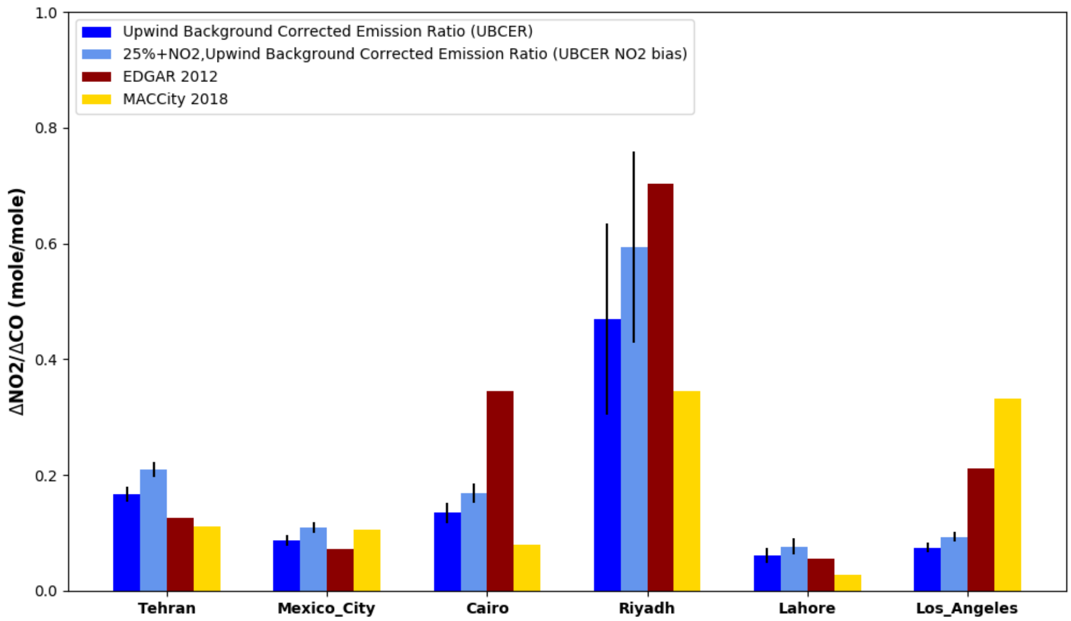

Figure 1. Comparison of TROPOMI-derived Upwind Background Corrected emission ratio shown in blue shades, to corresponding emission ratios from the EDGAR (red) and MACCity (yellow) emission inventories for six *megacities*. Error bars represent 1σ uncertainties calculated using boot strapping method.

- Table 1 settings: I am puzzled by the different definitions for different cities, especially as Line 146 states that the settings are "not critical". So why are they different at all? Do you need to tune the area definitions in order to get the right results??? How do the results look like if a consistent setup is chosen for all cities? Why is the upwind area for Riyadh different for dlat and dlon by a factor of 30?

Author Response:

The sentence " Every city has a different size and different neighboring CO and NO2 emission sources and therefore the appropriate choice of radii for the background and outskirt areas varies between cities (detail explanation in Supplements Section 1). This is important mostly to have a significant signal from city emissions in CO and NO2. However, since the same regional definition is used for NO2 and CO, the enhancement ratio is not so sensitive to the details of the region selection." is added in the line 154 to 158.

We performed the sensitivity analysis where background and outskirt radius is increased by 10km for four times results in the ratio change by 20 % for Riyadh and < 10 % for all the cities (see Figure S21).

There was a typing error for dlat and dlon of Riyadh. To maintain the consistency now, I am using 1.0˚, 1.0˚ dlat and dlon for all the cities.

Page 6:

the methods are explained in plain words, but not illustrated for real data. So Fig. S2 should be moved to the main text, and the background/upwind regions etc. should be marked in this plot. In addition, the ERA wind vector should be added. The rotated patterns and the percentiles used for the second approach should be provided in a separate figure.

Author Response:

Figure1 is added in the paper, to illustrate the upwind background method for real data as suggested by the reviewer. For the plume rotation method, see the Figure S8.

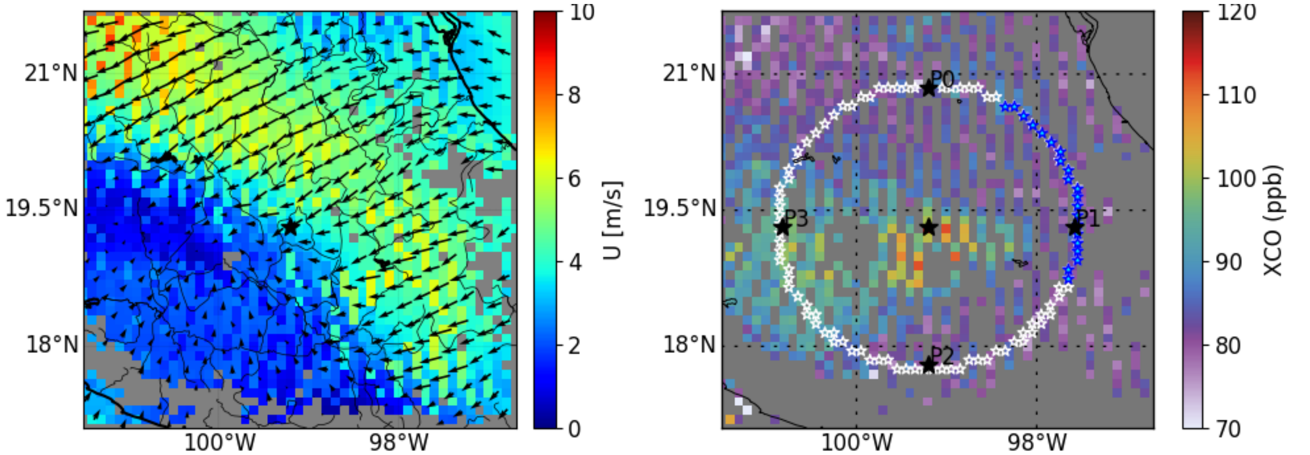

Figure 2. ERA interim average wind speed and direction from surface to 200m at the time TROPOMI overpasses (left) and TROPOMI derived CO total column over Mexico City (right) for 5[th] of June, 2018. The black star represents the centre of the city. In the right panel, the white circle is the background area for Mexico City and the blue section represents the upwind background area that we selected depending upon the wind direction in the core city area. P0,P1,P2 and P3 are the points where north, east, west and south wind directions intersects at the inner rim of the background area.

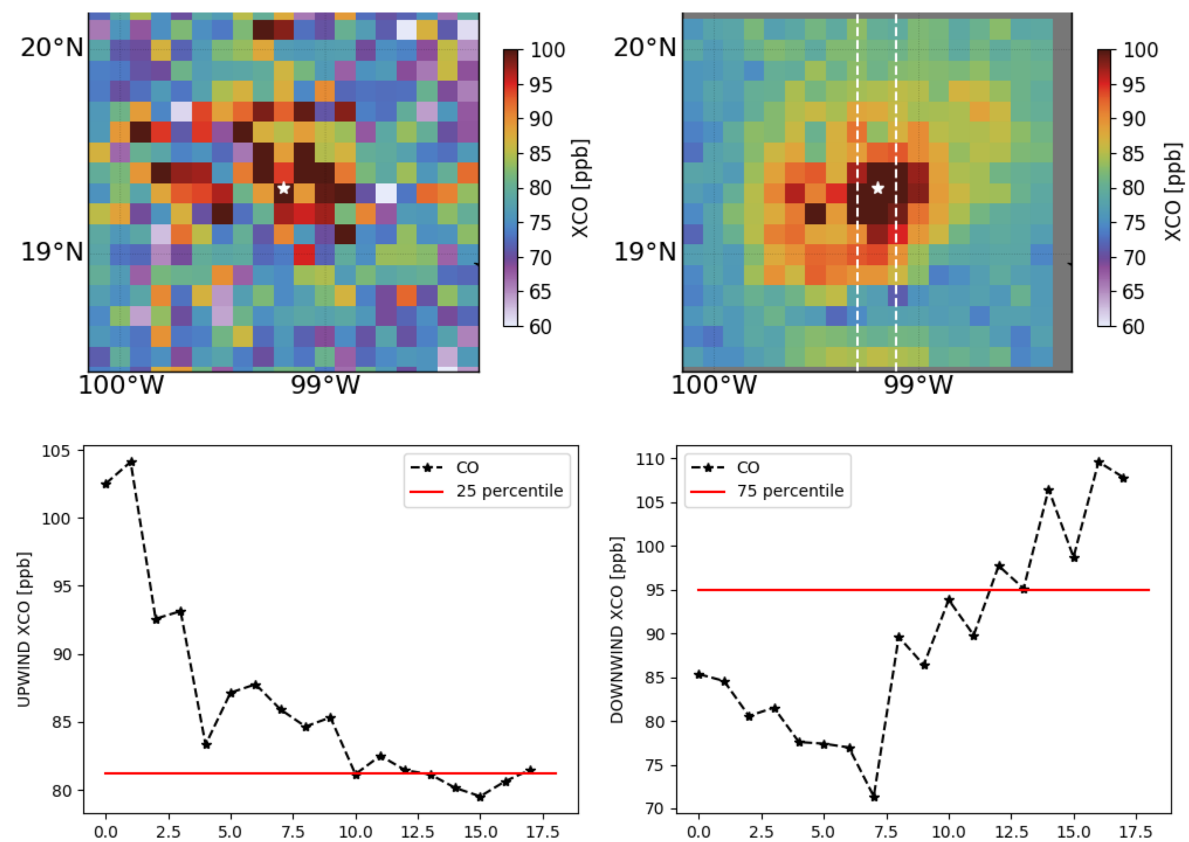

Figure S8. TROPOMI derived XCO a) averaged over June-August 2018 b) plume rotated over Mexico, c) Upwind and d) downwind region. The white star in top panel is the centre of Mexico City. The white lines in panel (b) represent the 20x100km^2 area to determine the column enhancement in the city. The area to the North and South of the city centre is upwind and downwind region respectively. The red line in panel c) and d) represents the 25 percentile and 75 percentile respectively. CO retrievals are gridded in 0.1˚x0.1˚

both methods compare columns "upwind" and "downwind" of the investigated cities. This approach requires that there *is* transport taking place. Wind speeds for Mexico City are quite low, as can also be seen in Fig. S2. So did you consider a minimum threshold for the wind speed? I expect that it would help to remove inconclusive days.

Author Response:

In this study we do not use a minimum threshold for the wind speed, because depending on the choice which would inevitably be quite arbitrary and a substantial amount of useful data may be lost. However, as explained before, by studying the ratio between NO2 and CO we are less sensitive to transport issues. Most important is to have a method the quantifies the difference between city and background in a way that is consistent between CO and NO2.

- **Line 157: with Eqs 2&3, daily ratios are calculated. But how is the total ratio (shown in Fig. 3) derived? Is it the mean of all daily ratios? This is by definition different from the second approach, where first CO and NO2 are averaged and then the ratio of means is calculated. Thus, also for approach 1, the ratio of means should be taken.**

Author Response:

In the upwind background method, the mean is taken of daily ratios. This method is favorable over the use of mean NO2 and CO in the plume rotation method, because it accounts for temporal correlations between NO2 and CO. However, for the plume rotation method we choose to stay consistent with Pommier et al (2013), which averages first since it is important when using noisy MOPITT data. The implication is that there is the inconsistency that the reviewer mentions. However, by including it in the comparison, we also implicitly test the robustness of the TROPOMI derived ratio to this methodological difference.

- Line 196: What emission database is used by CAMS? EDGAR? MACCity ? Or something else ? How far does this affect the following interpretation and discussion of CAMS OH? How do CAMS spatial patterns of CO and NO2 compare to TROPOMI? Please provide a Figure in the Supplement. Is TROPOMI CO and/or NO2 assimilated in CAMS?

Author Response:

CAMS is using MACCity for the anthropogenic emission. CAMS OH concentration depends upon the various chemical schemes used for the simulation rather than emission inventories (V. Huijen et al., 2019). The spatial pattern of NO2 of CAMS and TROPOMI are in good agreement for the six different cities (see Section 5 in Supplements Fig S11 to Fig S16). However, the spatial distribution of CO shows differences for Tehran, Cairo, Riyadh and Lahore (see Section 5 in Supplements Fig S13 to Fig S16). CAMS is using MOPPIT for CO and for NO_2, SCIAMACHY, GOME2 and OMI data are assimilated. TROPOMI CO and NO2 are not yet included in CAMS.

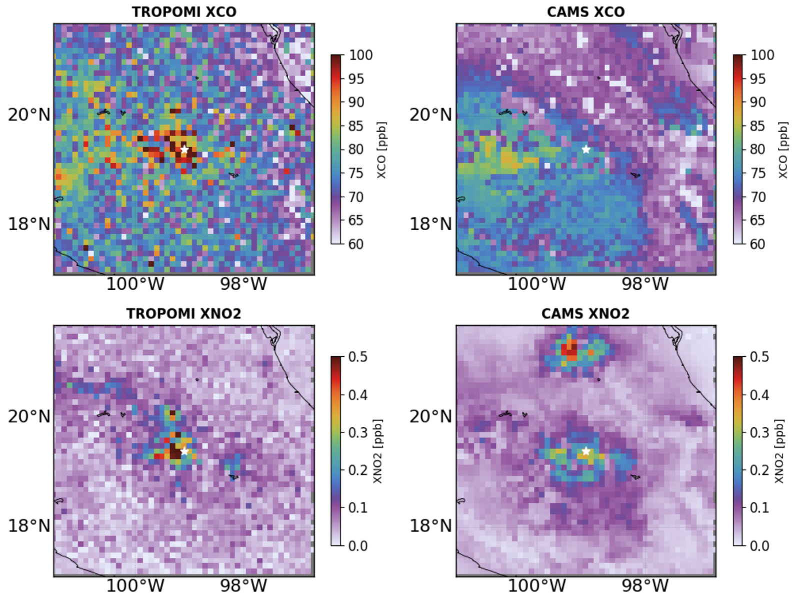

Figure S12. Collocated XCO (top) and XNO2 (bottom) averaged for June-August, 2018 over Mexico City and derived from TROPOMI (left) and CAMS (right). The white star represents the centre of the city. The enhancement of XCO and XNO2 in TROPOMI and CAMS collocates.CO and NO2 retrievals are gridded at 0.1˚x0.1˚ resolution.

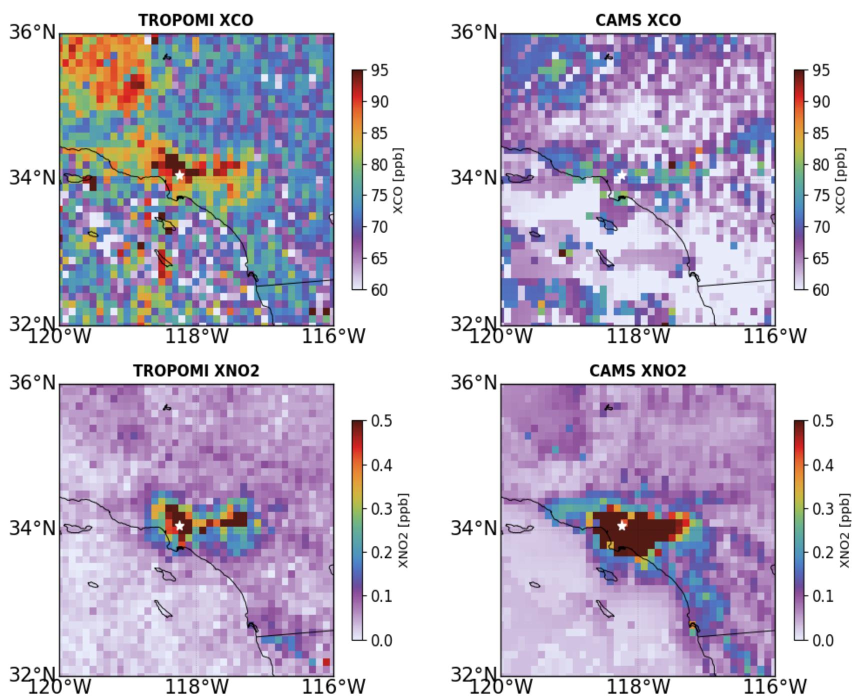

Figure S13. Same as FigS10 but over Los Angeles

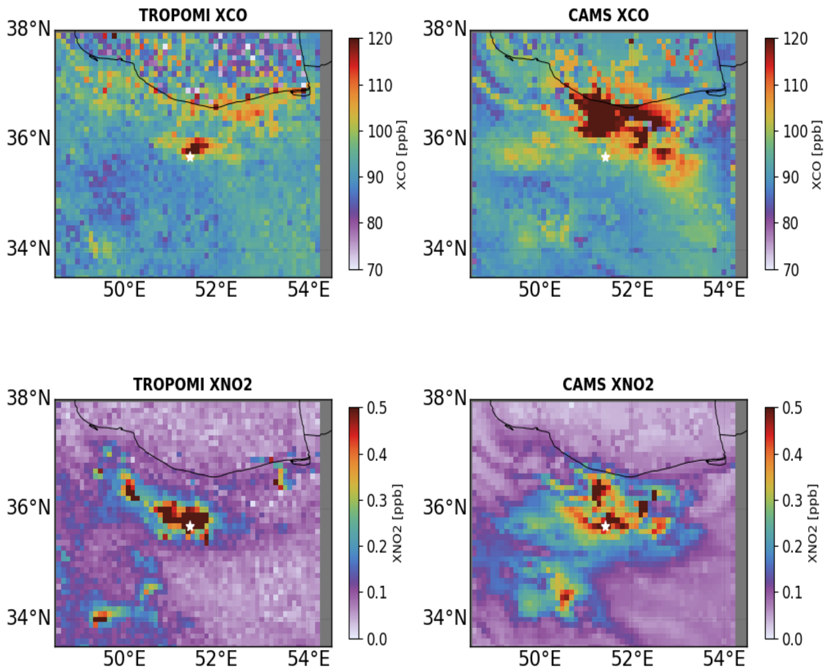

Figure S14. Collocated XCO (top) and XNO2 (bottom) averaged for June-August, 2018 over Tehran and derived from TROPOMI (left) and CAMS (right). The white star represents the centre of the city. The enhancement of CAMS XCO does not collocate with the TROPOMI XCO at the centre of city whereas NO2 enhancement collocates with each other. CO and NO2 retrievals are gridded at 0.1˚x0.1˚ resolution.

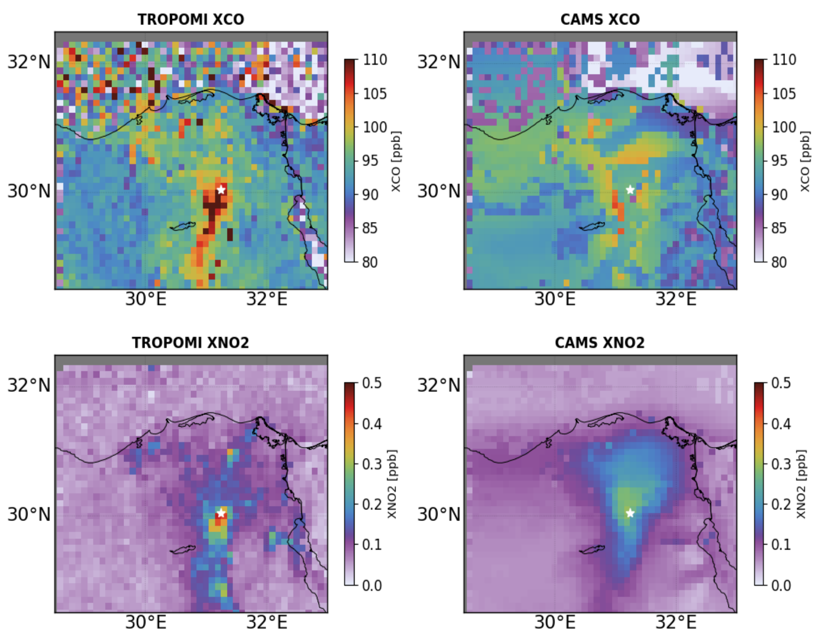

Figure S15. Same as Figure S12 but over Cairo

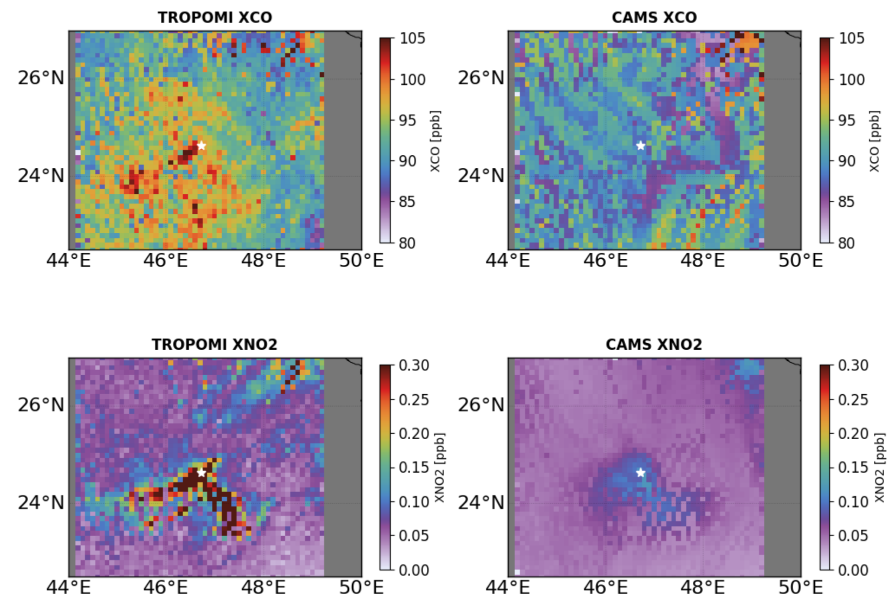

Figure S16. Same as Figure S13 but over Riyadh

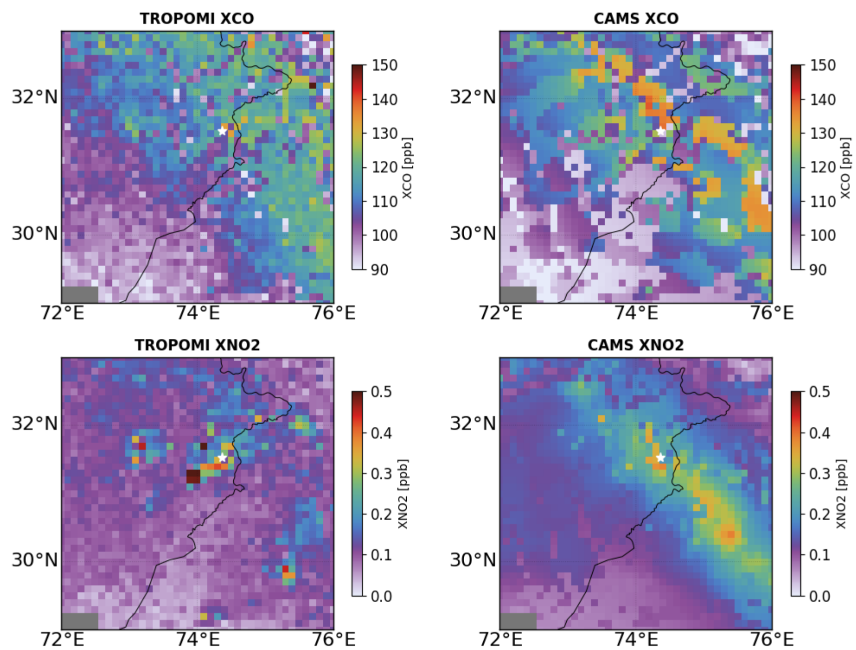

Figure S17. Same as Figure S13 but over Lahore

2.6.1: The bootstrapping approach evaluates the statistical uncertainty for the results with the chosen approach. But on top, there are also other uncertainties, like systematic effects introduced by the definition of radii etc. In particular the uncertainties of the wind direction and wind speed have to be discussed as well.

Author Response:

As suggested by the reviewer, we tested the sensitivity to the wind speed and direction by choosing different heights (i.e. 200m to 1000m), resulting in differences < 10 % for all the cities (see Figure S19, S20).

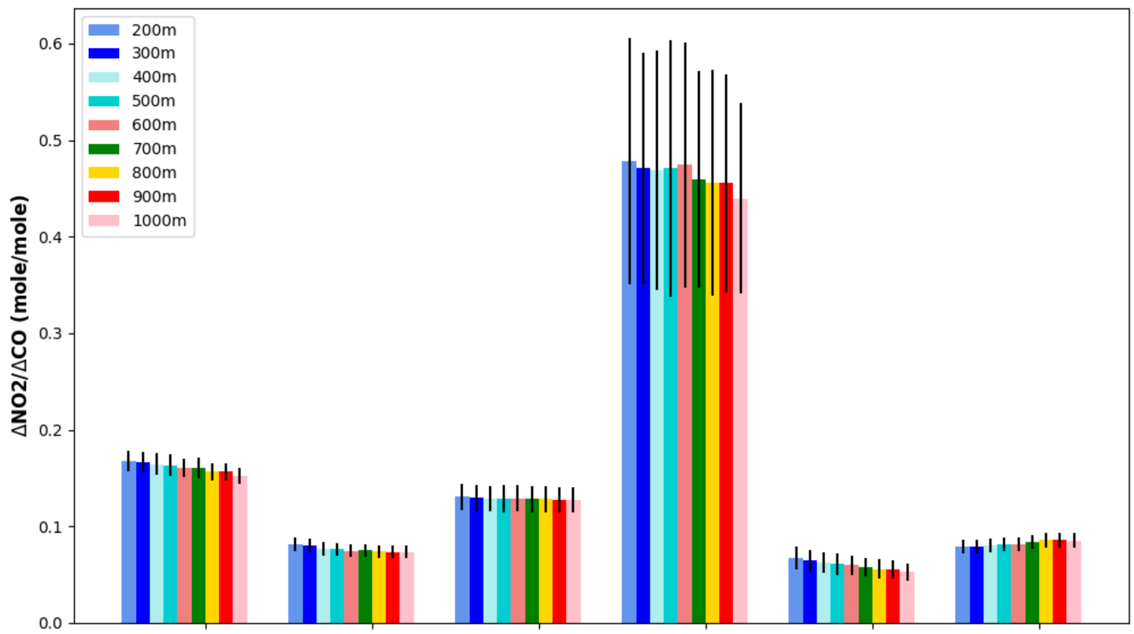

Figure S19. TROPOMI derived Upwind Background Corrected Emission Ratio for six megacities using average wind speed and direction calculated from surface till 200m to 1000m. The error bar represents 1σ uncertainties calculated using boot strapping.

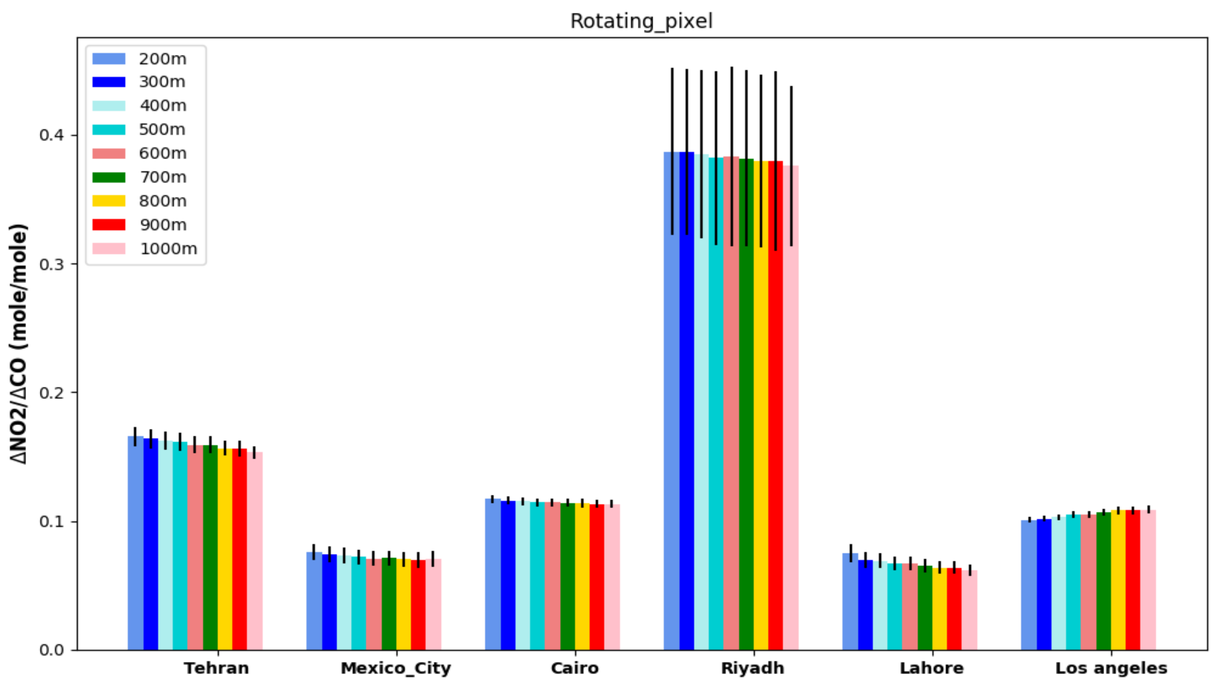

Figure S20. Same as Fig S11 but the emission ratio is Plume Rotation Corrected Emission ratio

We performed the sensitivity analysis where background and outskirt radius is increased by 10km for four times results in the uncertainty of 7-20 % in TROPOMI column enhancement ratio for six megacities (see Figure S21).

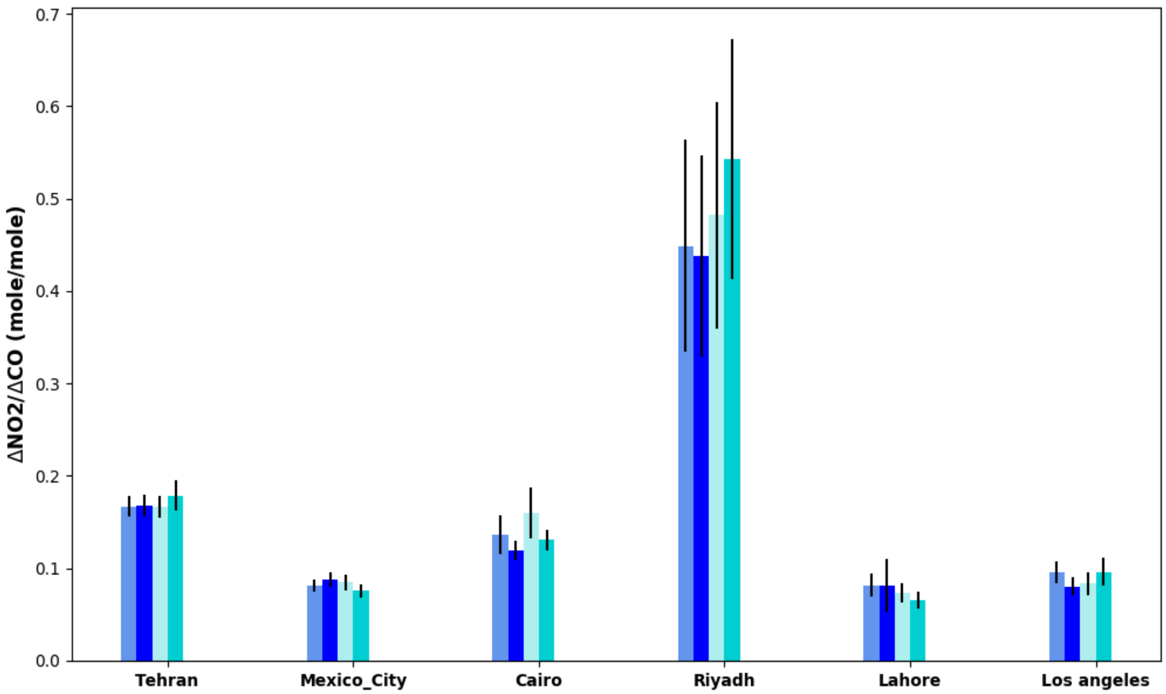

Figure S21. Upwind Background Corrected Emission ratio derived for six megacities using four different background and outskirt radius. For the initial step the outskirt and background radius for Tehran: 180 km and 190 km, Mexico City: 170km and 180km, Cairo: 135km and 145 km, Riyadh: 100km and 110 km, Lahore: 100km and 110km and Los Angeles: 200 km and 210 km. In every step the background and outskirt radius is increased by 10km.During this process dlat and dlon is 1.0˚,1.0 ˚.

Figure 3: Please estimate the uncertainties of NO2 lifetime and AK correction and provide error bars for the CER results as well. I expect that these uncertainties are far higher (and thus more relevant) than the purely statistical bootstrap uncertainties.

Author Response:

Table S3 shows the different sources of uncertainty (i.e. NO2 lifetime, Ak correction, wind direction and their contribution to the TROPOMI derived emission ratio). Error bars indicating the 1σ uncertainties in TROPOMI derived emission ratios are added in Figure 4. The following sentence is added to line 347 to 355 explaining the different sources of uncertainty and their effect on TROPOMI derived emission ratios:

"We calculated the wind direction and wind speed at different height i.e. 200m to 1000m and the ratio changes<10 % for all the cities(FigS19 and S20). The initial uncertainty for CAMS OH was ±50 % (V. Huijen et al., 2019). The bootstrapping method show that the concentration of OH varies from 8.0 – 15 % for six different megacities resulting similar uncertainty to the TROPOMI derived emission ratio. If the CAMS overestimate OH concentration systematically, the TROPOMI derived emission ratio will decrease. To estimate the effect of predefined areas as background, we simultaneously increase the outskirt and background radius by 10 km for all the cities for four times. The effect is about 20 % for Riyadh whereas for other cities, the effect is < 12 % (Fig S21). S5P TROPOMI NO2 retrievals have the largest contribution for the total uncertainty on satellite derived emission ratio. The wind direction and speed, boundary layer OH concentration, $A_{influence}$ correction and the predefined background setting contributes the negligible uncertainty on the TROPOMI derived emission ratio. The total

uncertainty calculated using error propagation method for TROPOMI derived emission ratio ranges from 27 to 35 % and the detail is provided in Table S3."

Table S3. Sources of uncertainties for TROPOMI derived emission ratio. The total uncertainty is derived by the error propagation.

City	S5P TROPOMI NO_2 uncertainty (%)	Wind direction and Wind speed (%)	Boundary layer OH concentration (%)	Predefined background area (%)	$A_{influence}$ correction (%)	Total effect on ER (%)
Tehran	25	1.5	8.4	10	1.2	±28.2
Mexico City	25	1.5	10	7	2.7	±27.9
Cairo	25	2.6	8.4	10	4.4	±28.6
Riyadh	25	2.0	12.5	20	4.1	±34.6
Lahore	25	6.5	15.0	12.0	0.4	±32.19
Los Angeles	25	4.0	8.3	12.5	4.2	±29.7

- 3.2ff: Please check the discussion and conclusions (a) for NO2 probably being biased low and (b) according to the quality of CAMS emissions and the agreement between TROPOMI and CAMS

Author Response:

Discussion section: Lines 346-348 discuss the importance of the bias in NO2. The sentence is as follows:" Additionally, TROPOMI underestimates NO_2 column by 7 % to 29.7 % relative to MAX-DOAS ground based measurement in European cities (Lambert, et al., 2019). However, since we don't know yet how representative this estimate is for the cities that we study so, the impact of the bias is accounted as an additional the source of uncertainty of 25% of the TROPOMI inferred NO2/CO ratio (see Table S3)".

Lines 318-320 describe the agreement between TROPOMI and CAMS over Mexico City and Los Angeles. The sentence is as follows:
"CAMS derived enhancement ratio for Mexico City differs by 5 % compared to UB and PR but for Los Angeles the ratio (0.094) is higher by 75% compared to UB and PR (0.034)."

Conclusion Section:
-Line 428 to 430 explains about the uncertainties and NO2 being biased low. The sentence is as follow: "The total uncertainty on TROPOMI derived emission ratio ranges from 27 to 35 %. The bias in S5P TROPOMI NO2 retrievals accounts for the major contribution for the uncertainties in the TROPOMI derived emission ratio".

Line 434 to 435 explains about the agreement between TROPOMI and CAMS over Mexico City. The sentence is "CAMS derived enhancement ratio over Mexico City differs by 5 % compared to UB and

PR". Line 438 to 439 explains about the disagreement between TROPOMI and CAMS over Los Angeles. The sentence is as follow:
"CAMS derived enhancement ratio for Los Angeles is higher by 75 % in contrast to UB and PR "

Minor comments:
- Lines 40-42: The references stating the high uncertainty of Chinese emissions are from a time period where development in China was vastly increasing. Meanwhile, NOx emissions have been reduced, and the awareness of air pollution has increased in China. I would thus assume that these high uncertainties do not hold any longer.
Author Response:

- The uncertainty in emission estimates of 2005 to 2008 is added in the sentence.

- Lines 60-61 Please provide refs to SCIAMACHY (Bovensmann) and TROPOMI (Veefkind).
- Changed as suggested

- **Line 70: Should be NOx emission**.
- Changed as suggested

- Line 75: Transport disperses NO2 and CO similarly, but the lifetime of NO2 is far shorter! See the different plume extents shown in Fig. S2.
Author Response:
The reviewer is right that the plume extents are different due to the difference on lifetime, which is the reason why we focus on the core city area rather than the full extent of the plume.

- Line 117: Avoid misreading as "the bias is low", e.g. "NO2 is biased low by about

- Changed as suggested

- **Table 1 lat/lon: Please provide consistent number of digits for lat/lon. .01_ should be accurate enough.**
- changed as suggested

- Fig. S1: I don't understand why there is a need for separating 4 different wind directions in the formalism; rotation matrix should work the same for all four cases!?

Author Response:

-changed as suggested and see Figure S7.

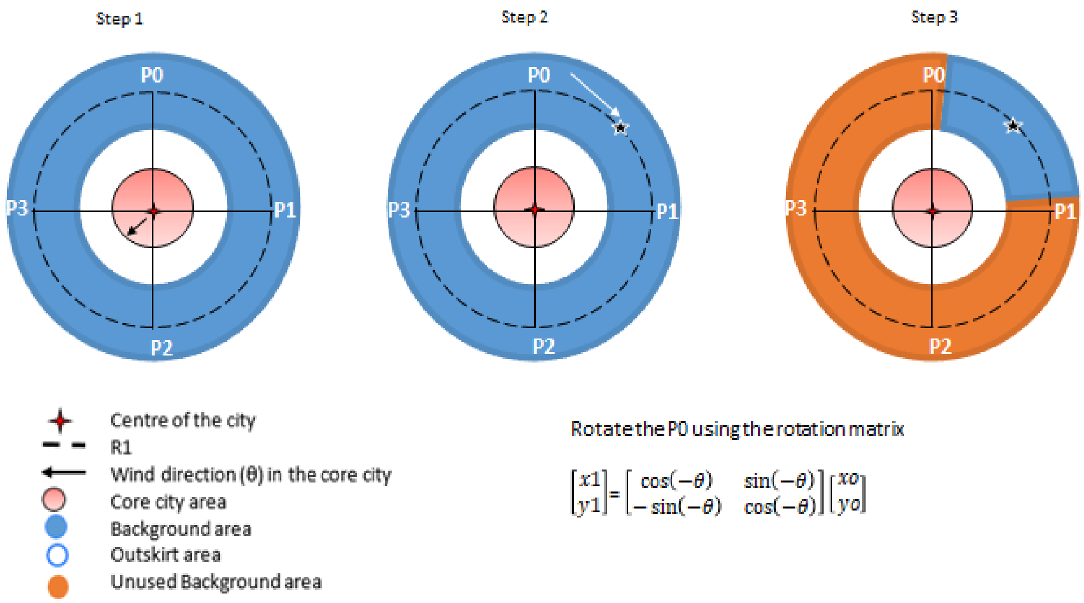

Figure S7. Schematic representation of the procedure used for selecting the upwind background. The centre of the city is represented by red star. The city, outskirt and background radii (see Table 1) are used to divide the city into three parts i.e. the core city (red circle), outskirt (white circle) and background region (blue circle), respectively. Step 1.Selection of radius R1, the mean of outskirt and background radii. Select the points P0, P1, P2, P3 where the north, east, south and west wind directions (**θ**) intersect at the outer rim of the dashed circle with radius R1. The black arrow symbolises an average wind direction over the core city region. Step 2.The rotation of P0 with **θ** in reference to the city centre and generate the new point. Step 3.Select the fraction of the upwind region. The Δlat and Δlon is provided in Table1.