#Reviewer 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 30% of the TROPOMI inferred NO2/CO ratio (see Table S3).

If the 30 % bias would apply to all the cities, then the TROPOMI derived emission ratios increase by factor of 1.30. In that case, TROPOMI-derived ratios remain in line with the emission inventories but the agreement would shift between EDGAR and MACCity depending on the city, for example in favor of EDGAR in case of Cairo and Riyadh. For Los Angeles, the difference between TROPOMI and the inventories would decrease, but remain significantly lower for TROPOMI.

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 following sentence was added (line 154-163): "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 (see Supplements Section 1 for details). Since the same regional definition is used for NO2 and CO, the enhancement ratio is not sensitive to the details of the region selection. Most important for the choice of radii is to catch the local enhancement in CO and NO2 to its full extend, to optimize the signal over noise and thereby the detection limit for urban emissions."

We performed a sensitivity test in which the background and outskirt radii were increased by 10km in four steps resulting in the ratio changes by < 15 % for all the cities (see Figure S20).

There was a typing error for Δlat and Δlon of Riyadh. To maintain the consistency now, I am using 1.0˚, 1.0˚ Δlat and Δlon 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.

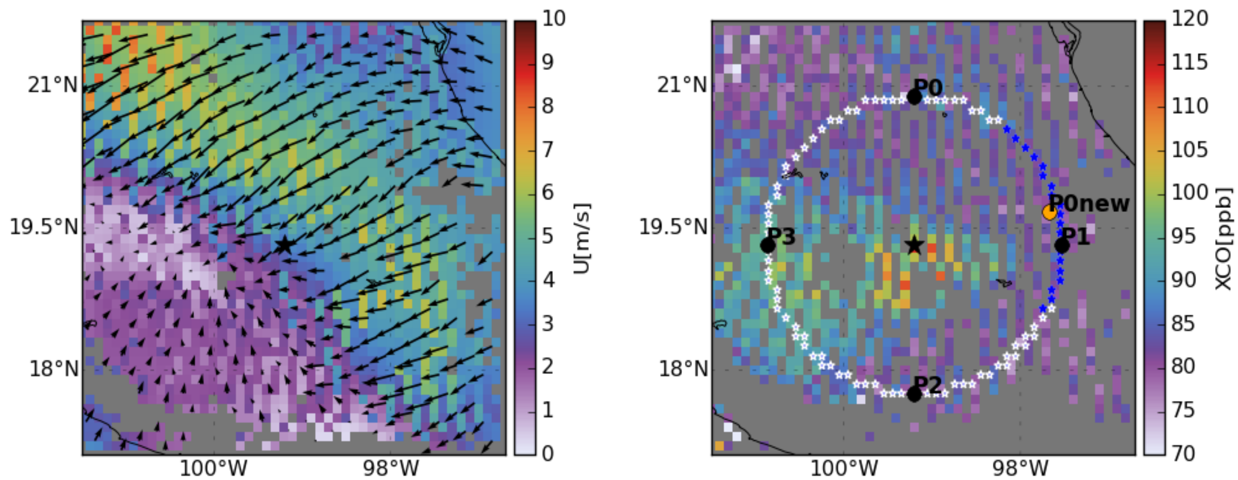

Figure 1. Average wind speed and direction from the surface to 200m from ERA Interim at the time of TROPOMI overpasses (left) and TROPOMI derived total column CO over Mexico City (right) for June 4th, 2018. The black star represents the centre of the city. In the right panel, the white circle is the background area for Mexico City and the blue section represents the upwind background area that we selected depending upon the wind direction (θ) in the core city area. P0, P1, P2 and P3 are the points where the north, east, west and south wind directions intersect with the inner rim of the background area. $P0_{new}$ is the new point generated by rotating P0 with θ in reference to the city centre

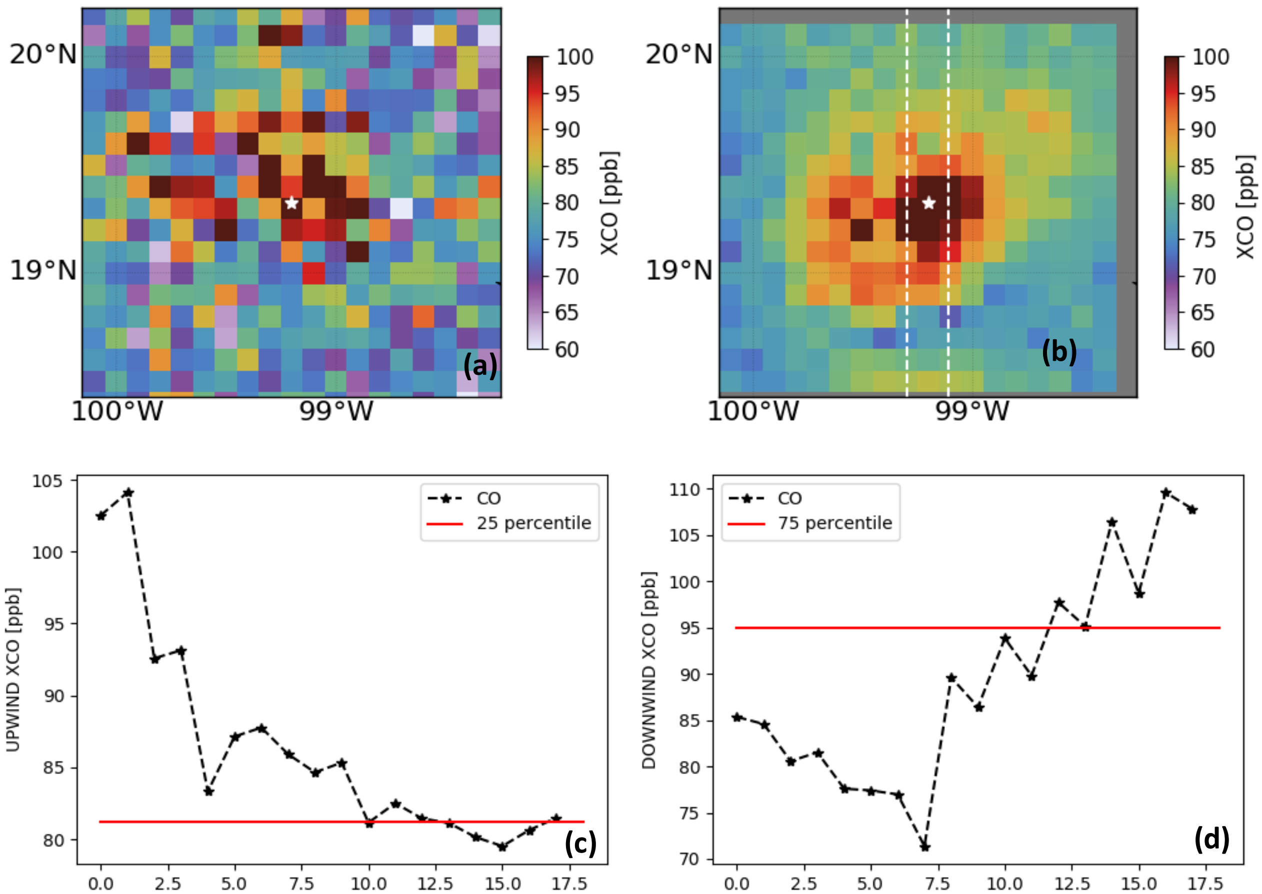

Figure S8. TROPOMI derived XCO a) averaged over June-August 2018 b) plume rotated over Mexico, c) Upwind and d) downwind region. The white star in top panel is the centre of Mexico City. The white lines in panel (b) represent the 20x100km^2 area to determine the column enhancement in the city. The area 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 alsobe 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 uses MACCity for the anthropogenic emission. CAMS OH depends upon the chemical scheme used for the simulation in addition to the emission inventories (V. Huijen et al., 2019). We do not discuss or interpret CAMS OH. We only use it to derive an estimate of the NO2 lifetime, which is valid within the uncertainty bounds of the reanalysis. The spatial patterns of CAMS and TROPOMI NO2 are in reasonably good agreement for the six different cities (see Section 5 in Supplements Fig S12 to Fig S17). However, larger differences are found for CO over Tehran, Cairo, Riyadh and Lahore (see Section 5 in Supplements Fig S14 to Fig S17). 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.

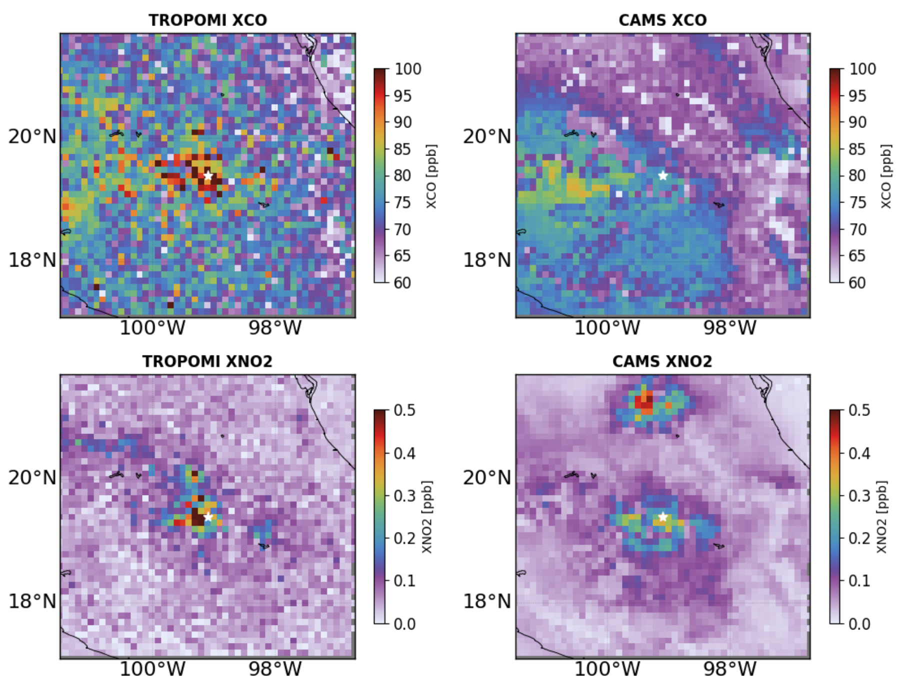

Figure S12. Collocated XCO (top) and XNO2 (bottom) averaged for June-August, 2018 over Mexico City comparing TROPOMI (left) and CAMS (right). The white star represents the centre of the city.

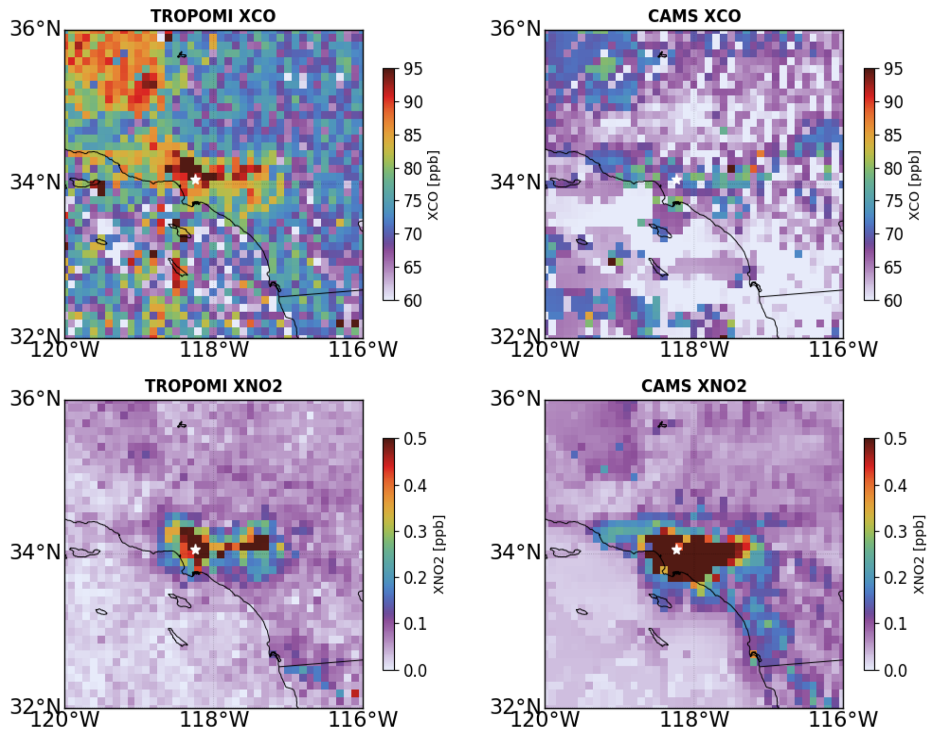

Figure S13. Same as FigS12 but over Los Angeles

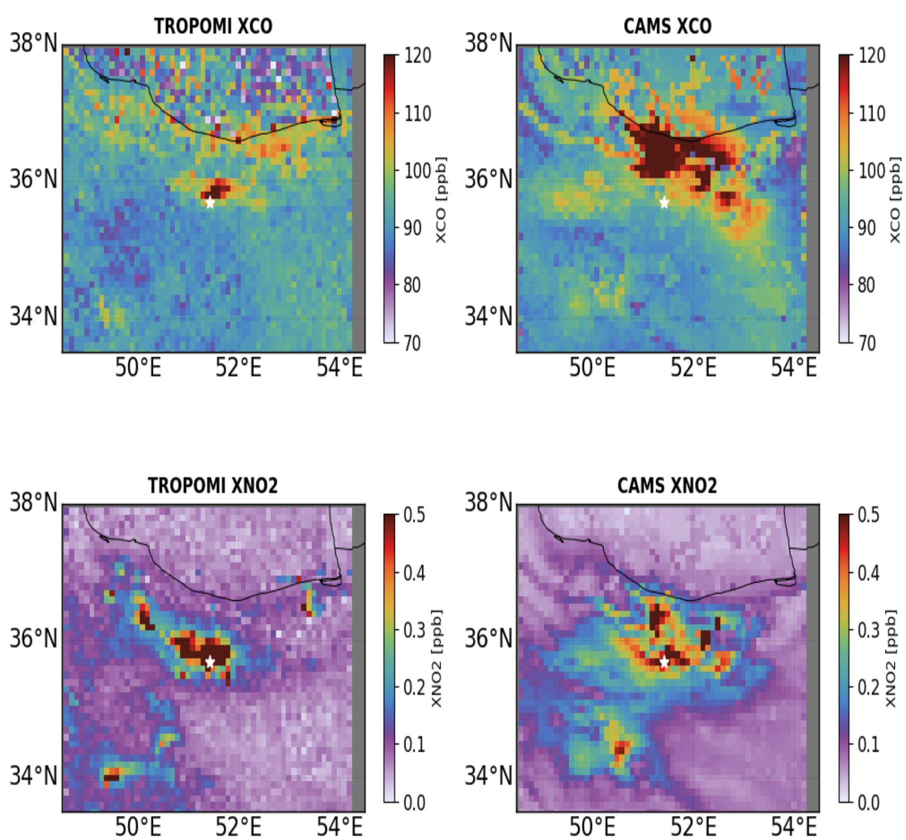

Figure S14. Same as Fig S12 for Tehran.

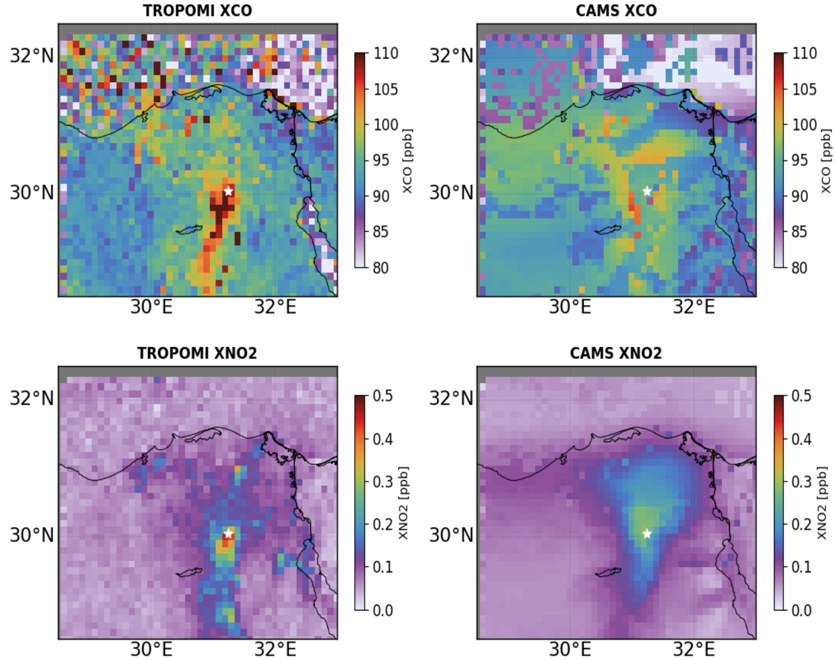

Figure S15. Same as Figure S12 but for Cairo

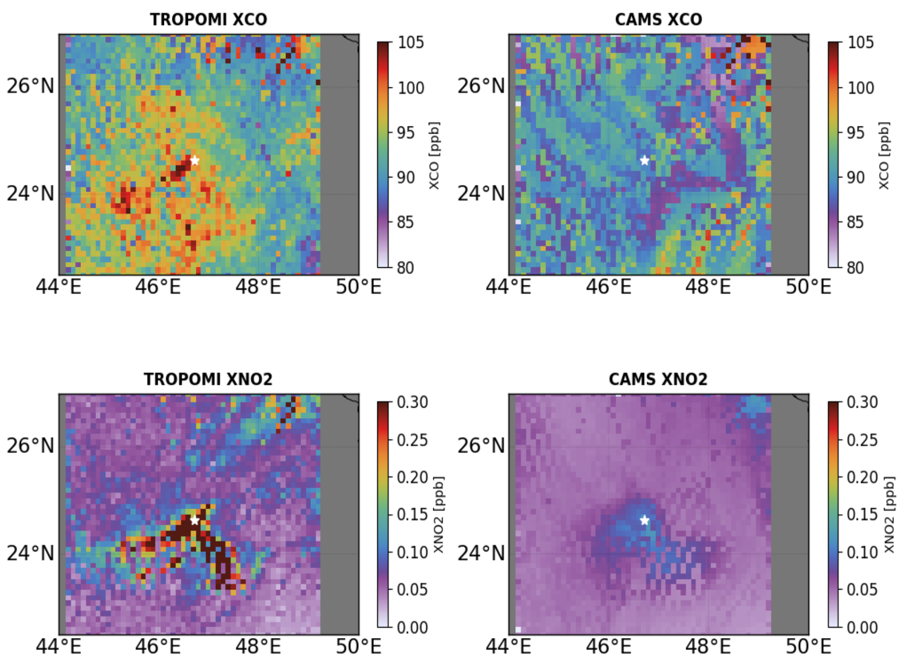

Figure S16. Same as Figure S12 but for Riyadh

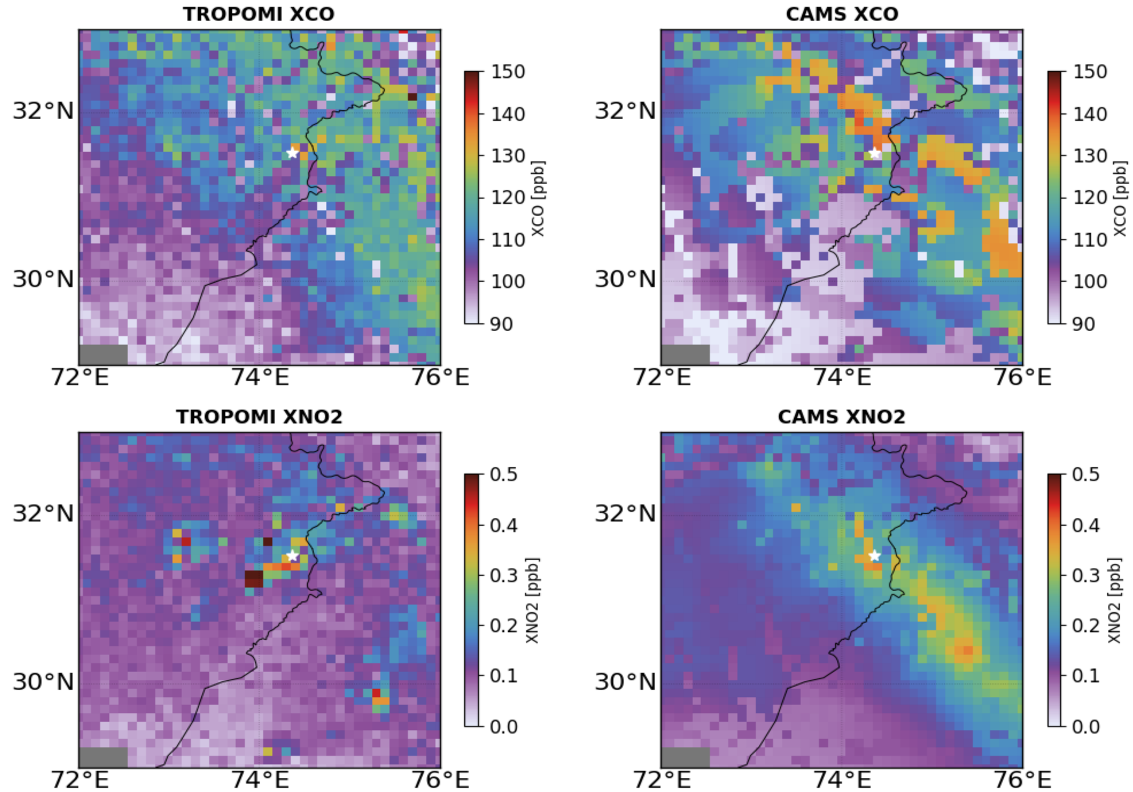

Figure S17. Same as Figure S12 but for Lahore

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 S18, S19).

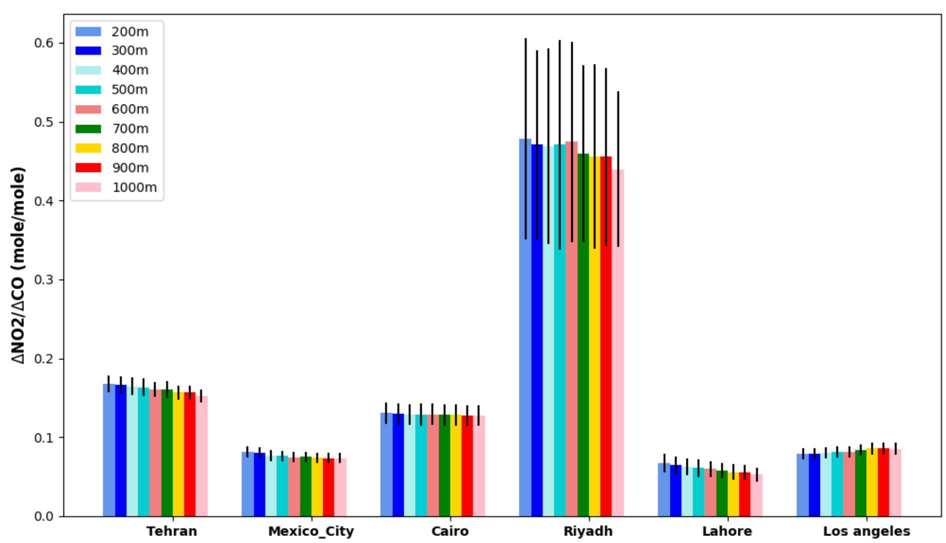

Figure S18. TROPOMI derived Upwind Background Corrected Emission Ratio for six megacities using average wind speed and direction calculated from surface till 200m to 1000m. The error bar represents 1σ uncertainties calculated using boot strapping.

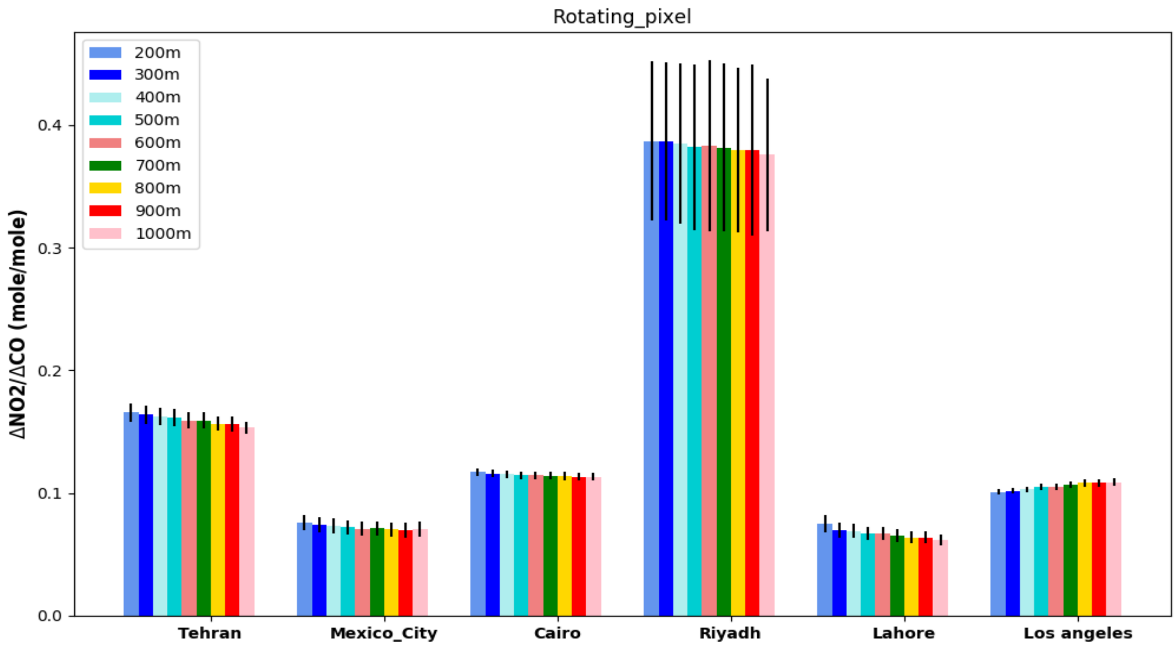

Figure S19. Same as Fig S11 but the emission ratio is Plume Rotation Corrected Emission ratio

We also performed a sensitivity analysis in which the background and outskirt radii were increased by 10km in four steps resulting in an uncertainty of 7 to 15 % in TROPOMI column enhancement ratios for the six megacities (see Figure S20).

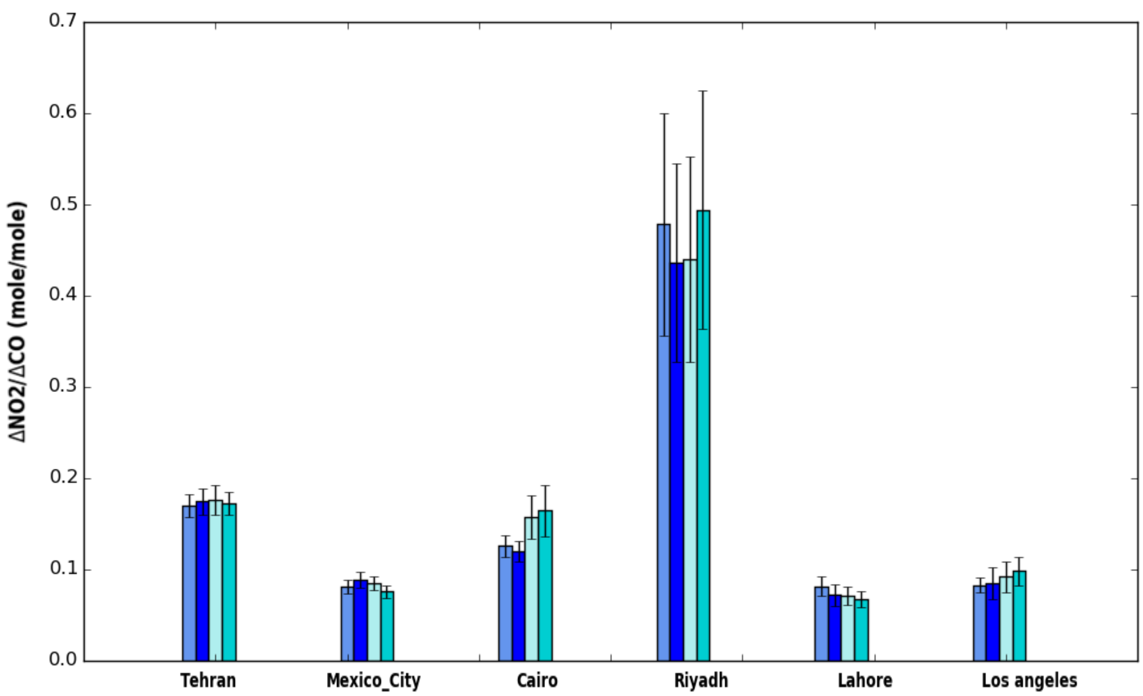

Figure S20. Upwind Background Corrected Emission ratio derived for six megacities using four different background and outskirt radius. For the initial step the outskirt and background radius for Tehran: 180 km and 190 km, Mexico City: 170km and 180km, Cairo: 135km and 145 km, Riyadh: 100km and 110 km, 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 1σ uncertainties in TROPOMI derived emission ratios are added in Figure 4. The following sentence is added to line 355 to 360 explaining the different sources of uncertainty and their effect on TROPOMI derived emission ratios:
"Additionally, TROPOMI underestimates the 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, the impact of this bias has been accounted for as an additional source of uncertainty of 30% in the TROPOMI inferred NO2/CO ratio (see Table S3). Compared to this number, other sources of uncertainty such as in the wind direction and speed (FigS18 and S19), boundary layer OH concentration, $A_{influence}$ correction and the predefined background setting (Fig S20) make only small contributions to the TROPOMI derived emission ratio. The total uncertainty in the TROPOMI derived emission ratio is calculated using error propagation (see Table S3) and ranges between 33 to 35.6%."

Table S3. Estimated uncertainties in TROPOMI derived emission ratios. The total uncertainty is derived by summing the individual components in quadrature

City	S5P TROPOMI NO_2 uncertainty (A) (%)	Wind direction and Wind speed (B) (%)	Boundary layer OH concentration (C) (%)	Predefined background area (D) (%)	$A_{influence}$ Correction (E) (%)	Total effect on ER $(\sqrt{(A^2+B^2+C^2+D^2+E^2)})$ (%)
Tehran	30	10.5	8.4	3.0	1.2	±33.03
Mexico City	30	7.5	10	9.0	2.7	±33.83
Cairo	30	2.3	8.4	13.0	4.4	±34.12
Riyadh	30	6.0	12.5	9.1	4.1	±34.5
Lahore	30	6.5	15.0	10.0	0.4	±35.6
Los Angeles	30	10.0	8.3	8.8	4.2	±34.1

- 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 355-360 discuss the importance of the bias in NO2. The sentence is as follows:" Additionally, TROPOMI underestimates the 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, the impact of this bias has been accounted for as an additional source of uncertainty of 30% in the TROPOMI inferred NO2/CO ratio (see Table S3)."

CAMS use multiple datasets i.e. MACCity emission inventory, SCIAMACHY, GOME2 and OMI data for CO and NO2. We don't know the influence of each of these datasets on the result. Therefore, we decided not to include CAMS in the comparison to TROPOMI derived emission ratios.

Conclusion Section:
- Lines 435 to 436 explain about the uncertainties due to NO2 being biased low:" The total uncertainty on TROPOMI derived emission ratio ranges from 33 to 35.6%.The bias in S5P TROPOMI NO2 retrievals has the most important contribution to the uncertainty in the TROPOMI derived emission ratio".

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:

- We have modified the sentence to clarify which years the uncertainty estimate referred to.

- 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.

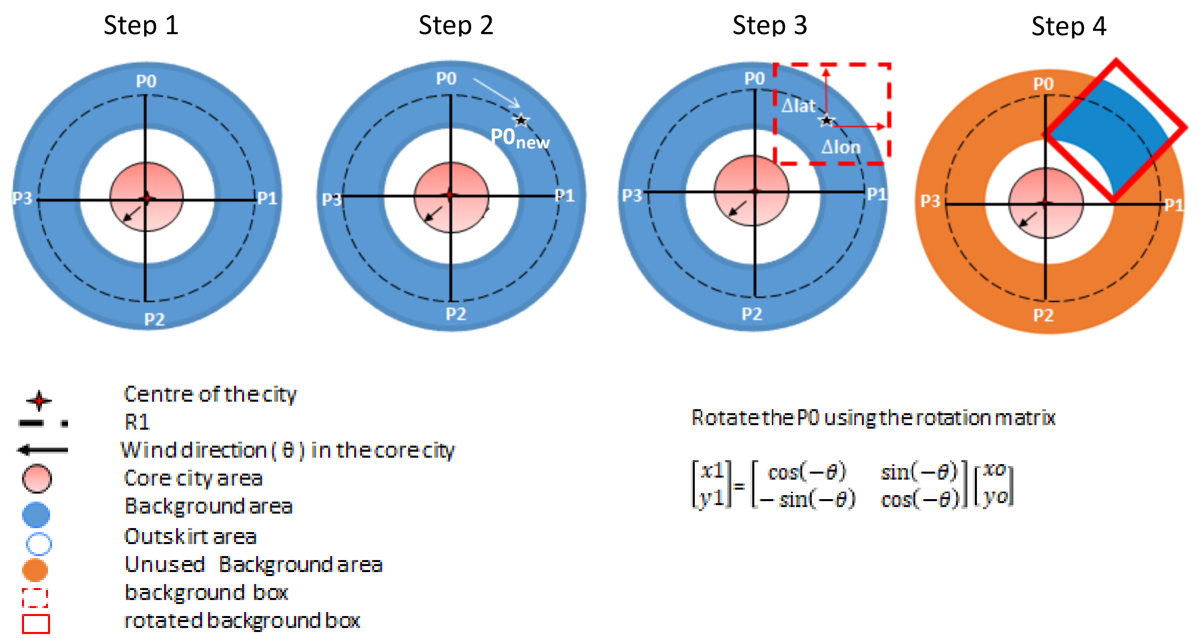

Figure S7. Schematic representation of the procedure used for selecting the upwind background. The centre of the city is represented by red star. The city, outskirt and background radii (see Table 1) are used to divide the city into three parts i.e. the core city (red circle), outskirt (white circle) 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. Step2. The rotation of P0 with **θ** in reference to the city centre and generate the new point ($P0_{new}$).Step 3. Select the square box in the background area using Δlat and Δlon provided in Table1. Step 4. Rotate the square box using wind direction (θ) in the core city area in reference to the $P0_{new}$ and select the fraction of the upwind region.