

Reviewer 1: Quantifying burning efficiency in Megacities using NO₂ /CO ratio from the Tropospheric Monitoring Instrument (TROPOMI)”

This manuscript presents a novel application of satellite measurements of CO and NO₂ to estimate regional-average burning efficiency for a number of large cities. The method is enabled by the capabilities of a relatively new satellite sensor and will likely interest many readers of ACP. With one major exception, the presented methods seem sound and the paper is generally well written.

Thank you for your time and pointing out issues to improve the paper.

General Comments:

To quantify the impact of this effect on Delta (XNO₂)/Delta (XCO) ratios, the authors introduce the variable A_{influence} in Eq. 6. It is unclear how this factor was derived or how it is calculated in practice; no derivation appears either in the main text or Appendices. Presumably, it somehow depends on the TROPOMI CO and NO₂ averaging kernels, but these dependences are not presented. There is a paragraph on the effects of the differing averaging kernels at the bottom of p. 11 (lines 282-290), but this paragraph only adds to the confusion since nowhere does it actually refer to the variable A_{influence}:

Author Response:

A_{influence} is the influence of the averaging kernel (A) on the TROPOMI observed NO₂/CO column ratio. It is derived by calculating Delta (XNO₂)/Delta (XCO) without and with the use of the averaging kernel applied the vertical profiles of NO₂ and CO from the CAMS reanalysis, as follows

$A_{influence} = \frac{(Without A - with A)}{Without A} \cdot 100\%$ and as mentioned in line 305. We have added Appendix C to further clarify how we derive the influence.

In the same paragraph, the authors report that “The CAMS simulated city enhancements averaged over June to August, 2018 did not compare well with TROPOMI for CO, possibly due to the coarse resolution of CAMS. Therefore, to calculate the averaging kernel impact, a few days were selected when CAMS CO and NO₂ enhancements did compare relatively well with TROPOMI.” This gives the impression that the authors’ method of analysing the effects of the averaging kernel differences for CO and NO₂ was based on a small number of ‘cherry-picked’ cases where the higher resolution of TROPOMI (compared to CAMS) was not an issue. Thus, it appears that the authors are probably underestimating the uncertainty of the averaging kernel-related error. I believe this entire issue requires more discussion and perhaps more analysis.

Author Response:

The reason for using CAMS is to have realistic vertical profiles of NO₂ and CO over cities, as those vertical profiles determine the impact that the averaging kernels will have on the ratio. What the reviewer calls “cherry picking”, is actually a selection of cases that is representative of the conditions for which we use TROPOMI data in our analysis. This way the CAMS derived averaging kernel impact is expected to be representative of the impact on the actual TROPOMI data we use. The question is how variable this impact is, since averaging kernels generally do not vary much, which has been tested for Tehran, Mexico City, Cairo, Riyadh, Lahore and Los Angeles. The results confirm that A_{influence} is about 10 -15 %. For Mexico City and Los Angeles, we used all the days from June-August, 2018, see Table S2.

Table S2. The CAMS derived influence of the averaging kernel (A) on the total column NO₂/CO ratio. The error bar represents 1σ uncertainties calculated by bootstrapping method.

City	Days	NO ₂ /CO A =1 (B)	NO ₂ /CO TROPOMI A (C)	Relative difference ((B-C)/B)*100 (%)
Tehran	July 03, 04 August 20	0.0158 ±0.002	0.013 ±0.002	9.0± 1.2
Mexico City	June to August	0.024 ±0.003	0.02± 0.0016	9.0±2.7
Cairo	August,09, 15,16,21,22,31	0.082 ± 0.0062	0.074 ±0.00861	10.0±4.4
Riyadh	June03,08,24,26, July 03,05,06,07, 12,19,22,24,28, August 18,19,20,22, 28,30	0.107859 ±0.01	0.09 ±0.03	10.3±4.1
Lahore	June 06 July 06 August 19, 29, 30	0.0047 ± 0.0003	0.003352 ±0.0005	9.0±0.4
Los Angeles	June to August	0.095 ±0.015	0.085 ±0.02	10.5±4.2

Specific Comments

- 1. The actual lifetimes of CO and NO₂ should be discussed somewhere, perhaps in the paragraph that begins on p. 3, l. 75**

The lifetime of CO and NO₂ is discussed in the section 2.5 NO₂/CO emission ratio, Line 200 to 205. There we also explain how we take the NO₂ loss by OH into account, which leads to a short lifetime of NO₂ at the local overpass time of TROPOMI. The much slower photochemical turnover of CO can be neglected on the temporal and spatial scale of our analysis.

- 2. p. 4, l. 103. Rodgers (2000) does not specifically discuss this type of retrieval algorithm and is not really an appropriate reference. Mathematically, averaging kernels play a different role in optimal estimation-based methods (as described by Rodgers) and Tikhonov regularization.**

The reference has been changed to Borsdorff(2018c). This reference is more appropriate as it explains how the CO total column is derived for TROPOMI

- 3. The chosen QA threshold values (0.75 for NO₂ and 0.7 for CO) would seem to allow low-clouds for CO retrievals but not for NO₂ retrievals. Are scenes with clouds excluded from this study because of the stricter QA threshold value for NO₂? Clouds could have a significant impact on the TROPOMI CO column averaging kernels.**

The application of SICOR algorithm to SCIAMACHY CO retrievals with low-level clouds increases the number of measurement with a limited impact on the retrieval quality (Borsdorff

et al., 2018a). In addition, we selected pixels that had valid retrievals for both NO₂ and CO. Therefore, CO and NO₂ will be influenced similarly by the residual availability of clouds.

4. Conceptually, the Upwind Background and Plume Rotation methods seem to have much in common. The text in Sections 2.4.1 and 2.4.2 should somewhere discuss expected differences in the outcomes from these two methods. Are there any obvious pros and cons to each method?

The Upwind Background method is used to calculate enhancement ratios based on single orbits, whereas in the Plume Rotation method column enhancement ratios are computed from CO and NO₂ columns that are averaged for three months. The plume rotation method is used primarily in reference to what was done in the past (Pommier et al 2013), using MOPITT data that had to be averaged for city signals to be detectable. The use of TROPOMI data has the advantage that city enhancements are detected already in single satellite overpasses, which the Upwind Background method helps exploiting. The use of the two methods allows us to quantify the robustness of the emission ratio that we derived from TROPOMI.

5. For the Plume Rotation method, why use the first quartile upwind and fourth quartile downwind concentrations (instead of simple averages for upwind and downwind regions)?

We use the method of Pommier et al., 2013 to compare our own approach with, but decided to make it less vulnerable to outliers by taking quartiles following Silva and Arellano, 2017 instead of 5 max and 5 min data.

Technical Corrections (partial list)

The numeral 2 should be subscripted in 'NO₂' (in Abstract and elsewhere).

Changed as suggested

Throughout the paper, 'mega cities' and 'mega-cities' should be replaced by 'megacities.'

Changed as suggested

p. 2, l. 48. 'depends' should be 'depend'

Changed as suggested

p. 2, l. 55. 'in respect' should be 'with respect'

Changed as suggested

p. 2, l. 66. 'precursor' should be capitalized

Changed as suggested

p. 3, l. 76. 'source' should be 'sources'

Changed as suggested

p. 8, l. 229. 'over passes' should be 'overpasses'

Changed as suggested

p. 8, l., 232. 'life time' should be 'lifetime'

Changed as suggested

p. 9, l. 244. missing Delta symbols before XNO2 and XCO

Changed as suggested

