

Reply to Referee#1

We thank you for your comments and suggestions to improve the paper. We have addressed your concerns as follows:

Page 3809

Line 5: Should read as "...several factors, such as the..." **Line 6: Should read as "...lower troposphere and..."** **Line 6: Should read as "In general, the..."** **Line 9: The term "well placed" is awkward. Perhaps "ideal".** **Line 23: "Therefore" is not needed. Should read as "In this paper, we..."** **Line 25: Acronyms such as TOMS, SBUV, OMI, MLS are never defined.**

The editorial corrections per your suggestion were included in the revised version. Line 9: "well placed" was replaced by "well suited". All the acronyms used in the paper have now been spelt out.

Page 3811

Line 3: The term "As mentioned above" should rarely (if ever) be used. There is rarely any need to repeat statements within a document. If this statement is essential in reminding the readers of an important fact, then simply state the fact without the "As mentioned above". **Line 5: Should read as "Eastern China and Eastern USA..."** **Line 5: Words like "this", "that" and "these" without specifying what the authors are talking about are vague. Perhaps edit to read as "This delineation was possible..."** **Line 15: Consider replacing "depiction" and "one" with "data"** **Line 23: Should read as "general, the plumes..."**

These changes have been incorporated in the revised text.

Line 23: Prior to discussing how far ozone plumes can be advected (Lines 23-26), it would be useful if the authors cited a typical lifetime of ozone in the troposphere

The typical lifetime of ozone in the troposphere is about 2-5 days in the boundary layer and increases to about 90 days in the middle troposphere. We have added this in the text.

Line 27: Should read as "...lower directly over Mexico City and a strong outflow..."

This has been changed as per suggestion.

Comment: The authors qualitatively reason that the ozone plumes are signatures of urban centers based on NO₂ plumes. Although NO₂ and O₃ have different tropospheric lifetimes, the concentrations downwind of the urban centers should correlate to some degree if in fact the statement presented on Page 3812 (Line 20) is true. To quantitatively address this issue, an NO₂/O₃ correlation map should be presented. Additional analyses could include a quantitative comparison of the ozone values upstream versus downstream of these urban centers. The conclusions of the paper are very hard to believe if the results of these quantitative analyses are not shown.

We should emphasize that the plumes seen in the TOR/TCO database can be very different for different cities. Often the maximum of the plume is not centered on the city unlike NO₂ plumes. This was meant to be an exploratory study rather than a comprehensive study of city plumes in satellite ozone data and thus a quantitative or modeling analysis was not attempted which are planned for future studies. In any case we have added a couple of figures to address your suggestions. The new Figure 2 shows a comparison between the upstream and downstream values of TOR/TCO for the 4 city plume examples shown in Figure 1. The TOR/TCO values were sampled along the black lines shown in Figure 1 (added in the revised version) roughly following the wind directions at the 4 cities. The red line in Figure 2 indicates the grid cell closest to the city location. Thus the TOR/TCO values to the left of this line denote the upstream values and the ones to the right denote the downstream values. The TOR and TCO pictures show generally similar behavior although with different amplitudes. There are significant and interesting differences among the cities. The maximum upstream/downstream difference can be seen for Mexico City with the TOR/TCO values increasing sharply near the city and downstream TOR/TCO values remaining nearly 5-7 DU larger than the upstream values for several hundred kms. This indicates the significant regional influence of the Mexico City pollution as has been observed also from aircraft observations. A similar extended plume can be seen for the New York City plume, the downstream TOR/TCO values differing from the upstream values by ~ 5 DU. In contrast the TOR/TCO values near Sao Paulo for October 2005 abruptly rise to very high values compared to the upstream values and seems to be contained within the vicinity of the city itself. Beijing also shows a less extended plume because of low wind speeds and a slow build up probably because of generally high ozone values in the upstream regions of Eastern China.

We have used these same upstream/downstream transects to study the correlation between TOR/TCO and NO₂ tropospheric columns. Figure 4 (added in revision) shows this correlation using all the sampled data for the 4 cities together. A distinct positive correlation can be seen between the ozone and NO₂ columns with a linear correlation coefficient of about 0.6. If we use only the downstream points for each city, then the correlation coefficient increases to 0.7. Note that these are monthly averaged values obtained from two different platforms. The correlation between the tropospheric columns of the two species should depend upon whether the ozone production is NO_x limited or VOC limited and this can vary significantly from city to city and also possibly from event to event. Thus Sillman et al. (1993) in their simulations found the slopes of the O₃-NO_y curves to be different for plumes related to New York-Connecticut, Boston-Maine and over Lake Michigan. In particular for New York city outflow plumes which were observed ~110 km downwind of the city, Kleinman et al. (2000) found a ratio of NO_x/NO_y of about 0.2-0.3 implying that much of the NO_x had been consumed in the plume which had aged by about 3-4 hours. The peak NO_x concentration of ~7 ppbv however was sufficient for active photochemistry in a VOC-sensitive regime leading to a ozone concentration of 100 ppbv. In Mexico City, Tie et al. (2009) found the aged outflow plume to be strongly NO_x sensitive, while the plume is VOC-sensitive near the city. In addition, the correlation will also depend upon the meteorological conditions given the different lifetimes of the two species.

Page 3812

Line 1: Should read as "...low column ozone directly over the city is likely..." Line 18: It would be useful if the authors cited a typical lifetime of NO₂ in the troposphere

Line 1: the sentence was changed as suggested. Lifetime of NO₂ is of the order of hours to days depending on the location, altitude and season. This has been added in the text.

Page 3814

Line 15-16: The sentence "However a caveat should be pointed out in this context." should be removed. Line 22: "etc" should be removed

These changes have been incorporated in the revised version.

Page 3815

Line 12: Combine the sentences starting with "Figure 5" and "These" to remove the use of the vague term of "These" Line 23: Should read as "In fact, Fresno and Visalia are just below..."

The sentence was changed to: "Figure 7 shows a few examples of urban signatures in climatological depictions averaged for the years (2004-2008) of available OMI/MLS measurements". The text was revised in this section to take into account the second referee's suggestion as well. Thus the sentence in line 23 now appears elsewhere and we have deleted "In fact" to frame the sentence properly in the new context.

Page 3816

Line 9: What does the term "away from the city" mean? This statement needs more clarification. For instance, how far from the city? Why was this distance chosen? What would result if the authors choose another distance for the analysis? In which direction is "away from the city" either in geographic coordinates or in relation to the mean wind direction? What would result if the direction "away from the city" changes? This analysis seems to be arbitrary without proper discussion and the results may be considerably biased based on the chosen parameters.

The purpose of (old) Figure 6 was to study the time series of ozone VMR in the vicinity of the city in comparison to a background location. As you have suggested in your earlier comment, one should notice a difference between the upstream and downstream values. However the wind direction is likely to change significantly with season and it is not easy to do this comparison as a function of time. The choice of the distance of this location is also impacted by presence or absence of nearby urban areas. Further, the city might even be the receptor of pollution from outside at certain times of the year. Given all these uncertainties, we had chosen the distance to be 5 degree east or west of the city in the upstream direction as determined for each city from the (old) Figure 5. For Johannesburg this was increased to 10 deg because of the larger plume. A better way to define the background would be taking the average ozone VMR over $\pm 5^\circ$ in latitude and longitude around the city grid cell (excluding the city cell itself), although it may contain the contributions from the outflow plumes from the city at certain times. A similar method was used by Massie et al. (2006) for studying the CO and aerosol plumes at Mexico City

from satellite observations. We are now showing the time series using this background average and have changed the discussion in the text accordingly.

Line 19: The term “footprint” must be defined if it is used. The authors have been using the term “signatures” and it is suggested that they remain consistent.

The word has been changed to ‘signatures’.

References:

Kleinman, L. I., Daum, P. H., Imre, D. G., Lee, J. H., Lee, Y-N., Nunnermacker, J., Springston,

S. R., Weinstein-Lloyd, J. and Neuman, L., Ozone production in the New York City urban plume, *J. Geophys. Res.*, 105, D11, 14495-14511, 2000.

Massie, S. T., Gille, J. C., Edwards, D. P. and Nandi, S., Satellite observations of aerosol and CO over Mexico City, *Atmos. Environ.*, 40, 6019-6031, 2006.

Sillman, S., Samson, P. J. and Masters, J. M.: Ozone production in urban plumes transported over water: Photochemical model and case studies in the northeastern and Midwestern United States, *J. Geophys. Res.*, 98, D7, 12687-12699, 1993.

Tie, X., Madronich, S., Li, G., Ying, Z., Weinheimer, A., Apel, E. and Campos, T.: Simulation of Mexico City plumes during the MIRAGE-Mex field campaign using the WRF-Chem model, *Atmos. Chem. Phys.*, 9, 4621-4638, 2009.