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**Urban ozone
signature from
satellite
measurements**

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Are there urban signatures in the tropospheric ozone column products derived from satellite measurements?

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Abstract

In view of the proposed geostationary satellite missions to monitor air quality from space, it is important to first assess the capability of the current suite of satellite instruments to provide information on the urban scale pollution. We explore the possibility of detecting urban signatures in the tropospheric column ozone data derived from TOMS/SBUV and OMI/MLS satellite data. We find that distinct isolated plumes of tropospheric ozone near several large and polluted cities around the world may be detected in these data sets. The ozone plumes generally correspond with the tropospheric column NO₂ plumes around these cities as observed by the SCIAMACHY instrument. Similar plumes are also seen in tropospheric mean ozone mixing ratio distribution after accounting for the surface and tropopause pressure variations. The total column ozone retrievals indicate fairly significant sensitivity to the lower troposphere over the polluted land areas, which might help explain these detections. These results indicate that UV measurements may, in principle, be able to capture the urban signatures and may have implications for future missions using geostationary satellites.

1 Introduction

The issue of urban pollution and how it affects the regional and even global environment has become increasingly important in view of the fact that nearly 50% of the world population is predicted to be concentrated in large urban centers in the near future (Gurjar et al., 2008). Recent modeling efforts indicate that both horizontal transport within the boundary layer as well as vertical lifting of pollution from megacities can be very important for regional scale pollution (Lawrence et al., 2007). A few megacities like Mexico City have been studied extensively in this context using ground based and aircraft campaigns (e.g., Molina et al., 2007), but future monitoring of urban pollution is likely to employ satellite remote sensing. In this context, it is important to determine if the enhanced pollution signature of cities can, in the first place, be detected using

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the currently available remote sensing observations from space. This question has assumed added significance in view of the planned geostationary satellites which will make high time resolution measurements around many cities.

The detection of urban pollution signatures from space is linked to several factors like the lifetime of the species, the sensitivity of the measurements to the lower troposphere as well as the vertical profile of the species. In general the sensitivity to the boundary layer is only modest, ~15–30% for UV (O_3 , SO_2 , HCHO), <20% for CO measurements in the thermal infra red (TIR) increasing to 40–60% in the visible (NO_2) (Martin, 2008). NO_2 is well placed for studying urban pollution because of its short lifetime, the high source strength compared to the background and relatively enhanced sensitivity. Indeed clear signature of city scale enhancements in NO_2 and even their weekly variations have been studied using the space based observations (Beirle et al., 2003; Boersma et al., 2009). Even for CO with a longer lifetime and reduced sensitivity to the boundary layer, Clerbaux et al. (2008) have shown that some cities can be detected in the time averaged MOPITT thermal IR retrievals if the thermal contrast is high and the city pollution is trapped by nearby mountains. Some city signatures in CO were also observed over India (Kar et al., 2008). NO_2 and CO are both precursors of ozone, which is the primary component of smog and directly affects the health of humans and the ecosystem. However, detection of ozone signature for cities using the tropospheric column data is more difficult because only about 10% of the total ozone column resides in the troposphere. As such, urban scale enhancements in ozone from satellites have so far been explored only in a few cases (Shim et al., 2008; Eremenko et al., 2008). Therefore in this paper we address the question if any possible signature of cities can be detected in the existing satellite ozone data. In previous studies using tropospheric ozone residuals from TOMS/SBUV measurements, the regional nature of ozone pollution has been identified and related to the interannual variability of El Niño in Northeast India (Fishman and Balok, 1999; Fishman et al., 2003, 2005) and to crop productivity in the Midwestern US (Fishman et al., 2009). Although the use of these instruments were not intended to observe such small scale features in

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the troposphere, it is apparent from these studies that clear lower atmospheric signals are present in these datasets. This study expands on these findings by concentrating on even smaller scale studies to explore the potential utility of such measurements for studying urban-scale features. Such a study is critical for determining whether or not the current observational techniques might meet the needs of future instruments on a geostationary platform.

2 Data

The primary technique employed to obtain the tropospheric ozone column from satellite measurements is to subtract the stratospheric column ozone (SCO) from the concurrent total ozone column measured by a TOMS like instrument and over the years several such products have become available (Fishman et al., 1990, 2003; Ziemke et al., 2006; Schoeberl et al., 2007). We have used the data products derived using two of these methods. In the empirically corrected tropospheric ozone residuals (TOR), the SCO is determined by an empirical correction applied to the tropospheric profile as retrieved by the SBUV measurements and subsequently the resulting SCO is subtracted from the TOMS total column measurements (Fishman and Balok, 1999). Monthly mean fields of TOR are available and encompass the time period from 1979–2005 with missing data for the years 1994–1996. We also use the tropospheric column ozone (TCO) database derived by Ziemke et al. (2006) from the OMI and MLS instruments after cross calibrating the MLS SCO with OMI SCO obtained by the convective cloud differential method. These data have similar spatial resolution as the TOR data ($1^\circ \times 1.25^\circ$) and are available from October 2004 onwards as monthly means. We also use tropospheric column abundances of NO_2 retrieved by the DOAS algorithm from SCIAMACHY measurements (Richter et al., 2005) as well as the OMI level 2 data for total ozone and efficiency factor. The wind data have been taken from the NCEP reanalyses.

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3 Results

3.1 Urban signatures in TOR and TCO data

As mentioned above, the empirically corrected TOR data delineated high tropospheric ozone in regions of the globe with high population densities like the Indo Gangetic basin, Eastern China, Eastern USA (Fishman et al., 2003). This was possible because of the high spatial resolution afforded by that technique using the TOMS/SBUV measurements. Figure 1 shows some examples of possible urban plumes detected in the tropospheric ozone residual data. The upper panels show the TOR distribution in the vicinity of Beijing (August 2005), New York (July 2005), Sao Paulo (October 2005) and Mexico City (February 2005). In the lower panels, the same cities are shown in the TCO data. Enhanced tropospheric ozone observed simultaneously in both the datasets indicates that the plumes may be associated with the cities and not artifacts of the retrieval. Note that the upper limit of the color scale has been changed for each city in order to clearly delineate the plumes. Except for Sao Paulo, the other cities show stronger plumes in the TOR depiction than in the TCO one and generally show similar orientation of the plumes. The winds shown in the plots correspond to 850 hPa for Beijing, New York and Sao Paulo and 700 hPa for Mexico City. For Beijing the winds are rather weak in August 2005 which would tend to promote ozone photochemistry. For New York City, the orientation of the ozone plume is consistent with the prevailing westerlies and extends over the Atlantic Ocean. Ozone plumes extending north east to Massachusetts and Maine originating from the New York plume in summer have been reported from air craft measurements (Sillman et al., 1993; Spicer et al., 1979). In general the plumes at all the cities are quite extended. Ozone plumes associated with cities have been observed hundreds of kms away, from in-situ measurements and are generally understood as the result of photochemical production downwind from a city (Sillman et al., 1993; Goncalves et al., 2009, etc.). In particular, note that both TOR and TCO values are lower over Mexico City itself and a strong outflow plume following the wind direction can be seen to the north east of the city extending into the Gulf of

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Mexico. The low column ozone over the city itself is likely due to the high elevation of the city which is surrounded by mountain ridges on three sides and has a broad opening to the north. Low values of TOR over mountains and other elevated regions have been observed earlier in the empirically corrected TOR data (Fishman et al., 2003).

5 This outflow plume to the north-east of the city has been observed persistently for various months and years of observation in the TOR as well as TCO data. In particular, the location of the outflow plume is quite similar to the aged or secondary Mexico City plume as observed by the in-situ aircrafts during March 2006 under the MILAGRO campaign (Shim et al., 2009; Mena-Carrasco et al., 2009). The simulations of Mexico
10 City outflow indicates that the ozone production could be about 6 times more efficient in the aged plume than in the young plume near the city (Tie et al., 2009). The city ozone anomaly is about 10 DU in TOR distribution compared to the background for Beijing, New York and Sao Paulo and 5–6 DU for the Mexico City outflow plume.

15 Figure 2 shows the corresponding urban plumes at the 4 cities as observed in the tropospheric column NO_2 retrievals from the SCIAMACHY instrument for the same months. The NO_2 columns are more sensitive to the boundary layer and the city plumes are observed in the immediate vicinity of the urban centers because of the low lifetime of NO_2 . Note the general correspondence between the tropospheric ozone and NO_2 column plumes as well as the much larger maximum values in both species for Beijing
20 and New York. This would lend credence to the plumes seen in TOR/TCO distributions being the signatures of city ozone plumes.

3.2 OMI layer efficiency

25 The capability to detect significant ozone anomalies in the lower troposphere from the residual ozone technique should be linked to the vertical sensitivity of the total ozone column retrievals from the TOMS or OMI measurements. The latter has been quantified in terms of an “efficiency factor” and is reported in the level 2 data products of TOMS

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(version 8) and OMI. In this scheme the best ozone column retrieved is formulated as

$$\text{Best Ozone} = \sum_{j=0}^{11} E_j x_j + (1 - E_j) a_j \quad (1)$$

where E is the efficiency factor, x is the true ozone profile and a is the a priori ozone profile. The efficiency factor is reported in the TOMS/OMI level 2 products as “layer efficiency” at 11 levels corresponding to the mid points of the Umkehr layers (at 2.8, 7.9, 12.5, 17.0, 21.3, 25.8, 30.4, 35.2, 40.2, 45.5, and 51.0 km) and represents the sensitivity of the total ozone column to perturbations in a particular layer.

Figure 3 shows the monthly mean profiles of the OMI layer efficiency near Beijing (August 2005), New York (July 2005), Sao Paulo (October 2005) and Mexico City outflow plume (February 2005) up to 20 km as obtained from the OMI level 2 database for nearly cloud free conditions (reflectivity <0.3). Note that at 2.8 km, the layer efficiency is near 0.5–0.6 with highest values for Beijing and New York. These values may not be negligible, particularly for cities with strong ozone pollution compared to the surrounding areas. Ozone formation often takes place in the upper levels within the planetary boundary layer (PBL) (Goncalves et al., 2009) and the PBL height should be an important factor for sensing the low altitude pollution, with higher PBL heights in summer giving access to the low altitude information. The layer efficiency increases sharply to about 0.8 by 6 km and local convection may also loft the low altitude pollution higher up making the urban pollution amenable to detection by a TOMS/OMI like instrument.

Figure 4 shows the global distribution of the layer efficiency at 2.8 km for the month of June 2005 from OMI retrievals for that month. We have used only the near nadir pixels (pixel numbers from 20 to 40 from each swath) and reflectivity less than 0.3 (nearly cloud free) for this plot. As can be seen, the layer efficiency at 2.8 km can be near 0.6–0.7 over parts of China, Eastern USA and India with low values over mostly mountain areas. It should be mentioned that TCO data from OMI/MLS have been filtered for clouds and only pixels with reflectivity less than 0.3 have been used for their derivation (Ziemke et al., 2006), although this was not done for the TOR database. We have further examined the total ozone columns from OMI for the examples shown in Fig. 1 and

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found that clear signatures of the city plumes could be seen in the total ozone columns for Beijing, Sao Paulo and Mexico City and they were not affected by cloud filtering. The a priori ozone fields employed in the OMI retrievals were also quite zonal around cities and are unlikely to affect the results.

5 The efficiency factors in TOMS data are analogous to the total column averaging kernels. Liu et al. (2005) have retrieved ozone profiles as partial column ozone in the Umkehr layers from GOME radiance measurements. The averaging kernels for these profiles at 3 km indicate strong sensitivity near the surface for the tropics (their Fig. 3). Liu et al. (2009) have recently retrieved ozone profiles from OMI measurements which
10 indicate sensitivity down to 800–900 hPa range. Other works also indicate that residual tropospheric columns using OMI measurements may provide information about ozone in the 900–650 hPa levels (Chatfield et al., 2008a,b). Thus it appears that UV instruments like TOMS and OMI may indeed have useful sensitivity to the lower tropospheric information contrary to the currently held view which may explain the observation of
15 the urban scale signature in the TOR/TCO distributions as shown above. However a caveat should be pointed out in this context. While we have detected these urban plumes in either TOR or TCO data for many other cities not shown here, they show up sporadically and not for all times at any particular city. A number of factors could be responsible for this, including stratospheric variability masking tropospheric signatures as well as stratosphere-to-troposphere exchange (STE) processes at mid to high latitudes, merging of plumes from nearby cities in generally polluted regions, sensitivity variations with season, local topography and meteorology etc. Further, a city downwind from a major biomass burning region may have large free tropospheric ozone which might again swamp the city signature, as was noted also for CO signature of
20 cities (Clerbaux et al., 2008). On the other hand, isolated cities away from polluted continents may have better chances of detection, as was indeed observed with often clear signals at Jakarta, Colombo, Taipei, etc. (not shown).
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3.3 Urban signatures in ozone VMR data

As noted above for Mexico City, the elevated topography in several cases may inhibit observations of the urban plumes in TOR/TCO in the close vicinity of the cities themselves. The pressure variation both at the surface and the tropopause may affect the TOR/TCO calculations. Ziemke et al. (2001, 2006) have corrected for this by defining a pressure-averaged quantity to represent the tropospheric ozone. They use the following formula to obtain a mean tropospheric volume mixing ratio (VMR, in ppmv):

$$\text{Mean VMR} = 1.27 \left(\text{TCO} / (P_{\text{surface}} - P_{\text{tropopause}}) \right) \quad (2)$$

where TCO is in DU and P is in hPa. We have used this mean VMR data product, which is also available along with the TCO data from the OMI/MLS method. The use of mean VMR rather than the column data is likely to impact primarily signals from those cities which are in the vicinity of mountains. Figure 5 shows a few examples of these urban signatures. These are all climatological depictions averaged over the years 2004–2008 of available OMI/MLS measurements. In this case, the maximum in the VMR plume is seen essentially centered on the location of Mexico City (September climatology, top left panel). A distinct signature of Mexico City was observed in the VMR data for many months with available data as was also the case for TOR or TCO data for this city. The upper right panel shows two distinct but close plumes near the cities of Fresno and Visalia in the San Joaquin valley with rather high mean VMR values near 80 ppbv (June climatology). Similar plumes were observed in the climatological mean VMR data in July and August as well and are thus likely to be robust features and are consistent with reports of high ozone pollution in the San Joaquin valley in California. In fact Fresno and Visalia figure just below Los Angeles and Bakersfield in the list of US cities with highest ozone pollution at the surface level (<http://www.stateoftheair.org/2009/city-rankings/>). The maximum of the Fresno plume appears to be centered over the Yosemite National Park while the maximum of the Visalia plume occurs over the Sequoia National Park. On clear summer days upslope flows can transport the valley pollution over the Sierra Nevada as has been observed over Alpine valleys as well as

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over the Zagros mountains in the Middle East (Henne et al., 2004; Kar et al., 2006). This unusually large and persistent tropospheric ozone pollution over the Yosemite and Sequoia National Parks has implications for the ecosystem there. The Johannesburg and Santiago plumes were also observed for several other months (lower right panel).

5 The Johannesburg and Mexico City plumes in particular have also been observed in CO (Clerbaux et al., 2008).

Figure 6 shows the time series of the mean tropospheric VMR at Mexico City, Fresno, Santiago and Johannesburg for the period of OMI data (2004–2009). Also plotted is a corresponding time series at a location away from the city. For Mexico City and
10 Santiago, an ozone anomaly over the city can be seen in most months of the year with somewhat higher values in local summer as might be expected from enhanced photochemistry. Over Fresno the enhancement is seen primarily in summer and the anomaly over Johannesburg is lower than in the other cities. The latter may partly be attributed to the fact that in summer, the mid tropospheric ozone peak over Johannes-
15 burg is due to long range transport of air from Central Africa (Diab et al., 2003). As for TOR and TCO, signatures of several other cities around the globe could be detected, albeit sporadically, in the VMR database.

4 Conclusions

In this paper, we addressed the question of detecting possible footprints of cities in the tropospheric ozone columns for the first time as it should have important implica-
20 tions for future geostationary missions. It was found that city signatures could indeed be detected in the TOR, TCO as well as mean tropospheric VMR data bases as enhanced ozone plumes with general similarity to corresponding NO₂ tropospheric column plumes. An interesting result was the high mean ozone VMR plumes observed
25 over the Yosemite and Sequoia National Parks in summer resulting from the city plumes of Fresno and Visalia. An analysis of the OMI retrieval layer efficiencies indicated a fair degree of sensitivity to the lower troposphere which along with lifting of the boundary

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layer pollution by convection or topographic venting could possibly explain these features. However, a caveat is that these plumes were observed mostly sporadically and not at all times. These results should have interesting implications for satellite remote sensing of urban pollution. Further investigation is needed to understand the various factors influencing the detection of these plumes and to characterize them using data on other concurrent trace species.

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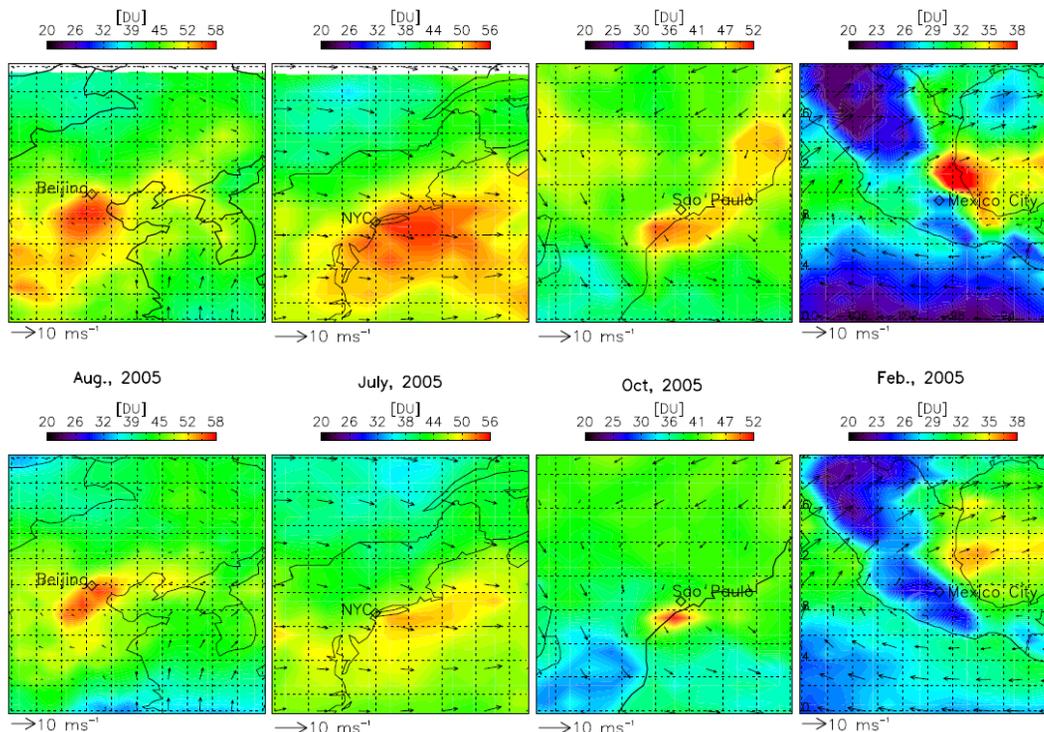


Fig. 1. Examples of urban scale signatures of tropospheric ozone pollution in monthly mean TOR (upper panels) and TCO data (the lower panels) for Beijing (August 2005), New York (July 2005), Sao Paulo (October 2005) and Mexico City (February 2005). Note that the upper limits of the color scales have been changed for each city to optimize the detection of the individual plumes because of different pollution levels in various cities. The monthly mean winds (at 850 hPa for Beijing, New York, Sao Paulo and at 700 hPa for Mexico City) are from the NCEP reanalyses.

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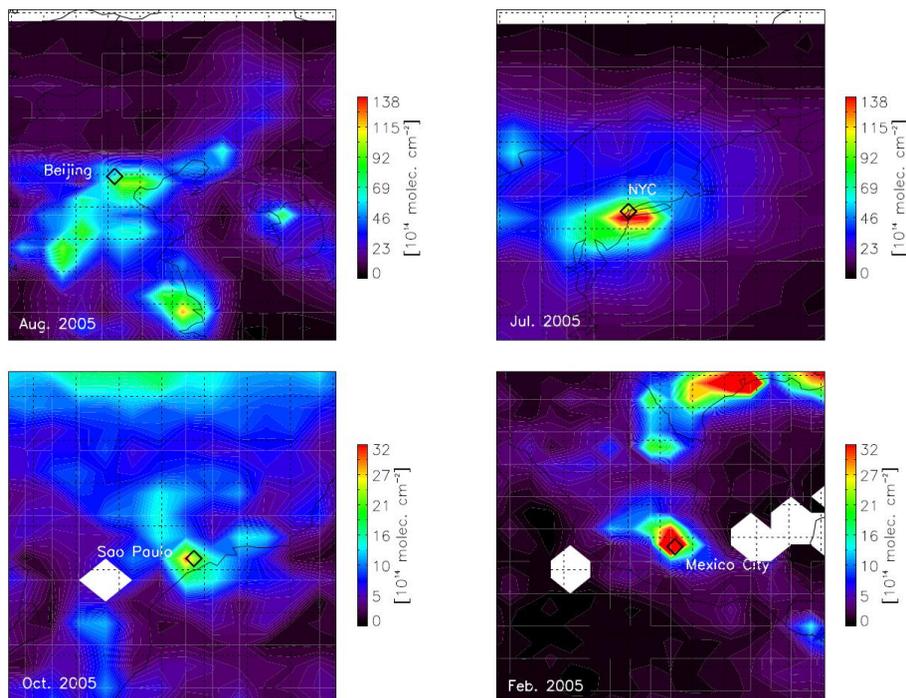


Fig. 2. The city plumes as observed in the SCIAMACHY tropospheric NO₂ columns for the same months as in Fig. 1. As in Fig. 1, the upper limit of the color scales has been varied to bring out the city plumes more clearly.

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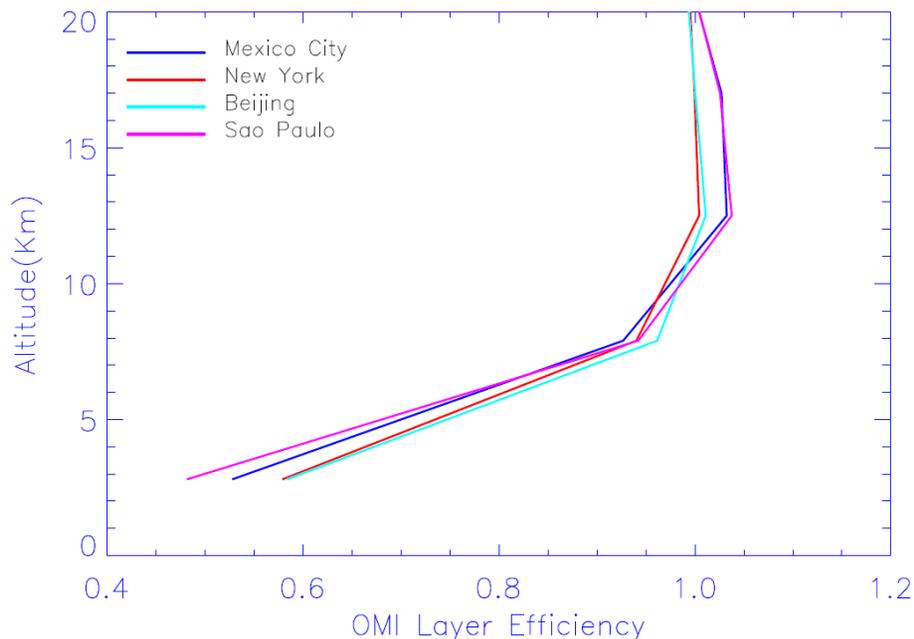


Fig. 3. Layer efficiency profiles near New York (July 2005), Beijing (August 2005), Sao Paulo (October 2005) and Mexico City outflow plume (February 2005) from the OMI level 2 data for reflectivity less than 0.3.

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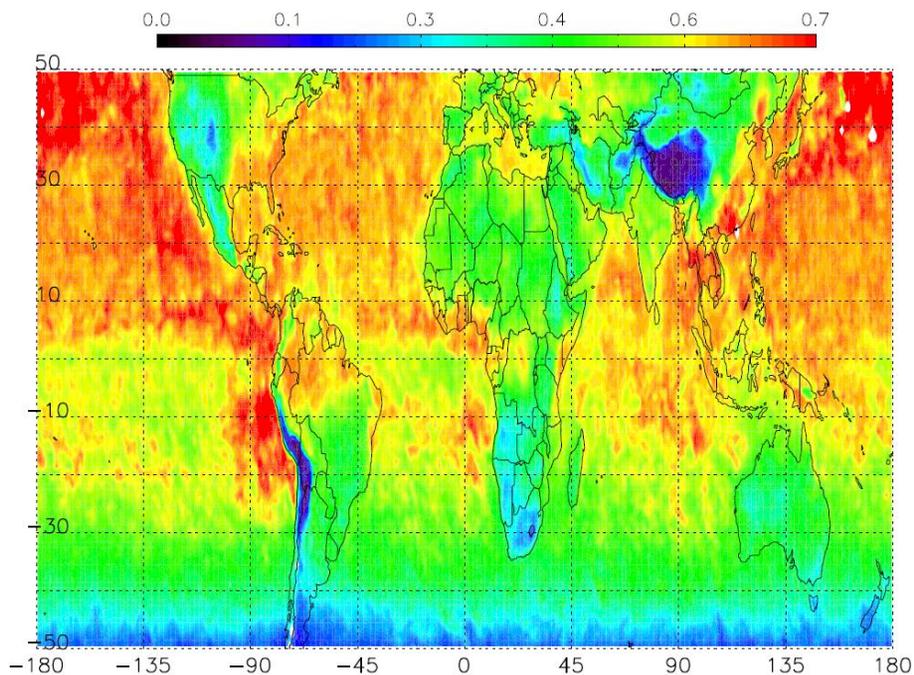


Fig. 4. Global distribution of the layer efficiency at 2.8 km for June 2005 from OMI level 2 data for reflectivity less than 0.3. Only the pixel numbers from 20 to 40 from each swath were used.

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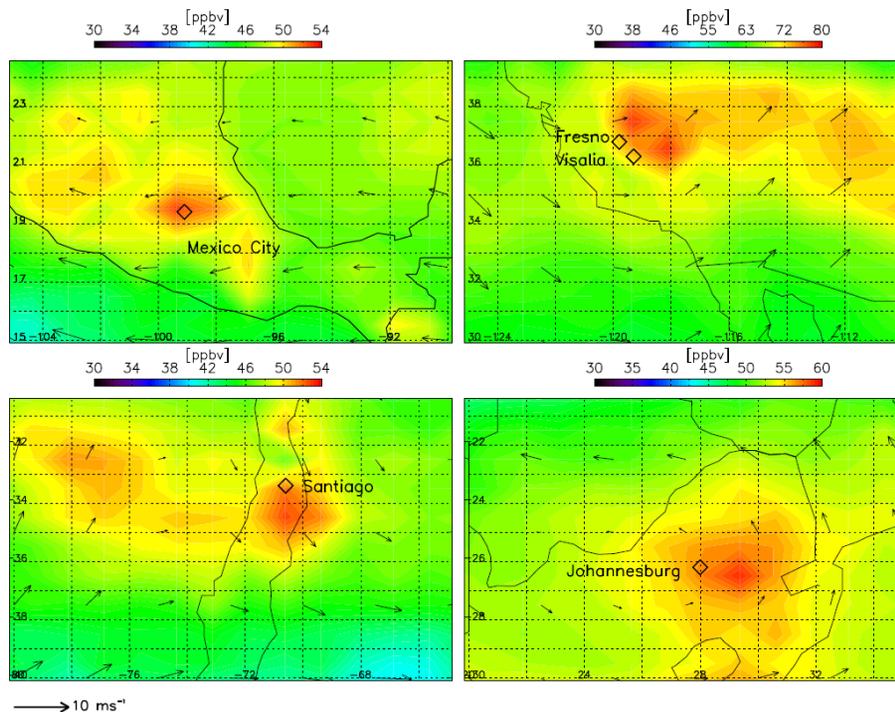


Fig. 5. Signatures of Mexico City (September), Fresno and Visalia (June), Santiago (February) and Johannesburg (January) in the climatological mean (2004–2008) tropospheric ozone VMR normalized for pressure variations at the surface and the tropopause. The NCEP reanalyses winds at 700 hPa are averaged for the corresponding months between 2004–2008.

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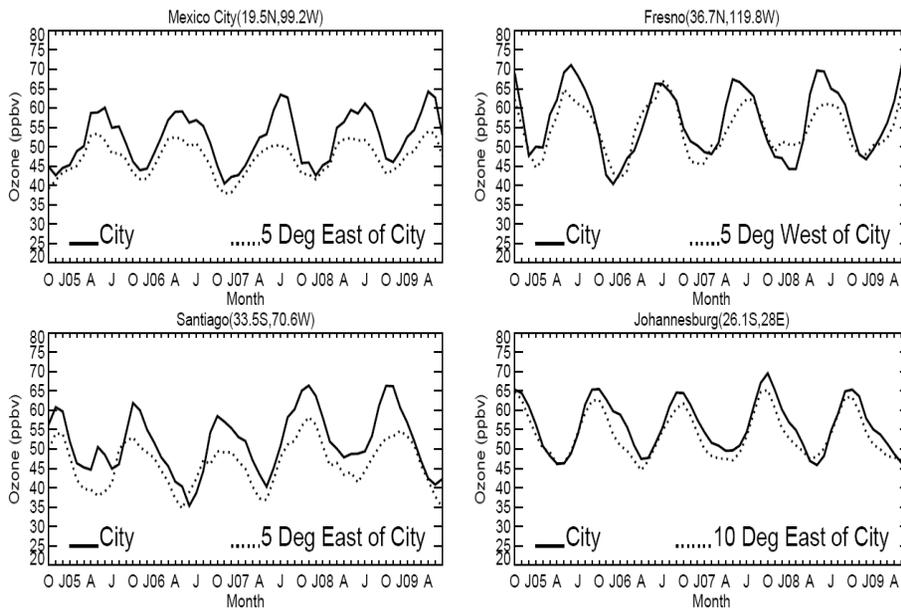


Fig. 6. Time series of monthly mean tropospheric ozone VMR at the 4 cities shown in Fig. 5 along with corresponding time series at locations away from the city (3 month running average).

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