

## Auxiliary Material

Cities investigated but deemed to have insufficient data for our particular analysis included: Beijing, Moscow, Cologne, Phoenix, Lagos, Riyadh, Paris, Dhaka, Karachi, Shanghai, Sao Paulo, New York City, Mexico City, Guangzhou, and Delhi.

All uncertainties reported and shown are one-sigma. For Fig. 1, and Figs. S1 & S4 error bars are generated by using a single sounding one-sigma error of 1.25 ppm, reduced by the square root of the number of observations in each averaging bin. The 1.25 ppm is derived in two manners. It is the higher end of the retrieval error for a single sounding (as small as 0.7 ppm). Also, on five different occasions, seven or greater observations were made within the LA basin. On these days, the standard deviation in  $X_{CO_2}$  ranges from 0.70 to 1.25 ppm. We have elected to use the more conservative 1.25 ppm value. For Fig. 2 we simply plot the reported retrieval error for each given sounding.

We investigated the potential role of systematic retrieval biases between the Los Angeles basin and the desert background soundings. Biases can occur due to different aerosol loadings as well as substantially different underlying surface albedos. We consider the overall enhancement of  $\sim 3.2$  ppm very robust. It does not exhibit a succinct seasonal cycle but is manifested as a rather constant offset throughout all seasons. Owing to the high temporal variability of aerosols in the LA basin, any aerosol related bias would, however, show a strong seasonal dependence, which is not observed. Furthermore, we find no significant correlation between aerosol optical depth and the basin-desert difference (Fig. S5). We also investigated systematic differences in retrieved surface pressure, albedo, and  $O_2$  A-band signal levels, all related to potential systematic

biases [Wunch et al., 2011]. Application of the suggested correction scheme for ACOS B2.9 data [Wunch et al., 2011] results in a statistically insignificant change in the basin-desert difference in Los Angeles and the city-rural difference in Mumbai (0.1 ppm in both cases). In addition, none of these three variables correlate with the basin-desert difference (Fig. S5). We thus conclude the differential enhancements reported are robust and significant.