Continental outflow of ozone pollution as determined by ozone-CO correlations from the TES satellite instrument

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Introduction

Space-based observations of tropospheric ozone and carbon monoxide (CO) from the Tropospheric Emission Spectrometer (TES) launched aboard the Aura satellite in July 2004 [Beer, 2006] offer new information on the continental outflow and intercontinental transport of ozone pollution through the correlation between ozone and CO concentrations. Measurements of ozone-CO correlations at continental outflow sites and from aircraft have been widely used to estimate the efficiency of ozone formation and export [Parrish et al., 1993, 1998; Honrath et al., 2004] and to test global model simulations of these processes [Chin et al., 1994; Li et al., 2002]. However, in situ observations are sparse. TES offers a global perspective. We examine here the tropospheric ozone-CO correlation patterns observed by TES in July 2005, with focus on outflow from the northern mid-latitudes continents, and examine the capability of a global 3-D tropospheric ozone model (GEOS-Chem) to reproduce these features as a test of the model simulation of global human influence on tropospheric ozone.

## TES Observations and GEOS-Chem Simulations

TES Observations

- TES aboard the EOS Aura satellite is a Fourier transform infrared emission spectrometer with high spectral resolution (0.1 cm^{-1}) and a wide spectral range (650-2250 cm^{-1}) [Beer et al., 2001].
- TES standard products (global surveys) consist of 16 orbits of nadir vertical profiles every other day. The degrees of freedom for signal (a measure of vertical sensitivity) for the vertical profiles is between 2-3 for ozone in the troposphere, and 1-2 for CO [*Worden et al.*, 2004].
- We use ozone and CO retrievals for 4-31 July, 2005, including 14 global surveys.

The TES retrieval process extracts concentrations from the observed radiances using the optimal estimation method [Rodgers, 2000].

$$\hat{x} = x_a + A(x - x_a) + G\mathring{a}$$

where the retrieved profile ($\hat{x}$), a priori (x_a), and true profile (x) are in logarithms of mixing ratios, ε is the radiance measurement noise, and G is the gain matrix converting this noise to retrieval error. The figure to the right shows a sample averaging kernel matrix (A) for TES nadir retrievals of ozone and CO.

23°N, 78°W from the global survey on 4-5 July 2005
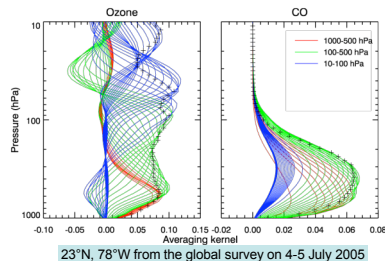
We focus our analysis on the 618 hPa level where TES has good sensitivity for both ozone and CO centered in the middle troposphere, with little influence from the stratosphere.

GEOS-Chem Simulations

- Driven by GMAO GEOS-4 assimilated meteorological fields
- 2° × 2.5° horizontal resolution, 30 eta levels in vertical
- Tropospheric ozone-NO_x-CO-VOC-aerosol chemistry
- Fire emissions are from the Duncan et al. [2003] climatological biomass burning inventory but allocated daily for the TES observation period on the basis of MODIS fire counts.
- Model profiles are sampled along the TES orbit track at the observation time, and then vertically smoothed with the TES averaging kernels.

Ozone and CO concentrations at 618 hPa for July 2005. Values are averaged on a 4° × 5° grid.
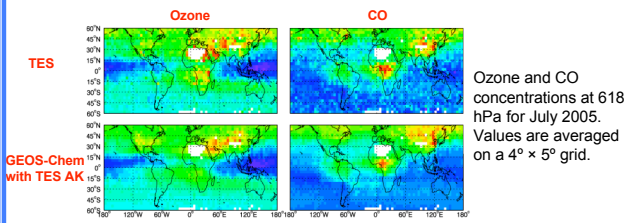
Ozone-CO correlations in Continental Outflow
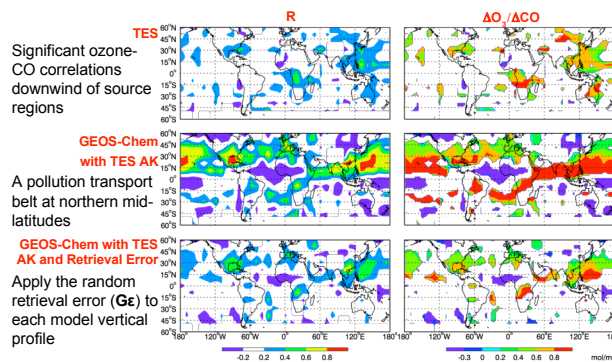
The figure below shows TES and simulated ozone-CO scatterplots for two 10° × 10° gridboxes over the southeastern U.S. and the Asian Pacific Rim, representative respectively of North American and Asian free tropospheric outflow.
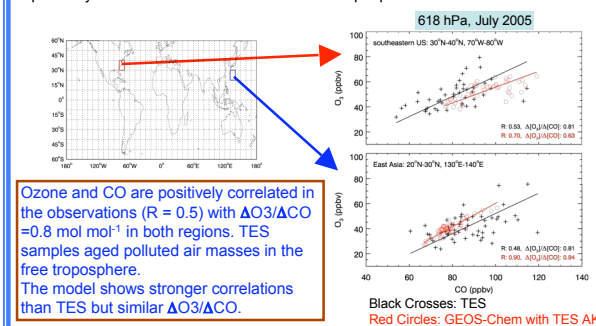
Ozone and CO are positively correlated in the observations (R = 0.5) with $\Delta O3/\Delta CO$ =0.8 mol mol^{-1} in both regions. TES samples aged polluted air masses in the free troposphere.
The model shows stronger correlations than TES but similar $\Delta O3/\Delta CO$.

Black Crosses: TES
Red Circles: GEOS-Chem with TES AK

Global Distribution of Ozone-CO Correlations

The figure below shows the global distribution of ozone-CO correlation coefficients R and enhancement ratios $\Delta O_3/\Delta CO$ in the TES data at 618 hPa for July 2005. The data are binned into 10° × 10° grid cells, and results are interpolated in the plot. White regions correspond to |R| < 0.2.

Significant ozone-CO correlations downwind of source regions

A pollution transport belt at northern mid-latitudes

Apply the random retrieval error ($G\varepsilon$) to each model vertical profile

Simulated ozone-CO correlations are similar to those observed by TES after degradation of the model fields to account for random retrieval error $G\varepsilon$. Simulated correlations without this retrieval error are much stronger and stretch across northern mid-latitudes, thus identifying retrieval error as a limiting factor for observing ozone-CO correlations from space. Recent warm-up of the TES optical bench should decrease retrieval error for CO.

Deriving Ozone-CO Correlations from TES data

- We determined ozone-CO correlations in the TES data by binning the observations for July 2005 into 10° × 10° grid cells. Each cell has 20-60 data points from which to derive the correlation.
- To remove the effect of the geographically variable a priori constraints used in the TES retrieval, the TES and model profiles are reprocessed using a universal a priori:

$$\hat{x}' = \hat{x} + (I - A)(x'_a - x_a)$$

where x_a is the original TES a priori constraint and x'_a is the universal a priori from averaging x_a in the 60°N - 60°S band.
- Ozone-CO linear regressions for these reprocessed fields are determined using the reduced major axis method; ozone-CO enhancement ratios ($\Delta O_3/\Delta CO$) are determined as the slope of the regression line.

Conclusions and Future work

- The TES observations for July 2005 show significant ozone-CO correlations downwind of continents with large industrial and biomass burning sources.
- The GEOS-Chem model reproduces the ozone-CO correlation patterns and enhancement ratios observed by TES, providing an important test of the model simulation of continental outflow and intercontinental transport of ozone pollution.
- Analysis of the model correlation fields shows that reducing retrieval error would increase the strength of ozone-CO correlations seen from space.
- We will extend the study to other seasons.
- Correlation in the ozone and CO retrieval errors (as for example due to clouds or temperature) still needs to be characterized.
- Use TES error analysis to estimate expected ozone-CO correlations.