1. Introduction

Estimating the global abundance and regional distribution of hydroxyl radical (OH) is challenging. Past empirical approaches have used methyl chloroform, methane, CH4 and other alternatives as atmospheric tracers, showing upon their well-quantified reaction rates with OH. Here we propose a new method that draws upon multi-year time series of fire emissions and column carbon monoxide (CO) observations.

Several recent findings provide the foundation for the development of our method:

Finding 1: Global fires are sporadic in nature, with high emissions events distributed widely in northern tropical and southern regions as a result of year-to-year variability in both climate and land use processes (Figure 1).

Finding 2: The fire-induced `impulses' in CO explain most of the interannual CO anomalies in both surface and column observations. For example, using the GEOS-Chem model and the Global Fire Emissions Database (GFEDv2), we found that fires explained 56% of CO variability at surface stations between 30°N and 30°S, 51% between 30°N and 30°S, 58% between 30°N and 90°N, and 59% in the period of 1997-2007 (Figure 2).

Finding 3: The sensitivity of CO variability to changes in OH is low over source regions and increases toward remote ocean regions. CO is relatively insensitive to OH over source regions because diffusive and advective atmospheric transport occurs more rapidly than reaction with OH. In remote ocean areas, the path length from source regions is large, and so air parcels have been exposed to the cumulative effects of OH oxidation for relatively long time intervals (Figure 3).

2. Model and Methodology

We used the GEOS-Chem global chemical transport model version v8-01-02. This model has a horizontal resolution of 4° in latitude, 5° in longitude and 47 layers in vertical. The driving wind is from assimilated meteorological dataset for the Goddard Earth Observing System (GEOS) version 5 at NASA Data Assimilation Office. Our approach for optimizing OH has two steps: adjust CO flux and optimize global OH.

Step 1: Adjust CO Flux

A Bayesian inversion model was used first to optimize GFEDv2 fire emissions using MOPITT observations over source regions where the sensitivity to emissions is high, but the sensitivity to OH is low, shown in Figure 3. The inversion model here uses anomaly data (both annual mean cycle and trend are removed).

Step 2: Optimize Global OH

Using the optimized CO fluxes, a series of offline GEOS-Chem simulations are conducted with prescribed OH levels that varied between 0.4 and 2.0 times the predicted OH distribution from a full chemistry run. We then calculated Taylor scores and RMSEs for each OH level. The OH level corresponding to the highest Taylor score (or lowest RMSE, shown in Figure 4) was identified as the optimal OH level.

Figure 5 shows the global distributions of correlation coefficients and ratios of standard deviations with MOPITT3 CO observations and model simulations. The maximum Taylor score or the minimum RMSE is corresponding to the optimized OH level. Here, we obtained the best OH level (area-weighted in the region 60°S-60°N) is 0.6 that means OH for current GEOS-Chem model is over-estimated by 40%.

Figure 6 shows the Taylor scores and RMSEs are calculated with different OH levels based on MOPITT3 CO observations and model simulations. The maximum Taylor score or the minimum RMSE is corresponding to the optimized OH level. Here, we obtained the best OH level (area-weighted in the region 60°S-60°N) is 0.6 that means OH for current GEOS-Chem model is over-estimated by 40%.

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3. Conclusions

- Errors in forward CO simulations are large over tropical South America, Africa and equatorial Asia. These errors can be effectively reduced through adjusting fluxes with a CO inversion model.
- Changing OH can improve correlation between model simulations and observations, but it has a larger positive effect in the ratio of standard deviations and root mean square differences.
- Both correlations with MOPITT and magnitudes of anomalies of CO in the remote areas are improved by decrease OH levels.
- The decay of CO anomalies from source to remote ocean regions contains variabilities in both OH effects and rates of atmospheric mixing.

Acknowledgement

This study is supported by NASA grant NNX08AF84G. The GEOS-Chem model is managed by the Atmospheric Chemistry Modeling group at Harvard University (http://acmg.seas.harvard.edu/geos/). For any question, please contact Mingquan Mu (mumin@uci.edu).