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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Free troposphere ozone and carbon monoxide over the North Atlantic for 2001–2011" *by* A. Kumar et al.

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We thank you for your valuable feedback on the manuscript titled "Free Troposphere Ozone and Carbon Monoxide over the North Atlantic for 2001-2011". Our responses to your comments and suggestions are provided below.

Comment 1: "The most important problem is the selection of key scientific issue. The authors tried to find an evidence to show whether the decreased emission in North America can influence the ozone and CO trend over the Atlantic Ocean. In fact, as a main downwind area of the high emission regions over North America, it is easy to understand the linkage, which has already been pointed out in some previous works (e.g. Oltmans et al., 2006). However, in this work the authors tried to prove this conclusion in

a quite complicated way. The worse problem is that because of data limitation and the shortcomings in their modeling and analysis, they didn't logically provide evidences to support the conclusions. This referee suggests that the authors should do more data analysis and modeling works, with a focus on the inter-annual variation (year-to-year difference) in ozone and CO besides the trend issue."

The fact that North American emissions can affect O3 and CO over the Atlantic ocean is indeed straightforward but the major point of this study is to quantify how much each of the major global source regions (not only North America but also other regions like Asia and Europe) have contributed to the background ozone and CO over the North Atlantic as well as the possible trends. During the study period (2001-2011), the anthropogenic emissions have decreased in some regions (e.g. North America & Europe) but increased in some other regions such as Asia. So the overall effects on the North Atlantic free troposphere as well as the specific contributions from various regions are not straightforward at all. We think the 11-yr time period is long enough that our statistical analysis should pick up the possible trend signal instead of the inter-annual variations. There are some known issues/biases of the model, but our conclusions were largely based on the statistical analysis of the measurement data and our modeling results were mainly used to help interpret the observations. Nevertheless, we have carried out some further analysis, as detailed in the response to comment 3 and Figure R1 below (Figure 2 in the revised manuscript).

Comment 2: "The data coverage looks quite bad, especially for CO. I don't know the limited data coverage was because of instrumentation problems or of the strict criteria to make the daily average (they used "only days with full 24 data availability"). Why the authors used such a strict criteria? In fact, from the perspective of observation it is quite normal that there are few hours' data missing because of zero or span calibration or instrument problems. In addition, the model output from GEOS-Chem have a resolution of 4-hour but the observation have a coverage of 24 hours."

Yes, the observations available from the Pico Mountain Observatory (PMO) do not

ACPD
13, C7757–C7764, 2013

span the entire study period (2001-2011) which is due to weather conditions at the station and other unavoidable factors. Operation of the remote station at the mountain top is very challenging, especially during wintertime. However, the data coverage is very good for the summer time. Also, there were periods (e.g. September 2004- August 2005) having near complete data which aided in studying the seasonal behavior of the species at the station. Our statistical analyses were based on daily (24h) averages of O3 and CO. Considering the diurnal variations for O3 and CO, we applied this 24 hour filter (i.e. only days with full 24 hour data availability were used) to ensure that the processed data are truly representative of daily (24h) averages. Otherwise, incomplete data points during a day (e.g. only daytime or night time data) may lead to unexpected bias in our results. This 24 hour filter did reduce the number of data points but we still used approximately 76% of all the measurement data for CO and 87% data for O3. We have added clarification in the text (Section 2.1, Lines 98-102)– "Considering the diurnal variations of CO and O3, we have applied a 24 hour filter to avoid unexpected bias – i.e. only days with full 24 hour data availability were used to calculate the daily average mixing ratios of CO and O3. The application of this filter resulted in the inclusion of approximately 76% (87%) of the total CO (O3) measurements in the final analysis." The GEOS-Chem output with 4-hour resolution were averaged to get the daily (24h) averages which is appropriate for comparison with the daily averages from observations. We have added clarification in the text (Sections 3.1, 3.2 & 3.3). Section 3.1, Lines 180-181: "These 4 hour instantaneous mixing ratios were used to compute the daily averages for both the species." Section 3.2, Lines 208-209: "These values were used to obtain the daily average for each CO tracer." Section 3.3, Lines 217-218: "The daily averages for all tracers were computed using these 4-hourly instantaneous values."

Comment 3: "To support the authors' conclusion, GEOS-Chem modeling still needs additional work. First, evaluation using one year data (September 2004-August 2005) is not enough. The comparison should be made for the entire period to see if the model can produce the year-to-year difference. Besides the Tagged simulation, a sim-

ACPD
13, C7757–C7764, 2013

ulation with fixed emission inventory during the 11 years should also to be conducted to see how much of the trend was caused by change in decadal variation in circulation patterns and how much of the trend was influenced by accumulated error during the numerical integration."

We have carried out further data analysis and model evaluation, as partly shown in the newly added section 4 in the revised MS. We have added in a new figure (Figure 2) showing the model simulated daily average O3 and CO for the 11-year period compared with measurement. For the original Figure 2 in the MS, we selected the period of September 2004-August 2005 for model evaluation because that full-year period has the best data coverage for both surface measurement and satellite observations.

Response to Minor Comments Comment 1: "For the FCNE simulation and Tagged simulation, the emission inventory should have the same data source."

The FCNE simulation was carried out using an older version of GEOS-Chem (v8.03.01) for which the biomass burning inventory GFEDv2 was used. We later updated to the newer version of GEOS-Chem (v9-01-02) which has updated biomass burning inventory of GFEDv3 and was used for the tagged simulation.

Comment 2: "The authors used two sections (sect. 2.2 and 2.3) to introduce satellite data. However, the data was only used in Fig.2. Can the data give more support for the discussions? For example, can the satellite data fill the gap in surface observation for some years?"

Good point. We actually ever thought about this, but the problem is that there are significant discrepancies between the satellite data and surface measurement data (as partly reflected in Figure 2 of the earlier manuscript) making it hard to "merge" these different observational datasets. On the other hand, the satellite data covers a shorter time period than the surface measurement so we couldn't use them alone for the trend analysis. Now we have combined the two sections (2.2 and 2.3) into one section in the text (2.2. Satellite data).

ACPD
13, C7757–C7764, 2013

Comment 3: “For the significant positive ozone trend in the upper troposphere, the authors attributed it to the change of lightning. Why not carried out sensitivity tests (with/without lightning) using GEOS-Chem?”

Point well taken. An additional 11-yr full chemistry simulation would take a long time so we have carried out analysis on the lightning flashes (and therefore lightning NOx emissions) for this time period and added discussion in the text (Section 5.2, Lines 347-349) “Harmonic regression analysis of monthly means of global lightning flashes archived from GEOS-Chem for 2001-2011 shows statistically significant increase over this period which points to an increase in the NOx produced.”

Comment 4: “Last paragraph of Section 4, the authors attributed to the overestimation of CO to low biases in CO emission. However, many reasons can cause this problem, e.g. parameterization of the boundary layer and vertical convection as well as chemical processes. It is worth to know the underestimation many existed in later-spring and summer. Emission shouldn’t have such large seasonal variation.”

Point well taken. Some CO sources (e.g. biomass burning emissions) do have large seasonal variations which may lead to higher biases in some seasons than others, but that’s beyond the scope of this study. So we have clarified this part in the text as (Section 4, Lines 226-229)- “The significant underestimate of CO by GEOS-Chem possibly reflects some low biases in the CO emission inventories used in the model. The model underestimates of CO have also been reported by previous studies [Bey et al., 2001; Duncan and Logan, 2008; Duncan et al., 2007; Val Martin et al., 2008].”

Comment 5: “The regression model fit to ozone: It looks that the amplitude of the fitting (i.e. a2) is too small if compared with observations.”

A possible reason for the sinusoids having a small amplitude than the fit for CO could be the greater variability in the O3 mixing ratios with respect to the mean as compared to CO. The coefficient of variation (CV) for O3 observations is 23.1 % while that for CO is 19.7% which points towards a higher noise to signal ratio for O3. If there is high

ACPD
13, C7757–C7764, 2013

variability in the data the regression model might not be able to capture all of that and consequently the R2 and the amplitude of the sinusoids could be smaller.

Comment 6: “For the different ozone trend in Asian, is there any reference support this point?”

To our knowledge, impacts on long-range transport of Asian pollution to the North Atlantic from climate change in the past decades have not been studied in the literature. The importance of possible impacts on tropospheric background ozone from climate change, as discussed in the MS, may warrant further studies on this topic.

Comment 7: “The conclusion seems too long. Please don’t repeat the results in this part but give key findings.”

We have made the conclusion section more concise now.

References

Bey, I., Jacob, D. J., Yantosca, R. M., Logan, J. A., Field, B. D., Fiore, A. M., Li, Q., Liu, H. Y., Mickley, L. J., and Schultz, M. G. (2001), Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation, Journal of geophysical research, 106(D19), 23 073-023 096. Duncan, B. N., and Logan, J. A. (2008), Model analysis of the factors regulating the trends and variability of carbon monoxide between 1988 and 1997, Atmos Chem Phys, 8(24), 7389-7403, doi:7310.5194/acp-7388-7389-2008. Duncan, B. N., Logan, J. A., Bey, I., Megretskaia, I. A., Yantosca, R. M., Novelli, P. C., Jones, N. B., and Rinsland, C. P. (2007), Global budget of CO, 1988–1997: Source estimates and validation with a global model, Journal of geophysical research, 112(D22), D22301, DOI: 22310.21029/22007JD008459. Val Martin, M., Honrath, R. E., Owen, R. C., and Li, Q. B. (2008), Seasonal variation of nitrogen oxides in the central North Atlantic lower free troposphere, Journal of Geophysical Research: Atmospheres (1984–2012), 113(D17), doi:10.1029/2007JD009688.

ACPD

13, C7757–C7764, 2013

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/13/C7757/2013/acpd-13-C7757-2013-supplement.zip

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Interactive comment on Atmos. Chem. Phys. Discuss., 13, 15377, 2013.

ACPD
13, C7757–C7764, 2013

Interactive Comment

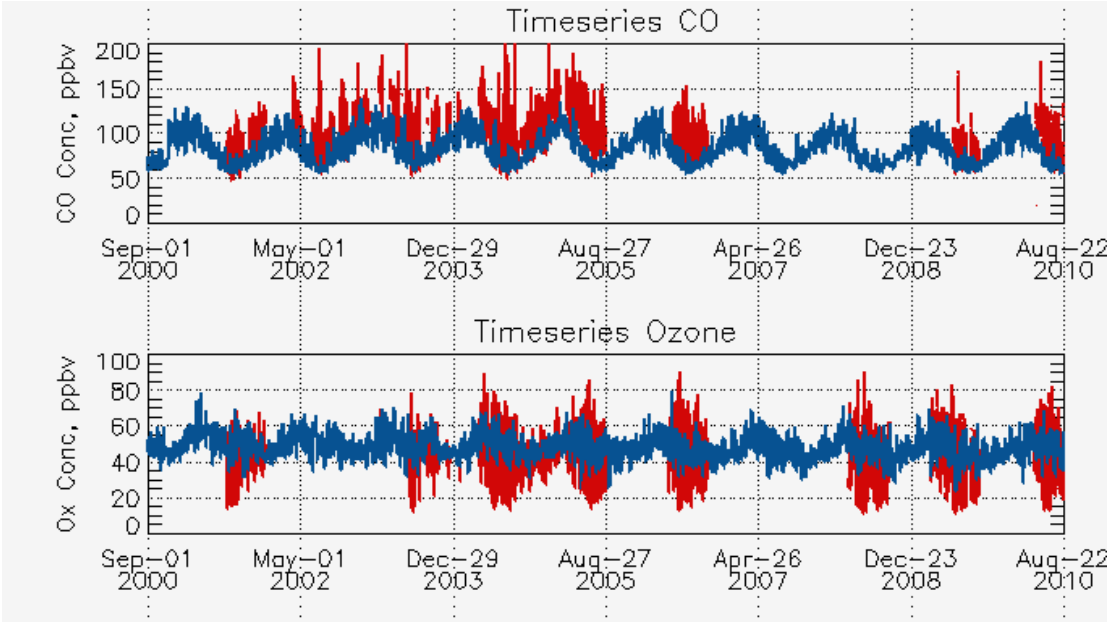

Fig. 1. Figure R1: Time series of GEOS-Chem output (blue) and Pico observations (red) for CO(top) and Ozone (bottom) respectively.

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