

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

Anonymous Referee #2

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Tropospheric ozone is an important trace gas from the perspectives of atmospheric chemistry, air quality and climate change. The concentration of tropospheric ozone has been found to be influenced by human activities and natural processes. So it is important to understand the trend of tropospheric ozone and the main cause in different region. In this work, the authors tried to investigate ozone and carbon monoxide trend over the North Atlantic based on data collected a mountain site during 2001-2011 and to explore the possible reasons based on chemical transport modeling using GEOS-Chem. The work itself is important for understanding the anthropogenic impact to the remote atmosphere. However, this referee thinks that it cannot be accepted for publication on Atmos. Chem. Phys. in its current forms before a substantial revision carried out according to following comments.

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Major comments:

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 interannual variation (year-to-year difference) in ozone and CO besides the trend issue.

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.

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 simulation 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.

Minor comments:

1.For the FCNE simulation and Tagged simulation, the emission inventory should have the same data source.

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?

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?

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.

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.

6.For the different ozone trend in Asian, is there any reference support this point?

7. The conclusion seems too long. Please don't repeat the results in this part but give key findings.

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