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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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Referee response 1

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: "What the observations are very important at this site, I am not convinced that they support a clear indication of a trend. The observations appear to be limited over this 10 year period, with some years having no data and most years having no seasonal information. So overall, it appears to me that the authors have largely based the trend analysis on the model results and set out to support or "prove" this trend

using the observations. The limited comparison of the data with the model (Figure 2), only compares monthly means, which provides very little insight into the models capabilities. I would suggest that the authors do a much better job at explaining what the observations really show, then to see if the model can reproduce the observations before talking about trends."

First of all, we would like to point out that our conclusions on possible trends are mainly based on the 10-yr observational data at the Pico site instead of the modeling results. Actually we first noticed the seemingly downward trends when analyzing the measurement data which motivated us to carry out further modeling work to confirm and more importantly to interpret those observations. Indeed, the Pico data are only available for this 10 year period and not all years have the full-year data. But this represents the best available dataset for this remote and unique station located in North Atlantic. The GEOS-Chem model has been extensively evaluated against observations in the literature and we have provided references in the manuscript. We have also included more model evaluation and discussion in Section 4. Finally, we have weakened our conclusions to "possible trends" or "likely trends".

Comment 2: "Don't understand how you can ïňĄt with sinusoidal given limited data on annual cycle. While this ïňĄt is statistically signiïňĄcant (given the large number of data points), from Figure 2, it does not appear to have a very high R2 and thus must be viewed with caution."

The seasonal cycles of O3 and CO are well known and documented, so we used the harmonic terms (with period of one-year) to account for these seasonal variations. Figure 3 in the updated manuscript shows the plot for the observations along with the model results and satellite data for the period of September 2004-August 2005. This period has the full-year in-situ data available. The figure clearly shows the seasonal variations for O3 and CO at the station and the time period of the cycles is one year. A similar seasonal cycle for CO was reported by [Val Martin et al., 2008] [Figure 3(d) (Pg D17307)]. As mentioned above, we have also weakened our conclusions to "possible

trends" or "likely trends".

Comment 3: "Only the seasonal cycle in the model is compared with the observations. This tells us very little. What other model evaluation for Pico have you done? The source analysis (table 4) depends critically on the model capturing day to day variations."

In addition to comparing the seasonal cycle between the model and observations we have also done model evaluation based on daily average O3 and CO mixing ratios, as shown in Figure 1 below (which was not included in the MS due to space limit). Some of these analyses have been summarized in section 4 of the manuscript: 1.) In general, GEOS-Chem was found to underestimate the observations with the magnitude during summer being 24.7 +/- 5.2 ppbv (1-ïĄş). This underestimation has also been reported by previous studies. 2.) For O3, the GEOS-Chem performance was much better. The model was 6.7 +/- 2.6 ppbv higher and showed the least agreement in winter and best agreement in summer and fall. 3.) Provided below is a timeseries plot of CO and O3 daily average mixing ratios from observations and GEOS-Chem which also points towards the facts mentioned in section 4 of the manuscript.

Comment 4: Model and satellite data are not independent, given apriori. What model is used in apriori and can you demonstrate that the satellite data show some reasonable agreement with the observations beyond the seasonal cycle.

The main purpose of our study is to examine the evolution of background O3 and CO over North Atlantic in the past decade. The seasonal or monthly average values are suitable for examining the long-term (e.g. decadal) trends. Nevertheless, we have updated the MS and included model evaluation based on daily O3 and CO values (Section 4). The satellite data have very limited coverage for the whole study period and hence were only used as supplementary reference in addition to the in-situ measurement. Detailed evaluation of the satellite data has been in literature (see references in the MS) and is beyond the scope of this study. The bottom line is that, even if we skip/remove

all the satellite data (i.e. only use the in-situ measurement for model evaluation), our major conclusion (regarding the possible trends of background O3 and CO) would not be affected at all.

RESPONSE TO DETAILED COMMENTS:

1) P15380, Line 9: "Decreasing trend is the second derivative. I don't think this is what you mean."

We determine the increasing vs. decreasing trends based on the a1 values from equation 1 in the MS – negative a1 values indicate decreasing trend (if it's statistically significant).

2) P15381, Line 10: "This is very puzzling. Every other mtntop station reports a diurnal cycle in upslope/downslope (e.g. MLO, MBO, Jungfraujoch). Comments?"

We meant to say that the measurement at the mountaintop station is generally good to represent the background concentrations. There are indeed diurnal cycles in upslope/downslope flows, as pointed out in Kleissl et al. (2007): "Diurnal cycles of wind direction on the mountain slope consistent with daytime upslope and nighttime downslope flow were found on 24% of the days during late June, July, and August 2004". We have clarified this sentence as (Section 2.1, Lines 79-81) "This station is well above the Marine Boundary Layer (MBL) during summertime and ideal for examining the atmospheric composition of the lower free troposphere (FT)."

3) P15381, Line 23: "There are biases in the day vs night AIRS data. See some of the recent AIRS analyses on this."

There is no discussion related to AIRS data in P15381.

4) P15389, Line 19: "Can you clarify the meaning of 1/3 in this context."

We have clarified this part as (Section 5.2, Lines 305-306) "Fig. 1 shows that the U.S. CO emissions have declined by about 1/3, from around 105 Tg in 2000 to 70 Tg in

2011."

5) P15391, Line 20+: "I find the model-observation comparison very limited (seasonal cycle) and not helpful in this regard. The satellite comparison is suspect because of the apriori influence. It would be more convincing to do daily comparisons of the model, obs and satellite. Without this, the results are not convincing."

As discussed above, the main purpose of our study is to examine the evolution of background O3 and CO over North Atlantic in the past decade. The seasonal or monthly average values are suitable for examining the long-term (e.g. decadal) trends. Nevertheless, we have updated the MS and included model evaluation based on daily O3 and CO values (Section 4). The satellite data have very limited coverage for the whole study period and hence were only used as supplementary reference in addition to the in-situ measurement. Detailed evaluation of the satellite data has been in literature (see references in the MS) and is beyond the scope of this study. The bottom line is that, even if we skip/remove all the satellite data (i.e. only use the in-situ measurement for model evaluation), our major conclusion (regarding the possible trends of background O3 and CO) would not be affected at all.

REFERENCES:

Kleissl, J., Honrath, R. E., Dziobak, M. P., Tanner, D., Martin, M. V., Owen, R. C., and Helmig, D., Occurrence of upslope flows at the Pico mountaintop observatory: A case study of orographic flows on a small, volcanic island, J. Geophys. Res., 112, 10, doi:10.1029/ 2006JD007565, 2007. Val Martin, M., R. Honrath, R. Owen, and Q. Li (2008), Seasonal variation of nitrogen oxides in the central North Atlantic lower free troposphere, Journal of Geophysical Research: Atmospheres (1984–2012), 113(D17).

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/13/C7765/2013/acpd-13-C7765-2013-supplement.zip

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

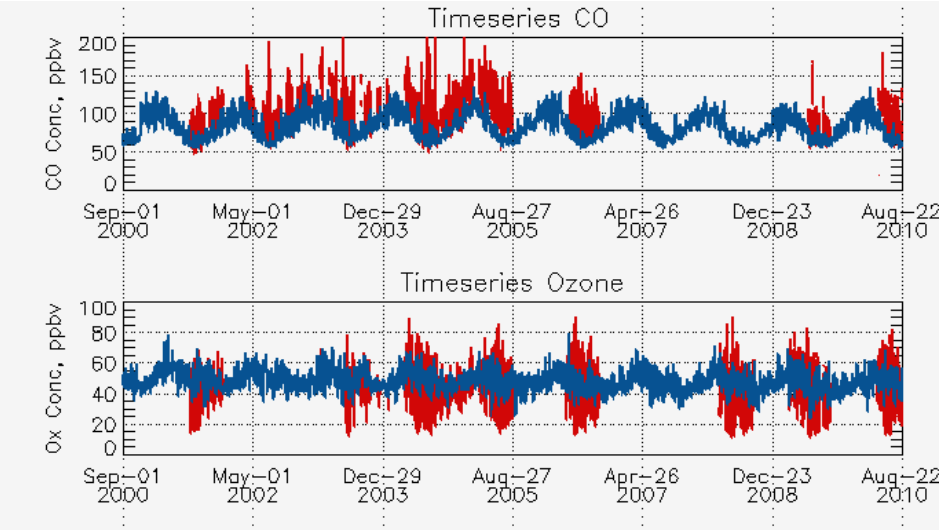

Fig. 1. Time-series showing CO (top) and O3 (bottom) mixing ratios from September 2000-August 2010. The observations are shown in red and GEOS-Chem output in blue.