Response to comments from referee #1 - A Lagrangian view of ozone production tendency in North American outflow in summers 2009 and 2010
MS Number: acp-2013-350

This manuscript presents a new modeling technique that builds on the methods of Owen and Honrath [2009] to fold FLEXPART retroplumes with GEOS-Chem forward simulations to isolate the chemical evolution of pollution plumes as they are transported from the eastern USA to Pico Mountain Observatory in the remote eastern North Atlantic Ocean. Overall I find the modeling technique to be sound and of value to the scientific community. However I have strong reservations regarding the overall conclusions. While I think it is very possible that the O_3/CO relationship can be driven as much by CO loss as by ozone production/destruction (based on the previous results of Real et al. 2008) I worry that the present study has reached the same conclusion due to some faulty assumptions. I elaborate on this concern below and also provide additional comments/suggestions that would improve the analysis. Without further analysis I have no confidence in the main conclusions of this manuscript. However, I think that if the authors revise their analysis as outlined below they will produce much more reliable estimates of the evolution of ozone and CO between the USA and PMO. My recommendation to the editor is that this paper be sent back to the authors for a major revision.

We thank the referee for providing valuable comments and suggestions to improve the analysis in our manuscript. Based on our study of d[O_3]/d[CO] evolution in two selected events, we found that ozone production may not be the only reason for the increase in d[O_3]/d[CO] during transport of pollution plumes. It can also be caused by photochemical loss of CO during transport. The work of *Real et al.* (2008) reached similar conclusions, and we used some pollution plume data that were also used in the work of *Real et al.* (2008). The referee provided several ways to improve the analysis in the manuscript to better support to the conclusions. We carefully considered the referee's comments and have revised the manuscript as described here.

Major comments:
The main conclusions of the analysis are drawn from Figure 10 which explores the mixing process between polluted air masses that travel from the eastern US to PMO. The analysis assumes that the pollution plumes from the USA mix with background air which the authors decide is characterized by the lowest 10% of ozone and CO mixing ratios measured at PMO during the summer. But why would the plumes only mix with clean "background" conditions. Couldn't the plumes just as easily mix with moderately polluted air? And where in the atmosphere are these clean "background conditions" found? Aged mid-latitude air does not have such low ozone (17 ppbv) and CO (63 ppbv) mixing ratios. Rather, these mixing ratios are typical of the tropical lower troposphere, so if they are found in the mid-latitudes they are associated with tropical air masses that have been recently advected into the mid-latitudes. If the authors ran FLEXPART retroplumes for these "background" events I am confident that they would show a tropical origin. So basically the authors are assuming that whenever a pollution plume leaves the eastern USA it mixes with tropical air. This might be true some of

the time, but it's far more likely that the plume would mix with aged mid-latitude air which has much higher ozone and CO mixing ratios. By examining the spatial extent of the retroplumes in Figure 3 we can get an idea of the range of air masses that mixed to produce the polluted air mass sampled at PMO. Neither of the retroplumes in Figure 3 indicates a contribution from tropical air. Therefore the authors need to seriously reconsider how they characterize background air. They should provide a range of background conditions and then see how robust their conclusions are.

In the original d[O_3]/d[CO] analysis, we used the lowest 10% of observed ozone and CO mixing ratios in a 3-month summertime period as the background over the North Atlantic. The referee believes the CO and ozone mixing ratios estimated in this way are too low to represent the tropospheric background in the mid-latitude region. The referee suspects such low mixing ratios were due to transport from tropic regions where background CO and ozone should be significantly lower than the background over the North Atlantic. Thus, the analysis in Fig. 10 could be invalid. To address this issue, we examined FLEXPART retroplumes for the periods when the lowest 10% mixing ratios were observed. The results confirmed the referee's conjecture, showing that most of the trajectories originated in tropical regions. We are grateful to the referee for providing this insight.

Because the estimation of the background was found to be incorrect, we revised our analysis and now use two criteria to identify North Atlantic background values specific to the summertime 2009 and 2010 CO and ozone measurements at PMO. First, a retroplume should have more than 50% of its residence time over the North Atlantic region (defined by latitude from 30 ºto 48 ºand longitude from -60 ºto -15 º) at ten days upwind from PMO. Given a typical transport time from North America to PMO is about 6-7 days (*Honrath et al.*, 2004), this threshold can insure that the CO and ozone mixing ratios observed during this period were aged mid-latitude North Atlantic air. Second, the retroplumes should have more than 80% of its residence time under a vertical height of 5 km at ten days upwind from PMO. In this way, stratospheric intrusion and any transport from the upper troposphere where ozone levels are much higher are ruled out. For times when qualified retroplumes occurred and for cases in which both CO and ozone values were available, averaged mixing ratio of each species was used as background in the revised Fig. 10. After applying these criteria, background CO and ozone mixing ratios were higher than the original estimates. The North Atlantic background CO for Event 2 in 2009 increased from 63 to 78 ppbv and ozone increased from 17 to 30 ppbv, which altered the simulated value of d[O_3]/d[CO] (L_1) from 1.05 to 1.15. For Event 6 in 2010, the updated background CO increased from 68 to 88 ppbv and ozone increased from 17 to 26 ppbv, which altered the simulated value of d[O_3]/d[CO] from 0.72 to 0.90. The new simulated values of d[O_3]/d[CO] are promising because they align better with the observed values (L_0, 1.37 for Event 2 and 0.99 for Event 6) for both events. The slope values for the other derived lines (L_2 and L_3) have been re-calculated accordingly. After we replaced the outdated plume data by recent MOZAIC/IAGOS data in the d[O_3]/d[CO] analysis (in response to the second major comment from this referee), the discussion and conclusions regarding d[O_3]/d[CO] during transport were still valid.

In the revised manuscript the new method used for determining the theoretical background replaced the original text at page 15163, line 13, sentences "The plume point …… observed in May-July 2009." The text now reads:

> "The plume point for Event 2 (red triangle in Fig. 10a) at PMO is determined by averaging the top 10% of O_3 and CO mixing ratios from measurements associated with this event. The background point is estimated from observed O_3 and CO mixing ratios at PMO during specific periods determined to be representative of the North Atlantic regional background. This determination is made by selecting periods with FLEXPART retroplumes having more than 50% of their residence time over the North Atlantic region (defined by latitude from 30 ºto 48 º, longitude from -60 ºto -15 º) and more than 80% of their residence time under a vertical height of 5 km at ten days upwind from PMO. Given a typical transport time from North America to PMO is approximately 6-7 days *(Honrath et al.*, 2004), this choice of upwind time insures that the CO and ozone mixing ratios selected were aged mid-latitude North Atlantic air. The average CO and O_3 mixing ratios for qualified periods were used as the background point (green circle in Fig. 10a)."

On the same page, line 18 "63 ppbv of CO and 17 ppbv of ozone" was replaced by "78 ppbv of CO and 30 ppbv of ozone"

Additionally, we replaced the text on page 15168, line 8, which reads "The lowest 10% of data …… 17 ppbv ozone." with

> "Similarly, the North Atlantic background CO and ozone for summertime 2010 were found to be 88 ppbv and 26 ppbv, respectively."

All slope values for L_1, L_2 and L_3 in Fig. 10 were updated throughout the manuscript.

Another problem with this analysis is that there is absolutely no evaluation of GEOS-Chem estimated ozone and CO over the eastern USA. If we don't know that the model is reasonably correct at the start of the pollution export episode, how can we have confidence that it correctly reproduces the chemical evolution of the plume? The authors state that 1) in situ measurements for model evaluation are not available, and 2) conclude that it is acceptable to instead use ozone and CO measurements from field missions conducted in 1993 and 2004 and adjust the CO data to account for emissions changes that have occurred over the 1993-2010 time period. These assumptions are incorrect and greatly lower my confidence in the conclusions of the paper. 1) There are plenty of in situ measurements across the eastern USA to evaluate GEOS-Chem. There are many rural ozone sites in the National Park Service and CASTNET databases (many at high elevations) that are suitable for evaluating a course model such as GEOS-Chem: http://epa.gov/castnet/javaweb/index.html http://ard-request.airresource.com/ TES ozone and CO retrievals for the mid- and lower troposphere can also be used above the USA and the North Atlantic. Also, MOZAIC aircraft profiles are available, as discussed below. 2) Trying to characterize ozone and CO above the eastern US in 2009 and 2010 using data from 1993 and 2004 is a bad idea. Even though the authors acknowledge that emissions have changed over this time period their method of adjusting the measured CO to account for

the change is not acceptable when they could produce a very nice summertime ozone/CO climatology for the eastern US using MOZAIC ozone and CO profiles from 2008-2011, as described below. Furthermore, it's not just CO that has decreased greatly, but so have NOx and VOCs which have greatly changed the photochemistry over the eastern US and resulted in very large decreases in ozone (see Cooper et al. 2012, and He et al. 2013). So ozone and CO from 1993 and 2004 cannot be used to characterize the eastern USA in 2009 and 2010, and the authors need to use the freely available MOZAIC data.

Cooper et al. (2012), Long-term ozone trends at rural ozone monitoring sites across the United States, 1990 ̶C2010, J. Geophys. Res., 117, D22307, doi:10.1029/2012JD018261.

He et al. (2013), Trends in emissions and concentrations of air pollutants in the lower troposphere in the Baltimore/Washington airshed from 1997 to 2011, Atmos. Chem. Phys. Discuss., 13, 3135-3178.

The MOZAIC program (now called IAGOS) has used commercial aircraft to measure ozone around the globe since 1994 and CO since 2001. I checked the IAGOS database: http://www.iagos.fr/web/rubrique40.html and there are plenty of flights to the eastern US (Boston, Philadelphia, Detroit, Atlanta) during the summers of 2008-2011. These ozone and CO profiles can be used to produce climatological ozone and CO values for the eastern US lower troposphere. These values need to replace the extremely outdated ozone and CO values from 2004 and 1993 that are currently used in this manuscript. Furthermore the MOZAIC profiles from June 2009 and July 2010 can be used to evaluate GEOS-Chem's estimates of ozone and CO exported from the US in the two case studies examined by this manuscript.

We thank the referee for suggesting several valuable data sources to improve our d[O_3]/d[CO] analysis. We have looked into each source the referee recommended and were able to incorporate the MOZAIC/IAGOS flight measurements to improve the d[O_3]/d[CO] analysis in the manuscript. As a result, several paragraphs in Sect. 5.1 and 5.2 in the manuscript were revised. We were not able to find useful data corresponding to our events from the other sources suggested by the referee.

The referee raised two issues in this comment. One was a lack of evaluation of the GEOS-Chem results. With regards to this, we looked for records of both Event 2 in 2009 and Event 6 in 2010 in the data resources suggested by the referee. Unfortunately, we were not able to find time- and location-appropriate data to verify the CO and ozone mixing ratios in GEOS-Chem in the free troposphere over the eastern U.S. Both the CASTNET and National Park Service databases were measurements at surface sites. Although some sites are located on top of mountains, e.g., the site at Rocky National Park, there were no elevated surface site values at the location and time that the events occurred. We also examined satellite data for CO and ozone. The satellite-measured profiles were not able to clearly detect CO and ozone mixing ratios in the lower troposphere and showed nearly identical mixing ratios from the surface up to 5 km. The closest observation was a MOZAIC flight measured profile at Philadelphia Airport, which was measured a day before the 2009 event occurred and

approximately 800 kilometers away.

Although we were not able to find any data that are directly linked to the studied events, we evaluated simulations of CO and ozone in GEOS-Chem by two other means. First, during the discussion of the GEOS-Chem model in Sect. 2.3, we added references about GEOS-Chem CO and ozone simulations over North America and the North Atlantic to document GEOS-Chem performance in simulation over the eastern U.S. Second, we cited a work by our colleagues (*Kumar et al.*, 2013), which used the same GEOS-Chem simulations for their study. The manuscript by *Kumar et al*. (2013) discussed in detail observed CO and ozone trends at PMO and compared the results with both GEOS-Chem simulations and satellite data. They found that GEOS-Chem underestimated CO over the North Atlantic, but successfully captured seasonal cycles and decreasing trends in CO and ozone at PMO. In the revised Fig. 10, the upwind $d[O_3]/d[CO]$ estimated by GEOS-Chem falls within the slope range estimated based on MOZAIC measurements (see below). Overall, we believe the GEOS-Chem simulation used in this study is credible, and the results from the $d[O_3]/d[CO]$ analysis using the GEOS-Chem data provides a useful representation of the range of ozone and CO behaviors. Associated changes to the manuscript begin at line 21, page 15147, and read,

> "The performance of the model in simulating CO and ozone has been comprehensively evaluated (e.g., *Jacob et al.*, 1993; *Bey et al.*, 2001; *Wang et al.*, 2009). Simulation of CO was found to be significantly affected by emission inventories used, but model results generally show reasonable agreement with data from various observational sites or networks around the world (*Duncan et al.*, 2007). GEOS-Chem has also been applied to simulate outflow events from North America *(Liang et al.*, 1998*; Li et al.*, 2004*; Auvray and Bey*, 2005; *Hudman et al.*, 2009). *Li et al.* (2005) used GEOS-Chem to characterize the major outflow pathways from North America to the North Atlantic and the model successfully captured ozone chemistry during convective lifting of pollution plumes. In the work by *Millet et al*. (2006), GEOS-Chem was used to estimate CO in outflows from the U.S. to Chebogue Point."

At line 5, page 15148, we also added,

> "These GEOS-Chem simulations are a subset of those used in *Kumar et al.* (2013), in which the observed CO and ozone trends at PMO were compared with both GEOS-Chem simulations and satellite data from AIRS and TES. They found that GEOS-Chem underestimated CO over the North Atlantic, but successfully captured the seasonal cycles and decreasing trends of CO and ozone at PMO."

The second concern in this comment was about the outdated plume data that we used in the $d[O_3]/d[CO]$ analysis (H-points in Fig. 10). While we explored all of the data sources recommended by the reviewer, we ultimately decided to use the referee's suggestion to produce a climatology of CO and O_3 from MOZAIC/IAGOS flights. The climatology consisted of CO and ozone profiles measured at two airports from 2008 to 2010. The reason for including data from 2008 is because there were only a few flight measurements available in late September for 2010. We included profiles in 2008 to compensate the lack of data in early summer of 2010. Profiles at Philadelphia and Atlanta were used to produce

climatological ozone and CO values for the plumes of Event 2 and Event 6, respectively, because these airports were closest to the origin of each event. Available CO and ozone mixing ratios from 2 to 3 km above the surface were extracted and considered as lower free troposphere air. The top 10% of CO mixing ratios at this level were treated as highly polluted plumes, and the selected CO and simultaneously measured ozone were used as upwind plume data for the two events. In the revised Fig. 10, these CO and ozone values were connected to the new North Atlantic background levels (discussed above) to produce a range of upwind $d[O_3]/d[CO]$ slopes (see hatched area in revised Fig. 10). Although the new slope range was slightly different from what we derived from the old plume data (for Event 2 in 2009 the updated slope range is from 0.48-0.78 vs. 0.65-0.83 using the old plume data; for Event 6 in 2010 the updated range is from 0.51-0.92 vs. 0.67-0.89), our conclusion was unchanged. We replaced the outdated plume data and incorporated these recent MOZAIC data in the manuscript. A list of relevant revisions is provided below.

Two paragraphs starting from line 12, page 15165 and line 5, page 15166, Table 2 and Equations 3 and 4 were removed from the manuscript.

At line 2, page 15163, we replaced the sentence "In order to evaluate the upwind $d[O_3]/d[CO]$ …… ICARTT (2004) research campaigns." with

> "In order to evaluate the values of upwind $d[O_3]/d[CO]$, we compare the values to upwind slopes simulated by GEOS-Chem and observational data from the Measurements of OZone, water vapour, carbon monoxide and nitrogen oxides by in-service AIrbus aircraft (MOZAIC) Program."

At line 12, page 15165, we added a paragraph to introduce how the upwind slope range was calculated by using MOZAIC data:

> "We also investigated upwind pollution plumes by using flight measurements obtained from the MOZAIC Airborne Program. To estimate the composition of the Event 2 plume when it was exported from eastern North America, we chose the CO and ozone profiles from Philadelphia, the closest MOZAIC airport to the location of the event origin (see Fig. 3) from 2008-10. The 2008 data were included because there were only a few profiles collected in summertime 2010. CO and ozone mixing ratios measured from altitude 2 km to 3 km were considered to represent lower free troposphere air. The top 10% of CO mixing ratios at this level were treated as highly polluted air and were used to determine the potential range of the initial plume conditions for Event 2. By connecting these CO and ozone mixing ratios in the plume with the North Atlantic background, we obtained a range in upwind $d[O_3]/d[CO]$ from 0.48 to 0.79 (H-range in Fig. 10a). This range is a little larger than those of recent satellite observations downwind of eastern North America (0.4-0.6; *Voulgarakis et al.*, 2011), but consistent with values measured near coastal areas observed during the ICARTT Campaign (0.81 and 0.72; *Zhang et al.*, 2006)."

At line 3, page 15169, we replaced the sentence "Other upwind slope values …… range from 0.67 to 0.80 (Fig. 10b)." with

"We also estimated the CO and ozone composition for Event 6 plume when it was exported from North America by using the same approach as done for Event 2. Profiles from Atlanta were used because it was the closest MOZAIC airport to the location of export in Event 6. By connecting the estimated CO and ozone composition and the background point, we obtained a range of upwind $d[O_3]/d[CO]$ from 0.51 to 0.92 (H-range in Fig. 10b)."

We added MOZAIC program into the acknowledgement:
"The authors acknowledge the European Commission for the support to the MOZAIC Project (1994-2003) and the preparatory phase of IAGOS (2005-2012). The authors also thank all scientists involved in the program and Valérie Thouret's help in getting access to the data."

Minor comments:

1) The first few lines of the Introduction discuss ozone properties but are not referenced. Please provide some overview references.

References on tropospheric ozone were added through lines 2 – 5 on page 15143.

2) Line 6 page 15146 What is the base year for the EDGAR inventory?

With regards to the EDGAR inventory, we modified the sentence at line 6 page 15146 as follows: "The model was coupled with CO emissions from the Emissions Database for Global Atmospheric Research (EDGAR version 3.2; *Olivier and Berdowski*, 2001) and the Global Fire Emissions Database (GFED v3.1, daily averaged fire emissions; *Mu et al.*, 2011)."

3) Line 11 page 15146 GFS winds are used at 1 degree resolution. But GFS winds have been available at half degree resolution for years. Why not use the half degree winds?

The primary reason is because we do not expect significant improvement by using GFS data with higher resolution. In this study, GEOS-Chem simulations were set up at a 2 by 2.5 degree resolution, so FLEXPART runs with much higher resolution may not necessarily improve folded retroplume results, which is a combination of the two models.

4) Line 27 page 15146 Here and throughout the paper the term "outputs" is used, which is not a word. Instead please use "output" or some other expression.

All "outputs" are replaced by "output" throughout the paper.

5) Line 24 page 15147 Here it says the model was only run for January-July 2010. So where does the 2009 model output come from?

We thank the referee for capturing this typo. The model was run for January-July for both 2009 and 2010. This statement was fixed in the revised manuscript.

6) Line 16 page 15148 I'm not sure what is meant by "entrywise"

A reference (*Charles*, 1989) was added to the revised manuscript. The entrywise product of a matrix is also called the Hadamard Product. A brief explanation can be found on a wiki website:
http://en.wikipedia.org/wiki/Hadamard_product_(matrices)#CITEREFHornJohnson1985
Charles, J. (1989), *Matrix Theory and Applications*, the American Mathematical Society Phoenix, Arizona, United States of America.

7) Line 10 page 15153 Here model data in the middle of the North Atlantic is used to determine when boundary layer air rises to PMO. Why use coarse, unreliable model data in the middle of the Atlantic when you could easily use in situ specific humidity time

series to tell when moist and relatively warm boundary layer air is advected to the site?

The Sheppard Model was used to calculate the height of the dividing streamline for verification of mechanical lifting to the top of Pico Mountain. If the height of a dividing streamline is lower than boundary layer height, the marine boundary layer air can be brought to PMO by mechanical lifting of the mountain slope. This method has been tested and applied at PMO in a published work for the same purpose (*Kleissl et al.*, 2006). The GFS meteorology data were used in the model calculation because it is the best we can get for this purpose. In addition, *in situ* humidity measurements may not be able to exactly reflect the origin of air. From the authors' personal experiences, PMO is frequently immersed in clouds. This phenomenon is not necessarily correlated with mechanically-lifted upslope flow. Clouds can be formed at the level of PMO in the absence of upslope flow. On the other hand, mechanical lifting may not lead to cloud formation.

8) Line 20 page 15153 If you ran GEOS-Chem at 2x2.5 degrees, why would you extract ozone and CO from a different model run at 4x5 degrees?

The 4x5 GEOS-Chem runs (with the same fully coupled ozone-NO_x-VOC-aerosol chemistry) were conducted for a much longer time period (10 years) than the 2x2.5 simulations. These 4x5 results were used to assist in selecting the time periods for the 2x2.5 runs. For the purpose of saving computing resources and time, the 2x2.5 degree GEOS-Chem simulations were only conducted for time periods covering the selected events.

9) Line 15 page 15155 Here the authors say that they identified warm conveyor belts using NOAA daily weather maps. But these maps only show surface fronts and 500 hPa isoheights. They do not show WCBs. The only way to say for sure if a WCB was present is to check archived satellite images at: http://locust.mmm.ucar.edu/imagearchive/

We thank the referee for providing better meteorological information for identification of WCBs. We looked into the satellite images through the link provided by the referee and successfully identified the WCB on 9 June 2009 for Event 2 (http://locust.mmm.ucar.edu/imagearchive/, under Satellite/Satellite IR national). In the revised manuscript, we replaced the link for the NOAA weather map with the satellite image.

10) Page 15158 Instead of combing FLEXPART with GEOS-Chem, why not just use the GEOS-Chem adjoint [Zhang et al., 2009] to achieve the same results?

Zhang, L., D. J. Jacob, M. Kopacz, D. K. Henze, K. Singh, and D. A. Jaffe (2009), Intercontinental source attribution of ozone pollution at western U.S. sites using an adjoint method, Geophys. Res. Lett., 36, L11810, doi:10.1029/2009GL037950.

The referee provided another option for studying chemical evolution in transported pollution plumes, the GEOS-Chem adjoint model. GEOS-Chem and FLEXPART have been set up and run smoothly for several years at Michigan Technological University. Many works published

previously by the PMO group were built upon results from these two models. On the other hand, the GEOS-Chem adjoint model has its own separate code package and requires completely different model set-up from the regular GEOS-Chem model. We had more confidence in using tools with which we are familiar with and have expertise. More importantly, the combination of the two models achieved a semi-Lagrangian view of comprehensive chemical and physical information during transport of pollution plumes, including mixing ratios of chemical spices, transport pathways, ozone production rates and NMHC aging. It would have required much more time and effort for us to develop the GEOS-Chem adjoint model to gather equivalent information. Finally, we are happy to be able to develop alternative approaches that can benefit the community by providing a) options/alternative research tools, b) opportunities to inter-compare/cross-validate different approaches.

Two other papers that need to be considered when discussing the background of the O_3/CO relationship are:
Chin et al. (1994), Relationship of ozone and carbon monoxide over North America, J. Geophys. Res., 99, 14,565-14,573.
Cooper et al., PROPHET 1998 meteorological overview and air-mass classification, J. Geophys. Res., 106, 24,289-24,299.

Chin et al. noted that a typical ozone/CO slope in the USA is 0.3 and attributed this to photochemistry. But Cooper et al. showed that transport alone could also produce a slope of 0.3, as the authors of this manuscript mention on lines 17-20, page 15151. Table 2 There are quite a few other papers that discuss transport of ozone and CO to the North Atlantic, though not all in summer. Was the intention that this table just focus on summer events? A paper that was omitted is:

Berkowitz et al. (1996), Synoptic patterns associated with the flux of excess ozone to the western North Atlantic, J. Geophys Res., 101, 28923-28933.

And a good springtime study is:
Prados et al. (1999), Transport of ozone and pollutants from North America to the North Atlantic Ocean during the 1996 AEROCE intensive, J. Geophys. Res., 104,

We thank the referee for kindly providing several relevant papers addressing ozone-CO correlation over the North America. Although this study focused on ozone-CO correlation in summertime, we decided to cite these papers in the revised manuscript in Sect. 3.1.2 in which the background of ozone-CO correlation was discussed.

With regards to this comment, at line 8, page 15151, we added:

> "Significant correlation between CO and ozone was found over eastern North America. A d[O_3]/d[CO] of 0.3 was speculated to represent a uniform characteristic of boundary layer air over eastern North America in summertime (*Cooper et al.*, 2001;*Chin et al.*, 1994)."

On the same page, at line 10, we added

> "At Nova Scotia, *Berkowitz et al*. (1996) observed transported pollution plumes that originated from urban areas of North America and $d[O_3]/d[CO]$ ranged from 0.19 – 0.30 ($R^2 > 0.5$). During the 1993 summer NARE intensive, a range in $d[O_3]/d[CO]$ from 0.25 to 0.28 was observed over eastern North America and the North Atlantic (*Parrish et al.*, 1993; *Daum et al.*, 1996; *Fehsenfeld et al.*, 1996a). In a spring study, Prados et al. (1999) reported a $d[O_3]/d[CO]$ of 0.21 ($R^2 = 0.19$) during intensive aircraft measurements between the U.S. and Bermuda. "

References cited during the authors' revision:

Auvray, M., and Bey, I.: Long-range transport to Europe: Seasonal variations and implications for the European ozone budget, Journal of Geophysical Research-Atmospheres, 110, 22, D1130310.1029/2004jd005503, 2005.

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Cooper, O. R., Moody, J. L., Thornberry, T. D., Town, M. S., and Carroll, M. A.: PROPHET 1998 meteorological overview and air-mass classification, Journal of Geophysical Research-Atmospheres, 106, 24289-24299, 10.1029/2000jd900409, 2001.

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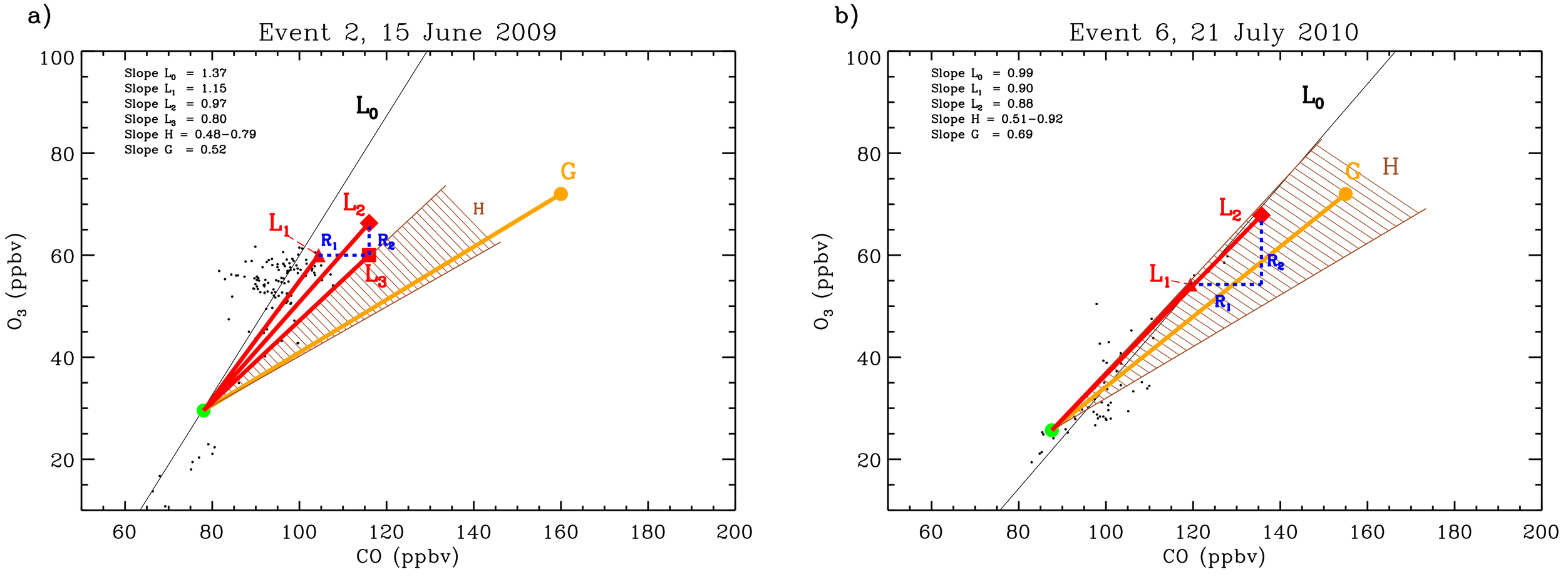
a)
Event 2, 15 June 2009
Slope L_0 = 1.37
Slope L_1 = 1.15
Slope L_2 = 0.97
Slope L_3 = 0.80
Slope H = 0.48–0.79
Slope G = 0.52
L_0
L_1
L_2
L_3
R_1
R_2
H
G
O_3 (ppbv)
CO (ppbv)
b)
Event 6, 21 July 2010
Slope L_0 = 0.99
Slope L_1 = 0.90
Slope L_2 = 0.88
Slope H = 0.51–0.92
Slope G = 0.69
L_0
L_1
L_2
R_1
R_2
G
H
O_3 (ppbv)
CO (ppbv)

Revised Fig. 10. Calculations of $d[O_3]/d[CO]$ for the two selected events (a – Event 2; b – Event 6). Background levels for the central North Atlantic (green circles), and simulated upwind pollution plume data points (red symbols) are defined in the text. Solid black lines (L0) indicate two-sided regression results of observation data (black dots) at PMO. The brown shaded area indicates the range in upwind lines derived using MOZAIC flight measurements. Line G has an upwind slope that is derived from the GEOS-Chem captured plume. Adjusted lines L1-L3 for each event demonstrate the variation in $d[O_3]/d[CO]$ according to chemical transformation pathways R1-R2 during transport.