Photochemical evolution of continental outflow to Pico Mountain (Azores): Integrating observations with CTM simulations in lower-FT outflow

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1 Synopsis

An analysis of the consistency of lower-FT observations with chemical transport model (CTM) simulations in aged U.S. export.

- Measurements: Pico Mountain lower-FT observatory, ∼4–8 days downwind of the N. American east coast.
- CTM simulations: GEOS-Chem simulations for 2005.
- Events occurred in both observations and CTM simulations.
- Analyzed using new tools that combine CTM simulations with FLEXPART retroplumes to probe the evolution of CO, O_3, and OH in the export events in conjunction with the observations.
  - FLEXPART retroplumes define where the air ultimately sampled at Pico was, at specified times upwind.
  - By convolving GEOS-Chem output with the retroplume, one can calculate the mixing ratio that would reach Pico if all chemistry ceased from that point forward.
  - This allows probing the chemical evolution in the CTM in a semi-lagrangian fashion.
  - This analysis is shown in section 4.
  - (More info: see Handout: Owen et al. AGU poster.)

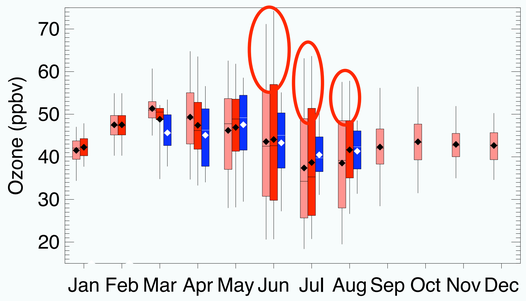
2 Evidence of discrepancies

Comparisons between in-situ observations and GEOS-CHEM simulations sampled at the location of the Pico station reveal several differences deserving further study. Two such comparisons are shown here.

O_3 seasonal cycle.

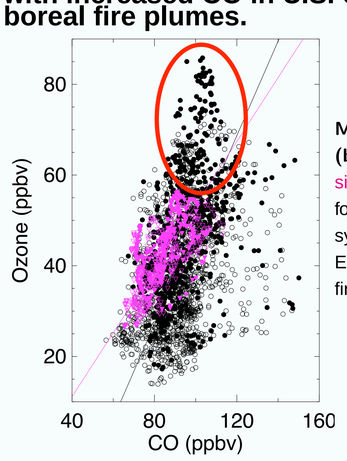
Shown: Pico 2001–2005, Pico 2005, and GEOS-Chem, 2005.

Boxes show middle 50%, whiskers show middle 90%.



- Observed O_3 variability is greatest during summer.
  - The magnitude of this increase is underestimated by the model.
  - Our focus here is on the high summertime O_3 observations.

Most high-O_3 observations are associated with increased CO in U.S. outflow or boreal fire plumes.



Measurements (black) and simulations (magenta) for summer 2005. Solid symbols indicate U.S., European, or boreal fire outflow.

3 Characterization of clear U.S. outflow events

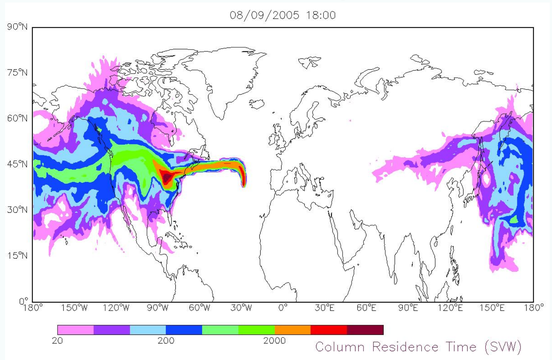
Continuous measurements allow sampling of clear outflow events, among many additional events that mix anthropogenic and biomass-burning impacts.

- Two clear events of U.S. outflow in the lower free troposphere occurred during August 2005.

- **Transport pathway: from FLEXPART**

FLEXPART retroplume residence times: well-organized transport from the east coast.

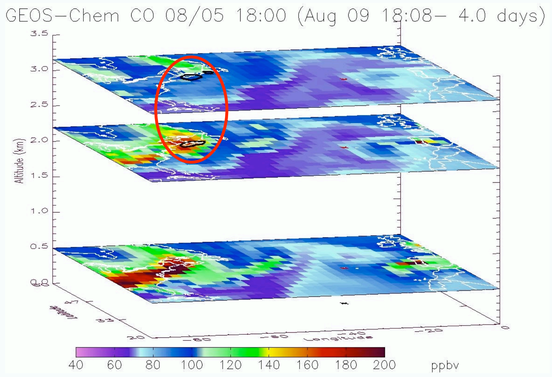
Case 1: Arrives 18:00 Aug. 9, 2005



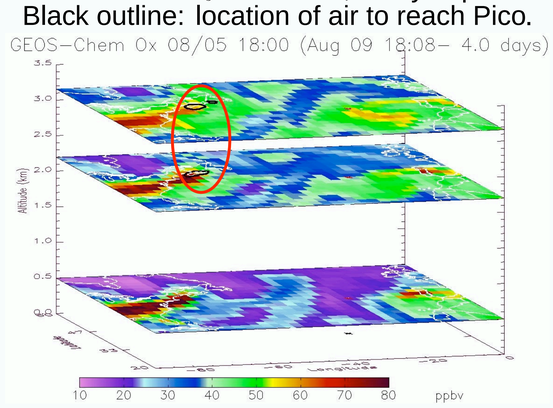
FLEXPART indicates average height over the ocean = 2.5 to 4 km.

- **The event is simulated by GEOS-Chem.**

GEOS-Chem CO simulation, 4 days upwind. Black outline: location of air to reach Pico.



GEOS-Chem O_3 simulation, 4 days upwind. Black outline: location of air to reach Pico.

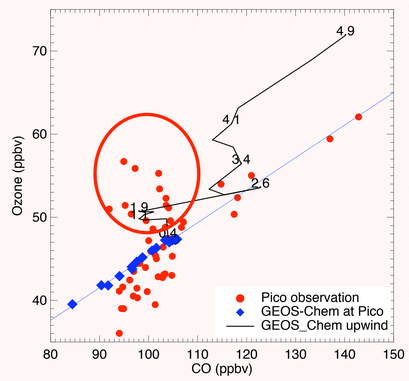


4 Analysis and interpretation combining observations, FLEXPART, and GEOS-Chem

Ozone vs CO, at Pico and upwind in GEOS-Chem

Case 1. August 9–11 event (transport shown in part 5 to the left).

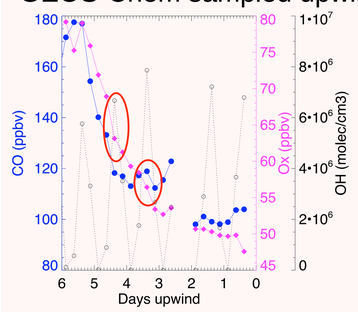
O_3 vs CO



Red circles: Pico observations,
Blue diamonds: GEOS-Chem simulations at Pico.

- GEOS-Chem simulates correlated CO and O_3 enhancements during this event.
- The Pico observations also exhibit enhanced CO and O_3, with 2 key differences:
  - The range of enhancement is larger than simulated. This could reflect dilution in the model grid cells.
  - There is a group of observations with much higher O_3 than expected from the GEOS-Chem O_3-CO slope and the observed CO.
- To investigate the cause, we convolved the GEOS-Chem simulations using the FLEXPART retroplume. This produced the
  Black line: FLEXPART retroplume sampling of GEOS-Chem simulations to give mixing ratios at Pico if all chemistry stopped after the indicated number of days upwind.
- The upwind GEOS-Chem values (numerals) lie above and to the left of the blue line extrapolated from the GEOS-Chem (at Pico) points. This indicates the importance of net O_3 destruction in the simulations.
- For the circled Pico observations, [O_3] corresponds to levels simulated up to 3.5 days upwind (2 to 4 days after export), but CO values are within the range simulated at Pico (and below levels simulated 3 days upwind).

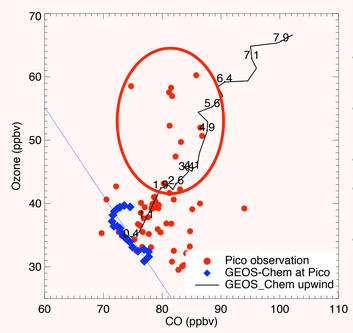
GEOS-Chem sampled upwind



Inspection of the FLEXPART-sampled GEOS-Chem simulations provides additional insight.
The plot to the left shows the CO and O_3 that would reach Pico if all chemistry ceased after the indicated number of days before arrival.
Red: GEOS-Chem upwind CO.
Blue: GEOS-Chem upwind Ox.
Black dotted line: Average OH at the upwind locations.

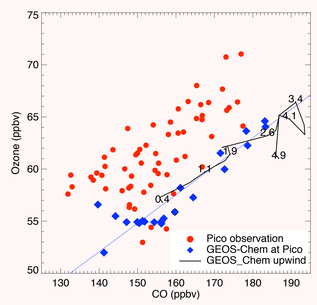
- The CO and O_3 lines would be flat and horizontal if no chemistry occurred and GEOS-Chem transport matched FLEXPART transport. (See Owen et al. poster handout for details.)
- The rapid drop starting at −5 days corresponds to export from N. America and indicates that dilution in GEOS-Chem exceeds that in FLEXPART.
- Additional O_3 loss due to net photochemical destruction (circled) is evident between days 5 and 4, and between days 4 and 3. (Daytime can be identified by peaks in upwind [OH].)

Case 2: Aug 3–5



- This event traveled north of Pico, then looped around the back of the Azores high. Travel time from N. America was ∼7 days.
- GEOS-Chem simulated the event, but its pathway diverged from FLEXPART's 3 to 4 days upwind of Pico. As a result, the GEOS-Chem at Pico points are do not show the event.
- Using the upwind FLEXPART+GEOS-Chem method, that does not prevent analysis of this event.
- The upwind time series (not shown) indicates significant simulated O_3 loss in this event also. The total O_3 decline after export was ∼25 ppbv, similar to Case 1.
- The O_3 levels observed at Pico during this event (circled) are again consistent with O_3 simulated by GEOS-Chem well upwind (4–6 days upwind, or 1–3 days after export). This suggests that O_3 destruction was overestimated.

Contrasting springtime case: April 21–24.



- Significant enhancements in CO (and in HCs and NO_y, not shown) occurred during this spring event. However,
- only a ∼10 ppbv drop in O_3 is simulated, mostly due to dilution. (Note that Y-axis range is reduced in this plot.)
- The observed and simulated O_3-CO relationships agree well during this event. The absence of a significant discrepancy here is consistent with the lack of simulated O_3 loss in transit.

Conclusions

- Ozone enhancements observed at Pico during well-defined U.S.-outflow events are larger than expected based on GEOS-Chem simulations.
- Analysis of simulations upwind of Pico indicates that O_3 peaks observed in the analyzed events are similar in magnitude to O_3 simulated well upwind.
  - This suggests that O_3 is more efficiently transported over the N. Atlantic Ocean than simulated, i.e. with less gross destruction or with greater gross production.
  - This discrepancy is probably not limited to the GEOS-Chem CTM (*e.g.*, [1]).
- The method used here—convolving lagrangian particle model retroplume simulations with 3-D CTM simulations to investigate transport and photochemistry—should be broadly useful for analysis of models and measurements.
- Although this analysis was limited to anthropogenic impacts, boreal wildfires affect Pico O_3 levels to a similar or even greater degree [12–14].

Pico station status

- Michigan Tech donated the station infrastructure to Portugal in June, 2006, in order to jump-start development of a permanent Portuguese station.
- Continuing station development is aimed toward creating a long-term Global Atmosphere Watch (GAW) observatory. See the Helmig poster for information on an EU effort to do this, and see the Fialho poster for more information on Azores atmospheric studies.
- Measurements in the near future are dependent on new funding.

References

Handouts are available for the large-font papers.

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Acknowledgements

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