Intercontinental Source Attribution of Ozone Pollution at Western U.S. Sites Using an Adjoint Method

Intercontinental source attribution of ozone pollution at western U.S. sites using an adjoint method

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[1] We use the GEOS-Chem chemical transport model and its adjoint to quantify source contributions to ozone pollution at two adjacent sites on the U.S. west coast in spring 2006: Mt. Bachelor Observatory (MBO) at 2.7 km altitude and Trinidad Head (TH) at sea level. The adjoint computes the sensitivity of ozone concentrations at the receptor sites to ozone production rates at $2^\circ \times 2.5^\circ$ resolution over the history of air parcels reaching the site. MBO experiences distinct Asian ozone pollution episodes; most of the ozone production in these episodes takes place over East Asia with maxima over northeast China and southern Japan, adding to a diffuse background production distributed over the extratropical northern hemisphere. TH shows the same Asian origins for ozone as MBO but no distinct Asian pollution episodes. We find that transpacific pollution plumes transported in the free troposphere are diluted by a factor of 3 when entrained into the boundary layer, explaining why these plumes are undetectable in U.S. surface air. **Citation:** 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.

1. Introduction

[2] Intercontinental transport of ozone pollution is becoming a major issue as countries at northern mid-latitudes strive to meet increasingly stringent air quality standards [*Task Force on Hemispheric Transport of Air Pollution*, 2007]. Ozone is produced in the troposphere by photochemical oxidation of CO and volatile organic compounds (VOCs) in the presence of nitrogen oxides ($NO_x \equiv NO + NO_2$). It has a lifetime of days in the boundary layer but weeks in the free troposphere [*Wang et al.*, 1998], enabling transport on the intercontinental scale. Intercontinental source attribution for ozone pollution at a given site is made difficult by the complexity and non-linearity in the chemistry, the multiplicity of sources and time scales, and the general lack of structure of the ozone background especially in surface air [*Goldstein et al.*, 2004]. We present here a new approach using the adjoint of a chemical transport model (CTM) and apply it to examine the detail of intercontinental influence on ozone pollution at two U.S. west coast sites.

[3] Previous CTM studies of intercontinental influence on surface ozone have used either ozone tracers tagged by production region [*Li et al.*, 2002; *Jaeglé et al.*, 2003; *Derwent et al.*, 2004; *Sudo and Akimoto*, 2007] or sensitivity simulations with perturbed emissions [*Jacob et al.*, 1999; *Yienger et al.*, 2000; *Wild and Akimoto*, 2001; *Derwent et al.*, 2008; *Duncan et al.*, 2008; *Fiore et al.*, 2009]. These source-oriented methods are computationally limited in the spatial resolution of the source region that they can achieve. The CTM adjoint offers a far more computationally efficient approach for a receptor-oriented problem such as source attribution of ozone at a given site. A single run of the adjoint model can compute the sensitivity of ozone concentrations at a given location and time (or an average over a spatial domain and time interval) to the global distribution of sources over the spatial and temporal resolution of the model. The method has been applied previously to pollutant transport to Hawaii [*Vukićević and Hess*, 2000; *Hess and Vukićević*, 2003], intercontinental transport of aerosol to the United States [*Henze et al.*, 2008], and regional sensitivity analyses for ozone pollution episodes [*Elbern and Schmidt*, 2001; *Hakami et al.*, 2006; *Nester and Panitz*, 2006].

[4] We use here the GEOS-Chem CTM and its adjoint to estimate source contributions to surface ozone pollution in spring 2006 at two nearby sites on the U.S. west coast, one at high altitude (Mt. Bachelor Observatory, Oregon) and one at sea level (Trinidad Head, California). The NASA/INTEX-B aircraft campaign over the northeast Pacific taking place at that time provided a detailed characterization of transpacific transport of ozone and its precursors [*Singh et al.*, 2009]. The GEOS-Chem simulation was previously evaluated in detail with INTEX-B as well as concurrent satellite and ground-based data, lending confidence in its representation of transpacific transport [*Zhang et al.*, 2008]. Mt. Batchelor Observatory and Trinidad Head are standard reference sites for background air entering the United States [*Goldstein et al.*, 2004; *Jaffe et al.*, 2005; *Oltmans et al.*, 2008]. The altitude difference between the two sites allows us to explore the dilution effect as Asian pollution plumes transported mainly in the free troposphere are entrained down to affect U.S. surface air.

2. GEOS-Chem Model and Its Adjoint

[5] The GEOS-Chem CTM (http://www.as.harvard.edu/chemistry/trop/geos/) [*Bey et al.*, 2001] is driven by assim-

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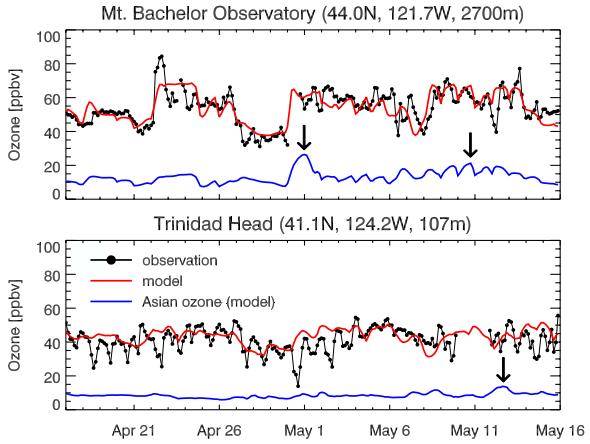

Figure 1. Time series of 3-hourly averaged ozone concentrations at (top) Mt. Bachelor Observatory and (bottom) Trinidad Head during the INTEX-B period (April 17–May 15, 2006). Model results (red) are compared to observations (black). The contribution of ozone produced over Asia in the model is also shown (blue). Black arrows indicate the Asian pollution events discussed in the text.

ilated meteorological data from the Goddard Earth Observing System (GEOS)-4 of the NASA Global Modeling and Assimilation Office (GMAO). Details of its application to simulate satellite, aircraft, and ground-based observations of ozone and its precursors during INTEX-B (April 17–May 15, 2006) are given by *Zhang et al.* [2008]. The GEOS-4 dataset has a temporal resolution of 6 hours (3 hours for surface variables and mixing depths), a horizontal resolution of 1° × 1.25°, and 55 layers in the vertical. We degrade the horizontal resolution to 2° × 2.5° for input to GEOS-Chem. We use Asian anthropogenic emissions from *Zhang et al.* [2009] for the year 2006. U.S. anthropogenic emissions are from the National Emission Inventory for 1999 (NEI 99) by the U.S. Environmental Protection Agency (EPA) (http://www.epa.gov/ttn/chief/net/). Transport of ozone from the stratosphere is simulated using the "Synoz" boundary condition of *McLinden et al.* [2000], which imposes a global cross-tropopause ozone flux of 495 Tg a^{-1}.

[6] *Zhang et al.* [2008] used a GEOS-Chem simulation with detailed NO_x-VOC chemistry for comparison to INTEX-B observations and for sensitivity analyses, but also archived daily 3-D fields of ozone production rates and loss frequencies to reproduce the ozone simulation results using tagged tracers of source regions. This tagged ozone tracer technique offers a computationally efficient approach for tracking the transport of ozone produced in different regions, and has been applied in a number of model studies [*Wang et al.*, 1998; *Li et al.*, 2002; *Sudo and Akimoto*, 2007]. We use it here in our adjoint model application. We define "Asian ozone" as ozone produced over Asia (8°N–55°N, 70°E–152°E) throughout the tropospheric column. 80% of this production is in the lower troposphere below 700 hPa. Asian ozone defined in this way does not discriminate between anthropogenic and natural sources, nor does it quantitatively resolve ozone production from Asian precursors downwind of the continent. It assigns the source geographically rather than by precursor emissions. The latter would be more precise for Asian anthropogenic source attribution but would require an adjoint model with full chemistry. *Zhang et al.* [2008] previously found by combining the two approaches that half of anthropogenic Asian influence on surface ozone in the western U.S. is from production in the Asian lower troposphere, with the rest from production in the free troposphere and downwind.

[7] The adjoint model of GEOS-Chem was constructed and tested by *Henze et al.* [2007] in work directed at constraining aerosol sources, and was further developed and applied by *Kopacz et al.* [2009] in an inverse analysis of CO emissions. We use the transport component of the adjoint including advection, boundary layer mixing, and convection [*Henze et al.*, 2007; K. Singh et al., Towards the construction of a standard adjoint GEOS-Chem model, paper presented at High Performance Computing and Simulation Symposium, Soc. for Model. and Simul. Int., San Diego, Calif., 2009]. We add self-adjoint ozone chemistry with archived ozone production rates and loss frequencies. The resulting model is used to compute the sensitivity of ozone concentrations at selected receptor sites to 3-D ozone production rates at 2° × 2.5° resolution for different time lags and over the history of air parcels reaching the site.

3. Time Series of Ozone at U.S. West Coast Sites

[8] We use ozone measurements from Mt. Bachelor Observatory (MBO, 44.0°N, 121.7°W, 2700 m) and Trinidad Head (TH, 41.0°N, 124.2°W, 107 m). MBO is a mountain site in central Oregon that is particularly sensitive to Asian influences due to its exposure to the free troposphere [*Jaffe et al.*, 2005; *Weiss-Penzias et al.*, 2006; *Wolfe et al.*, 2007]. TH on the northern California coast is widely used as a surface background site for the United States [*Goldstein et al.*, 2004; *Oltmans et al.*, 2008; *Parrish et al.*, 2009]. The TH ozone measurements were obtained from http://www.esrl.noaa.gov/gmd/obop/thd/.

[9] Figure 1 shows the 3-hourly observed and modeled time series of ozone at MBO and TH for the INTEX-B period. There is good agreement between the measurements and GEOS-Chem. The mean observed concentration at MBO is 54 ± 10 ppbv, compared with 53 ± 9 ppbv in the model, while the mean observed concentration at TH is 41 ± 7 ppbv, compared with 43 ± 5 ppbv in the model. The model cannot reproduce the low ozone levels often observed at TH at night due to local deposition under stratified conditions [*Goldstein et al.*, 2004], but the synoptic-scale variability is well captured.

[10] The contribution of ozone produced over Asia in the model ("Asian ozone") at the two sites is also shown in Figure 1. It averages 13 ± 3.6 ppbv at MBO and 8.4 ± 1.4 ppbv at TH. This is somewhat larger than the Asian anthropogenic ozone enhancement derived by *Zhang et al.* [2008] from a sensitivity simulation with Asian anthropogenic emissions turned off in the same model with full-chemistry (9 ± 3 ppbv at MBO). The difference is due to natural production over Asia contributing to Asian ozone as defined here; see section 2 for further discussion. The weaker and less variable contribution at TH than at MBO can be explained by dilution of free tropospheric plumes during entrainment in the boundary layer [*Hudman et al.*, 2004]. Model Asian ozone at MBO shows a maximum

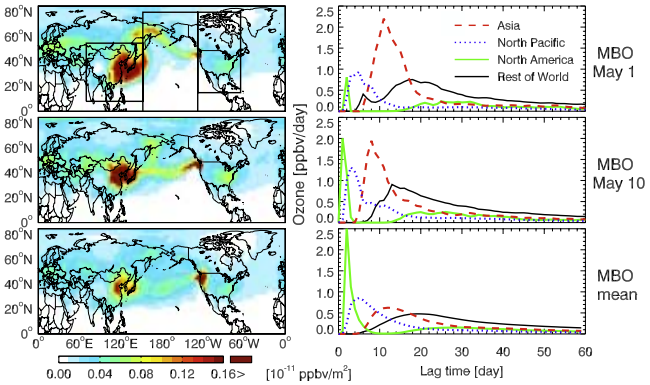

Figure 2a. Sensitivity of ozone concentrations at Mt. Bachelor Observatory, Oregon (MBO, 2.7 km altitude) to ozone production worldwide as inferred from the GEOS-Chem adjoint model: Asian pollution events at MBO ((top) May 1, 2006, at 00 UT and (middle) May 10, 2006, at 18 UT) as highlighted in Figure 1 and (bottom) the mean for the INTEX-B period (April 17–May 15, 2006). (left) The sensitivities integrated in time, over the depth of the tropospheric column and at the 2° × 2.5° grid resolution of the model. (right) The time-dependent sensitivities (going back in time) to ozone production over Asia, the North Pacific, North America, and Rest of World (as indicated by rectangles).

event on May 1 (26 ppbv) and a broader event on May 6–11, consistent with independent analyses of Asian pollution plumes observed at MBO during INTEX-B [*Wolfe et al.*, 2007; *Zhang et al.*, 2008]. Asian ozone at TH shows maximum influence on May 12, reflecting subsidence of the May 6–11 MBO plumes.

4. Fine Geographical Source Attribution for Ozone

[11] Figure 2 shows the sensitivities of ozone concentrations at MBO and TH to the global distribution of ozone production rates for the previous two months, as inferred from the GEOS-Chem model adjoint. The left panels show the integrals of the production rates over time and over the tropospheric column depths at the 2° × 2.5° horizontal resolution of the model. They show the amount of ozone produced in each grid square and transported to the receptor site with chemical loss accounted for during transport. Summing these values globally over all 2° × 2.5° grid squares approximates the ozone concentrations simulated by GEOS-Chem at the receptor site; there is a 10%–15% residual that reflects production in the stratosphere and tropospheric production at time lags larger than 2 months. The right panels show the time-dependent sensitivities to production over Asia (8°N–55°N, 70°E–152°E), North Pacific (0°N–80°N, 152°E–232°W), North America (15°N–80°N, 232°W–295°W), and Rest of World. Similar sensitivity spectra have been shown by *Vukićević and Hess* [2000]. Integrating under these curves gives the total contributions of ozone production in these regions to the ozone concentrations at the receptor site.

4.1. Asian Pollution Events

[12] Figure 2a (top) show the sensitivities of ozone concentrations at MBO for the transpacific ozone pollution events of May 1 at 00 UT and May 10 at 18 UT. Most of the ozone production contributing to MBO ozone on those days took place over East Asia, with maxima over the northeast China plain and southern Japan. We also find significant production over the North Pacific during plume transport. The May1 plume took a more northerly and higher-altitude route than the May 10 plume, resulting in less ozone production over the Pacific [*Zhang et al.*, 2008]. Both plumes show a secondary maximum of ozone production just off the west coast of United States, where subsidence of air masses causes decomposition of Asian PAN (peroxyacetylnitrate, a thermo-unstable NO_x reservoir species) and drives further ozone production [*Kotchenruther et al.*, 2001; *Heald et al.*, 2003; *Hudman et al.*, 2004; *Zhang et al.*, 2008]. In addition to these direct Asian pollution influences, both plumes show a significant background contribution to ozone from diffuse production in the extratropical northern hemisphere.

[13] The sensitivity spectra on Figures 2a (right) and 2b (right) show the transport timescales from production region to the receptor site. We see for the two Asian pollution episodes (top two panels) that ozone produced over North America had an immediate impact on MBO; this mostly reflects the decomposition of PAN in the subsiding air mass as discussed above rather than North American emissions. The North American contribution also shows a weak secondary peak at 20 days that reflects ozone produced in the United States and transported in the westerly atmospheric circulation.

[14] We find that ozone production over Asia begins to impact MBO after a 6-day time lag and that maximum Asian influence for the two events is at time lags of 8–11 days. This is consistent with previous studies showing that Asian pollution plumes can be transported across the Pacific in 5–10 days [*Yienger et al.*, 2000; *Stohl et al.*, 2002]. We related these time lags to observed cold front passages over eastern Asia on April 21 and May 3, lifting Asian pollution in warm conveyor belts (WCBs) that enables rapid transport across the Pacific [*Liu et al.*, 2003]. The Asian sensitivity spectra also show a long tail, similar to the North American spectra and indicating the impact on background ozone in addition to direct transport.

[15] The May 12 event at TH (Figure 2b, top) shows similar source attribution as the May 10 event at MBO and

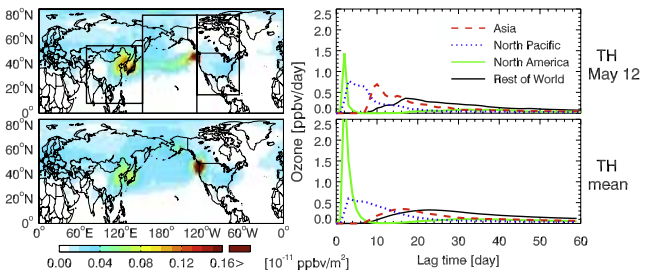

Figure 2b. Same as Figure 2a, but for Trinidad Head, California (TH, sea level). The event is for May 12, 2006, at 15 UT.

can be interpreted as subsidence of the free tropospheric plume that affected MBO on May 6–11. The Asian influence is much weaker at TH. Its sensitivity spectrum peaks at a time lag of 10 days, 2 day after that at MBO. Integrating the Asian sensitivity spectra for the events at MBO and TH over time lags of 5–20 days (direct transport component as opposed to background), we find a factor of 3 dilution effect as the plume mixes down to the surface (15 ppbv at MBO vs. 5 ppbv at TH). *Hudman et al.* [2004] previously estimated a factor of 10 dilution between the free troposphere and surface air for Asian dust plumes observed over the western United States.

4.2. Mean Conditions at MBO and TH

[16] Figure 2a (bottom) shows the source attribution for the mean ozone concentration at MBO during the INTEX-B period. The patterns are similar to the Asian pollution events previously discussed but the influence of direct Asian transport is weaker. Background production is mainly north of 20°N. Fine structure in the contributions from source regions in Asia can still clearly be distinguished, with maximum contributions from eastern China (5 ppbv) and Japan (1 ppbv) as derived by summing the corresponding grid squares. The Japanese contribution as identified from the adjoint model is mainly from boundary layer production and hence associated with local anthropogenic emissions. Its contribution to transpacific pollution to the U.S. is higher than would be expected from its NO_x emissions (0.7 Tg N a^{-1}) relative to China (6.4 Tg N a^{-1}) [*Zhang et al.*, 2009]. Export of Japanese pollutants into the westerly flow of the North Pacific is more efficient than for China [*Wild et al.*, 2004]. The mean ozone concentration at MBO is also sensitive to sustained production over the North Pacific from Asian pollution at 25°N–40°N, and particularly off the North American west coast, as previously discussed by *Zhang et al.* [2008].

[17] Figure 2b shows the sensitivity of the mean ozone concentration at Trinidad Head to production upwind. North American production is more important than at MBO. Asian influence is weaker than at MBO but still shows the Eastern China – southern Japan dipole. Asian ozone by summing the sensitivities over Asia is 8 ppbv at TH, consistent with studies using source-oriented methods [*Jaeglé et al.*, 2003; *Goldstein et al.*, 2004; *Zhang et al.*, 2008]. The peak in Asian influence is at a time lag of 16 days, as compared to 12 days at MBO, reflecting the delay and dilution during entrainment from the free troposphere to the surface. The mean transport time from Asia, calculated as the sensitivity-weighted mean time lag [*Vukićević and Hess*, 2000], is 23 days for MBO and 27 days for TH, comparable to the mean transport time of 2–3 weeks from East Asia to the western North America surface previously estimated by *Liu and Mauzerall* [2005].

[18] In summary, we have shown that an adjoint model analysis can provide detailed geographical and temporal information on intercontinental pollution influences at specific receptor sites. Such information can be used to better determine the sources of this intercontinental pollution, down to the scale of individual source countries and urban areas. For policy purposes it will be important to attribute intercontinental ozone pollution to the actual emissions of ozone precursors, in particular NO_x, taking advantage of the fine resolution enabled by the adjoint. This requires an adjoint of the model chemical mechanism to resolve the non-linearity on ozone production and hence a more elaborate calculation than was presented here.

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