External influences on ozone air quality: Intercontinental transport, changing climate, and stratospheric exchange

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Numerous collectors of PAN, NOy and O3 measurements
GFDL Atmospheric Model Development Team, TF HTAP Modeling Team

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The U.S. ozone smog problem is spatially widespread, affecting ~120 million people [U.S. EPA, 2010].

4th highest daily max 8-hr O₃ in 2008

Counts with monitors violating newly proposed primary O₃ standard in range of 0.060-.070 ppm (2006-2008 data)

http://www.epa.gov/air/airtrends/2010/

http://www.epa.gov/air/ozonepollution/pdfs/20100104maps.pdf
Newly proposed standards exceeded in spring in west (and SE) U.S. at “background” sites

Number of days with daily max 8-hr $O_3 > 60$ ppb

SPRING

SUMMER

2007-2009 averages using hourly $O_3$ from CASTNet database (www.epa.gov/castnet/data.html)

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Brief overview of tropospheric ozone and external influences on air quality

1. INTERCONTINENTAL TRANSPORT
   - NMVOCs: CO, CH₄
   - OH + NOₓ
   - Atmospheric cleanser

2. WARMING CLIMATE
   - T↑
   - PAN: (CH₃C(O)OOONO₂)
   - Ozone

3. STRATOSPHERE
   - Lightning
   - "Background" ozone

Human activity
Fires
Land biosphere
Continent
Ocean

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Evidence of intercontinental transport at northern midlatitudes: 2001 Asian dust event

Dust leaving Asian coast, April 2001

Reduced Visibility from Asian Dust over Glen Canyon, Arizona, USA

Intercontinental ozone transport is difficult (impossible?) to observe directly at surface [e.g., Derwent et al., 2003; Fiore et al., 2003; Goldstein et al., 2004; Jonson et al., 2005]

Estimates rely on models
Wide range in prior estimates of intercontinental surface ozone source-receptor (S-R) relationships

Assessment hindered by different (1) methods, (2) reported metrics, (3) meteorological years, (4) regional definitions

Few studies examined all seasons

Studies in TF HTAP [2007] + Holloway et al., 2008; Duncan et al., 2008; Lin et al., 2008

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Task Force on Hemispheric Transport of Air Pollution (TF HTAP): Multi-model studies to quantify & assess uncertainties in N. mid-latitude source-receptor relationships to inform CLRTAP

BASE SIMULATION (21 models):
- horizontal resolution of 5 \times 5^\circ or finer
- 2001 meteorology
- each group’s best estimate for 2001 emissions
- methane set to 1760 ppb

SENSITIVITY SIMULATIONS (13-18 models):
- -20% regional anthrop. NO_x, CO, NMVOC emissions, individually + all together (=16 simulations)
- -20% global methane (to 1408 ppb)

TF HTAP, 2007, 2010; Sanderson et al., GRL, 2008; Shindell et al., ACP, 2008; Fiore et al., JGR, 2009, Reidmiller et al. ACP, 2009; Casper Anenberg et al., ES&T, 2009; Jonson et al., ACPD, 2010
Large inter-model range; multi-model mean generally captures observed monthly mean surface $O_3$

- Many models biased low at altitude, high over EUS+Japan in summer
- Good springtime/late fall simulation
Model ensemble annual mean decrease in surface O\textsubscript{3} from 20% reductions of regional anthrop. O\textsubscript{3} precursors

Fiore et al., JGR, 2009

Spatial variability over continental-scale receptor region (NA) (see also Reidmiller et al, 2009; Lin et al., 2010)

\[ \Sigma 3 \text{ foreign} = 0.45 \text{ ppbv} \]

“import sensitivity”

Fiore et al., JGR, 2009
Seasonality of surface ozone response over North American and Europe to -20% foreign anthrop. emissions

1. Spring max due to longer O₃ lifetime, efficient transport [e.g., Wang et al., 1998; Wild and Akimoto, 2001; Stohl et al., 2002; TF HTAP 2007]

2. Response typically smallest to SA emissions (robust across models)

3. Similar response to EU & EA emissions over NA Apr-Nov (varies by model)

4. NA>EA>SA over EU (robust across models)

Fiore et al., JGR, 2009
Monthly mean import sensitivities (surface $O_3$ response to foreign vs. domestic emissions)

- SA fairly constant ~0.5
- 1.1 (EA), 0.7 (EU) during month with max response to foreign emissions
- 0.2-0.3 during month of max response to domestic emissions

Fiore et al., JGR, 2009
Models differ in estimates of surface $O_3$ response to foreign emission changes... which are best?

O$_3$ decrease from -20% foreign emissions

No obvious relationship btw “base-case bias” and magnitude of source-receptor relationship (e.g., NA$\rightarrow$EU)

$y = 0.0041x + 0.2954$

$R^2 = 0.0929$
Models likely differ in export of $O_3 +$ precursors, downwind chemistry (PAN [Emmerson and Evans, ACP, 2009]), and transport to receptor region.

$\text{NO}_x + \text{VOC} \rightarrow \text{PAN, } O_3$

$\text{CH}_3\text{C(O)OONO}_2$

$\text{NO}_x \rightarrow O_3$

$\text{N}_2\text{O}_5$

$\text{HNO}_3$

$\text{O}_3$

$\text{NO}_y$ partitioning (e.g., PAN vs. HNO$_3$) influences $O_3$ formation potential far from source region.

Observational evidence for $O_3$ production following PAN decomposition in subsiding Asian plumes [e.g., Heald et al., JGR, 2003; Hudman et al., JGR, 2004; Zhang et al., 2008; Fischer et al., 2010]

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Compare models with PAN measurements at high elevation sites in Europe (3 sites) and WUS (2 sites)

c/o Emily Fischer, U Washington

Mount Bachelor (2.76 km)
Jungfraujoch (3.58 km)

1. No observations from 2001; difficult to constrain models
2. How different are VOC inventories in the models?
3. What do the models say about the relative roles of regional anthrop. VOC emissions at measurement sites?

Model differences in lightning NOx affect PAN + impact of anthro. emis. [Fang et al., in review] in addition to anthrop. sources, chemistry, and transport
Strong sensitivity of exported EU \( \text{O}_3 \) to large spread in EU NMVOC inventories (anthrop. \( \text{NO}_x \) fairly similar across models)

Do the relative contributions from different source regions in the model correlate with NMVOC emissions?

\( R^2 = 0.5035 \)

Fiore et al., JGR, 2009
Relative influence of regional O$_3$ precursors on PAN at Mount Bachelor (OR), as estimated by the HTAP models

4 example HTAP models sampled at Mount Bachelor

Fraction of total PAN from source region
EU NA EA SA

More EA influence

More EU influence

Mount Bachelor, April: $r^2=0.85$

EA/EU PAN influence correlates with EA/EU AVOC emissions

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Model differences in relative contributions of source regions to PAN at Jungfraujoch, Switzerland

4 example HTAP models sampled at Jungfraujoch

- Fraction of total PAN from source region:
  - EU
  - NA
  - EA
  - SA

- More EU influence
- More NA influence

Jungfraujoch, APRIL: $r^2=0.40$

Wide range of EU NMVOC inventory contributes to model discrepancies

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Major uncertainties from sub-grid processes: Deep convection at the leading edge of the convergence band and associated pollutant export missing in global model [Lin et al., ACP, in press]

How well do models represent mixing of free trop. air to the surface?
Summary of external influence on air quality #1: Hemispheric Transport of O$_3$

- Benchmark for future: Robust estimates + key areas of uncertainty
- “Import Sensitivities” (\(\Delta O_3\) from anthrop. emis. in the 3 foreign vs. domestic regions): 0.5-1.1 during month of max response to foreign emis; 0.2-0.3 during month of max response to domestic emissions
- Variability of O$_3$ response to emission changes within large HTAP regions
- Potential for PAN, NOy, other species, to help constrain model O$_3$ response to emission changes
- Role of “missing” processes (e.g., mesoscale)
External influence on air quality #2: Changing climate

Observations c/o Jenise Swall and Steve Howard, U.S. EPA

Strong relationship between weather and pollution implies that changes in climate will influence air quality

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Pollution build-up during 2003 European heatwave

CO and O₃ from airborne observations (MOZAIC)

Above Frankfurt (850 hPa; ~160 vertical profiles)

Stagnant high pressure system over Europe
(500 hPa geopotential anomaly relative to 1979-1995 for 2-14 August, NCEP)

Ventilation
(low-pressure system)

Carlos Ordóñez, Toulouse, France Contribution to GEMS
GEMS-GRG, subproject coordinated by Martin Schultz
Model estimates of climate change impact on U.S. surface ozone [Weaver et al., BAMS, 2009]

ROBUST FINDINGS:
1. Increased summer O$_3$ (2-8 ppb) over large U.S. regions
2. Increases are largest during peak pollution events

KEY UNCERTAINTIES:
1. Regional patterns of change in meteorological drivers
2. Isoprene emissions and oxidation chemistry
3. Climate signal vs. interannual variability
4. Future trajectory of anthrop. emissions (not shown here)
Summertime surface $O_3$ changes in a warmer climate in the new GFDL chemistry-climate model (AM3)

20-year simulations with annually-invariant emissions of ozone and aerosol precursors

**Present Day Simulation** ("1990s"): observed SSTs + sea ice (1981-2000 mean)

**Future Simulation** ("A1B 2090s"): observed SSTs + sea ice + average 2081-2100 changes from 19 IPCC AR-4 models

Previously noted degradation of summertime EUS $O_3$ air quality e.g., reviews of Jacob and Winner, Atmos. Environ. 2009 and Weaver et al., BAMS, 2009

Previously noted decrease of lower troposphere background $O_3$ e.g., Johnson et al., GRL, 2001; Stevenson et al., JGR, 2006

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Preliminary future climate simulations suggest more days with $O_3 > 60$ ppb in western US in spring.

FUTURE: ▲ individual years — 20-yr mean (climate change only)

PRESENT: ● individual years — 20-yr mean

Increase in background? Strat. $O_3$?

More local destruction or smaller imported background?

How does “background” ozone in WUS respond to climate change?

Warmer climate may increase strat-to-trop influence at northern mid-latitudes [e.g., Collins et al., 2003; Zeng and Pyle, 2003; Hegglin and Shepherd, 2009; Li et al., 2009]

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External influence #3: Stratospheric O$_3$ in surface air…
Uncertain and controversial

Models differ in approaches and estimates for strat. O$_3$ in surface air [e.g., Roelofs and Lelieveld, 1997; Wang et al., 1998; Emmons et al., 2003; Lamarque et al., 2005]

Hess and Lamarque, JGR, 2007: 2 approaches to estimate O$_3$-strat contribution in surface air

VIEW #1: Fiore et al., JGR, 2003

Mar-Oct 2001 U.S. daily mean afternoon surface O$_3$

Model
Observed

1. Prior interpretation strat. source in WUS [Lefohn et al., 2001]) underestimated role of regional (+hemispheric) pollution
2. Direct strat. intrusions to surface are rare and should not compromise O$_3$ standard attainment (GEOS-Chem model year 2001)

VIEW #2: Langford et al., GRL, 2009

Observations of direct strat. influence on surface O$_3$ at Front Range of CO Rocky Mtns in 1999; could lead to O$_3$ standard exceedances

→Important to determine frequency of direct strat intrusions
→Focus in next few slides on variability

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Observations suggest influence of stratosphere on lower tropospheric ozone over Europe in winter-spring

$r = 0.77$ Jungfraujoch vs. ozonesonde

*Ordóñez et al., GRL, 2007*
MOZART-2 anomalies in O$_3$ and in strat. O$_3$ tracer are correlated in winter-early spring at Jungfraujoch

Only meteorology (and lightning NO$_x$) vary in this simulation [Fiore et al., 2006]
Model indicates a role for stratospheric influence on interannual variability in O₃ at WUS sites in March

- Observations
- Model (MOZART-2) total O₃
- Model (MOZART-2) stratospheric O₃ tracer

Can we exploit times when stratospheric influence dominates observed variability to determine “indicators” of enhanced strat O₃ contribution?
Potential for developing space-based “indicator” for day-to-day variability in strat $O_3$ influence at WUS sites?

OMI/MLS products: trop. column $O_3$ (TCO) and strat. column $O_3$ (SCO) [Ziemke et al., 2006].

Correlate daily anomalies in March in TCO and SCO with those at Mesa Verde CASTNET site, with ground site lagged (8 day lag shown below)

Signal of strat influence in upwind trop for WUS? or simply free trop signal?

“strat source region” for WUS?

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Concluding thoughts…
External influences on surface $O_3$ at N. mid-latitudes

- Intercontinental transport occurs year-round; peaks in spring
  → Uncertainties in spatiotemporal variability, role of sub-grid processes
  → Need better constraints on response to emissions perturbations

- Warming climate expected to degrade air quality in polluted regions
  → Uncertainties in regional climate response and isoprene-$NO_x$ chemistry
  → Competing influences on tropospheric background ozone

- Stratospheric $O_3$ influence peaks in early spring, at high altitude sites
  → Uncertainties in contribution to surface air and variability
  → Need better process understanding on daily to decadal time scales

- Implications for attaining ever-tightening air quality standards
- Potential insights from long-term \textit{in situ} obs, satellite, models into role of meteorology/ climate versus emissions on observed variability and trends

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