Challenges in quantifying intercontinental pollution influences on surface ozone in the western US

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with Lu Hu, Johan Schmidt, and former students/postdocs Lin Zhang, Rynda Hudman, Colette Heald, Yevgenii Rastigejev, Jingqiu Mao, Emily Fischer, Peter Zoogman
Back to my San Joaquin Valley days...

The $\text{H}_2\text{SO}_4$-$\text{HNO}_3$-$\text{NH}_3$ System at High Humidities and in Fogs

1. Spatial and Temporal Patterns in the San Joaquin Valley of California

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Tule fog seen from above

We fight acid fog!
Emerging interest in intercontinental ozone pollution in the late 1990s driven by

- 1997 change in NAAQS from 0.12 to 0.08 ppm
- Rapidly rising Asian emissions
NO$_x$ emissions observed from space

http://disc.sci.gsfc.nasa.gov/giovanni
NO$_x$ emission trends observed from space

OMI NO$_2$ trend
2005-2010

fractional change

Verstraeten et al. [2014]
Successive versions of the GEOS-Chem global model used over the years to simulate intercontinental ozone pollution.


from European anthropogenic emissions

from Asian anthropogenic emissions

Lin Zhang, Harvard
Intercontinental ozone pollution in western US: what models tell us

- Mechanism involves transport in free troposphere and subsidence
  - Free troposphere has strong westerly winds, long ozone lifetime

- Influence is largest in March-May
  - That’s when transport and chemistry are most favorable

- Influence is largest in Intermountain West
  - High elevation, arid terrain, deep PBL mixing

- MDA8 ozone increases in spring by ~10 ppb (surface), ~15 ppb (elevated sites)
  - Half is due to methane, half to NO$_x$ from other continents
  - NO$_x$ influence is mainly from Asia, and is ~half through PAN
  - 2xAsian emissions increases ozone by 1-2 ppb (surface), 2-4 ppb (elevated)
  - Canada/Mexico influence adds 1-5 ppb

- Influence is mainly manifested by elevated background
  - Relatively little day-to-day variability, no large “intercontinental pollution events”

But how do we test the models?
Conceptual picture of ozone production in transpacific Asian pollution plumes

GEOS-Chem ozone production efficiency (OPE) per unit NO\textsubscript{x}
In May 2002 at 2-4 km altitude

Hudman et al. [2004]

5-10% export of NO\textsubscript{y} mainly as PAN

Subsidence over E Pacific:
PAN $\rightarrow$ NO\textsubscript{x}$\rightarrow$HNO\textsubscript{3}
OPE 60-80
strong $\Delta$O\textsubscript{3}

U.S. boundary layer

Asian boundary layer
(OPE $\sim$ 5)

E. Asia Pacific United States
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Simulation of ozone at remote western US sites

NA background ≡ simulation with no anthropogenic sources in N America

- Most ozone is from non US sources
- Intercontinental pollution contributes ~15 ppb with little day-to-day variability
- Model reproduces timing but not magnitude of stratospheric intrusion spike

Zhang et al. [2014]
Ozone from stratospheric intrusions is not all natural

The “produced” definition is correct for diagnosing natural stratospheric ozone, the “transported” definition is correct for diagnosing intrusions

~ 50% of ozone in stratospheric intrusions originates from the troposphere

Model inability to capture intrusion structure is due to numerical diffusion

Zhang et al., [2014]
Asian pollution ozone at Mt. Bachelor, Oregon

No observed Asian events in 2006… but an event in 2008

Asian pollution ozone = 9 ± 2 ppb

Asian pollution events are occasionally observed…but are less spectacular than stratospheric events

Zhang et al. [2008], Ambrose et al. [2011]
Remote sea-level sites show less ozone variability than mountain sites

Reflects 3-10x dilution during entrainment of free tropospheric air

Mt. Bachelor Observatory
(122W, 44N, 2.7km)

Trinidad Head (124.2W, 41N, 107m)

Observed (2006)  GEOS-Chem

Subsiding conditions are also conducive to fast PBL ozone production – elevated ozone is not necessarily background

Zhang et al. [2009]
Large Asian ozone plumes are seen from aircraft… but GEOS-Chem cannot reproduce them

Observations of Asian plumes arriving over California

**MAY 5**

- CO
- O$_3$
- PAN
- HNO$_3$
- NO$_x$

**MAY 5** plume at 6 km: High CO and PAN, no O$_3$ enhancement

**MAY 17**

- CO
- O$_3$
- PAN
- HNO$_3$
- NO$_x$

**MAY 17** subsiding plume at 2.5 km: High CO and O$_3$, PAN → NO$_x$ → HNO$_3$

Hudman et al. [2004]
Layered structure of transpacific Asian ozone pollution

TRACE-P flights, 26-27 Feb 2001

Lidar ozone profiles below the aircraft:

Think of transpacific Asian pollution plumes as free tropospheric layers ~1-km thick, stretching over 1000s of km in horizontal, 1-2 weeks old

Heald et al. [2003]
Eulerian models cannot preserve intercontinental layers

2-D pure advection $\frac{\partial C}{\partial t} = -u \nabla C$ of inert Asian plume at 4 km altitude

- Advection equation should conserve mixing ratio
- It does in uniform flow but not in divergent/shear flow
- Increasing resolution yields only marginal improvement

Need more creative approaches: embedded Lagrangian plumes, adaptive grids

Rastigejev et al. [2010]
Global simulation of tropospheric ozone: a moving target

In Zhang et al. [2011, 2014] GEOS-Chem was near-perfect! But alas, since...

Add Br chemistry; ozone decreases by 5-10 ppb

Add new HO\textsubscript{x} aerosol chemistry: ozone decreases by 0-10 ppb

Improve isoprene chemistry: ozone increases by 5-10 ppb

Update meteorological fields: all hell breaks loose

Mao et al. 2013ab; Johan Schmidt (in prep.)
Lu Hu (in prep.)

Trinidad Head
Most recent GEOS-Chem version (10.01, April 2015 release): our best simulation of global tropospheric ozone so far

Global comparison to 500 hPa ozonesonde data

Mean bias ± StdDev

Each point represents monthly mean $O_3$ at 500 hPa for a specific site

Lu Hu (in prep.)
Most recent GEOS-Chem version (10.01, April 2015 release): our best simulation of global tropospheric ozone so far

Comparison to bias-corrected OMI satellite data at 400-700 hPa for 2011-2013

Lu Hu (in prep.)
PAN is a major vehicle for intercontinental ozone pollution
...but successful simulation is a big challenge

PAN at 30-60N in spring
GEOS-Chem: background
Aircraft data: circles

Global contributions to PAN
In GEOS-Chem: complex VOC emissions/chemistry

Fischer et al. [2013]
Inability of models to simulate multi-decadal ozone trends
Long-term records at European mountain sites

1870-1990: Marenco et al. [1994]

Preindustrial ozone models

1870-1990: Marenco et al. [1994]

Model error is likely in
- Natural sources and trends: lightning NOx, isoprene, fires…
- Chemistry: halogens, HOx, HONO, PAN, low-NOx regime…
Looking ahead: geostationary observation of ozone air quality

Geostationary constellation launch in 2018-19

TEMPO       Sentinel-4       GEMS

TEMPO will include first ozone observation in the weak visible Chappuis bands:

UV       IR       Vis

~3 km

air scattering       thermal contrast       surface

this will provide observation sensitivity down to the surface

Natraj et al. [2011]
Observation system simulation experiment (OSSE) to test utility of TEMPO for monitoring/interpreting ozone events in western US

Use a CTM to produce
A virtual atmosphere

Sample this virtual atmosphere on TEMPO observing schedule

Use TEMPO averaging kernel matrix to simulate what TEMPO would see

TEMPO synthetic ozone data
Assimilate TEMPO in monitoring system for surface ozone

“Truth” = AM3-Chem model

Forecast model: GEOS-Chem

TEMPO

CASTNet synthetic data

Data assimilation (Kalman filter)

Assimilated 3-D ozone field on forecast model grid

Zoogman et al. (2014)
Utility of TEMPO for monitoring high-ozone days

OSSE for April-June 2010

Number of days with MDA8 ozone > 70 ppbv

“Truth”  GEOS-Chem (no assimilation)

with surface data  with surface + TEMPO data

Zoogman et al. (2014)
Utility of TEMPO to diagnose stratospheric intrusions

Stratospheric ozone intrusion over New Mexico, 13 June 2010

Zoogman et al. (2014)
Why do we have this problem?

- As plume stretches to \( W \sim \Delta x \), advection algorithm becomes highly diffusive.
- Finer grid resolution only delays the problem; improvement \( \sim \Delta x^{-1/2} \).

Improving resolution of free tropospheric layers by a factor 2

\[ \Rightarrow \text{requires increasing grid resolution by factor of 4} \]
\[ \Rightarrow \text{increases computer time by a factor } \sim 100 \]

Need more creative solutions: embedded Lagrangian plume, adaptive grid.

Rastigejev et al. [2010]