Integrated analysis of the impact of long-range transport of midlatitude pollution on ozone abundances in the Arctic troposphere

T. Walker, M. Parrington, D. B. A. Jones
University of Toronto

D. K. Henze
University of Colorado at Boulder

J. R. Worden, K. W. Bowman
NASA Jet Propulsion Lab

J. Bottenheim, K. Anlauf, J. Davies, D. W. Tarasick
Environment Canada

A. M. Thompson
Pennsylvania State University

Thomas Walker
University of Toronto

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O$_3$ is not emitted directly
Produced through hydrocarbon oxidation in the presence of NO$_x$ (NO + NO$_2$)
Atmospheric lifetime (O$_3$): ~3 weeks
Atmospheric lifetime (NO$_x$): ~1-2 days

Ozone in the Arctic reflects either direct transport of ozone from lower latitudes and the stratosphere or in situ production (driven by NO$_x$ transported into the region as PAN)
Model Description and Data Sources

**GEOS-Chem**: global chemical transport model
- NOₓ-HOₓ-VOC chemistry
- dynamics driven by GEOS meteorological data
- results shown here are from v7-02-04, resolution 4°x5° horizontal, 30 vertical levels
- tagged-Oₓ simulation permits regional source attribution
- adjoint permits detailed sensitivity analysis

**Tropospheric Emission Spectrometer (TES)**:
- high resolution IR Fourier transform spectrometer
- 3.2 – 15.4 μm spectral range
- O₃ profiles assimilated into GEOS-Chem using suboptimal Kalman filter between 60oS - 60oN

**Ozonesondes**:
- vertical profiles of ozone, temperature, pressure
- launched roughly weekly, intense campaigns in spring

**Surface measurements at Alert**:
- hourly average O₃
- measurements of PAN every half hour
Monthly mean O$_3$ profiles above Eureka

- high ozone values in free troposphere (eg. ~500 hPa) appear in model earlier than observed
- the model significantly underestimates ozone abundances in the middle and upper troposphere in summer
Model Simulation of Surface $\text{O}_3$:
Agreement with Surface Data

- model reproduces seasonal cycle in surface ozone at Alert (2001)
- highest values in spring; lowest in summer
- slight overestimate in early spring; slight underestimate in fall
- depletion events due to halogen radical chemistry not represented in model
Model Simulation of Surface PAN: Agreement with Surface Data

- model reproduces seasonal cycle in surface PAN at Alert (2001)
- accumulation during winter leading to spring maximum; sharp drop to very low values in summer
- slight underestimate in spring
- seasonal cycle is apparent across the region, not just at Alert
Fractional $O_3$ Contribution from Midlatitude Source Regions

seasonal and vertical distributions of the fractional contribution to ozone profiles above Eureka (2005)

- in summer the dominant midlatitude contributions are from North America and Siberia
Satellite constraints on ozone transport into the Arctic

TES $O_3$ profiles are assimilated into GEOS-Chem equatorward of 60° between July 1 – Aug 31, 2006

- Parrington et al. [JGR, 2008] showed that over North America, the assimilation reduced the mean differences between the model and ozonesondes to less than 5% in the middle troposphere.
- Assimilation of TES $O_3$ increases concentrations across Northern mid-latitudes, with largest enhancements over Siberia and North America.
- The assimilation provides an improved description of tropospheric $O_3$ at midlatitudes and thus an improved boundary condition for $O_3$ transport into the Arctic.
Assimilation of O$_3$ at midlatitudes corrects high latitude bias

- assimilation of TES O$_3$ results in significant correction to free troposphere ozone at high latitude sites
- mean bias between 400-500 hPa is reduced from +7.1 to -1.9 ppbv at Eureka, and from +11.5 to +1.9 ppbv at Ny Alesund
- little change to boundary layer
Ozone fluxes into the Arctic

- monthly mean flux during July 2006
- change to flux due to assimilation is concentrated in the upper troposphere and lower stratosphere
- monthly mean net northward flux across 60°N changes from 1900 to 9700 kg/s with the assimilation
- net downward flux across 300 hPa north of 60°N changes from 320 to 760 kg/s with the assimilation (not shown)
Summary of Results

- model reproduces surface ozone and PAN seasonal cycles, but underestimates free tropospheric ozone in the summer
- midlatitude continental source regions do contribute significant fractions of simulated ozone at Eureka
- assimilation of ozone data from TES into GEOS-Chem equatorward of 60°N enhances midlatitude ozone abundances in the model, particularly over North America and Siberia, which reflects, in part, stratosphere-troposphere exchange captured by the TES measurements.
- improved midlatitude boundary conditions in the assimilation results in increase transport of ozone into the Arctic, mainly in the UTLS; the mean bias in ozone relative to ozonesondes decrease from +9.2 to -0.1 ppbv between 400-500 hPa over Eureka and Ny Alesund
- assimilation increases net horizontal flux across 60°N from 1900 to 9700 kg/s, and increases net downward flux in the Arctic from 320 to 760 kg/s
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EXTRAS
Source regions for tagged $O_x$ simulation
Why Tropospheric Ozone in the Arctic?

1. **RADIATIVE IMPACT**
   - direct greenhouse gas
   - affects atmospheric lifetime of other greenhouse gases
   - expected to increase

2. **SENSITIVE ENVIRONMENT**
   - largest temperature trends are observed at high latitudes

3. **AIR QUALITY**
   - constituent of photochemical smog and of “Arctic haze”

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**GLOBAL MEAN RADIATIVE FORCINGS**

- RF Terms:
  - Long-lived greenhouse gases
  - Nitrous oxide ($N_2O$)
  - Methane ($CH_4$)
  - Halocarbons

- RF values (W m$^{-2}$):
  - $CO_2$: 1.66 [1.49 to 1.83]
  - $N_2O$: 0.48 [0.43 to 0.53]
  - $CH_4$: 0.16 [0.14 to 0.18]
  - Halocarbons: 0.34 [0.31 to 0.37]

- Spatial scale:
  - Continental
  - Global

- LOSU:
  - Med - Low

**GLOBAL TEMPERATURE TRENDS**

- Surface
- Troposphere

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from IPCC AR4, 2007
Ozone Production

\[
\begin{align*}
\text{RH} + \text{OH} & \rightarrow^{(02)} \text{RO}_2 + \text{H}_2\text{O} & \text{(R1)} \\
\text{RO}_2 + \text{NO} & \rightarrow \text{RO} + \text{NO}_2 & \text{(R2)} \\
\text{RO} + \text{O}_2 & \rightarrow \text{R'CHO} + \text{HO}_2 & \text{(R3)} \\
\text{HO}_2 + \text{NO} & \rightarrow \text{OH} + \text{NO}_2 & \text{(R4)} \\
2(\text{NO}_2 + h\nu) & \rightarrow^{(02)} \text{NO} + \text{O}_3 & \text{(R5)} \\
\text{RH} + 4\text{O}_2 & \rightarrow \text{R'CHO} + 2\text{O}_3 + \text{H}_2\text{O} & \\
\end{align*}
\]

**hydrocarbon-limited case:**
termination reaction
\[
\text{HO}_2 + \text{HO}_2 \rightarrow \text{H}_2\text{O}_2 + \text{O}_2 \quad \text{(R6)}
\]
yielding
\[
\text{P(O}_3\text{)} = 2k_4 \left\{ \frac{\text{P(HO}_x\text{)}}{k_6} \right\}^{1/2} [\text{NO}]
\]

**NO\text{\textsubscript{x}}-limited case:**
termination reaction
\[
\text{HO}_2 + \text{NO} + \text{M} \rightarrow \text{HNO}_3 + \text{M} \quad \text{(R7)}
\]
yielding
\[
\text{P(O}_3\text{)} = \frac{2k_4 \text{P(HO}_x\text{)}}{k_6 [\text{NO}_2] [\text{M}]}
\]
Adjoint model

Consider a linear expansion about a background set of inputs $x_o$:

$$ y = M(x) = M(x_o) + \frac{\partial M}{\partial x}(x - x_o) $$

where $\frac{\partial M}{\partial x}$ is the tangent linear model (TLM).

Given a small perturbation $\Delta x$, the TLM can calculate the perturbation to the state $\Delta y$:

$$ \Delta y = TLM(\Delta x) $$

Likewise, given the perturbed state, the adjoint model (transpose of TLM) calculates the required change in inputs

$$ \Delta x = TLM^T(\Delta y) $$

Example: $\Delta y$ could be the difference between simulated and observed ozone and $\Delta x$ could be the required change in NO$_x$ emissions to obtain that perturbed state.