Factors controlling global tropospheric ozone: Roles of isoprene chemistry, tropospheric halogen chemistry, deep convection, and lightning NO$_x$ sources

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With
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Global simulation of tropospheric ozone is a moving target

Global tropospheric ozone burden in GEOS-Chem benchmark outputs

- Update isoprene chemistry (+5%)
- Add Br chemistry (-5%)
- Update lightning NO\textsubscript{x} (+8%)

Model release date

ACCMIP 15 model mean (Young et al., 2013)
Introducing the new generation of meteorological fields generally reduces free tropospheric $O_3$ (up to 15 ppbv)

Difference of simulated ozone between **NEW** (GEOS-FP) and **OLD** (GEOS-5) meteorological fields

New meteorological field (since June 2012):
• Higher temporal and horizontal resolution
• Much weaker convection
• Lower temperature $\rightarrow$ lower global isoprene emission (by 20%)
Do recent updates improve our ability to simulate tropospheric ozone?

**Approach:**
Evaluate GEOS-Chem with the observations for global tropospheric ozone, and diagnose model weaknesses, using:

- Ozonesondes $\rightarrow$ vertical gradient
- Civil aircraft $\rightarrow$ upper troposphere
- Satellite data $\rightarrow$ global distribution

Evaluating global $O_3$ simulation with sondes

Ozonesonde launch at the South Pole

Credit: NOAA

Data sources: WOUDC, NOAA GMD for year 2013; total 1849 sondes (40 sites)
Spring: widespread model low biases in northern mid-latitude

3-7 km (400-700 hPa)

Model bias (%)
Spring: widespread model low biases in northern mid-latitude
3-7 km (400-700 hPa)

Summer: obviously high biases in the U.S.; low biases in Japan and polar regions
OMI ozone data quality is not significantly degraded from 2008 (not shown).

OMI ozone data from Xiong Liu (CFA, Harvard-Smithsonian)

OMI O$_3$ data was corrected for biases (3.6 ppbv) relative to in situ ozonesonde data following the method by Zhang et al. [2010]
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Improved simulation of global tropospheric ozone compared to previous versions

**Model bias (GC-OMI)**

- **MAM**: Mean bias -1.5 (7.8)
- **JJA**: Mean bias -0.1 (4.3)
- **SON**: Mean bias 0.1 (4.6)
- **DJF**: Mean bias -0.3 (6.9)

**Hu et al., [in prep.]:** v10-01, 2x2.5, GEOS-FP, 2013, updated isoprene chemistry, with Br, ‘Linoz’

**Model bias (Old GC-OMI)**

**Zhang et al., [2010]:** v8-01-04, 4x5, GEOS-4, 2006, ‘old’ isoprene chemistry, no Br, ‘Synoz’
Improved ozone simulation is a result of advancing scientific knowledge of atmospheric processes controlling tropospheric ozone.

Chemical production of ozone is mostly sensitive to meteorological fields, lightning NO$_x$ source, and isoprene chemistry.
(New) tropospheric halogen chemistry mostly impacts high latitude $O_3$

Schmidt et al., JGR [in review]
Impact of isoprene chemistry on $O_3$ extends to free troposphere in the tropics (2-3 ppbv), where convection is strong.

‘Old’ isoprene chemistry:
- Dry deposition is the only loss pathway for isoprene nitrate
- Turning off ISOPOOH+OH$\rightarrow$IEPOX
- Turning off isomerisation and self-reaction pathways of ISOPO2
Impact of isoprene chemistry on $O_3$ extends to free troposphere in the tropics (2-3 ppbv), where convection is strong.

Impact of isoprene chemistry on $O_3$ in the Eastern U.S. is most significant in the boundary layer (5-8 ppbv).

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Eastern U.S. summer ozone overestimation is likely caused by NO$_x$ bias in the U.S. NEI emission inventory (SEAC$^4$RS)

- U.S. NEI NO$_x$ emissions are likely overestimated by a factor of two:
  - Both NEI2005 and NEI2011 have such a problem
  - This seems to be a problem since NEI1999 (Hudman et al., 2007)
- U.S. lightning NO$_x$ sources need to be reduced by half
Lightning NO$_x$ sources affect ozone formation: not only in tropics, but also in mid-high-latitude.

- Large uncertainties remain for the magnitude of lightning NO$_x$ sources in some regions (e.g., U.S., East Asia)
- More observational constraints are needed
Conclusions

• Tropospheric ozone simulation is mostly sensitive to model treatments in lightning NO$_x$ source and isoprene chemistry, and selection of meteorological inputs.

• GEOS-Chem has improved global tropospheric ozone simulation over the years, reflecting integrated improvements of the above controlling processes.

• GEOS-Chem underestimates tropospheric O$_3$ in the mid-high-latitudes (>50°) in winter and spring, likely due to adding Br chemistry and insufficient stratosphere-troposphere exchange.

• GEOS-Chem overestimates summer O$_3$ in the Eastern U.S., but this is caused by an overestimated U.S. NEI NO$_x$ emission. NO$_x$ emission is far more uncertain than current estimates.
Thanks!
Global comparison to OMI satellite data

Each point represents seasonal mean O₃ at 400-700hPa for a model grid (2x2.5)

Bias at northern mid-high-latITUDE partly due to satellite error

Bias-corrected OMI ozone [ppbv]
Well simulated ozone in the free troposphere (400-700 hPa)

Zonally averaged free tropospheric ozone

Bias at mid-high-latitude (>45 °) partly due to satellite error
Comparisons with sondes are generally consistent with what OMI tells us for the mid-troposphere.

Bias at mid-high-latitude (>45 °) partly due to satellite error.