Current research activities at Harvard
What GEOS-Chem aims to be:
1. An open-access, easy-to-use, state-of-science, well-supported atmospheric chemistry model for interpreting observations and advancing atmospheric chemistry knowledge;
2. An atmospheric chemistry module to serve Earth system models and data assimilation systems
GEOS-Chem development is driven by its user community

Model Support Team (Harvard, Dalhousie, U. Colorado)

Model version updates

Contribute developments, report bugs

Support

Model development

Set priorities

Implement priorities

Benchmark updates

GEOS-Chem Steering Committee

Model scientist: Jacob (Harvard)
   co-scientist: Martin (Dalhousie)
Adjoint model scientist: Henze (U. Colorado)
Nested model scientists: Wang (Tsinghua/UH), Zhang (PKU)
Engineer: Yantosca (Harvard)
Aerosols WG:
   Heald (MIT), Alexander (UW), Pierce (CSU), Yu (SUNYA)
Chemistry WG:
   Henderson (EPA), Evans (York), Mao (U. Alaska), Hu (U. Montana)
Emissions and Deposition WG:
   Lin (PKU), Fischer (CSU), Millet (U. Minnesota), Marais (U. Leicester)
Chemistry-Ecosystems- Climate WG:
   Liao (NIUST), Tai (CUHK), Murray (U. Rochester), Geddes (Boston U)
Carbon WG:
   Jones (U. Toronto), Bowman (JPL)
Adjoint and Data Assimilation WG:
   Wang (U. Iowa), Henze (U. Colorado)
Transport WG:
   Liu (NIA), Molod (NASA)
Hg and POPs WG:
   Holmes (FSU), Fisher (U. Wollongong)
GCHP WG: Martin (Dalhousie), Eastham (MIT)
GMAO Liaison: Keller (NASA)
At large: Kasibhatla (Duke)

Emphasize grass-roots development, fast innovation, traceability, user support
Input meteorological data
NASA GEOS fields, 1980-present
0.25°×0.3125° or 0.5°×0.625°, 72 vertical levels

GEOS-Chem solves 3-D chemical continuity equations on global or nested Eulerian grid

Modules
- emissions
- transport
- chemistry
- aerosols
- deposition

Applications
- Model adjoint
- Tropospheric and stratospheric chemistry+aerosols, aerosol microphysics, carbon gases, mercury, POPs, isotopes…
- New major version releases every year: version 12.0.0 (June 2018) includes updated chemistry for SOA, isoprene, halogens, mercury, new emissions…
- Grid-independent architecture for MPI and coupling to Earth System models, transparent to GEOS-Chem Classic users
Errors in off-line and coarser-grid transport simulations
Comparisons of on-line (GEOS-5 c360) and off-line (GEOS-Chem) $^{222}$Rn simulations

$^{222}$Rn at c360 (July 2013) mixing ratios, mBq SCM$^{-1}$

- off-line 0.25$^\circ$x0.3125$^\circ$
- vs. on-line c360:
- % difference

- off-line 2$^\circ$x2.5$^\circ$
- vs. 0.25$^\circ$x0.3125$^\circ$:
- % difference

How to cure?
- Re-diagnose convection off-line
- Replicate transport on cubed-sphere grid
- Facilitate use of high-resolution off-line model
- Facilitate use of on-line model

Yu et al., GMD 2018
GEOS-Chem high performance (GCHP):
Massively parallel off-line capability with cubed-sphere advection using distributed-memory MPI parallelization

1-month simulation of troposphere-stratospheric chemistry at c180 (~50 km) resolution requires only 1 day of wall time with 540 cores

Ozone at 4 km altitude, July 2016

Aerosol optical depth (AOD), July 2016

Eastham et al., GMD 2018
GEOS-Chem as on-line chemical module for Earth system models (ESMs)

any 3-D grid specified at run time

Off-line GEOS-Chem CTM

Advection

Mixing Convection

coupler

coupler

Chemistry (FlexChem):
\[ \frac{dC}{dt} = P - L - D \]

Emissions (HEMCO):
\[ \frac{dC}{dt} = E \]

coupler

coupler

GEOS-Chem chemical module
GEOS-Chem as on-line chemical module for Earth system models (ESMs)

any 3-D grid specified at run time

Chemistry (FlexChem): \( \frac{dC}{dt} = P - L - D \)

Emissions (HEMCO): \( \frac{dC}{dt} = E \)

GEOS-Chem chemical module

Earth System Model with GEOS-Chem chemistry

Dynamics, chemical transport

Coupler
GEOS-Chem as on-line chemical module for Earth system models (ESMs)

any 3-D grid specified at run time

Off-line GEOS-Chem CTM

Advection

Mixing

Convection

GEOS-Chem chemical module

Chemistry (FlexChem): \[ \frac{dC}{dt} = P - L - D \]

Emissions (HEMCO): \[ \frac{dC}{dt} = E \]

Earth System Model with GEOS-Chem chemistry

Dynamics, chemical transport

CTM and ESM use exactly the same GEOS-Chem code

GEOS-Chem CTM users contribute model advances

Advances are incorporated into GEOS-Chem

ESM GEOS-Chem module always stays referenced to latest version

Keller et al., GMD 2014; Long et al., GMD 2015
Full-year tropospheric chemistry simulation at c720 (~12 km) resolution using on-line GEOS-Chem in NASA GEOS-5 ESM

August 1, 2013

Hu et al., GMD 2018

GEOS-Chem has also been coupled to BCC ESM, WRF, NCAR CESM2

What about IFS?
- “80% done” by Seb Eastham, another 1-week visit would do it he says
Running GEOS-Chem on the cloud

GEOS-Chem Classic is now operational on the AWS cloud, GCHP is on the way.

Many advantages for access, reproducibility, sharing of model versions and data.
Inversion of 2010-2015 GOSAT methane data

Analytical inversion using GEOS-Chem forward model with joint Bayesian optimization of
- methane emissions (4°x5°)
- 2010-2015 trends (4°x5°)
- annual global OH concentration

Emission trends: tropical increases

OH trend: -2% contributes to methane increase

Maasakkers et al. [in prep]
New inventory of methane emissions from fuels

- EDGAR has large errors for oil/gas emissions
- EDGARv4.3.1 has no sectoral breakdown beyond “fuel” which includes oil+gas+coal

Our inventory:
1. Follows UNFCCC-reported national emission totals
2. Includes detailed subsectoral breakdown

Scarpelli et al., in prep.
Satellite measurements of methane as proxy for global OH

\[
\frac{dm_{CH4}}{dt} = E - k[OH] m_{CH4} + \text{minor terms}
\]

Distribution of tropospheric methane + OH loss rate (GEOS-Chem model)

Loss pattern has broad meridional and seasonal signatures, distinct from emission signatures in inversions of methane satellite data

Zhang et al., APCD 2018
Ability of satellite methane data to constrain OH and its trend

OSSE using satellite observations in the SWIR (TROPOMI) and TIR (CrIS)
In Maasakkers et al. inversion framework with ACCMIP OH distributions

- Combination of SWIR and TIR enables better separation of global emissions and OH
- Global emissions and OH concentrations, and their trend, can be separately retrieved

Zhang et al., ACPD 2018
Interpreting ozone pollution trends in China
Extensive network observations starting in 2013

Mean 2013-2017 anthropogenic trend after removing meteorological variability

Li et al., in prep.
Anthropogenic drivers of 2013-2017 trend in summer MDA8 ozone

Ozone increases are mostly due to PM decrease affecting HO$_2$ aerosol uptake

Li et al., in prep.
Free troposphere makes major contribution to OMI tropospheric NO$_2$

Mean NASA SEAC$^4$RS NO$_2$ and NO/NO$_2$ ratio over Southeast US (Aug-Sep 2013)

- Model NO/NO$_2$ bias in upper troposphere implies error in NO-NO$_2$-O$_3$ kinetics or positive interference in observations from unaccounted NO$_x$ reservoir
- This needs to be resolved for interpretation of OMI NO$_2$ columns and trends

Silvern et al., GRL 2018
Evaluation with cloud-sliced upper tropospheric OMI NO₂

450-280 hPa aircraft NO₂ data (Mar-Aug)

- OMI upper tropospheric NO₂ noisy but consistent with aircraft
- Strong correlation with LIS/OTD lightning
- Model has too high background, too weak enhancements – need better understanding of upper tropospheric chemistry

Marais et al., ACPD 2018
Fine-scale 4DVar inversion of satellite HCHO over Southeast US to optimize isoprene emissions

OMI HCHO column with 37% low bias corrected

Relative decrease of MEGAN-2 isoprene emissions

GEOS-Chem adjoint

$0.25^\circ \times 0.3125^\circ$

Isoprene, ppb

HCHO, ppb

Now implementing MEGAN-3 into GEOS-Chem

Kaiser et al., ACP 2018
Sulfate aerosol should now be titrated by ammonia in eastern US… but it isn’t

Data: EPA NEI & MASAGE

Emission Ratio
\[ \text{NH}_3 / \text{SO}_2 \]

Wet Deposition
Ammonium-sulfate Ratio
\[ [\text{NH}_4^+] / [\text{S(VI)}] \]

Aerosol
Ammonium-sulfate Ratio
\[ [\text{NH}_4^+] / [\text{S(VI)}] \]

Data: NADP

Excess ammonia

Major departure from standard sulfate-ammonium aerosol thermodynamics

Silvern et al., ACP 2017
Long-term trends in observations depart from thermodynamics

Averages for Southeast US summer 2013

Wet Deposition Fluxes (kg ha⁻¹)

- Sulfate
  - '03 to '06: 4.0
  - '06 to '09: 3.0
  - '09 to '12: 2.0
  - Trend: -6.1% a⁻¹

- Ammonium
  - '03 to '06: 0.8
  - '06 to '09: 0.6
  - '09 to '12: 0.4
  - Trend: +5.8% a⁻¹

CSN Aerosol Concentrations (μg m⁻³)

- Sulfate
  - '03 to '06: 6.0
  - '06 to '09: 4.0
  - '09 to '12: 2.0
  - Trend: -8.0% a⁻¹

- Ammonium
  - '03 to '06: 2.0
  - '06 to '09: 1.5
  - '09 to '12: 1.0
  - Trend: -8.5% a⁻¹

- Ammonium-sulfate Ratio [NH₄⁺]/[S(VI)] (mol mol⁻¹)
  - '03 to '06: 2.5
  - '06 to '09: 2.0
  - '09 to '12: 1.5
  - Trend: +3.0% a⁻¹

Ammonium-sulfate aerosol ratio decreases as sulfate decreases!

Silvern et al., ACP 2017
Possible retardation of thermodynamic equilibrium by organic aerosol

**Aerosol has changed over past decade**
from sulfate-rich to organic-rich

Evidence of phase separation, kinetic limitation to NH₃ uptake

Retardation/modification of equilibrium by organics
could affect uptake of other gases, including water
Two models for formation of secondary organic aerosol

Classical model for reversible uptake by pre-existing organic aerosol

Alternative model for irreversible uptake by aqueous aerosol
Aqueous-phase mechanism for organic aerosol from isoprene: the short version

Marais et al. [2016]
Observations show correlation of IEPOX SOA with sulfate

Correlations with sulfate in SEAC$^4$RS and at Centerville, Alabama research site

Sulfate ↓ → Aerosol volume ↓ → Aerosol acidity ↓ → IEPOX SOA ↓

Suggests that SO$_2$ emission controls decrease organic aerosol as co-benefit

Marais et al. [2016]
Representations of SOA in GEOS-Chem

• Default: simple linear parameterization
  - Isoprene SOA: emitted as 3% of isoprene
  - Terpene SOA: emitted as 10% of terpenes
  - Anthropogenic SOA: produced from emitted CO with fixed rate constant
  - Fire SOA: produced from emitted CO with fixed rate constant

  No chemistry required, does as well as advanced schemes

• Advanced:
  - VBS for all SOA
  - Aqueous-phase production coupled to isoprene chemistry for isoprene SOA
Tropospheric radical chemistry initiated by sea-salt chloride

Reactive tropospheric halogen sources in GEOS-Chem

Large effect on ozone, mostly initiated by

\[
\text{HOBr} + \text{Cl}^- + \text{H}^+ \rightarrow \text{BrCl} + \text{H}_2\text{O} \quad \text{(remote)}
\]

\[
\text{N}_2\text{O}_5 + \text{Cl}^- \rightarrow \text{ClNO}_2 + \text{NO}_3^- \quad \text{(polluted)}
\]

Xuan Wang, Lei Zhu (in prep)
Sensitivity of global-scale plume transport to model resolution

Numerical diffusion of inert free tropospheric plumes transported for 8 days with FV3 dynamical core (advection only) at horizontal resolution c384 to c48 (0.2° to 2°) and vertical resolution L160 to L20 (80 m to 0.6 km in free troposphere)

Optimal grid resolution ratio is $\Delta x / \Delta z \sim 1000$, but current models have $\Delta x / \Delta z \sim 20$

Zhuang et al., ACP 2018
SWIR observations of methane from space

- GOSAT: 10 km pixels spaced by 250 km, 3-day return time
- TROPOMI: global daily coverage with 7×7 km² pixels (nadir)
- geoCARB: geostationary view of the Americas with 3×3 km² pixels 2-3x/day
- GHGSat: targeted view of 12×12 km² scenes with 25×25 m² pixels
Retrieving point source emission rates from high-resolution remote sensing of instantaneous methane plumes

GHGSat-D (50x50 m² pixels) over Lom Pangar Dam, Cameroon

WRF large-eddy simulation at 50x50 m² resolution

AVIRIS-NG over Four Corners (Frankenberg et al. 2016)
Methods for inferring point source rates $Q$ from instantaneous observation of column plume enhancements $\Delta \Omega$

1. Gaussian plume inversion

$$Q = U \Delta \Omega(x, y) \sqrt{2\pi \sigma_y(x)} e^{-\frac{y^2}{2\sigma_y(x)^2}}$$

Fails for plumes < 10 km due to non-Gaussian behavior

2. Source pixel mass balance

$$Q = \frac{U W p}{g \Omega_a} \Delta \Omega$$

Fails for pixels < 1 km because eddy flow dominates ventilation

3. Cross-sectional flux

$$Q = U_{eff} \int_{plume\ width} \Delta \Omega(x, y) dy$$

4. Integrated mass enhancement (IME)

$$Q = \frac{1}{\tau} \int_{plume\ area} \Delta \Omega(x, y) dx dy = \frac{U_{eff}}{L} \int_{plume\ area} \Delta \Omega(x, y) dx dy$$

Both methods require estimates of plume size $L$ and effective wind speed $U_{eff}$
Testing the methods with independent set of LES plumes

Ability to retrieve “true” source rate $Q$ from instrument of precision $\sigma$

IME method

$x$-sectional flux method

<table>
<thead>
<tr>
<th>Method</th>
<th>Instrument precision</th>
<th>If no local wind data</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1%</td>
<td>3%</td>
</tr>
<tr>
<td>IME</td>
<td>0.07 t h$^{-1}$ + 5%</td>
<td>0.13 t h$^{-1}$ + 7%</td>
</tr>
<tr>
<td>x-sectional flux</td>
<td>0.07 t h$^{-1}$ + 8%</td>
<td>0.18 t h$^{-1}$ + 8%</td>
</tr>
</tbody>
</table>

• IME method better than $x$-sectional flux method
• Sources > 0.5 t h$^{-1}$ (75% of US GHGRP) can be usefully retrieved
• Lack of local wind data can dominate error at low winds

Varon et al., AMTD 2018