Constraining urban-to-global scale estimates of black carbon distributions, sources, regional climate impacts, and co-benefit metrics with advanced coupled dynamic - chemical transport - adjoint models

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**NOAA:** Georg Grell*

*supported by EPA STAR
Innovation in BC modeling & data assimilation

1. Science
2. Tools
3. Decision Support
Science Objectives

1. Rank and constrain the contributions of transport, deposition, aerosol properties, and emissions to uncertainty in estimates of BC distributions in urban and remote areas and of radiative forcing.

2. Improve model representation of BC distributions at urban to regional scales through assimilation of surface, aircr and remote sensing measurements.

3. Assess the range of BC uncertainties in climate impacts metrics, and develop novel metrics for air quality and climate impacts that reflect the competing effects of pollutants and account for propagation of uncertainties in sources, transport, and radiative processes.
Objectives: Tools & Methods

Develop and apply tools that improve model representation of BC distributions at urban to regional scales through assimilation of surface, aircraft and remote sensing measurements.

1. Global to regional: GEOS-Chem adjoint & 4DVAR
2. Urban to global, coupled: WRF-Chem & GSI 3DVAR
3. LES to global, coupled: WRF-Chem adjoint & GSI 4D
California: regional BC transport w/intense forest fire episodes and complex meteorology

- Constrain uncertainty in BC emissions & vertical distributions using in situ observations
- U.S. application of sectoral BC co-benefits metrics

India & East Asia: high urban & regional BC burdens, strong direct & indirect radiative forcing

- Constrain large, highly uncertain emissions inventory
- Sectoral BC co-benefits metrics across urban to regional scales

Arctic: long-range BC transport critical to global climate

- Constrain and compare uncertainty in BC emissions & long-range transport in regional & global models
- National/sectoral co-benefits metrics @ GCM scale
TOOLS
Developing a comprehensive open-source community toolkit for BC across scales

Direct Radiative Forcing

Emissions constraint & source attribution

Semi-direct & indirect effects

Climate response

Emissions control strategies

1D models

Co-benefits metrics and uncertainty quantification

Urban
Regional
Intercontinental

CMAQ, STEM

WRF-Chem

GEOS-Chem
What do adjoint chemistry + climate models do?

\[ \delta E_1 \]
\[ \delta E_2 \]
\[ \vdots \]
\[ \delta E_{10^5} \]
\[ \delta RF \]
Uncertainty Assessment & ConstrainUti

1. Total Uti uncertainty U
2. Adjoint sensitivities Uti quantify uncertainties Uti
3. 4DVAR constrainUti uncertainty Uti

Uncertainty (µg/m³) ti

- emissions ti
- concentration ti
- radiativti
BC Direct, Indirect, Semi-Direct Forcing Sensitivities: online, in situ, adjoint

Aerosol direct effects in WRF-Chem

- Aerosol composition, mass & size (MOSAIC, SORGAM, GOCART)
- Optical averaging
- Optical thickness (tau_e)
- Asymmetry parameter (g_{aer})
- Single-scattering albedo (ws)
- Optical Properties: 300, 400, 600, 900 nm

Reference: Fast et al. (2006)

Aerosol indirect effect in WRF-Chem

- Temp Tendency due to SW radiation
- Net flux @ surface (GSW)
- Upward TOA flux
- Cloud Optical Thickness
- Cloud fraction
- Pressure
- Water (gas, liquid, ice)

Mix-Activate
- Aerosol (MOSAIC, SORGAM, GOCART)
- Water (vapor & cloud)
- Droplet number

Updated droplet number

Updated aerosol properties (MOSAIC, SORGAM, GOCART)

References: Gustafson et al. (2007), Chapman et al. (2009)
APPLICATIONS
ASSIMILATING AAOD TO IMPROVE BLACK CARBON EMISSIONS & CONCENTRATIONS

Anthropogenic BC Emissions @ 0.67° x 0.5°

Bond, 1.0°x1.0°, 2000

SEAC4RS, 0.1°x0.1°, 2012

INTEX-B, 0.5°x0.5°

MEIC, 0.5°x0.5°

Mg BC/yr

(Bond et al., 2007) (Zhang et al., 2009) (Lu et al., 2011) (Zhang et al., 2012)
GEOS-Chem simulated AAOD vs. OMI Observed AAOD

April

GEOS-Chem minus OMI

OMI observed data counts

OMI aer optical depth (AA an atmospheric measurement of aerosol par al., 2007], w much better spa temporal constrain BC

OMI AAOD L2 da corrected Chem aer
Anthropogenic emissions optimized using GEOS-Chem adjoint + MEIC_SEAC$^4$RS

April

Priori

Posteriori

Posteriori - Priori

October
Optimized AAOD evaluation, April 2006

- Posterior AAOD consistent with AERONET AA observations in April in Beijing and eastern Indo-Gangetic Plain (70°-90°E).
- Large discrepancies over India and Thailand even after data assimilation.
- Residual errors associated with limited and sparse OMI AAOD observations, inconsistencies with AERONET products.
Optimized Surface BC Concentrations

April

Priori

Posteriori

October

Priori

Posteriori

R2=0.57
y=0.15x+0.762
R2=0.61
y=0.68x+0.501

R2=0.75
y=0.24x+0.728
R2=0.81
y=0.29x+0.593
Take Home Points: Constraining BC in GEOS-Chem Adjoint using OMI AA

1. **Science: attribution & reduction in uncertainty**
   - April: higher anthropogenic emissions after optimization (up to 500%). BB increase significant across south Asia
   - October: +10-50% over eastern China. Higher BB over Indonesia
   - Observational counts near sources determine effectiveness of inversion

2. **Tools: ready for operational use**
   - top-down, spatially resolved emissions inversion
   - many choices for AOD products & DA methods

3. **Policy: better upstream inputs; further improv by DA requires more satellite and surface obs**
INITIAL APPLICATION OF THE WRF ADJOINT FOR BC: FOREST FIRE SMOKE DURING ARCTAS-CARB

WRF-Chem Adjoint Development

Adjoint: chemical transport
✓ Surface-air interactions (LSM, SFCLAY)
✓ Turbulent Mixing (PBL)
✓ Emissions and Deposition
✓ Chemistry (BC aging + sulfate)

Adjoint: climate forcing by aerosols
• Radiation
• Microphysics
• Cumulus Convection

I/O mechanism for long duration chemical inversions (>3-6hr)
3 hour Derivative Tests

\[ \frac{\partial [BC]}{\partial X} \]

TL and AD agree to 8+ digits
Sensitivity demonstration: ARCTAS-CARB

Time Period
Jun 20 00Z to Jun 27 09Z, 2008
(Friday to Friday)

Observations

- **IMPROVE**
  - Jun 20, 23, 26
  - Daily avg. @ ~40 sites (local time)
  - 0.025 $\mu$ g/m$^3$ mdl
  - 7-50% uncertainty (conc. dependent)

- **ARCTAS-CARB (DC-8)**
  - Jun 20, 22, 24, 26 (~midday)
  - Single Particle Soot Photometer (SP2)
  - 10 sec sample rate
  - 0.01 $\mu$ g/m$^3$ mdl
  - 30% uncertainty

Model Configuration

- **WRF-Chem V3.6**
  - 18km – 79 x 79 x 42 levels
  - GOCART Aerosols

- **IC/BC**
  - MET BCs: 32km NARR Reanalysis
  - CHEM spun up from Jun 15
  - $[BC]_{\text{bound}} = 0.01 \mu$ g/m$^3$

- **Emissions**
  - NEI2005 Anthro Emissions (4km)
  - FINN BB Emissions (1km) w/plumerise

- **Physics**
  - ACM2 PBL Mixing (7)
  - Pleim-Xiu SFCLAY & LSM (7)
  - radiation*: RRTM LW (1) & GSFC SW (2)
  - no microphysics, no cumulus convection

*Forward model only for radiation in this application
Total Emitted Black Carbon (Jun 20-26, 2008)

Anthropogenic - NEI 2005

Weekday Diurnal Patterns

x 10^3 kg
Total Emitted Black Carbon (Jun 20-26, 2008)

WRAP Diurnal Pattern

Burning - FINN

[Map and graph showing emission patterns with coordinates and emission values in kg]
Model to Obs. Comparison

**ARCTAS-CARB, 2008:**

June 20, 22, 24, 26

**IMPROVE** Sites; June 20, 23, 26

![Map and Graph](image)
Model to Obs. Comparison

![Graph showing model to observation comparison](image)

- **ARCTAS**
- Model [BC] (µg m⁻³) vs. DC-8 Observed [BC] (µg m⁻³)
- Altitude
- Obs. Height / PBLH
- PBLH (m)
- Residual = Model - Obs.
Model/Obs. Misfit
structural & parametric uncertainties

\[ J = \frac{1}{2} \sum_k \left[ H_k(M_k(x)) - y_k \right]^T R^{-1} \left[ H_k(M_k(x)) - y_k \right] \]

Cost Function
Net Diurnal Sensitivities

\[ \frac{\partial \ln J}{\partial \ln E_n} \]

ANTHRO emissions are too high, except during evening rush hour

BURNING peak timing/prominence could be off
PBL Mixing Implications

ANTHRO - WEEKDAY

BURNING

Fully Normalized Sens.

Local Time (hr)

PBL Height (m)
Take Home Points: WRF Adjoint

1. **Tool**: Developed adjoint of NWP + chemistry model
2. **Decision Support**: Local/regional source attribution
3. **Science**
   - Enables determination of model process vs. emission contributions to errors
   - Sensitivities of direct & indirect radiative forcing to emissions (ongoing adjoint development)
ASSIMILATION OF NEXT GENERATION GEOSTATIONARY RETRIEVALS & MULTI-PLATFORM INVERSION OF WILDFIRE EMISSIONS


Saide, P.E., et al. Revealing important nocturnal and day-to-day variations in fire smoke emissions through a novel multiplatform inversion. Submitted.
East Asia pollution episodes

- April 26 – May 9 2012,
  3 types of events:
  - Dust
  - Anthropogenic
  - Biomass burning

- WRF-Chem v3.5 CBM-Z, 8bin MOSAIC, anthropogenic emissions (MIX), biomass burning (QFED2), dust (GOCART), MACC boundary conditions

- Regional modeling system resolves episodes but underestimates particle loads
3DVAR experiments

- Objective: Assess performance of adding GOCI AOD to a system already assimilating MODIS AOD
- System: WRF-Chem + GSI for MOSAIC sectional aerosol model (Saide, ACP 2013) supports assimilation of multiple data sources
- GSI AOD assimilation every 3 hours: MODIS, MODIS + GOCI
- GOCI assimilated only over ocean

Lee et al., RSE 2010, Park et al., ACP 2014
Improved assimilation from geostationary updates every 3 hours

- Evaluation @AERONET DRAGON + PM$_{10}$ network
- GOCI fills gaps in MODIS (clouds, ocean glint)
Forecasting episode AOD & PM$_{10}$: from marginal to skillful

Geostationary DA improves AQ episode forecasts: all hours, all sources
Multiplatform inversion of the 2013 Rim Fire smoke emissions during the SEAC4RS campaign

- WRF-Chem v3.5, 12km CONUS
- CBM-Z, 4 bin MOSAIC
- NRL AOD assimilated every 3h
Rim Fire smoke during SEAC4RS
Simultaneously improve model fit to multiple measurements:
- SEAC4RS in-situ (OA, CO, extinction) and remote sensing (DIAL-HSRL Extinction) from 2 flights
- AERONET AOD (1, 2, 3, 4)
- Ground-based CO (1, 5, 6)
- Satellite AOD

2 met boundary conditions (FNL, NARR)
Inversion Process

- Constrained emissions factor of 3 higher
- FNL & NARR inversions consistent

![Correction factors](image)

![Emissions](image)

<table>
<thead>
<tr>
<th>Flight Extinction</th>
<th>Flight CO</th>
<th>AERONET AOD</th>
<th>Ground CO</th>
<th>Satellite AOD</th>
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</thead>
<tbody>
<tr>
<td>Initial Observable FB=0.73 FE=0.91</td>
<td>FB=0.46 FE=0.54</td>
<td>FB=0.67 FE=0.70</td>
<td>FB=0.20 FE=0.40</td>
<td>FB=0.58 FE=0.66</td>
</tr>
<tr>
<td>Constrained Observable FB=0.17 FE=0.58</td>
<td>FB=0.25 FE=0.35</td>
<td>FB=-0.01 FE=0.45</td>
<td>FB=0.00 FE=0.34</td>
<td>FB=0.01 FE=0.50</td>
</tr>
</tbody>
</table>
Inversion Results

- Flight + Ground higher correction factors than satellite
- Nocturnal emissions up to 26x higher
Inversion Results

Day to day variability in constrained emissions correlates to daily burned area from airborne infrared obs.
Impacts on smoke $\text{PM}_{2.5}$, AOD, radiative forcing

- **Maximum changes, posterior – prior**
- **Methods support constraint across observed $\text{PM}_{2.5}$ range:**
  4 orders of magnitude
1. Science
   - Constraining fire emissions with multiple data sources can better characterize fires and improve air quality predictions
   - Assimilating next gen geostationary AOD improves air quality forecasts

2. Tools
   - Practical additions to GSI + any model
   - Next: GSI + WRFPLUS adjoint = BC 4DVAR

3. Decision Support
   - Fast track to better inputs for monitor siting, forecasting, SIP, policy analysis applications
A conceptual model for the link between Central American biomass burning aerosols and severe weather over the south central United States

Jun Wang\textsuperscript{1,2,3,6}, Susan C van den Heever\textsuperscript{4} and Jeffrey S Reid\textsuperscript{5}
Case Study: 27 April 2011 outbreak

“By many metrics, the tornado outbreak on 27 April 2011 was the most significant outbreak since 1950”
BB smoke, stronger tornadoes

- BC absorption contributes ~40-80% to enhancements in tornado parameters
- Mechanism: strengthening the capping inversion by heating layer above clouds

ADJOINT METRICS FOR AIR QUALITY & CLIMATE CO-BENEFITS

SN Spak, DK Henze, F Lacey, EA Minor, GR Carmichael. Adjoint health and climate co-benefits metrics identify optimal local and global air pollution control policies. In preparation.
Global aerosol direct radiative forcing & population-weighted PM$_{2.5}$ sensitivities from every sector & region

\[ \delta E_1 \]
\[ \delta E_2 \]
\[ \vdots \]
\[ \delta E_{10^5} \]

Calculated very efficiently with the
**GEOS-Chem adjoint** (Henze et al., 2007) + **LIDORT** (Spurr, 2002)
RCP2000 sensitivities: concentrations

Population-weighted BC concentrations : emissions
($\mu g/m^3/kg/year$, log scale)

\[
\text{Sensitivity}_{SLFCA} = \frac{\partial \ln \text{Concentration}[SLFCA]}{\partial \ln \text{Emissions}[SLFCA]}
\]
RCP2000 sensitivities: SLCF

Arctic BC DRF : emissions (W/m²/kg/year or %/%, log scale)

\[ \text{Elasticity}_{SLFC} = \frac{\Delta SLFC / SLFC}{\Delta \text{Emissions}[SLFCA] / \text{Emissions}[SLFCA]} = \frac{\partial \ln SLFC}{\partial \ln \text{Emissions}[SLFCA]} \]
Global BC DRF: emissions
(W/m²/kg/year, log scale)

\[ \text{Elasticity}_{\text{SLFC}} = \frac{\Delta \text{SLFC}}{\text{SLFC}} = \frac{\partial \ln \text{SLFC}}{\partial \ln \text{Emissions[SLFCA]}} = \frac{\Delta \text{Emissions[SLFCA]}}{\text{Emissions[SLFCA]}} \]
Reducing uncertainties: adding regional refinement

Global BC DRF by Latitude Band / Global Average
Co-benefits metrics

Modelers calculate:

\[
Elasticity_{SLFC} = \frac{\Delta SLFC / SLFC}{\Delta \text{Emissions}[SLFCA] / \text{Emissions}[SLFCA]} = \frac{\partial \ln \text{SLFC}}{\partial \ln \text{Emissions}[SLFCA]}
\]

\[
Sensitivity_{SLFCA} = \frac{\partial \ln \text{Concentration}[SLFCA]}{\partial \ln \text{Emissions}[SLFCA]}
\]

We’re adding:

\[
\text{Forcing Amplifier}_{SLFCA} = \frac{\partial \ln \text{SLCF}[SLFCA]}{\partial \ln \text{Concentration}[SLFCA]} = \frac{Elasticity_{SLFC}}{Elasticity_{SLFCA}}
\]

\[
\text{Emissions Reduction Efficiency}_{SLFCA1,SLFCA2} = \frac{\text{Forcing Amplifier}_{SLFCA1}}{\text{Forcing Amplifier}_{SLFCA2}}
\]

+ impacts, cost:benefit, uncertainties
Where to focus BC on climate vs AQ?

Global BC DRF by latitude band: Population-weighted BC (W•m/µg, log scale)

\[ \text{Forcing Amplifier}_{SLFCA} = \frac{\partial \ln \text{SLCF}[SLFCA]}{\partial \ln \text{Concentration}[SLFCA]} = \frac{\text{Elasticity}_{SLFC}}{\text{Elasticity}_{SLFCA}} \]
Design policies to reflect regional differences?

Global BC DRF by latitude band: Population-weighted BC (W•m/µg, log scale)

\[
\text{Forcing Amplifier}_\text{SLFCA} = \frac{\partial \ln \text{SLCF}[\text{SLFCA}]}{\partial \ln \text{Concentration}[\text{SLFCA}]} = \frac{\text{Elasticity}_{\text{SLFC}}}{\text{Elasticity}_{\text{SLFCA}}}
\]
Where to focus on BC:SO$_2$ for climate?

Global BC:SO$_2$ Emissions Reduction Efficiency

Manage the ratio

Reduce BC
The 75 Largest Cities

- Population
- Total GDP
- GDP by sector:
  - Commodities
  - Construction
  - Business/Finance
  - Manufacturing
  - Local/non-market
  - Trade/tourism
  - Transportation
  - Utilities
- Sources: Brookings Institute Global MetroMonitor, PWC Global City GDP rankings 2008-2025

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## Top 10 Highest BC Sensitivities

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<tr>
<th>Global DRF</th>
<th>Per Capita Global DRF</th>
<th>Arctic DRF</th>
<th>Global FA</th>
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North American Cities

Population Weighted BC

BC DRF Arctic
2012


Tsao, C.C., J.E. Campbell, M.A. Mena-Carrasco, S.N. Spak, G.R. Carmichael, Y. Chen. Biofuels that cause land-use change may have much larger non-GHG air quality emissions than fossil fuels, *Environ. Sci. Technol.* 46, 10835–10841.

2013


2014


Submitted


In preparation


Ongoing: Year 4

- WRF-Chem adjoint development
  - public release with climate coupling
  - publication

- Co-benefits Applications
  - Global futures: quantify impacts of proposed BC-specific emission mitigation policy measures
  - Impacts of emissions inversion: Asia
  - Process uncertainties: CA
Conclusions

1. **Science**
   - Emissions the greatest parametric uncertainty, and the most readily reduced through assimilation

2. **Tools**
   - We have advanced BC adjoint and DA capabilities for AQ + climate, extended them to multi-scale coupled modeling in WRF-Chem, and incorporated them in public model releases and operational DA platforms
   - synergy between non-linear sensitivity tools & emerging observing systems for understanding and reducing uncertainties in BC

3. **Policy analysis & decision-making support**
   - Adjoint metrics identify where and how to most quickly improve climate & AQ
Thank you!