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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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 BC radiative forcing.

2. Improve model representation of BC distributions at urban to regional scales through assimilation of surface, aircraft 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 co-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: WRFPlus-Chem adjoint, WRFDA-Chem 4DVAR
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

Co-benefits metrics and uncertainty quantification

Urban Regional Intercontinental Global

1D models

WRF-Chem

CMAQ, STEM

GEOS-Chem
Uncertainty Assessment & Constraint

1. Total uncertainty
2. Adjoint sensitivities quantify uncertainties
3. 4DVAR constrained uncertainty

Constraint on:
- emissions
- concentrations
- radiative forcing
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 inventories
- 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
AND THEN, SCIENCE HAPPENED

BASIC & APPLIED
GLOBAL & REGIONAL
TOOLKIT & RESEARCH RESULTS

2015-2016
California: regional BC transport w/intense forest fire episodes and complex meteorology

- Constrain uncertainty in BC emissions & vertical distributions using in situ observations
  - Rim Fire + fire attribution + tornadogenesis

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

- Constrain large, highly uncertain emissions inventories
  - Eastern China Haze AQ:climate feedbacks
  - Geostationary assimilation + Central Asia BC

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 + Megacity co-benefits metrics
ASSIMILATING AAOD TO IMPROVE BLACK CARBON EMISSIONS & CONCENTRATIONS

Uncertainties in 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°, 2006

MEIC, 0.5°x0.5°, 2010

(Bond et al., 2007) (Zhang et al., 2009) (Lu et al., 2011) (Zhang et al., 2012)
Grid-scale BC emissions constraints

How to isolate BC when assimilating AAOD?

Use model
BC/total AAOD
in each layer

Use model
BC/total AAOD
in each column

Use model
absolute OC and Dust AAOD
in each column

Use OMI retrieval aerosol classification: smoke, sulfate, dust
(Torres et al., 2013)
Seasonal, spatial differences in AAOD
GEOS-Chem simulation vs. OMI retrieval

OMI aerosol absorption optical depth (AAOD) is an atmospheric column measurement of absorbing aerosol particles [Torres et al., 2007], which provides much better spatial and temporal coverage to constrain BC.

OMI AAOD L2 data were corrected using GEOS-Chem aerosol layer height.
Anthropogenic emissions optimized using GEOS-Chem adjoint + MEIC_SEAC$^4$RS

April

Priori

Posteriori

Posteriori - Priori

October

Priori

Posteriori

Posteriori - Priori

$10^4$ Kg
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 AAOD

1. Science: attribution & reduction in uncertainty
   - Seasonal: higher anthropogenic emissions after optimization (up to 500%), BB increase significant, spatially variable
   - Observational counts near sources determine effectiveness of inversion
   - Extension to U.S. long-term trends (submitted)

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 improvement by DA requires more satellite & surface obs


Findings:

• Secondary inorganic 70% of PM$_{2.5}$ mass, but BC absorption matters: up to 50% of net aerosol feedbacks

• Aerosol feedbacks important: lower PBL, less sun, change in surface temperature, wind speed/direction, net increase in PM$_{2.5}$

• January 2013 haze: 690 premature deaths, 45K acute bronchitis, 24K asthma cases

• $254$ MM losses = 0.08% of Beijing GDP
DEVELOPMENT & INITIAL APPLICATION OF THE WRF ADJOINT FOR BC: FOREST FIRE SMOKE DURING ARCTAS-CARB


J.J. Guerette, CU-Boulder
Biomass burning emissions: uncertainty abounds

\[ \Sigma E_{BC,BB} = 5031 \times 10^6 \text{ g} \]
\[ dx = 18 \text{ km} \]

Darmenov and da Silva (2013)

\[ \Sigma E_{BC,BB} = 488 \times 10^6 \text{ g} \]

Wiedinmyer et al. (2011)

\[ \Sigma E_{BC,ANT} = 548 \times 10^6 \text{ g} \]

ANTHROPOGENIC:
National Emission Inventory (NEI2005)

\[ \Sigma E_{BC,ANT} = 548 \times 10^6 \text{ g} \]

BC Emissions, Jun 20-27, 2008:
\times 10 spread similar to:
• Zhang et al. (2014)
  (7 inventories in Sub-Saharan Africa)
• Fu et al. (2012)
  (2 inventories in Southeast/East Asia)
Bayesian Inversion Framework

\[ P(x|y^0) \propto P(x) P(y^0|x) \]

Gaussian distributions:
1. Prior
   \[ P(x) \propto \exp\left[-\frac{1}{2}(x-x^b)^T B^{-1}(x-x^b)\right] \]

2. Model/Obs. mismatch
   \[ P(y^0|x) \propto \exp\left[-\frac{1}{2}(y-y^o)^T R^{-1}(y-y^o)\right] \]

\[ y = G(x) \]

- \( x \) – control variable
- \( y^0 \) – observations
- \( y \) – modeled observations
- \( B \) – Background covariance
- \( R \) – Model/Obs. covariance

3D: \( x \in \mathcal{R}^{n_x \times n_y \times n_t} \)

4D, sparse

2D (spatial) + 1D (temporal) fields of chemical fluxes

Model + Observation Operator
WRFDA-Chem Model Family

**WRF**: Advection, diffusion, PBL mixing, convection, microphysics, radiation

\[ G(\chi) \]

-**Chem**: Chemical transformation, advection, emission, fire plume rise, dry/wet deposition, vertical mixing (PBL and cumulus)

[Skamarock et al. (2008)]

[Grell et al. (2005)]

**WRFPLUS**: Tangent Linear (TLM) and Adjoint (ADM) models for meteorology

**Chem**: ADM/TLM of PBL transport, emissions, dry deposition, and GOCART aerosols

[Guerrette and Henze (GMD, 2015)]

**WRFDA**: 4D-Var for meteorology (initial and boundary conditions), many in-situ and remote obs. types

**Chem**: 4D-Var for chemical emissions control variables and in-situ observations

[Barker et al. (2005); Huang et al. (2009); Zhang et al. (2014)]

[Guerrette and Henze, in review, ACPD]
WRF-Chem Adjoint Development

Adjoint: chemical transport

- Surface-air interactions (LSM, SFCLAY)
- Turbulent Mixing (PBL)
- Emissions and Deposition
- Chemistry (BC aging + sulfate)
- Observation operators (BC, sulfate, dust)
Development milestones

- Implemented ADM/TLM
- Verified gradients by comparison to finite difference approximations
- Calculated model/observation variance
- Reduced memory requirements for simulations with $\Delta t > 6$ hr using second order checkpointing

$$\frac{\partial [BC]}{\partial x}$$

$$\text{diag}(R)_i = \sigma_i^o^2 = \sigma_{\text{model},i}^2 + \sigma_{\text{instrument},i}^2$$

Stochastic variance from 156 ensemble members
June 22, 2008 ARCTAS-CARB Case Study

- **BC measurements**
  - NASA DC8 SP2 on 22 June
  - 241 grid-scale measurements
  - [Kondo et al., 2011; Sahu et al., 2012]

- **Domain**
  - 18 km, $80 \times 80 \times 4$

- **Prior emissions**
  - FINN biomass burning emissions, x3.8 relative uncertainty
  - NEI2005 anthro. emissions, x2.0 relative uncertainty
  - Assumed temporal and spatial error correlations

- **Inversion Configuration**
  - 5 outer iterations x10 inner iteration for $x^a$
  - 1 outer iteration x50 inner iterations for $P^a$
  - 24 hour assimilation window from 00Z, 22 June

MODEL – OBS = $y - y^o$

-Guerrette and Henze, 2015, GMD-
22 June model, 4DVAR, inversion x2 emissions inventories

[Guerrette and Henze, in review, ACP]
Biomass Burning BC Constraint

- Relative increments are correlated in space (as expected; $L_{x,y} = 36$ km)
- Absolute increments are disperse, collocated with large priors or large residual errors
- Consistent sign of increments across broad areas due to sparse observations + enforced emission correlation

[Guerrette and Henze, in review, ACPD]
Take Home Points: WRFDA-Chem Adjoint

1. **Tools:** Developed adjoint of NWP + chemistry model. WRFDA-Chem 4D-Var is **fully functional** for BC and other tracer emission inversions. These tools lay the foundation for 4D-Var in a coupled meteorology chemistry model and for operational air quality forecasting.

2. **Decision Support:** Local/regional source attribution

3. **Science:** Enables determination of model process vs. emission contributions to errors
ASSIMILATION OF NEXT GENERATION GEOSTATIONARY RETRIEVALS & MULTI-PLATFORM INVERSION OF WILDFIRE EMISSIONS


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
- SEAC4RS in-situ + remote sensing ground observations assimilated

WRF-Chem
  - Full Chemistry
  - Hourly Tracers

OBS Sensitivities

Tracer Sensitivities

Variational Inversion

Correction factors

Airborne
Ground
Satellite
Impacts on smoke PM$_{2.5}$, AOD, SLCF

- Maximum changes, posterior – prior
- Methods support constraint across observed PM$_{2.5}$ range:
  4 orders of magnitude
Operational real-time GSI assimilation & inversion in ORACLES forecasting

Instrumentation & Models  NASA Earth-Venture-Suborbital-2 project

GO TO CROSS-SECTION

ORACLES Forecast, 00h = 12UTC, 21/08/2016

Domain  -  Africa  +
Pressure  -  860 mb  +
Start  36  hour
End  +  56  hour

Aerosols:  ACAOD  AOD
AOD WFA  AOD IFA  AOD DIFF
EXT  EXT WFA  EXT IFA
EXT DIFF  NUM  PM2.5

Tracers:  BB Age Mean
BB Age Mode  BB Col Age Mean
BB Col Age Mode  CO BB
CO col BB  BB day <10
BB day -10  BB day -9  BB day -8
BB day -7  BB day -6  BB day -5
BB day -4  BB day -3  BB day -2
BB day -1  BB day 0  BB day 1
BB day 2  BB day 3  CO Anthro
CO col Anthro

Meteorology:  CLD BASE  CLD TOP
CLD TOP DIFF  LWP LWP
LWP DIFF  LWP+IWP
NUM DROP  NUM DROP DIFF
PBL Height  PRECIP CONVY
PRECIP TOT  RH T T DIFF
Take Home Points: GSI Assimilation

1. Science
   - Constraining fire emissions with multiple data sources can better characterize fires, improve AQ predictions
   - Assimilating next gen geostationary AOD improves AQ forecasts

2. Tools
   - Attribution + inversion in coupled AQ + climate model using tracer + concentration + climate sensitivities
   - Practical additions to GSI + any model

3. Decision Support
   - Fast track to better inputs for monitor siting, forecasting, SIP, policy analysis applications
Case Study: 27 April 2011 outbreak

The tornadoes of spring 2011 in the USA: an historical perspective

Charles A. Doswell III1,2, Gregory W. Carbin3 and Harold E. Brooks4

historical record of tornadoes in the USA. The past offers considerable insight into the deadly tornado events of this past spring, and may also provide a glimpse into the tornado that strikes in an area with little or no human population may have minimal societal impact even if it is large and violent; these rural events are likely to be under-

Service Assessment

The Historic Tornadoes of April 2011

“By many metrics, the tornado outbreak on 27 April 2011 was the most significant outbreak since 1950”
Where there’s smoke…

WRF-Chem simulations indicate that the presence of smoke during this event results in lower cloud bases and stronger low-level wind shear which indicate higher probability of tornadogenesis and tornado intensity and longevity.

Why?
- Optical thickening of shallow clouds due to increase CCN
- Enhancing of the capping inversion due to soot absorption

Saide et al., GRL 2015b
Analyzing a decade of outbreaks

Updated near-operational NWP approach for SLFC & regional attribution: WRF with aerosol-aware microphysics (AAM) (Thompson and Eidhammer, 2014) with WRF-Chem primary emissions

Aerosol Number concentration [#/cm³] Saide et al., JGR 2016
Found: how, when smoke impacts

- Large spread in smoke effects on environmental conditions, from negligible impacts to large intensification.
- 27 Apr 2011 intensification consistent with WRF-Chem MOSAIC

FIRE ON+INVERSION – FIRE OFF

Saide et al., JGR 2016: Higher likelihood of tornado formation and intensity
RCP2000 sensitivities: SLCF

Global BC DRF : emissions
(W/m²/kg/year, log scale)

\[
Elasticity_{SLFC} = \frac{\Delta SLFC / SLFC}{\Delta Emissions[SLFCA] / Emissions[SLFCA]} = \frac{\Delta \ln SLFC}{\ln Emissions[SLFCA]}
\]
Where to focus BC on climate vs AQ?

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

Forcing Amplifier_{SLFC} = \frac{\ln SLCF_{SLFC}}{\ln Concentration_{SLFC}} = \frac{\text{Elasticity}_{SLFC}}{\text{Elasticity}_{SLFC}}
Design policies to reflect urban/regional differences in sources, mixtures, impacts?

Global BC:SO\textsubscript{2} Emissions Reduction Efficiency

\[
\text{Emissions Reduction Efficiency}_{SLFCA1,SLFCA2} = \frac{\text{Forcing Amplifier}_{SLFCA1}}{\text{Forcing Amplifier}_{SLFCA2}}
\]
## 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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North American Cities
Conclusions: Science

- Start here: emissions are the greatest parametric uncertainty, and readily reduced through assimilation, globally, across scales.
- Nested solutions: constraining biomass burning emissions represents a potential fast track to improve rates + spatial/temporal attribution for other sectors, globally, across scales.
- Climate surprises: BC direct + indirect SLCF can influence tornado likelihood, severity.
Conclusions: 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

• Shared experimental recipes for BC sensitivity, DA, attribution using AQ + climate models in real world conditions, averages, extremes

• Synergies between non-linear sensitivity tools & emerging observing systems for understanding and reducing uncertainties in BC
Conclusions: Decision support

• Adjoint co-benefits metrics: just add emissions to identify where and how to most quickly improve climate & AQ together

• BC SLFC:AQ and BC:SO$_2$ management vary by orders of magnitude globally and among megacities

• Trans-boundary BC-specific SLFC may have broad environmental policy implications
Thank you!
Publications

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


2015 (continued)


Submitted


Zhang, L., D. K. Henze, G. A. Grell, O. Torres, H. Jethva, L. N. Lamsal, What factors control the trend of increasing AAOD over the United States in the last decade?

In preparation


Xu, J., et al., Source attribution of Arctic black carbon constrained by aircraft and surface measurements.