Inverse modeling and sensitivity studies in atmospheric chemistry: aerosols, emissions, and direct radiative forcing

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Areas of interest

Model sensitivity:
How do model estimates in atmospheric chemistry depend upon numerous uncertain parameters?

Data assimilation:
How can large sets of gas- and aerosol-phase measurements be used to best constrain our estimate of the chemical state of the atmosphere?

Mitigation strategies:
What are the most effective strategies for controlling air quality and climate change?

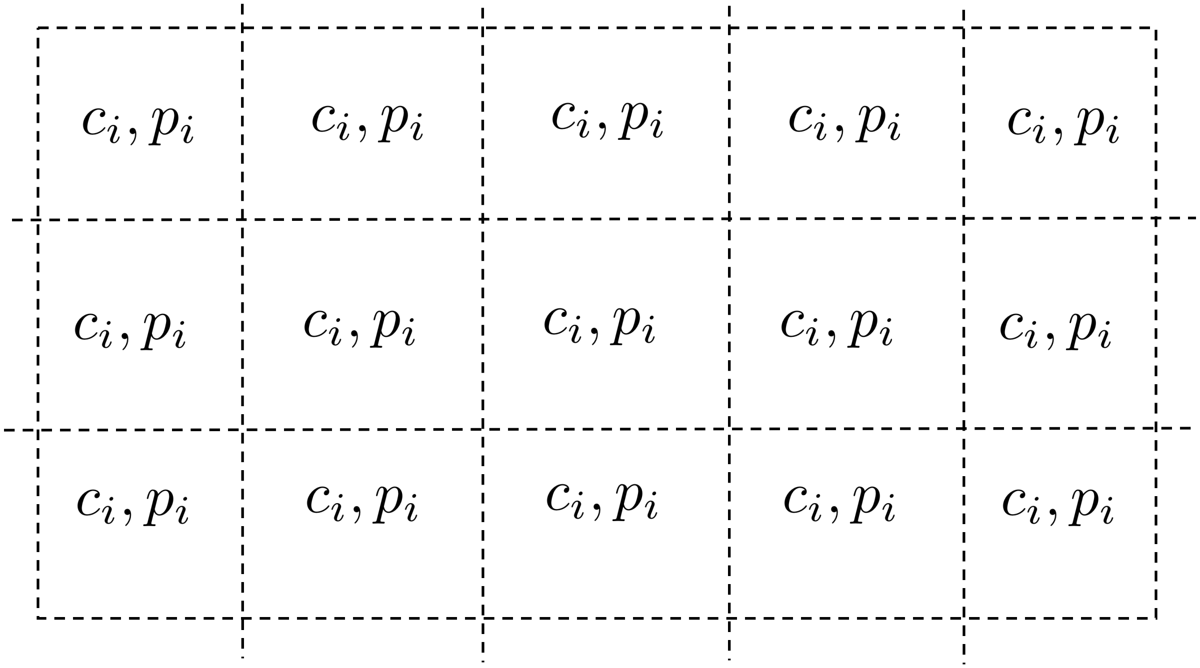
Address these issues using adjoint models

Adjoint modeling: ??

So what is an adjoint model?

… a different way of thinking about model sensitivity...

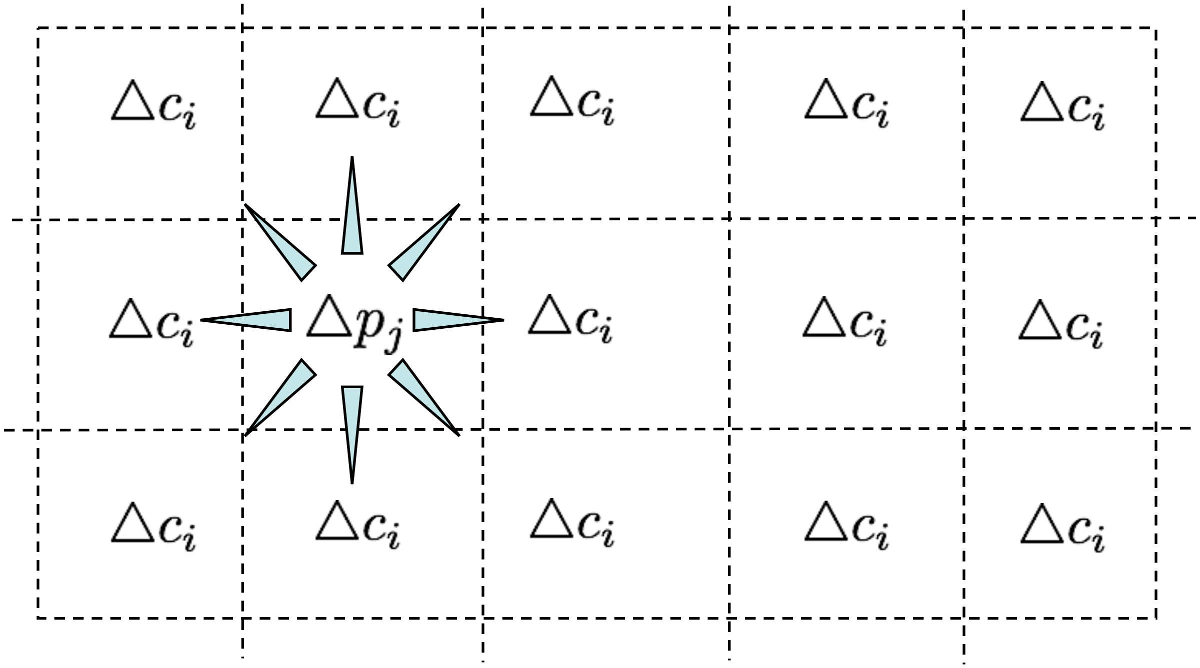
Forward sensitivity



c_i, p_i	c_i, p_i	c_i, p_i	c_i, p_i	c_i, p_i
c_i, p_i	c_i, p_i	c_i, p_i	c_i, p_i	c_i, p_i
c_i, p_i	c_i, p_i	c_i, p_i	c_i, p_i	c_i, p_i

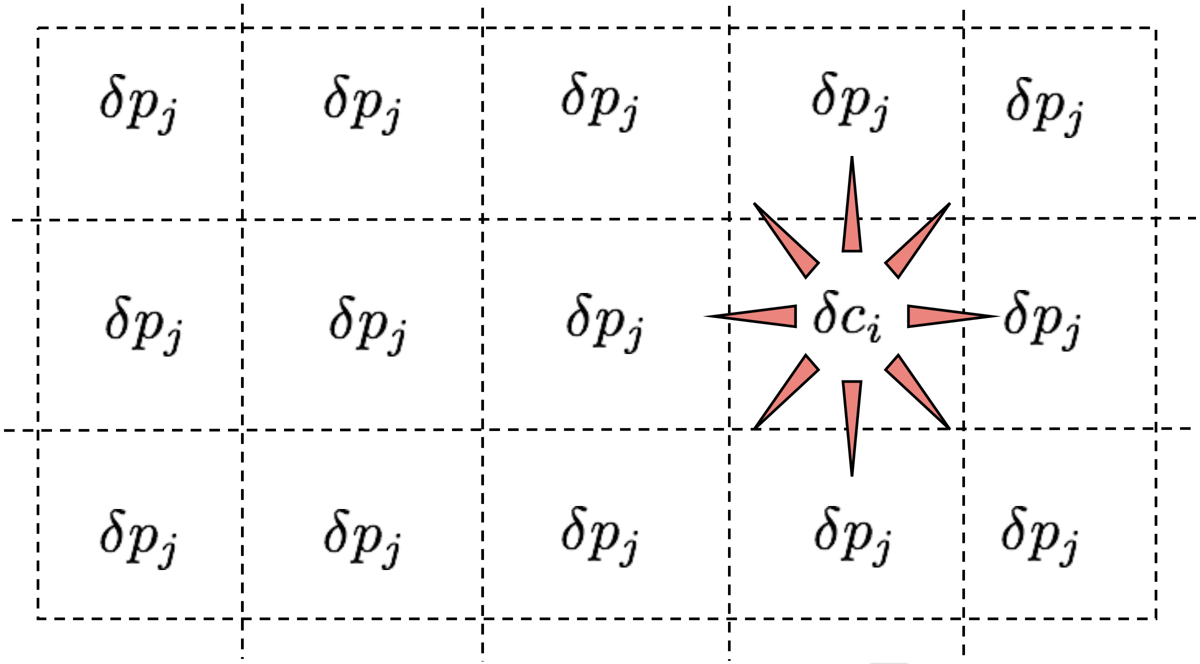
Model estimates, c_i
Model parameters, p_i

Forward sensitivity



$$\frac{\partial \mathbf{c}}{\partial p_j} = \begin{bmatrix} \frac{\partial c_1}{\partial p_1} & \cdots & \frac{\partial c_1}{\partial p_J} \\ \vdots & \ddots & \vdots \\ \frac{\partial c_I}{\partial p_1} & \cdots & \frac{\partial c_I}{\partial p_J} \end{bmatrix} \mathbf{u_j}$$

Adjoint sensitivity



$$\begin{bmatrix} \frac{\partial c_1}{\partial p_1} & \cdots & \frac{\partial c_1}{\partial p_J} \\ \vdots & \ddots & \vdots \\ \frac{\partial c_I}{\partial p_1} & \cdots & \frac{\partial c_I}{\partial p_J} \end{bmatrix}^T \mathbf{u_i} = \frac{\partial c_i}{\partial \mathbf{p}}$$

Adjoint modeling applications

Depending on “model response,” can be used for:

Sensitivity analysis: quantifying influence of uncertain model parameters (emissions, reaction rates, …)
Response = Average concentrations of X in location Y...

Inverse modeling: using large data sets, optimizing parameters on resolution commensurate with forward model.
Response $\sim$ *sum(model – obs)*^{2}

Attainment studies: assessing the effectiveness of emissions changes on an air quality
Response = total amount of nonattainment

Adjoint modeling: History

From principles of functional analysis (Hilbert)

Used extensively for optimal control problems (Lions, 1971)
- nuclear reactor design (classified?!)
- oceanography (Tziperman and Thacker, 1989)
- meteorology (Derber, 1985)
- aeronautics (Giles and Pierce, 2000)

Atmospheric chemistry
- proposed for tracer analysis (Marchuk, 1974)
- stratospheric chemistry (Larry et al, 1995)
- tropospheric chemistry (Elbern and Schmidt, 1999)

Aerosols
- microphysics, 2D (Henze et al., 2004; Sandu et al., 2005)
- black carbon (tracer) (Hakami et al., 2005)
- coupled thermodynamics and chemistry (Henze et al., 2007)
- AOD (offline chemistry) (Dubovik et al., 2008)

Adjoint modeling applications

Depending on “model response,” can be used for:

Sensitivity analysis: quantifying influence of uncertain model parameters (emissions, reaction rates, …)

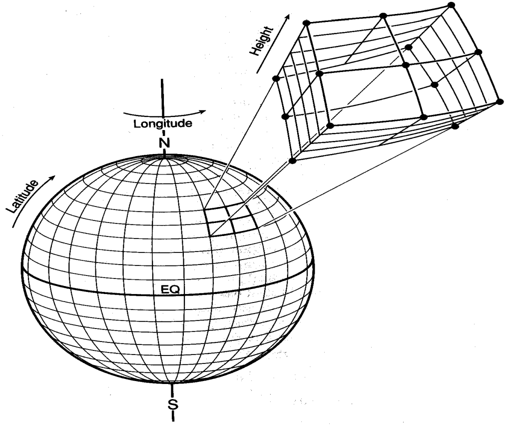
Inverse modeling: using large data sets, optimizing parameters on resolution commensurate with forward model.

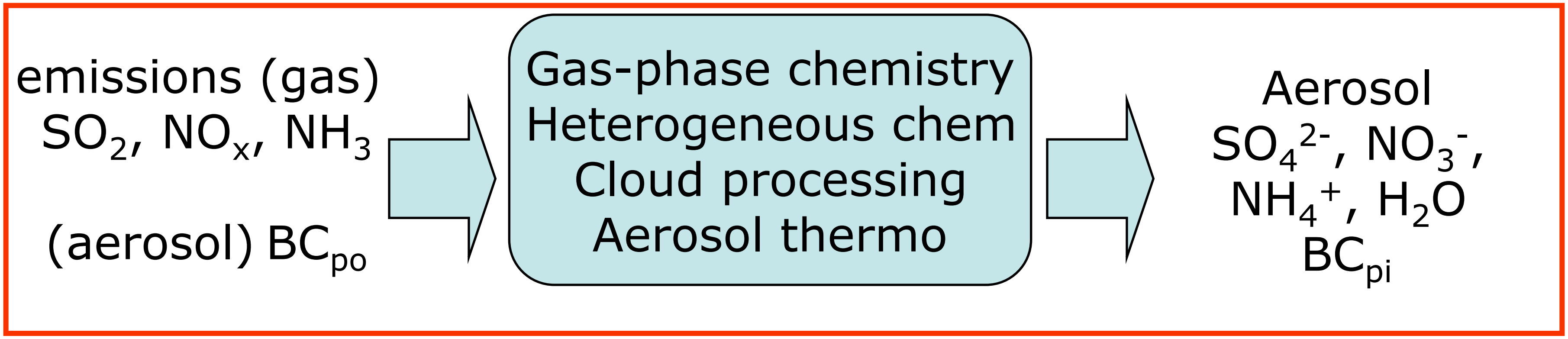
Attainment studies: assessing the effectiveness of emissions changes on an air quality or radiative forcing goal.

GEOS-Chem: Chemical Transport Model

Model Description, v6-02-05 (*Bey et al.*, 2001; *Park et al.*, 2004)

- GEOS-3 Assimilated meteorology
- 4°x5°(Global) resolution, 30 levels
- HO_x - NO_x - HC gas-phase chemistry
- Inorganic aerosol (secondary)
- Carbonaceous aerosol (primary)
- Sea salt
- Dust, SOA





emissions (gas)
SO_2, NO_x, NH_3

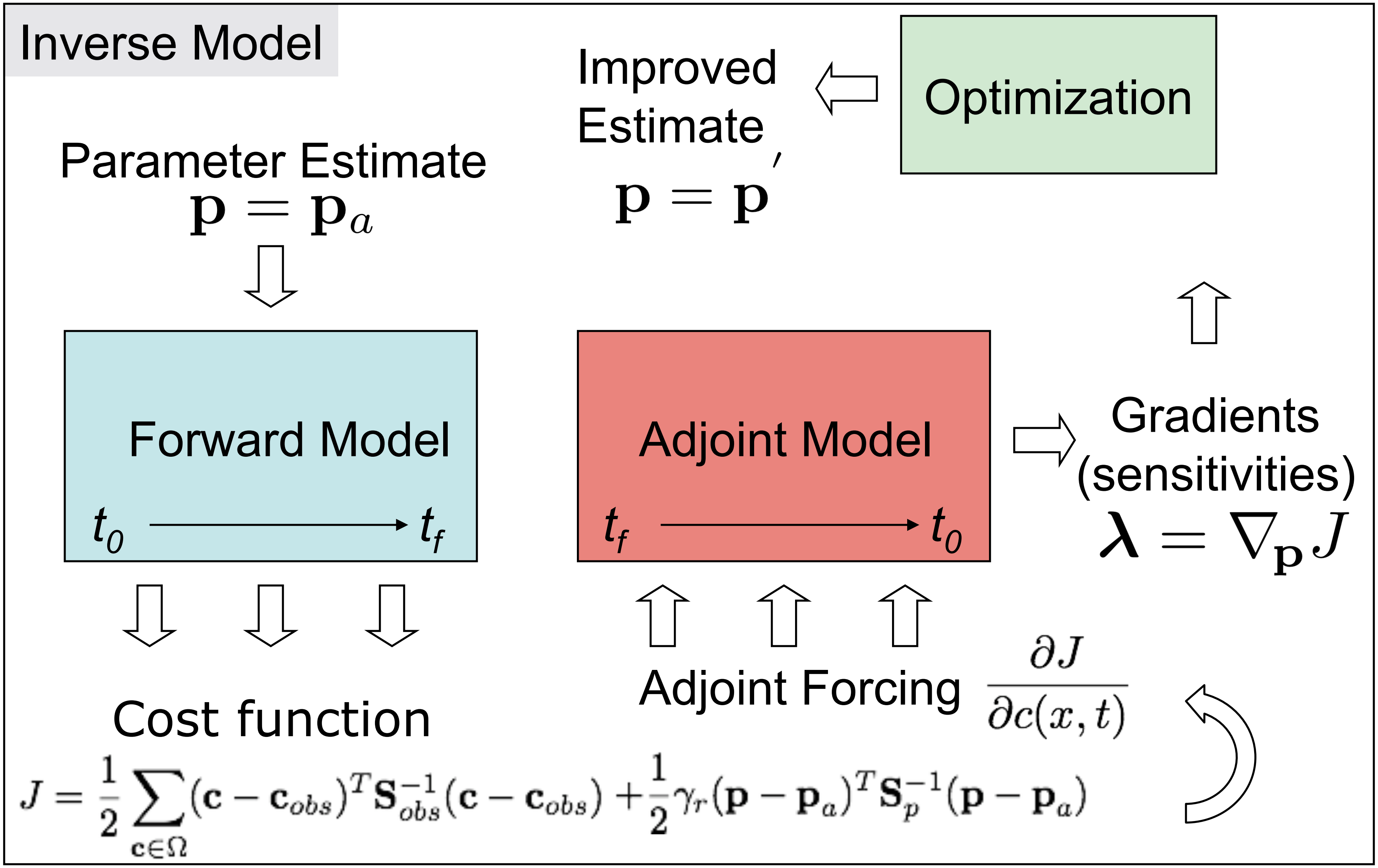
(aerosol) BC_{po}

Gas-phase chemistry
Heterogeneous chem
Cloud processing
Aerosol thermo

Aerosol
SO_4^{2-}, NO_3^-,
NH_4^+, H_2O
BC_{pi}

All included in adjoint of GEOS-Chem (Henze et al., 2007)

Inverse Modeling using Adjoint Model



Inverse Model

Parameter Estimate
$\mathbf{p} = \mathbf{p}_a$

Improved Estimate
$\mathbf{p} = \mathbf{p}'$

Optimization

Forward Model
$t_0 \longrightarrow t_f$

Adjoint Model
$t_f \longrightarrow t_0$

Gradients (sensitivities)
$\boldsymbol{\lambda} = \nabla_{\mathbf{p}} J$

Adjoint Forcing $\frac{\partial J}{\partial c(x,t)}$

Cost function

$$J = \frac{1}{2}\sum_{\mathbf{c}\in\Omega}(\mathbf{c}-\mathbf{c}_{obs})^T\mathbf{S}_{obs}^{-1}(\mathbf{c}-\mathbf{c}_{obs}) + \frac{1}{2}\gamma_r(\mathbf{p}-\mathbf{p}_a)^T\mathbf{S}_p^{-1}(\mathbf{p}-\mathbf{p}_a)$$

Inverse Modeling: 4D-Var

4D Variation Data Assimilation (Kalnay, 2003):

- Optimize parameters at resolution of forward model
- Forward model equations are strong constraints

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Applications with GEOS-Chem adjoint:

- Top down NH_3 emissions estimates using surface obs. of sulfate and nitrate (IMPROVE)

- Top down emissions estimates using remote sensing
    - MOPITT CO (Kopacz et al., 2008)
    - SCIAMACHY SO_2, NO_2 (C. Lee, C. Shim, Q. Li, R. Martin)
    - TES O_3 (K. Singh, A. Sandu, K. Bowman)

- Potential for combining aerosol and gas-phase obs.

Adjoint modeling applications

Depending on “model response,” can be used for:

Sensitivity analysis: quantifying influence of uncertain model parameters (emissions, reaction rates, initial conditions, …)

Inverse modeling (Data Assimilation): using large data sets, optimizing parameters on resolution commensurate with forward model

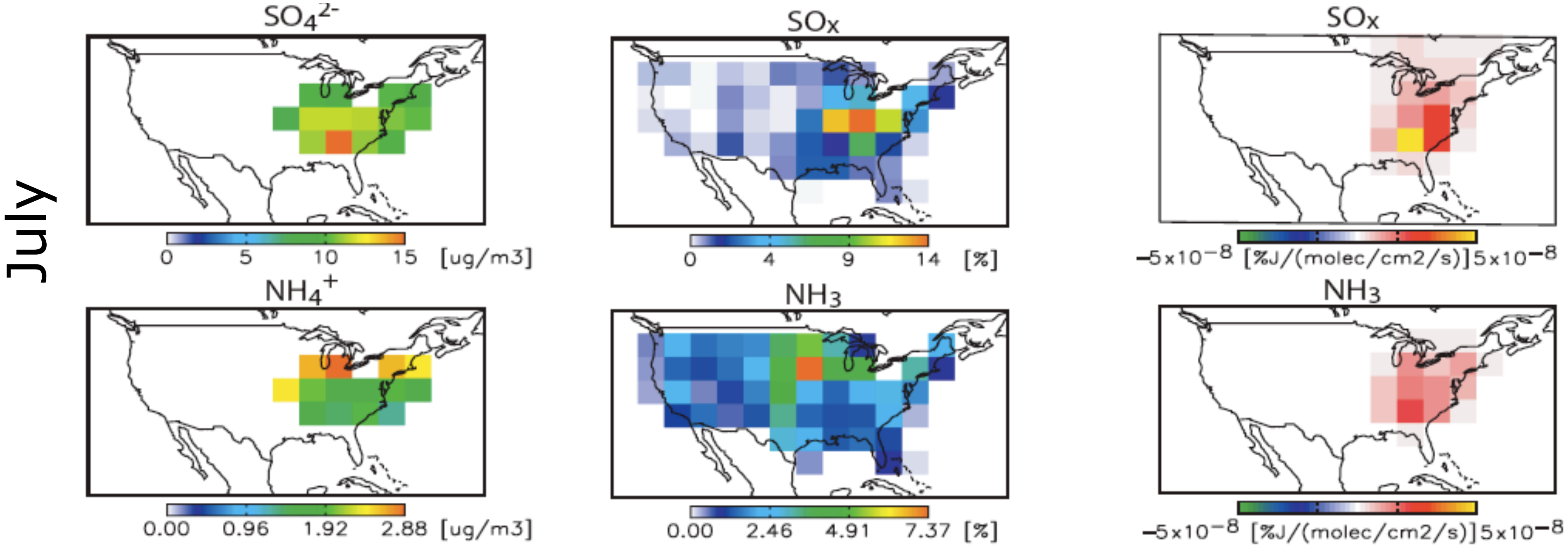
Attainment studies: assessing the effectiveness of emissions abatement on an air quality or radiative forcing goal.

AQ Attainment

Consider metric of $\mathrm{PM}_{2.5}$ air quality,

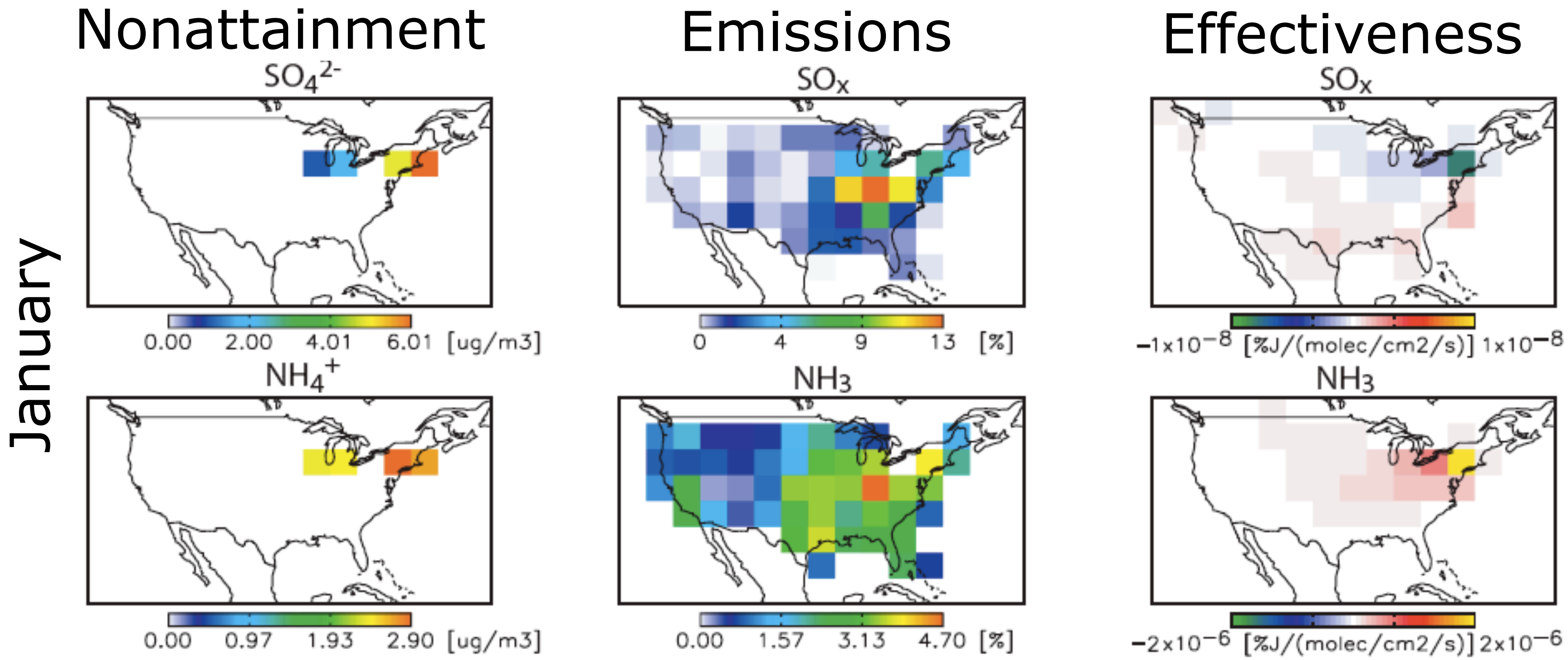
$$J = \frac{1}{2}\sum \mathrm{MAX}[(\mathrm{inorganic\ PM}_{2.5})_{24\mathrm{h}} - 10\mu\mathrm{g/m}^3, 0]^2$$

Nonattainment Emissions Effectiveness



Effectiveness = adjoint sens = $\frac{\partial J}{\partial E} \times \frac{100\%}{J} \approx \frac{\triangle J[\%]}{E[molec/cm2/s]}$

AQ Attainment



Nonattainment

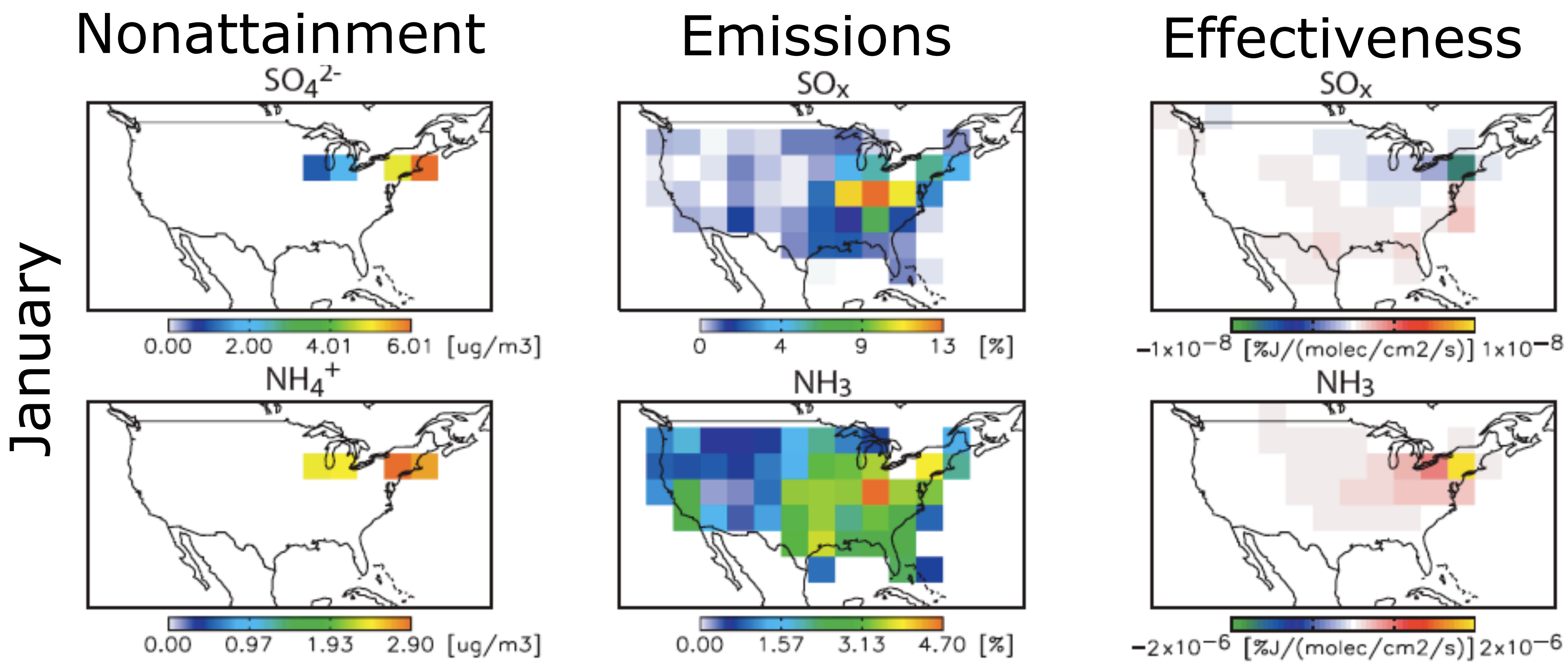
Emissions

Effectiveness

January

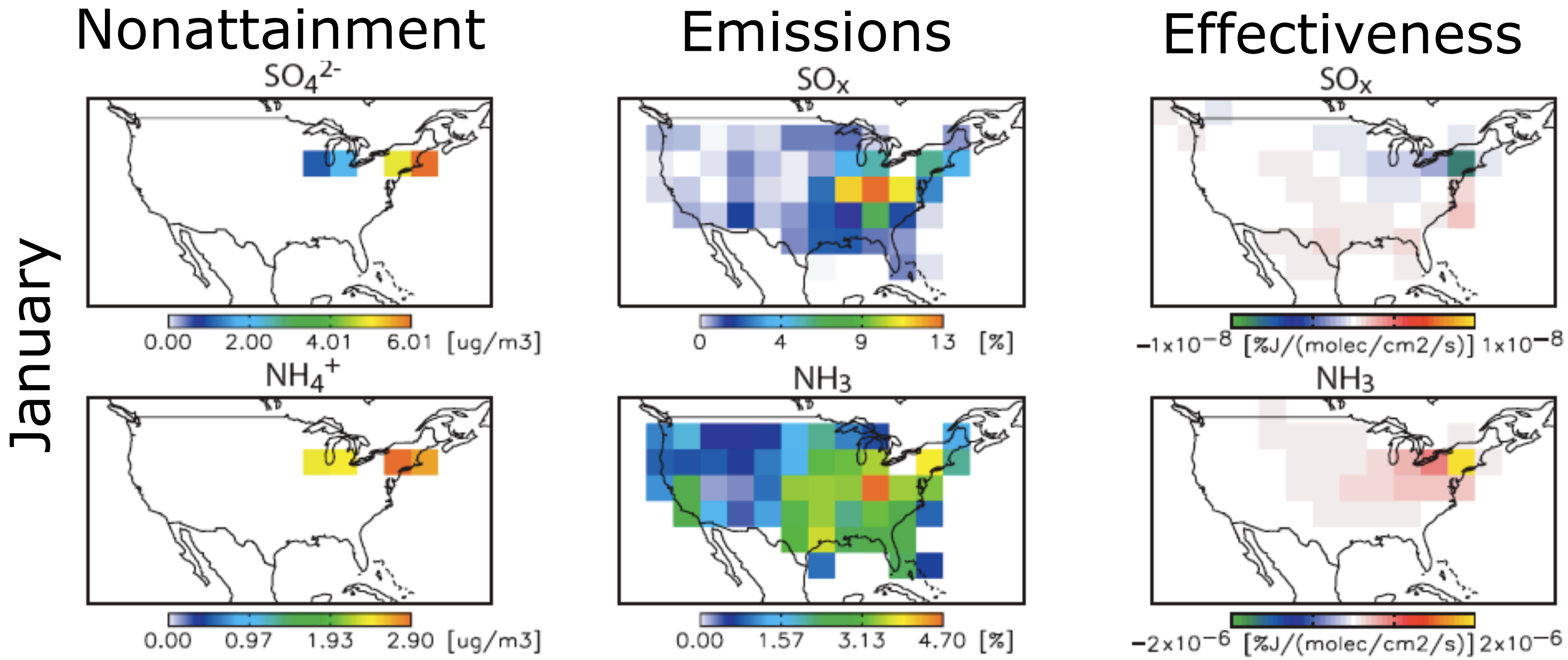
$(NH_4^+)_2SO_4^{2-}$ (mw = 132) → $2NH_4^+NO_3^-$ (mw = 2x80)

AQ Attainment



West et al. 1999; Vayenas et al., 2005; Pinder et al., 2007

AQ Attainment



Effectiveness of emissions controls have critical spatial, seasonal, sector and chemical variability

Final comments on adjoint sensitivities

Disadvantages

- sensitivities ≠ source attribution

Advantages

- Computational efficiency

- No perturbation to forward model
    - sensitivities around current model state
    - relevant for policy (+/- 10-30% Δemission)

- Models can be first conditioned to observations using 4D-Var

- Estimates of emissions influence side-by-side with estimates influence of other parameters (ex: D_{dry})

The end