Uncertainty Reduction in Atmospheric Composition Models by Chemical Data Assimilation

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Information feedback loops between CTMs and observations: data assimilation and targeted meas.

Chemical kinetics

Transport

Meteorology

CTM

Data Assimilation

Optimal analysis state

Observations

Targeted Observ.

Improved:
• forecasts
• science
• field experiment design
• models
• emission estimates

Aerosols

Emissions

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What is data assimilation?

The fusion of information from:
1. prior knowledge,
2. imperfect model predictions, and
3. sparse and noisy data,
to obtain a consistent description of the state of a physical system, such as the atmosphere.

Lars Isaksen (http://www.ecmwf.int)
Source of information #1: The prior encapsulates our current knowledge of the state

- The background (prior) probability density: \( P^b(x) \)
- The current best estimate: apriori (background) state \( x^b \).
- Typical assumption on random background errors

\[
\varepsilon^b = x^b - S(x^{\text{true}}) \in \mathcal{N} (0, B) .
\]

- With many nonlinear models the normality assumption is difficult to justify, but is nevertheless widely used because of its convenience.
Source of information #2: The model encapsulates our knowledge about physical and chemical laws that govern the evolution of the system

- The model evolves an initial state \( x_0 \in \mathbb{R}^n \) to future times

\[
x_i = \mathcal{M}_{t_0 \rightarrow t_i} (x_0).
\]

- Typical size of chemical transport models: \( n \in \mathcal{O}(10^7) \) variables.

- The model is imperfect

\[
\mathcal{S} (x^\text{true}_i) = \mathcal{M}_{t_{i-1} \rightarrow t_i} \cdot \mathcal{S} (x^\text{true}_{i-1}) - \eta_i,
\]

where \( \eta_i \) is the model error in step \( i \).
Source of information #3: The observations are sparse and noisy snapshots of reality

- Measurements $y_i \in \mathbb{R}^m$ ($m \ll n$) taken at times $t_1, \ldots, t_N$

$$y_i = H^t(x_{i \text{true}}) - \varepsilon^\text{instrument} = H(S(x_{i \text{true}})) - \varepsilon^\text{obs}, \quad i = 1, \ldots, N.$$ 

- Observation operators
  - $H^t$: physical space $\rightarrow$ observation space, while
  - $H$: the model space $\rightarrow$ observation space.

- The observation error

$$\varepsilon^\text{obs} = \varepsilon^\text{instrument} + H(S(x_{i \text{true}})) - H^t(x_{i \text{true}})$$

  - $\varepsilon^\text{obs}$ is the observation error.
  - $\varepsilon^\text{instrument}$ is the instrument error.
  - $H(S(x_{i \text{true}})) - H^t(x_{i \text{true}})$ is the representativeness error.

- Typical assumptions:

$$\varepsilon^\text{obs} \in \mathcal{N}(0, R_i); \quad \varepsilon^\text{obs}_i, \varepsilon^\text{obs}_j \quad \text{independent for} \quad t_i \neq t_j.$$
Result of data assimilation: The analysis encapsulates our *enhanced knowledge* of the state

- The analysis (posterior) probability density $P^a(x)$:

  
  Bayes: 
  
  $$P^a(x) = P(x|y) = \frac{P(y|x) \cdot P^b(x)}{P(y)}.$$ 

- The best state estimate $x^a$ is called the aposteriori, or the *analysis*.

- Analysis estimation errors $\varepsilon^a = x^a - S(x^{true})$ characterized by
  - *analysis mean error (bias)* $\beta^a = E^a[\varepsilon^a]$ 
  - *analysis error covariance matrix* 
    
    $$A = E^a \left[ (\varepsilon^a - \beta^a) (\varepsilon^a - \beta^a)^T \right] \in \mathbb{R}^{n \times n}$$

- In the Gaussian, linear case, Bayes posterior admits an analytical solution by Kalman filter formulas
Extended Kalman filter

- The observations are considered successively at times $t_1, \cdots, t_N$.
- The background state at $t_i$ given by the model forecast:

$$x^b_i \equiv x^f_i = M_{t_{i-1} \rightarrow t_i} \cdot x^a_{i-1}.$$ 

- Model is imperfect, but is assumed unbiased

$$\eta_i \in \mathcal{N}(0, Q_i)$$ 

- Model error $\eta_i$ and solution error $\varepsilon^a_{i-1}$ are assumed independent; solution error small, propagated by linearized model $M = M'(x)$

$$O(n^3) : \quad B_i \equiv P^f_i = M_{t_{i-1} \rightarrow t_i} P^a_{i-1} M^T_{t_i \rightarrow t_{i-1}} + Q_i.$$ 

- EKF analysis uses $H_i = H'(x^f_i)$:

$$O(nm) : \quad x^a_i = x^f_i + K_i (y_i - \mathcal{H}(x^f_i))$$

$$O(nm^2 + n^2m + m^3) : \quad K_i = P^f_i H^T_i (H_i P^f_i H^T_i + R_i)^{-1}$$

$$O(n^2m + n^3) : \quad A_i \equiv P^a_i = (I - K_i H_i) P^f_i.$$
Practical Kalman filter methods

- EKF is not practical for very large systems
- Suboptimal KF approximate the covariance matrices e.g.,

\[ B(\ell),(k) = \sigma(\ell) \sigma(k) \exp \left( \text{distance} \{ \text{gridpoint}(\ell), \text{gridpoint}(k) \}^2 / L^2 \right) \]

- Ensemble Kalman filters (EnKF) use a Monte-Carlo approach

\[
\begin{align*}
x_i^f[e] &= \mathcal{M}_{t_i-1 \to t_i} \left( x_{i-1}^a[e] \right) + \eta_i[e], & e = 1, \ldots, E \\
x_i^a[e] &= x_i^f[e] + K_i \left( y_i + \varepsilon_i^{obs}[e] - \mathcal{H}_i(x_i^f[e]) \right), & e = 1, \ldots, E.
\end{align*}
\]

- Error covariances \( P_i^f, P_i^a \) estimated from statistical samples
- EnKF issues: rank-deficiency of the estimated \( P_i^f \)
- EnKF strengths: capture non-linear dynamics, doesn’t need TLM, ADJ, accounts for model errors, almost ideally parallelizable
Maximum aposteriori estimator

Maximum aposteriori estimator (MAP) defined by

$$\mathbf{x}^a = \arg\max_{\mathbf{x}} \mathcal{P}^a(\mathbf{x}) = \arg\min_{\mathbf{x}} \mathcal{J}(\mathbf{x}) , \quad \mathcal{J}(\mathbf{x}) = -\ln \mathcal{P}^a(\mathbf{x}) .$$

Using Bayes and assumptions for background, observation errors:

$$\mathcal{J}(\mathbf{x}) = -\ln \mathcal{P}^a(\mathbf{x}) = -\ln \mathcal{P}^b(\mathbf{x}) - \ln \mathcal{P}(\mathbf{y}|\mathbf{x}) + \text{const}$$

$$\approx \frac{1}{2} (\mathbf{x} - \mathbf{x}^b)^T \mathbf{B}^{-1} (\mathbf{x} - \mathbf{x}^b) + \frac{1}{2} (\mathcal{H}(\mathbf{x}) - \mathbf{y})^T \mathbf{R}^{-1} (\mathcal{H}(\mathbf{x}) - \mathbf{y})$$

Optimization by gradient-based numerical procedure

$$\nabla_{\mathbf{x}} \mathcal{J}(\mathbf{x}^a) = \mathbf{B}^{-1} (\mathbf{x}^a - \mathbf{x}^b) + \mathbf{H}^T \mathbf{R} (\mathcal{H}(\mathbf{x}^a) - \mathbf{y}) ; \quad \mathbf{H} = \mathcal{H}(\mathbf{x}^b) .$$

Hessian of cost function approximates inverse analysis covariance

$$\nabla_{\mathbf{x}, \mathbf{x}}^2 \mathcal{J} = \mathbf{B}^{-1} + \mathbf{H}^T \mathbf{R}^{-1} \mathbf{H} \approx \mathbf{A}^{-1} .$$
Four dimensional variational data assimilation (4D-Var) I

- All observations at all times $t_1, \cdots, t_N$ are considered simultaneously
- The control variables (parameters $p$, initial conditions $x_0$, boundary conditions, etc) uniquely determine the state of the system at all future times
- 4D-Var MAP estimate via model-constrained optimization problem

\[
\mathcal{J} (x_0) = \frac{1}{2} \left\| x_0 - x_0^b \right\|_{B_0}^2 + \frac{1}{2} \sum_{i=1}^{N} \left\| \mathcal{H}(x_i) - y_i \right\|_{R_i}^2
\]

\[
x_0^a = \text{arg min} \mathcal{J} (x_0)
\]

subject to: $x_i = \mathcal{M}_{t_0 \rightarrow t_i} (x_0)$, $i = 1, \cdots, N$

- Formulation can be easily extended to other model parameters
Four dimensional variational data assimilation (4D-Var) II

- The large scale optimization problem is solved in a reduced space using a gradient-based technique.
- The 4D-Var gradient reads

\[
\nabla J_{x_0} (x_0) = B_0^{-1} (x_0 - x_0^b) + \sum_{i=1}^{N} \left( \frac{\partial x_i}{\partial x_0} \right)^T H_i^T R_i^{-1} (H(x_i) - y_i)
\]

- Needs linearized observation operators \( H_i = H'(x_i) \)
- Needs the transposed sensitivity matrix \( (\partial x_i/\partial x_0)^T \in \mathbb{R}^{n\times n} \)
- Adjoint models efficiently compute the transposed sensitivity matrix times vector products
- The construction of an adjoint model is a nontrivial task.
Correct models of background errors are of great importance for data assimilation

- Background error representation determines the spread of information, and impacts the assimilation results
- Needs: high rank, capture dynamic dependencies, efficient computations
- Traditionally estimated empirically (NMC, Hollingsworth-Lonnberg)

1. Tensor products of 1d correlations, decreasing with distance (Singh et al, 2010)
2. Multilateral AR model of background errors based on “monotonic TLM discretizations” (Constantinescu et al 2007)
3. Hybrid methods in the context of 4D-Var (Cheng et al, 2007)
What is the effect of mis-specification of inputs?

(Daescu, 2008) Consider a verification functional $\Psi(x_v^a)$ defined on the optimal solution at a future time $t_v$. $\Psi$ is a measure of the forecast error. What is the impact of small errors in the specification of covariances, background, and observation data?

\[
\nabla_{y_i} \Psi = R_i^{-1} H M_{t_0 \rightarrow t_i} \left( \left( \nabla^2_{x_0,x_0} J \right)^{-1} \nabla_{x_0} \Psi \right)
\]

\[
\nabla_{R_i(:)} \Psi = \left( R_i^{-1} (H(x_i^a) - y_i) \right) \otimes \nabla_{y_i} \Psi
\]

\[
\nabla_{x^b} \Psi = B_0^{-1} \left( \left( \nabla^2_{x_0,x_0} J \right)^{-1} \nabla_{x_0} \Psi \right)
\]

\[
\nabla_{B_0(:)} \Psi = \left( B_0^{-1} (x_0^a - x_0^b) \right) \otimes \nabla_{x^b} \Psi
\]
General framework for sensitivity analysis

Forward model equations link parameters and solutions:

\[ \mathcal{F}(x, \theta) = 0 \in \mathcal{H}_F \quad \mathcal{H}_F = \text{model constraint space, Hilbert: } \langle \cdot, \cdot \rangle_{\mathcal{H}_F} \]
\[ x \in \mathcal{H}_x \quad \mathcal{H}_x = \text{model state space, Hilbert: } \langle \cdot, \cdot \rangle_{\mathcal{H}_x} \]
\[ \theta \in \mathcal{H}_\theta \quad \mathcal{H}_\theta = \text{parameter space, Hilbert: } \langle \cdot, \cdot \rangle_{\mathcal{H}_\theta} \]

The response functional (QoI) associates a real value to each state

\[ \mathcal{I}(x) : \mathcal{H}_x \rightarrow \mathbb{R} \quad \left( \text{e.g., } \mathcal{I}(x) = \frac{1}{2} \| \mathcal{H}(x) - y \|^2_{\mathbb{R}^{-1}} \right) \]

Assumptions:

1. \( \mathcal{F}, \mathcal{I} \) are continuously Fréchet differentiable.
2. \( \mathcal{F}_x \) has a continuous linear inverse mapping. By IFT a Fréchet differentiable model solution operator \( x = \mathcal{M}(\theta) \) exists locally

\[ \mathcal{M} : \mathcal{H}_\theta \rightarrow \mathcal{H}_x \quad x = \mathcal{M}(\theta) \quad \mathcal{M}'(\theta) = -\mathcal{F}_x^{-1}(x, \theta) \cdot \mathcal{F}_\theta(x, \theta) \]
Formulation of the inverse problem as a model-constrained optimization problem

Find the optimal vector of parameters $\theta_{\text{opt}}$ such that:

$$
\theta^* = \arg \min_{\theta} \mathcal{J}(x)
$$

subject to $\mathcal{F}(x, \theta) = 0$.

Comments.

1. The cost function depends implicitly on the parameters:

$$
\mathcal{J}(x) = \mathcal{J}(\mathcal{M}(\theta)) = (\mathcal{J} \circ \mathcal{M})(\theta).
$$

2. Gradient-based optimization techniques require

$$
\nabla_{\theta} \mathcal{J} = \mathcal{M}'^*(\theta) \cdot \nabla_x \mathcal{J}
$$

3. Difficulty: model solution operator is only defined implicitly.
Direct (forward) vs. adjoint sensitivity analysis

1. **Tangent linear model** is obtained by Frèchet differentiation

\[
(TLM) : \quad F_\theta(\theta, x) \cdot \delta \theta + F_x(\theta, x) \cdot \delta x = 0 \in \mathcal{H}_F.
\]

\[
\delta \mathcal{J} = \langle \nabla_x \mathcal{J}, \delta x \rangle_{H_x} = \langle \nabla_\theta \mathcal{J}, \delta \theta \rangle_{H_\theta}.
\]

**Comment.** \( \nabla_x \mathcal{J} \) by direct differentiation. One TLM solution provides one inner product. To find the entire gradient \( \nabla_\theta \mathcal{J} \) ...
Direct (forward) vs. adjoint sensitivity analysis

1. Tangent linear model is obtained by Fréchet differentiation

\[
(\text{TLM}) : \quad \mathcal{F}_\theta(\theta, x) \cdot \delta \theta + \mathcal{F}_x(\theta, x) \cdot \delta x = 0 \in \mathcal{H}_F.
\]

\[
\delta \mathcal{I} = \langle \nabla_x \mathcal{I}, \delta x \rangle_{\mathcal{H}_x} = \langle \nabla_{\theta} \mathcal{I}, \delta \theta \rangle_{\mathcal{H}_\theta}.
\]

Comment. \n\n\n\n\n2. Adjoint model obtained using duality:

\[
(\lambda \in \mathcal{H}_F^* \equiv \mathcal{H}_F) \iff \langle \lambda, \mathcal{F}_x \cdot \delta x \rangle_{\mathcal{H}_F} + \langle \lambda, \mathcal{F}_\theta \cdot \delta \theta \rangle_{\mathcal{H}_F} = 0 \in \mathbb{R} \quad \text{(by adjoint)}
\]

\[
\iff \langle \mathcal{F}_x^* \cdot \lambda, \delta x \rangle_{\mathcal{H}_x} + \langle \mathcal{F}_\theta^* \cdot \lambda, \delta \theta \rangle_{\mathcal{H}_\theta} = 0 \in \mathbb{R}.
\]

\[
\text{ADJ} : \quad \mathcal{F}_x^* \cdot \lambda = -\nabla_x \mathcal{I}(x).
\]

\[
\langle \nabla_x \mathcal{I}(x), \delta x \rangle_{\mathcal{H}_x} = \langle (\mathcal{F}_\theta)^* \cdot \lambda, \delta \theta \rangle_{\mathcal{H}_\theta} = \langle \nabla_{\theta} \mathcal{I}, \delta \theta \rangle_{\mathcal{H}_\theta} = \delta \mathcal{I}.
\]

Comment. Adjoint model does not depend on the particular perturbations \(\delta \theta, \delta x\), and needs to be solved only once.
Continuous and discrete adjoints of mass balance equations lead to different computational models

\[ \nabla y_0 \psi = \cdots + \sum_{k=1}^{N} \left( \partial y^k / \partial y^0 \right)^T \left( H_k \right)^T R_k^{-1} \left( H_k y^k - z_{obs} \right) \]

**Continuous forward model**

\[ \frac{dC_i}{dt} = -\bar{u} \cdot \nabla C_i + \frac{1}{\rho} \nabla (\rho K \cdot \nabla C_i) + \frac{1}{\rho} f_i(\rho C) + E_i \]

\[ C_i(t^0, x) = C_i^0(x), \quad t^0 \leq t \leq t^F \]

\[ C_i(t, x) = C_i^{IN}(t, x) \quad \text{on} \quad \Gamma^{IN} \]

\[ K \frac{\partial C_i}{\partial n} = 0 \quad \text{on} \quad \Gamma^{OUT} \]

\[ K \frac{\partial C_i}{\partial n} = V_i^{DEP} C_i - Q_i \quad \text{on} \quad \Gamma^{GROUND} \]

**Continuous adjoint model**

\[ \frac{d\lambda_i}{dt} = -\nabla \cdot (\bar{u} \lambda_i) - \nabla \left( \frac{\rho K \cdot \nabla \lambda_i}{\rho} \right) - \left( F^T (\rho C) \cdot \lambda_i - \phi_i \right) \]

\[ \lambda_i(t^F, x) = \lambda_i^F(x), \quad t^F \geq t \geq t^0 \]

\[ \lambda_i(t, x) = 0 \quad \text{on} \quad \Gamma^{IN} \]

\[ \bar{u} \lambda_i + \rho K \frac{\partial (\lambda_i / \rho)}{\partial n} = 0 \quad \text{on} \quad \Gamma^{OUT} \]

\[ \rho K \frac{\partial (\lambda_i / \rho)}{\partial n} = V_i^{DEP} \lambda_i \quad \text{on} \quad \Gamma^{GROUND} \]

**Discrete forward model**

\[ C^{k+1} = N_{[t,t+\Delta t]} \circ C^k \]

\[ N_{[t,t+\Delta t]} = T_{HOR}^{\Delta t} \circ T_{VERT}^{\Delta t} \circ R_{CHEM}^{\Delta t} \circ T_{VERT}^{\Delta t} \circ T_{HOR}^{\Delta t} \]

**Discrete adjoint model**

\[ \lambda^k = N_{[t,t+\Delta t]}^{*} \circ \lambda^{k+1} + \phi^{k+1} \]

\[ N_{[t,t+\Delta t]}^{*} = \left( T_{HOR}^{\Delta t} \right)^{*} \circ \left( T_{VERT}^{\Delta t} \right)^{*} \circ \left( R_{CHEM}^{\Delta t} \right)^{*} \circ \left( T_{VERT}^{\Delta t} \right)^{*} \circ \left( T_{HOR}^{\Delta t} \right)^{*} \]

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Discrete adjoints of advection numerical schemes can become inconsistent with the adjoint PDE

Change of forward scheme pattern:
- Change of upwinding
- Sources/sinks
- Inflow boundaries scheme

Example: 3\textsuperscript{rd} order upwind FD

Active forward limiters act as pseudo-sources in adjoint
Example: vminmod

\[ [\text{Liu and Sandu, 2005}] \]
Discrete Runge-Kutta adjoints can be regarded as “numerical methods” applied to the adjoint ODE

\[ y^{n+1} = y^n + h \sum_{i=1}^{s} b_i f(Y^i), \]

\[ Y^i = y^n + h \sum_{i=1}^{s} a_{i,j} f(Y^j) \]

**RK Method**

\[ \lambda^n = \lambda^{n+1} + \sum_{i=1}^{s} \theta^i \]

\[ \theta^i = h J^T(Y^i) \cdot \begin{bmatrix} b_i \lambda^{n+1} + \sum_{j=1}^{s} a_{j,i} \theta^j \end{bmatrix} \]

**Discrete RK Adjoint**

[Hager, 2000]
Discrete Runge-Kutta adjoints: error analysis

Local error analysis: The discrete adjoint of RK method of order $p$ is an order $p$ discretization of the adjoint equation. [Sandu, 2005]. This:
- works for both explicit and implicit methods
- true for arbitrary orders $p$

Global error analysis: The discrete adjoint (of a RK method convergent with order $p$) converges with order $p$ to the solution of the adjoint ODE. [Sandu, 2005] The analysis accounts for:
1. the truncation error at each step, and
2. the different trajectories about which the continuous and the discrete adjoints are defined

Stiff case: Consider a stiffly accurate Runge Kutta method of order $p$ with invertible coefficient matrix $A$. The discrete adjoint provides:
1. an order $p$ discretization of the adjoint of nonstiff variable
2. an order $\min(p,q+1,r+1)$ of the adjoint of stiff variable

[Sandu, 2005]
Properties of discrete adjoint LMM

1. For **fixed step sizes**
   - the discrete adjoint starting and ending steps, in general, are not consistent approximations of the adjoint ODE
   - the adjoint LMM is (at least) first order consistent with the adjoint ODE

2. For **variable step sizes** the adjoint LMM is not a consistent discretization of the adjoint ODE

3. The discrete **adjoint variable at the initial time** is an order p approximation of the continuous adjoint, where p is the order of the (forward) LMM method.

   [Sandu, 2007]
Uncertainty quantification using polynomial chaos and the STEM model

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ground emissions NOx (NO, NO2)</td>
<td>±20%</td>
</tr>
<tr>
<td>Ground emissions AVOC (HCHO, ALK, OLE, ARO)</td>
<td>±50%</td>
</tr>
<tr>
<td>Ground emissions BVOC (ISOPRENE, TERPENE, ETHENE)</td>
<td>±40%</td>
</tr>
<tr>
<td>Deposition velocity O3</td>
<td>±50%</td>
</tr>
<tr>
<td>Deposition velocity NO2</td>
<td>±50%</td>
</tr>
<tr>
<td>West Dirichlet B.C. O3</td>
<td>±5%</td>
</tr>
<tr>
<td>West Dirichlet B.C. PAN</td>
<td>±5%</td>
</tr>
</tbody>
</table>

Data assimilation. The Bayesian framework. UQ/UA for STEM [23/25]
Uncertainty apportionment with the STEM model

**Figure:** Top: New York. Bottom: Boston. 48 hrs ozone mean, standard deviation, and uncertainty (variance) apportionment.
Quantification of the probability of non-compliance with the NAAQS ozone maximum admissible levels

Figure: Boston 8hrs average ozone PDF shows a 68% probability of exceeding the maximum admissible level of 75 ppbv.
Ensemble-based chemical data assimilation is an alternative to variational techniques.

Chemical kinetics

Transport
Meteorology

CTM

Observations

Optimal analysis state

Targeted Observ.

Ensemble Data Assimilation

Improved:
• forecasts
• science
• field experiment design
• models
• emission estimates

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The Ensemble Kalman Filter (EnKF) popular in NWP but not extensively used before with CTMs

\[
y_{f}^{k} = M(i^{k-1}, y_{a}^{k-1})
\]

\[
y_{a}^{k} = y_{f}^{k} + P_{f}^{k} H_{k}^{T} \left( R_{k} + H_{k} P_{f}^{k} H_{k}^{T} \right)^{-1} \left( z_{obs}^{k} - H_{k} y_{f}^{k} \right)
\]

Specify initial ensemble (sample B)

Covariance inflation: Prevents filter divergence (additive, multiplicative, model-specific)

Covariance localization (limit long-distance spurious correlations)

Correction localization (limit increments away from observations)

[Constantinescu et al., 2007]

Ozonesonde S2 (18 EDT, July 20, 2004)
Ground level ozone at 2pm EDT, July 20, 2004, better matches observations after LEnKF data assimilation.

Observations: circles, color coded by O$_3$ mixing ratio

Forecast ($R^2=0.24/0.28$)  
Analysis ($R^2=0.88/0.32$)
The use of adjoints in large scale simulations: atmospheric chemical transport models

Optimal analysis state

Chemical kinetics

Transport Meteorology

CTM

4D-Var Data Assimilation

Observations

Targeted Observ.

Improved:
- forecasts
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- field experiment design
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- emission estimates

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Adjoint sensitivity analysis of non-attainment metrics can help guide policy decisions

Estimated contributions by state to violating U.S. ozone NAAQS in July 2004

[Hakami et al., 2005]
STEM: Assimilation adjusts $\text{O}_3$ predictions considerably at 4pm EDT on July 20, 2004

Observations: circles, color coded by $\text{O}_3$ mixing ratio

Ground $\text{O}_3$ (forecast)  

Ground $\text{O}_3$ (analysis)

[Chai et al., 2006]
Assimilation of elevated observations for July 20, 2004

NOAA P3 flight observations

Ozonesonde observations (Rhode Island)
The inversion procedure can be extended to emissions, boundary conditions, etc.

Texas: 4am CST July 16 to 8pm CST on July 17, 2004.

[Schiamacy, Aug. 4, 2011. IFIP UQ Workshop, Boulder, CO]

[NO2 emission corrections]

[O3 AirNow]

[NO2 Schiamacy]

[HCHO emission corrections]

[Zhang, Sandu et al., 2006]
Smallest Hessian eigenvalues (vectors) approximate the principal a posteriori error components

\[
\left( \nabla^2_{y^0, y^0} \Psi \right)^{-1} \approx \text{cov}(y^{opt})
\]

(a) 3D view (5ppb)
Assimilation of TES ozone column observations, August 2006. Lobatto-IIIC integrates stiff chemistry.

TES is one of four instruments on the NASA EOS Aura platform, launched July 14 2004.
Quality of TES ozone column data assimilation results for several methods (August 1-15, 2006)

[Singh, Sandu et. al., 2010]
Dynamic integration of chemical data and atmospheric models is an important, growing field

- the tools needed for 4D-Var chemical data assimilation are in place:
  - (adjoints for stiff systems, aerosols, transport; singular vectors, parallelization and multi-level checkpointing schemes, models of background errors)
- all algorithms are on a solid theoretical basis
- the ensemble filter methods show promise
- STEM, CMAQ, GEOS-CHEM have been endowed with data assimilation capabilities
- the tool strengths have been demonstrated using real (field campaign) data; ambitious science projects are ongoing