Progress in Inverse Modeling of Aerosols Using the Adjoint Method

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QuickTime?and a
TIFF (Uncompressed) decompressor
are needed to see this picture.

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Goal

Provide improved estimates of the magnitude and the variability (spatial and temporal) of emissions of aerosols and aerosol precursors by performing a 4-D variational assimilation study in which aerosol measurements (surface, satellite) are used as input for an adjoint model of a large scale CTM.

Presentation Overview

Adjoint models of aerosols

- Detailed adjoint of growth due to condensation / evaporation
- Hybrid model (forward and reverse sensitivities)
- Adjoint of gas - aerosol equilibrium

Adjoint CTMs

- Fitting adjoint aerosol routines into CTMs
- Which pieces to work on next

Adjoint of Cond / Evap

Test System

- Three components

  Species 1 : fine mode, condensing
  Species 2 : coarse mode, evaporating
  Species 3 : broad mode, not evaporating or condensing.

- Distribution discretized into 8 bins

  D_p min = 0.039 μm
  D_p max = 10.0 μm

Adjoint of Cond / Evap

Forward Model: evolution due to condensation / evaporation of an aerosol distribution governed by the aerosol dynamics equation

$$\frac{\partial p_i}{\partial t} = H_i p - \frac{1}{3}\frac{\partial}{\partial \mu}[H p_i]$$

where $p_i = p_i(\mu,t)$ is the mass distribution of the ith species
$H_i = H_i(p_1, \ldots, p_Q; \mu, t)$ is the growth rate for the ith species
$H = \sum_{i=1}^{Q} H_i, \quad p = \sum_{i=1}^{Q} p_i$

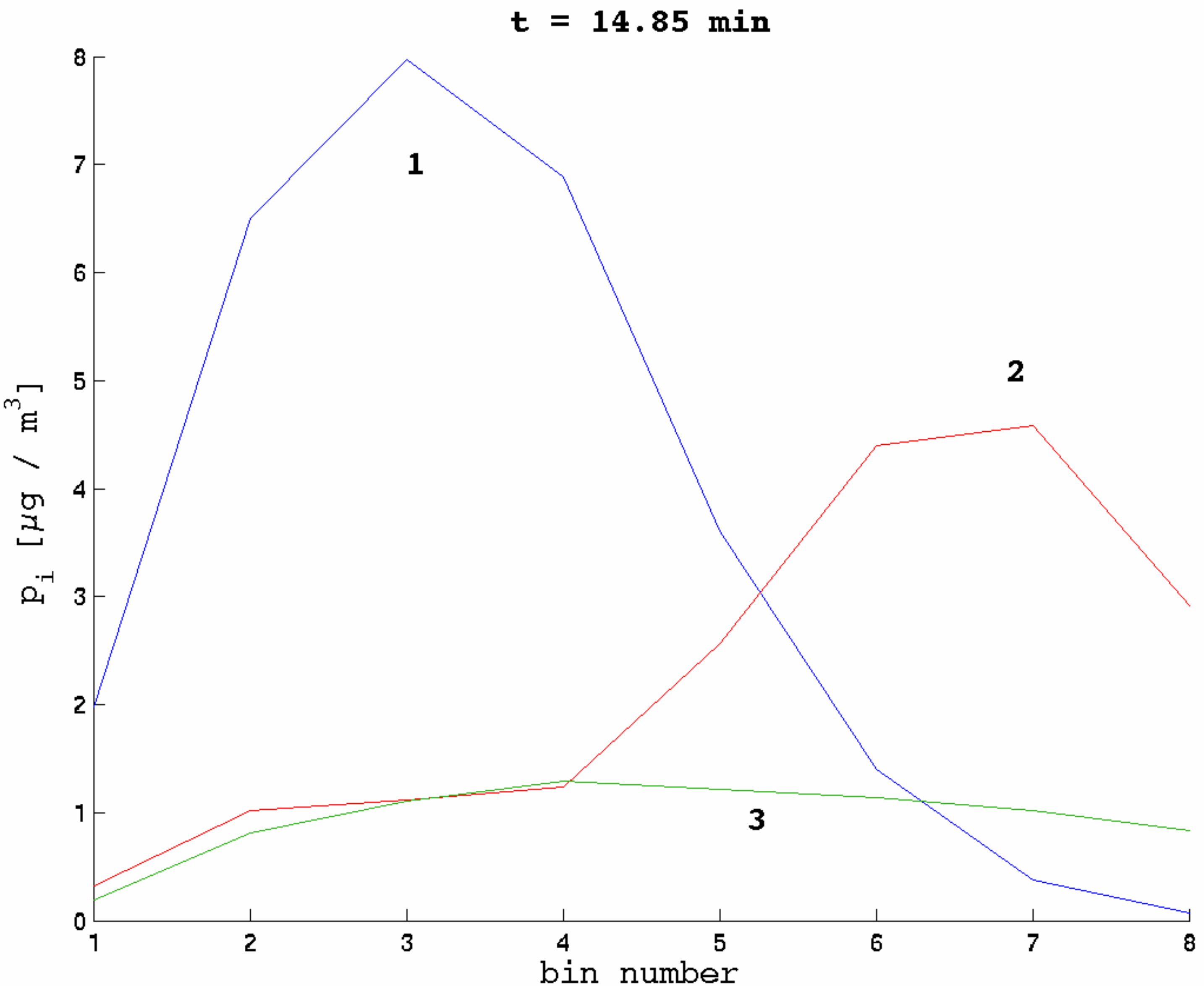
with "ideal" growth rate

$$H_i = \frac{2\pi D_p D_i}{m\left(1 + \frac{2\lambda}{\alpha_i D_p}\right)}(g_i - x_i c_i^\circ)$$

where g_i = gas phase concentration of the ith species
c_i^0 = pure component surface vapor concentration of the ith species

Adjoint of Cond / Evap

Forward Model



Adjoint of Cond / Evap
Inverse Modeling Problem

Goals

- Recover the initial distribution
- Recover the pure component vapor concentrations, c_i^0

Implementation

- Compare continuous vs discrete formulation
- Attempt inversion with sparse observations
  (time, size space, chemical resolution)

Adjoint of Cond / Evap

Continuous Adjoint Model

Take variation of augmented cost function

$$\delta\mathcal{J} = \int_{t^0}^{T}\int_0^{\infty}\sum_{i=1}^{n}\frac{\partial J_0}{\partial p_i}\delta p_i(\mu,t)\,d\mu\,dt - \int_{t^0}^{T}\int_0^{\infty}\sum_{i=1}^{n}\delta\lambda_i(\mu,t)(LHS_{p_i} - RHS_{p_i})\,d\mu\,dt$$

$$-\int_{t^0}^{T}\int_0^{\infty}\sum_{i=1}^{n}\lambda_i(\mu,t)\delta(LHS_{p_i} - RHS_{p_i})\,d\mu\,dt$$

Collected remaining terms yields adjoint equation

$$\frac{\partial\lambda_i}{\partial t} = -\sum_{j=1}^{n}\lambda_j H_j - p\sum_{j=1}^{n}\lambda_j\frac{\partial H_j}{\partial p_i} - \frac{1}{3}\sum_{j=1}^{n}p_j\frac{\partial\lambda_j}{\partial\mu}\frac{\partial H}{\partial p_i} - \frac{H}{3}\frac{\partial\lambda_i}{\partial\mu} - \frac{\partial J_0}{\partial p_i}$$

Solve this numerically from $t = T$ to $t = t^0$

$$\lambda_i(\mu,T) = 0$$
$$\lambda_i(\mu,t^0) = \nabla_{p_i^0}\mathcal{J}$$

Adjoint of Cond / Evap

Discrete Adjoint Model

Discretize the governing equation

$$[p_i]_j^k = F_j(p_i^{k-1}, g_i^{k-1})$$

$k = 1, \ldots, N$ = # time steps
$i = 1, \ldots, Q$ = # species
$j = 1, \ldots, S$ = # of size bins
$[p_i]_j^k$ = particulate concentration of species i in bin j at time step k
p^k = vector of all particulate concentrations
g^k = vector of all gas concentrations

Express the desired gradient using the chain rule

$$\nabla_{p^0} J = \left[\frac{\partial p^1}{\partial p^0}\right]^T \left[\frac{\partial p^2}{\partial p^1}\right]^T \cdots \left[\frac{\partial p^N}{\partial p^{N-1}}\right]^T \left[\frac{\partial J(p^N)}{\partial p^N}\right]$$

Adjoint of Cond / Evap

Discrete Adjoint Model

Define the adjoint variable λ as

$$\lambda^k = \left[\frac{\partial p^N}{\partial p^k}\right]^T \left[\frac{\partial J(p^N)}{\partial p^N}\right] = \nabla_{p^k} J$$

Calculate the desired gradient using the iterative loop

DO k = N, 1, -1

$$\lambda^{k-1} = \left[\frac{\partial p^k}{\partial p^{k-1}}\right]^T \lambda^k$$

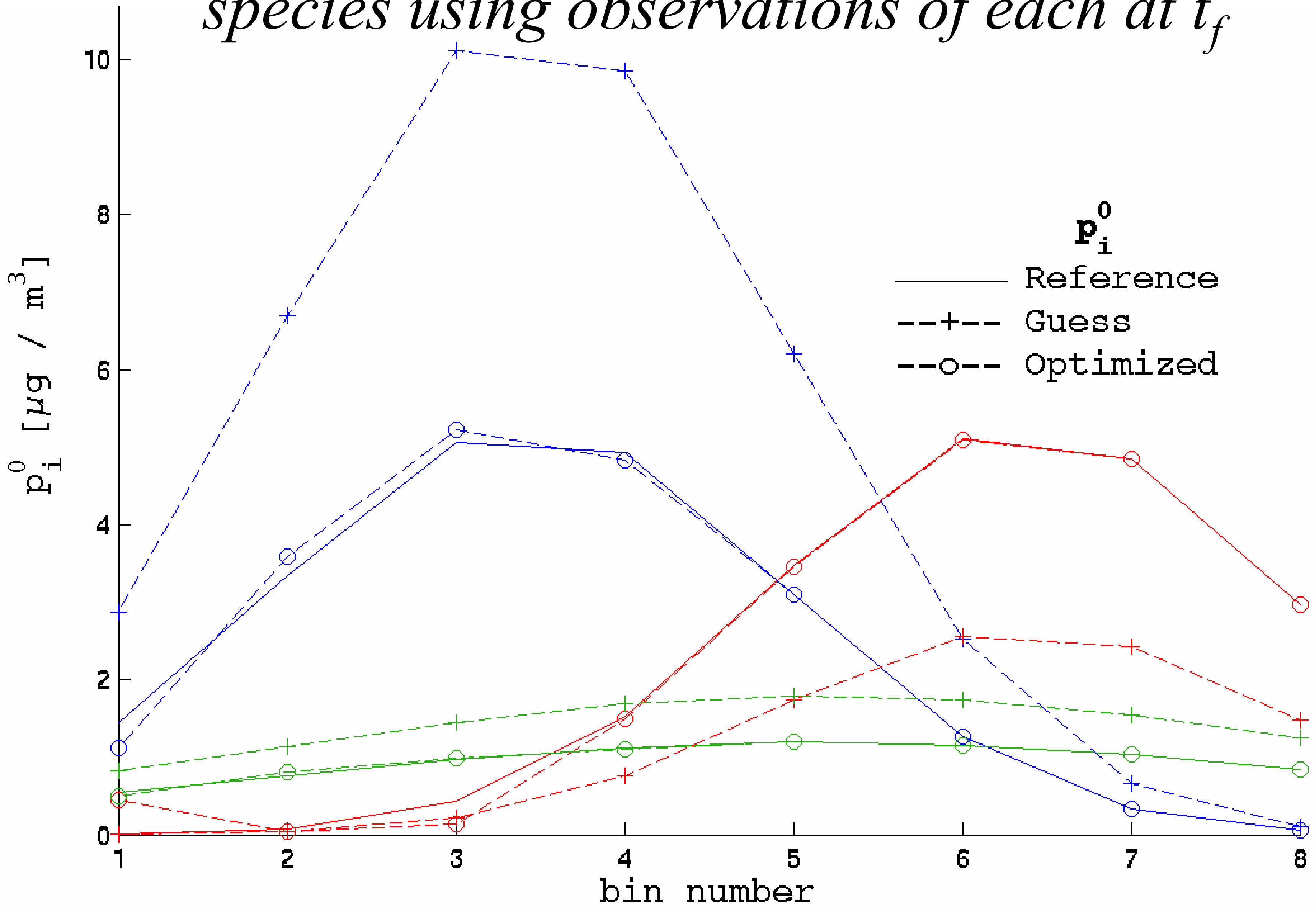
ENDDO

where $\left[\frac{\partial p^k}{\partial p^{k-1}}\right]^T$ is found at each step using TAMC[1]

[1] Giering, R., and T. Kaminski, Recipes for Adjoint code Construction, *ACM Transactions on Mathematical Software*, *24* (4), 437, 1998.

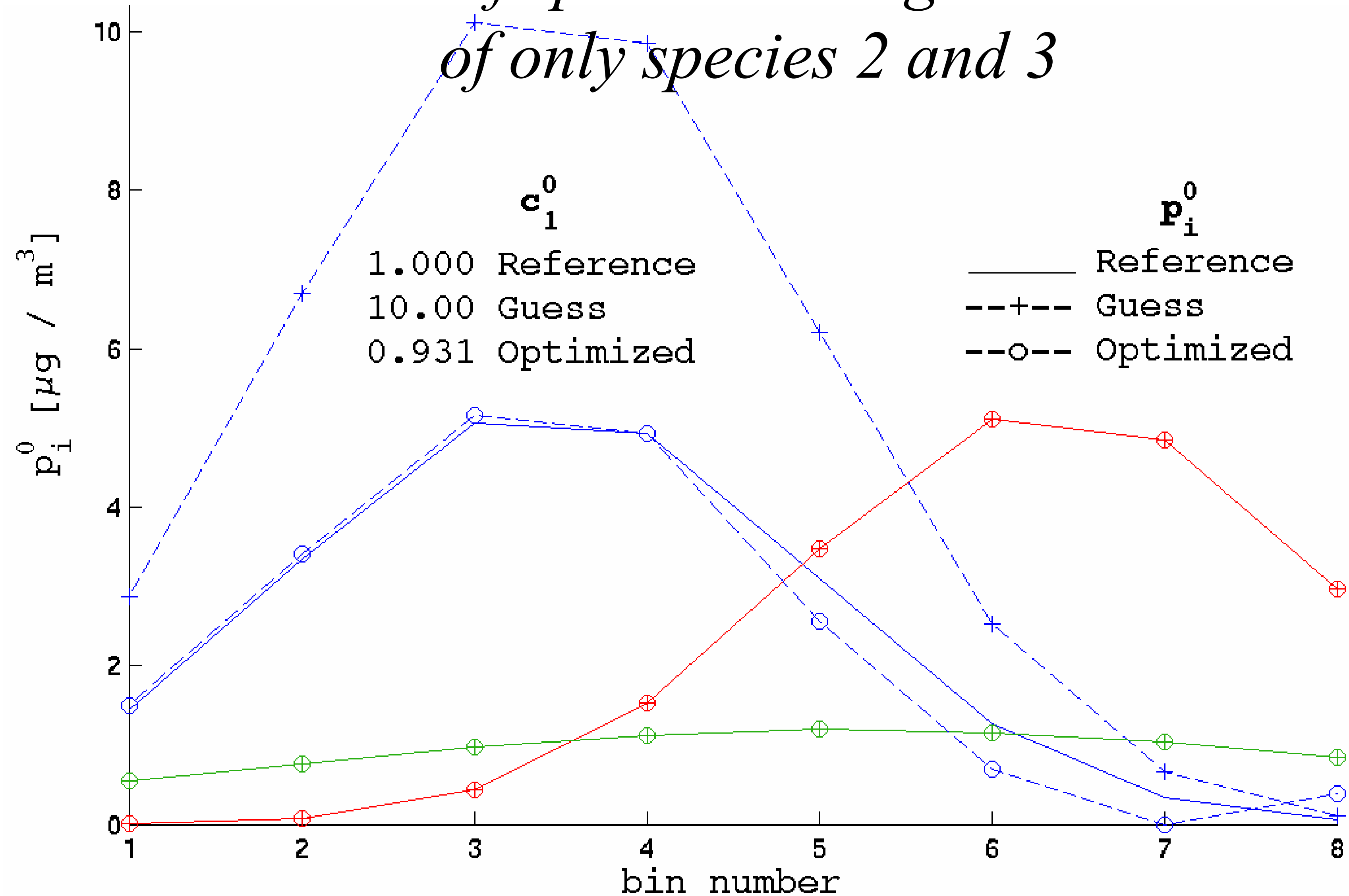
Adjoint of Cond / Evap

Recovering initial distributions of all three species using observations of each at t_f



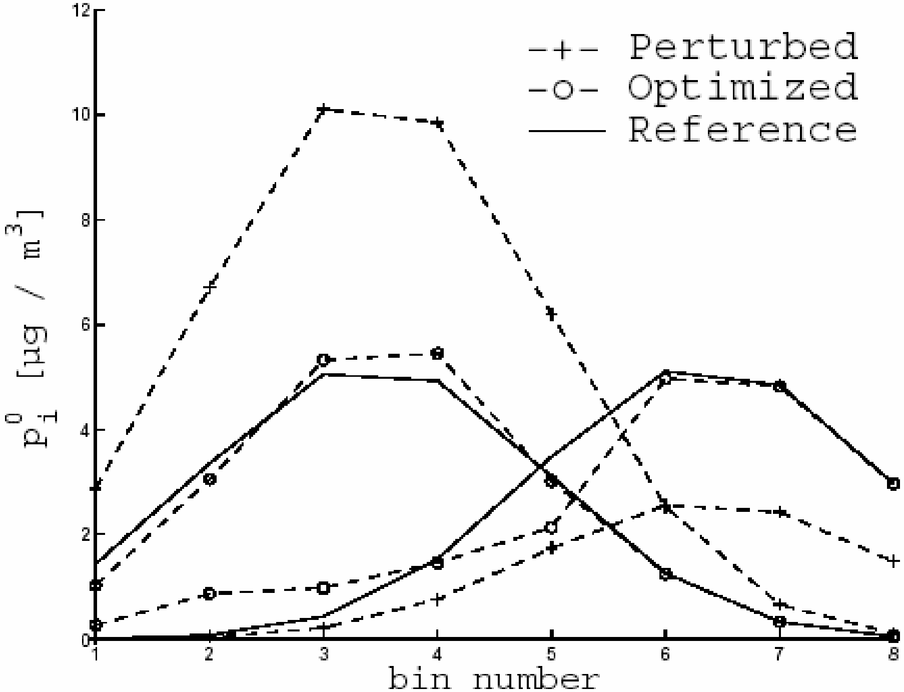
Adjoint of Cond / Evap

Recovering initial distribution and pure vapor concentration of species 1 using observations at t_f of only species 2 and 3

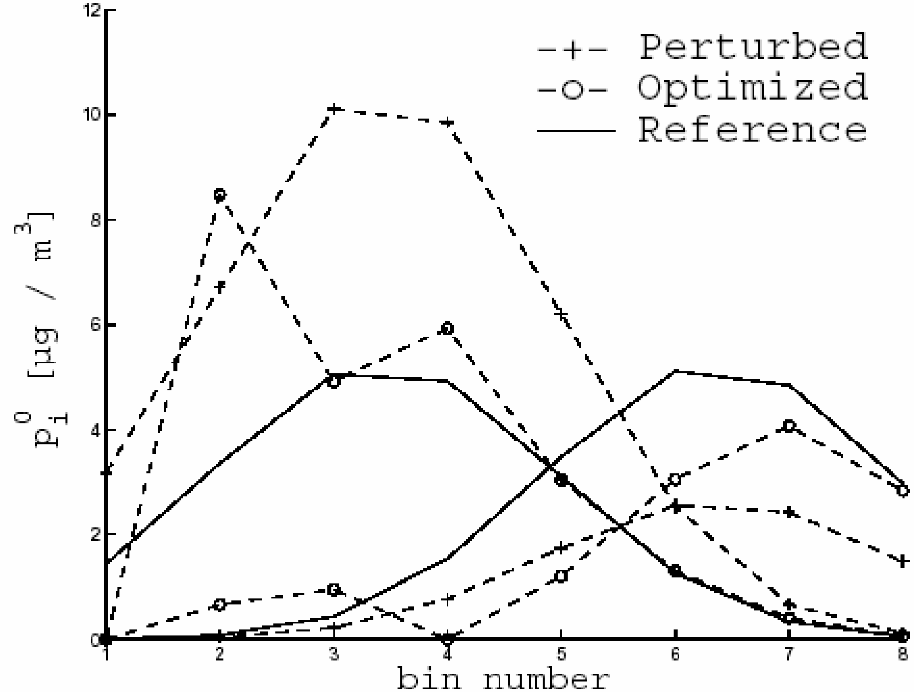


Adjoint of Cond / Evap
Discrete vs Continuous

Discrete



Continuous



Long Assimilation Period

The crux of inverse aerosol modeling:
Thermodynamics

Highly nonlinear behavior

Difficult to derive analytical expressions for chemical equilibrium

=> can't derive continuous adjoint

Difficult to construct discrete adjoint code

- Complex data relationships hinder manual construction
- Use of commands (RETURN, GOTO, …) not amenable to AD

Inverse Thermodynamics

Possible Approaches

Simplify the thermodynamics

- As used in large scale models (GEOS-CHEM)
- Limited chemical possibilities (MARS)

Obtain more robuts AD tools

- ADIFOR 3.0
- TAF ($$)

Calculate derivatives using alternative methods

- Hybrid approach
- Tabulated offline analysis

Inverse Thermodynamics
Hybrid Approach

Forward differentiation using TAMC

- Tangent linear derivative:

$$\mathrm{TAMC}(y, x, v) = \frac{\partial y}{\partial x} \cdot v$$

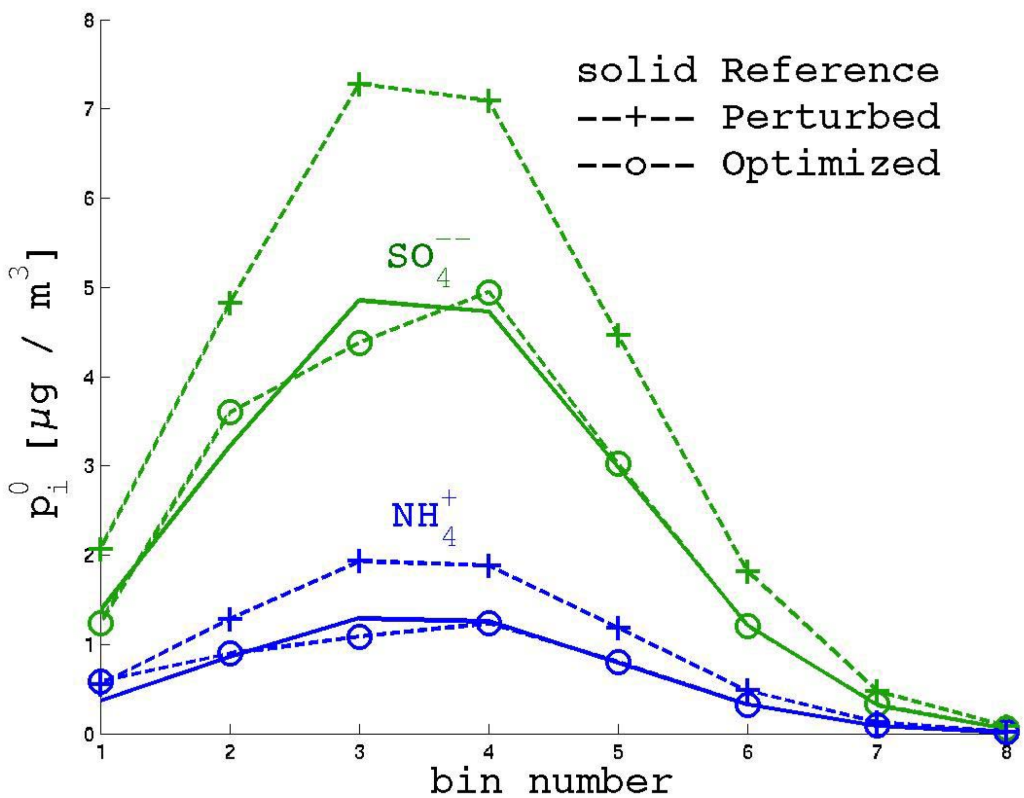
- The full Jacobian:

$$\mathrm{TAMC}(c_i^\circ, p_i^{k-1}, I_{n\times m}) = \frac{\partial c_i^\circ(p_i^{k-1})}{\partial p_i^{k-1}}$$

Use the transpose of this for the adjoint routine

Inverse Thermodynamics
Hybrid Approach

Recovering NH_4^+ and SO_4^{--} at $t = 0$ based on measurements after 1 hour (NH_3 condensing)



Inverse Thermodynamics

Simple Equilibrium Treatment

Emissions (NH_3, NO_x, SO_2, VOCs)

↓

CTM

↓

Total Concentrations (NH_3, HNO_3, SO_4^{2-})

↓

THERMO

Aerosol Phase ← → **Gas phase**

Aerosol Phase

NO_3^-, NH_4^+, SO_4^{2-}

H_2O, HSO_4^-

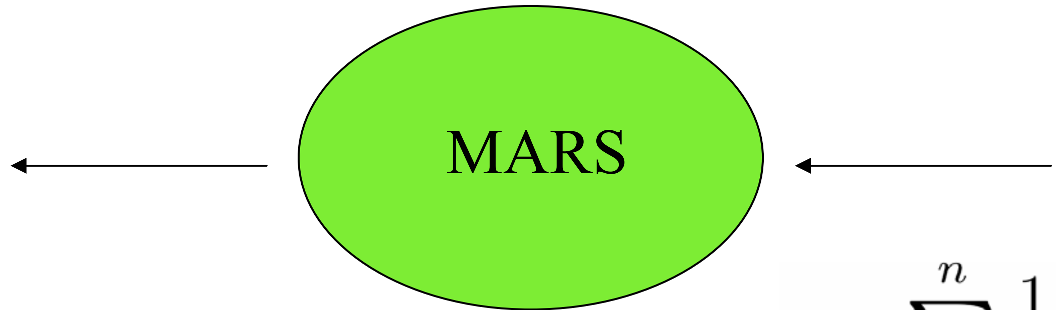
Gas phase

HNO_3, NH_3

Inverse Thermodynamics

Adjoint of Equilibrium Model

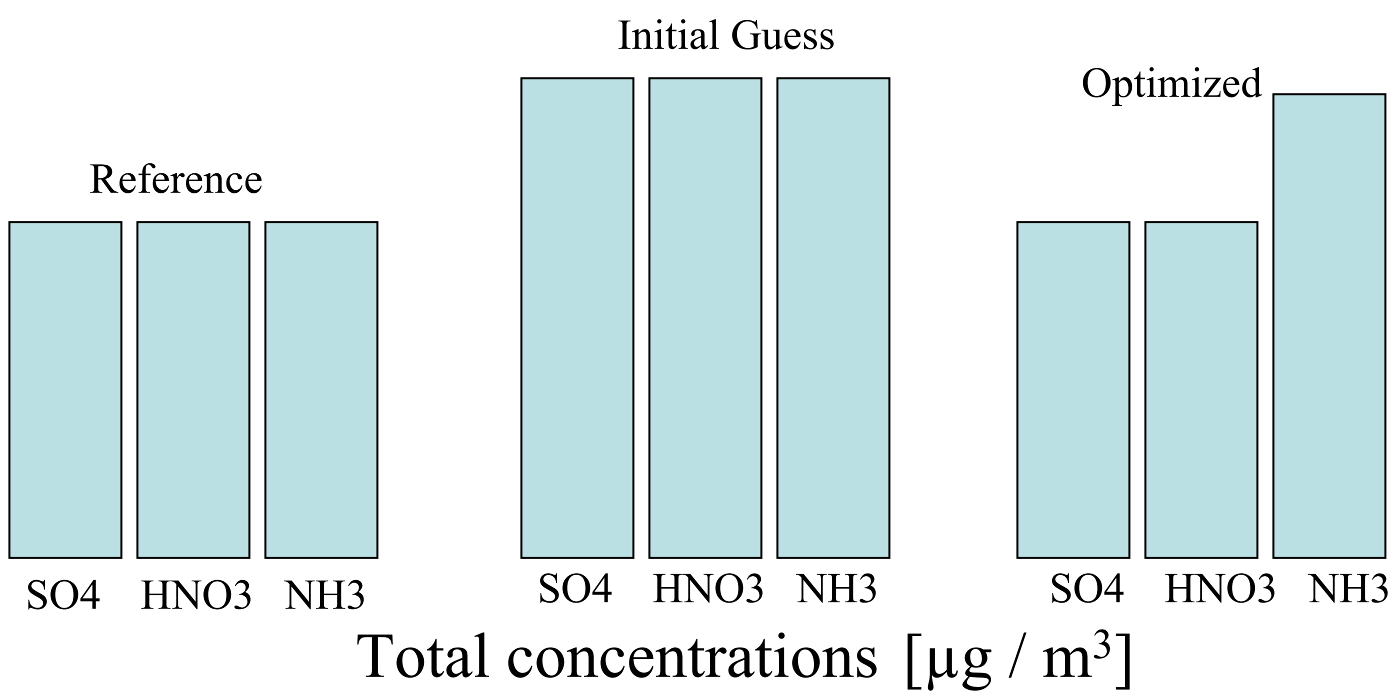
$$\frac{\partial J}{\partial c_{i,tot}}$$



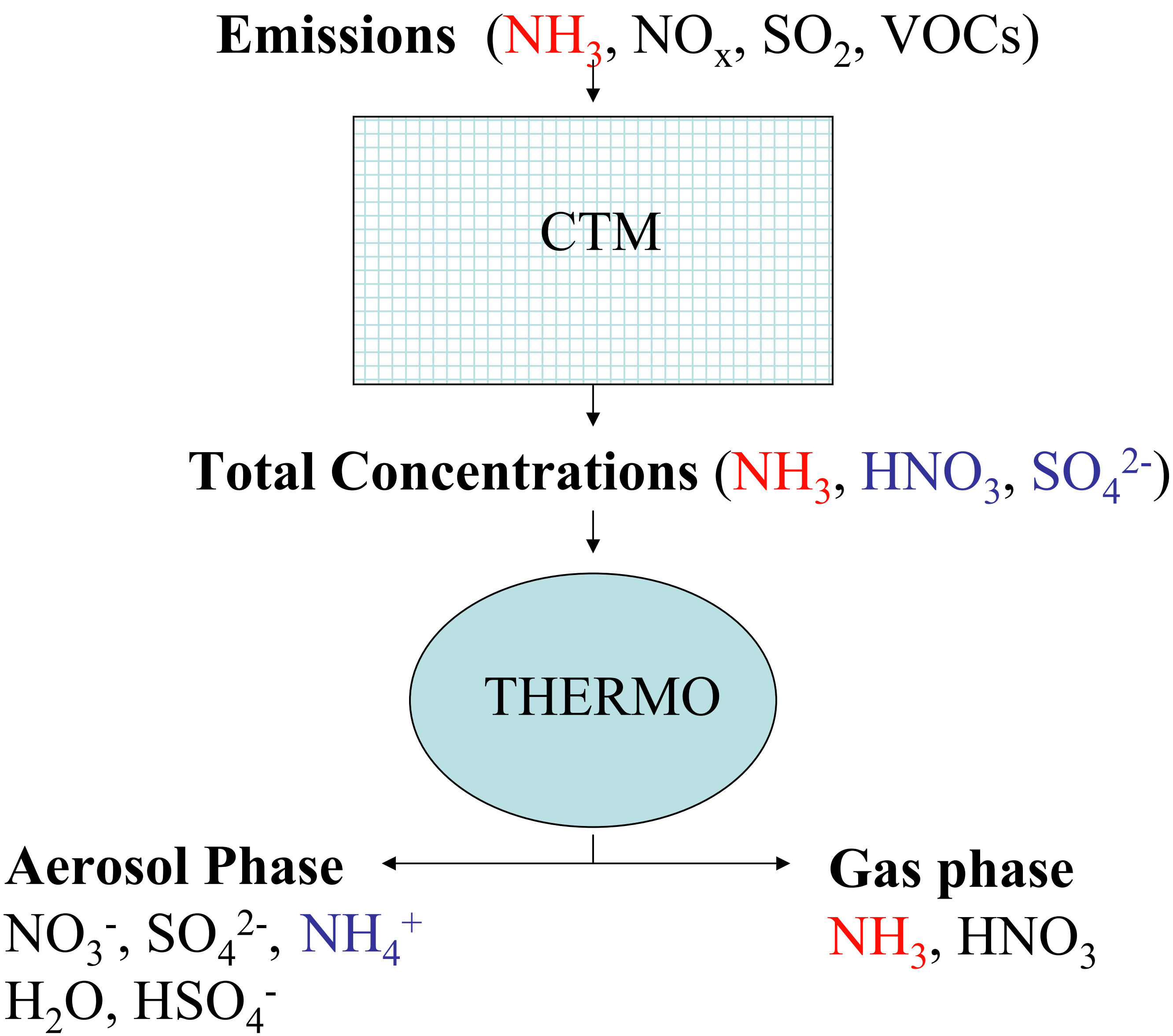
$$\frac{\partial J}{\partial p_i}$$

$$J = \sum_{i=1}^{n} \frac{1}{2}(p_i - p_i^{obs})^2$$

Results



Constraining NH_3



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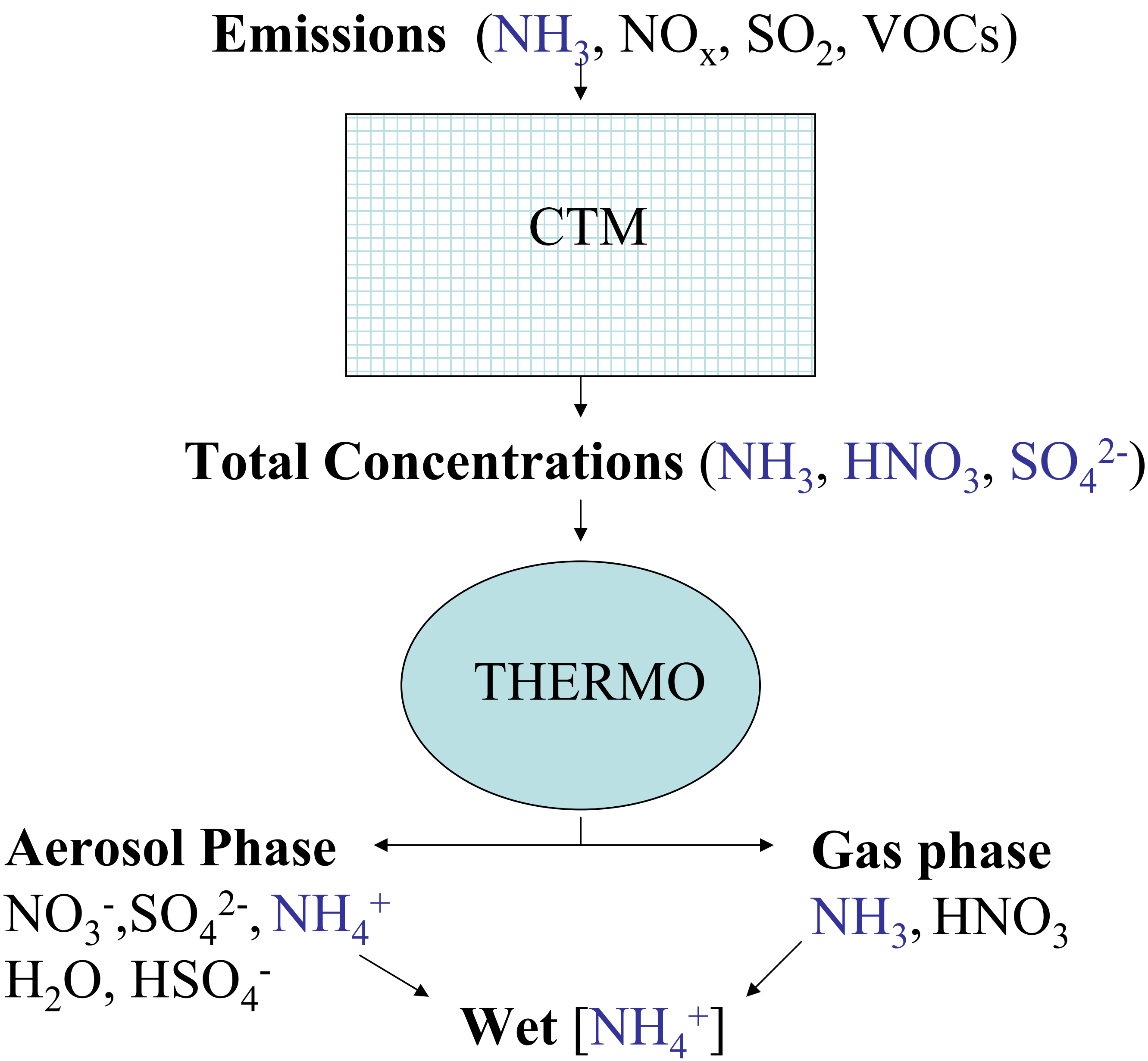
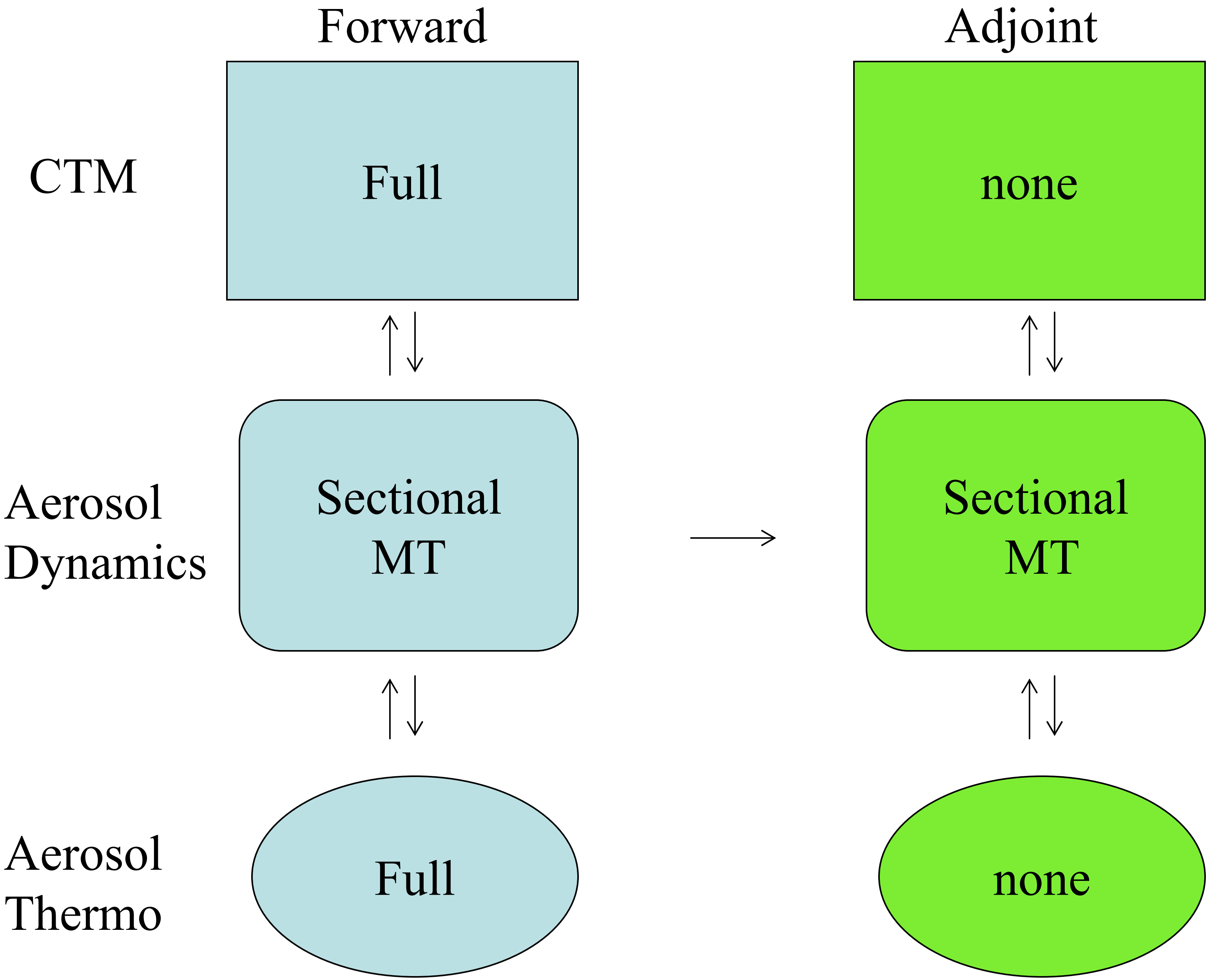
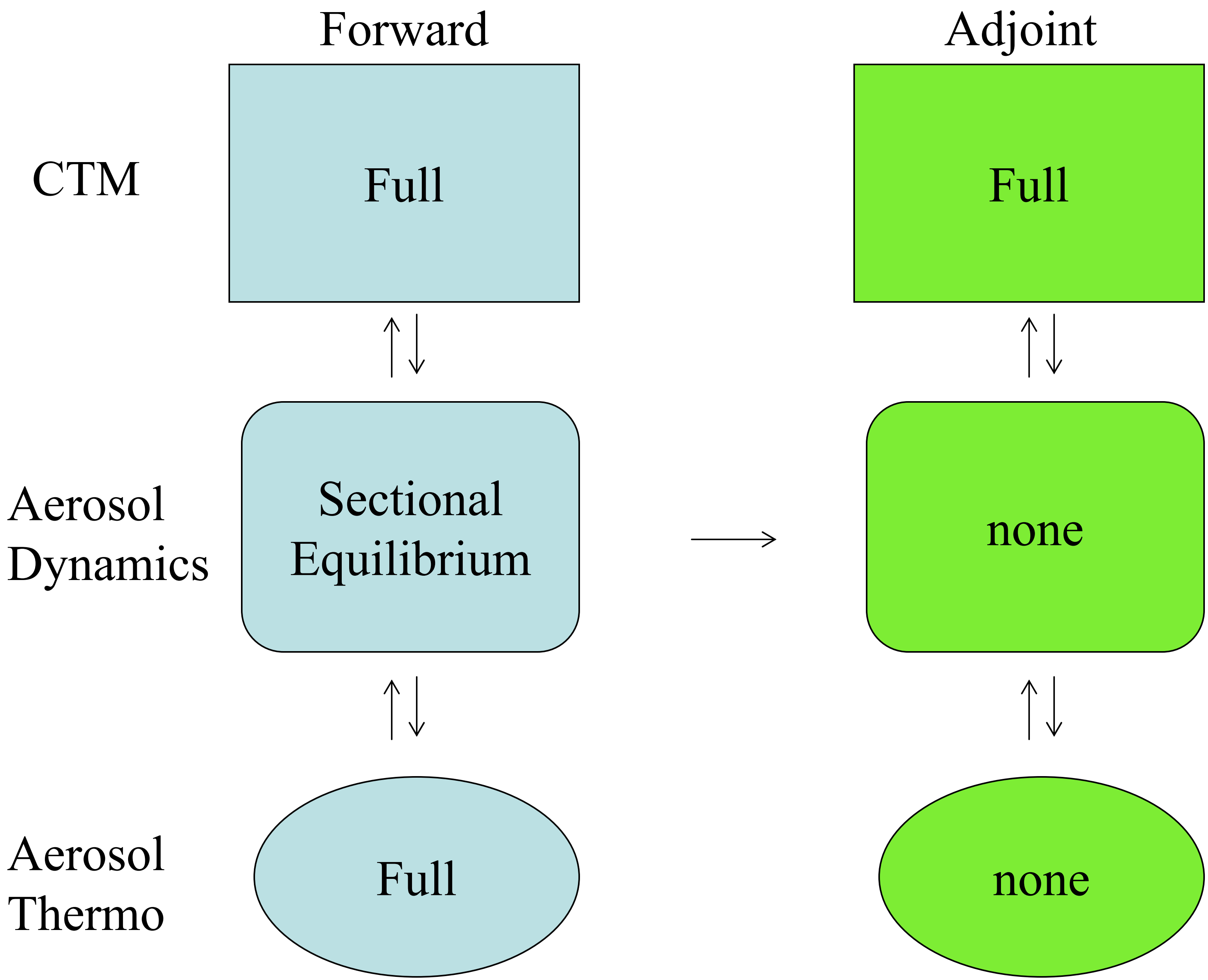


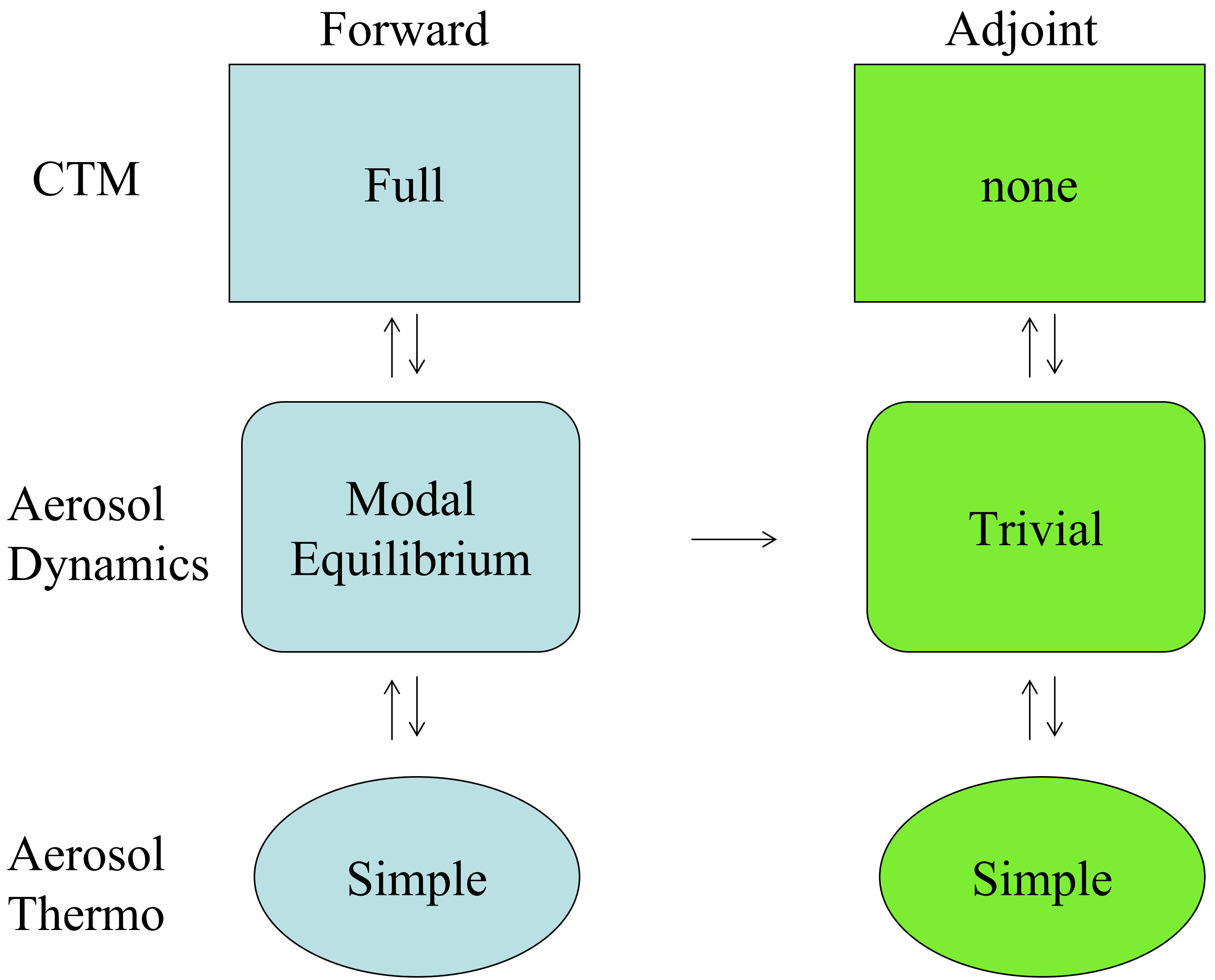
Local Scale (CIT Model)



Regional Scale (STEM)



Global Scale (GEOS-CHEM)



Future work

Regional Scale

- Adjoint of equilibrium, sectional aerosol dynamics

Global & Local Scale

- Adjoint of the rest of the model

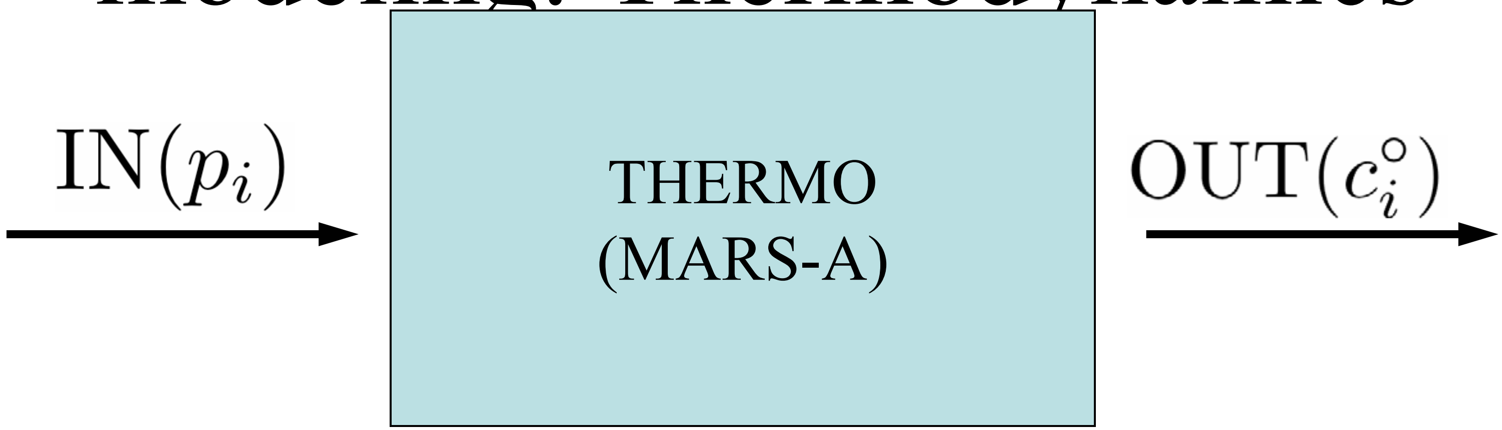
All Scales

- Decide on treatment of thermodynamics
  - simplify
  - approximate
  - hybrid

Previous inverse modeling studies of PM

	Mendoza-D. et al, 2002	*Gilliland et al, 2003*	*Park et al, 2003*
Model	URM	CMAQ	GEOS-CHEM
Domain	Eastern U.S.	Eastern U.S.	U.S.
Period	5/24-29, 7/11-19 1995	1990	1998
Data	IMRPOVE, AIRS, PAM	NADP (Wet NH_4^+)	IMRPOVE
Method	DDC + R.R.	Kalman Filter	Lin. Reg.
Emission	CO, SO_2, NO_x, NH_3, VOC, ORGF	NH_3 monthly	OC and EC yearly by source

The crux of inverse aerosol modeling: Thermodynamics



$$\lambda^{k-1} = \left[\frac{\partial F(c_i^\circ(p_i^{k-1}), g_i^{k-1})}{\partial p_i^{k-1}} \right]^T \lambda^k$$

$$= \left[\frac{\partial c_i^\circ(p_i^{k-1})}{\partial p_i^{k-1}} \right]^T \left[\frac{\partial F}{\partial c_i^\circ(p_i^{k-1})} \right]^T \lambda^k$$

What comes next?

- More robust thermodynamic inverse
- Inverting "equilibrium mode" aerosol dynamics
- Extend from λ to emissions

$$\nabla_E \mathcal{J} = \int_\tau \frac{\partial F(p(\tau))}{\partial E} \lambda(\tau) d\tau$$

Inverse Modeling

Given velocity field from meteorological data

- Guess the Emission Flux
- Run model simulation
- wait wait wait....
- Compare model predicted surface concentrations to surface measurements
- Guess the Emission Flux...

$$J = \int_{\Omega} \int_{\tau} (|N - N_{obs}|)\delta(\Omega - \Omega_{obs})\, d\Omega\, dt$$

or

- Develop inverse model to find direct relation

$$\Delta J = \iint \left\{ v^{*} \Delta E \right\}_{z=0} d\omega\, dt$$

Our inverse modeling approach

- Kalman filter
  - coarse constraint of regional emission scale factors
  - quantify model & estimation error
- Adjoint method
  - refine constrains of emissions with spatial / temporal resolution
  - generate discrete adjoint using AD

A Simple Example

Consider perturbation in the solution, Δu

$$L\Delta u = Q\Delta\alpha$$

$$L = \left(\frac{\partial}{\partial t} - \frac{\partial f}{\partial u}\right)_{\alpha} \quad Q = \left(\frac{\partial f}{\partial \alpha}\right)_{u}$$

$$\Delta J = \int_{\tau} \left(\frac{\partial g}{\partial u}\right)_{\alpha} \Delta u dt + \int_{\tau} \left(\frac{\partial g}{\partial k}\right)_{u} \Delta \alpha dt$$

Remember that $L\Delta u$ is a very expensive term to evaluate.

Just in Time Mathematics presents....
The Adjoint Method

How to avoid multiple model runs

- Gov'n equation

$$\frac{\partial u}{\partial t} = f(u,\alpha)$$

- Cost function

$$J = \int_{\tau} \underbrace{(|u - u_{obs}|)\delta(t - t_{obs})}_{\text{g}} dt$$

A Simple Example

Introduce the adjoint operator

$$\int_{t_0}^{t_f} \Delta u L^* v^* dt = \int_{t_0}^{t_f} v^* L \Delta u dt - [\Delta u v^*]_{t_o}^{t_f}$$

Force the adjoint equation at the observation point

$$L^* v^* = \left(\frac{\partial g}{\partial u}\right)_\alpha$$

Substitute back into the adjoint operator equation

$$\int_{t_0}^{t_f} \Delta u \left(\frac{\partial g}{\partial u}\right)_\alpha dt = \int_{t_0}^{t_f} v^* L \Delta u dt - [\Delta u v^*]_{t_o}^{t_f}$$

A Simple Example

Run the model once to obtain base case values

Find the adjoint solution

The adjoint solution is invariant to changes in parameters and boundary conditions.

Now have the cost function in terms of known quantities.

$$\Delta J = \int_{t_0}^{t_f} v^* Q \Delta \alpha dt - [\Delta u v^*]_{t_o}^{t_f}$$

A Simple Example

Formulate the adjoint operator

$$\int_{t_0}^{t_f} \Delta u L^* v^* dt = \int_{t_0}^{t_f} v^* L \Delta u dt - [\Delta u v^*]_{t_o}^{t_f}$$

$$\therefore L^* = -\frac{\partial}{\partial t} - \frac{\partial f}{\partial u}$$

Solve the adjoint equation

$$L^* v^* = \delta(t - t_{obs})$$

$$\{v^*\}_{t_f} = 0$$

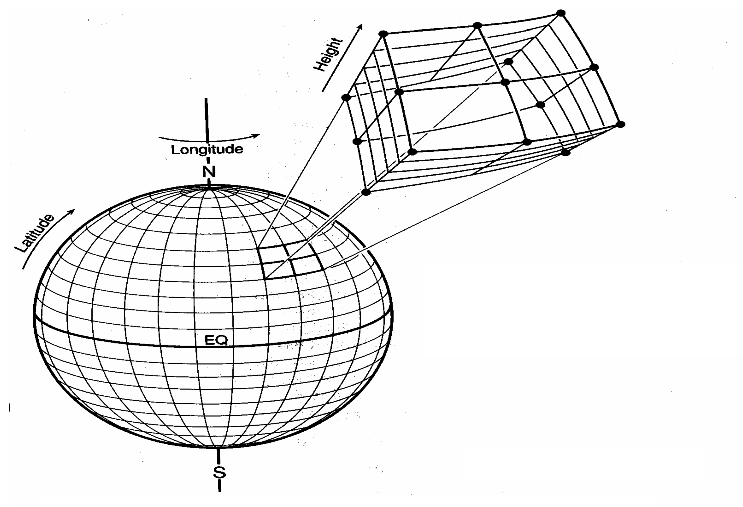
Inverse Modeling

The Adjoint Method

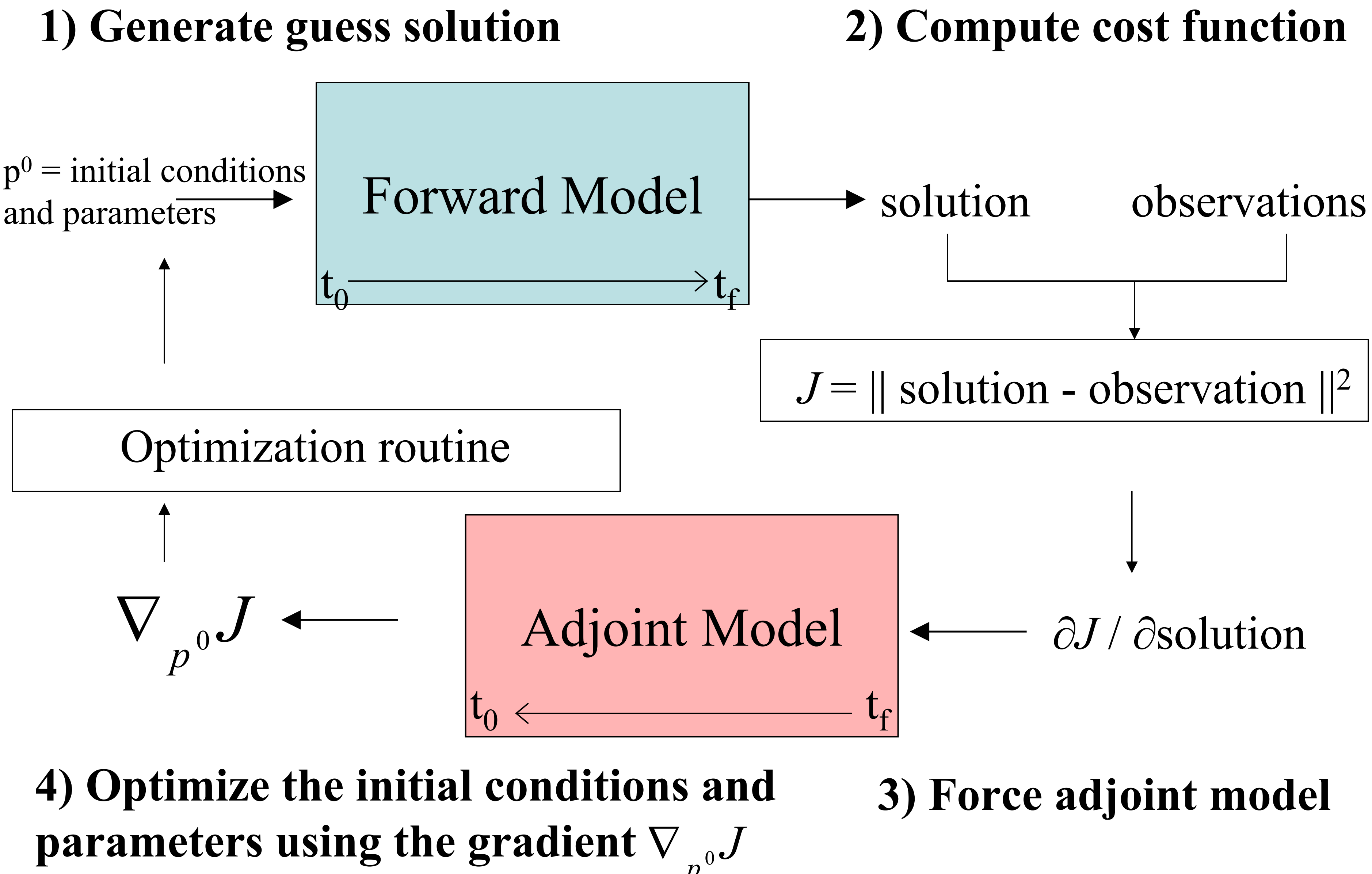
- Variational calculus, optimal control theory (*Hilbert, 50's*)
- Suggested for use in atmospheric data assimilation
  (Marchuk, 71)
- Used in Fluid Mechanics *(Pironneau, 74),* Oceanography
  (Tziperman and Thacker, 89)
- Applied to gas phase species in the atmosphere
  (Fisher and Long, 95)
- Can we use it to recover information about aerosols?
  (Henze et al, 2004)

Forward model: GEOS-CHEM

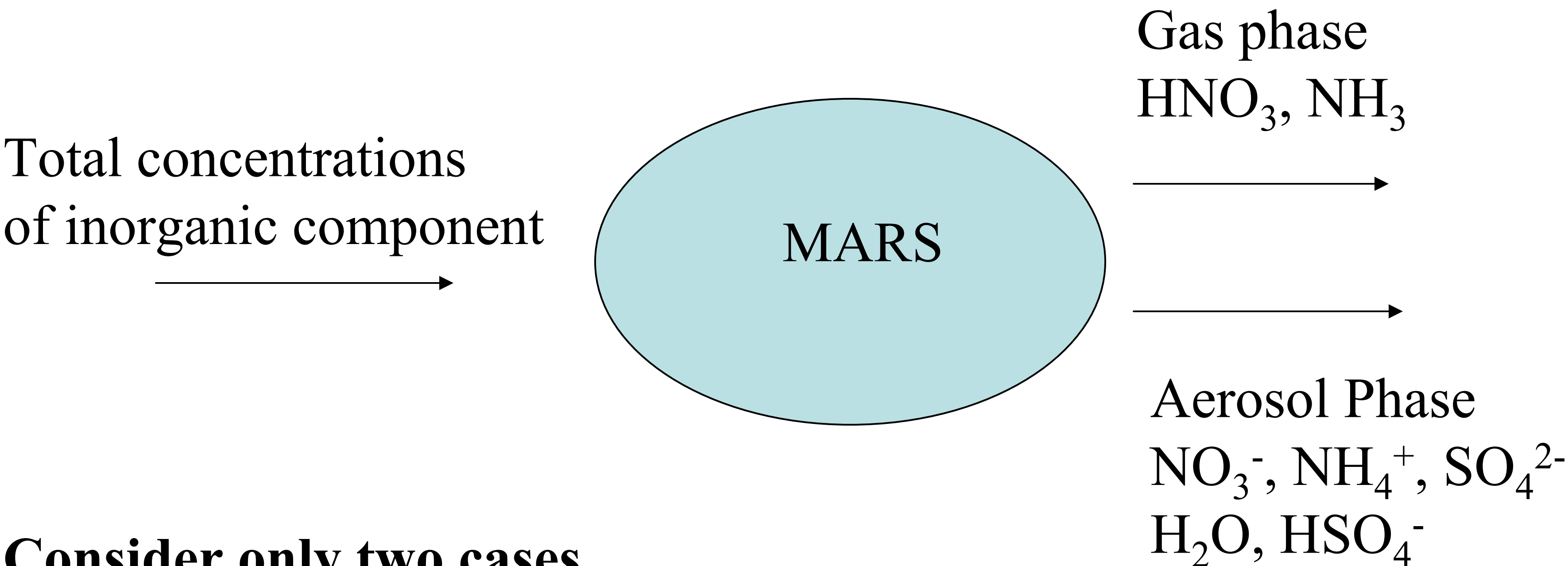
- 1°x1° (U.S.) nested in 4°x5° (Global) resolution, 55 levels
- Gas - aerosol interactions
  - photolysis frequencies
  - heterogeneous chemistry
  - gas/particle fractionation
- Externally mixed aerosols
  - H_2SO_4-HNO_3-NH_3 system driven by
  - organic carbon (OC)
  - elemental carbon (EC)
  - soil dust (four size classes)
  - sea salt (two size classes)
  - mechanistic SOA (in progress)



Inverse Modeling Using Adjoint Method



Simple Thermodynamic Module



Consider only two cases

- Sulfate rich
- Sulfate poor

Bromely Method for activity coef

ZSR Method for water

Previous inverse modeling study of PM

- *Park et al*, 2003: Constrain carbonaceous aerosol using 7 emission parameters, **multiple linear regression**
- *Gilliland et al*, 2003: Constrain single NH3 emission parameter using **Kalman filter**
- *Mendoza-Dominguez & Russell*, 2001: Constrain several emission scale factors using **Kalman filter**.

More detail required to account for regional variation