Applications of Satellite Measurements and Modeling for Air Quality

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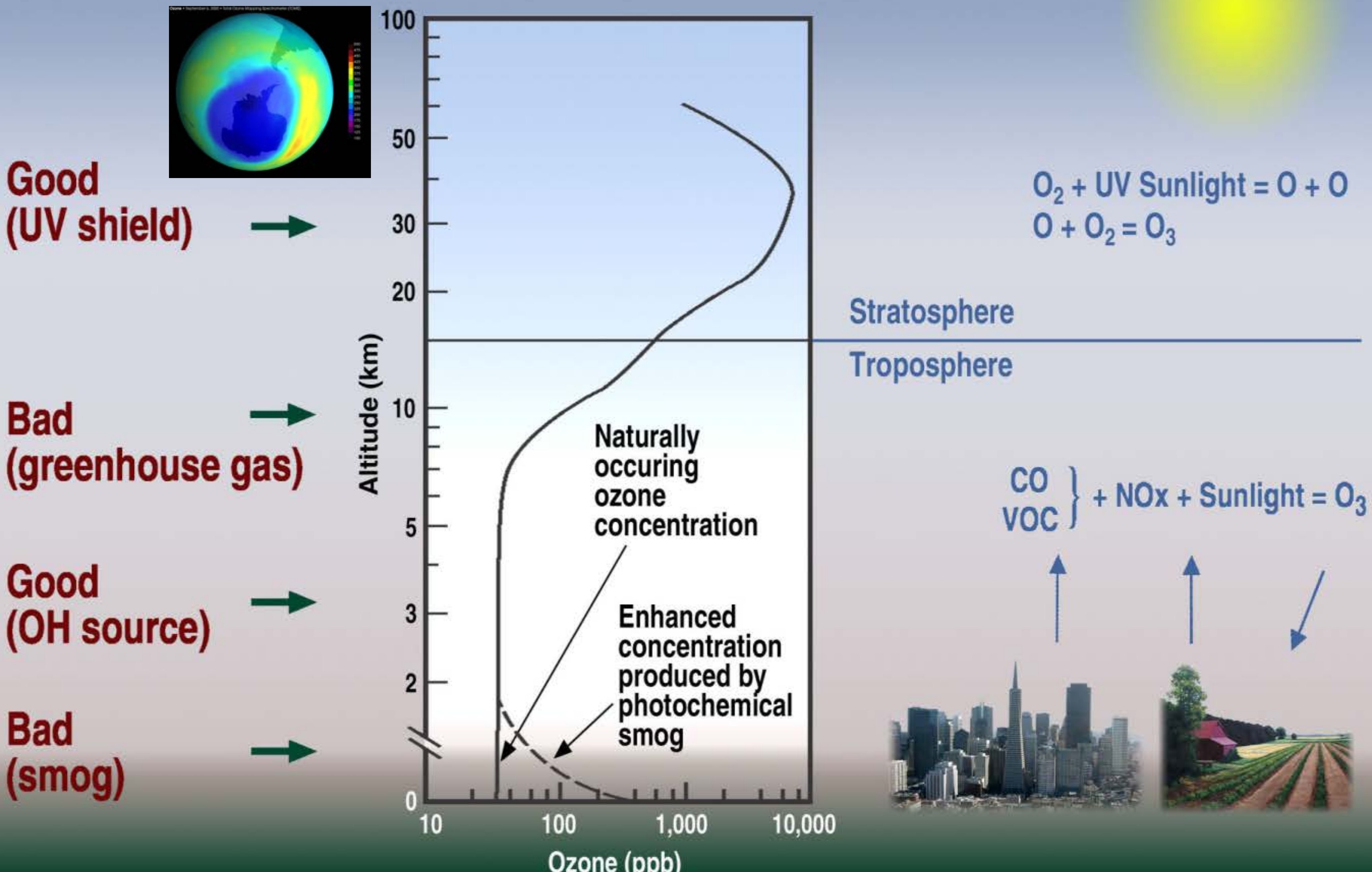
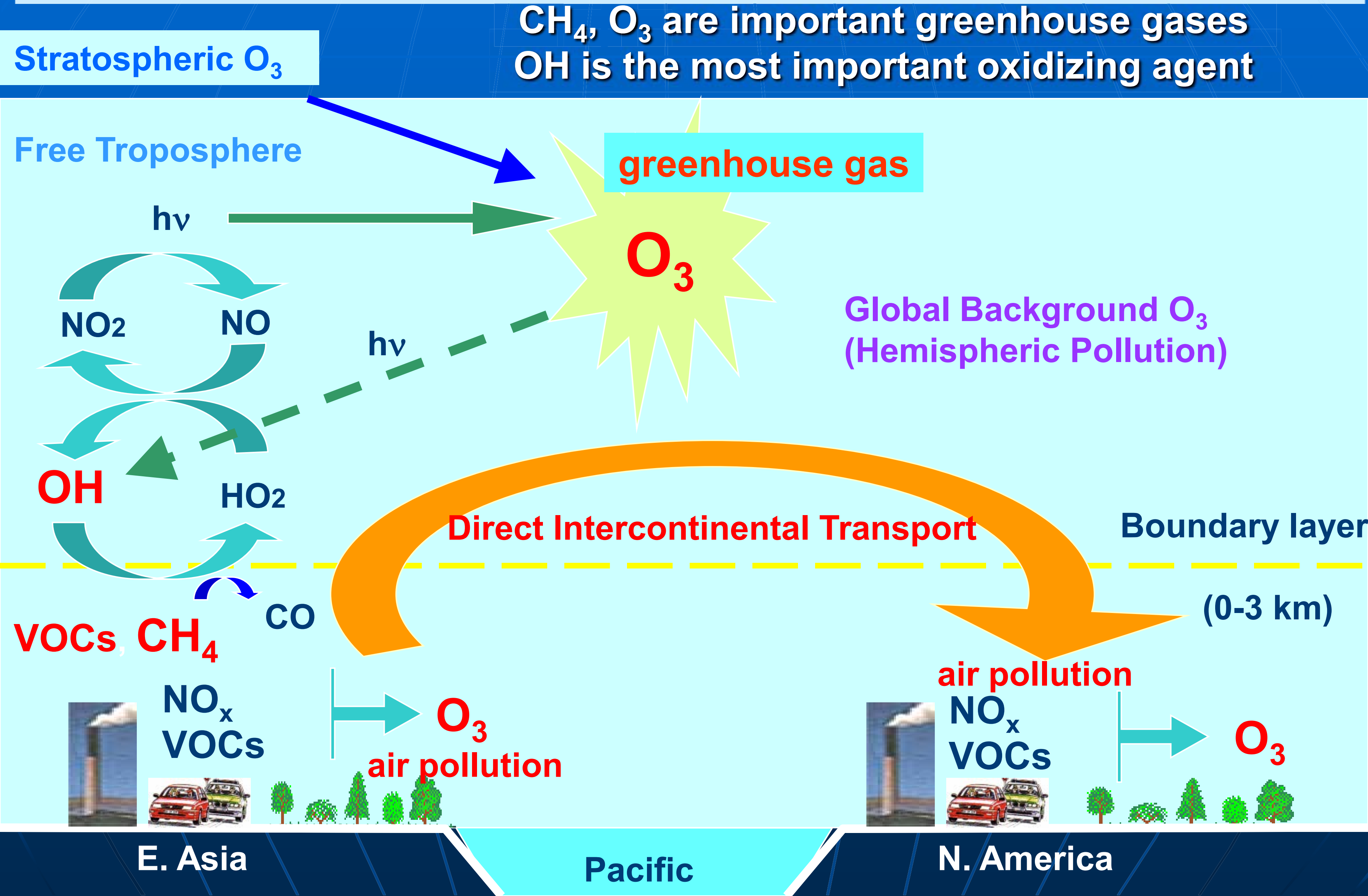
Atmospheric O_3 (Why O_3?)

OZONE: “GOOD UP HIGH, BAD NEARBY”



Prediction of O_3 is challenge

O3: Air Quality and Climate impact

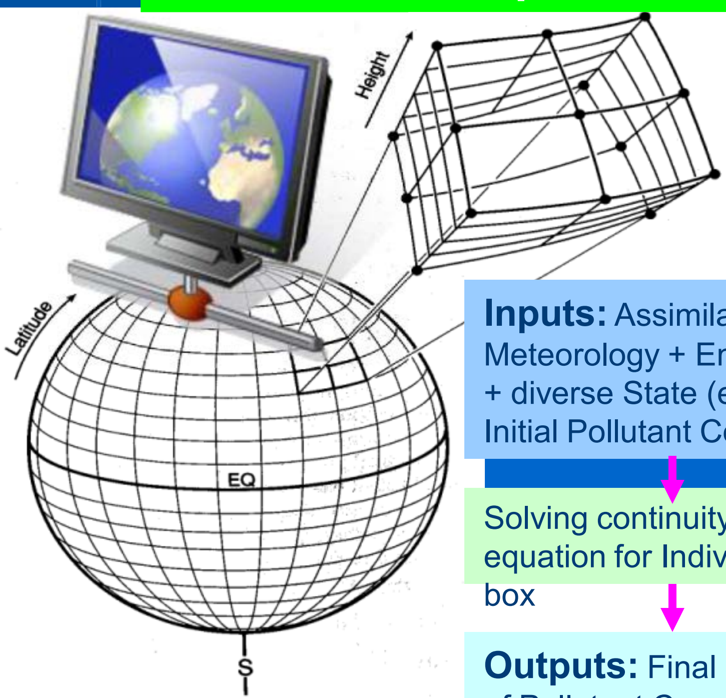


Quantitative Understanding of Air Pollution Chemistry

Accurate attribution of the factors is essential to air pollution policy!

- ➢ Background from Natural emissions (VOCs, NOx, …)
- ➢ Industrial/urban pollutants emissions
- ➢ Meteorological Impact (e.g., Stratospheric O_3 influence, rainouts)
- ➢ Atmospheric Chemical Kinetics (any missing mechanism?)

Chemical Transport Model



Inputs: Assimilated Meteorology + Emissions + diverse State (e.g., Initial Pollutant Conc.

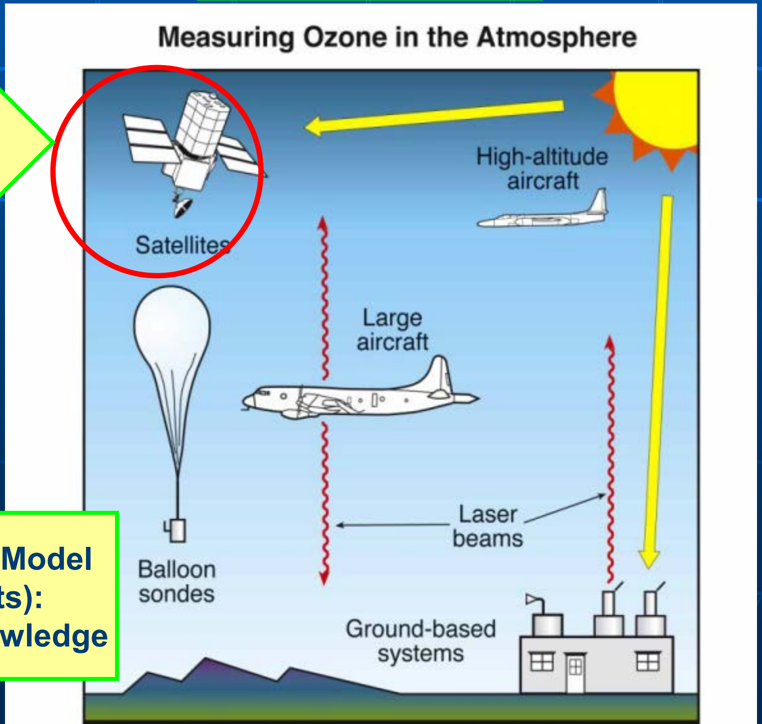
Solving continuity equation for Individual box

Outputs: Final State of Pollutant Conc.

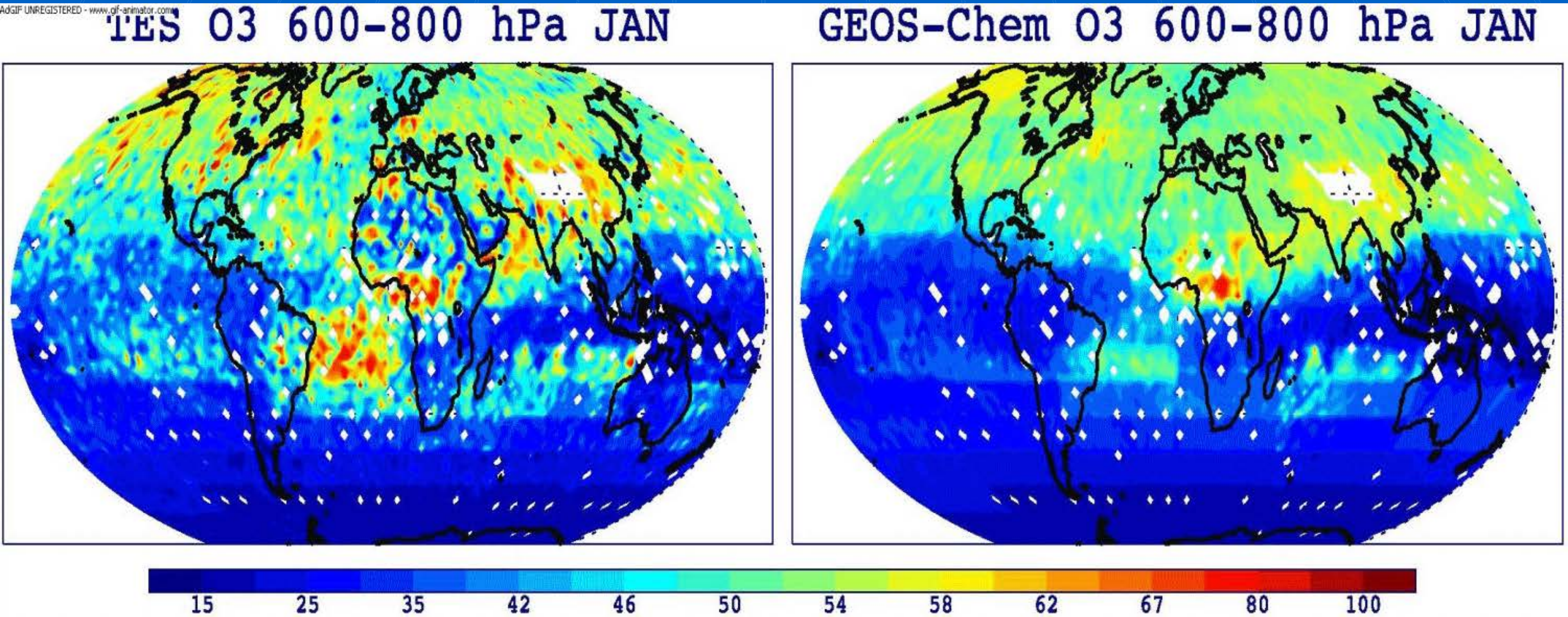
Understand Obs. Constrain Satellite Data

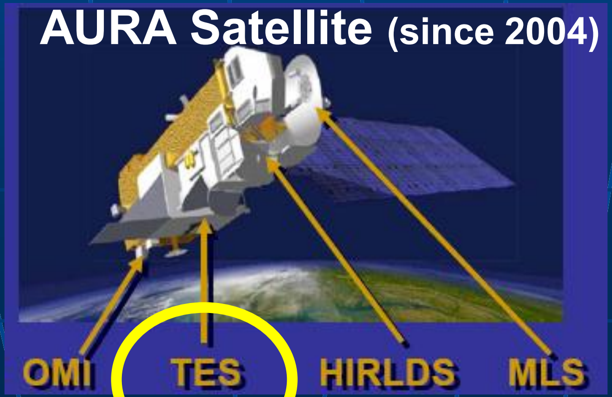
Constrain Most of Model Variables (Inputs): Increasing our knowledge

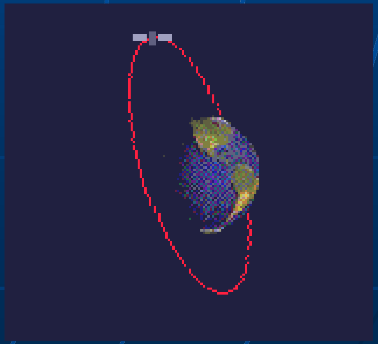
Observations



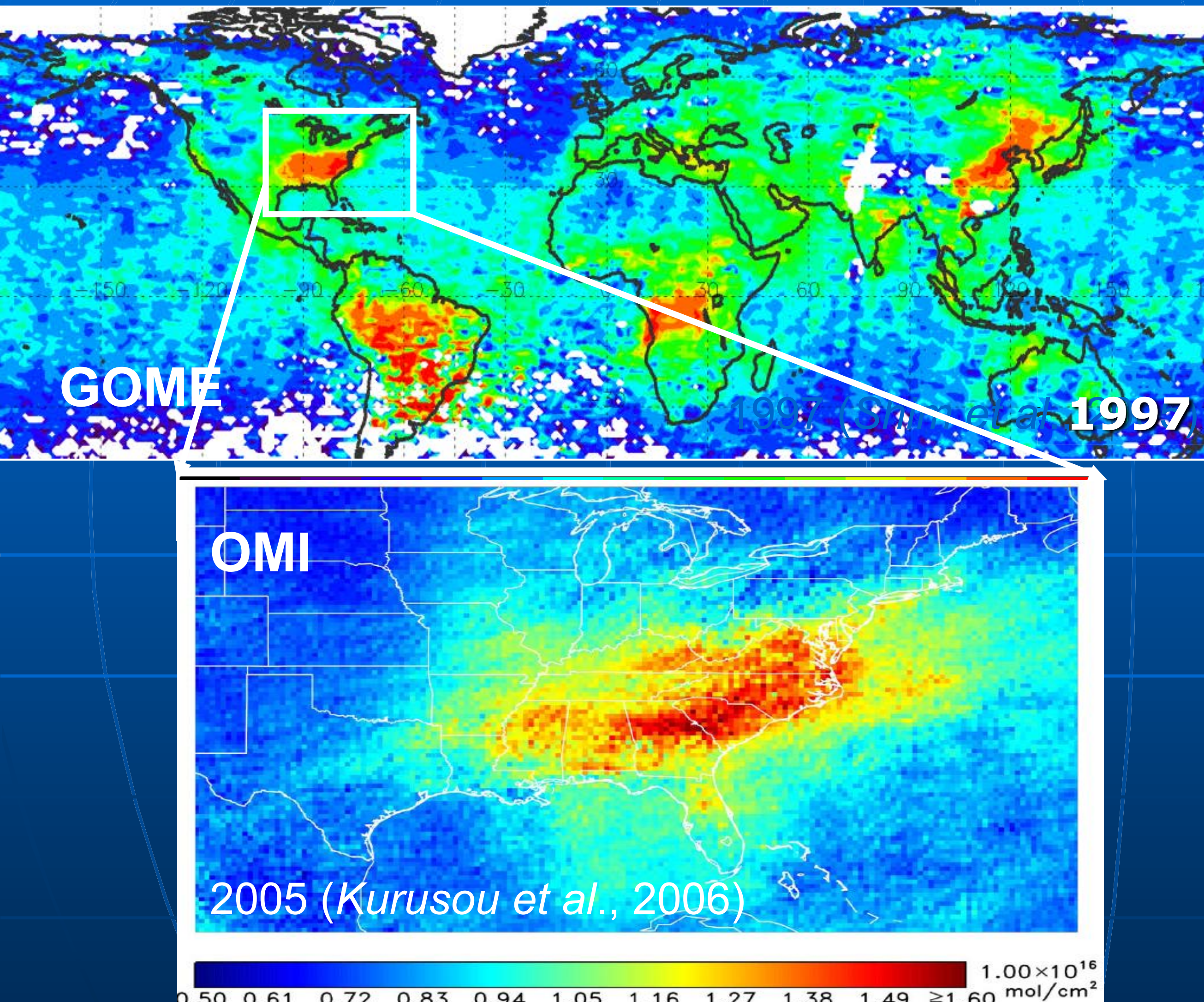
Global tropospheric O3 distribution by TES in 2006







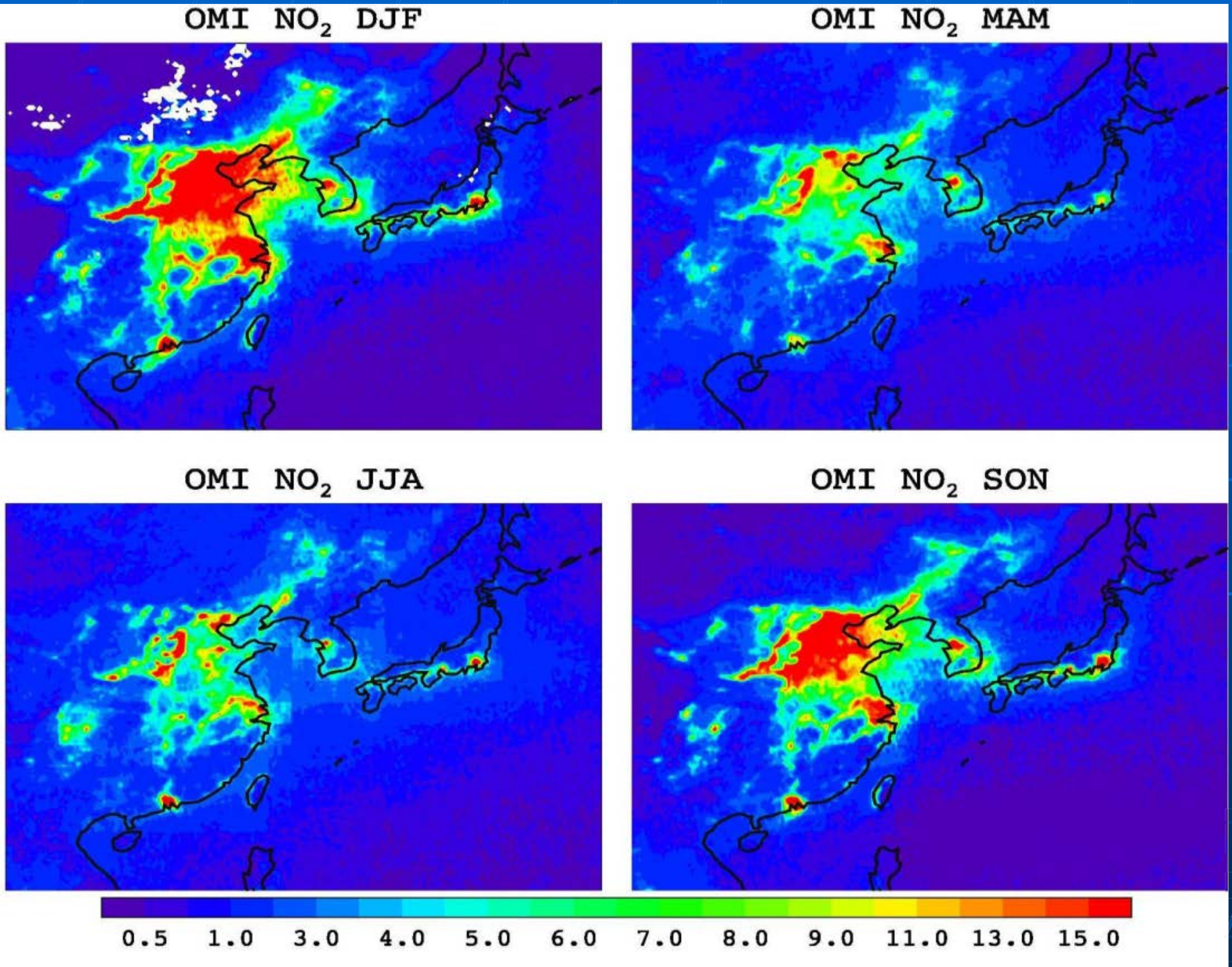
Improving resolution in satellite data (GOME vs OMI HCHO)

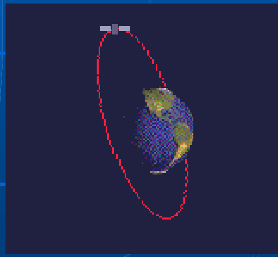


Satellite Instruments	Foot print	scale
GOME (1995)	320 x 80 km	Continental ~ global
TOMS(1996)	280 x 100 km	Continental ~ global
SCIAMACHY (2002)	60 x 30 km	Regional ~ continental
OMI (2004)	13 x 24 km	regional

Finer scale study

Utilizing Satellite Measurements: East Asian pollution (Nitrogen Dioxide from OMI in 2006)

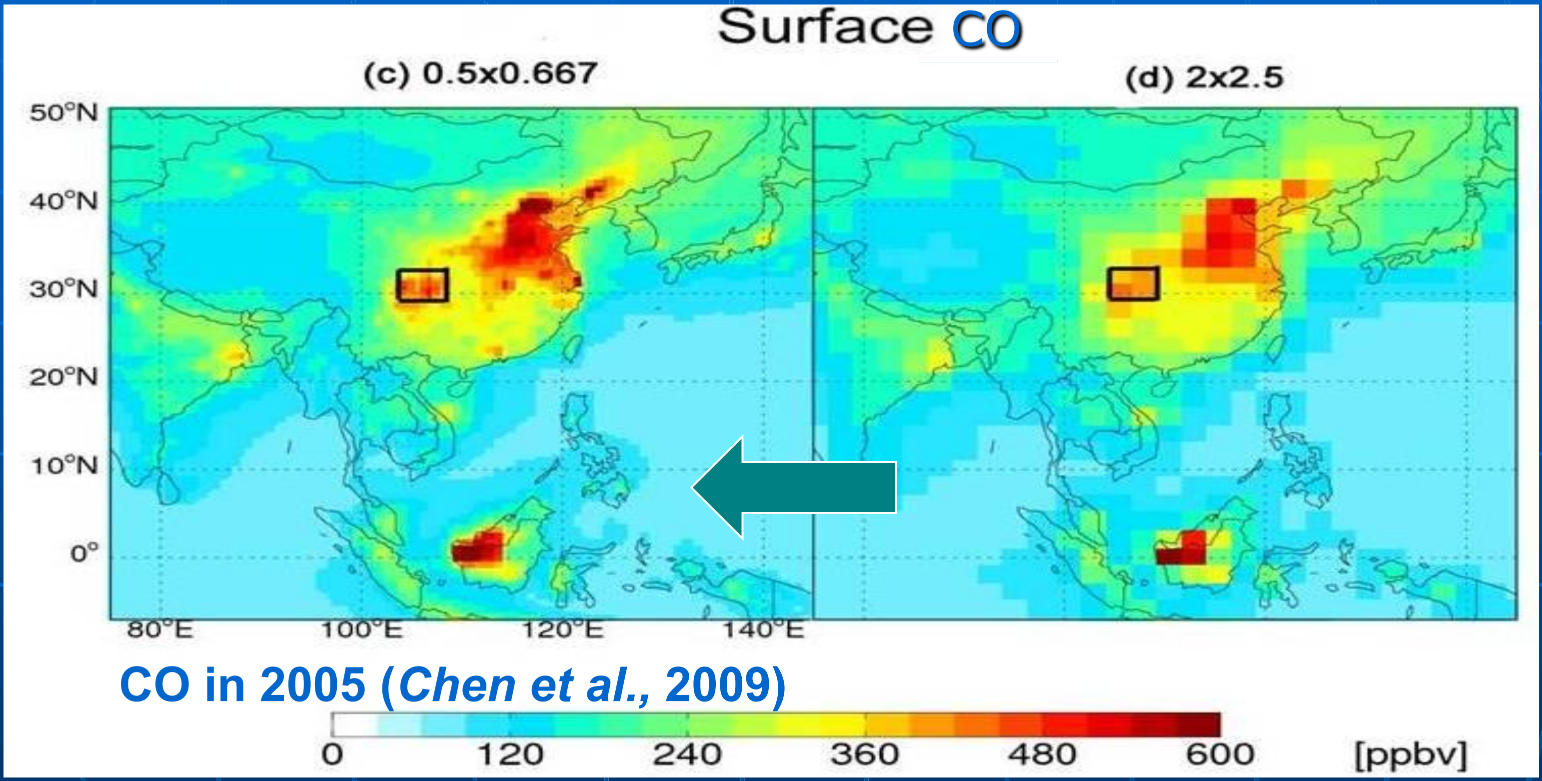




- Seasonal energy use
- NO2 lifetime
- Monsoon effect

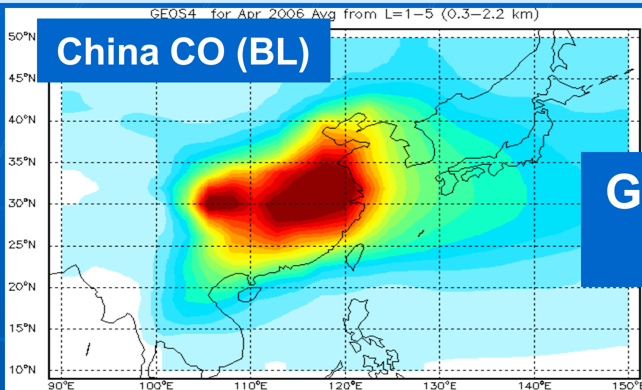
Improving horizontal resolution of CTM

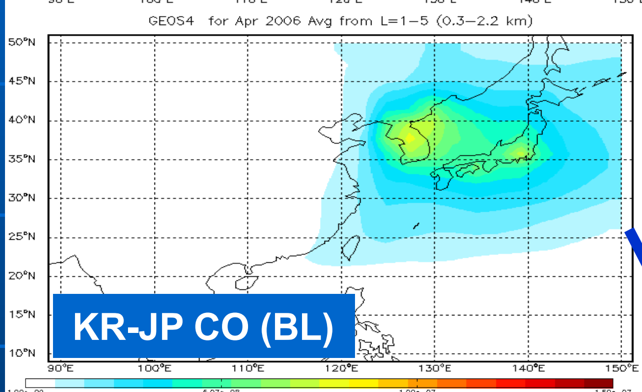
- Nested grid GEOS-Chem simulation (left)



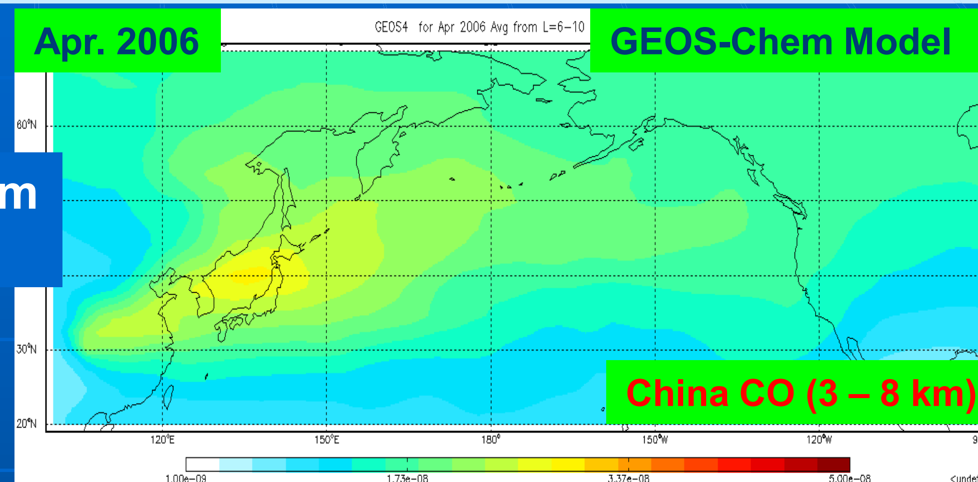
➢ Progress on the remote sensing technique and modeling capability is going to cover regional ~ global scale study

Utilizing CTM + Satellite Obs.
: Analysis of East Asian Pollution w/ TES CO (Seoul)

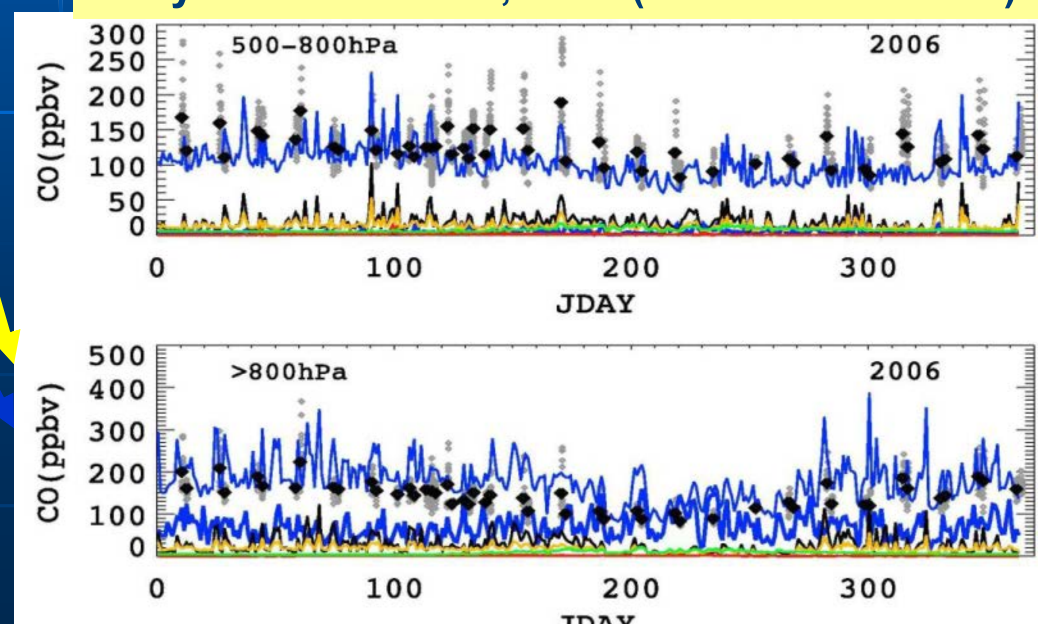




GEOS-Chem Model



Daily CO over Seoul, 2006 (Satellite vs Model)



Grays: TES Satellite Instr.
- ◇ Daily TES avg CO
- Model CO (all sources)
- China industry/urbane
- China Biofuel Burning
- KR-JP anth
- Biosphere
- BB

Topic 2: Improving emissions estimation

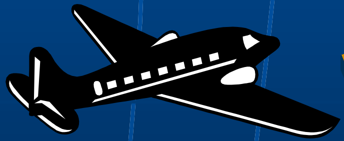
2 approaches to emissions estimation

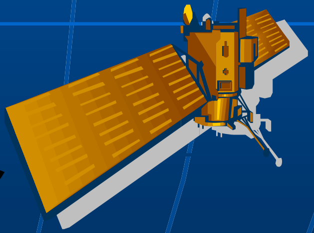
"Bottom up" emissions estimates

Creating detailed emissions inventories at a model resolution

"Top-down" emissions estimates

From observations + Inverse modeling w/ atmospheric observations





need for accurate emission estimates for regulatory purposes

Forward (CTM) vs Inverse model

- Infer a numerically optimized model variables in the given system derived from true states

"Causes (x)"

"Effects (y)"

Problem: Model variable amounts (more uncertain)
"bottom-up" a priori

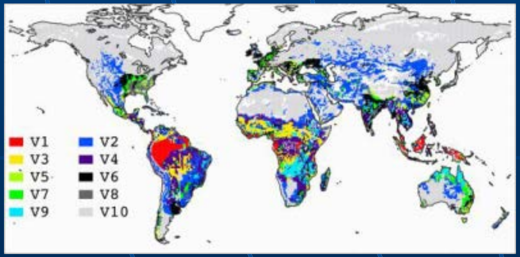
Forward model
(ex. CTM)
$y = Fx + e$
$(\mathbf{x} = x_1 + x_2 + ..)$

Prediction (e.g., modeled chemical species.)

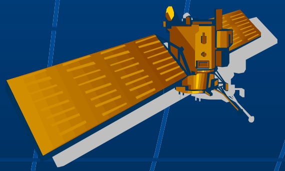
Solution: Optimized model variables
"Top-down" a posteriori

Inverse model

True State of Data (e.g., observed data)



Linearization
Optimization
$K = dy/dx$



Adjoint Inverse Modeling

Objective: calculate model parameters by minimizing the mismatches between observation and model prediction (cost function J(x))

minimize $J(\mathbf{x}) = (\mathbf{F}(\mathbf{x}) - \mathbf{y})^T \mathbf{S}_\varepsilon^{-1} (\mathbf{F}(\mathbf{x}) - \mathbf{y}) + (\mathbf{x} - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a)$

compute $\nabla_\mathbf{x} J(\mathbf{x}) = 2\underbrace{(\nabla_\mathrm{x} \mathbf{F}(\mathbf{x}))^\mathrm{T}}_{\text{adjoint of forward model}} \mathbf{S}_\varepsilon^{-1} (\mathbf{F}(\mathbf{x}) - \mathbf{y}) + 2\mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x_a})$

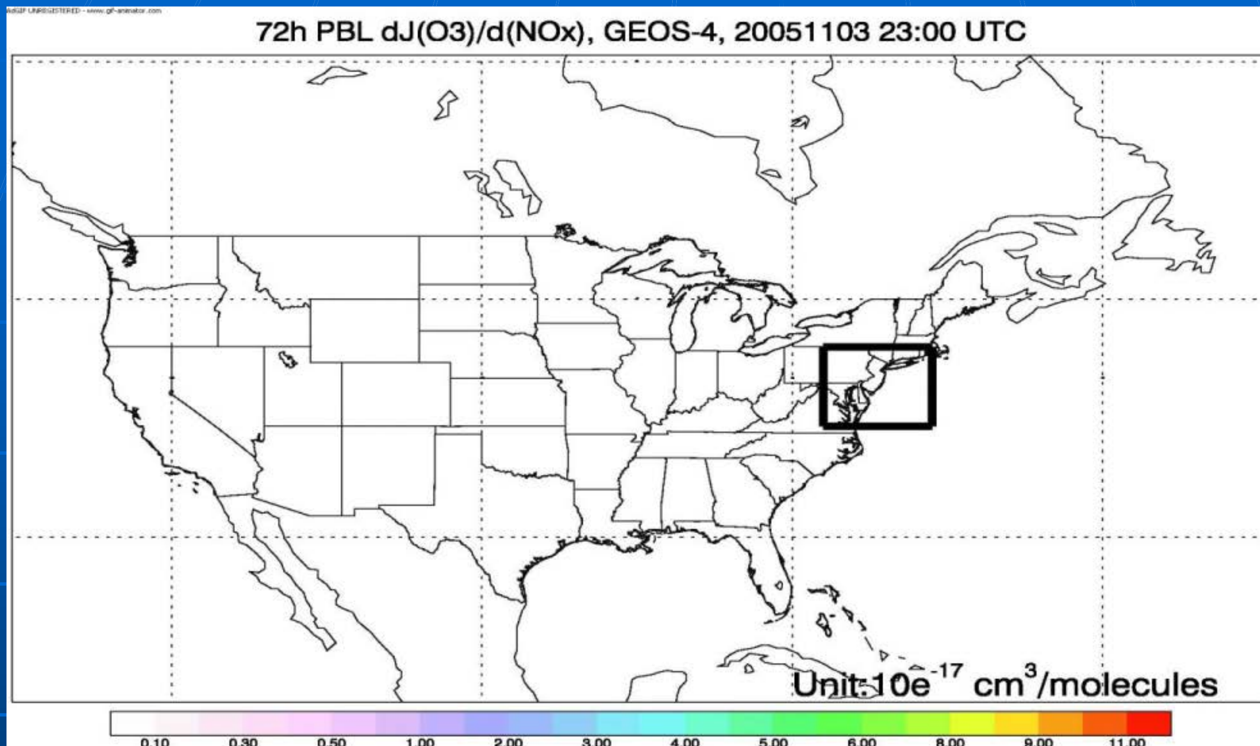
Rodgers, 2000

Adjoint method

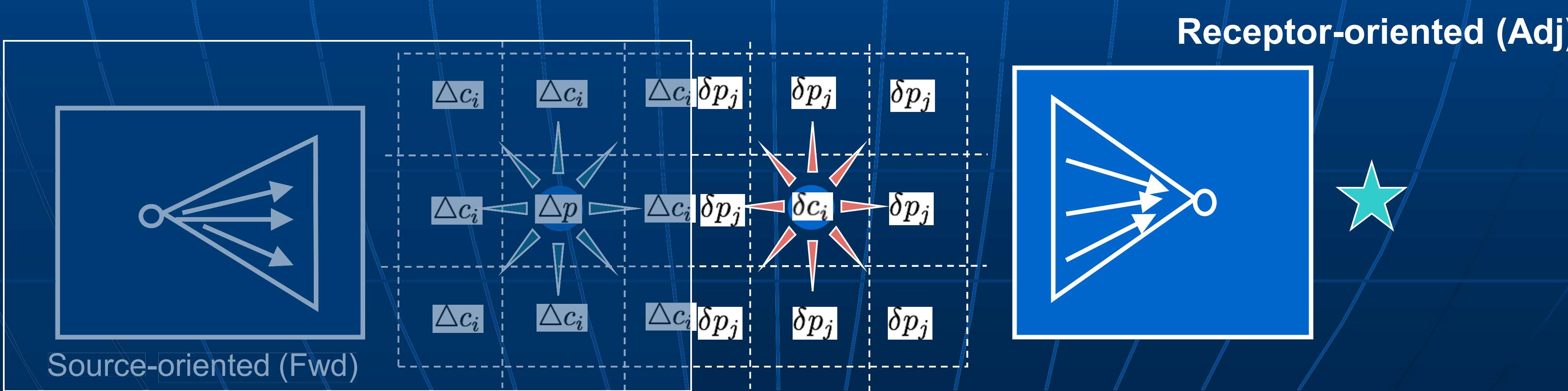
-Compute gradients $\nabla_x J(\mathbf{x})$ wrt model parameters by estimating adjoint (backward) forcing at the receptor, propagating backward in time and space to the initial condition (e.g. surface emissions). Advantage?

-Use optimization (steepest-descent) algorithm iteratively until the gradient reaches the minimum → find $\hat{\mathrm{x}}$

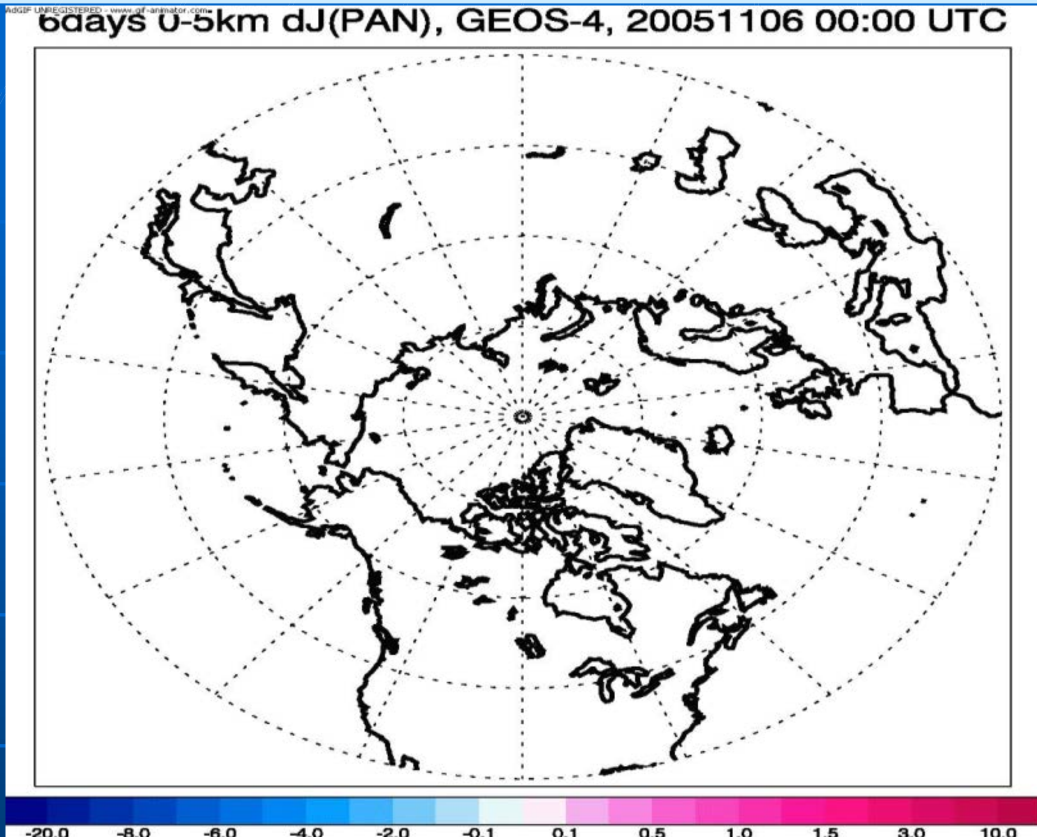
Adjoint Sensitivity (Case I: E. US)



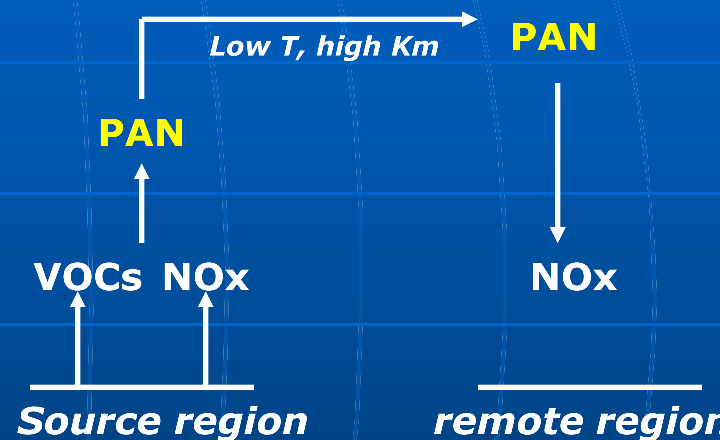
- 72 hrs adjoint sensitivity of PBL O3 over NYC ~ D.C wrt PBL NOX concentration (2005 11/03)

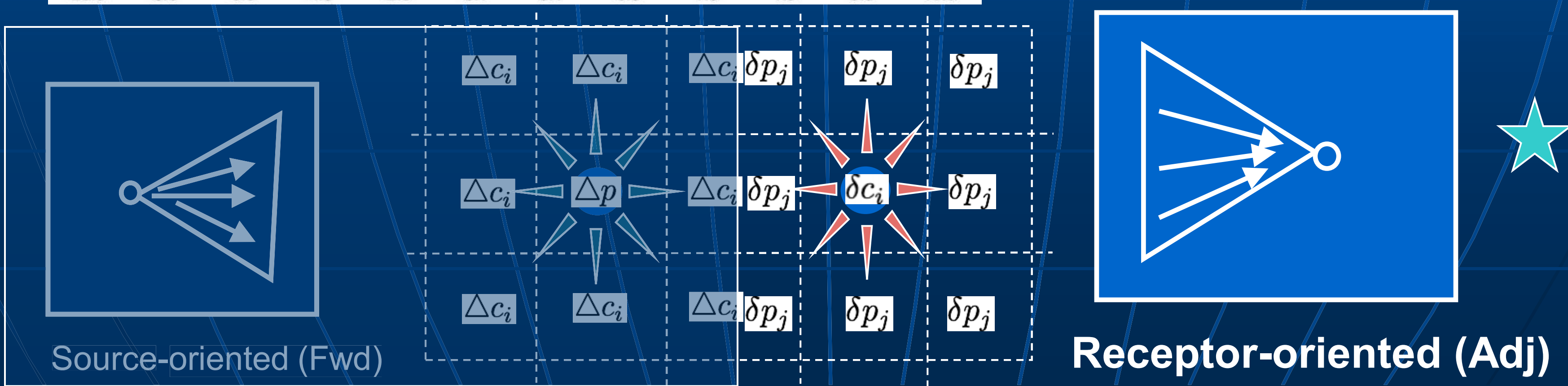


Adjoint Sensitivity (Case II: N. Pacific)

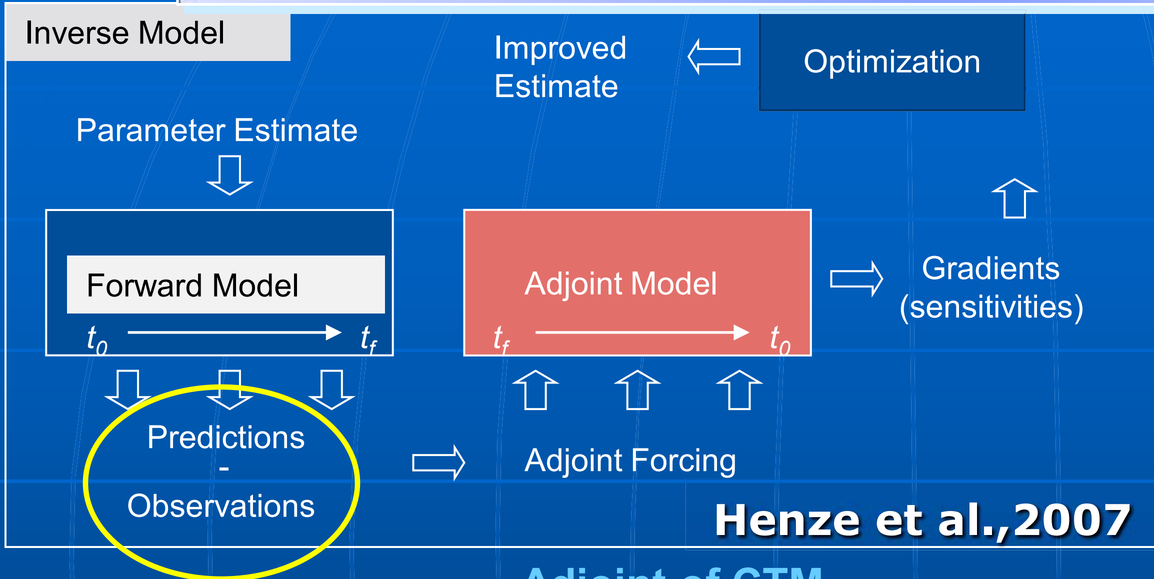


- 5 days adjoint sensitivity of PAN over **Alaska** at 0 ~ 5km height (2005 11/05)





Adjoint Inverse Modeling (iterative way)



Cost function

$$J(\mathbf{x}) = (\mathbf{F}(\mathbf{x})-\mathbf{y})^{T}\mathbf{S}_{\Sigma}^{-1}(\mathbf{F}(\mathbf{x})-\mathbf{y}) + \gamma(\mathbf{x}-\mathbf{x}_{a})^{T}\mathbf{S}_{a}^{-1}(\mathbf{x}-\mathbf{x}_{a})$$

Adjoint of CTM

$$\mathbf{K}^{T} = \left(\frac{\partial \mathbf{y}_{n}}{\partial \mathbf{y}_{n-1}}\frac{\partial \mathbf{y}_{n-1}}{\partial \mathbf{y}_{n-2}}\cdots\frac{\partial \mathbf{y}_{1}}{\partial \mathbf{y}_{0}}\frac{\partial \mathbf{y}_{0}}{\partial \mathbf{x}}\right)^{T} = \left(\frac{\partial \mathbf{y}_{0}}{\partial \mathbf{x}}\right)^{T}\left(\frac{\partial \mathbf{y}_{1}}{\partial \mathbf{y}_{0}}\right)^{T}\cdots\left(\frac{\partial \mathbf{y}_{n-1}}{\partial \mathbf{y}_{n-2}}\right)^{T}\left(\frac{\partial \mathbf{y}_{n}}{\partial \mathbf{y}_{n-1}}\right)^{T}$$

where

$$\mathbf{K} = \frac{\partial \mathbf{y}_{n}}{\partial \mathbf{x}} = \frac{\partial \mathbf{y}_{n}}{\partial \mathbf{y}_{n-1}}\frac{\partial \mathbf{y}_{n-1}}{\partial \mathbf{y}_{n-2}}\cdots\frac{\partial \mathbf{y}_{1}}{\partial \mathbf{y}_{0}}\frac{\partial \mathbf{y}_{0}}{\partial \mathbf{x}}$$

and y is obs. x is a state vector

During the reverse integration for each iteration, the adjoint model calculates the gradient of cost function to seek the minimum of cost function initiated by the “adjoint forcing” (error weighted difference b/w model predictions and observations by steepest-descent algorithm based on successive calculations

gradient

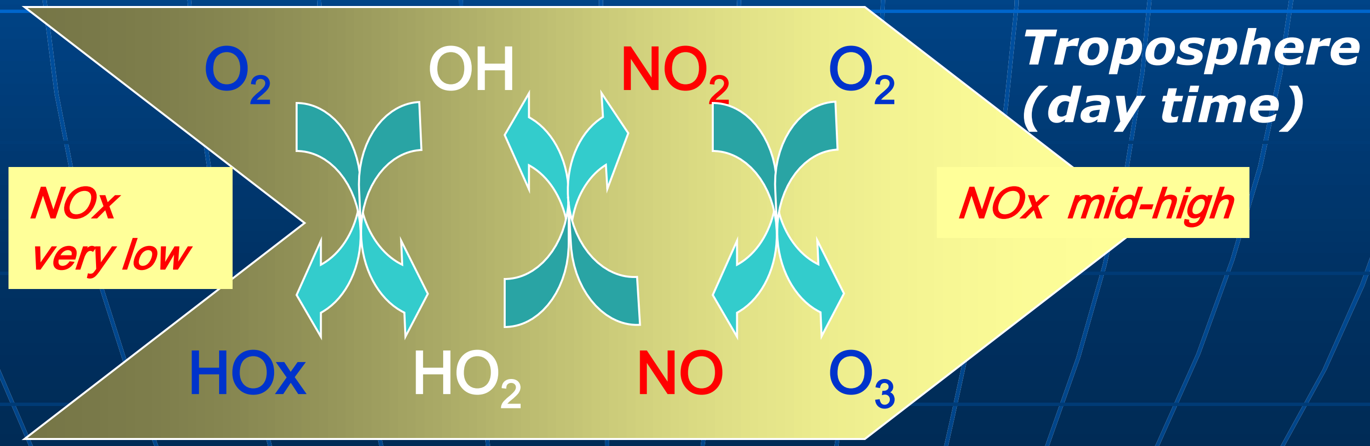
$$\nabla_{x}J(\mathbf{x}) = 2\nabla_{x}\mathbf{F}^{T}\,\underline{\mathbf{S}_{\Sigma}^{-1}(\mathbf{F}(\mathbf{x})-\mathbf{y})} + 2\gamma\mathbf{S}_{a}^{-1}(\mathbf{x}-\mathbf{x}_{a})$$

Adjoint forcing

Inverse modeling for Constraining Global NOx emissions

- Critically affect the capacity to produce O_3 via photochemical processes
  - controlling oxidizing power of atmosphere
    - influence on the lifetime of other GHG (e.g., CH_4)

- Limits in our understanding of NOx emission budget
  (1) A wide variety of sources
  (industry/urban > biomass burning/soil > lightning > etc.)
  (2) Large temporal & spatial variability
  (3) Less understanding in upper tropospheric features (convection, lightning/aircrafts, transport etc..)



Global NOx emissions (GEOS-Chem a priori, 2005 Nov)

NOx emissions	2005 (a priori)	2001 (a)	2005 (b)	%
Industry/urban	23.6 **(1998)** **GEIA**	24	27.9 **(2000)** **EDGAR**	56 ~ 62
Biofuel	2.02 (1995)	2.2	2.03 (2000)	4.5 ~ 5
Soil/fertilizer	5.06 (c)	5.77	5.5	12 ~13
Biomass Burning	6.7 **(Climatological)**	6.5 (d)	5.41 **(GFEDV2)**	10 ~16
Lightning/aircraft	4.2 (f)	4.7	4.5	~10
Total **(Tg/yr)**	42 (v6)	43	45 (v7)	100

(a): adapted from Park et al., (2004)
(b): GEOS-Chem v7-1-3.
(1998): GEIA anthropogenic emission inventory for year 1985 scaled to 1998 by CO2 emission trends [Bey et al., 2001; Marland et al., 1999].
(2000): EDGAR anthropogenic emission inventory based on 2000.
(c): Based on Yienger and Levy et al., (1995).
(d): Climatological monthly biomass burning data (Duncan et al., 2003).
(e): Monthly GFEDv2 biomass burning data.
(f): Based on Wang et al., (1998)

Retrieval of SCIAMACHY NO_2 Columns to map NOx emissions
(from Dalhousie Univ.)

Spectral Fit (429-452 nm)

Total Slant Column

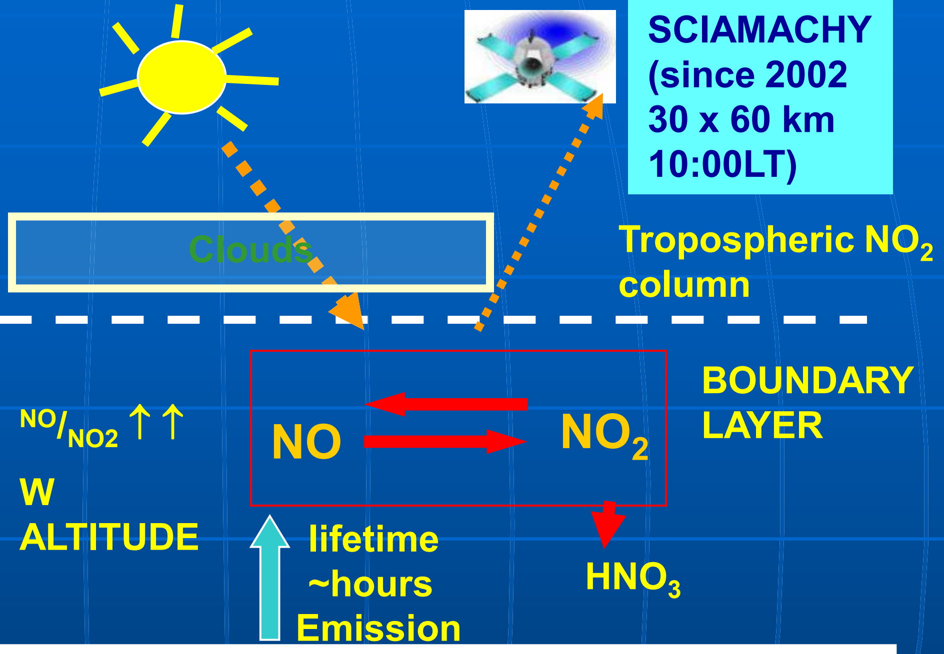
Remove Stratosphere

Tropospheric Slant Column

Calculate AMF

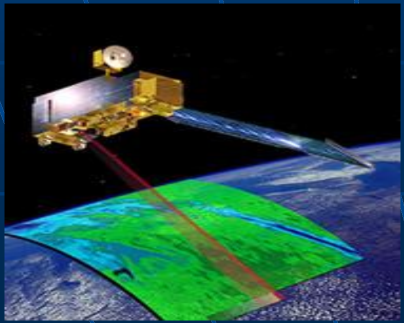
(geometry + NO2 vertical profile + signal sensitivity + aerosol & cloud effect)

Tropospheric NO2 Column



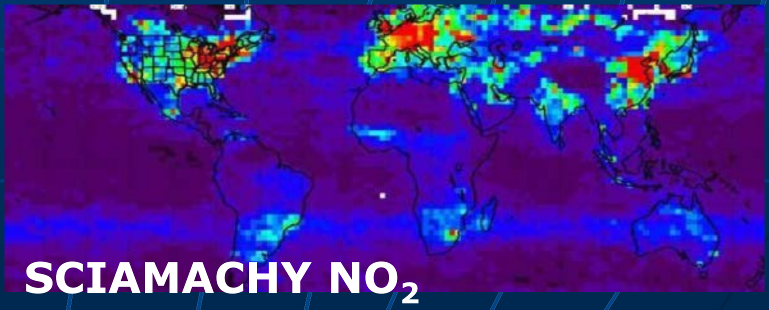
NITROGEN OXIDES (NO_x)

SCIAMACHY



Data retrieval

Global NO2 columns



Adjoint Inversion

- Objective
  - Inversion of NOx emissions with consideration of physiochemical feedbacks with direct computing of parameter's sensitivity
  - Comparison with "top-down" emissions estimates (or mass balance approach) derived from satellite observations (e.g., Martin et al., 2003; 2006)

- Advantage
  - Can consider the chemical and physical feedbacks during optimization
    - → quantifying the parameter's sensitivity w.r.t. model predictions
  - Optimization control

$$J(\mathbf{x}) = (\mathbf{F}(\mathbf{x}) - \mathbf{y})^{\mathrm{T}} \mathbf{S}_{\Sigma}^{-1} (\mathbf{F}(\mathbf{x}) - \mathbf{y}) + \gamma (\mathbf{x} - \mathbf{x}_{\mathrm{a}})^{\mathrm{T}} \mathbf{S}_{\mathrm{a}}^{-1} (\mathbf{x} - \mathbf{x}_{\mathrm{a}})$$

- Disadvantage
  - Still computationally expensive
    - Ex) 64 Intel Itanium2 processors (SGI architecture with LINUX)
      - 1.5 GHz clock speed with 1MB Cache + 1GB RAM
      - With parallel computing (8 CPUs)

      → Each iteration for one month time window (2°x2.5°, globally) takes 44 hours.

Data (Nov. 2005)

- SCIAMACHY NO_2 from Dalhousie Univ (reprocessed data), filtered cloud fraction > 40%.

- CTM, GEOS-Chem is developed by Harvard Univ. and NASA.
- Adjoint of GEOS-Chem v6-2-5 & GEOS-4 & full chemistry (by D. Henze) with 2°x2.5° horizontal resolution
- Time window: one month (Nov. 2005)← a week x 4

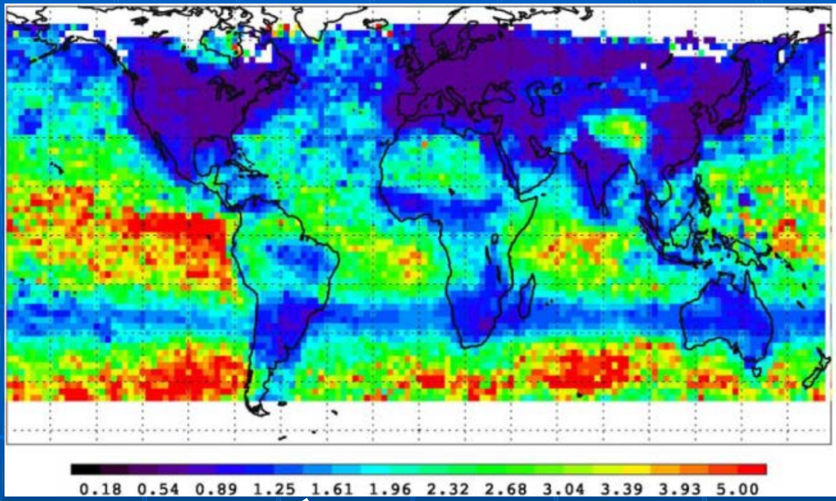
- Emissions (NOx)
  - GEIA anthropogenic NOx emission (scaled to 1998) ←→2005
  - Climatological Biomass Burning (Duncan et al., 2003)
  - Biofuel emissions (Yevich et al., 2003)
  - Soil NOx (Yienger and Levy(1995) & Wang(1998))
  - Lightning NOx (Cloud Top Height; Price and Rind(1998) & Pickering (1998)): only consider the total emissions for opt.
  - Do not optimize the emission scheme → total amount of each type

Error Specification

State vector errors (%)

	N.Am	E.U.	Asia	S.Am.	Africa	R.W
Ind1	50	50	100	100	100	100
Ind2	50	50	100	100	100	100
Light	200	200	200	200	200	300
Soil	150	150	150	150	150	150
BB	150	150	150	150	150	150
BF	100	100	100	100	100	100

Relative instrument errors (N/S)



Observation error = *e1* + *e2* + *e3*

* ***e1***: retrieval error from instrument (SCIAMACHY)

* ***e2***: representation error : ~0.7 of *e1* (~4.0×10^{13} molec/cm^2)

* ***e3***: model transport error (from *Jones et al*., 2003)

→ ~0.8 of *e1* (~4.5×10^{13} molec/cm^2)

total obs. error is about factor of ~2.5 of instrumental (retrieval) error

→ Same quantity of errors were applied to mass-balance approach

Inversion Results

Iteration	Norm of grad.	Cost func. ratio
1	3.405D+03	1
2	1.857D+03	0.90
3	7.530D+02	0.73
4	3.180D+02	0.62
5	1.485D+02	0.52
6	1.112D+03	0.46
7	1.948D+02	0.403
8	8.180D+01	0.397

Cost Function

$$J(\mathbf{x}) = (\mathbf{F}(\mathbf{x}) - \mathbf{y})^T \mathbf{S}_\Sigma^{-1} (\mathbf{F}(\mathbf{x}) - \mathbf{y}) + \gamma (\mathbf{x} - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a)$$

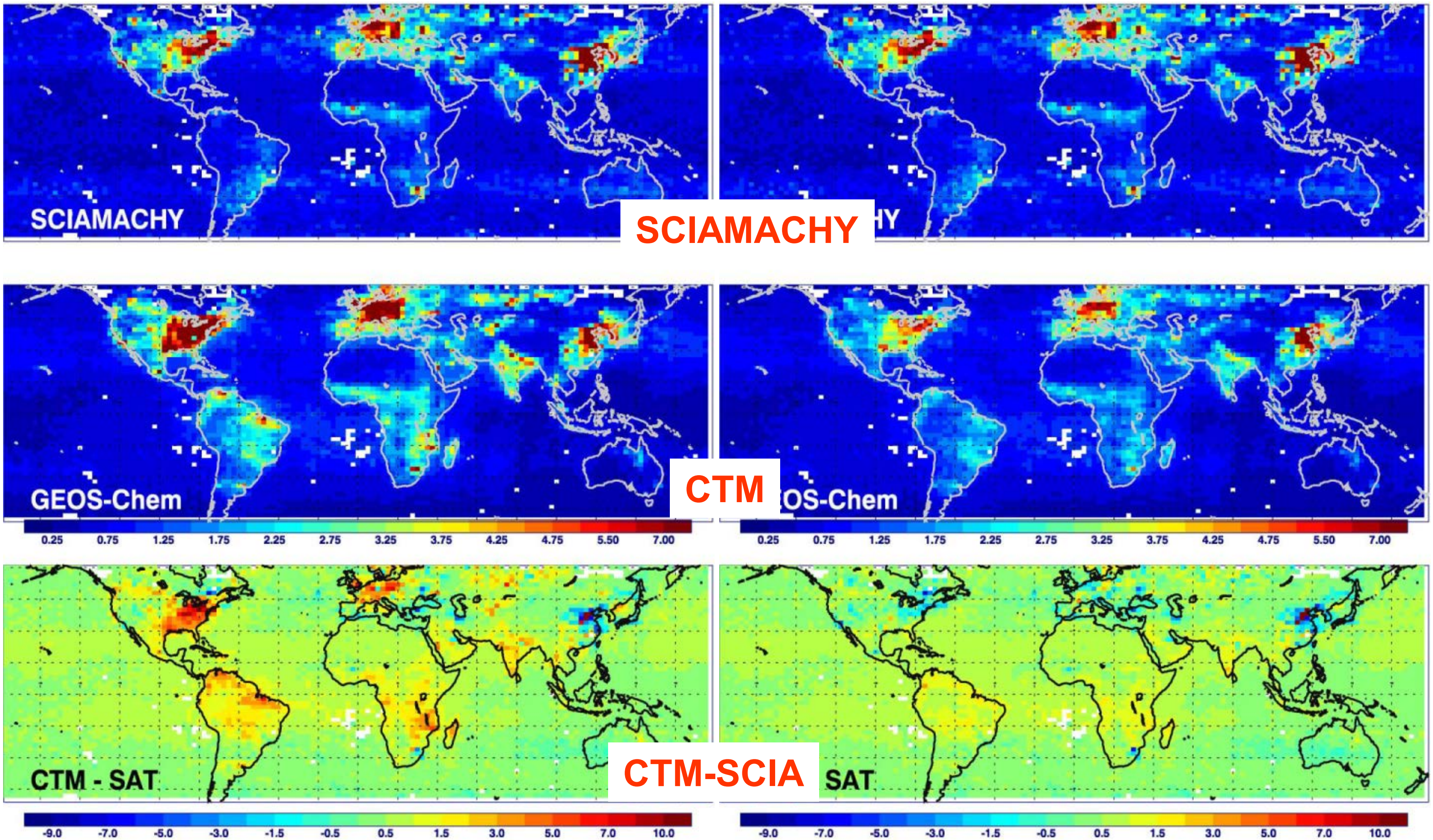
Gradient

$$\nabla_x J(\mathbf{x}) = 2 \nabla_x \mathbf{F}^T \mathbf{S}_\Sigma^{-1} (\mathbf{F}(\mathbf{x}) - \mathbf{y}) + 2\gamma \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a)$$

Cost function from obs. Vs from a priori = ~10: 1

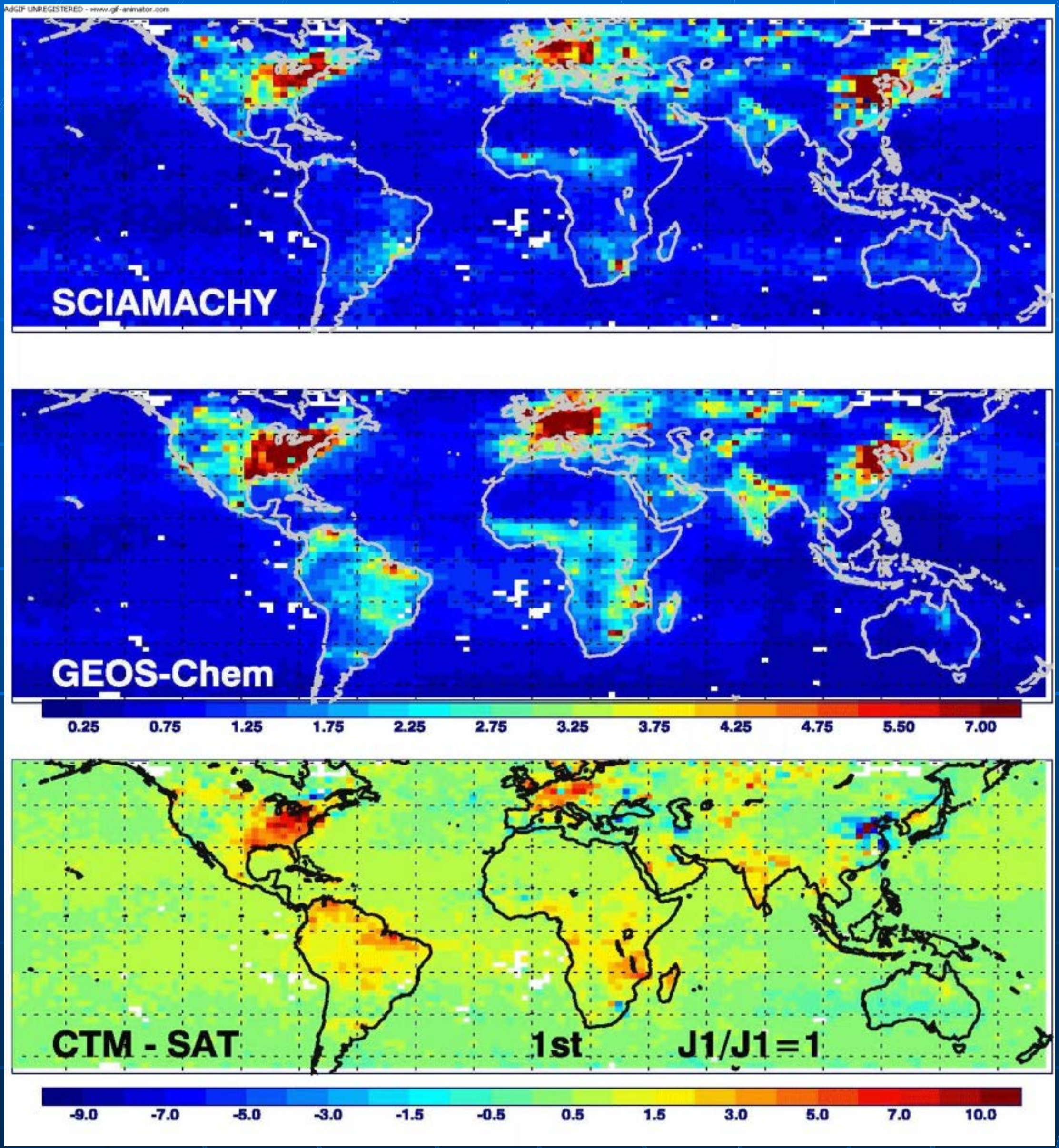
Now cost function reached ~ 0.40 of initial value after 7th iteration

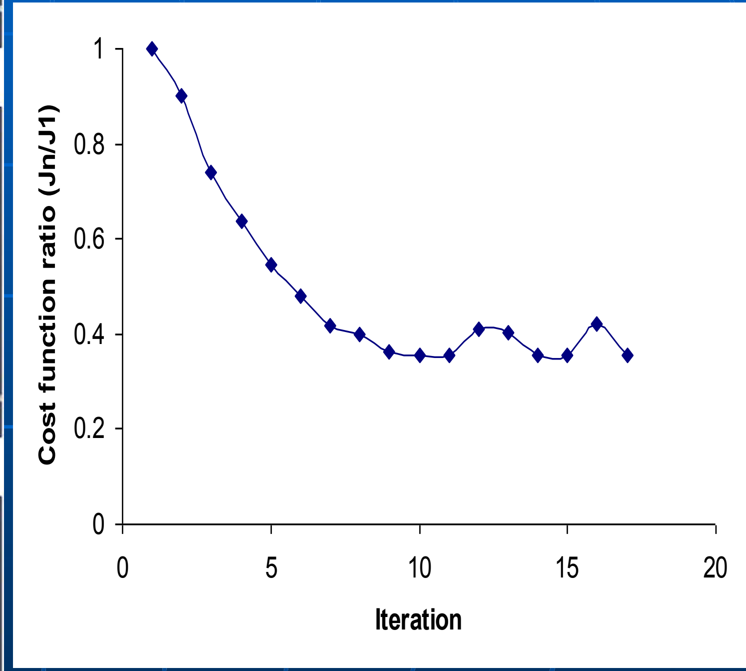
NO_2 columns: a priori vs A posteriori (Unit: 10^{15} molecules/cm^2) Nov. 2005



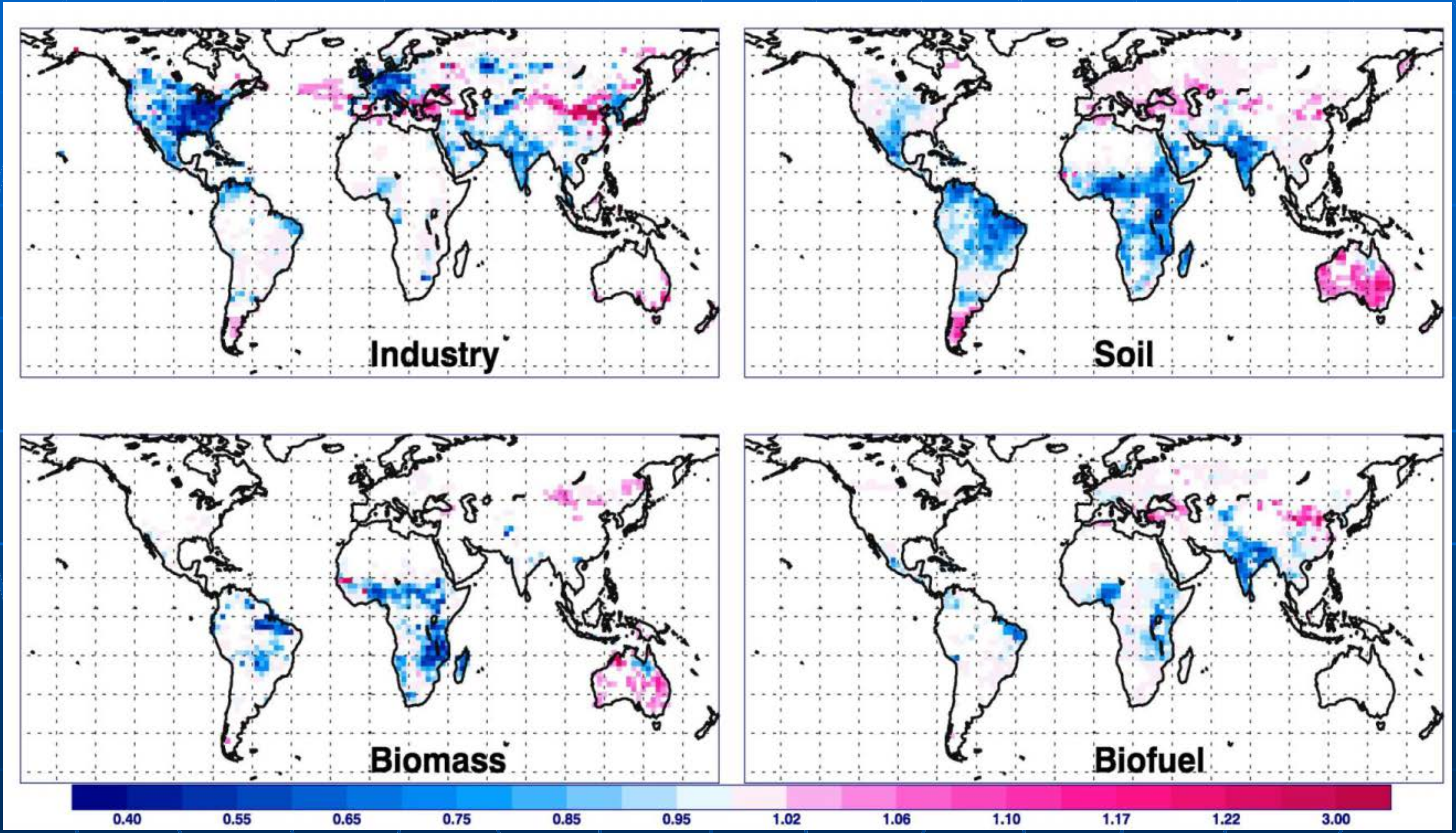
Initial (a priori) *8th iteration (a posteriori)*

Inversion Results (cost func. Reduction)

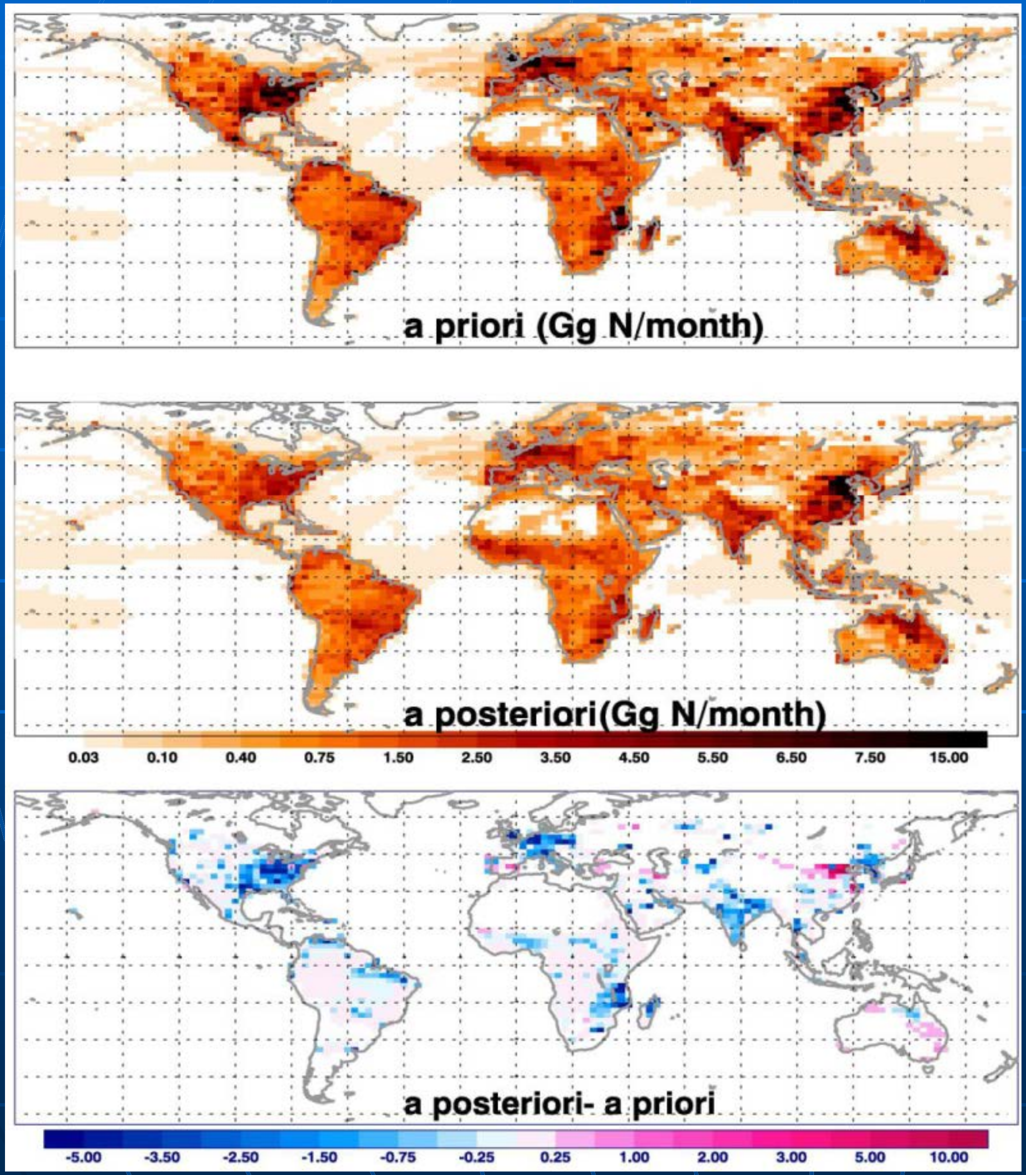




Inversion Results : NOx emissions ratio
(*a posteriori / a priori*)



Total NOx emissions (by adjoint method, Nov 2005)



- Large reductions in N. Ame, Europe, and India
- Moderate reductions in Africa and S. Ame
- China shows mixed features (central China: 10% higher)

Unit: Gg N/ month

A priori / A posteriori (unit: Gg N/ month; Nov 2005)

	N.Am	E.U.	E.Asia	India	S.Ame	Africa	Aus.	R.W.	Total
IND1	430 (209)	220 (154)	384 (342)	95.4 (50.)	74.3 (58)	103 (64)	12.6 (13.6)	165 (123)	1484 (1013)
IND2	176 (85)	202 (136)	7.5 3.6	0	0	14.5 (8.)	9.7 (10.2)	32 (18)	442 (261)
Light.	21.8	10	5.3	1.6	62.3	53.4	12.3	23.3	190
Soil	33.4 (30)	15.4 (15.3)	11.8 (11.6)	34.1 (25.2)	98.8 (85)	142 (115)	38.7 (39.2)	24.8 (24)	399 (345)
BB	5.8 (5.6)	5.1 (5.1)	22.3 (19.2)	3.7 (3.)	126 (77)	276 (173)	94.2 (89.4)	45 (42)	578 (413)
BF	7.5 (6.9)	18.5 (18.2)	51.8 (50.0)	33.3 (25)	14.6 (13.4)	31.5 (27.7)	0.66 (0.65)	24 (23)	182 (164)
Total	682 (364)	490 (357)	535 (481)	202 (130)	391 (286)	652 (451)	169 (166)	338 (273)	3456 (2507)

- Total global NOx emissions are lower by ~28% (Nov 2005): Annual proj. (42 vs 31 Tg N/yr)
- N.Ame (~48%) and Europe (~30%) have significant reduction in industrial NOx emissions (2005 vs 1998)
- A posteriori BB emissions are well matched with GFEDv2 (global total)

A priori / Mass-Balance / Adjoint (unit: Gg N/ month; Nov 2005)

	a Priori	Mass-Balance	Adjoint
N. Ame.	682	424 (-38%)	364 (-47%)
Europe	490	410 (-16%)	357 (-27%)
E. Asia	535	687 (+28%)	481 (-10%)
India	202	126 (-38%)	130 (-36%)
S. Ame.	391	249 (-36%)	286 (-27%)
Africa	652	458 (-30%)	451 (-31%)
Aus.	169	237 (+40%)	166 (-2%)
Global	3456	2877 (-19%)	2507 (-28%)

Conclusions

- Satellite measurements and CTM can study regional ~ global scale air quality and inverse modeling technique can better estimate the initial condition of model inputs from the observations
- According to the adjoint inversion, the N.Ame and European anthropogenic NOx emissions are greatly reduced by 48% and 30% (1998 vs 2005)
- Significant increase in Chinese industrial NOx emissions in 21st is evident by SCIAMACHY, but the adjoint inversion does mixed features
- Natural a priori NOx emissions overestimated (Nov. 2005) and a posteriori biomass burning emission is closer to a newer GFEDv2 inventory in global total

More ..

- Validation of a posteriori emissions by comparison with recent emissions inventory for specific regions (EPA, EMEP, Streets, etc..) will be continued.

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