Adjoint Inversion of Global NOx emissions with SCIAMACHY NO_2

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I. Abstract

We explore the use of satellite observations to constrain the global NOx emissions. The GEOS-Chem chemistry and transport model, driven by assimilated NASA/GMAO GEOS-3 meteorological data (1979-2003), and its adjoint are applied for global inversions of SCIAMACHY tropospheric NO_2 columns. The targeted state vector contributing to tropospheric NO_2 column includes NOx emissions from fossil fuel, biomass burning, biofuel burning, soil, lightning, and NH_3 oxidation. We conducted a "pseudo inversion" of SCIAMACHY NO_2 data from November 2005 using GEOS-Chem results for November 2001, for which the GEOS-Chem adjoint is available (for GEOS-3). Our focus with this pseudo inversion is on demonstrating the utilities of the adjoint approach with satellite observations. Preliminary results from this inversion are presented here. The model overestimation of NOx over the Eastern US and Central Europe are improved by reducing significantly *a posteriori* fossil fuel emissions. The *a posteriori* biomass burning NOx emissions are higher over Siberia and Australia, while biofuel burning NOx contributions are lower over Eastern Europe and Asia. However, these changes may be largely attributed to temporal mismatches between model year (2001) vs. observations year (2005). The implementation of an adjoint for GEOS-Chem driven by GEOS-4 reanalysis (2003 and beyond) is ongoing.

II. Introduction

Atmospheric nitrogen oxide radicals (NOx = NO + NO_2) have profound influences on both tropospheric ozone, a key factor in air quality and climate change, and the hydroxyl radical, the primary oxidizing agent and scavenger of many species. Traditional bottom-up estimates of NOx emissions are highly uncertain due to extrapolation of high spatiotemporal variability of emission fluxes. Additional top-down constraints based on satellite observations are necessary to accurately represent the spatial and temporal variations in emissions. Here we derive top-down NOx emissions with SCIAMACHY tropospheric NO_2 column data using a variational approach with GEOS-Chem adjoint [*Henze et al.,* 2007]. This method allows the use of finer spatial-temporal distribution of emissions and can account for nonlinear chemistry. It relies on the exact and efficient evaluation of the gradient of a cost function with respect to the control variables. The results from the adjoint inversion will be compared with those from a mass-balance method [*Martin et al.,* 2003], thus providing information on the 'smearing' effect due to transport (significant in fall and winter with longer NOx lifetime) in the latter approach.

III. Methodology

SCIAMACHY NO_2 Retrieval

➢On board ESA/ENVISAT satellite launched in March 2002.
-Sun-synchronous orbit, crossing equator 10:00 AM local time in the descending node.
-Nadir view with resolution of 30 km x 60 km (6days for global coverage).

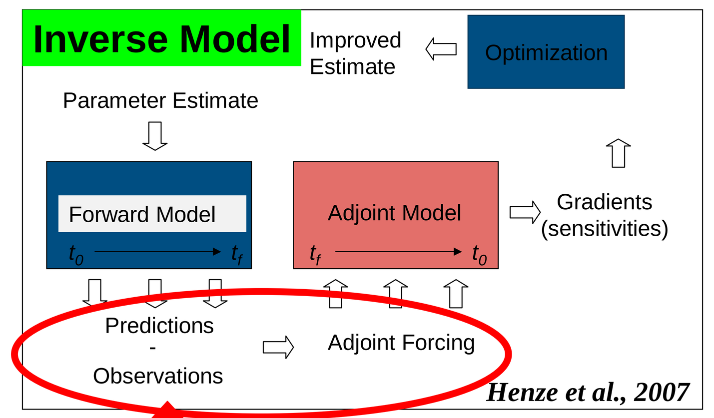
(1) Obtaining total slant columns of NO_2 by direct fitting backscattered radiance spectra (429 – 452nm) with reference spectra [*Chance,* 1998; *Martin et al*., 2002].

(2) Removing stratospheric columns by subtracting central Pacific NO2 columns from total columns [*Martin et al*., 2002] → Tropospheric NO_2 slant columns.

(3) Calculating air mass factor (AMF) to convert tropopospheric NO_2 slant columns into vertical tropospheric NO2 columns [*Palmer et al.,* 2001; *Spurr et al.,* 2001, 2002].
*** Integral of the relative vertical distrution of NO_2 (shape factor) ← GEOS-Chem data.**
*** Shape factor is weighted by local sensitivity of backscattered radiance to NO_2 (scattering weights). ← Linearized Discrete Ordinate Radiative Transfer (LIDORT) model.**

(4) Cloud correction with Fast Retrieval Scheme for Cloud Observables (FRESCO)

GEOS-Chem CTM and its adjoint



Observational operator

1. Mapping model grid to SCIAMACHY grid
2. Applying SCIAMACHY averaging kernel to GEOS-Chem data
3. Calculating cost function & adjoint forcing

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

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

Adjoint forcing

FORWARD MODEL: GEOS-Chem (v6-2-5)

➢Driven by GEOS-3 assimilated meteorological fields (2001 data) from NASA GMAO.
➢ 2°x 2.5° horizontal resolution with 30 vertical laye rs.
➢ O3-NOx-Hyrocarbon chemical mechanisms [*Bey et al*., 2001].
➢Anthropogenic emissions from GEIA inventory [*Benkovitz et al.,* 1996]
➢Climatological monthly biomass burning data [*Duncan et al*.,2003]
➢Soil NOx emissions from *Yienger and Levy* [1995] and *Wang et al*.[1998].
➢Lightning NOx emissions [*Wang et al*.,1998] parameterized by Cloud Top Height (CTH) [*Price and Rind*, 1992] with vertical profiles from *Pickering et al*.[1998]
➢Biofuel burning emissions [*Yevich and Logan*, 2003]

ADJOINT MODEL

Adjoint model is the transpose of its Jacobian Matrix K.

$$\mathbf{K} = \frac{\partial \mathbf{y}_n}{\partial \mathbf{x}} = \frac{\partial \mathbf{y}_n}{\partial \mathbf{y}_{n-1}} \frac{\partial \mathbf{y}_{n-1}}{\partial \mathbf{y}_{n-2}} \cdots \frac{\partial \mathbf{y}_1}{\partial \mathbf{y}_0} \frac{\partial \mathbf{y}_0}{\partial \mathbf{x}}$$

where x is state vector and y is observation and n is given time step.

$$\mathbf{K}^T = \left(\frac{\partial \mathbf{y}_n}{\partial \mathbf{y}_{n-1}} \frac{\partial \mathbf{y}_{n-1}}{\partial \mathbf{y}_{n-2}} \cdots \frac{\partial \mathbf{y}_1}{\partial \mathbf{y}_0} \frac{\partial \mathbf{y}_0}{\partial \mathbf{x}} \right)^T = \left(\frac{\partial \mathbf{y}_0}{\partial \mathbf{x}} \right)^T \left(\frac{\partial \mathbf{y}_1}{\partial \mathbf{y}_0} \right)^T \cdots \left(\frac{\partial \mathbf{y}_{n-1}}{\partial \mathbf{y}_{n-2}} \right)^T \left(\frac{\partial \mathbf{y}_n}{\partial \mathbf{y}_{n-1}} \right)^T$$

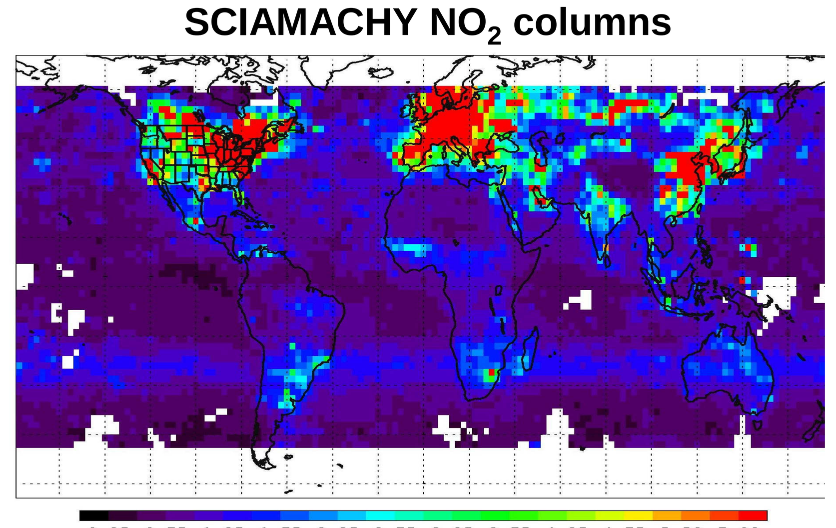
During above reverse integration for each iteration, the adjoint model calculates the gradient of cost function to seek iteratively the minimum of cost function initiated by the "adjoint forcing" (error weighted difference between model predictions and observations).

State vector: Fossil Fuel (industry), Fossil Fuel (point sources), Soil, Lightning, Biomass burning, Biofuel burning, and NH3 oxidation.

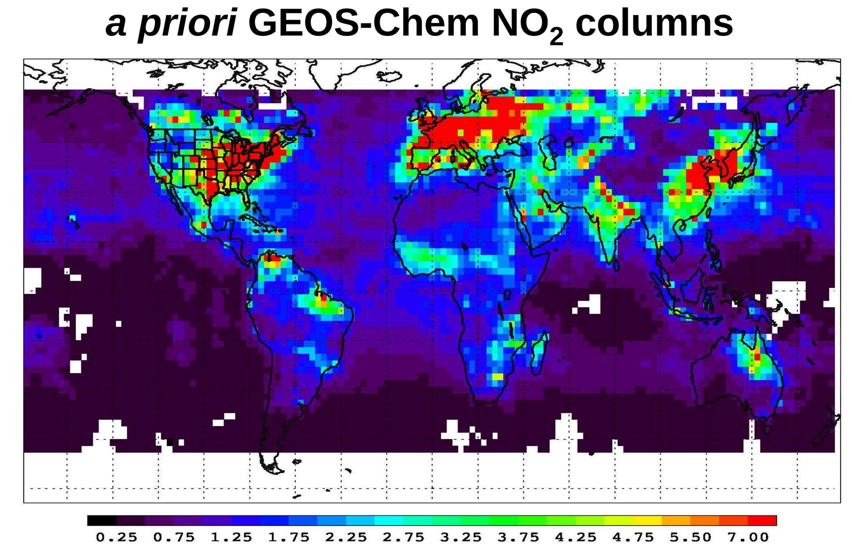
IV. Results

Comparison between SCIAMACHY and GEOS-Chem tropospheirc NO_2 columns (Nov. 2005)

SCIAMACHY NO_2 columns

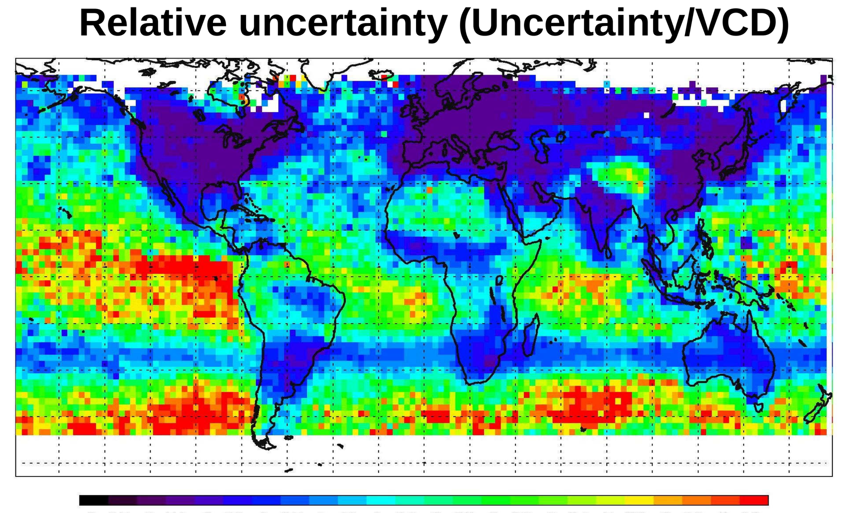


a priori GEOS-Chem NO_2 columns

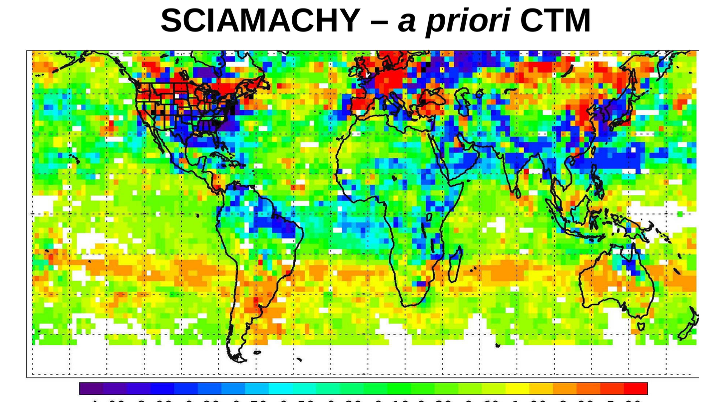


10^{15} molecules/cm^2

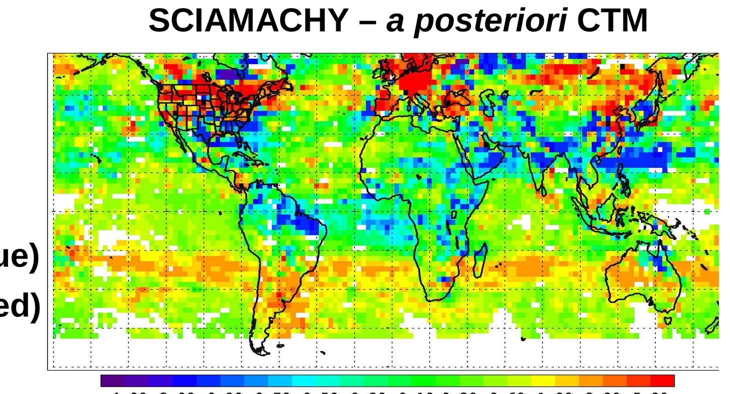
Relative uncertainty (Uncertainty/VCD)



SCIAMACHY – *a priori* CTM



SCIAMACHY – *a posteriori* CTM

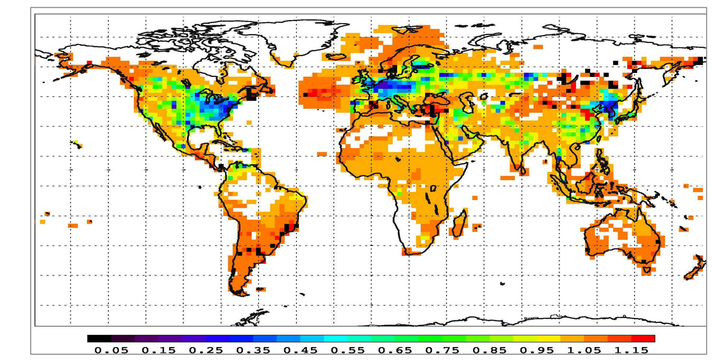


10^{15} molecules/cm^2

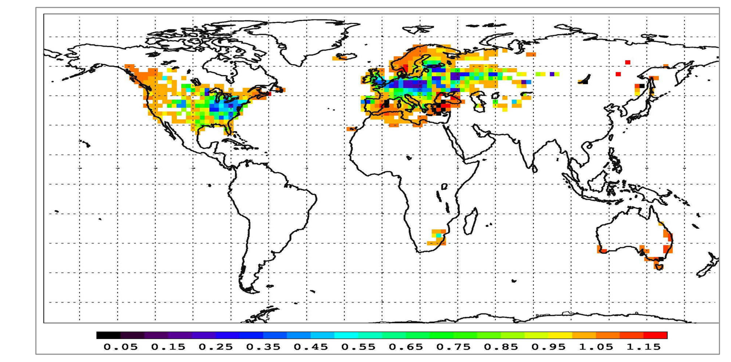
➢Cost function reduction: ~50% after 6 iterations.
➢Some improvements over the overestimated regions (Blue)
➢Little improvements over the underestimated regions (Red)

Resulting Emission Scaling Factors (*a posteriori/ a priori*)

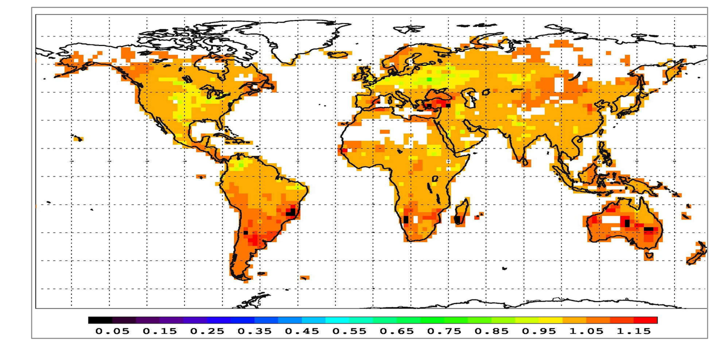
Fossil Fuel (industry)



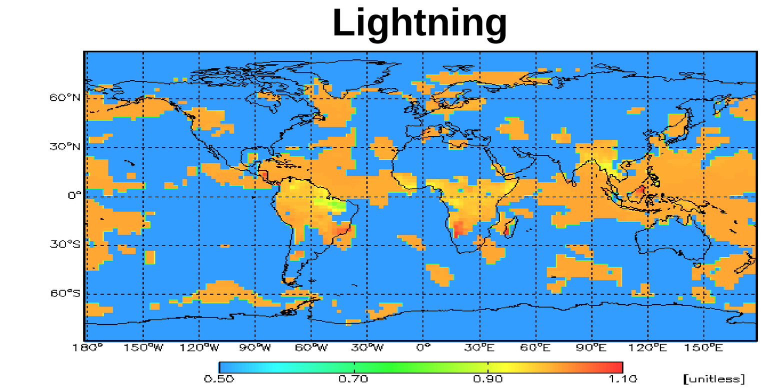
Fossil Fuel (point sources)



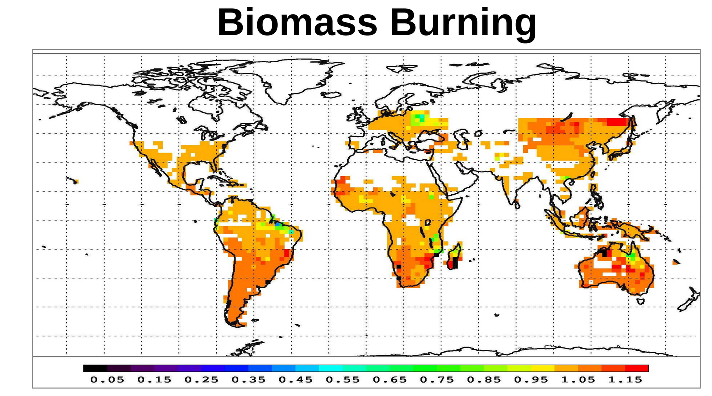
Soil



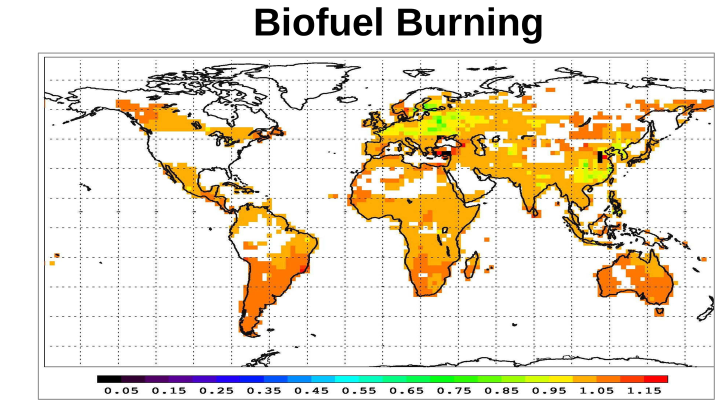
Lightning



Biomass Burning



Biofuel Burning



Global NOx emissions (reference & *a priori*)

Sources	Tg N/yr	%
Fossil fuel	19.9 (24)	31
Biomass B.	12.0 (~9)	18.7
Soil	20.2 (~6)	31.5
Lightning	8.0 (4.7)	12.5
NH_3 oxidations	3	~5
Ocean	<1	<1
Aircraft	~0.5	<1
stratosphere	~0.1	<1
Total	~64 (43)	100

Based on Logan et al. (1983), IPCC (1994), and Davidson et al. (1991)
Reds in ( ) are from a priori GEOS-Chem in 2001 [Park et al., 2004].

➢Significant reductions in *a posteriori* fossil fuel NOx emissions over the Eastern US and Central Europe.
➢ Slightly higher *a posteriori* biomass burning Nox emissions over Siberia and Australia.
➢ Slightly reductions in *a posteriori* soil NOx emissions over central Us and Europe.
➢ *a posteriori* lightning NOx emissions are most uncertain due to temporal mismatches between the model (2001) and the observation (2005).
➢ No changes for NOx sources from NH_3 oxidation.

V. Conclusions & ongoing research.

We presents here some preliminary results of a global NO_2 inversion using the adjoint of GEOS-Chem to constrain various global NOx emissions with SCIAMACHY tropospheric NO_2 columns. Some reductions of discrepancy between the model and the observations may be attributed to the mismatches of emissions and meteorological fields of different years (2001 for the model and 2005 for the observations). The full implementation of adjoint of GEOS-Chem driven by GEOS-4 reanalysis (for 2003 and beyond) are ongoing.

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