First Steps Towards Incorporating Aerosol-Cloud Interactions in Reanalyses

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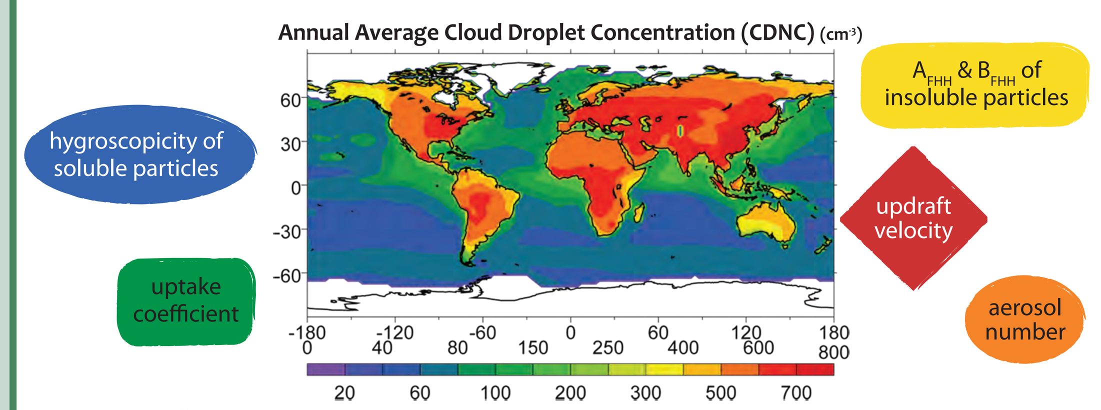
Motivation



Clouds in the Climate System

Through clouds, aerosol concentrations affect the *radiative heat balance* of the earth as well as the *distribution of precipitation*. Drivers of cloud droplet formation include the affinity of aerosol for water, the updraft velocity of the column, and the concentration of aerosol. As aerosol composition or concentrations change, so too may the cloud droplet number concentration (CDNC).

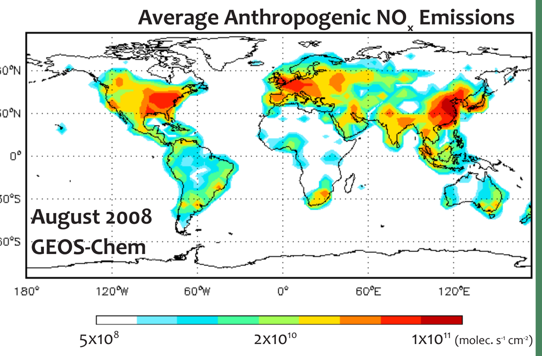
Linking Emissions to Cloudiness

Anthropogenic activity significantly alters emissions of species leading to aerosol formation, including inorganic aerosol precursors such as NO_x and SO_2. Advances in the computational resources have begun to allow the integration of chemical processes with climatic ones. For instance, the coupling of chemical transport models (CTMs) such as GEOS-Chem, which is driven by NASA GMAO's GEOS-5, with the meteorological models formerly used in an offline manner provides a significant opportunity for reanalyses through data assimilation of chemical as well as meteorological variables.

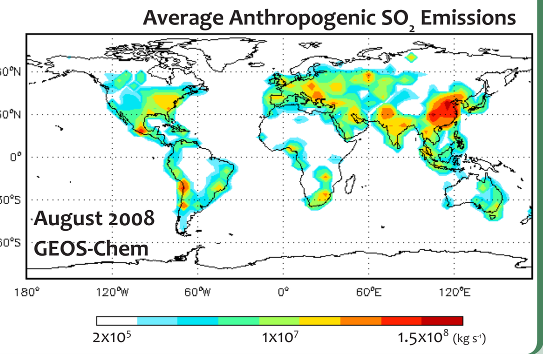
Advancing Reanalyses with CTM Adjoints

The promise of adjoint-based 4D-Var data assimilation with coupled chemistry-climate models motivated the development of the adjoint of a cloud droplet activation parameterization and an inorganic aerosol thermodynamic equilibrium model.

Average Anthropogenic NO_x Emissions



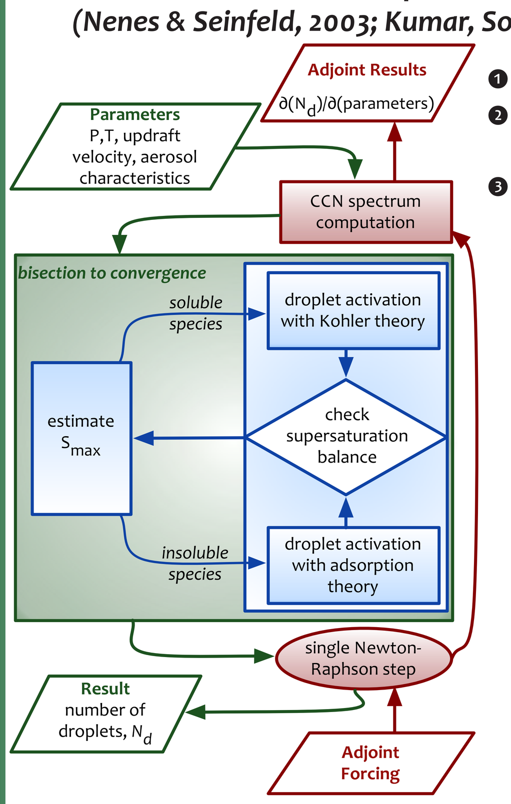
Average Anthropogenic SO_2 Emissions



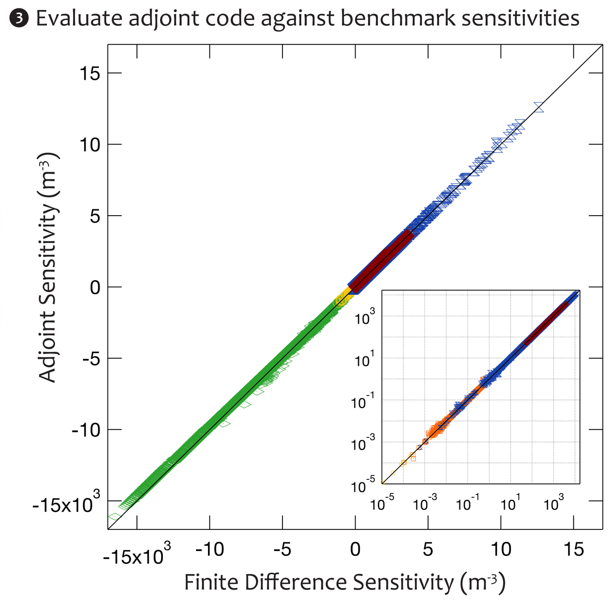
Adjoint Development

Cloud Droplet Activation Parameterization

(Nenes & Seinfeld, 2003; Kumar, Sokolik, & Nenes, 2009; Karydis et al., 2012)

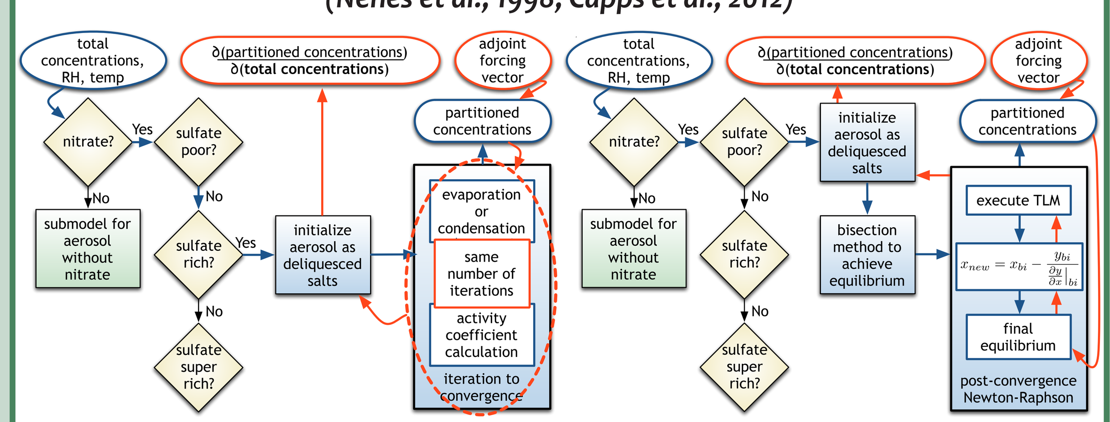


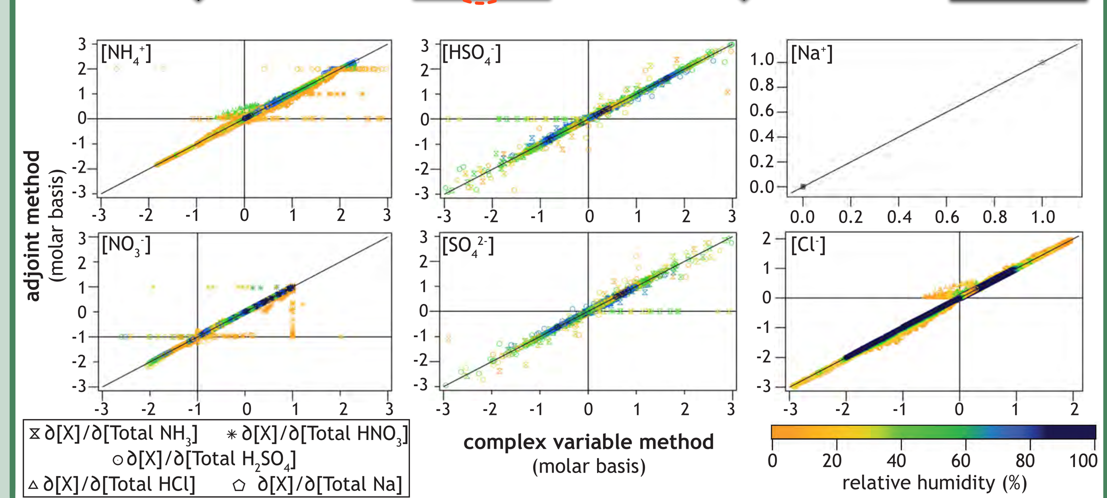
1. Determine the desired sensitivities
2. Manipulate code for application of TAPENADE, an automatic differentiation tool (Hascoët & Pascual, 2004)
3. Evaluate adjoint code against benchmark sensitivities



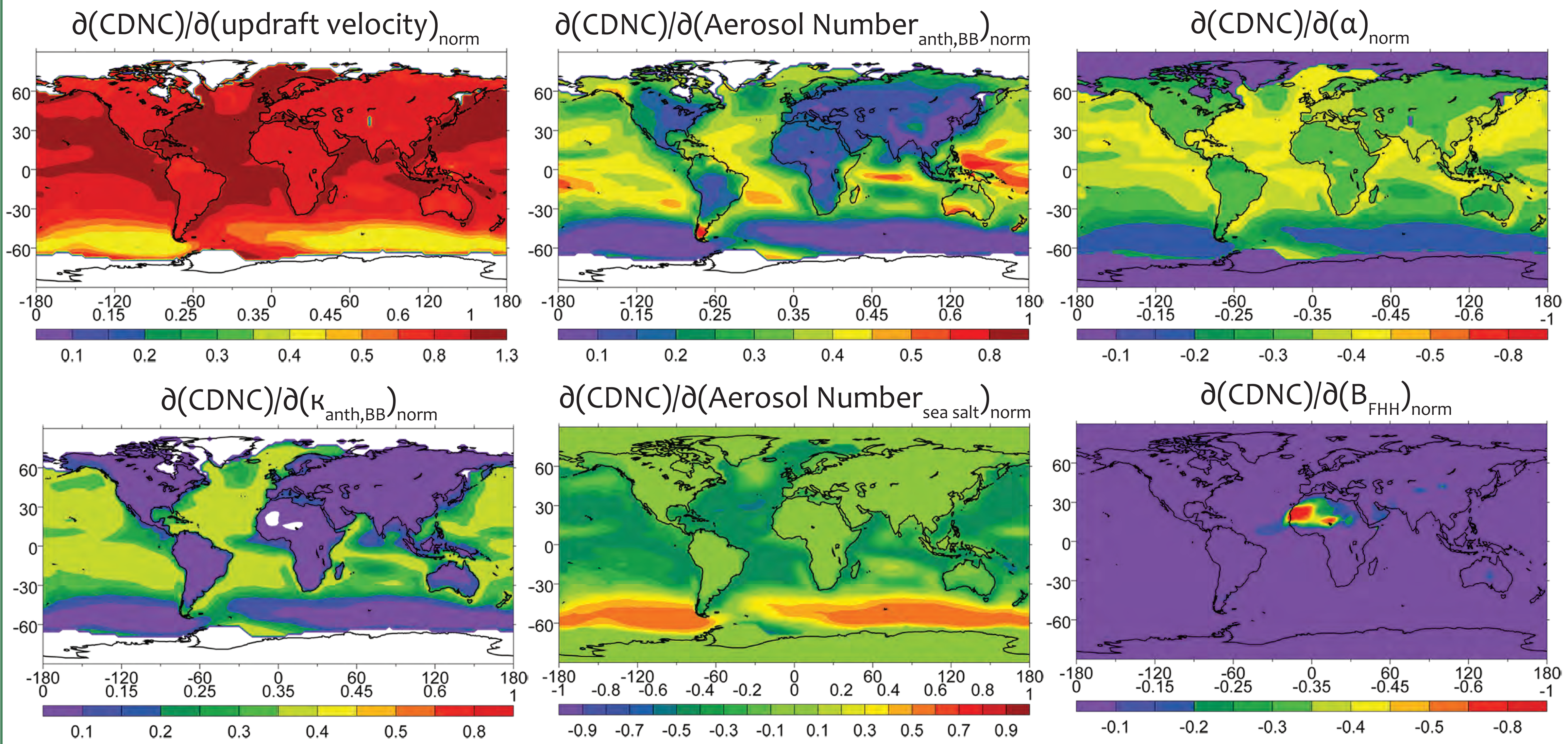
ISORROPIA: inorganic aerosol thermodynamic equilibrium model

(Nenes et al., 1998; Capps et al., 2012)





Elucidating Cloud Droplet Activation Influences



Applying the Parameterization Adjoint

GEOS-Chem was executed for the year December 2007 - December 2008 with an additional year of spin-up. The **annual average aerosol concentrations from 970 hPa** were used to drive the cloud droplet activation parameterization and its adjoint. Based on the grid cell type, an updraft of 0.15 m/s over oceans and 0.3 m/s over land was assumed. Sensitivities of CDNC with respect to each of the parameterization input variables were determined for these annual average concentrations.

Calculation of Normalized Cloud Droplet Number Concentration Sensitivity to Parameterization Input Variable

$$\frac{\partial(\text{CDNC})}{\partial(\text{Updraft})_{\text{norm}}} = \frac{\partial(\text{CDNC})}{\partial(\text{Updraft})} \times \frac{\text{Updraft}}{\text{CDNC}}$$

Evident from the Annually Averaged Sensitivities

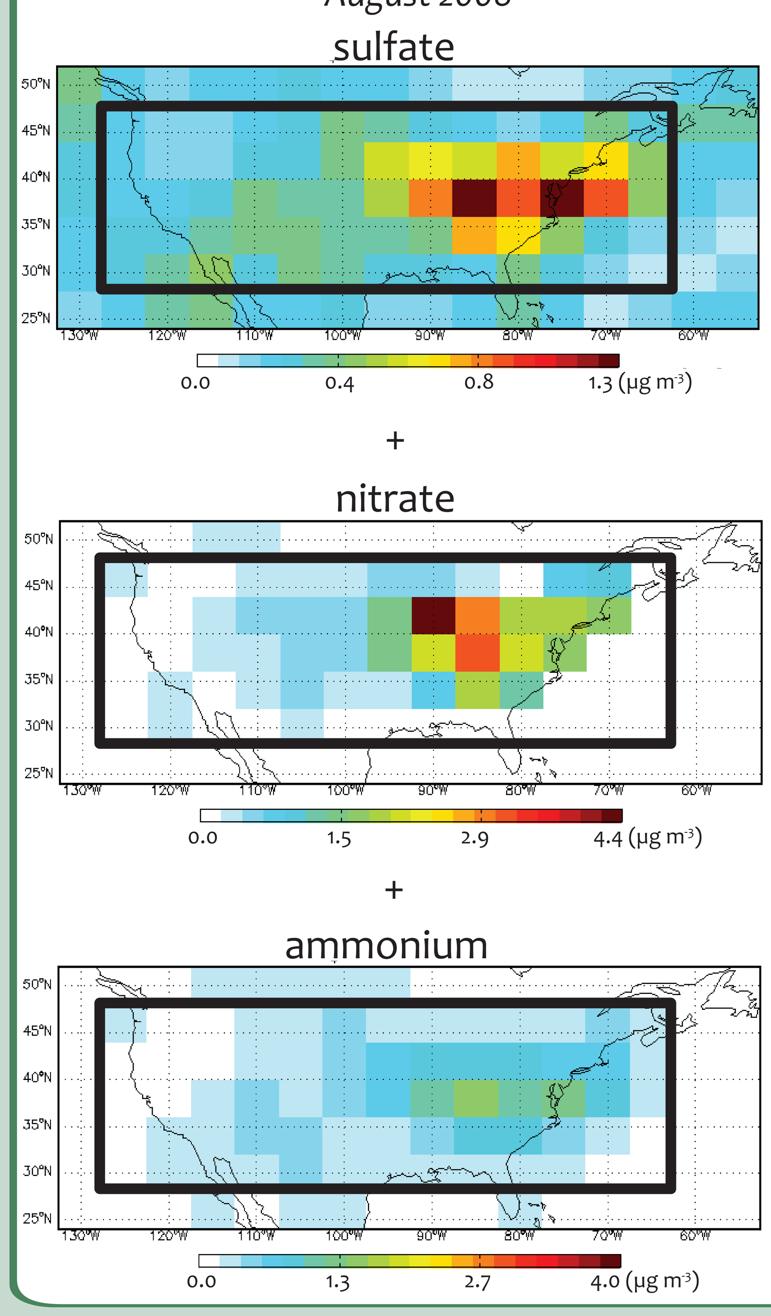
CDNC sensitivities **vary the most for the number concentration** of anthropogenic or biomass burning (bb) aerosol as well as the adsorption parameter, B_{FHH}, for dust. The **effect of large cloud condensation nuclei (CCN)** diminishing the maximum supersaturation is evident in the negative sensitivities caused by dust (B_{FHH}) and sea salt (Aerosol $\text{Number}_{\text{sea salt}}$). The consistently **large magnitude** of the sensitivity to **updraft velocity** implies that accurately estimating this value is important.

Tracing Aerosol Concentration Changes to Emissions

The **adjoint of GEOS-Chem** (Henze et al., 2007) equipped with **ANISORROPIA** (Capps et al., 2012) is used to reveal the influence of anthropogenic emissions on fine mode inorganic aerosol over the continental U.S. for August and February 2008. Sensitivities of concentrations with respect to emissions obtained with the adjoint are able to drive a 4D-Var optimization when observations are available.

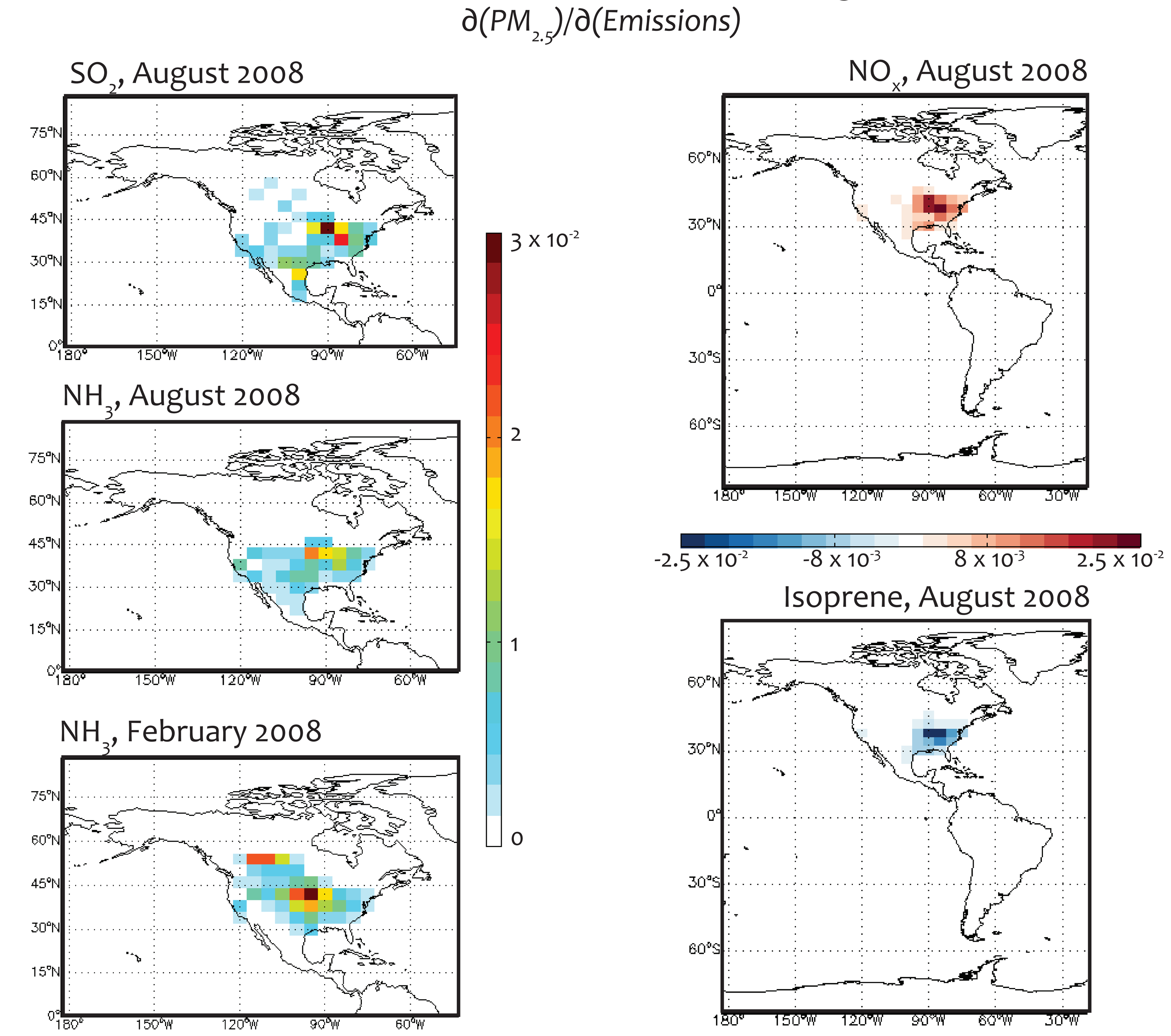
Concentration of Fine Mode Aerosol

August 2008



Sensitivity of U.S. Fine Mode Aerosol to Anthropogenic Emissions

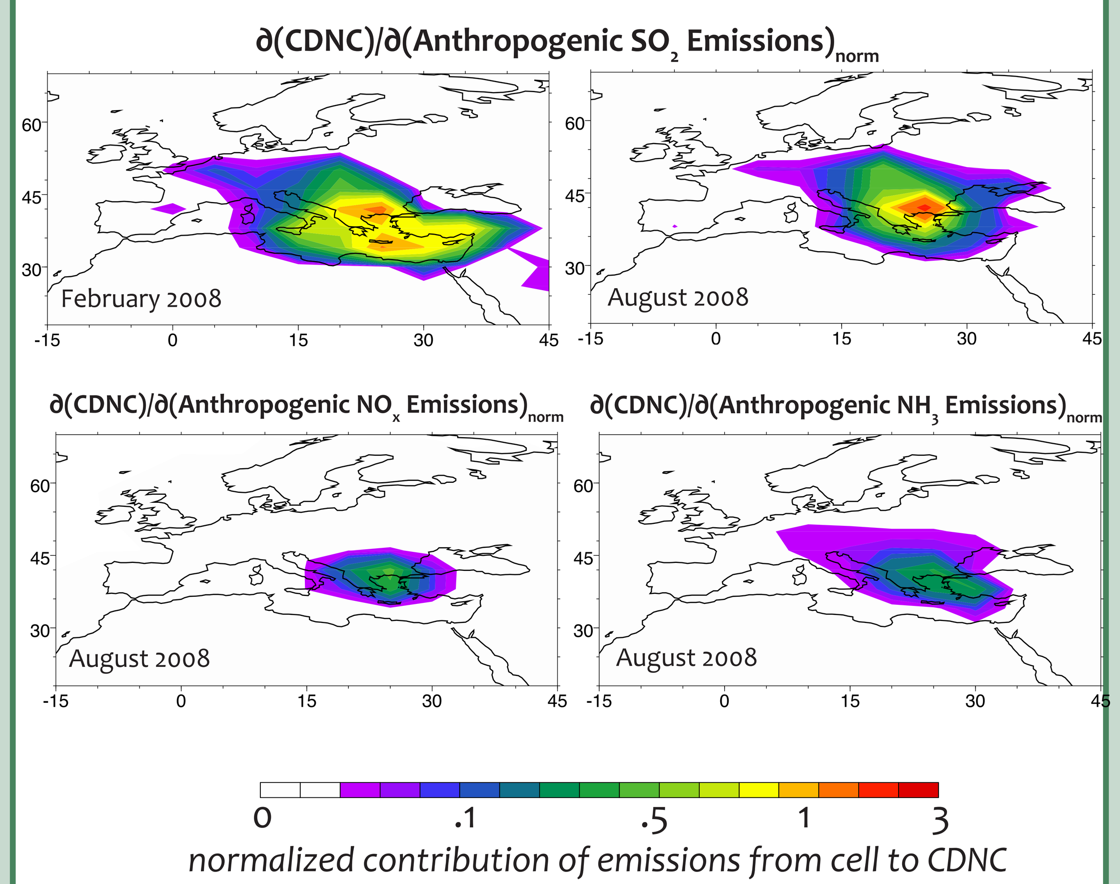
$\partial(PM_{2.5})/\partial(Emissions)$



Cloud Droplet Sensitivity to Emissions

$$\frac{\partial(\text{CDNC})}{\partial(NO_{x,\text{an emis}})} = \frac{\partial(\text{CDNC})}{\partial(\text{Aerosol Number})} \times \frac{\partial(\text{Aerosol Number})}{\partial(NO_{x,\text{an emis}})}$$

Emissions from different sectors of specific species affect cloud droplet number concentrations distinctly. The role of unique sources is approximated by propagating uncertainties in a multiplicative manner. Here, the sensitivity of the cloud droplet number concentration over the eastern Mediterranean region with respect to inorganic aerosol precursor emissions is investigated with the expression shown above.



Next Steps

Coupled chemistry-climate models with full adjoints will provide another means of assessing and refining understanding of the role of aerosol in the climate system. The ***GEOS-5*** meteorological model and data assimilation system is currently being ***augmented to include the cloud droplet activation parameterization of Kumar et al. (2009)***. The ongoing development of a ***column model for GEOS-Chem*** is a step in the direction of coupling these two very complex modeling systems. Within this integrated framework, employing the adjoint models developed for these models independently, including ANISORROPIA and the adjoint of the droplet activation parameterization may lend insight into the ***dynamics of aerosol-cloud interactions***.

$$\frac{\partial(\text{CDNC})}{\partial(NO_{x,\text{an emis}})} = \frac{\partial(\text{CDNC})}{\partial(\text{Aerosol Number})} \times \frac{\partial(\text{Aerosol Number})}{\partial(NO_{x,\text{an emis}})}$$

$$\left.\frac{\partial(\text{CDNC})}{\partial(NO_{x,\text{an emis}})}\right|_{\text{online}}$$

Calculation of CDNC sensitivities concurrent with the sensitivity of aerosol to emissions for each time step and location will enhance the specificity of the result. Addtionally, precision in the association with specific emissions may increase as sensitivities are ***directly propagated*** through a complete adjoint rather than multiplicatively.

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for more information

ANISORROPIA: the adjoint of the aerosol thermodynamic model ISORROPIA (Capps et al., ACP, 2012)
Adjoint sensitivity of global cloud droplet number to aerosol and dynamical parameters (Karydis et al., ACPD)

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