Inverse Modeling of NH_3 Precursor Emissions Using the Adjoint of CMAQ

Matthew Turner[1], Daven Henze[1], Amir Hakami[2], ShunLiu Zhao[2]
[1]University of Colorado at Boulder, Department of Mechanical Engineering, CO, USA
[2]Carleton University, Department of Civil and Environmental Engineering, Ottawa, Canada

Introduction

Air quality models are utilized by the US EPA to develop emission control regulations for air quality improvements, but air quality models contain large amounts of uncertainty. Developing a constraint on emissions using the 4D-Variational data assimilation technique is one method of reducing the uncertainty in sources of aerosols. In an effort to provide more accurate predictive capabilities, the current CMAQ adjoint model will be updated to include an adjoint of aerosol dynamics. Inverse modeling will be performed on the model domain to constrain spatial variability and magnitude of NH_3 emissions. A combination of speciated aerosol concentrations taken from a network of air quality surface monitoring stations, and NH_3 profiles from the TES instrument aboard the Aura satellite will be used. Ammonium measurements will be taken from the AMoN monitoring stations, which provides two-week average measurements of ammonia.

Objectives

This project focuses on reducing uncertainty in sources of aerosols by developing a constraint on emissions using observations with the 4D-Var data assimilation technique. Through this approach, this project will address the following research objectives:

1. More accurately distinguish between natural and anthropogenic sources of aerosol.
2. More accurately distinguish between local and long-range sources of aerosol.
3. Better predict the effects that policy change will have on the future evolution of atmospheric composition.

Ammonium sulfate and ammonium nitrate consist of approximately half of the average mass concentration in the United States. Our analysis will allow us to quantify their contribution to air quality, from uncertainty in their precursor emissions to effectiveness of their control strategies. Inverse modeling of additional species that contribute to $PM_{2.5}$ is not considered here as their formation mechanism is uncertain, or there are not enough widespread observations to warrant data assimilation at this time.

Motivation

Particulate Matter (PM) is an air pollutant consisting of a mixture of solid and liquid particles suspended in the air. Knowledge of PM concentrations is important for many reasons, two of which are that PM has an adverse effect on human health, and PM also has an effect on climate change.

•Currently no other regional chemical transport model has adjoint that includes aerosols.

•Previous inverse modeling studies of NH_3 emissions used ammonium (NH_4^+) concentrations, as ammonia concentration data was not available (Gilliland et al., 2003)

- Confirmed anticipated strong seasonal differences in NH_3 emissions
- Results suggested USEPA 1990 National Emission Inventory for NH_3 was approximately 20% too high.

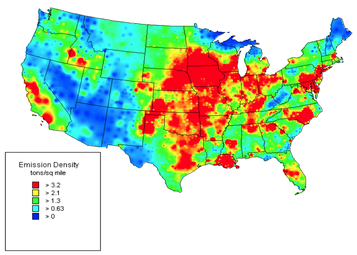
•Current estimates of NH_3 emissions can reach over 3 tons/sq mile yr (see Figure 1)

- Uncertainty in global NH_3 emissions estimated to be 27-38%, with regional uncertainties surpassing 100% (Beusen et al, 2008)

•Total global NH_3 emissions have increased by a factor of 5 since pre-industrial times (IPCC, 2007)

- Global NH_3 emissions are projected to increased by a factor of 10 between pre-industrial times and 2050

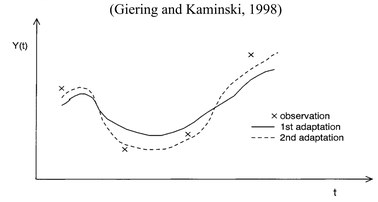
Figure 1: Density map of 1998 ammonia emissions (US EPA, 2000)



What Are Adjoints?

- Adjoint models are used for studies that require an estimate of sensitivity of a model output with respect to an input.
- In air quality studies, adjoints are often used for data assimilation
  - Atmospheric chemical transport models have large amount of uncertainty
  - An adjoint model used in conjunction with the 4DVar data assimilation technique allows model trajectory to be brought as close as possible to observed data (see Figure 2) by varying control variables (emission scaling factors, initial condition scaling factors, etc.)
- To quantify misfit of model prediction, a cost function is introduced
  - Cost function is reduced through an iterative process
- Adjoint method has 2 main advantages over finite difference:
  - Especially for large number of parameters, adjoint model saves run time
  - Computed gradient is exact

Figure 2: Schematic representation of variational methods (Giering and Kaminski, 1998)



CMAQ Adjoint Development

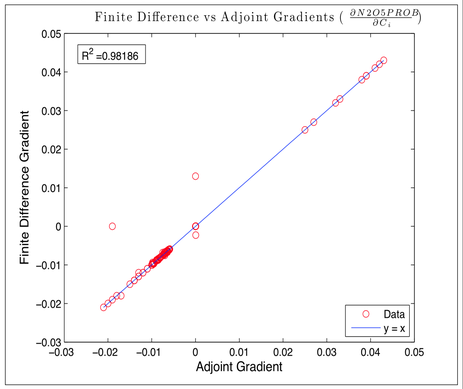
Development Goals

- Consider each aerosol module separately
- Construct adjoint manually
- Validate adjoint via comparison to finite difference

Heterogeneous Chemistry

- Adjoint developed for HETCHEM and N2O5PROB
  - HETCHEM calculates heterogeneous conversion of N_2O_5 to HNO_3
  - N2O5PROB calculates the N_2O_5 heterogeneous reaction probability
- Two-sided finite difference approach was used
- Gradient data generated using aerosol concentrations in first vertical layer of grid cell containing Los Angeles, CA.
  - Simulation performed between 0:00 UTC June 3, 2004 and 0:00 UTC June 7, 2004
- Finite difference gradients compared to adjoint gradients (see Figure 2)
  - Majority of points lie on $y = x$ line, with few outliers
  - Point corresponding to adjoint gradient -0.019 and finite difference gradient 0 is calculated gradient with respect to nitrate corresponding to initial conditions
    - Finite difference fails as finite difference perturbation is outside numerical precision of computer
  - Two other outliers both correspond to adjoint gradients of 0
    - These outliers caused by nonlinearities in forward model

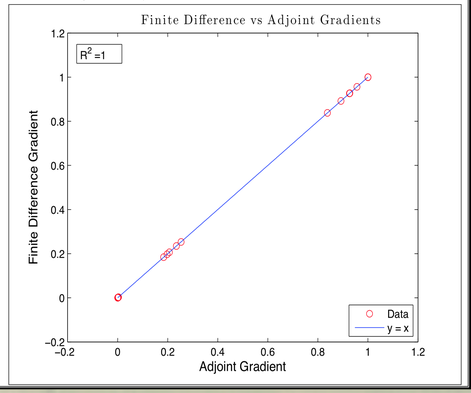
Figure 2: Adjoint verification for subroutine N2O5PROB



Coagulation

- Adjoint developed for GETCOAGS
  - Calculates coagulation rates using approximate algorithm for 2nd moment
- Finite difference gradients compared to adjoint gradients (see Figure 3)
  - All points lie on $y = x$ line, with no outliers
  - R^2 value exactly 1
  - Data shows some points with positive finite difference gradient and negative adjoint gradient.
    - These points are on order 10^{-29}
    - Discrepancies caused by round-off and Truncation within the forward model

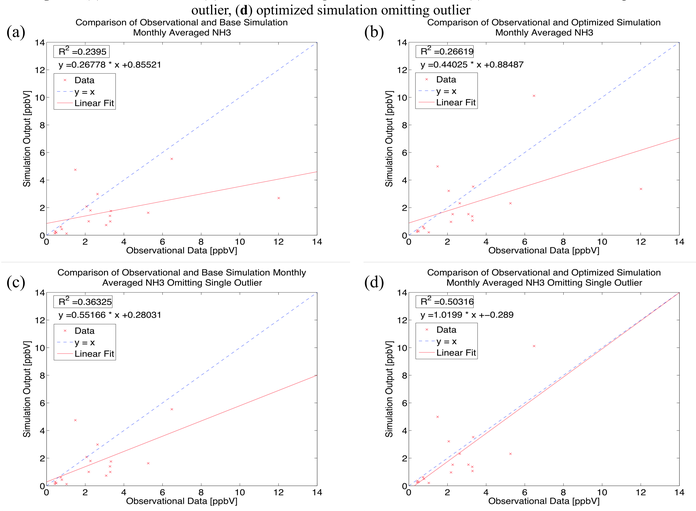
Figure 3: Adjoint verification for subroutine GETCOAGS



Inverse Modeling

- Inverse modeling of NH_3 performed using optimized scaling factors provided by GEOS-Chem
- GEOS-Chem adjoint run on 4° x 5° horizontal grid
- Optimized emissions scaling factors obtained through inverse modeling within GEOS-Chem
  - Ammonia concentration from TES instrument aboard NASA's Aura satellite were used.

Figure 4 (**a**) base simulation, (**b**) simulation with optimized scaling factors, (**c**) base simulation omitting outlier, (**d**) optimized simulation omitting outlier



- CMAQ simulation run on 36 km horizontal grid for April, 2008
- Simulation results compared to ammonia concentrations from AMoN
- Figure 4a is comparison of observed data to CMAQ simulation without scaling factors.
  - For majority of data points, CMAQ under predicts the concentration of NH_3.
- Figure 4b is comparison of observed data to CMAQ simulation that uses optimized scaling factors
  - Although simulation results do not match observations, model predictions using scaling factors are closer to observations than model predictions without scaling factors
- Figures 4c and 4d are comparisons of observed data to model predictions for no scaling factors (c) and simulations with scaling factors (d) after removal of single outlier
  - Statistical analysis shows cook's distance for outlier is 0.519, which is much larger than critical cook's distance of $\frac{4}{n}$ where n is number of observations
  - In this case, the critical cook's distance is 0.222.

Conclusions

- Adjoint code has been generated and verified for both HETCHEM and GETCOAGS aerosol modules.
- Based on GEOS-Chem and CMAQ simulations, it can be concluded that these models generally under predict concentrations of ammonia.
- When compared to independent observations, inverse modeling is demonstrated to improve bias, but not variance; suggesting that model variance of NH3 may be owing to other issues such as transport, deposition, and resolution error.

References

1. AB Gilliland, RL Dennis, SJ Roselle, and TE Pierce. Seasonal NH3 Emission Estimates for the Eastern United States Based on Ammonium Wet Concentrations and an Inverse Modeling Method. J. Geophys. Res. Atmos., 108(D15):4477, Jan 2003.
2. AHW Beusen, AF Bouwman, PSC Heuberger, G Van Drecht, and KW Van Der Hoek. Bottom-up Uncertainty Estimates of Global Ammonia Emissions from Global Agricultural Production Systems. Atmos. Environ., 42(24):6067–6077, Jan 2008.
3. US EPA. National Air Pollutant Emission Trends Report, 1900-1998. Mar 2000.
4. KL Denman, G Brasseur, A Chidthaisong, P Ciais, PM Cox, RE Dickinson, D Hauglus- taine, C Heinze, E Holland, D Jacob, U Lohmann, S Ramachandran, PL da Silva Dias, SC Wofsy, and X Zhang. 2007: Couplings Between Changes in the Climate System and Biogeochemistry. Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press., Jun 2007
5. R Giering and T Kaminski. Recipes for Adjoint Code Construction. ACM T. Math. Software, 24(4):437–474, Jan 1998.

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