Predicted Responses of Inorganic PM to Emission Changes For a Chemical Transport and Observation Based Models

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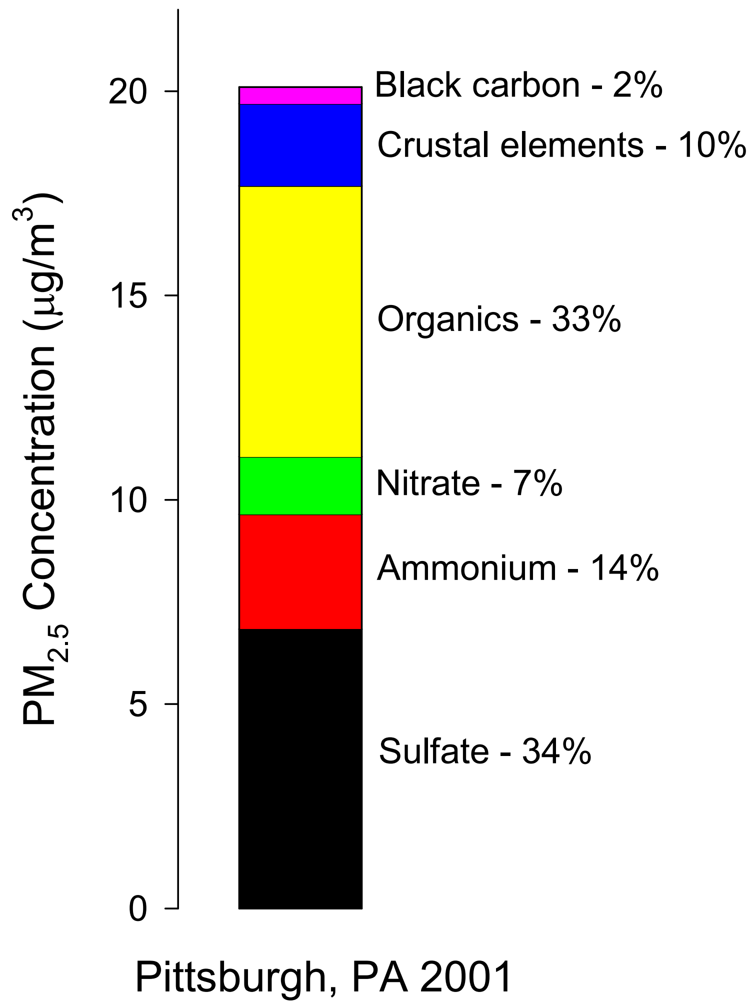
Introduction

- In the Eastern US, inorganic components account for approximately 50% of $PM_{2.5}$ mass.
- **OBJECTIVE:** Understand how inorganic $PM_{2.5}$ mass responds to changes in emissions of precursors.



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Approaches

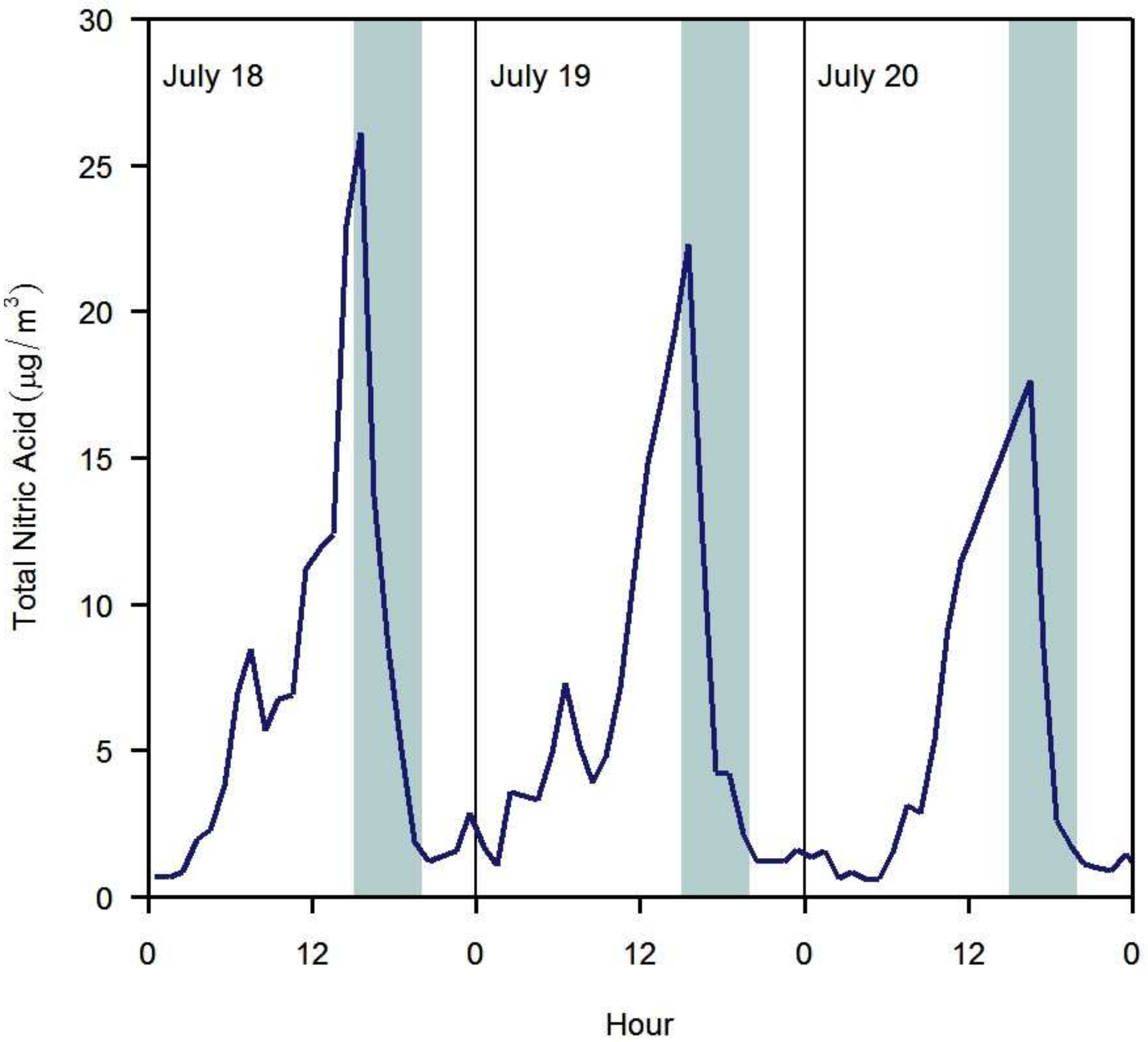
- 3-D chemical transport model (PMCAMx)
  - directly links emissions to $PM_{2.5}$ concentrations with detailed physics and chemistry
  - large uncertainties in inputs (emission inventories and assimilated meteorological data)
- Observation-based approaches are proposed as alternatives
  - rely on measurements of Temperature, RH, Sulfate, Total Nitric Acid, Total Ammonia (Total = $PM_{2.5}$ + gas)
  - simplify description of atmospheric transport processes

GFEMN

- Chemical equilibrium model
- Directly minimizes Gibbs Free Energy of system (gas and aerosol phases) [Ansari and Pandis, 1999]
- Limitations for use in evaluation of emission control strategies:
  - GFEMN implicitly assumes that when the concentration of one species is changed, the concentration of other species remain constant.
  - The partitioning of semi-volatile species between the gas and aerosol phases can affect their lifetime (and hence availability) in the atmosphere (Pandis and Seinfeld 1990).

Rapid Deposition of Nitric Acid

(Measurements for Pittsburgh, July 2001)



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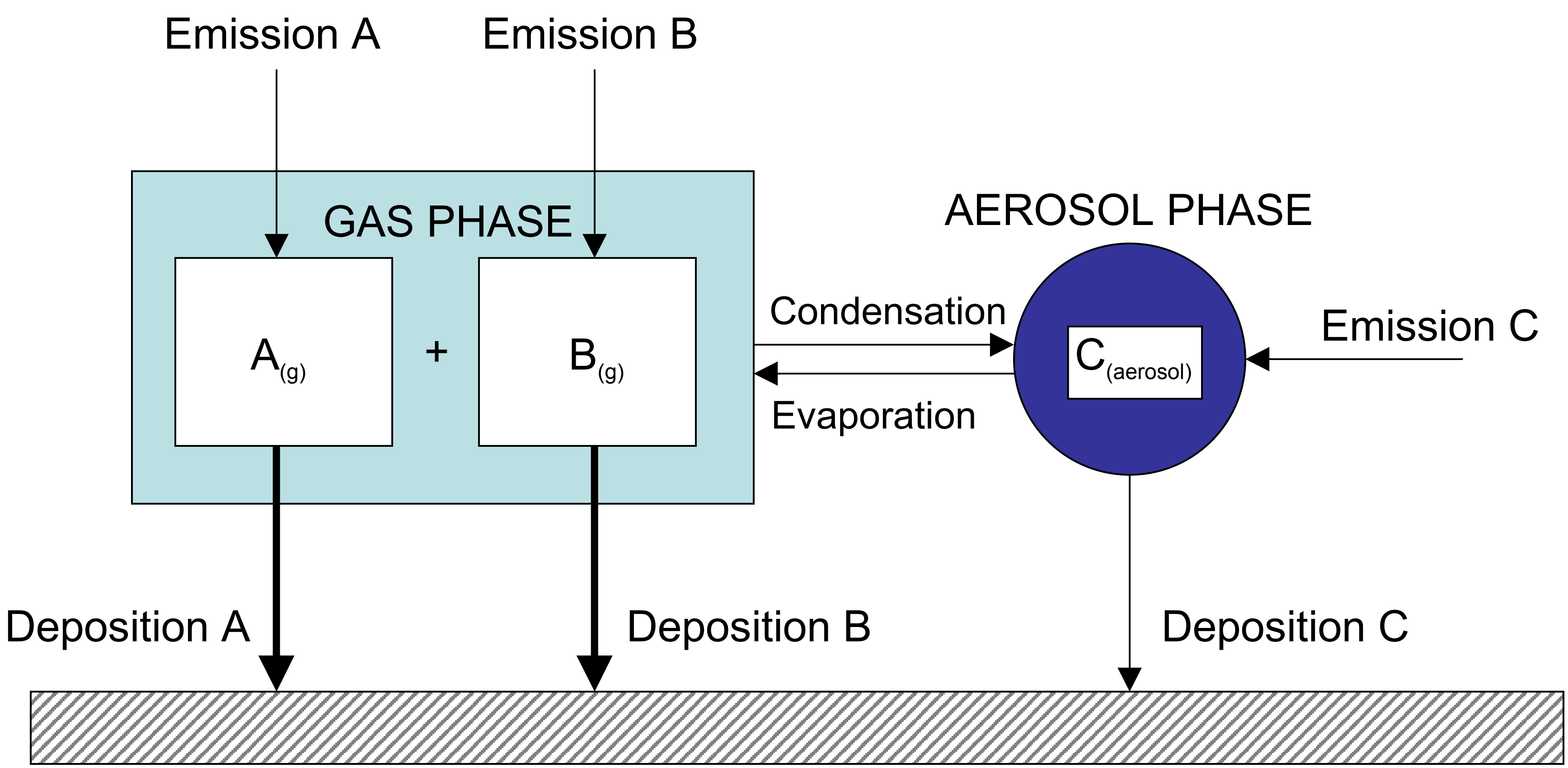
Interaction of Condensation/ Evaporation and Dry Deposition



adapted from Pandis and Seinfeld (1990)

Thermodynamic Model with Removal (TMR)

Eulerian box model formulation [Seinfeld and Pandis, 1998]

$$\frac{dc_i}{dt} = \left(\frac{\partial c_i}{\partial t}\right)_{\text{cond/evap}} - \frac{v_i}{H(t)} c_i + \frac{q_i}{H(t)} + R_i + \frac{c_i^0 - c_i}{\tau_r} + \frac{c_i^a - c_i}{H(t)} \frac{dH}{dt} \Phi\left(\frac{dH}{dt}\right)$$

- If we assume that the area of interest is relatively homogeneous $c_i \approx c_i^0 \approx c_i^a$, equation is simplified to

$$\frac{dc_i}{dt} = \left(\frac{\partial c_i}{\partial t}\right)_{\text{cond/evap}} - \frac{v_i}{H(t)} c_i + P_i(t)$$

- $P_i(t)$ is the chemical production rate with small contributions from horizontal advection and vertical entrainment, and is estimated from the observations
- Differential equation solved for (1) nitric acid and (2) $PM_{2.5}$ nitrate

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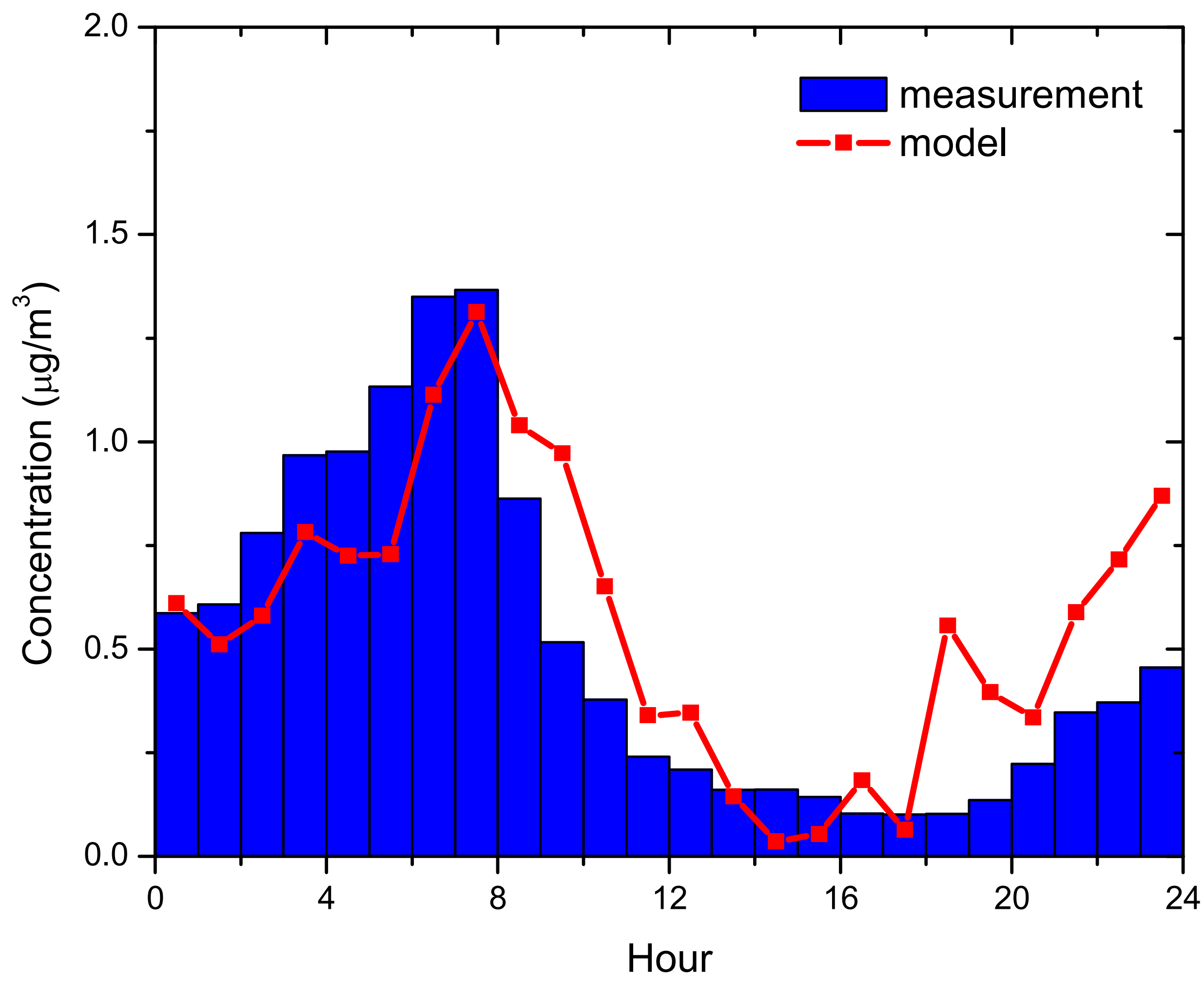
PMCAMx

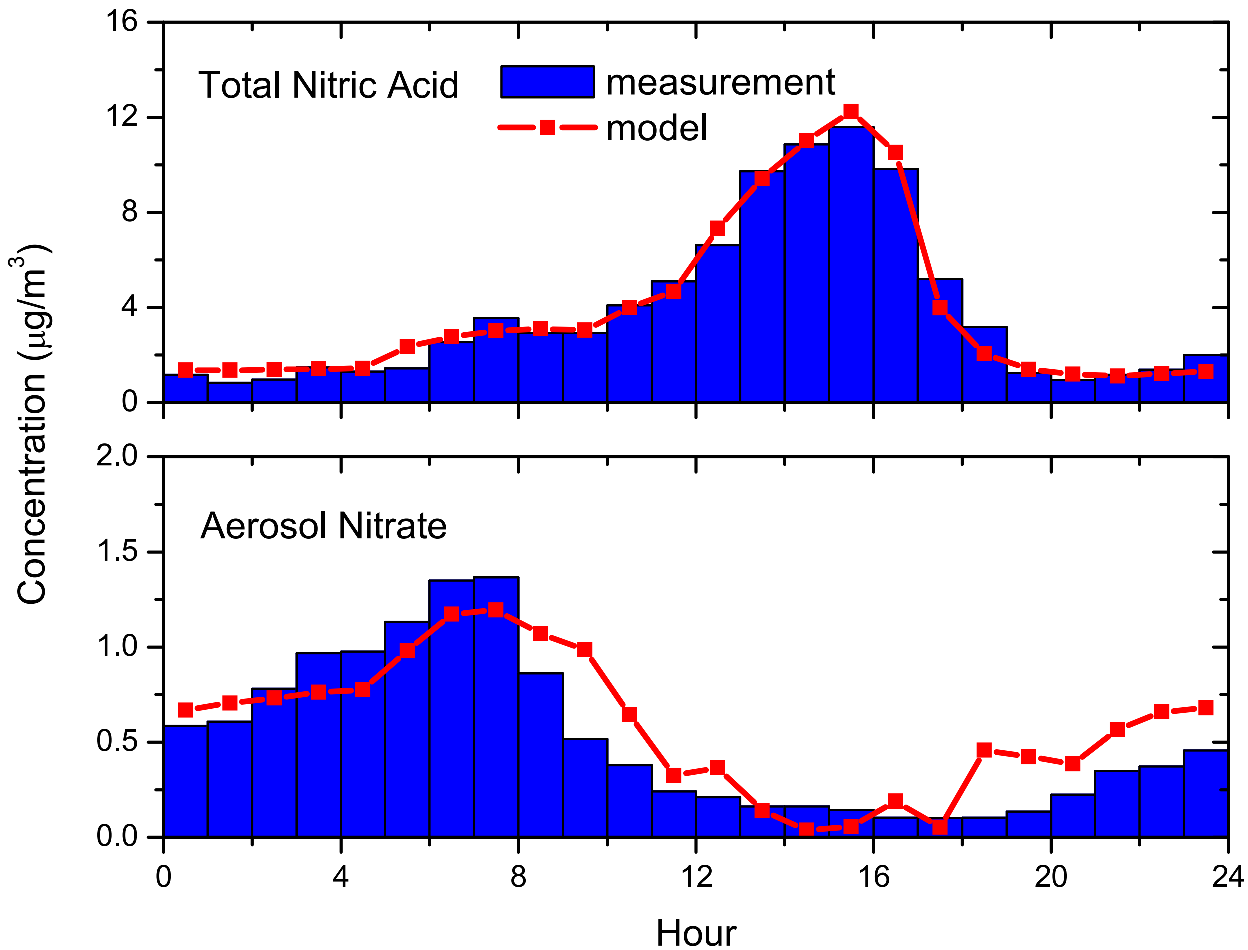
- 3-D Chemical Transport Model
  - Uses emission data and meteorological data to predict PM concentrations over a period of time
  - 3-D grid framework (36 km x 36km grid squares, 14 vertical layers)
- Modeling domain: Eastern US
- Uniform reductions in SO_2 emissions

GFEMN Evaluation for $PM_{2.5}$ Nitrate
(Pittsburgh, July 2001)



TMR Evaluation for $PM_{2.5}$ Nitrate

(Pittsburgh, July 2001)



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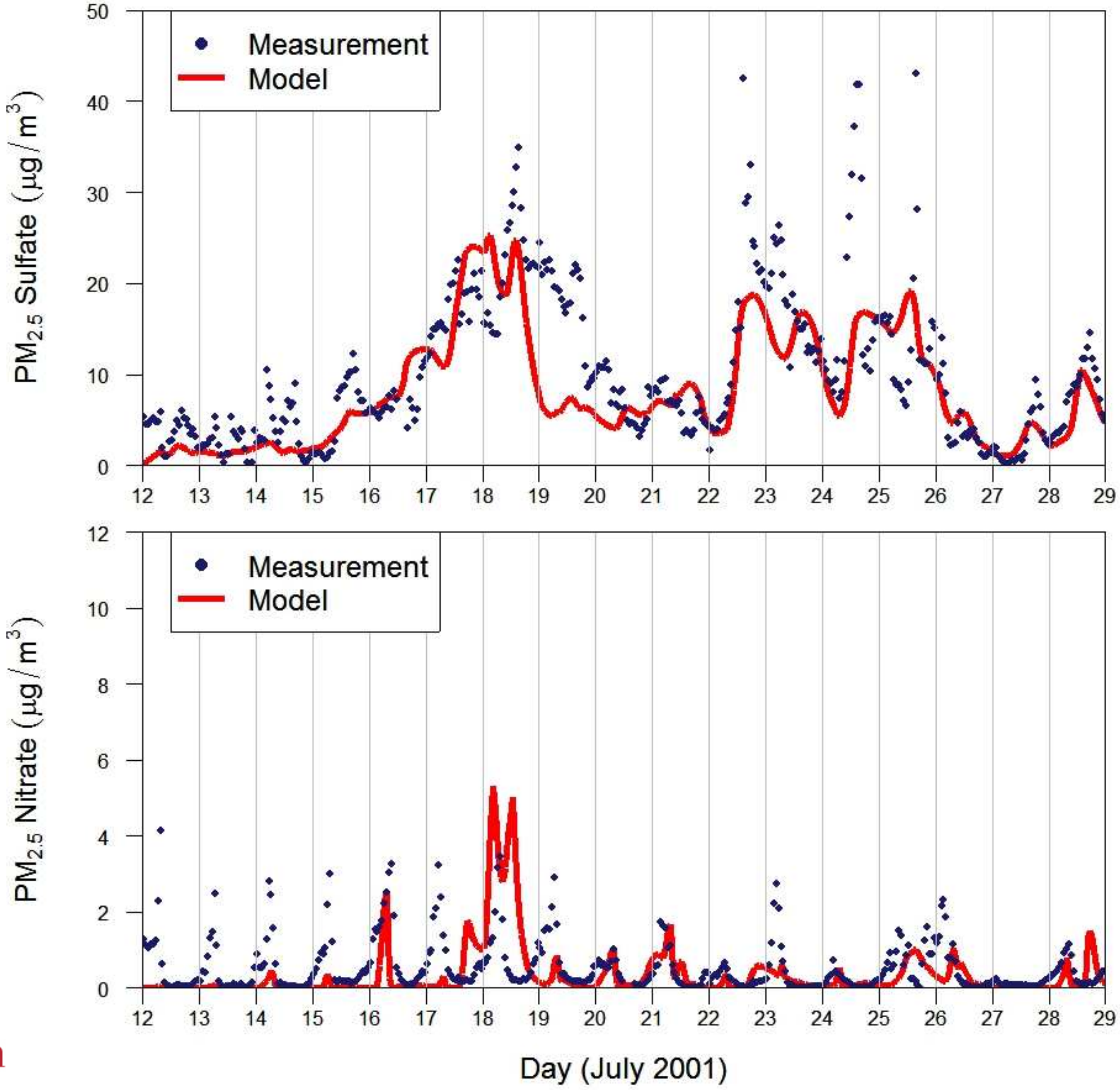
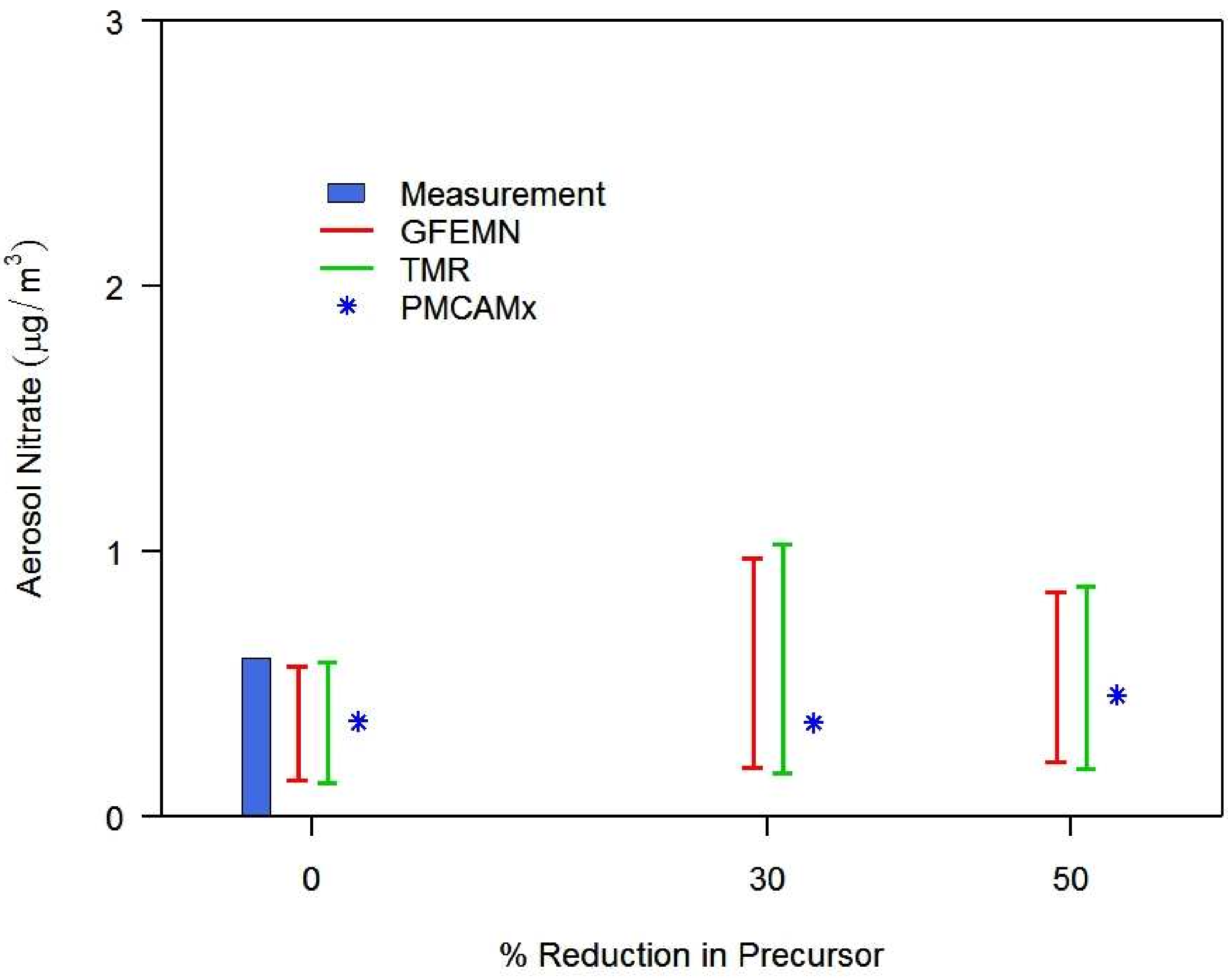
PMCAMx Evaluation
(Pittsburgh, July 2001)



Predicted Nitrate Concentrations
(Pittsburgh, July 2001 Conditions)



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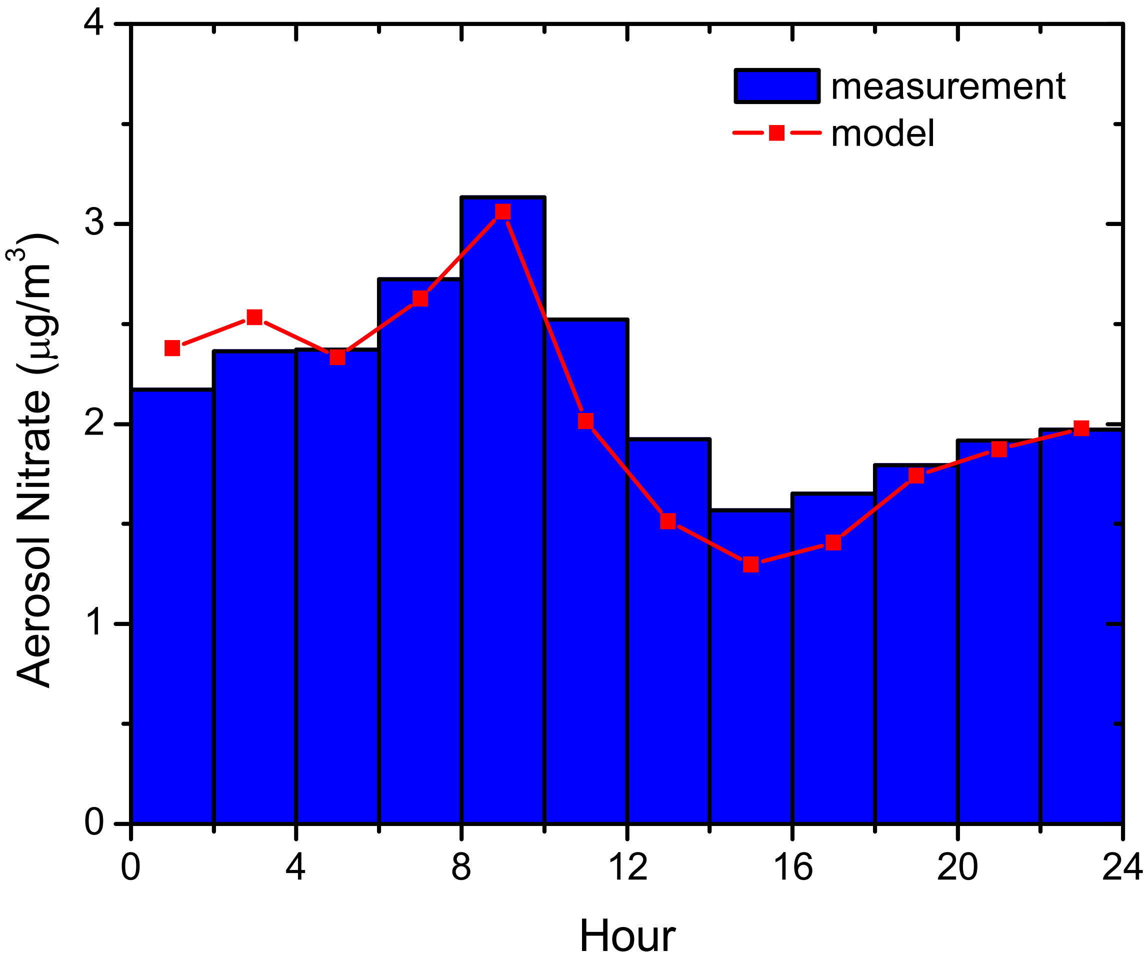
GFEMN Evaluation for $PM_{2.5}$ Nitrate
(Pittsburgh, January 2002)



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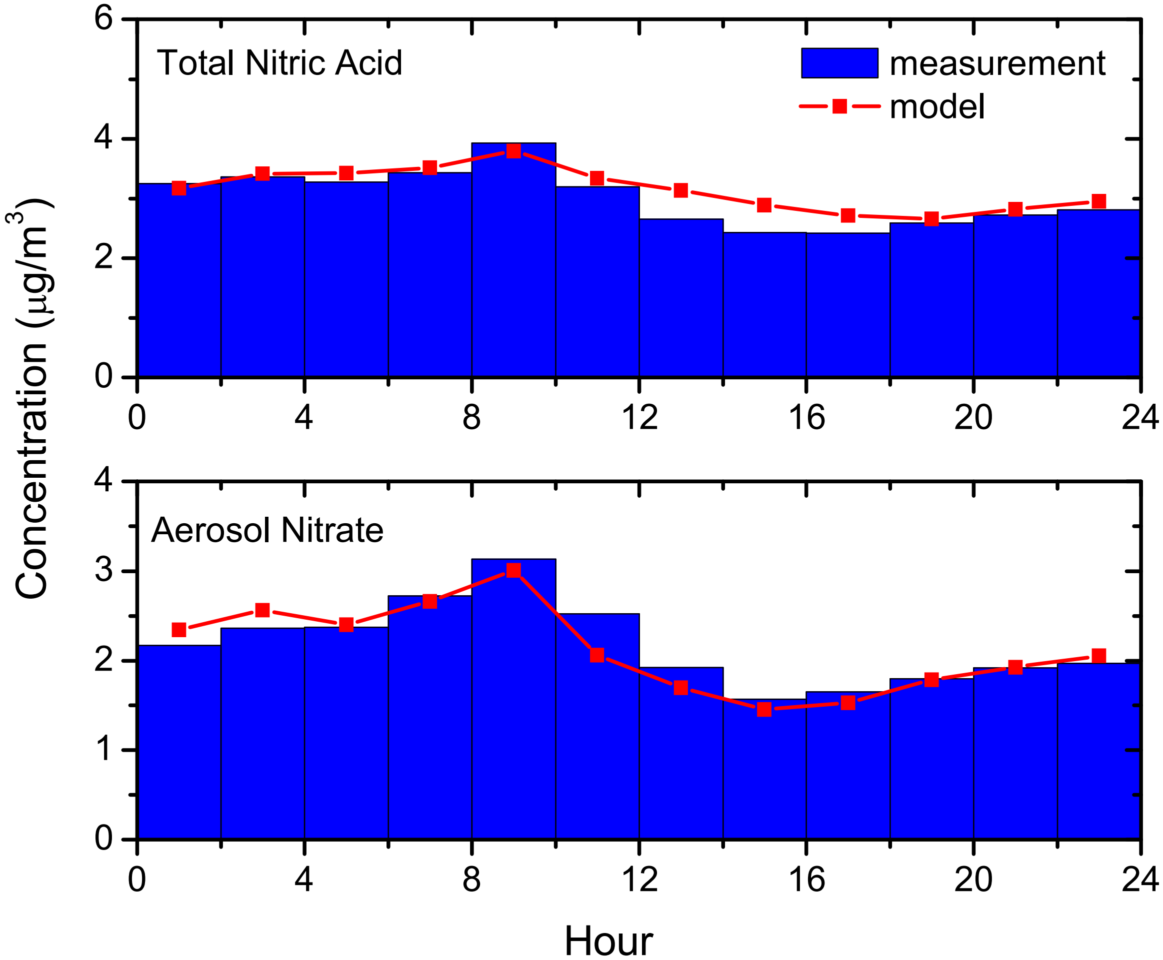
TMR Evaluation for $PM_{2.5}$ Nitrate
(Pittsburgh, January 2002)



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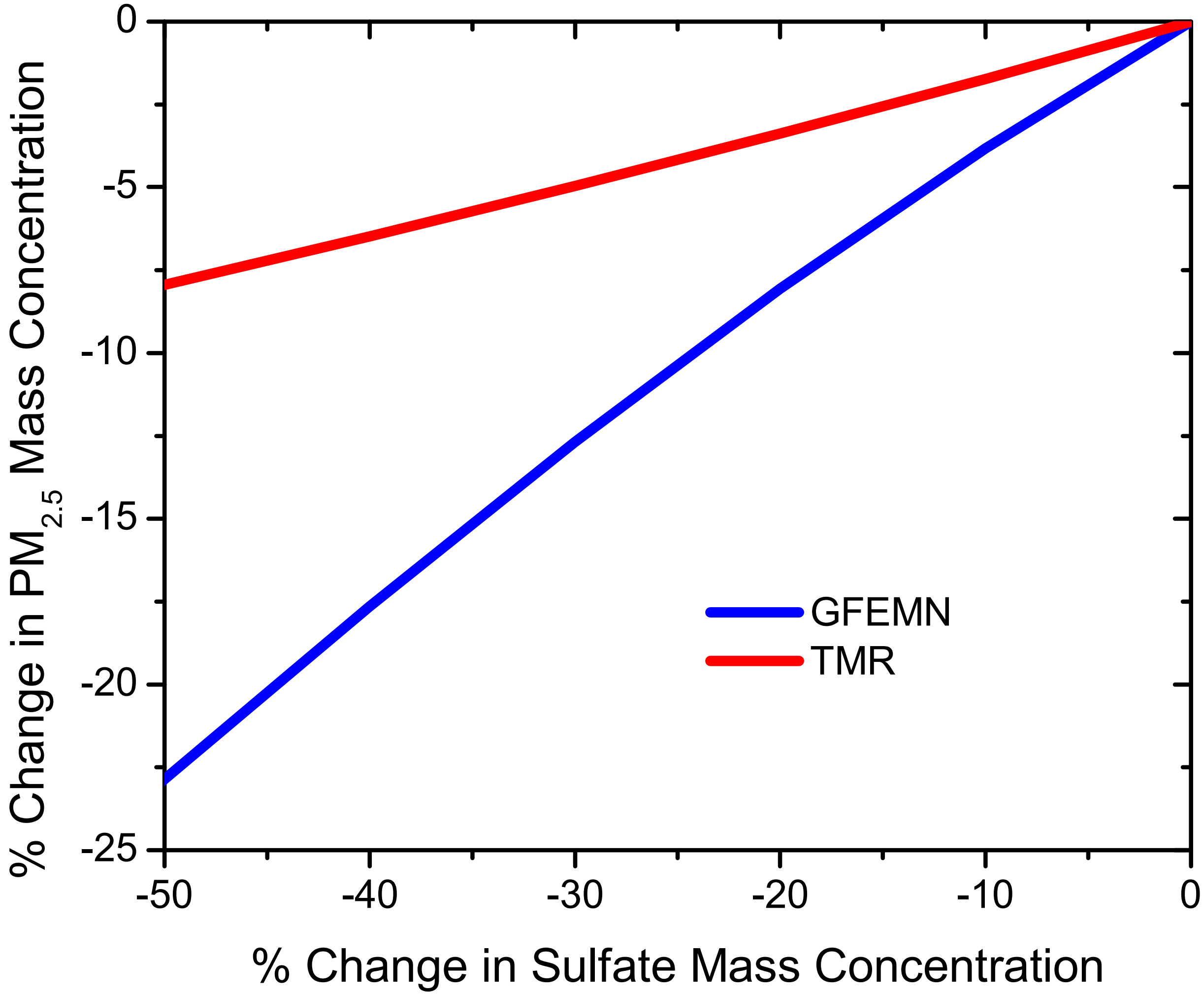
Predicted Inorganic $PM_{2.5}$ Response to Sulfate (Pittsburgh, Winter 2002)



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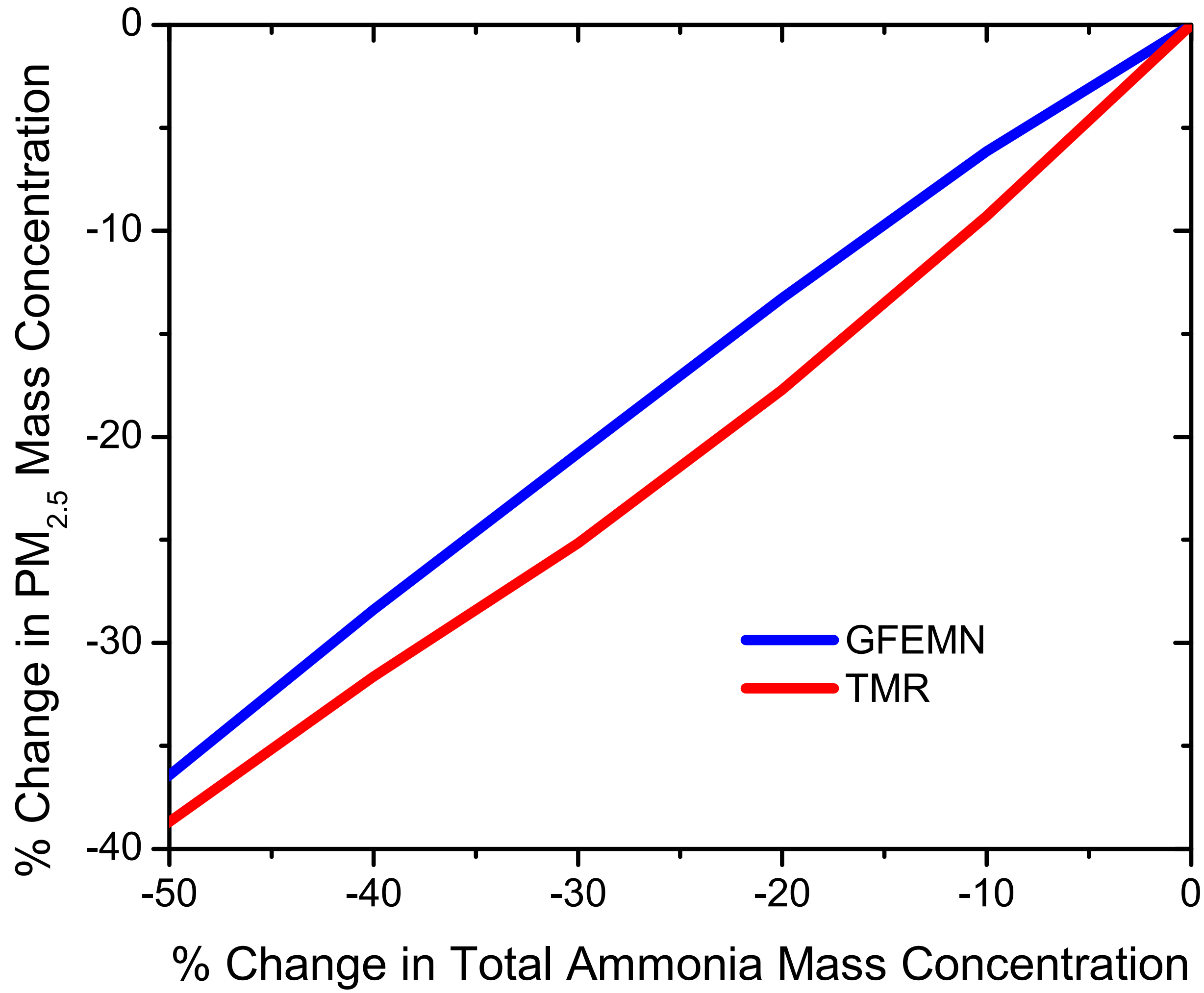
Predicted Inorganic $PM_{2.5}$ Response to Ammonia (Pittsburgh, Winter 2002)



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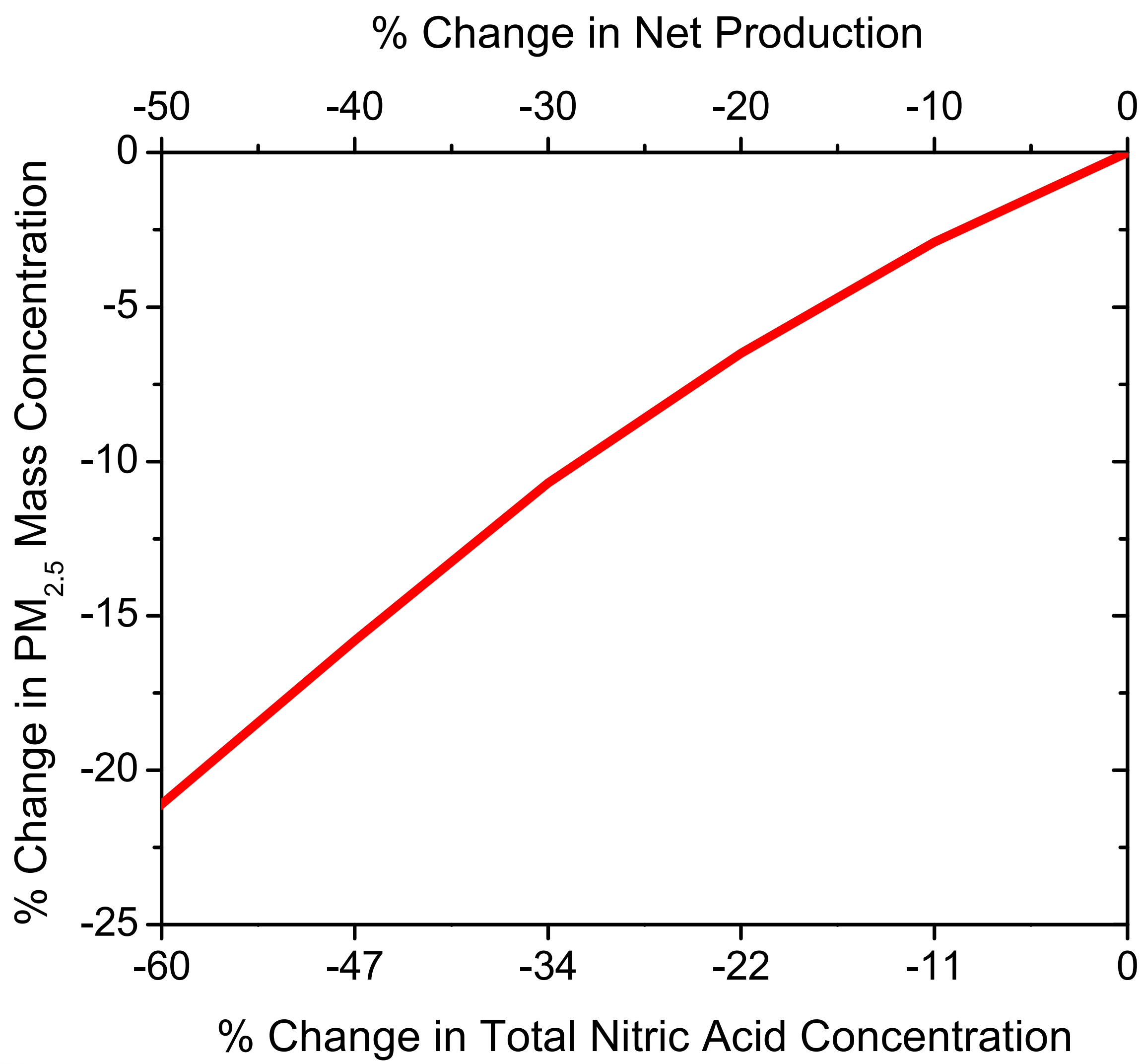
Predicted Inorganic $PM_{2.5}$ Response to Changes in Nitric Acid (Pittsburgh, Winter 2002)



Conclusions

- Observation based models (thermodynamic, TMR)
  - Take advantage of semi-continuous measurements
  - Require measurements of total ammonia and nitric acid (aerosol concentrations are not enough)
- All tools are able to reproduce the measurements for nitrate and nitric acid concentrations in summer and winter within experimental error
  - Different types of assumptions/uncertainties for each type of model
- Qualitative agreement provides some confidence in the results. Responses can be viewed as a range of potential outcomes

Conclusions (continued)

- July 2001 (Pittsburgh):
  - When SO_2 is reduced by 30%, sulfate is reduced by 23% (PMCAMx)
  - For reductions in sulfate up to 50%, nitrate substitution effect is small (less than 0.5 μg/m^3).
- January 2002 (Pittsburgh):
  - For up to 30% reductions in sulfate, inorganic $PM_{2.5}$ may be reduced by only 8% (substitution of approximately half the reduced sulfate by nitrate)
  - For 30% reduction in total ammonia, reductions in inorganic $PM_{2.5}$ predicted to be 20-25%.
  - For 30% reduction in total nitric acid, reduction in inorganic $PM_{2.5}$ predicted to be 11%.

Acknowledgements

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