ANISORROPIA:
Development of the Adjoint of ISORROPIA

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CMAQ-Adjoint Development Team

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Carleton University

Daven Henze and coworkers at
University of Colorado

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National Science Foundation
Graduate Research Fellowship

Conoco-Phillips
Model Development Support

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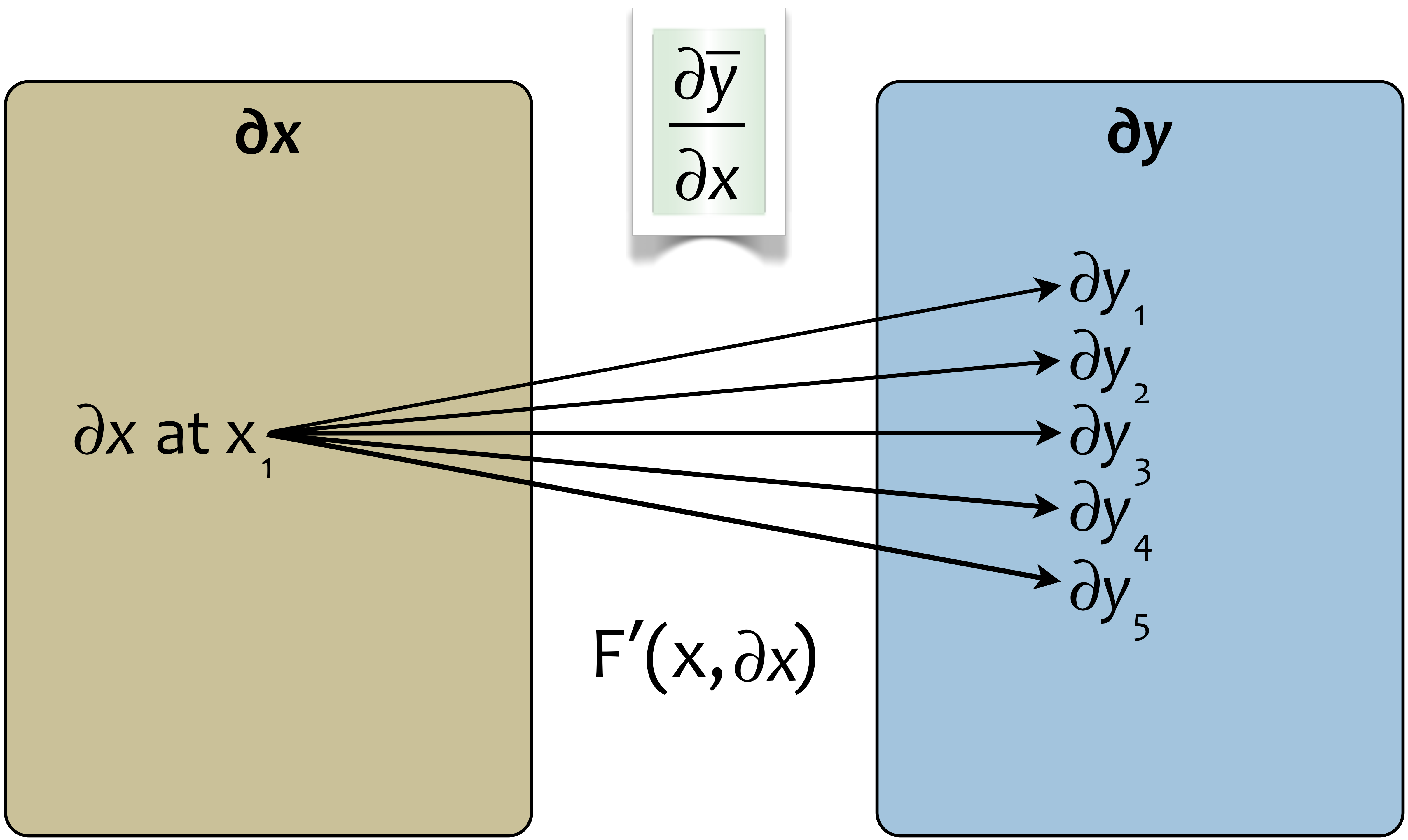
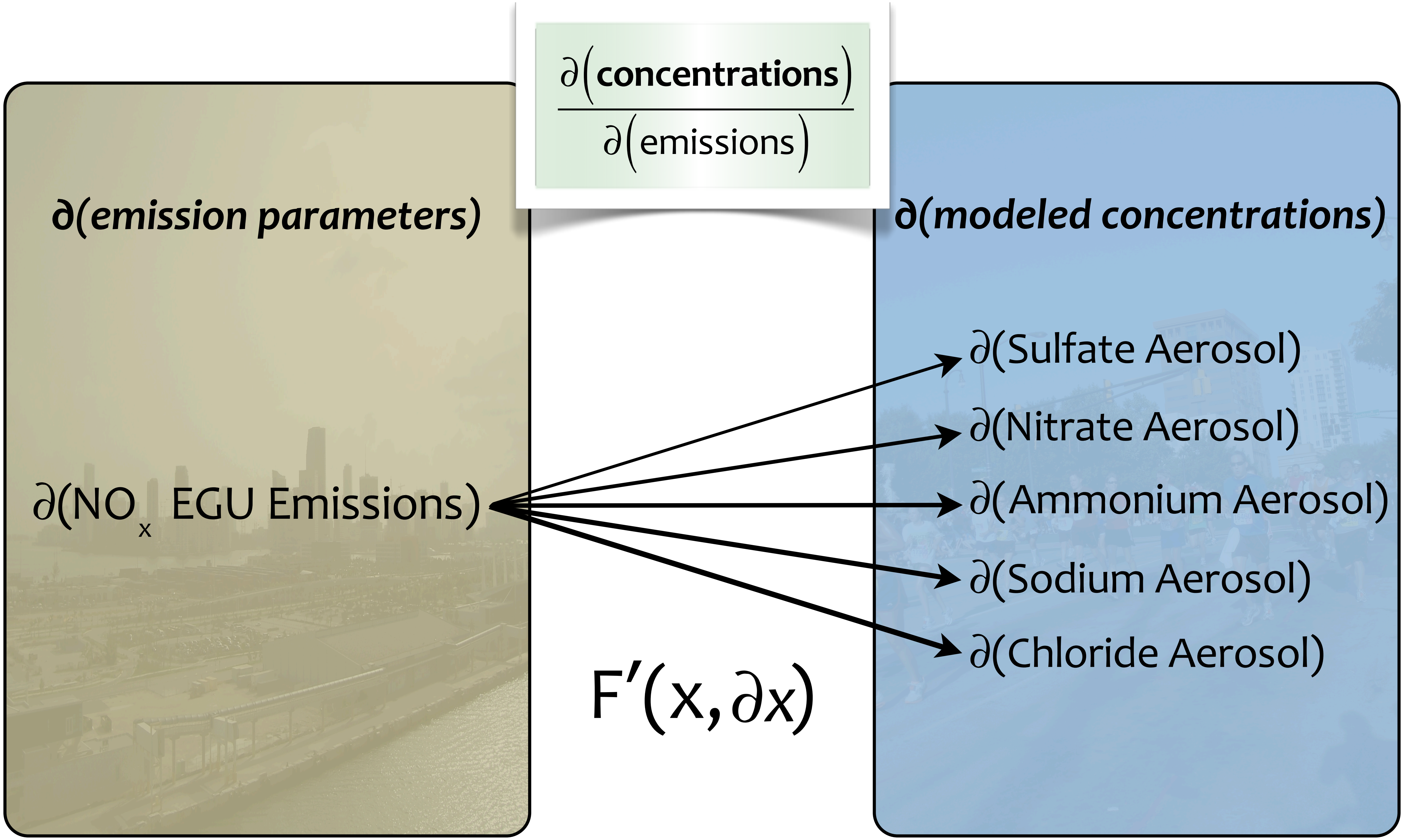
Direct Sensitivity Analysis:
Forward Method



Direct Sensitivity Analysis:
Forward Method



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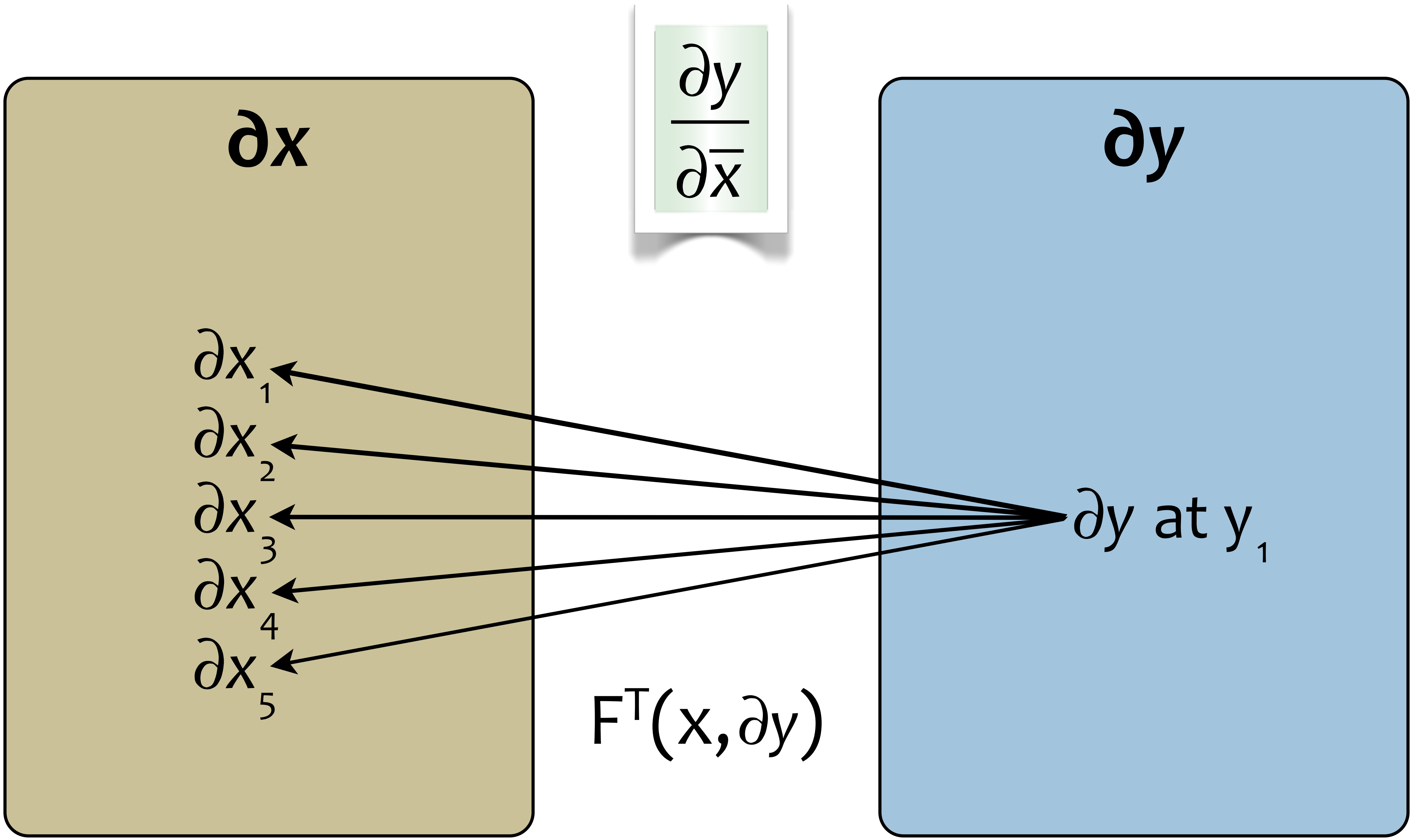
Direct Sensitivity Analysis:

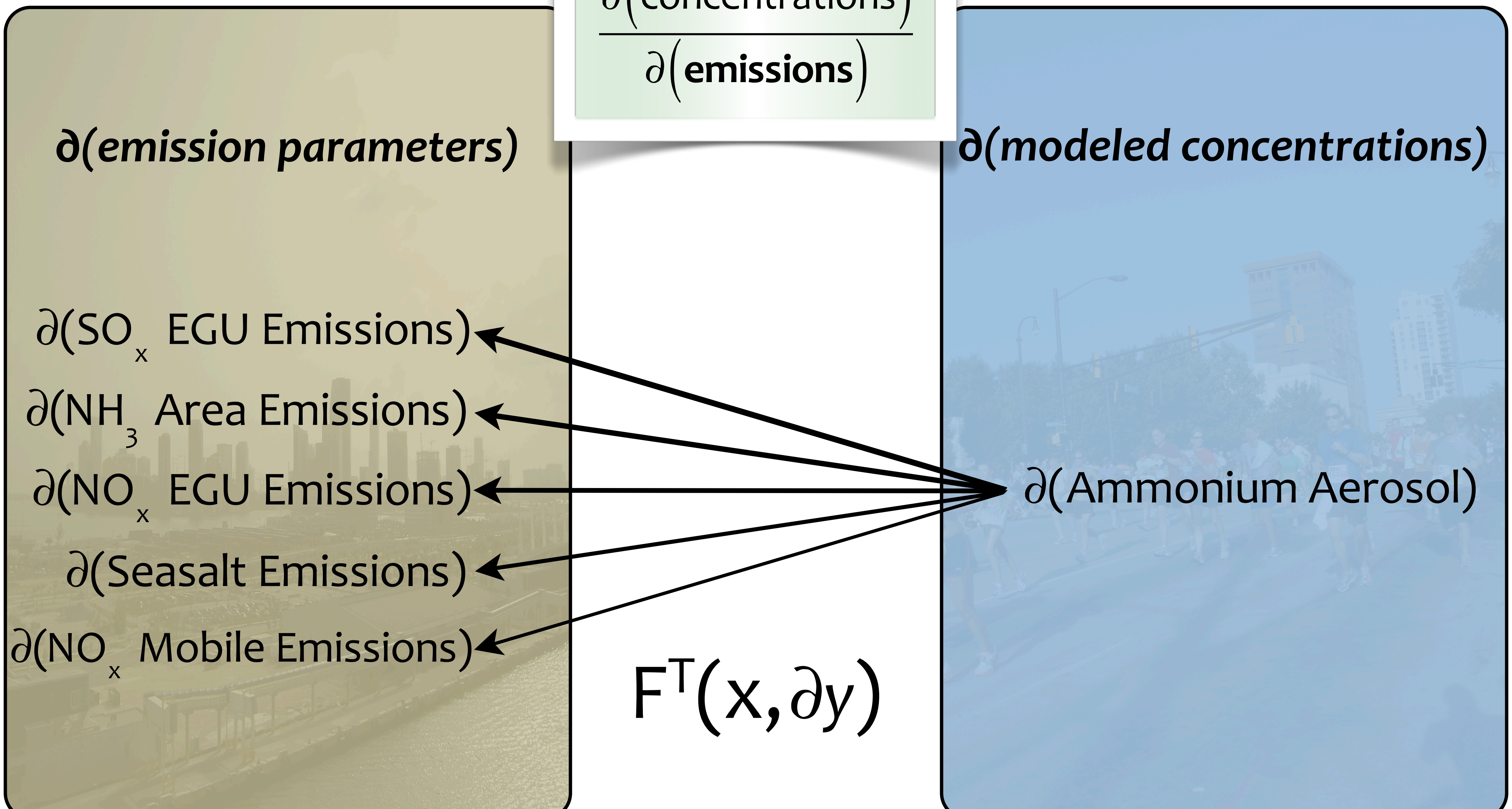
Reverse Method



Direct Sensitivity Analysis:

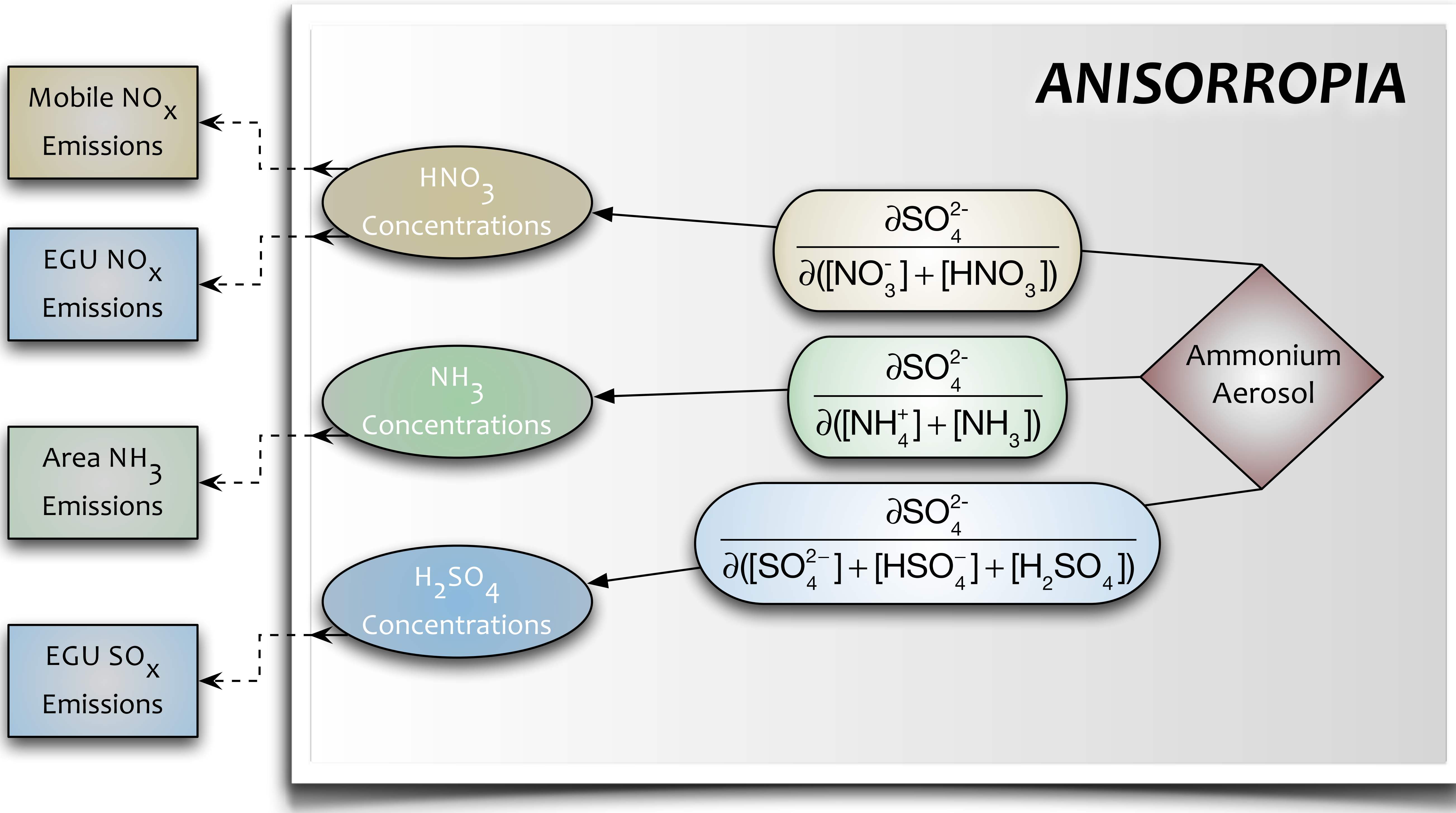
Reverse Method

$$\frac{\partial(\text{concentrations})}{\partial(\text{emissions})}$$



CMAQ-Adjoint Sensitivities

Inorganic Thermodynamic Equilibrium Contribution



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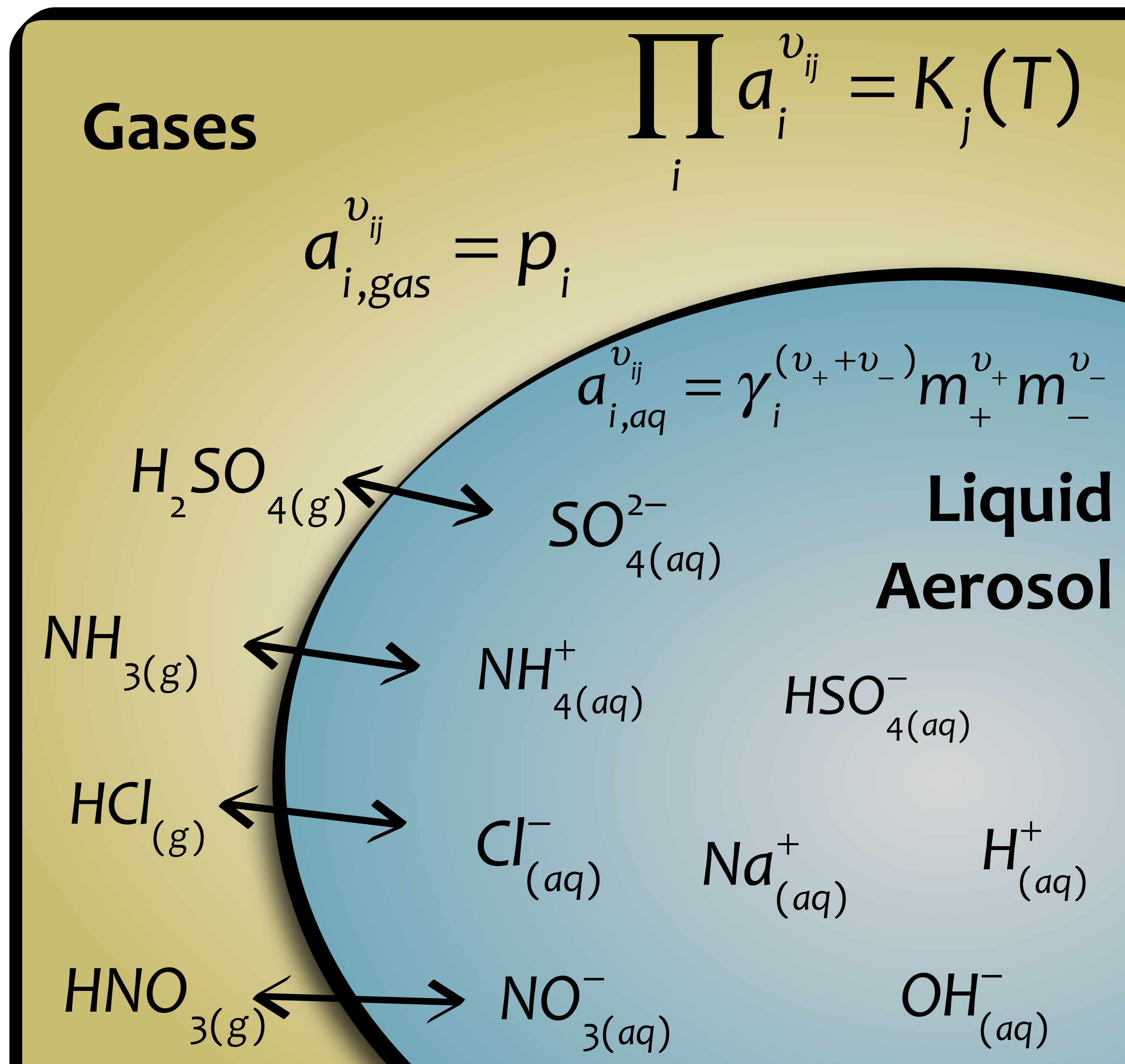
Strategy for ANISORROPIA Development

acidic aerosol

neutralized aerosol

ISORROPIA

(Nenes et al., 1998; Fountoukis et al., 2007)

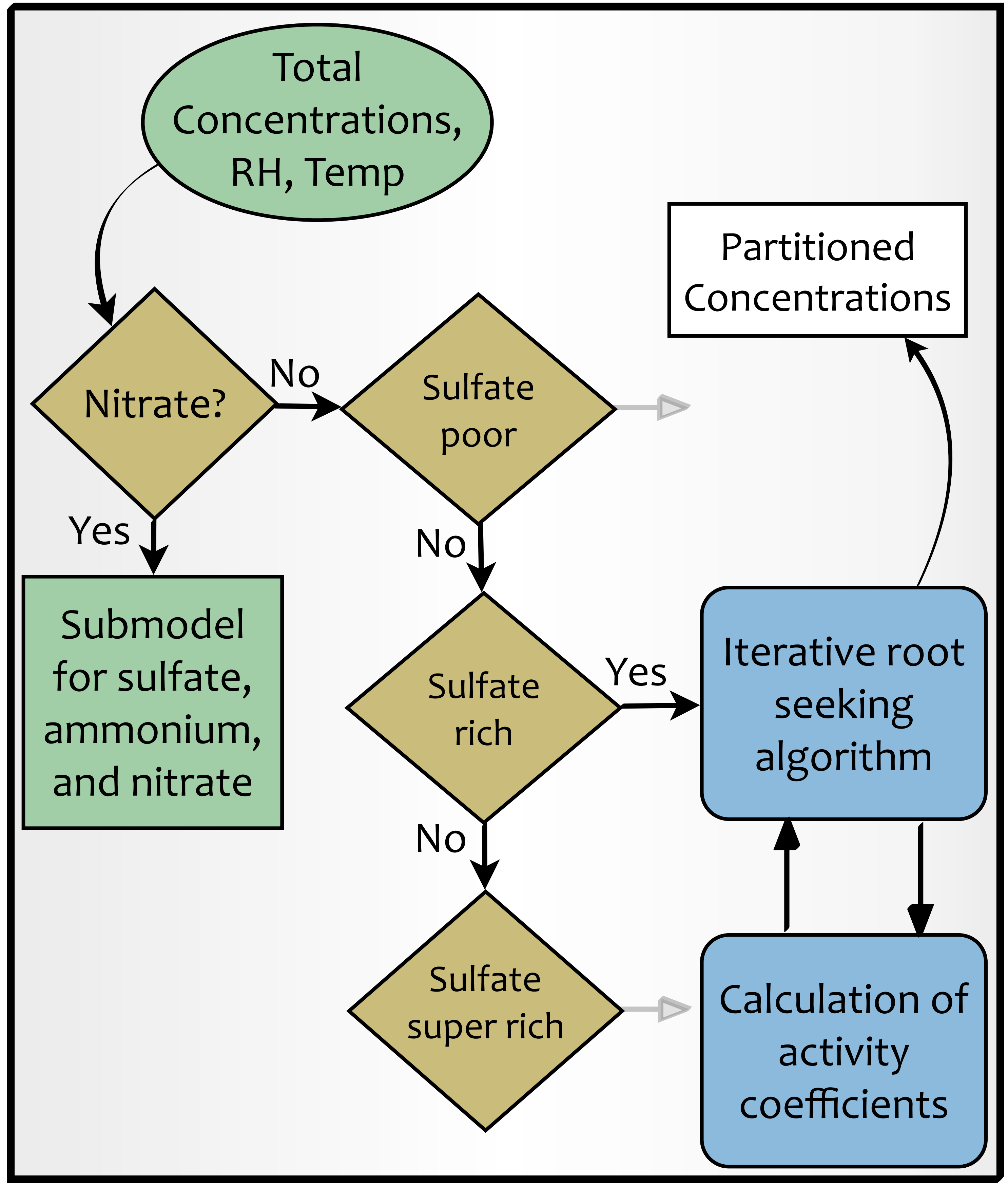


Strategy for ANISORROPIA Development

acidic aerosol

- ISORROPIA algorithm: *explicit solution, iteration on activity coefficients*
- Direct application of automatic differentiation tool, TAPENADE

Hascoët and Pascual, 2004



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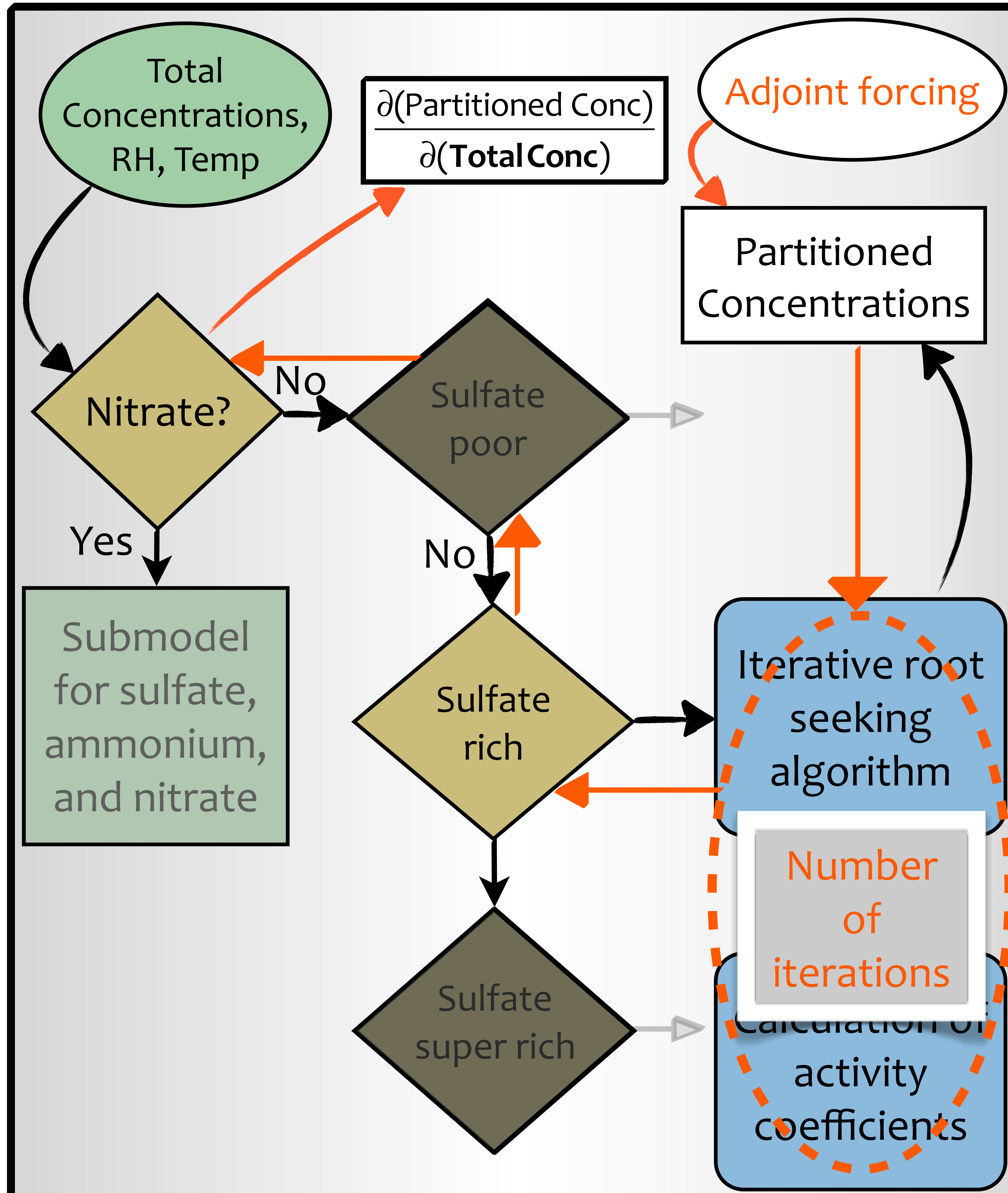
Strategy for ANISORROPIA Development

acidic aerosol

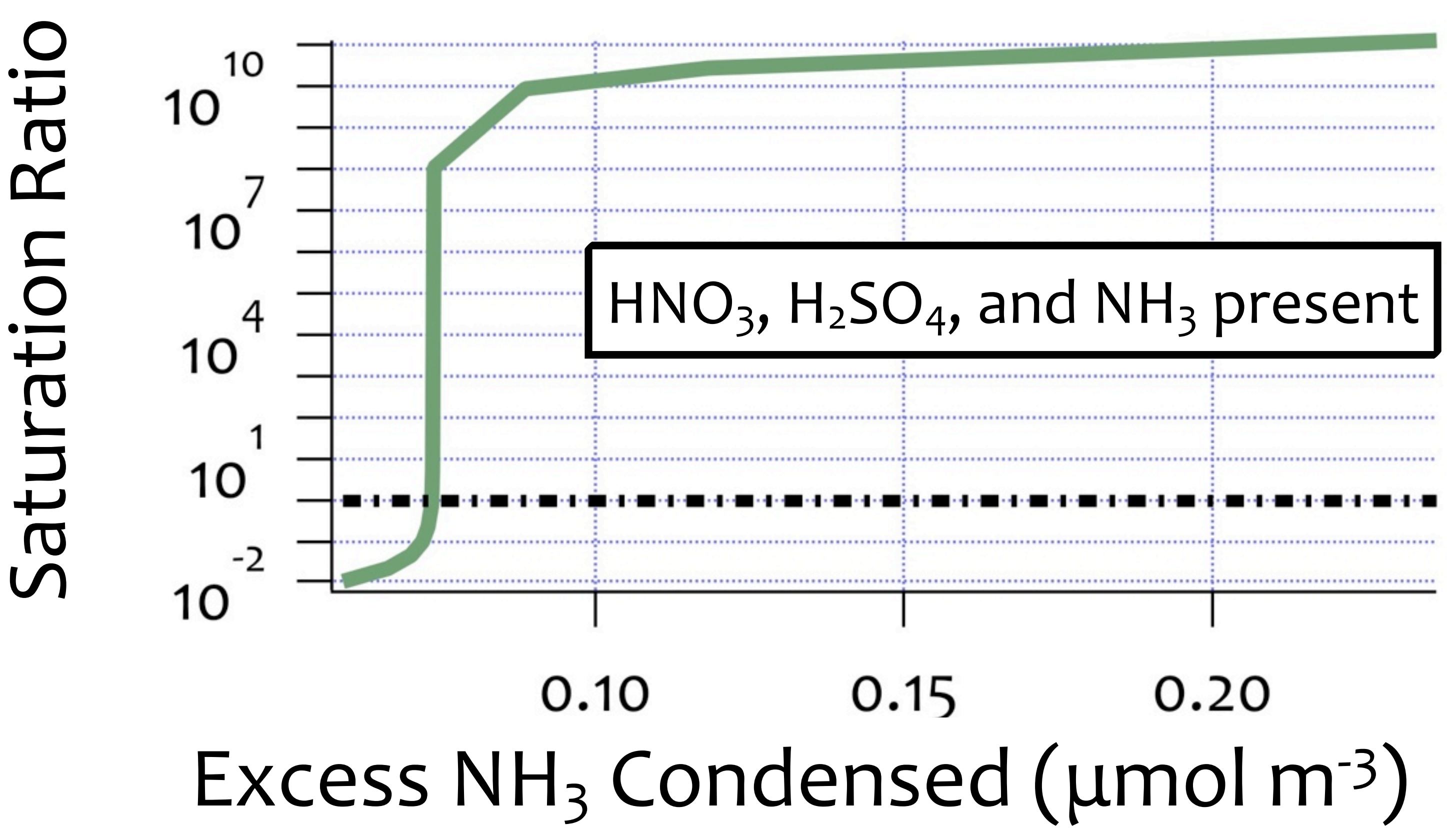
- ISORROPIA algorithm: *explicit solution, iteration on activity coefficients*
- Direct application of automatic differentiation tool, TAPENADE

$$\frac{\partial(\text{Partitioned Conc})}{\partial(\textbf{Total Conc})}$$

Hascoët and Pascual, 2004



Strategy for ANISORROPIA Development

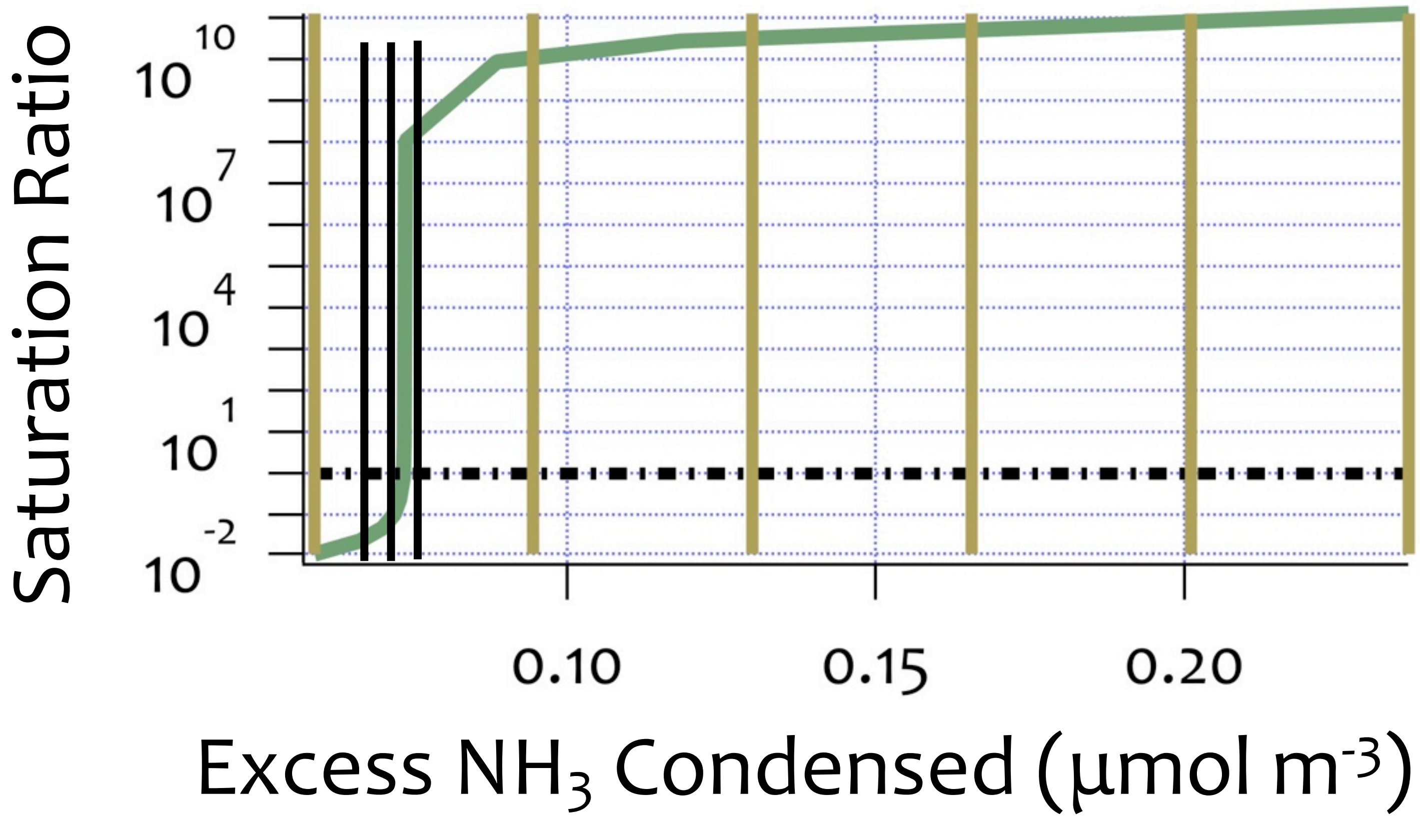


neutralized aerosol

- ISORROPIA algorithm:
  bisection method to minimize deviation of saturation ratio from 1

$$\text{Saturation Ratio} = \frac{\left[NH^{+}_{4,(aq)}\right]}{K\left[NH_{3,(g)}\right]\left[H^{+}_{(aq)}\right]\gamma}$$

Strategy for ANISORROPIA Development



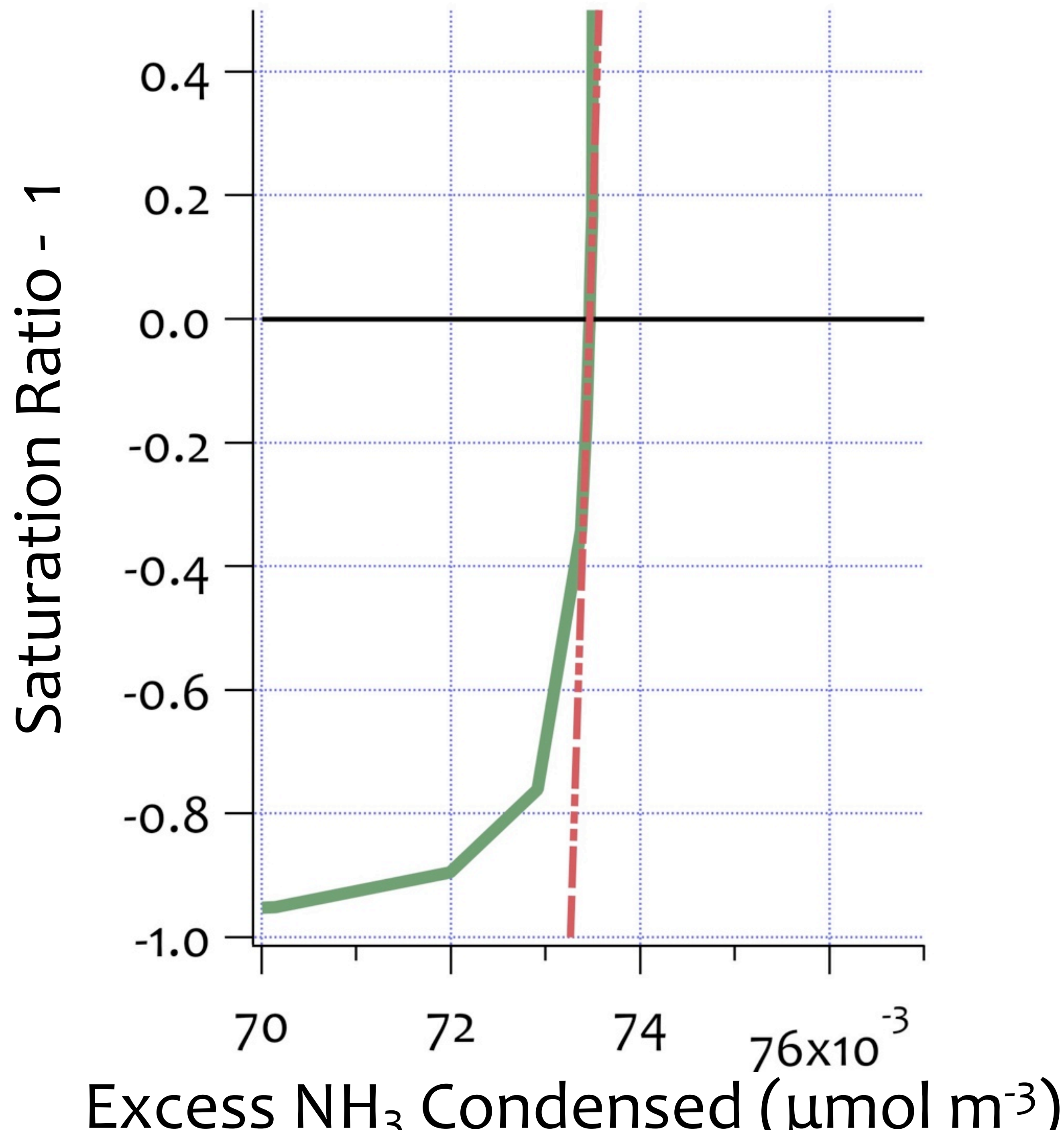
neutralized aerosol

- ISORROPIA algorithm:
  bisection method to minimize deviation of saturation ratio from 1

$$\text{Saturation Ratio} = \frac{\left[NH^{+}_{4,(aq)}\right]}{K\left[NH_{3,(g)}\right]\left[H^{+}_{(aq)}\right]\gamma}$$

- Derivative cannot be traced across bisection steps

Strategy for ANISORROPIA Development



neutralized aerosol

- Stable determination of root by bisection
- Apply single iteration of Newton-Raphson at root
  - $x_{n+1} = x_n - \dfrac{f(x_n)}{f'(x_n)}$
  - TAPENADE-produced forward sensitivity code
- Adjoint only of post-convergence N-R step

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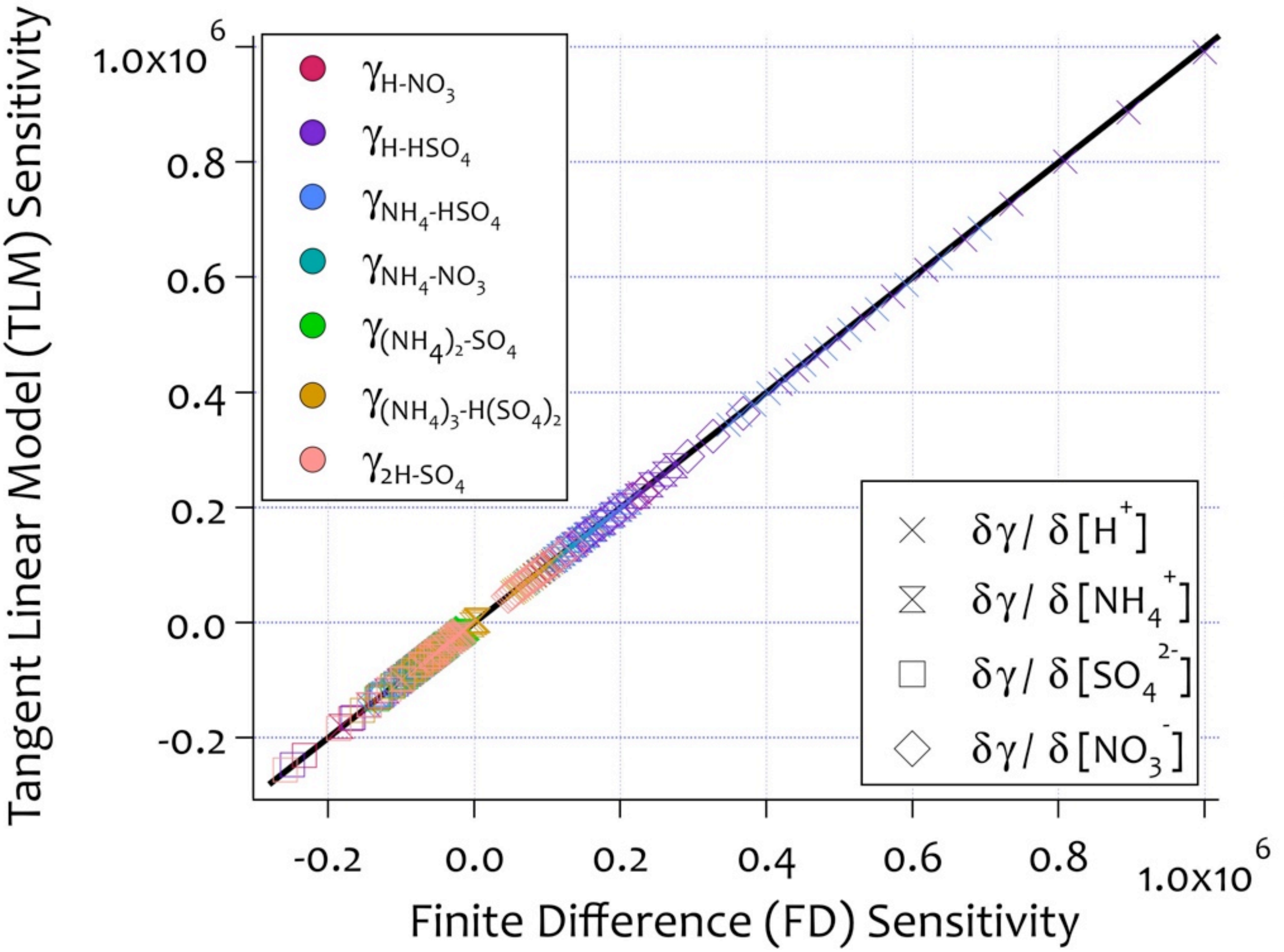
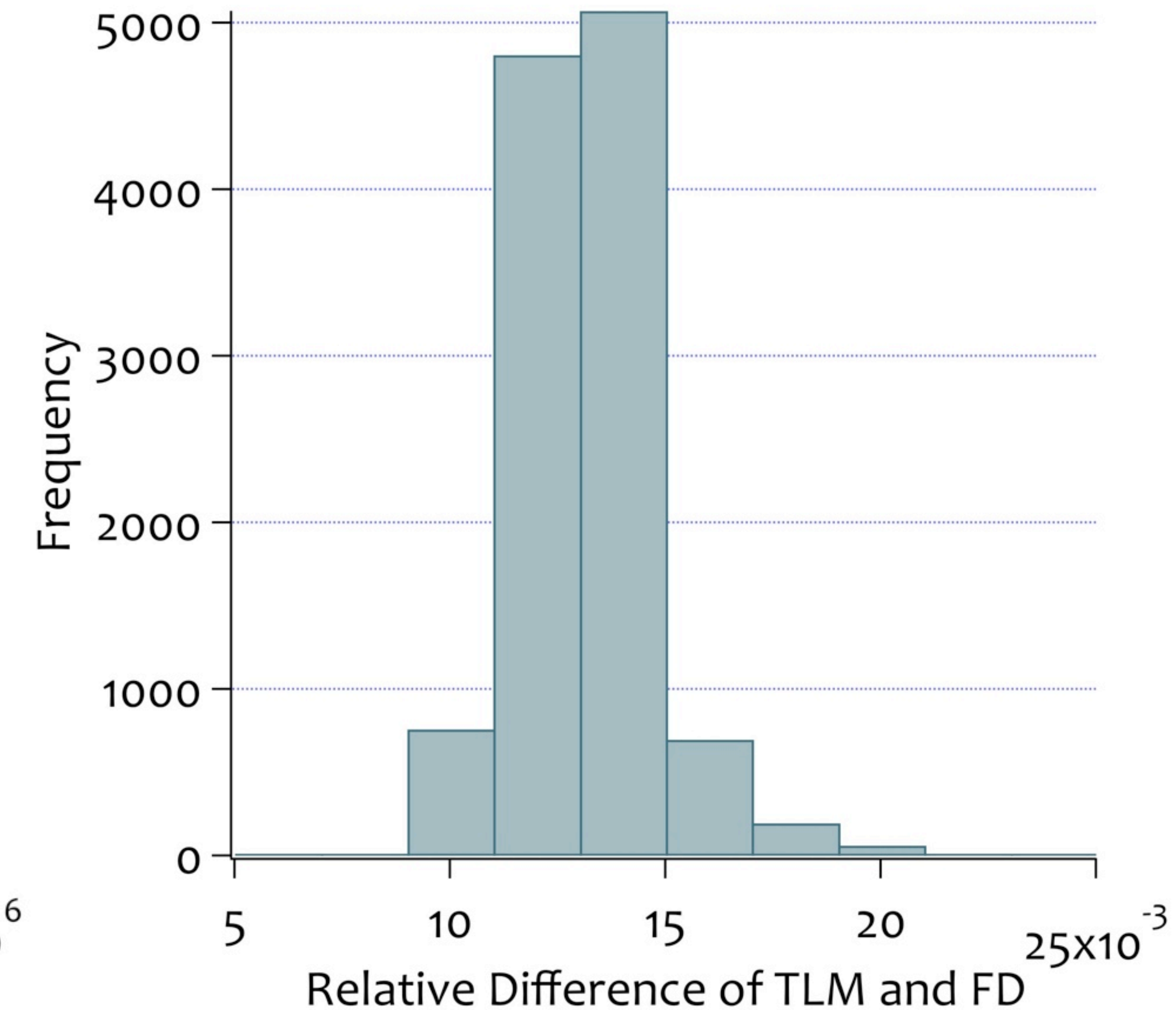
Bartholomew-Biggs, 1996

Activity Coefficient Sensitivities

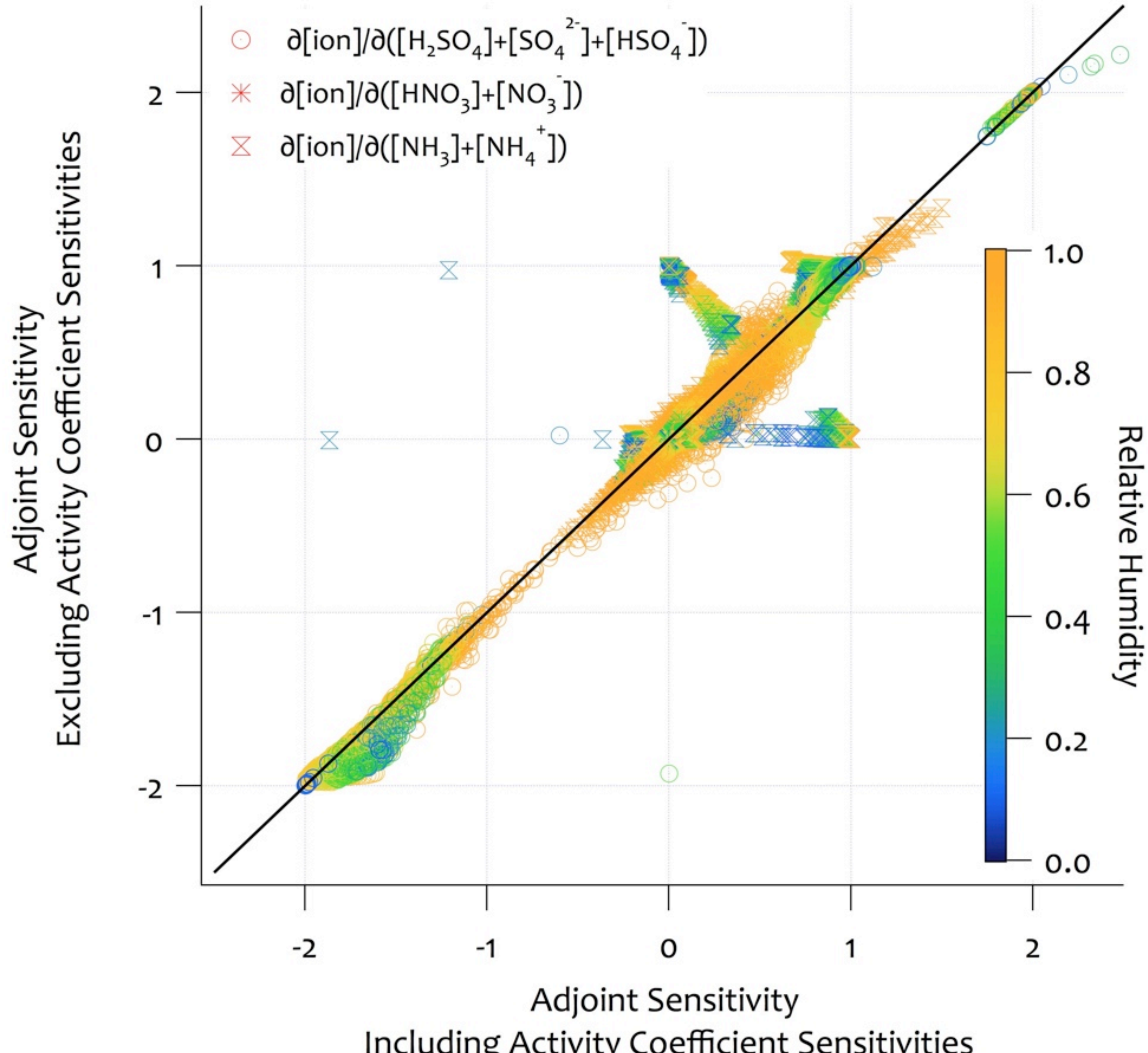


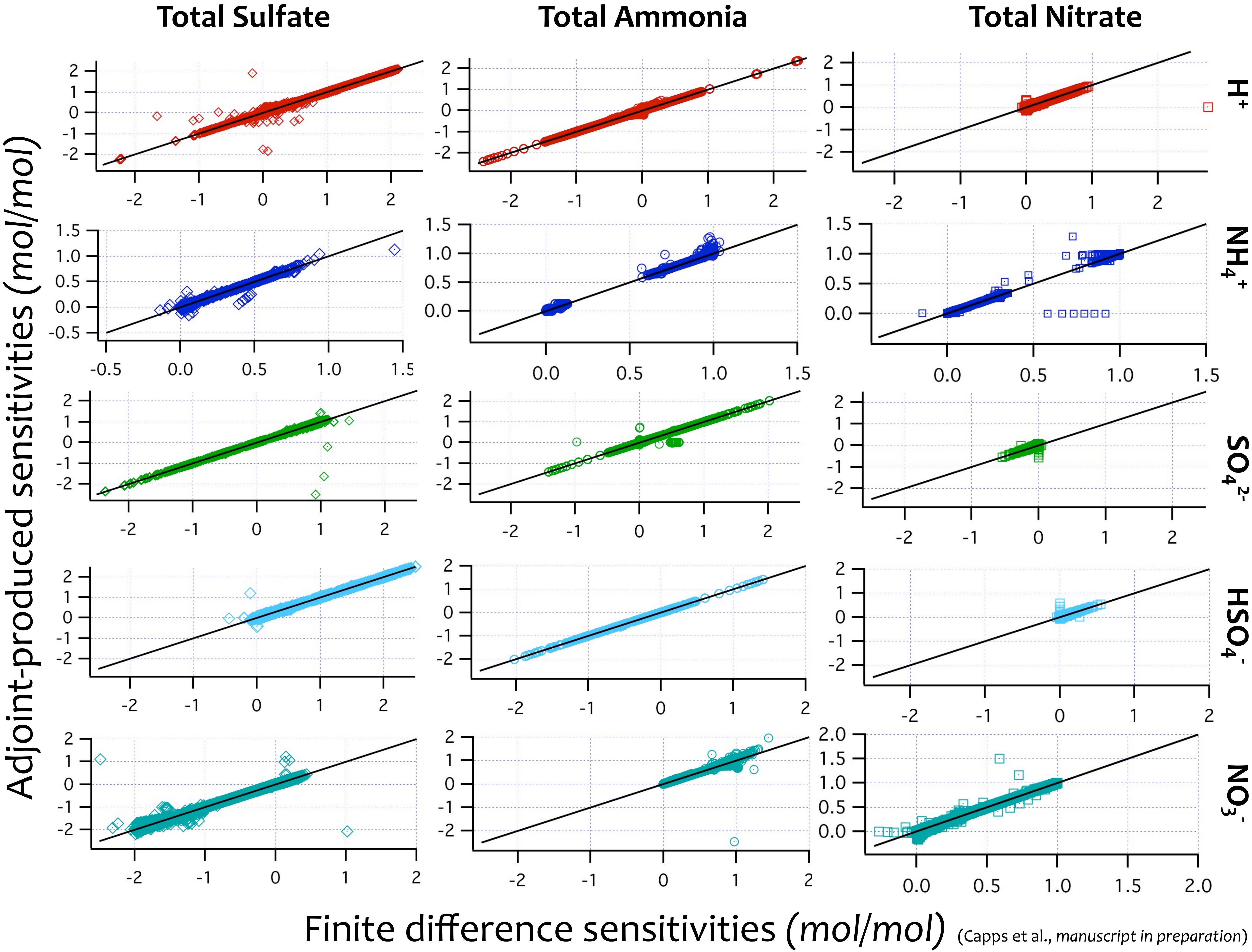


Differentiated code properly captures the sensitivity of activity coefficients involved in the calculation.

Activity Coefficient Sensitivities

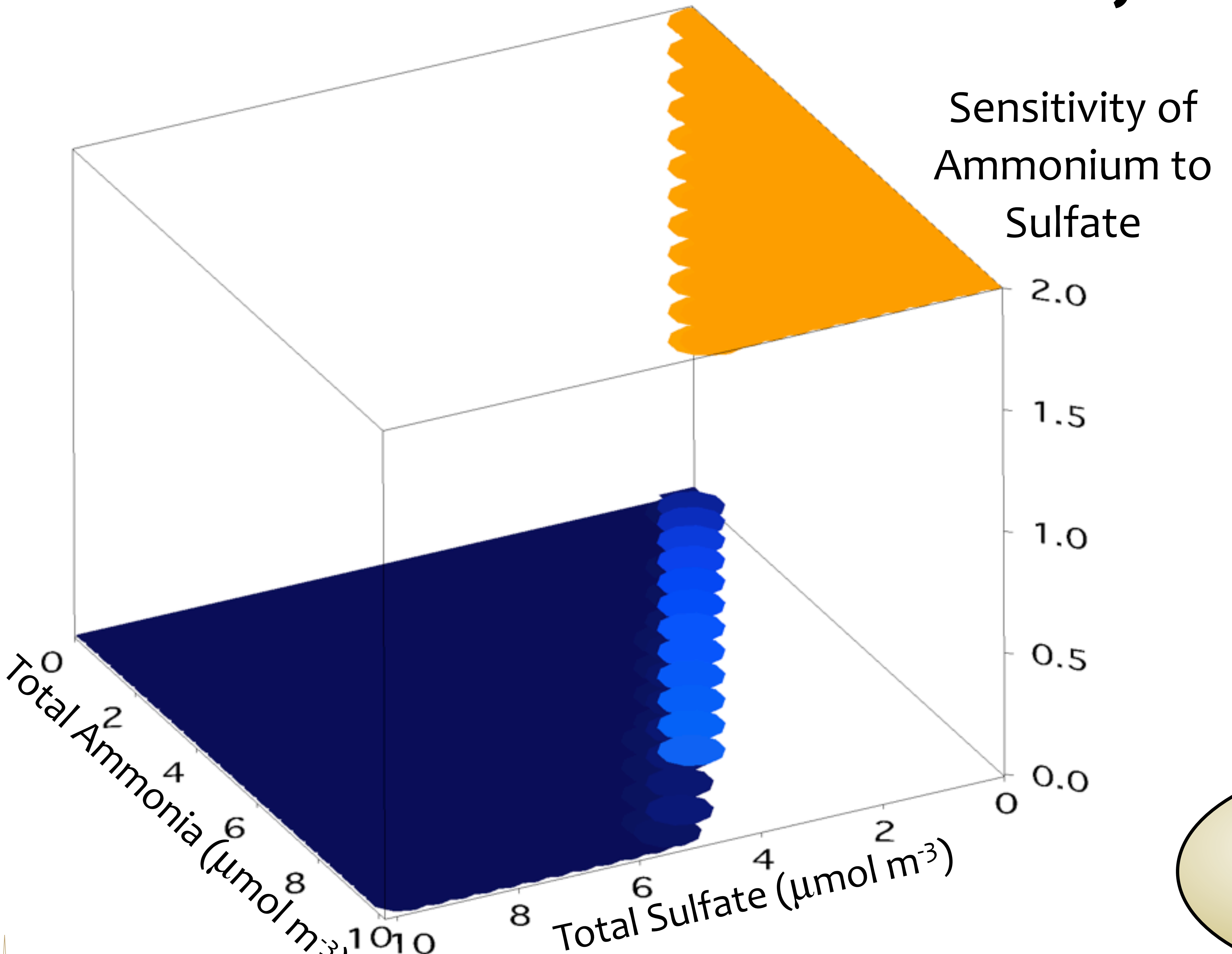
- For neutralized aerosol, accumulation of sensitivities through the activity coefficient calculations can be significant
  - Lower relative humidities
  - Sensitivities to total ammonia

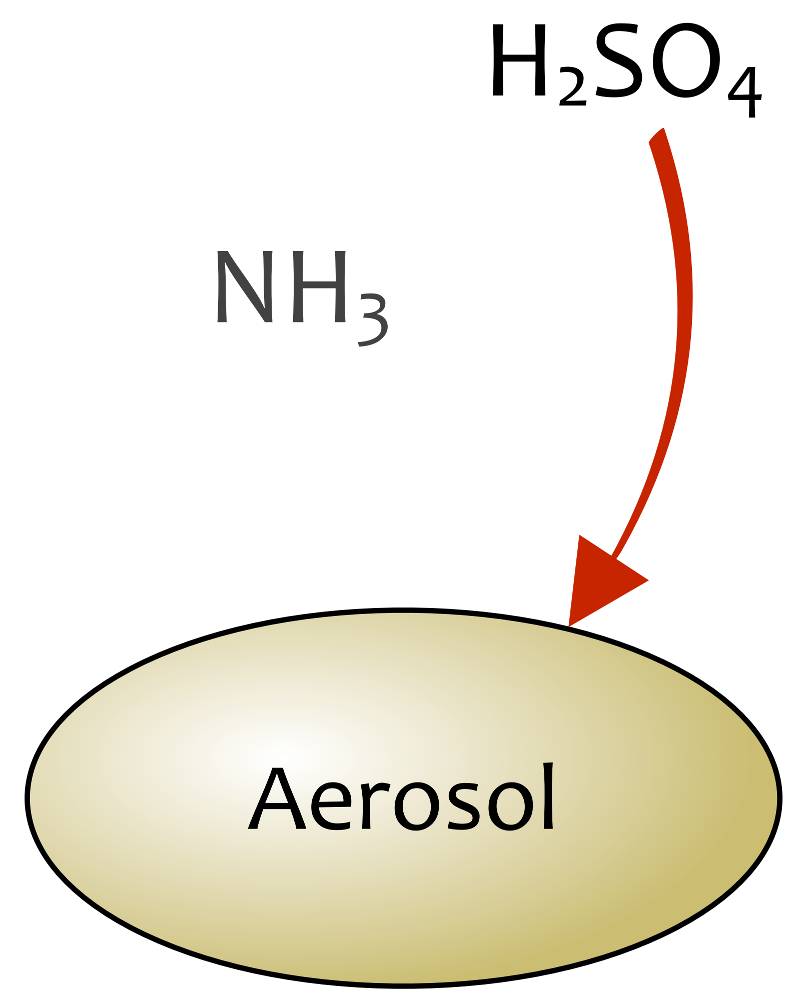




(Capps et al., *manuscript in preparation*)

Sensitivity Space: *Ammonium and Sulfate*





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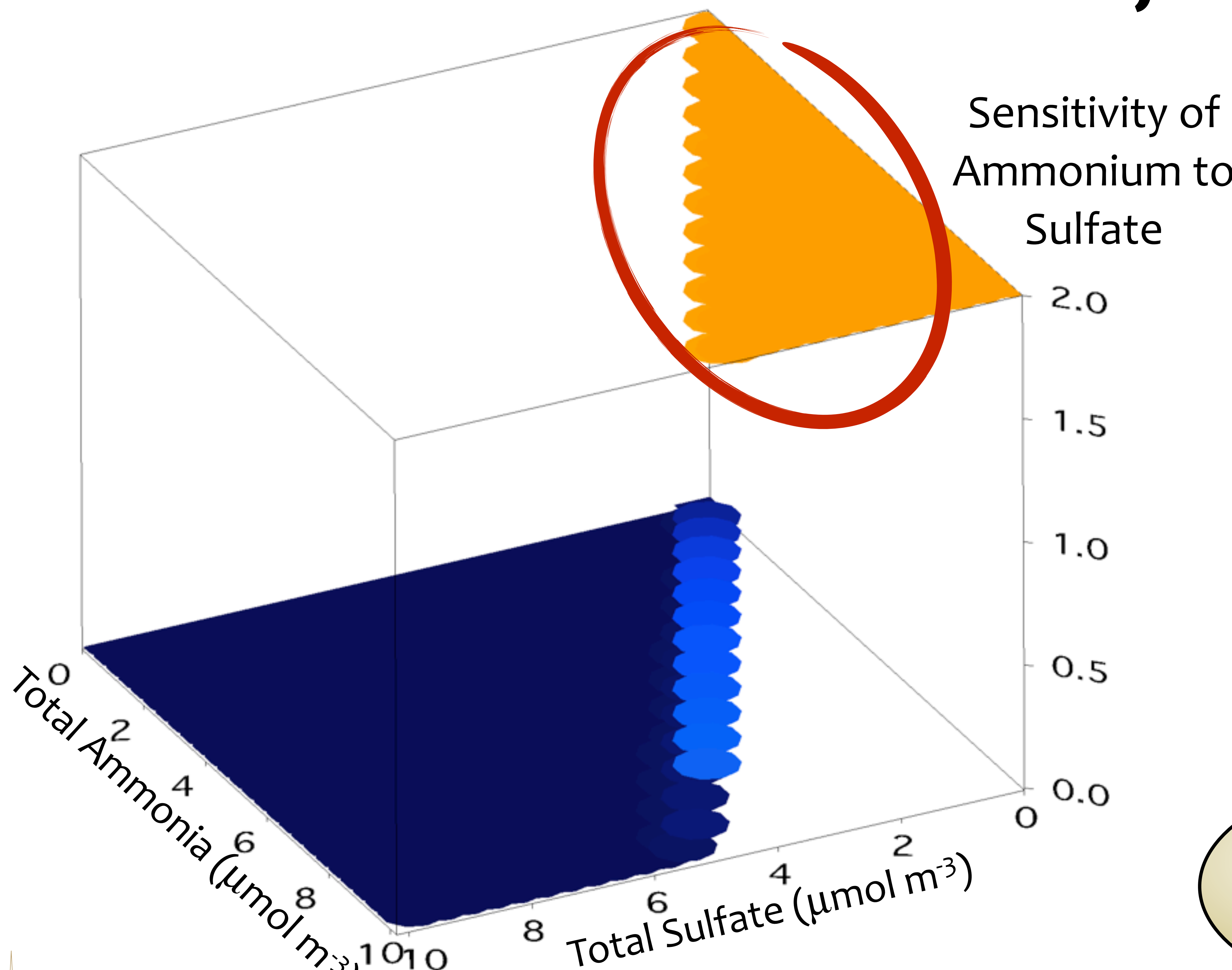

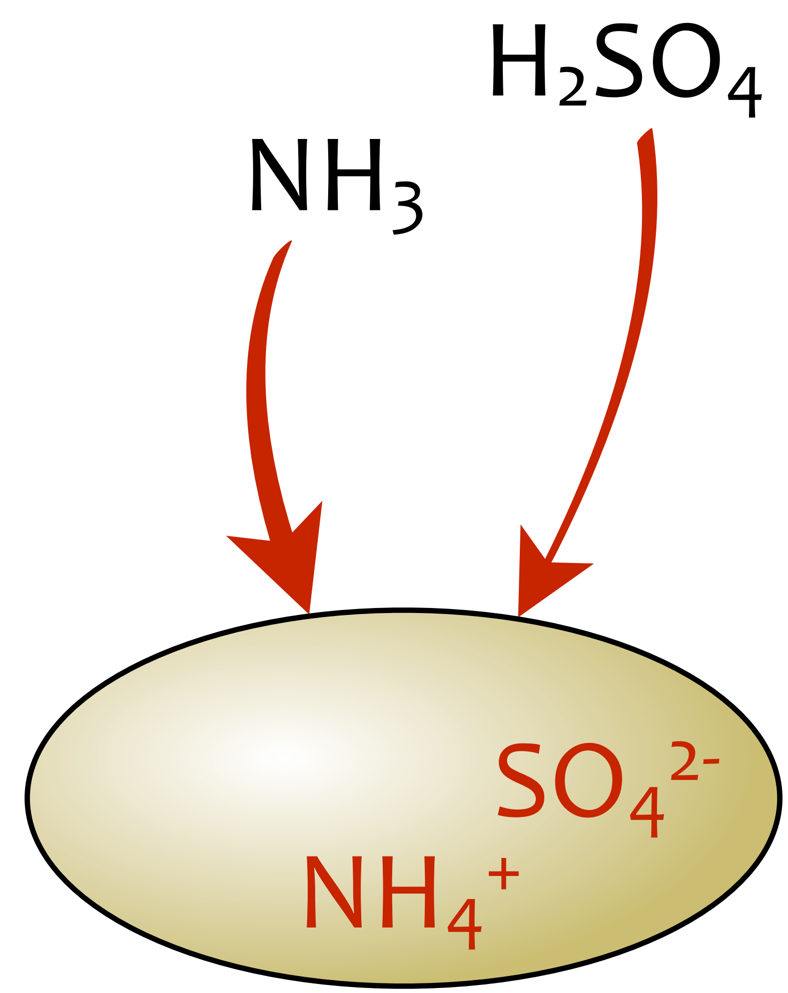

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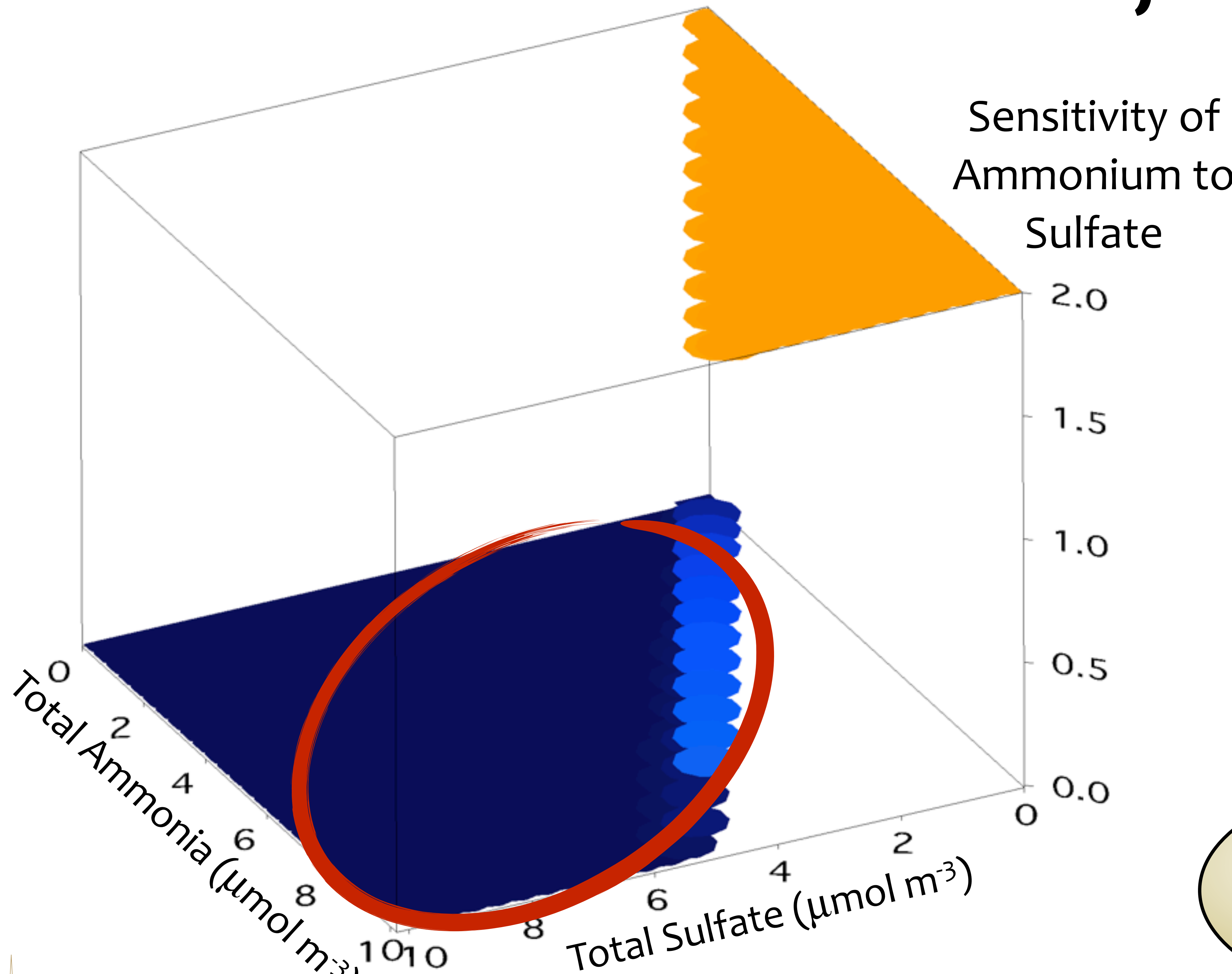

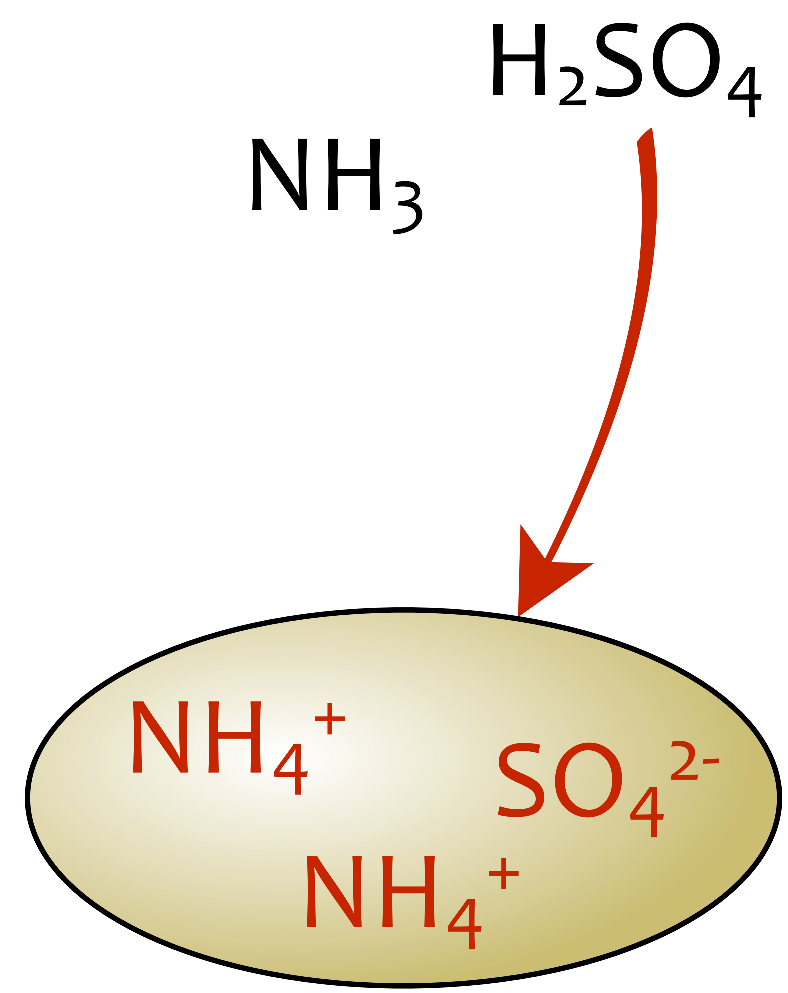

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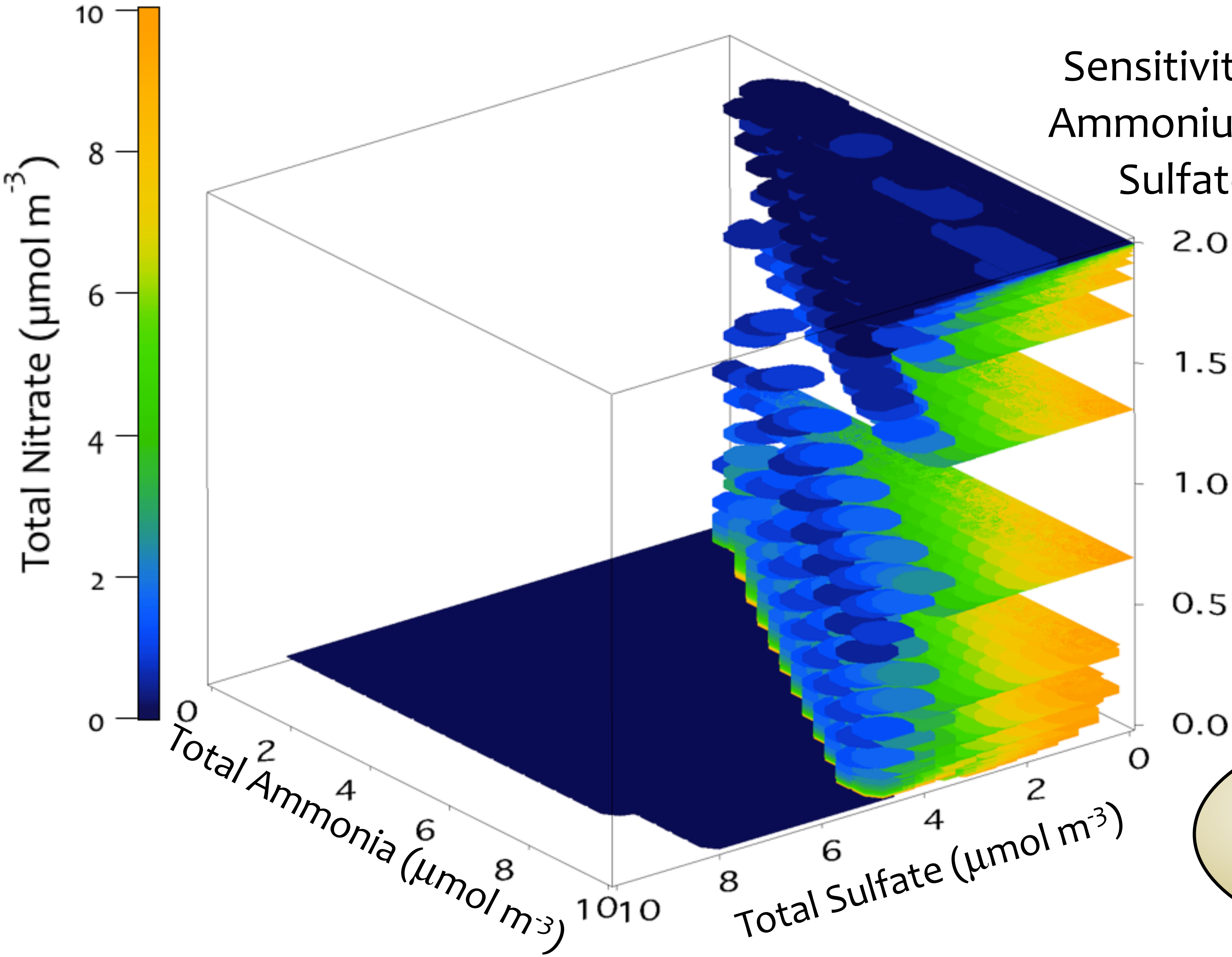
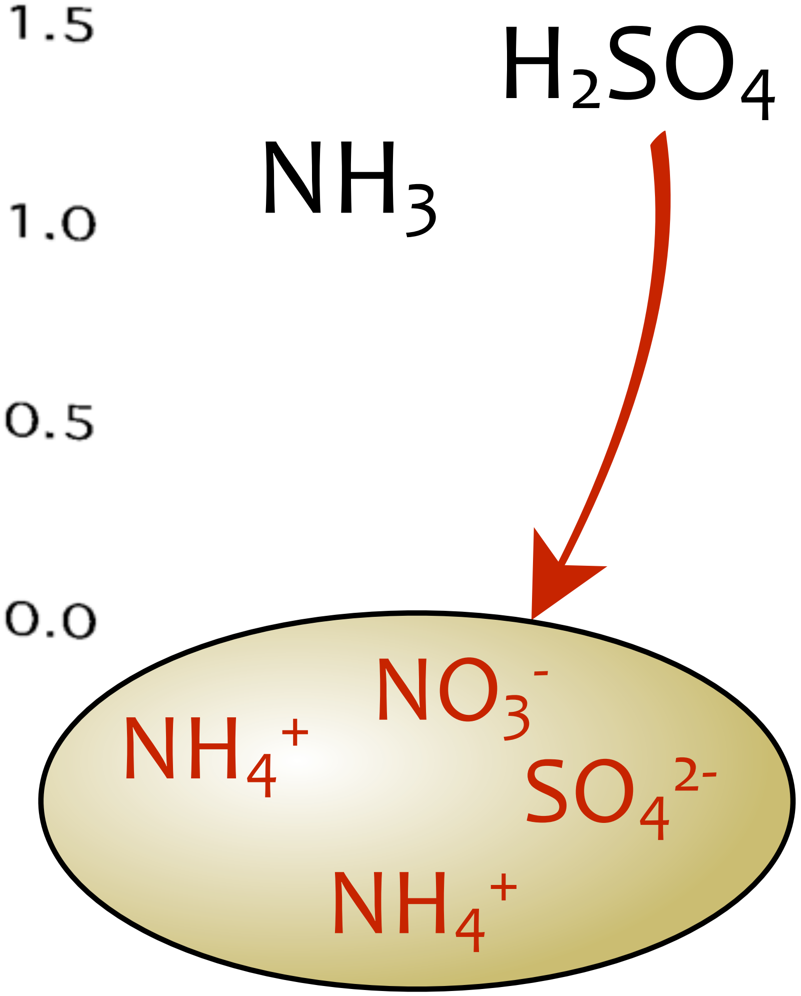
Sensitivity Space:

Ammonium, Sulfate, and Nitrate





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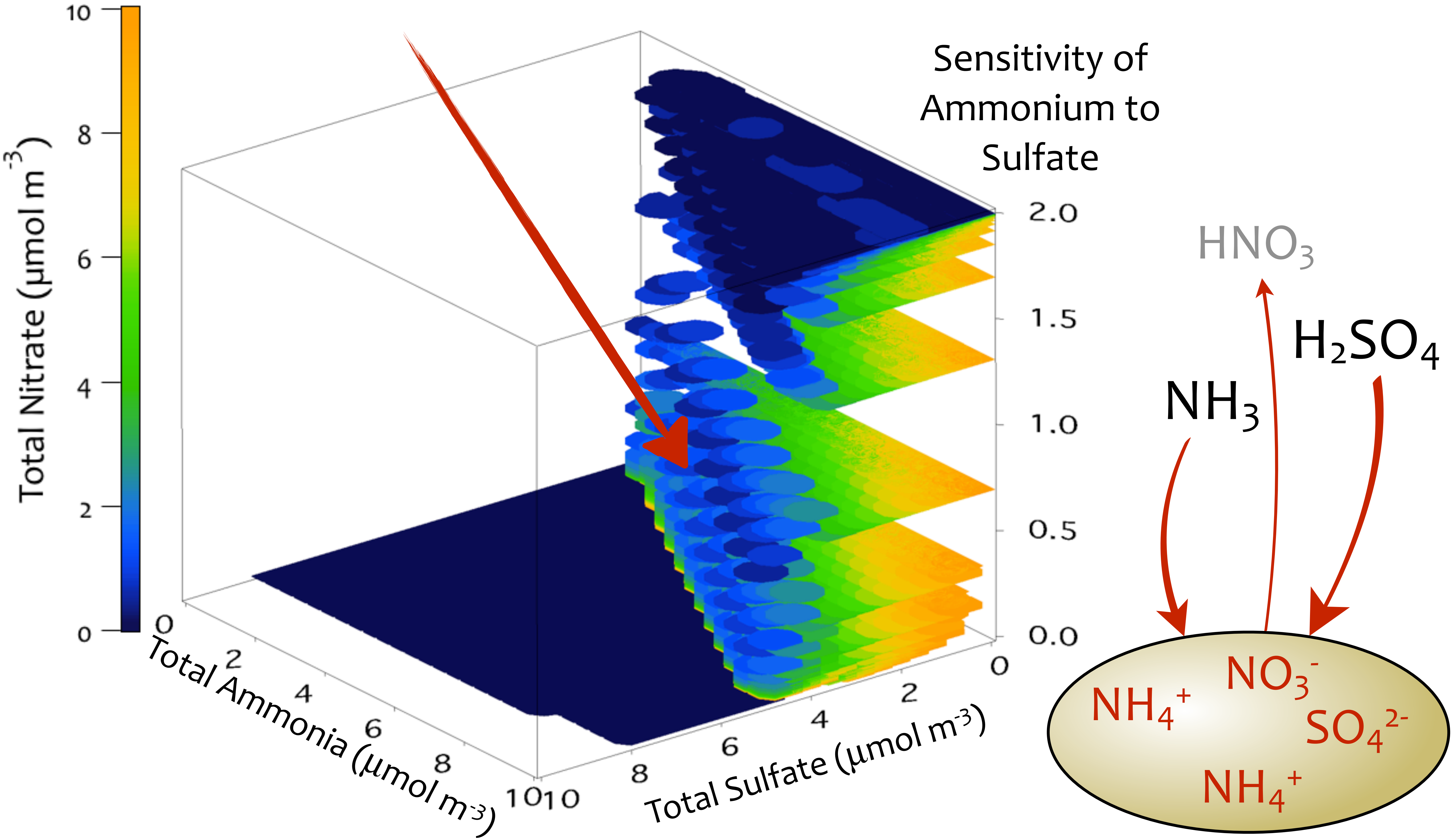

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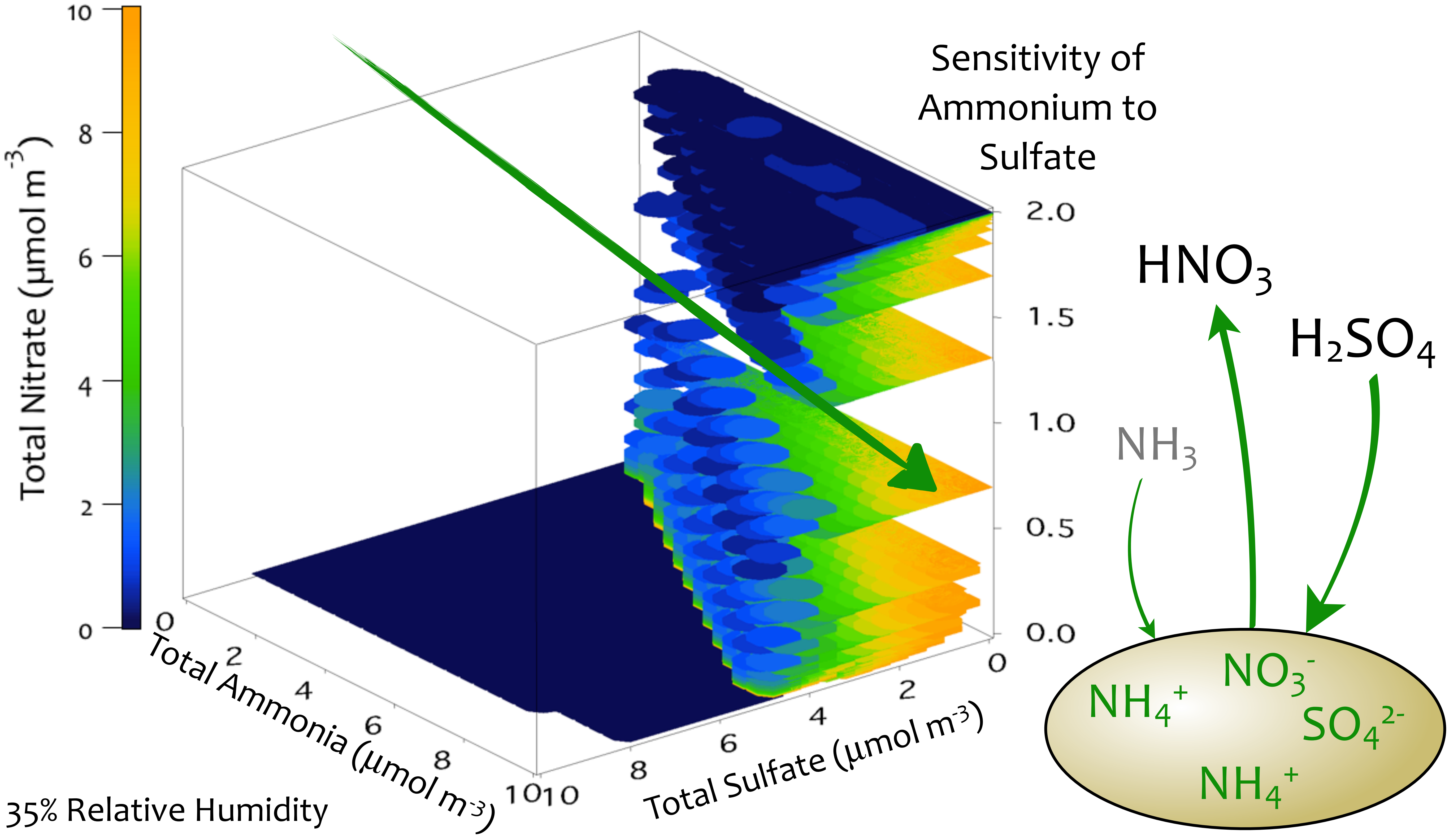
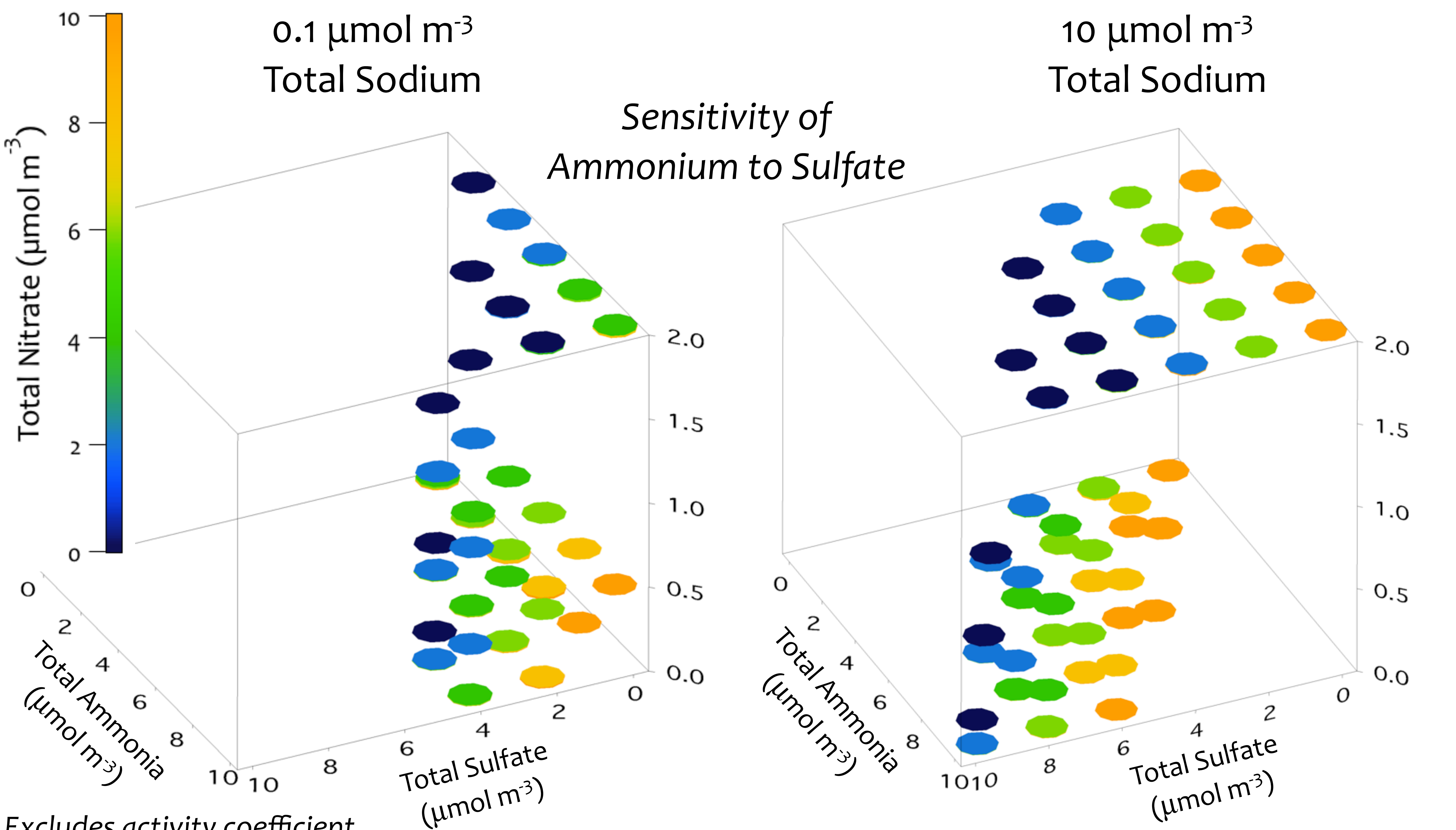


Sensitivity Space:
Ammonium, Sulfate, Nitrate, Chloride, and Sodium

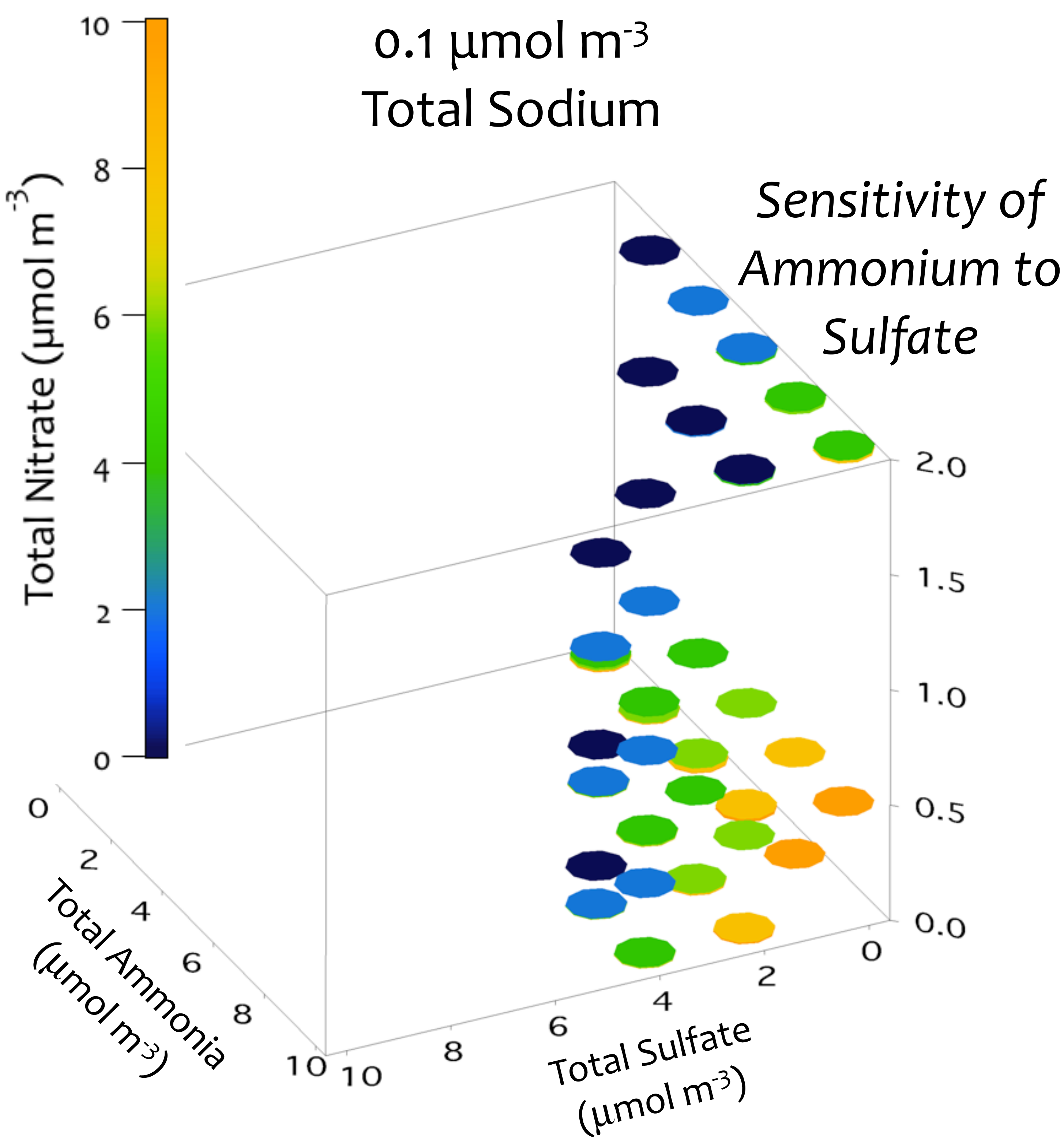


Excludes activity coefficient sensitivities in neutralized aerosol

0.1 μmol m^{-3} Total Chloride, 85% Relative Humidity

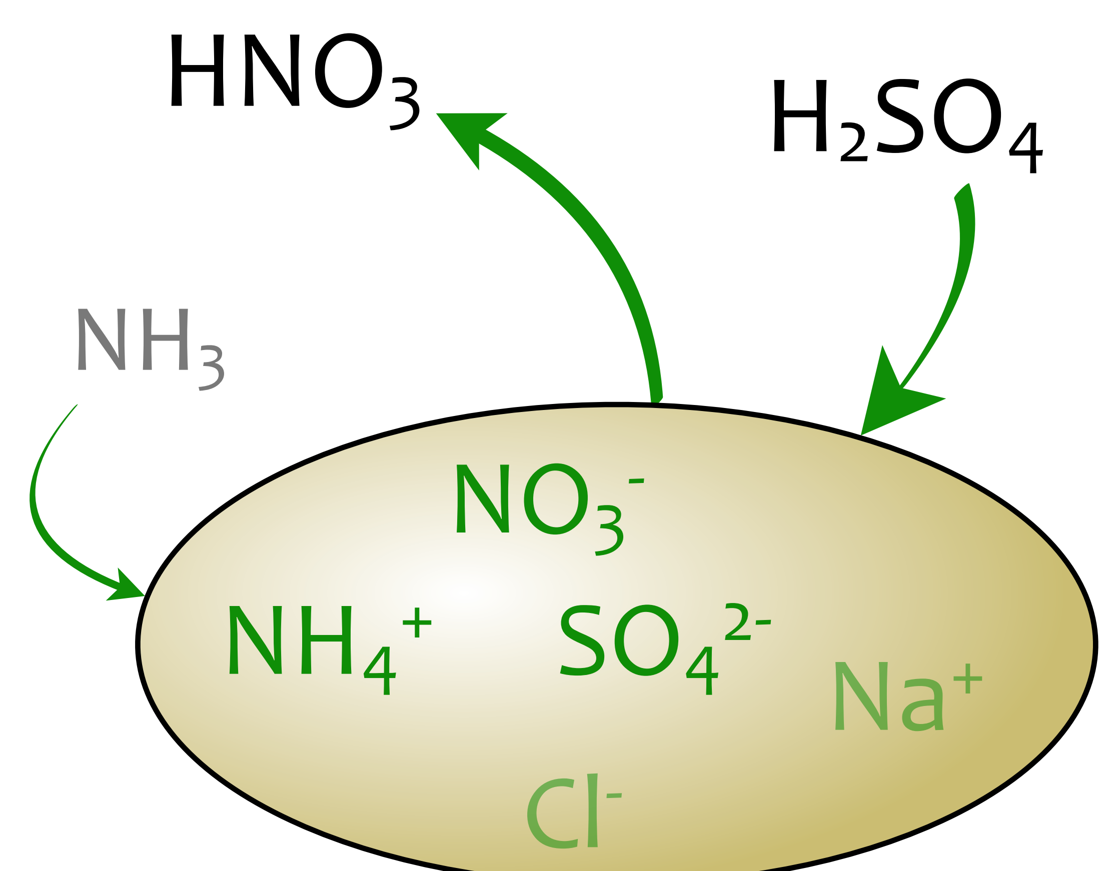
Sensitivity Space:

Ammonium, Sulfate, Nitrate, Chloride, and Sodium



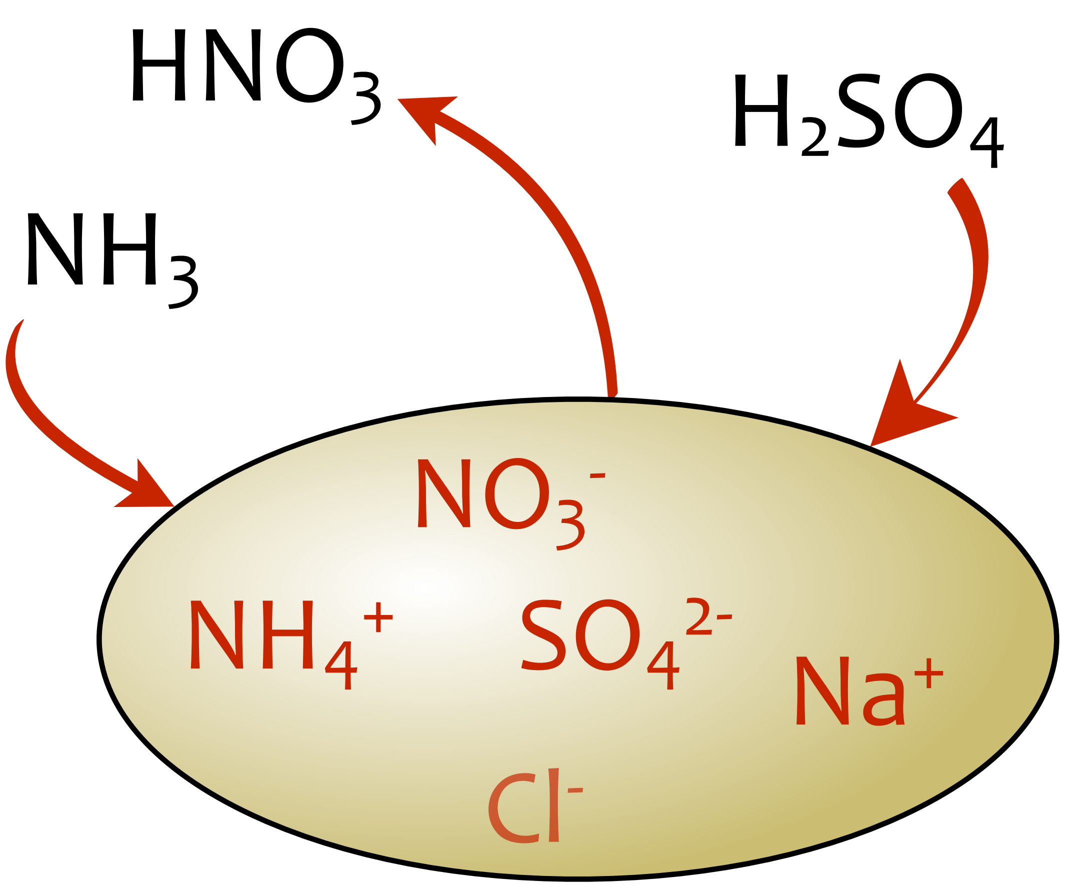
0.1 µmol m^{-3} Total Chloride, 85% Relative Humidity

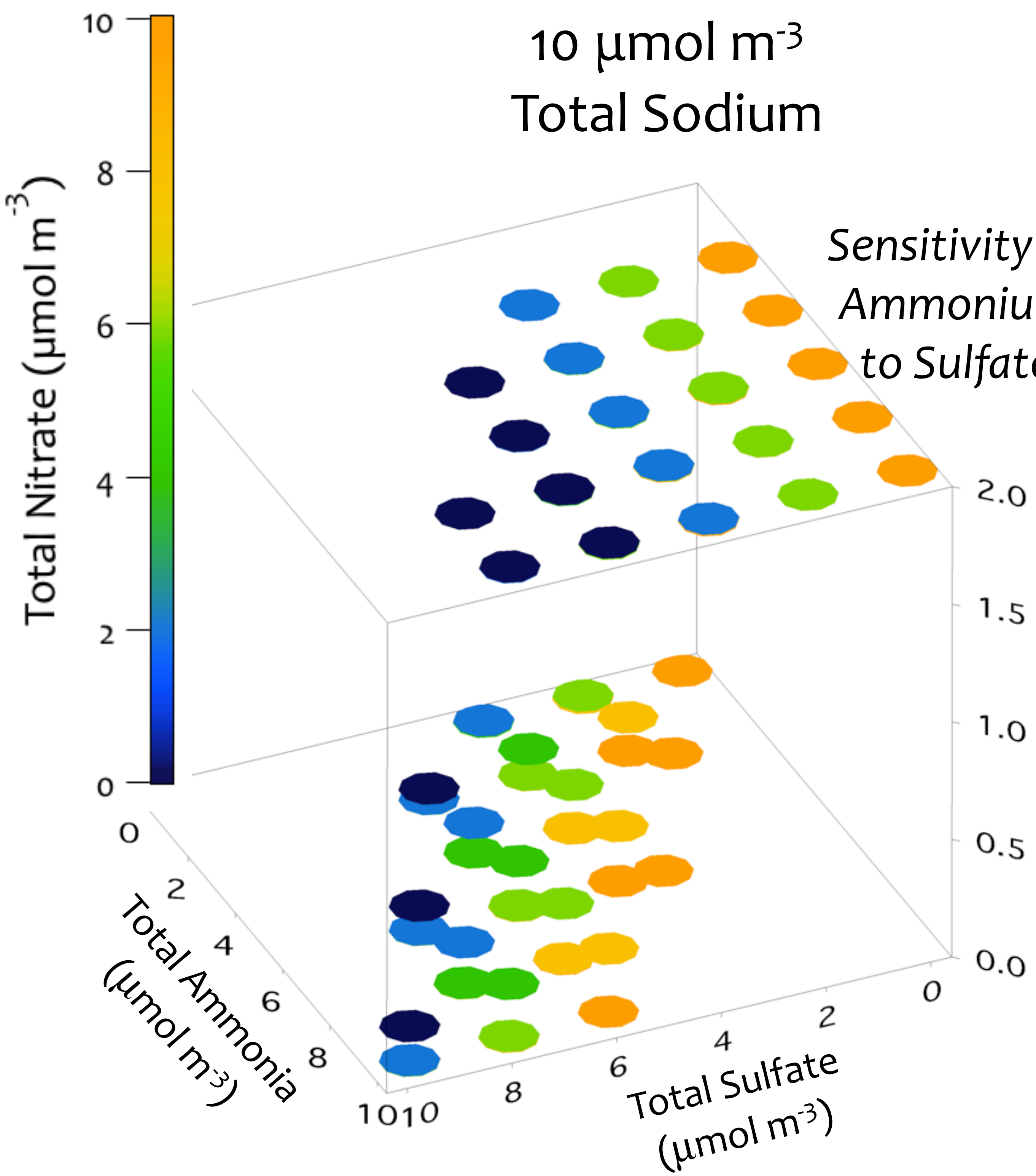
- Limited by 2:1 ammonium to sulfate ratio
- Buffering effect of nitrate



Sensitivity Space:
Ammonium, Sulfate, Nitrate, Chloride, and Sodium

- Sodium extends range of non-zero ammonium sensitivity
- Buffering effect of nitrate





0.1 µmol m-3 Total Chloride, 85% Relative Humidity

Future Work

- Attributing speciated aerosol concentration to sources requires implementation within the full CMAQ-Adjoint - *see ShunLiu Zhao's talk at 4:40 pm*

- Further refining routines with Na^{+}-Cl^{-}

- Extending to K^{+}-Mg^{2+}-Ca^{2+}

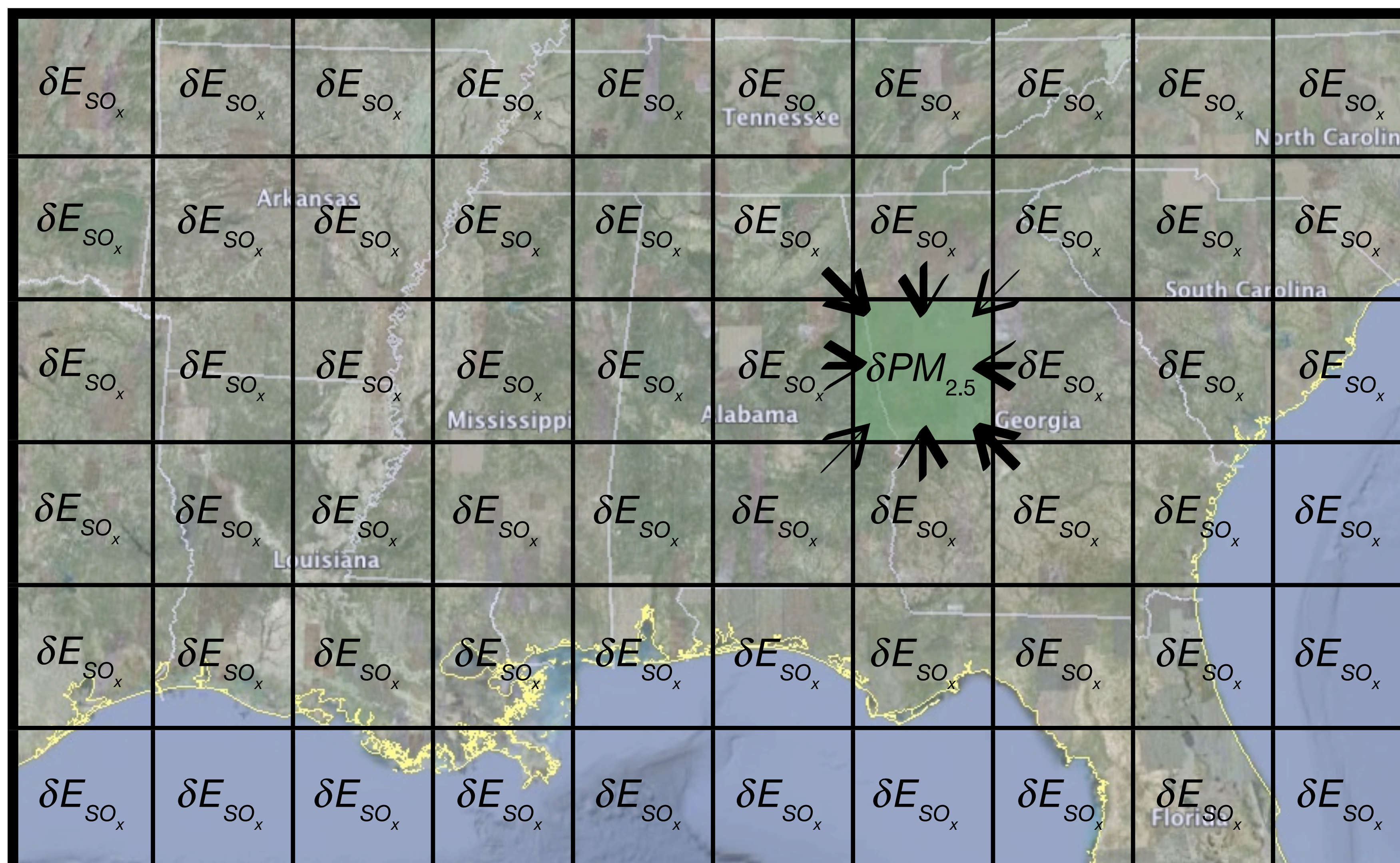


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image credit: Google Earth; adapted from Daven Henze's representation of sensitivity methods