Ammonia variability over the United States (and its potential influence on inorganic particulate matter)

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Particulate matter has large effects on people and the environment.

Substantial PM is formed through inorganic chemistry.

- Health
- Visibility
- Climate
- Nutrient Transport

$\text{SO}_2 \rightarrow \text{H}_2\text{SO}_4$

$\text{NO}_x \rightarrow \text{HNO}_3$

$\text{NH}_3$

$\text{NH}_3 \rightarrow \text{H}_2\text{SO}_4$

$\text{H}_2\text{SO}_4 \rightarrow \text{SO}_4^{2-}$

$\text{H}_2\text{SO}_4 \rightarrow (\text{NH}_4)_2\text{SO}_4$

$\text{HNO}_3 + \text{NH}_3 \rightarrow \text{NH}_4\text{NO}_3$

$f(T, RH)$
Given constant anthropogenic ammonia emissions,
Does the ammonia concentration change over this time period?

If so, What is the cause of this variability?
How does this contribute to the variability of surface PM?
Significant year-to-year variation in ammonia concentrations

IASI Satellite Observations [Van Damme et al., 2014]

GEOS-Chem Base Scenario

JJA Column

<table>
<thead>
<tr>
<th>Year</th>
<th>2008</th>
<th>2009</th>
<th>2010</th>
<th>2011</th>
<th>2012</th>
</tr>
</thead>
</table>

JJA Surface

<table>
<thead>
<tr>
<th>Year</th>
<th>2008</th>
<th>2009</th>
<th>2010</th>
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<th>2012</th>
</tr>
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</table>

Lack of averaging kernel from IASI product prevents direct quantitative comparison

Considerable model variability given fixed anthropogenic ammonia emissions.

What is the source of model variability in concentration?
Model column concentration variability dominated by meteorology

Sensitivity simulations: Anthropogenic $SO_x/NO_x$ emissions and meteorology constant from 2008

<table>
<thead>
<tr>
<th>JJA Column</th>
<th>2008</th>
<th>2009</th>
<th>2010</th>
<th>2011</th>
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</thead>
<tbody>
<tr>
<td>GEOS-Chem</td>
<td></td>
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<tr>
<td>Base Scenario</td>
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</tr>
</tbody>
</table>

Changes due to:
- Anthropogenic $SO_x/NO_x$ Reduction
- Meteorology Variability

Anomaly from 2008

- T [°C] RH [%] PBL H [m] - Surface Column Mean

$NO_x$ Emissions: All Soil Fertilizer
Model concentration variability dominated by meteorology

Sensitivity simulations: Anthropogenic $\text{SO}_x/\text{NO}_x$ emissions and meteorology constant from 2008

AMoN Surface Observation
GEOS-Chem Base Scenario

Due to Anthropogenic $\text{SO}_x/\text{NO}_x$ Reductions
Due to Meteorology Variability

AMoN Observations vs GEOS-Chem
JJA Only

Base  Fixed $\text{SO}_x/\text{NO}_x$ Emissions  Fixed Met
Model concentration variability dominated by meteorology

Range of seasonal ammonia mean column mass (max-min) from 2008-2012

Due to Anthropogenic $\text{SO}_x/\text{NO}_x$ Decreases

Due to Meteorology Variability

Base Scenario

Due to Anthropogenic $\text{SO}_x/\text{NO}_x$ Decreases

Due to Meteorology Variability

Base Scenario Sampled to IASI

IASI Observed

[Maps showing concentration variability in the U.S. with color scales indicating kg N km$^{-2}$]
Lack of model variability at surface suggests missing processes?

Range of seasonal ammonia mean surface concentrations (max-min)

2009-2012

AMoN Observed   Base Model Scenario

Livestock Population

Surface Temperature

Missing response of ammonia emissions to range of:

2005-2013

2008-2013

2011-2012
Conclusions and Implications

• Meteorology contributes to most of the year-to-year variation in simulated ammonia concentration in both surface and column.

• Lack of surface concentration variability in model could signal missing processes relating to emissions.

• Trend of shifting of ammonia from particle to gas phase due to lower $SO_x/NO_x$ emissions will affect lifetimes, transport of nitrogen.

Continuing Work

• Quantify impacts on PM formation.

• Investigate potential missing processes and impacts of updated and more variable emission inventories.
Extra Slides
IASI Ammonia Column Concentration [Van Damme et al., 2014]

- Morning overpass (9-10 am local time)
- 2008-2013 over North America
- 24 km footprint gridded to 0.5°x0.667° GEOS-5 grid

Filtered for:
1. Cloud Cover < 25%
2. $T_{\text{skin}} > -10$ °C
3. Post-grid Relative Error ≤ 75%
Seasonal mean ammonia column concentration

Changes due to:

Anthropogenic \(SO_x/NO_x\) Decreases

Anthropogenic \(SO_x\) Emissions Changes

Anthropogenic \(NO_x\) Emissions Changes
### Seasonal mean ammonia column concentration

<table>
<thead>
<tr>
<th>Changes due to:</th>
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<tbody>
<tr>
<td>Meteorology</td>
<td><img src="image1" alt="Map" /></td>
<td><img src="image2" alt="Map" /></td>
<td><img src="image3" alt="Map" /></td>
<td><img src="image4" alt="Map" /></td>
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<tr>
<td>Variability</td>
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<td><img src="image6" alt="Map" /></td>
<td><img src="image7" alt="Map" /></td>
<td><img src="image8" alt="Map" /></td>
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<tr>
<td>Surface T Changes</td>
<td><img src="image9" alt="Map" /></td>
<td><img src="image10" alt="Map" /></td>
<td><img src="image11" alt="Map" /></td>
<td><img src="image12" alt="Map" /></td>
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<tr>
<td>Surface RH Changes</td>
<td><img src="image13" alt="Map" /></td>
<td><img src="image14" alt="Map" /></td>
<td><img src="image15" alt="Map" /></td>
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<tr>
<td>PBL Height Changes</td>
<td><img src="image17" alt="Map" /></td>
<td><img src="image18" alt="Map" /></td>
<td><img src="image19" alt="Map" /></td>
<td><img src="image20" alt="Map" /></td>
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<tr>
<td>Soil NO\textsubscript{X} Emissions Changes</td>
<td><img src="image21" alt="Map" /></td>
<td><img src="image22" alt="Map" /></td>
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<td><img src="image24" alt="Map" /></td>
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<tr>
<td>Fertilizer NO\textsubscript{X} Emissions Changes</td>
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<td><img src="image26" alt="Map" /></td>
<td><img src="image27" alt="Map" /></td>
<td><img src="image28" alt="Map" /></td>
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</table>
**Inter-annual correlation (R) of seasonal means**

sampled to days with valid IASI retrievals

<table>
<thead>
<tr>
<th>Season</th>
<th>Scenario</th>
<th>Map</th>
<th>Correlation Bar Chart</th>
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<tr>
<td>JJA</td>
<td><strong>Base Scenario</strong></td>
<td><img src="image1.png" alt="Map" /></td>
<td><img src="image2.png" alt="Bar Chart" /></td>
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<tr>
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<td>Fixed Anthropogenic Emissions</td>
<td><img src="image3.png" alt="Map" /></td>
<td><img src="image4.png" alt="Bar Chart" /></td>
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<td>Fixed Meteorology</td>
<td><img src="image5.png" alt="Map" /></td>
<td><img src="image6.png" alt="Bar Chart" /></td>
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**JJA IASI vs GEOS-Chem Ammonia Column Mass**

- **Base Scenario**
  - Fixed Anthropogenic Emissions
  - Fixed Meteorology

Solid: $R > 0$, Shaded: $R < 0$
Inter-annual correlation (R) of seasonal means

JJA

GEOS-Chem Emission/Meteorology vs GEOS-Chem Ammonia Column Mass:

- Anthropogenic SO$_X$
- Anthropogenic NO$_X$
- Soil and Fertilizer NO$_X$
- Surface T
- Surface RH
- PBL Height

SE: Base Scenario
FE: Fixed Anthropogenic Emissions
FM: Fixed Meteorology

No Data -1.0 -0.5 0.0 0.5 1.0