SOURCE-RECEPTOR RELATIONSHIPS
FOR TOTAL NITRATE IN
NORTHEAST ASIA

A&WMA International Specialty Conference:
Leapfrogging Opportunities for Air Quality Improvement

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This study was performed as a part of LTP project

Joint Research Project on Long-Range Transboundary Air Pollutants in Northeast Asia (LTP project)

- This joint project started in 1995 by the initiative of NIER of Korea with the goal of addressing long-range transboundary air pollutants in Northeast Asia, China, Japan and Korea, by conducting a joint research and analysis.

- The source-receptor relationship among the three countries concerning total nitrate is a significant point in LTP project

- According to LTP expert meeting, this study will be completed in 2011

- This presentation provide the result of the first year
Design of Research
Model & Conditions

- CMAQ 4.6 & MM5 3.7
- Modeling domain
  - 20°N~50°N, 100°E~150°E
  - 60km horizontal grids
  - 24 vertical layers
  - 90 × 60 grid
- Modeling Period
  - March, July, October, and December in 2006
  - Concentration and dry/wet deposition of air pollutants
    - 1 hour resolution
    - 5 day pre-running
• Anthropogenic emissions for SO$_2$, NOx, NH$_3$, VOCs, CO, PM$_{10}$, PM$_{2.5}$
  – Emissions were prepared based on INTEX-B (Zhang et.al, 2007)
    • except NH$_3$
    • Base year 2006
  – Volcanic emissions for 2002 were included
    • Release height assumed to be about 1500m
  – Natural emissions are not taken into account
• Modeling emission inventories were produced by SMOKE v. 2.1
  – SAPRC99 chemical mechanism
  – no temporal variation (uniform)
  – Source sectors in INTEX-B and TRACE-P were reorganized based on EDGAR (Olivier et al.. 2005) and mapped with USEPA’s SCC, and GIS technique has been applied to convert gridded emissions into admin-based emissions for preparing USEPA’s IDA format
SMOKE Output

CMAQ ready format – Base case (60km×60km, ground level)
Emissions by Regions

Region I
- CO: 24,328,692 ton
- NOx: 3,192,879 ton
- PM$_{10}$: 2,422,458 ton
- SO$_2$: 2,922,261 ton
- VOCs: 3,171,543 ton
- NH$_3$: 1,602,414 ton

Region II
- CO: 91,674,167 ton
- NOx: 11,037,177 ton
- PM$_{10}$: 10,069,961 ton
- SO$_2$: 18,469,483 ton
- VOCs: 12,149,473 ton
- NH$_3$: 7,158,629 ton

Region III
- CO: 44,682,196 ton
- NOx: 5,821,856 ton
- PM$_{10}$: 5,167,885 ton
- SO$_2$: 9,401,411 ton
- VOCs: 7,036,937 ton
- NH$_3$: 4,573,795 ton

Region IV
- CO: 762,621 ton
- NOx: 983,221 ton
- PM$_{10}$: 282,591 ton
- SO$_2$: 449,727 ton
- VOCs: 622,033 ton
- NH$_3$: 195,338 ton

Region V
- CO: 5,319,829 ton
- NOx: 2,325,003 ton
- PM$_{10}$: 191,832 ton
- SO$_2$: 806,005 ton
- VOCs: 2,034,035 ton
- NH$_3$: 342,524 ton

* Not included volcanic emis.
## The Methodology of S-R Relationship

<table>
<thead>
<tr>
<th>No.</th>
<th>Method - scheme</th>
<th>Method - description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>100% A = Dep</td>
<td>Deposition due to emission in country A computed in the model run with emissions from country A only.</td>
</tr>
<tr>
<td>2.</td>
<td>All - Dep</td>
<td>Deposition due to emission in country A computed as a difference between the model run with all emissions and the model run with all emissions except emissions from country A.</td>
</tr>
<tr>
<td>3.</td>
<td>All - 90% A = Dep</td>
<td>Deposition due to emission in country A computed as a difference (multiplied by 10) between the model run with all emissions and the model run with all emissions except 10% emissions from country A.</td>
</tr>
<tr>
<td>4.</td>
<td>110% A - All = Dep</td>
<td>Deposition due to emission in country A computed as a difference (multiplied by 10) between the model run with all emissions (+10% emissions from country A) and the model run with all emissions included.</td>
</tr>
</tbody>
</table>

(Source: EMEP/MSC-W, 1999, COMPUTING SOURCE-RECEPTOR MATRICES WITH EMEP EULERIAN ACID DEPOSITION MODEL)
The Methodology of S-R Relationship

- The S-R formula of S-R relationship calculation

\[ R_{ij} = \frac{H_{ij}}{\sum_{i=1}^{n} H_{ij}} \times 100 \text{ (\%)} \]

- \( R_{ij} \) : the fractional contribution of i-th source on j-th receptor
- \( H_{ij} \) : the deposition amount at j-th receptor due to i-th source
  - \( H_{ij} \) is obtained as the difference between the result of control simulation (all emission sources) and the result of a simulation in which all emission sources except 20% NOx emissions from i-th source are considered

- The Total Nitrate
  - Sum of N from nitric acid(HNO$_3$), inorganic nitrate(NO$_3^-$) and organic nitrate(PAN)
SMOKE Output

CMAQ ready format – Perturbed emissions w.r.t region –

- Emissions perturbed for only NOx with 20% reduction rate

Base case: no reduction

Case 1: 20% reduction in reg. 1

Case 2: 20% reduction in reg. 2

Case 3: 20% reduction in reg. 3

Case 4: 20% reduction in reg. 4

Case 5: 20% reduction in reg. 5
Results
Simulation results
- NOx conc. -
**HNO$_3$(g) concentration**

**Simulation results**

**Monthly Averaged Air Temperature in July**
(At 1.5m, degree)

**Monthly Averaged Air Temperature in Dec.**
(At 1.5m, degree)

**Ranged Air Temperature in Mar.**
(At 1.5m, degree)

**Ranged Air Temperature in Jul.**

\[
\text{NH}_3(g) + \text{HNO}_3 \leftrightarrow \text{NH}_4^+ (aq) + \text{NO}_3^- (aq)
\]

\[
\text{NH}_3(g) + \text{HNO}_3 \leftrightarrow \text{NH}_4\text{NO}_3(p)
\]
Monthly Accumulated Precipitation

Simulation results

Monthly Accumulated Precipitation in Mar.

Monthly Accumulated Precipitation in Jul.

Monthly Accumulated Precipitation in Oct.

Monthly Accumulated Precipitation in Dec.
## Simulation results

### Deposition Amount of the Total Nitrate

#### March

<table>
<thead>
<tr>
<th>Region</th>
<th>I</th>
<th>II</th>
<th>III</th>
<th>IV</th>
<th>V</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dry</td>
<td>5.4</td>
<td>12.7</td>
<td>22.7</td>
<td>3.0</td>
<td>17.2</td>
<td>61.0</td>
</tr>
<tr>
<td>Wet</td>
<td>50.8</td>
<td>95.2</td>
<td>71.9</td>
<td>2.4</td>
<td>17.2</td>
<td>237.5</td>
</tr>
<tr>
<td>Total</td>
<td>56.2</td>
<td>107.9</td>
<td>94.6</td>
<td>5.4</td>
<td>34.4</td>
<td>298.5</td>
</tr>
</tbody>
</table>

#### July

<table>
<thead>
<tr>
<th>Region</th>
<th>I</th>
<th>II</th>
<th>III</th>
<th>IV</th>
<th>V</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dry</td>
<td>37.8</td>
<td>64.4</td>
<td>36.3</td>
<td>3.0</td>
<td>9.0</td>
<td>150.6</td>
</tr>
<tr>
<td>Wet</td>
<td>22.3</td>
<td>77.8</td>
<td>52.8</td>
<td>4.0</td>
<td>24.2</td>
<td>181.2</td>
</tr>
<tr>
<td>Total</td>
<td>60.1</td>
<td>142.2</td>
<td>89.2</td>
<td>7.0</td>
<td>33.3</td>
<td>331.8</td>
</tr>
</tbody>
</table>

#### October

<table>
<thead>
<tr>
<th>Region</th>
<th>I</th>
<th>II</th>
<th>III</th>
<th>IV</th>
<th>V</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dry</td>
<td>18.2</td>
<td>44.3</td>
<td>65.2</td>
<td>2.4</td>
<td>8.1</td>
<td>138.2</td>
</tr>
<tr>
<td>Wet</td>
<td>15.9</td>
<td>58.0</td>
<td>49.9</td>
<td>2.4</td>
<td>16.1</td>
<td>142.3</td>
</tr>
<tr>
<td>Total</td>
<td>34.0</td>
<td>102.4</td>
<td>115.1</td>
<td>4.8</td>
<td>24.2</td>
<td>280.5</td>
</tr>
</tbody>
</table>

#### December

<table>
<thead>
<tr>
<th>Region</th>
<th>I</th>
<th>II</th>
<th>III</th>
<th>IV</th>
<th>V</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dry</td>
<td>1.1</td>
<td>7.7</td>
<td>26.4</td>
<td>1.9</td>
<td>18.6</td>
<td>55.8</td>
</tr>
<tr>
<td>Wet</td>
<td>4.0</td>
<td>36.9</td>
<td>42.0</td>
<td>4.2</td>
<td>32.8</td>
<td>119.9</td>
</tr>
<tr>
<td>Total</td>
<td>5.1</td>
<td>44.6</td>
<td>68.4</td>
<td>6.0</td>
<td>51.4</td>
<td>175.7</td>
</tr>
</tbody>
</table>
The Total Nitrate

Total deposition

Simulation results
The Total Nitrate
Dry deposition

Simulation results
Simulation results

HNO

Dry deposition

Monthly Accumulated Dry Deposition of HNO3 in Mar.

Monthly Averaged Air Temperature in Dec.
(at 1.5m, degree)

kg/ha
**Contribution of S-R for the total nitrate deposition in Mar.**

<table>
<thead>
<tr>
<th>Receptor</th>
<th>Dry Deposition</th>
<th>Wet Deposition</th>
<th>Total Deposition</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Source I</td>
<td>I</td>
<td>II</td>
</tr>
<tr>
<td>March</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>I</td>
<td>37.3%</td>
<td>57.4%</td>
<td>3.2%</td>
</tr>
<tr>
<td>II</td>
<td>8.8%</td>
<td>70.6%</td>
<td>12.2%</td>
</tr>
<tr>
<td>III</td>
<td>1.8%</td>
<td>27.8%</td>
<td>68.4%</td>
</tr>
<tr>
<td>IV</td>
<td>16.0%</td>
<td>25.5%</td>
<td>4.0%</td>
</tr>
<tr>
<td>V</td>
<td>16.5%</td>
<td>19.2%</td>
<td>1.3%</td>
</tr>
</tbody>
</table>
Contribution of S-R for the total nitrate deposition in Jul.

<table>
<thead>
<tr>
<th>July</th>
<th>Dry Deposition</th>
<th>Wet Deposition</th>
<th>Total Deposition</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Source</td>
<td>Source</td>
<td>Source</td>
</tr>
<tr>
<td></td>
<td>I</td>
<td>II</td>
<td>III</td>
</tr>
<tr>
<td>I</td>
<td>60.3%</td>
<td>37.5%</td>
<td>0.1%</td>
</tr>
<tr>
<td>II</td>
<td>4.8%</td>
<td>76.4%</td>
<td>12.3%</td>
</tr>
<tr>
<td>III</td>
<td>0.8%</td>
<td>32.7%</td>
<td>63.8%</td>
</tr>
<tr>
<td>IV</td>
<td>0.5%</td>
<td>5.5%</td>
<td>3.0%</td>
</tr>
<tr>
<td>V</td>
<td>1.0%</td>
<td>8.9%</td>
<td>7.6%</td>
</tr>
</tbody>
</table>
**Contribution of S-R for the total nitrate deposition in Dec.**

### S-R relationship

<table>
<thead>
<tr>
<th>Receptor</th>
<th>Dry Deposition</th>
<th>Wet Deposition</th>
<th>Total Deposition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Source</td>
<td>Source</td>
<td>Source</td>
<td>Source</td>
</tr>
<tr>
<td>I</td>
<td>II</td>
<td>III</td>
<td>IV</td>
</tr>
<tr>
<td>I</td>
<td>55.7%</td>
<td>42.6%</td>
<td>0.9%</td>
</tr>
<tr>
<td>II</td>
<td>28.0%</td>
<td>51.2%</td>
<td>17.4%</td>
</tr>
<tr>
<td>III</td>
<td>2.5%</td>
<td>52.6%</td>
<td>44.3%</td>
</tr>
<tr>
<td>IV</td>
<td>32.8%</td>
<td>49.7%</td>
<td>1.1%</td>
</tr>
<tr>
<td>V</td>
<td>19.7%</td>
<td>46.1%</td>
<td>2.6%</td>
</tr>
</tbody>
</table>
Deposition by Region 2

- The difference between the result of base case simulation (all emission sources) and that of a simulation with all emission sources minus 20% NOx emissions from i-th source
  - Deposition amount by i-th source

![Total deposition decrease of the total nitrate by 20% NOx reduction in Region 2 in Mar.](image1.png)

![Total deposition decrease of the total nitrate by 20% NOx reduction in Region 2 in Jul.](image2.png)

![Total deposition decrease of the total nitrate by 20% NOx reduction in Region 2 in Oct.](image3.png)

![Total deposition decrease of the total nitrate by 20% NOx reduction in Region 2 in Dec.](image4.png)
• Impact of Central China exceeded self-contribution in northern China and Korea region

• Impact of Japan also evident in Korea even for non-summer months

• Depositions in coastal land areas overestimated due to grid allocation in S-R method

• Without temporal emission variation, seasonal meteorological and chemical variations still big influence in S-R relationship
On-going Work

• Review of S-R method
  – Optimum perturbation factor
  – EMEP-4 method

• Development of alternative aerosol module for CMAQ
  – ISOROPIA -> UHAERO

• Temporal variation
  – Apply monthly temporal allocator

• 2006 year-long simulation
• This research was supported in part by the ‘‘Development of common next-generation acid-deposition module and quantitative analysis for nitrogen oxide source-receptor relationship( I )’’ project sponsored by the Korea National Institute of Environmental Research (NIER) and we thank them for their support.
Thank you for your attention
S-R relationship for total reactive nitrogen deposition

- Meiyun Lin et al., 2008, “Long-range transport of acidifying substances in East Asia-Part II”, AE, 42(24)
  - Region-to-region source–receptor relationships for total reactive nitrogen deposition in East Asia.
  - CMAQ
    - Including anthropogenic, volcanic, soil NOx, biogenic VOCs sources
  - NOx perturbation factor of 25%

Vertical bars show the origin of substances deposited in each “receptor” region. Values given show percentage of total deposition in a “receptor” region contributed from different “source” regions. Red series is used for seven source regions in China, whereas blue series shows source regions in Southeast Asia.
• Meiyun Lin et al., 2008, “Long-range transport of acidifying substances in East Asia-Part II”, AE, 42(24)

Contribution from 25% reduced emissions in Central China to dry depositions of nitric acid and aerosol nitrate in January 2001, derived from the sensitivity simulations, 25RM_SN_NH3 (upper panels) and 25RM_SN (lower panels).

- **25RM_SN_NH3**: a 25% reduction of SO2, primary sulfate, NOx, primary nitrate, and NH3 simultaneously from regional emissions;
- **25RM_SN**: a 25% reduction of SO2, primary sulfate, NOx, and primary nitrate simultaneously from regional emissions.

Contribution from 25% reduced emissions in Central China to dry depositions of nitric acid and aerosol nitrate in January 2001, derived from the sensitivity simulations, 25RM_SN_NH3 (upper panels) and 25RM_SN (lower panels).
Simulation of Japan

Model description

Off-line coupled meteorology / chemical transport model (WRF/RAQM)

Weather Research and Forecasting model ver3.0 (Skamarock et al., 2008)
- Advanced Research WRF core
- PBL physics Mellor-Yamada-Janjic (Janjic, 2002)
- Cloud microphysics WSM5 (Lin et al., 1983)
- Cumulus convection Grell-Devenyi ensemble scheme (Grell and Devenyi, 2002)
- Long-wave radiation Rapid Radiative Transfer Model (RRTM, Mlawer et al., 1997)
- Short-wave radiation Dudhia scheme (Dudhia, 1989)
- Land-surface model Noah scheme (Chen and Dudhia, 2001)

Regional Air Quality Model (An et al., 2002; Han et al., 2006)
- Emission Provided by LTP secretariat
- Photolysis Madronich (1987)
- Photochemistry Condensed CB-IV (He and Huang, 1992)
- Gas-aerosol equilibrium ISSOROPIA (Nenes et al., 1998)
- Aqueous chemistry RADM (Chang et al., 1987)
- Dry deposition Walmsley and Wesely (1996)
- Wet deposition RADM (Chang et al., 1987)
Concentrations of (left) HNO$_3$ gas and (right) aerosol NO$_3^-$.
Concentrations of (left) HNO$_3$ gas and (right) aerosol NO$_3^-$

Seasonally mean HNO$_3$ conc. [ppb] in Spring 2006

Seasonally mean nitrate conc. [ug/m$^3$] in Summer 2006
Air Quality Model

- Air Quality model
  - CMAQ 4.6
  - Advection, horizontal and vertical diffusion
  - Gas-phase Chemical Mechanisms
    - SAPRC99 chemical mechanism
  - Aerosol Module
    - The 3rd generation CMAQ aerosol model
  - Dry/Wet deposition and cloud process
    - RADM
Meteorological Fields

- **MM5 v. 3.7**
  - The PSU/NCAR mesoscale model
- **Domain 1**
  - Horizontal grid
    - 180km grid size
    - 51×47 horizontal grid points
- **Domain 2**
  - Horizontal grid
    - 60km grid size
    - 109×82 horizontal grid points
- **Vertical properties**
  - 23 level of stretched vertical grids
  - Model top at 100hPa height
- **Using FDDA**
  (Four-dimensional data assimilation)

**Input fields**
- Source data : FNL
  - Spatial resolution : 1.0 ° X 1.0 °
  - Temporal resolution : 6h
- Domain for meteorological fields is larger than that for CMAQ domain
Gridded Emissions Data

- Annual emissions for 2006 -

- $\text{SO}_2$
- $\text{NOx}$
- $\text{CO}$

- $\text{PM}_{10}$
- $\text{VOCs}$
- $\text{NH}_3$
Locations of monitoring sites
Monitoring and Simulation Result

Validation of Simulation

SO₂ in Rishri

SO₂ in Ganghwa

NO₂ in Gosan

NO₂ in Ganghwa

Rhishri and Gosan located background area

Ganghwa located near urban area
Validation of Simulation

Monitoring and Simulation Result

- **O₃ in Oki**
  - Graph showing the observed (Obs.) and simulated (Simul.) concentrations of O₃ in Oki over months 1 to 12.

- **O₃ in Taean**
  - Graph showing the observed (Obs.) and simulated (Simul.) concentrations of O₃ in Taean over months 1 to 12.

- **CO in Taean**
  - Graph showing the observed (Obs.) and simulated (Simul.) concentrations of CO in Taean over months 1 to 12.

- **CO in Ganghwa**
  - Graph showing the observed (Obs.) and simulated (Simul.) concentrations of CO in Ganghwa over months 1 to 12.
Nitrate concentration

Simulation results

Nitrate Monthly Average Concentration in Mar.

Nitrate Monthly Average Concentration in Jul.

Nitrate Monthly Average Concentration in Oct.

Nitrate Monthly Average Concentration in Dec.
Dry Deposition Velocity of HNO$_3$

Simulation results

Monthly Averaged Deposition Velocity for HNO$_3$ in Mar.

Monthly Averaged Deposition Velocity for HNO$_3$ in Jul.

Monthly Averaged Deposition Velocity for HNO$_3$ in Oct.

Monthly Averaged Deposition Velocity for HNO$_3$ in Dec.
The total nitrate
- Sum of N from nitric acid, nitrate, PAN

Dry deposition
- Nitric acid >> Nitrate

Wet deposition
- Nitrate >> Nitric acid

PAN showed very low deposition amount
- Below 10% of nitrate plus nitric acid

Large contribution in Region II and III
- 89.2 ktonN ~ 142.2 ktonN, except December
  - Relatively low deposition in December

Region I, III and V had similar amount (44.6~68.4 ktonN)
- Most part of total nitrate deposition appeared in near ocean along coastal belt not inland.
The Total Nitrate
Wet deposition

Simulation results
Nitrate
Wet deposition

Monthly Accumulated Wet Deposition of Nitrate in Mar.

Monthly Accumulated Wet Deposition of Nitrate in Jul.

Monthly Accumulated Wet Deposition of Nitrate in Oct.

Monthly Accumulated Wet Deposition of Nitrate in Dec.
Results

S-R Relationships
## Contribution of S-R for the total nitrate deposition in Oct.

### Table: Total Nitrate Deposition in Oct.

<table>
<thead>
<tr>
<th></th>
<th>Dry Deposition</th>
<th>Wet Deposition</th>
<th>Total Deposition</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Source I</td>
<td>II</td>
<td>III</td>
</tr>
<tr>
<td>Receptor</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>I</td>
<td>40.6%</td>
<td>52.1%</td>
<td>3.5%</td>
</tr>
<tr>
<td>II</td>
<td>15.0%</td>
<td>74.7%</td>
<td>4.7%</td>
</tr>
<tr>
<td>III</td>
<td>5.5%</td>
<td>43.9%</td>
<td>45.2%</td>
</tr>
<tr>
<td>IV</td>
<td>22.1%</td>
<td>12.0%</td>
<td>2.2%</td>
</tr>
<tr>
<td>V</td>
<td>10.6%</td>
<td>8.9%</td>
<td>3.0%</td>
</tr>
</tbody>
</table>

### Diagrams:

1. **The total nitrate S-R relationship for dry deposition in Oct.**
2. **The total nitrate S-R relationship for wet deposition in Oct.**
3. **The total nitrate S-R relationship for total deposition in Oct.**

**Legend:**
- Source I
- Source II
- Source III
- Source IV
- Source V
Depositions by Region 4, 5