Emissions, Transport, and Evolution of Atmospheric Pollutants from China: An Observational Study

Can Li
Department of Atmospheric and Oceanic Science
University of Maryland
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Dissertation Committee:
Dr. Zhanqing Li
Dr. Russell R. Dickerson
Dr. Sheryl H. Ehrman
Dr. Nickolay A. Krotkov
Dr. Ross J. Salawitch
Dr. Jeffrey W. Stehr
Beijing AERONET AOD: 0.3 (440 nm)

NASA EOS/Aqua MODIS

Beijing AERONET AOD: 1.8 (440 nm)
Industrialization and Urbanization

Source: Chinese Bureau of Statistics

<table>
<thead>
<tr>
<th>Year</th>
<th>GDP (billion RMB)</th>
<th>Power Generation (billion KWH)</th>
<th>Coal Production (million ton)</th>
<th>Population (million)</th>
<th>Number of Vehicles (million)</th>
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</thead>
<tbody>
<tr>
<td>1978</td>
<td>365</td>
<td>257</td>
<td>618</td>
<td>302</td>
<td>1.36</td>
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<tr>
<td>1990</td>
<td>1867</td>
<td>612</td>
<td>1080</td>
<td>577</td>
<td>5.51</td>
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<td>2006</td>
<td>21087</td>
<td>2866</td>
<td>2373</td>
<td>1314</td>
<td>37.0</td>
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</tbody>
</table>

- **GDP (billion RMB)**
  - Industry
  - Urban

- **Power Generation (billion KWH)**
  - Industry
  - Urban

- **Coal Production (million ton)**
  - Industry
  - Urban

- **Population (million)**
  - Industry
  - Urban

- **Number of Vehicles (million)**
  - Industry
  - Urban
• Near Emission Sources…
  – **Health effects** [WHO and UNDP, 2001];
  – **Loss of crop yield** [Chameides et al., 1999a, 1999b];
  – **Damage to ecosystems** [e.g., Yang et al., 2001];
  – **Poor visibility in cities**;
  – **Impacts on weather and climate** [e.g., Menon et al., 2002; Qian et al., 2006; Xu et al., 2006; Yu et al., 2002] – may further influence ecosystems;
  – **Estimated economic loss** – 25 billion USD in 2004 (1.2% of GDP [China EPA and China CBS, 2006], 7% of GDP [World Bank, 1997])

• Downwind…
  – **Influence air quality of N America** [e.g., Jaffe et al., 2003];
  – **Affect weather and climate downwind** [Zhang et al., 2007];

**Factors determine the large-scale impacts?**
Scientific Questions and Approaches

• Amount of pollutants emitted – emission inventories for China: how good are they? - Ground measurements near source regions

• Pollutants even if emitted in large amount, have minimal large-scale effects until lofted out of the boundary layer:

• What are some important meteorological mechanisms lofting pollutants over China? - Aircraft measurements over inland China.

• Evolution of pollutants: how do we track pollution plumes on their transport pathway? - Application of satellite observations.
Rural Site ~70 km east-southeast of Beijing.

One of the supersites of EAST-AIRE: (East Asian Study of Tropospheric Aerosols: an International Regional Experiment)

Mixed agricultural land, residence, and light industry

Routinely measured: radiation, sky condition, aerosol optical thickness (AOT), meteorology.

First intensive field campaign of EAST-AIRE (March, 2005): Trace gases \((\text{O}_3, \text{CO}, \text{SO}_2, \text{NO}_y)\), aerosol scattering and absorption, aerosol chemical composition.
Overall, Polluted

<table>
<thead>
<tr>
<th>Location</th>
<th>CO (ppm)</th>
<th>SO$_2$ (ppb)</th>
<th>NO$_2$ (ppb)</th>
<th>Aerosol Scattering (Mm$^{-1}$ or $10^5$ m$^{-1}$)</th>
<th>Aerosol Absorption (Mm$^{-1}$ or $10^5$ m$^{-1}$)</th>
<th>Al (µg/m$^3$)</th>
<th>Fe (µg/m$^3$)</th>
<th>Pb (µg/m$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MD</td>
<td>0.22</td>
<td>2.6</td>
<td>8.1</td>
<td>68</td>
<td>4.7</td>
<td>0.21</td>
<td>0.15</td>
<td>0.005</td>
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<tr>
<td>YRD</td>
<td>0.65</td>
<td>15.9</td>
<td>13.9</td>
<td>154</td>
<td>18.3</td>
<td></td>
<td></td>
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<tr>
<td>XH</td>
<td>1.09</td>
<td>17.8</td>
<td>26.0</td>
<td>358</td>
<td>65.4</td>
<td></td>
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<tr>
<td>XH</td>
<td>0.85</td>
<td>75</td>
<td>7.2</td>
<td>9.5</td>
<td>9.5</td>
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</tr>
<tr>
<td>XH</td>
<td>0.059</td>
<td>3.3</td>
<td>4.1</td>
<td>4.1</td>
<td>4.1</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Pb in North American Cities in 1970s
0.34-1.81 µg/m$^3$ [Ondov et al., 1982]

“XH”, Xianghe, this study; “MD”, Eastern Shore, MD [Stehr et al., 2000]; “YRD”, Yangtze River Delta Region [Wang et al., 2002; 2004]; “IL”, Bondville, IL [Delene and Ogren, 2002]; “SDZ”, Shangdianzi, ~100 km NE of Beijing [Yan et al., 2008]; New Castle, NH [Pike and Moran, 2001]; Fort Meade, MD [Chen, 2002]
Pollution Responds to Weather

Source: [Streets et al., 2003]
Pollutant Ratios: Comparison to Emission Inventory

Table 3.1. Observed and national inventory pollutant ratios (ppbv/ppbv)

<table>
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</tr>
</thead>
<tbody>
<tr>
<td>CO/(\text{SO}_2)</td>
<td>35.8</td>
<td>72.1</td>
<td>13.0</td>
<td>17.5</td>
<td>7.7</td>
</tr>
<tr>
<td>CO/(\text{NO}_x)</td>
<td>27.2</td>
<td>25.4</td>
<td>16.7</td>
<td>22.7</td>
<td>8.6</td>
</tr>
</tbody>
</table>

\(^a\) Use \(\text{SO}_2\) and \(\text{NO}_x\) emission data from *Streets et al.* [2003].

\(^b\) Emission inventory is regional.

Large uncertainties in emission inventories? CO underestimated? \(\text{SO}_2\) overestimated? \(\text{SO}_2\) estimates are believed to be less uncertain...

**Measurements at a single site vs. Inventories for large areas (national/regional). Let’s look closer.**
Spatial Distribution of Pollutant Ratio

Group measurements based on backward trajectories calculated using NOAA/ARL HYSPLIT model and NCEP global reanalysis data.

The region West and Southwest of Xianghe:
- CO/\text{SO}_2: 37 vs. 10-17
- CO/\text{NO}_x: 22 vs. 15-24

At least more populated regions seem to have problems …

Also some large uncertainties in emissions from mega cities in China including Beijing…

Small boilers? Traffic? Biofuel?

Source: [Streets et al., 2003, 2006]
Emissions from a Small Coal-Fired Boiler

Small coal-fired boiler ~50 m east of instruments for radiator heat. Running record, wind direction, peak in $\text{SO}_2$ concentration.

Correlation analysis derived emission factors and inventory values:

- $\text{SO}_2$: 15.4 g $\text{SO}_2$/kg vs. 3.29-54.8 g $\text{SO}_2$/kg in inventory [Streets et al., 2003];
- $\text{NO}_x$: 1.8 g $\text{NO}_2$/kg vs. 1.19-2.24 g $\text{NO}_2$/kg [Streets et al., 2003];
- CO: 31 (50) g CO/kg vs. 74-124 g/ CO/kg [Streets et al., 2003, 2006];

Need to check emissions from other sources, especially automobiles…
Source of Aerosols: Black Carbon

Estimated annual BC emissions from China:
1300-2600 Gg/yr

Inventory: 1049 Gg/yr
[Streets et al., 2003]

Better agreement compared to a similar study for India
[Dickerson et al., 2002]

Single Scattering Albedo (SSA): Morning: \(~0.81\), Afternoon: \(~0.85\)
Source of Aerosols: Elements

Calculation of enrichment factor:

$$EF_X = \frac{\left( \frac{X}{Al} \right)_{\text{Aerosol}}}{\left( \frac{X}{Al} \right)_{\text{Crust}}}$$

Abundant crustal elements: soil particles

Enriched pollution tracer elements: anthropogenic emissions

Both soil and anthropogenic sources
Source of Aerosols: Secondary Species

Synoptic change in nitrate/sulfate ratio

\[ \text{SO}_x = \text{SO}_4^{2-} + \text{SO}_2, \text{SO}_4^{2-}/\text{SO}_x \text{ and } \]
\[ \text{NO}_3^-/\text{NO}_y \text{ reflect the fraction of SO}_2 \]
\[ \text{and NO}_x \text{ oxidized} \]

\[ \text{NO}_3^-/\text{NO}_y \text{: mainly through gas-phase} \]
\[ \text{SO}_4^{2-}/\text{SO}_x \text{: some other mechanism …} \]
Source of Aerosols: Secondary Species

Backward trajectories (200, 500, and 1000 m above ground) and EOS/MODIS satellite cloud product synchronized in time and space ($\pm 15$ min and $\pm 2.5^\circ$ lat/lon)

Cloud interaction?

Trajectory RH $\geq 80$

Cloud top near the trajectory height or thick clouds above trajectory height

~70% of samples with relatively high $\text{SO}_4^{2-}/\text{SO}_x$ (or low $\text{NO}_3^-/\text{SO}_4^{2-}$) are possibly related to cloud interaction. In-cloud processing of $\text{SO}_2$ could be important during transport behind cold fronts.
Aircraft campaign
Location: Shenyang; 
~600 km NE of Beijing; 
~6 million people; 
Industrialized region; 
April 1-12, 2005, 8 flights

DU: Dobson Unit: $2.69 \times 10^{20}$ molecules/m²

Aura/OMI SO₂ (2005-06) [Krotkov et al., 2007]

SO₂ emissions [Streets et al., 2003]
April 5, 2005: Flight Ahead of Cold Front
Polluted PBL: almost 20 ppb \( \text{SO}_2 \) and 500 Mm\(^{-1} \) aerosol scattering – SW flow from polluted region in the lower atmosphere, also dust emissions due to strong surface winds.

Pollutants in FT may travel far downwind: *the lofting mechanism*?
Lofting by Convective Clouds over Upwind Industrial Region

MODIS true color image for 04 UTC April 4, 2005 with 2500 m back trajectory superimposed (no precipitation observed on April 4 over the industrial region marked by red disk).
April 7, 2005: Behind Cold Front

[Graphs showing data on temperature, SO₂, aerosol scattering, and O₃ vs. altitude]
Comparing the climatology over N China and the NE US: Similar springtime occurrence of convective clouds (7% vs. 9%), but much less rain in N China.

Dry convection can be very important in exporting pollutants from China, but needs more flights and numerical simulation.

Observe the April 5 episode at larger scale? Where would the pollution plume travel to? How would the plume evolve during transport?
From Space: Lots of Dust and SO$_2$, and over a Large Region

Aura/OMI Operational SO$_2$ April 5

Aura/OMI Aerosol Index April 5

On the Aircraft: Lots of Dust
Initialization of Trajectories: Air Parcel “Tagging”

Model: NOAA/ARL HYSPLIT model;

Data: NCEP Global Reanalysis;

Time: 05UTC, April 5, 2005;

Region: 35-49N, 117-134E;

Forward trajectories initiated from the center of grids $0.5 \times 0.5^\circ$ in size;

Eight Levels: every 500 m from surface to 4000 m AGL;

Each trajectory tagged with MODIS-retrieved AOD and OMI-retrieved SO$_2$, scaled with aircraft-measured vertical profile.
Snapshots of SO$_2$ Distribution by Aura/OMI

First-order correction for:
SO$_2$ vertical profile;
Aerosol and cloud interference;
Viewing geometry
[Krotkov et al., 2007]
Measurements over small area; Detailed vertical information

Measurements over large area without in-situ measurements; Signal/noise ratio may be insufficient for part of the plume

Initial vertical distribution for tracer model

Assuming conserved tracers; Find the part of the plume with strongest signals.

Satellite observes the part of the plume with strongest signals; Tracer Model estimates the fraction observed by the satellite; Aircraft provides information for trajectory and satellite
Observing Part of the Plume, Quantifying the Whole Plume

Select a polygon box (covers a good part of the trajectory-projected main body of the plume)

Estimate the fraction of the plume covered by this box (using trajectory-projected SO$_2$ distribution, total trajectory SO$_2$ remains constant)

Determine the SO$_2$ mass within the box (with satellite data, e.g., April 6)

Use the fraction to weigh the satellite-determined SO$_2$ mass

Each point: a grid with trajectory-projected column SO$_2$ > 0.5 DU

Uncertainties in trajectory calculation, mismatch between satellite and trajectory: five boxes different in size and shape
Change of Total SO$_2$ Loading of the Whole Plume and AOD near the Core Part of the Plume

The satellite sensitivity to SO$_2$ may change during transport: different threshold values.
Aerosols and SO$_2$ initially outside the plume region on April 5 may add to the SO$_2$ and aerosol loadings in polygon boxes on April 6 and 7 – outside contribution.
Hygroscopic growth of aerosols.
SO₂ mass decays with time: overall lifetime 1-4 day, in the range of modeling studies [Berglen et al., 2004; Chin et al., 2000; Koch et al., 1999]; *Assuming all lost SO₂ converts into ammonium sulfate*, would add to ~0.1-0.2 in AOD near plume core on April 6, ~0.2-0.4 on April 7.
In this case, decrease in dust AOD and increase in sulfate AOD are the two major processes in GOCART model contributing to total AOD change near the core part of the plume.
Conclusions

• Substantial uncertainties in emission estimates for China – CO emissions may be higher than expected;
• Heavy aerosol loading (mixed soil and anthropogenic origins), low SSA – intense local solar heating of the lower atmosphere;
• The synoptic change in air pollution driven by mid-latitude cyclones – feedback of pollution on weather systems?
• Over the continent, dry convection may be an important mechanism for long-range transport – over the oceans, WCB may be more important;
• Satellite shown a useful tool in tracking the transport and evolution of pollution plume on regional scale, but needs to be combined with trajectory and chemical transport models, and has large uncertainties.
Future Work

• More field campaigns: add to the data set;
• Emission inventory: factor analysis; CMB method;
• Numerical simulation using chemical transport models: evaluate emission inventories and study transport of pollutants;
• Improved satellite retrievals: quantitative application of satellite data with better uncertainties;
• Applying satellite data to track pollution plumes and their large-scale impacts: e.g., their interaction with clouds in downwind regions.
Thank you!

Questions? Comments?