The Impact of Asian Emissions on Local/Regional/Global Air Quality

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China’s energy needs are projected to continue to grow at substantial rates, and with a continued reliance on coal – Sulfur emissions may follow the same trend.
Motivating Questions

- What are the current and projected trends in energy and emissions?
- How are these impacting human health? Food security?, etc.
- What are the transboundary impacts of Asian emissions within Asia?
- What are the region-specific science issues that need to be addressed (e.g., aerosol interactions)?
- What are present and future impacts of Asian emissions on global air quality? on US air quality?
- To what extent are air pollution and climate change issues linked? And how can these be exploited?
- What cost-effective options are available to address these concerns?
We develop and use a variety of chemical transport models:

- Global (GFDL, Chip Levy)
- Regional (STEM)
- Integrated Assessment (RAINS-Asia)
Transfer of pollutants in the atmosphere

- Pollution may be transported over long distances (ATMOS)
- Sulphur deposition calculated by MATCH model using 'Conventional Development Scenario' emissions
Transboundary Pollution Issues are of Growing Importance

[Graphs and charts showing emission changes and deposition contributions over years.]
Aerosols are Pervasive Throughout Asian Environments

Mineral Dust

BC from Pollution & Fires
A Key Science Issue: Chemistry/Aerosol/Regional Climate Coupling

Radiative Forcing

Anthropogenic Biogenic Tropospheric O$_3$ Stratospheric input

Gas-phase Tropospheric Chemistry

Heterogeneous NOx/NOy chemistry Free radical scavenging? Alteration of Actinic flux

Tropospheric Aerosols

Sea salt Mineral dust Biomass burning particles Sulfates (via gas-phase chemistry) Carbonaceous particles NH$_3$

Tropospheric Clouds

Cirrus

Stratus

Cloud processing

CCN

Flux of gaseous species to clouds

Generation of inorganic and organic aerosols by gas-to-particle conversion and nucleation

Direct effect

Alteration of cloud albedo (indirect effect)

modified after J.H.Seinfeld 1999
What is the Importance of Heterogeneous Chemistry on Mineral Aerosol Surfaces in the Troposphere?

*Answer Requires:*

- Knowing what chemistry occurs on/in the aerosol.
- Quantifying mineral surfaces in space and time.
- Assessing how mineral aerosols age; and how surfaces activate/deactivate, take on water, etc.
- Quantifying aerosol/radiation interactions.
- Resolving chemical composition as a function of size.
Chemical Role of Aerosol Particles in the Atmosphere

Can change the chemical balance of the atmosphere in two ways

Sink

\[ \text{NO}_2 + \rightarrow \text{NO}_2 \]

Reactive Surface

\[ \text{NO}_2 + \rightarrow \text{Q} + \text{NO} \]

\[ \text{H(a)} + \text{NO}_2 \rightarrow \text{HONO} \]
Experimental Considerations

- **Spectroscopic measurements** to provide both qualitative (what reactions are possible) and quantitative information
  - Provide mechanistic information on the molecular level
  - Need to have techniques that can detect gas-phase and surface-bound species
    > Transmission FT-IR Spectroscopy
    > Diffuse Reflectance UV-vis Spectroscopy
    > Mass Spectrometry

- **Kinetic measurements** to provide quantitative information
  - Determine uptake coefficients (sticking coefficients, reaction probabilities) \( \gamma \)
    > Knudsen cell apparatus

- **Provide data as input for global atmospheric models** - removal rate of gas-phase species \( j \)
  \[
  k_j = \int_{r_1}^{r_2} k_{d,j}(r) n(r) \, dr
  \]
  \[
  k_{d,j} = \frac{4\pi^2 D_{j} V}{1 + K_n (\lambda + 4(1 - \gamma)/3\gamma)}
  \]

  \( n(r)dr \) = number density of particles between \( r \) and \( r+dr \)
Scheme for Aldol Condensation
Reaction on Oxide Particles
(Al$_2$O$_3$, Fe$_2$O$_3$, TiO$_2$, CaO, and MgO)

Li et al., JGR, in press, 2001
Summary of Combined Laboratory and Modeling Study

- Spectroscopic probes of gas-phase and adsorbed species along with kinetic measurements provide the necessary information to evaluate reactions of potential importance in the troposphere
  - reaction mechanisms, surface coverage, saturation
  - uptake coefficients
- Diffusion of gases into powdered samples can have a very significant effect on the measured uptake coefficient for powdered samples
  - multiple collisions amplify the observed uptake coefficient
- Atmospheric implications of uptake measurements determined from box-model analysis
  - heterogeneous pathways are competitive with other carbonyl loss mechanisms (e.g. reaction with OH radical)
Three-Dimensional Combined Transport/Chemistry Analysis (STEM-III)

On/Off Line Transport Model

**Boundary Condition**
- Met. Data (ECMWF)
  - Topo
  - Landuse
  - SST

**Emission Data**
- RAINS-ASIA
- China Map
- Energy sectors, fuels, LPS
- On-line: Emission markers; mineral and sea salt emissions

**Transport**
- 3D advection
- Vertical Diff.
- Cloud, Precip.

**Reaction**
- Gas phase
- Aqua. phase

**Removal**
- Dry dep.
- Wet dep.

**Aerosol**
- (size resolved; kinetic and thermodynamic modules)
  - SOA to be added

**3D Chemical Transport Model**
- STEM-OFLT (off-line transport)
- RAMS-ONLT (on-line transport)

Forecasts of 3-D tracer and aerosol fields; and detailed process oriented analysis of observations
Dust, Sulfate, CO, Lightning NO$_x$ during March-April 2000
Model is Able to Capture Many Important Observed Features

Calculated Fine and Coarse Mode Aerosol Distributions in the Boundary Layer; PEM-WEST B
Calculated Fine and Coarse Mode Aerosol Distributions in the Boundary Layer; PEM-WEST B
INTERACTIONS OF SO$_2$ WITH MINERAL AEROSOL CHANGE SULFATE SIZE DISTRIBUTION AS WELL AS THE CHEMICAL LIFETIMES OF SULFUR. THESE INTERACTIONS HAVE IMPLICATIONS FOR RADIATIVE FORCING

Song et al., JGR in press
Calculated Fine and Coarse Mode Aerosol Distributions at 5 km; PEM-WEST B
Calculated Fine and Coarse Mode Aerosol Distributions at 5 km; PEM-WEST B
The Presence of Mineral Aerosol Alters the Partitioning of Nitric Acid

Song et al. J. Atmos. Chem., in press
Impact of Aerosols on the Photochemical Oxidant Cycle Through Photolysis Rates

<table>
<thead>
<tr>
<th>Season</th>
<th>Impact on O3</th>
<th>ΔSulfate</th>
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<th>ΔMineral</th>
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%Change NO2 Photolysis

Ozone variations (%) due to dust

Mineral Optical Depth

%Change

Tianjin

Local time (hour)

NOx
NMHC
O3
HOx
UVB
Nitric Acid Reactions with Calcium Carbonate are not Limited to the Surface, and Change the Water Uptake Properties and Particle Morphology

Goodman et al., JGR, 105:29053, 2000
Mineral Aerosol Perturb Trace Gas Cycles in Many Ways

- Increase SO$_2$ to sulfate conversion rates.
- Increase the importance of dry deposition.
- Decrease the lifetime of SO$_x$, NO$_y$ and VOC?
- Control the partitioning of semi-volatile species (e.g., HNO$_3$).
- Influence precipitation pH.
- Provide reaction channels which may: recycle No$_x$; produce particulate nitrate; make longer chain VOCs; provide radical sources; and indirectly/directly Perturb O$_3$; and alter water uptake.
- Alter photolysis rates.
April 1998 Asian Dust Event

Transport of East Asian Dust Pall Across the Pacific

Courtesy of Rudolf Husar, Washington University, St. Louis: http://capita.wustl.edu/Asia-FarEast/
The Changing Air Quality of the Northern Hemisphere
Pacific Basin

- Pressures are from the Pacific Rim Countries and beyond.
- Changing patterns and growth rates of energy use and resulting emissions are the primary forcing factors -- with East and West following different paths.
- Impacts are local, regional (East and West), basin-wide, and global.
- Complexities in transport and chemistry over the Pacific greatly challenge present modeling and measurement efforts.
Starting....

GFDL/GCTM
Contribution of Fossil Fuel Burning to Tropospheric Ozone

Yienger, et al, JGR 2000
Asian Contribution to Total Ambient Ozone over Central California
500mb (6 hour averages)

ppb
Dec Jan Feb Mar Apr May Jun

Yienger et al.,
JGR, 26931,
2000

940mb (6 hour averages)

ppb
Dec Jan Feb Mar Apr May Jun

Yienger et al.,
JGR, 26931,
2000
Further Quantification Requires

- *Continued laboratory studies*: understanding the role of water, surface reactions, etc.

- *Ambient measurements*: mineral aerosol size distributions; ammonia concentrations; aerosol chemical composition as a function of size (including the large particles!); dry and wet deposition of the major cations.
ACE-Asia (NSF) & TRACE-P (NASA)

Spring 2001 Experiments

NASA/GTE DC-8
Development and Application of Chemical Weather Forecasting System over East Asia

Itsushi Uno (RIAM/Kyushu-U), Gregory R. Carmichael (UI/CGRER)

We are developing and applying an operational chemical weather forecasting system based on 3-D on-line regional scale chemical transport model fully coupled with RAMS (Regional Atmospheric Modeling System, Pielke et al., 1992). This system consists of several important components; i) operational global forecast data set access to NCEP and JMA, ii) RAMS weather forecast for 72-96 hours based on the NCEP & JMA data as a lateral boundary condition, iii) On-line chemical transport calculation of important chemical tracers (SO$_2$/SO$_4$, mineral dust, black carbon and sea-salt, etc.) and iv) post-processing of “chemical weather forecast” results with 2/3-D graphics into the WWW-page.

One of the main purposes of this system is to understand the regional transboundary air pollution and to schedule/design the operational field monitoring campaign during the ACE(Aerosol Characterization Experiment)-Asia and Trace-P intensive observations.
Application & Model Validation

• ACE-Asia & TRACE-P Field Campaign Planning
• Field Observation Design
• Traffic (Aviation)
• Visibility
• Environmental Assessment

• Model Validation for Episodic Application
  – Yellow Sand (Kosa) April 1998 & March 2000
  – SO$_2$/SO$_4$ climate simulation

Simulated Kosa Onset April 98
Observed sulfate concentration at Osaka compared with model results show a good agreement, and the intermittency during the winter season and the periodicity typical of spring/fall rainy seasons, when the alternance of high/low pressure systems characterize the meteorology of the region, is nicely reproduced by the numerical model (RAMS on-line transport model).

Modeled twelve hours averaged concentration (straight line) and daily averaged observation (dot line)
Several useful atmospheric tracer to understand the origin of air mass.

Tracers/Markers:
- SO2/Sulfate
- DMS
- BC
- OC
- Volcanic
- Megacities
- CO fossil
- CO-Biomass
- Ethane
- Ethene
- Sea Salt
- Radon
- Sea Salt
- Lightning NOx
- Dust 12 size bins
Asia Megacity Tracers

1) White Isosurface: 10% CO from Megacities
2) Green Isosurface: 4 ppb O3 from Megacities
3) Colored Contours for CO from all sources at 990 mb (sigma)
4) Wind vectors at 950 mb (sigma)
Rainy Meso Front divides air mass
Frontal Outflow is the Key Transport Mechanism and needs to be Better Quantified
Frontal Outflow: Dust (brown) & Sulfate (green)
Elevated dust layer comes first and then dense dust layer appears in PBL (below 2km) associated with the movement of low-pressure system.
Comparison of Predicted Dust and BC Distribution to Satellite Information
April 21

POES (dust)

TOMS AI overlaid Dust (Yellow Sand) and BC

TOMS-IA (Color shading)  
Dust Isosurface (Yellow)  
BC Isosurface (Orange)  
Precipitation (Contours)

TOMS AI and Precip
Fly here to sample high O₃
Methodology for Asian Emission Estimates

- Energy Use
- RAINS-Asia Model
- Emission factors, Regulations
- Activity data
- Other human activities
- Biomass burning
- Biogenic, Volcanic...
- Natural emissions

- Anthropogenic emissions
- "Total" emissions
- Emission Controls

“Total” emissions
Organization of Emissions Data

National (23 countries)
- Historical: 75-95 (5-yr)
- Current: 90-00 (1-yr)

Regional (94 regions)
- Projections: 95-30 (5-yr)

Urban (22 cities)

Point (355 sources)
- 1° x 1° down to 1 km x 1 km

Species:
- SO₂, NOₓ, SO₂, BC, NH₃, NOₓ, PM₁₀, OC, CO, PM₂.5, NMVOC

Gridded emissions

Lat/long
Emission Data

Overview

Emission inventories are being developed in support of the Ace-Asia and Trace-P Experiments. An overview of the Methodology for Asian Emission Estimate and Organization of Emissions data are shown below.

Methodology for Asian Emission Estimates

- Energy Use
- Other Human Activities
- Biomass Burning
- Emissions from, Emissions to, and Emissions by anthro. sources
- Natural emissions

Organization of Emissions Data

- National (20 countries)
- Regional (64 regions)
- Urban (25 cities)
- Point (355 sources)
- Historical (1940-1995)
- Projected (2000-2020)

Datasets

<table>
<thead>
<tr>
<th>Level</th>
<th>Species</th>
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<tr>
<td>Regional</td>
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<td>2) CO</td>
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<td>3) NMVOC</td>
</tr>
<tr>
<td></td>
<td>4) NOx</td>
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<tr>
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<td>5) SO₂</td>
</tr>
<tr>
<td>1° x 1°</td>
<td>1) Black Carbon</td>
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<td>3) NMVOC</td>
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<td>4) NOx</td>
</tr>
<tr>
<td></td>
<td>5) SO₂</td>
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</table>

Gridding Capabilities

We will put procedures and examples about the gridding here.

Gridding Procedure for Black Carbon
SULFUR EMISSIONS IN ASIA ARE CHANGING, REGION BY REGION, IN DIFFERENT WAYS
UNEXPECTED DECREASES IN CHINA’S SULFUR EMISSIONS --- CAN THESE REDUCTIONS BE SUSTAINED?

New Two-Zone Control Policy

Streets et al. Atmos. Env., 2000

SEPA
The Rising Role of the Transport Sector in Asia

A NO$_x$ projection for East Asia
Source: Klimont et al., IIASA
Emissions of VOC in East Asia
(Mt VOC)

Million tons

- Other
- Combustion (industry, residential)
- Chemical industry
- Solvent use

1990 1995 2030-CLE 2030-NFC
Structure of VOC emissions in 1995 for selected countries
Ratio of NO$_x$ to VOC emissions in East Asia

1995

2030(CLE)
Emissions of ammonia in East Asia

(Mt NH₃)
Impacts of Human Activities are Felt on Scales from Local to Global
Energy Growth in Asia & Its Links to Emissions

- **Bad News:** Growth will have wide-ranging and increasing impacts on the environment at all scales.
- Projections present a very pessimistic picture.
- **Better News:** Projections and present practices will not be followed.
- There are ways to decouple energy growth from economic and population growth.
- Key may be to recognize linkages between urban pollution and climate change.
The Future: A Close Integration of Measurements and Models

Data:
- Chemical Composition of the atmosphere
- Aerosol composition, size and number
- Optical properties/radiances
- Meteorological fields
- Combined met/chem quantities: fluxes, tendencies, sensitivities

Models:
- Coupled Chemistry
- Aerosol
- Meteorological Radiation
- Regional/Global

Measurements:
- Surface
- Satellites
- Lidar
- Sondes
- Mobile platforms - planes, ships, vans
- Laboratory

INTEGRATION
ASSIMILATION
INTERPRETATION

Optimal Analysis State to Support:
- Interpretation of data
- Air quality forecasting
- Mission/experiment planning
- Identification of science deficiencies
- Monitoring Network Design
- Scenario studies
- Feedback evaluation
- Design of improved retrieval algorithms

Utilities/tools for data mining and analysis for use by a broad community
Acknowledgements

• **Funding From:**
  - NASA
  - NSF
  - DOE/ACP
  - The World Bank & ADB
  - CRIEPI/Japan
  - World Meteorological Organization
Thank You !!!

This picture, adapted from Katsushika Hokusai’s masterpiece “The Great Wave off Kanagawa,” artistically displays the spirit of supercomputing. Complex phenomena, such as waves on the surface of a fluid, are modeled by covering space with a grid and then solving the laws of physics at discrete points on that grid. The finer the grid, the closer the numerical simulation is to the actual solutions of the mathematical laws of physics.
Impacts Lead To Action

Xiangtan, China - received over 2 Million Yuan compensation from the neighboring cities due to yellowing of paddy fields in 1999.

Decreased Crop Yields

New Two-Zone Control Policy

China Emissions are changing in unanticipated ways!
Asia presently has ~1 billion urban dwellers, projected to grow to ~3 billion in 2025.

10 Asian Megacities will account for ~40% of GNP in 2025.

Each 1 million urban inhabitants emit average of 25,000 tons of CO₂ every day (six times global per capita average).

Indoor and Outdoor air pollution pose severe human health concerns.

4%/yr urban growth rate in Asia-Pacific region.

23 megacities in 1995 (17 in developing countries) to 36 in 2025 (23 in Asia)