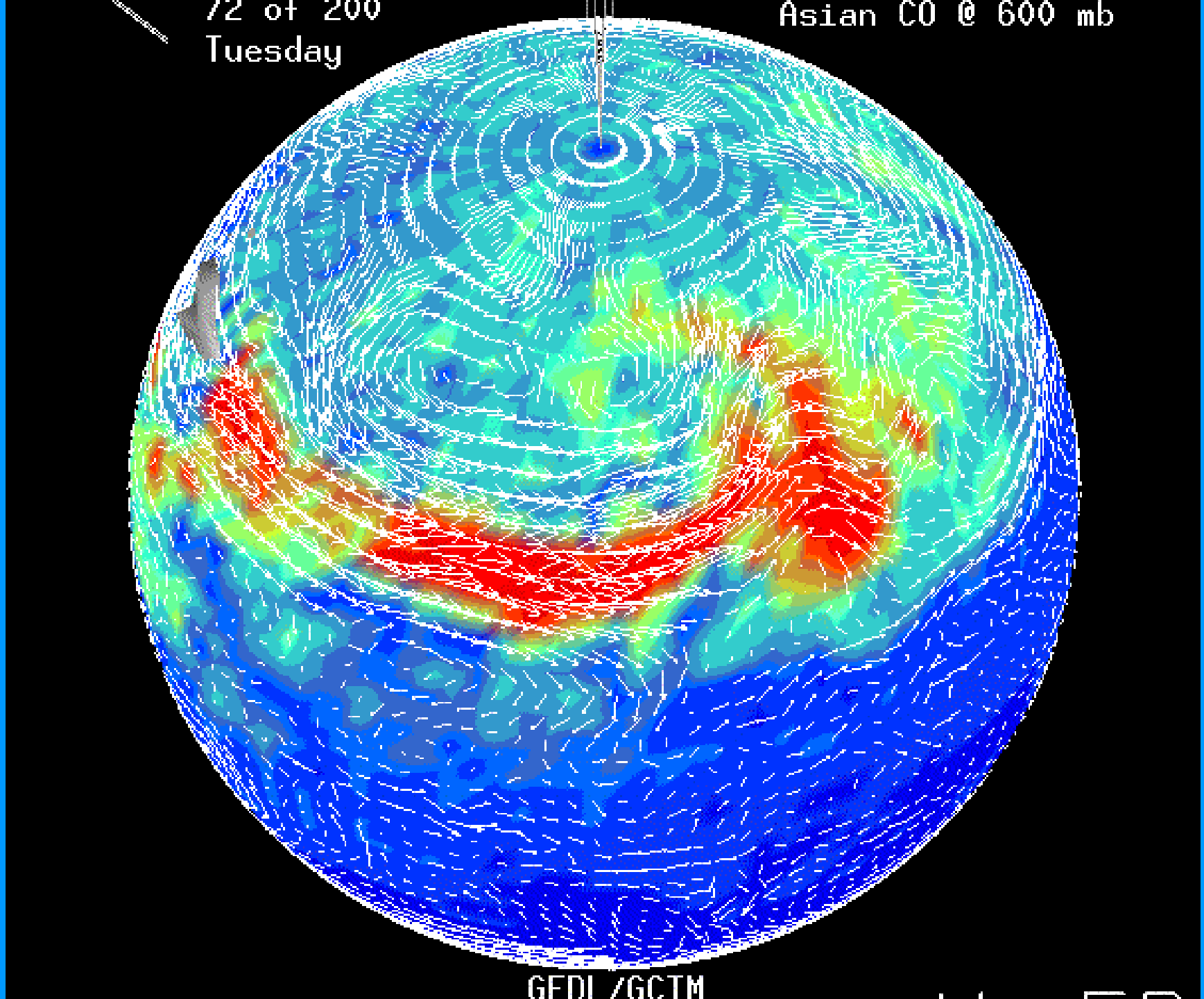
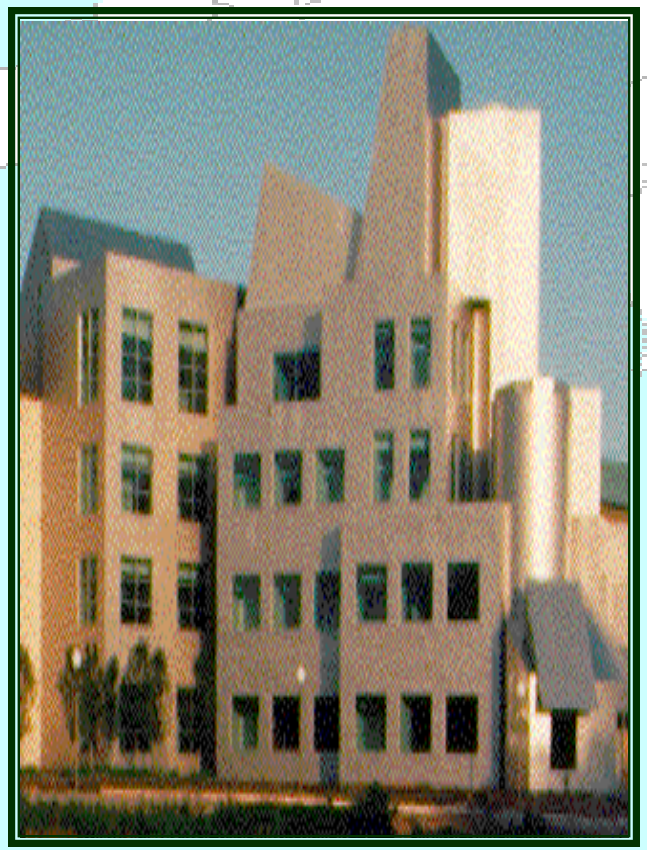

72 of 200
Tuesday
Asian CO @ 600 mb
GFDL/GCTM

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



Gregory R. Carmichael

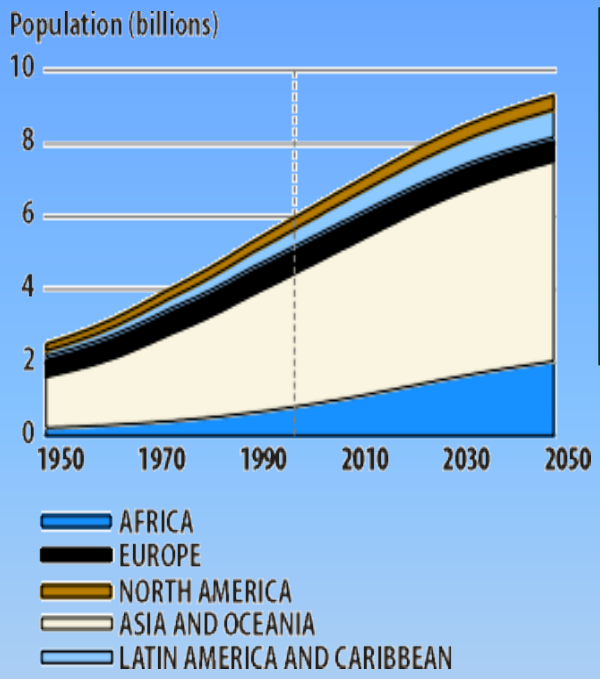
Department of Chemical & Biochemical Engineering

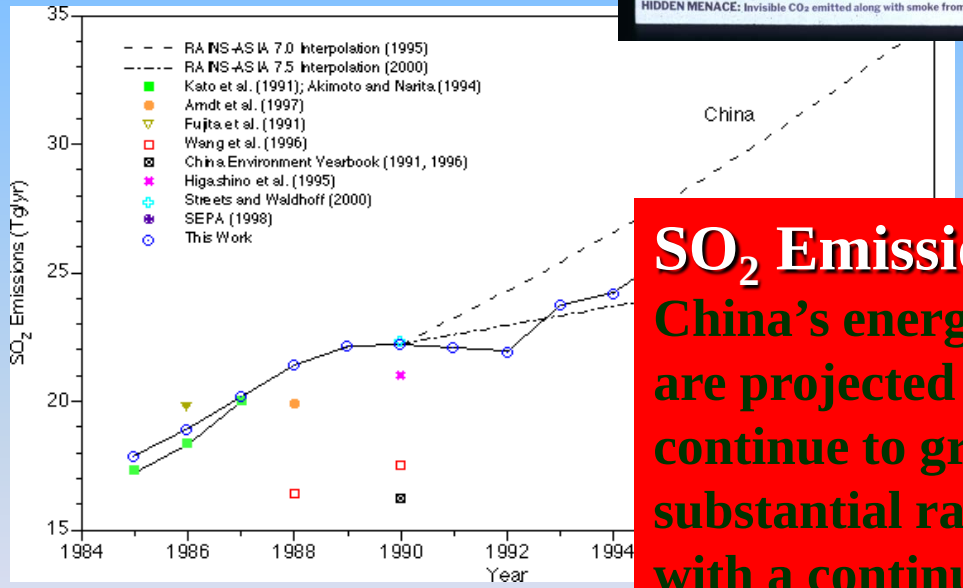
Center for Global & Regional Environmental Research and the

University of Iowa

CIT Seminar Jan. 2001

Air Quality Engineering: A Focus on Asia





SO_2 Emissions -- 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

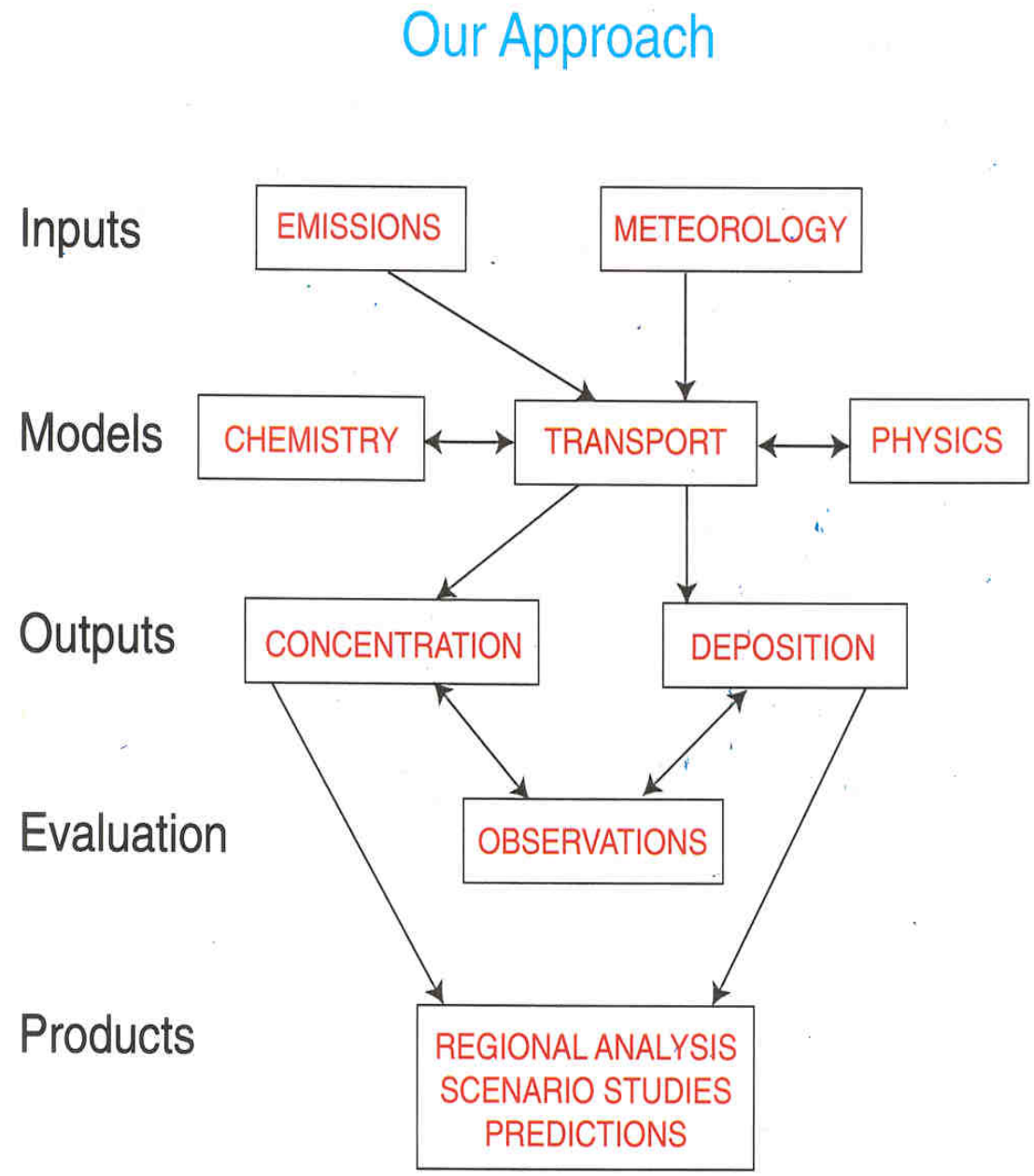
Local to Global Impacts

Motivating Questions

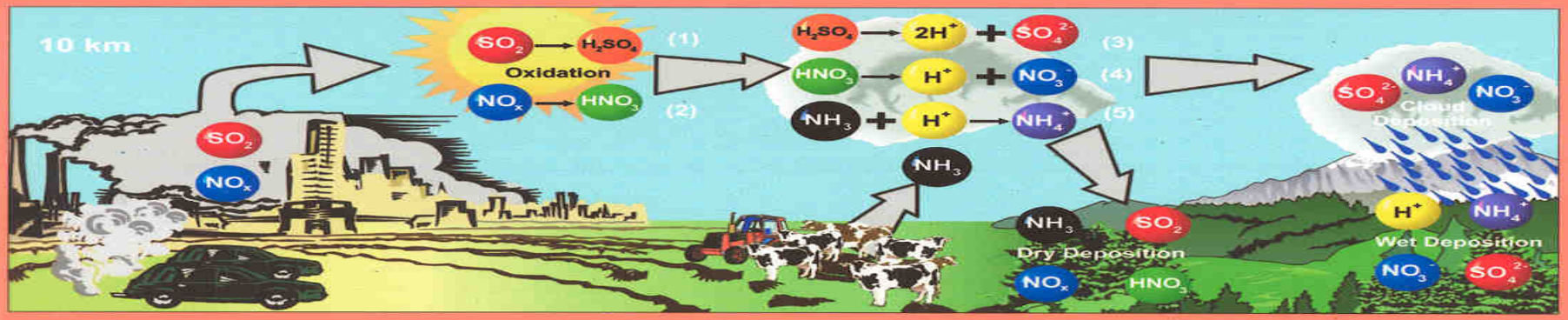
- What are the current and projected trends in energy and emissions?
- How are these impacting human health? Food security?, etc.
- What are the transboundary impacts of Asian emissions within Asia ?
- What are the region-specific science issues that need to be addressed (e.g., aerosol interactions)?
- 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:

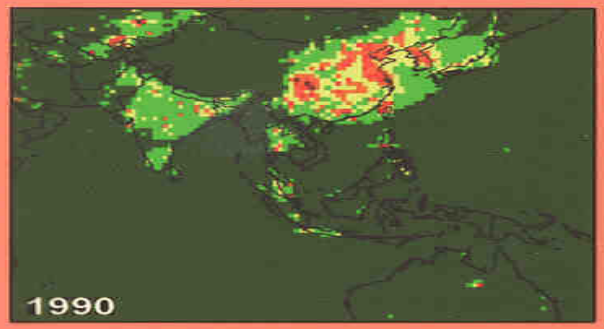
- Global (GFDL, Chip Levy)
- Regional (STEM)
- Integrated Assessment (RAINS-Asia)

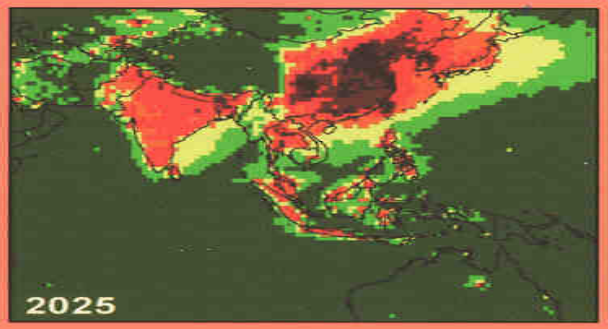


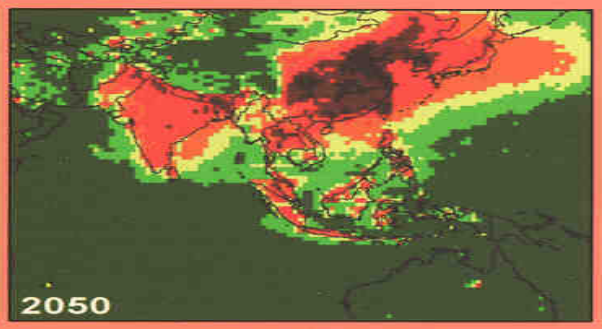
Transfer of pollutants in the atmosphere

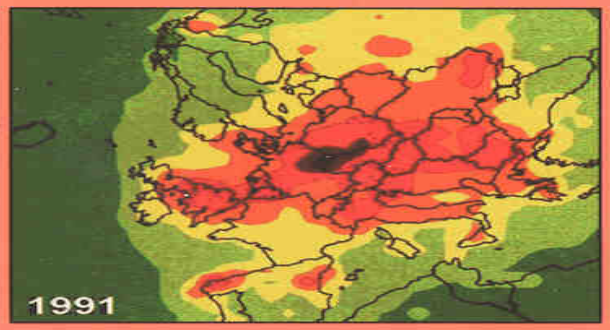


- Pollution may be transported over long distances
- Sulphur deposition calculated by MATCH model using 'Conventional Development Scenario' emissions









$mg.S.m^{-2}.yr^{-1}$

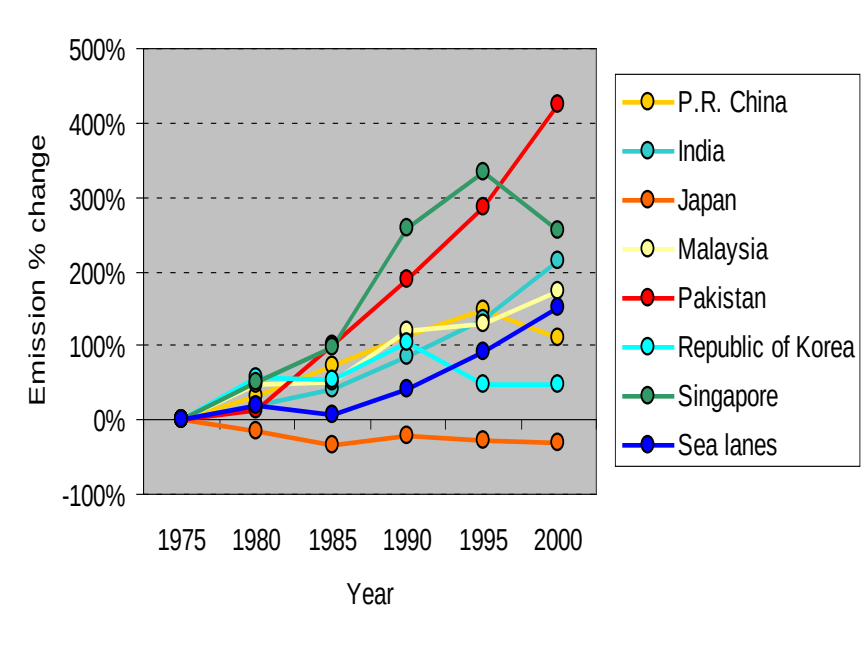
- 0 - 199
- 200 - 499
- 500 - 999
- 1000 - 1999
- 2000 - 4999
- 5000 - 9999
- 10000+

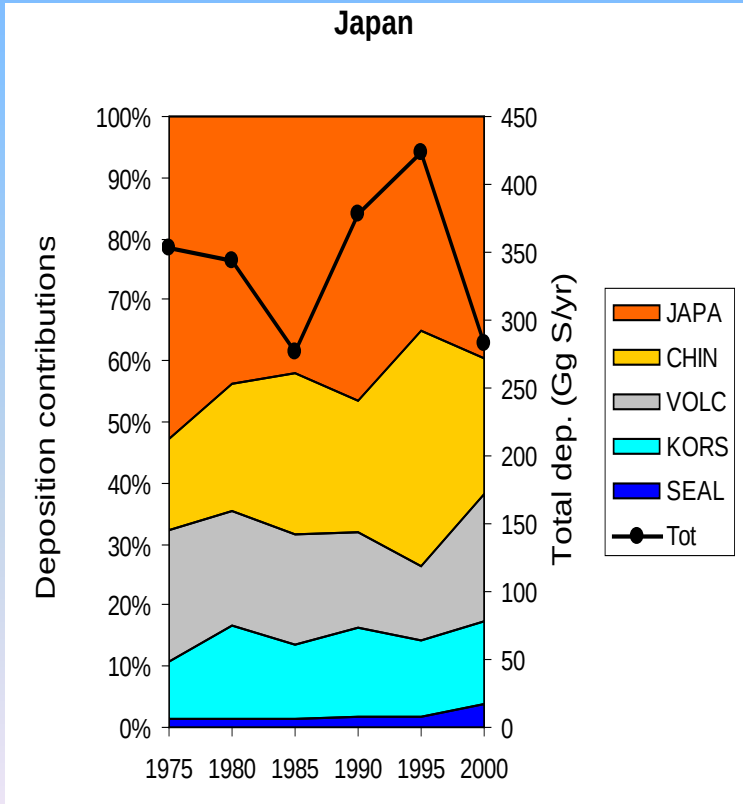
SEI STOCKHOLM ENVIRONMENT INSTITUTE

Atmospheric Environment Issues in Developing Countries

Sida

Transboundary Pollution Issues are of Growing Importance

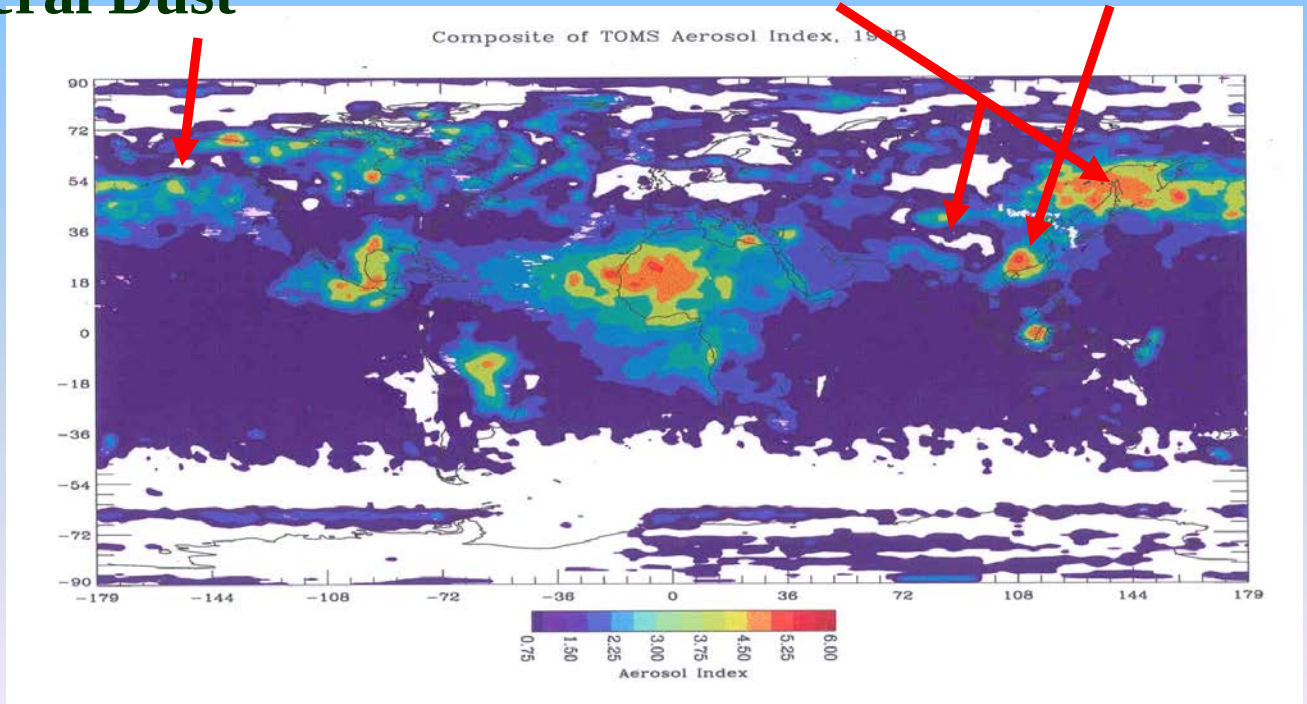




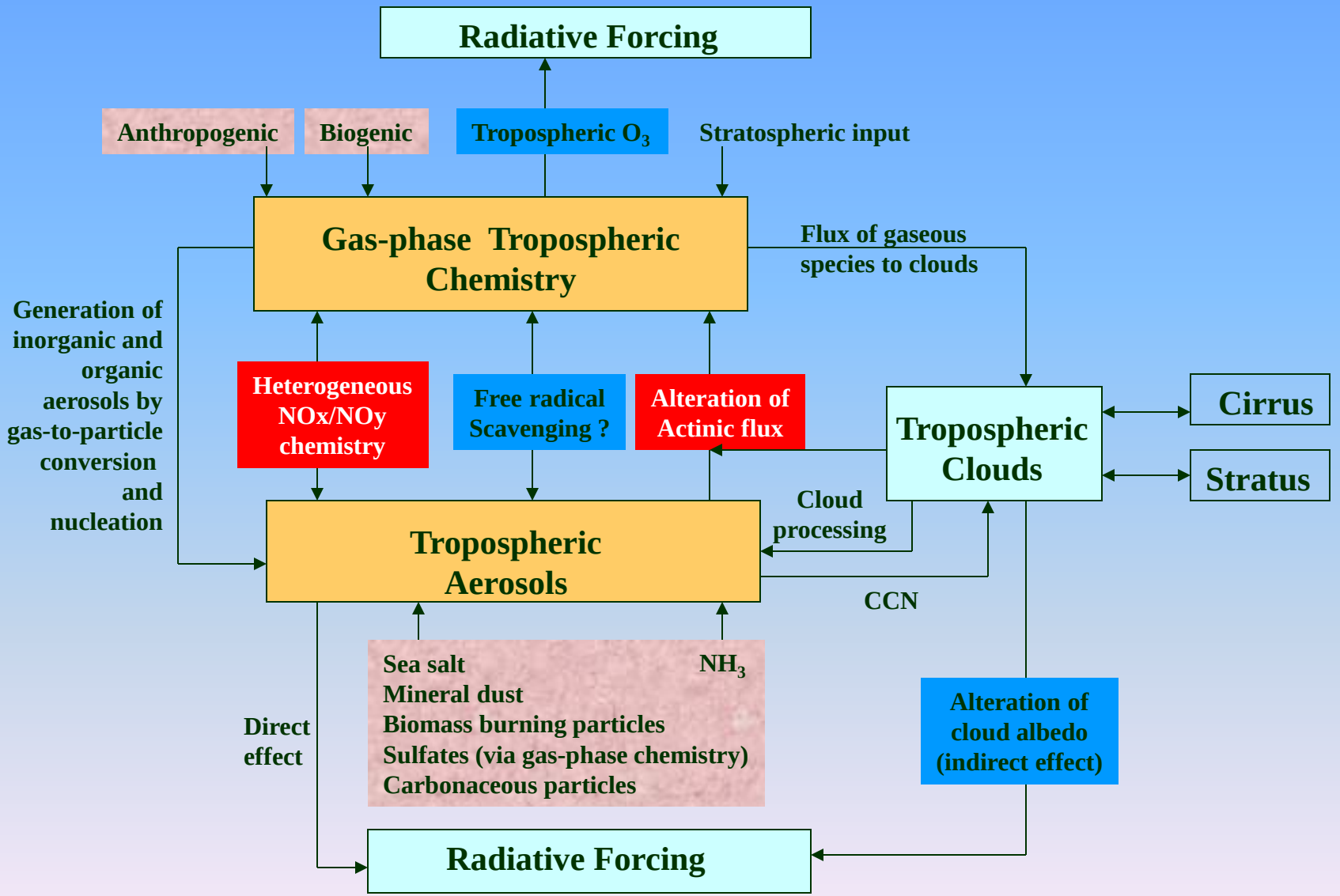
Aerosols are Pervasive Throughout Asian Environments

Mineral Dust

BC from Pollution & Fires



A Key Science Issue: Chemistry/Aerosol/Regional Climate Coupling



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.

THE APPROACH

MODELING ↔ LABORATORY

FIELD STUDY

(Vicki Grassian –leader)

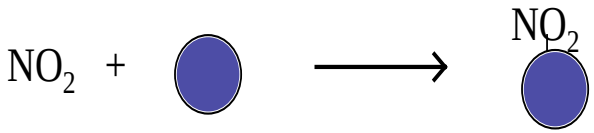
- 3D STEM-III transport/chemistry/deposition model
- Dynamics of Aerosol Processes
- Combined Kinetic/Thermodynamic Approach
- New Approach for Secondary Organic Aerosol Partitioning
- Sensitivity Analysis and Improved Numerical Methods

- Spectroscopic Measurements
- Detailed Reaction Mechanisms
- Kinetic Measurements
- Surface Coverages
- Adsorption Isotherms

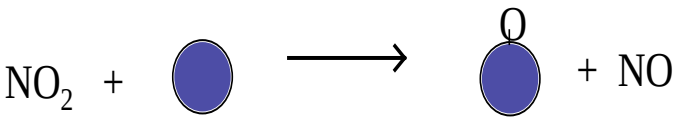
Chemical Role of Aerosol Particles in the Atmosphere

Can change the chemical balance of the atmosphere in two ways

Sink

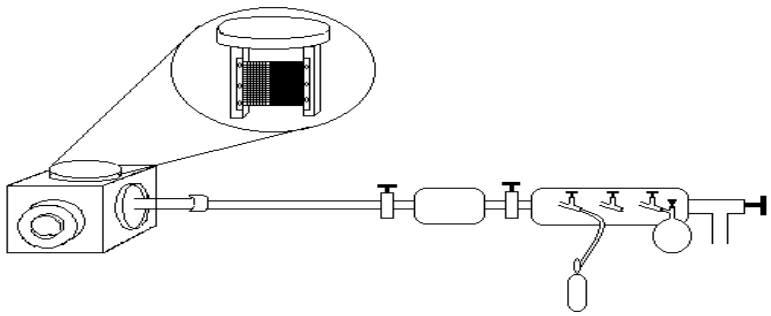


Reactive Surface



$H(a) + NO_2 \longrightarrow HONO$

Experimental Considerations

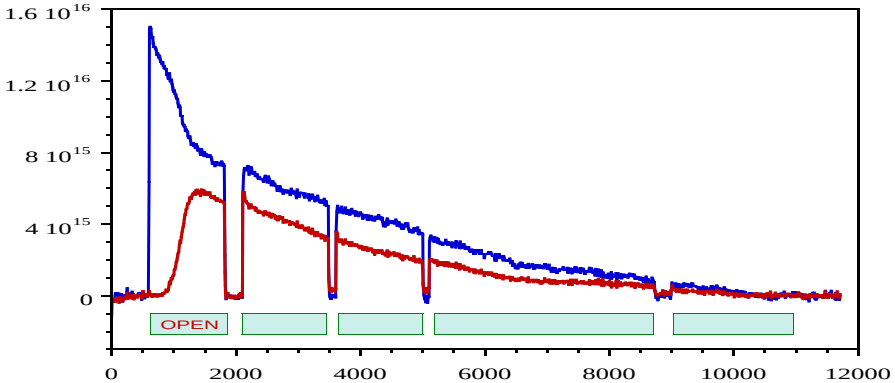


- **Spectroscopic measurements** to provide both qualitative (what reactions are possible) and quantitative information
  - Provide mechanistic information on the molecular level
  - Need to have techniques that can detect gas-phase **and** surface-bound species

Transmission FT-IR Spectroscopy
Diffuse Reflectance UV-vis Spectroscopy
Mass Spectrometry

- **Kinetic measurements** to provide quantitative information
  - Determine uptake coefficients (sticking coefficients, reaction probabilities) γ

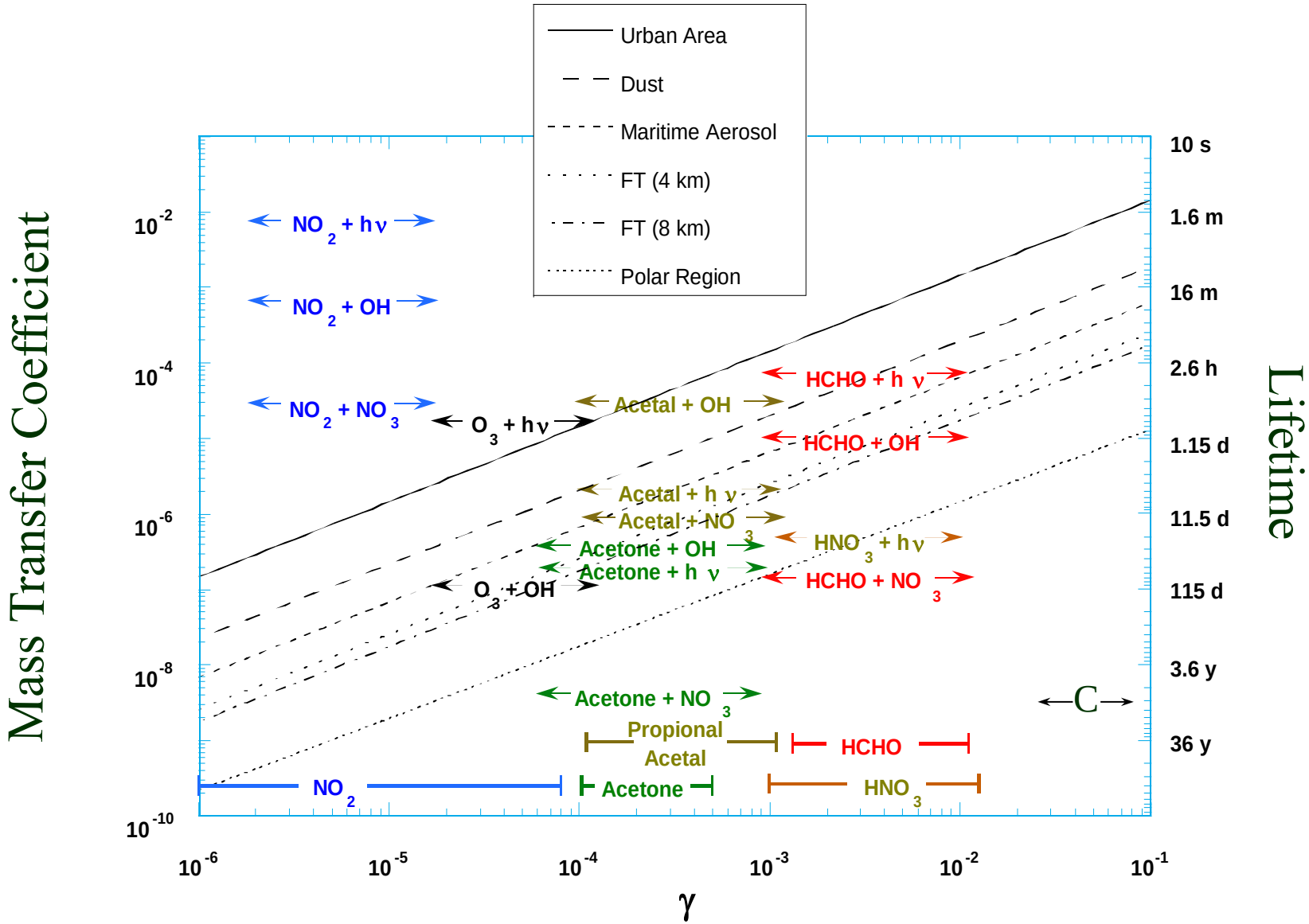
Knudsen cell apparatus



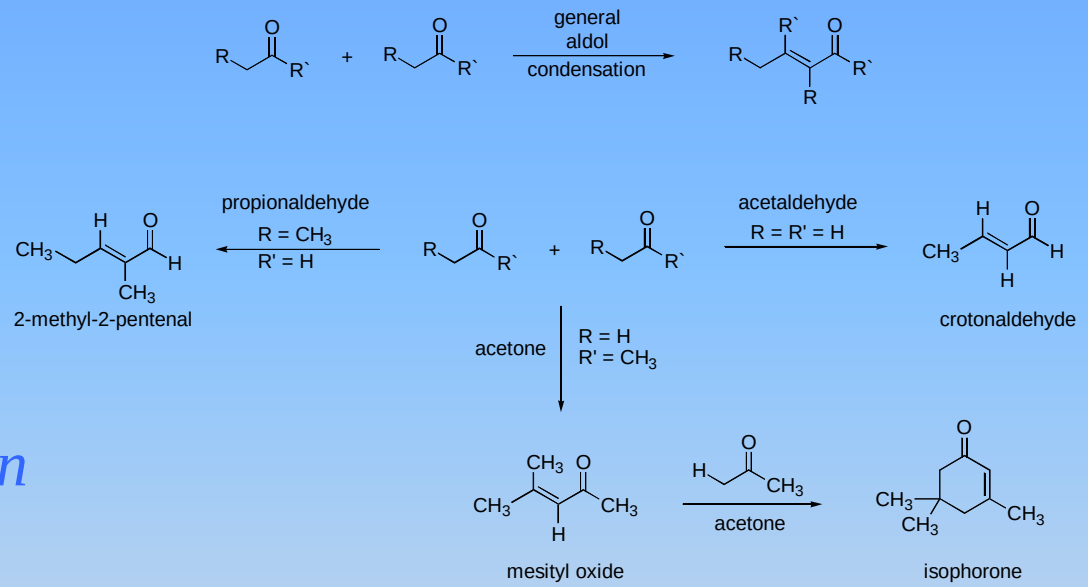
- **Provide data as input for global atmospheric models** - removal rate of gas-phase species j

$$k_j = \int_{r_1}^{r_2} k_{d,j}(r)\, n(r)\, dr \qquad k_{d,j} = \frac{4\pi r^2 D_j V}{1 + K_n(\lambda + 4(1-\gamma)/3\gamma)}$$

n(r)dr = number density of particles between r and r+dr


Urban Area
Dust
Maritime Aerosol
FT (4 km)
FT (8 km)
Polar Region
Mass Transfer Coefficient
Lifetime
γ
10 s
1.6 m
16 m
2.6 h
1.15 d
11.5 d
115 d
3.6 y
36 y
NO2 + hν
NO2 + OH
NO2 + NO3
O3 + hν
Acetal + OH
HCHO + hν
HCHO + OH
Acetal + hν
Acetal + NO3
Acetone + OH
Acetone + hν
HNO3 + hν
HCHO + NO3
O3 + OH
Acetone + NO3
Propional
Acetal
HCHO
NO2
Acetone
HNO3
C

Scheme for Aldol Condensation Reaction on Oxide Particles (Al_2O_3, Fe_2O_3, TiO_2, CaO, and MgO)



Li et al., JGR, in press, 2001

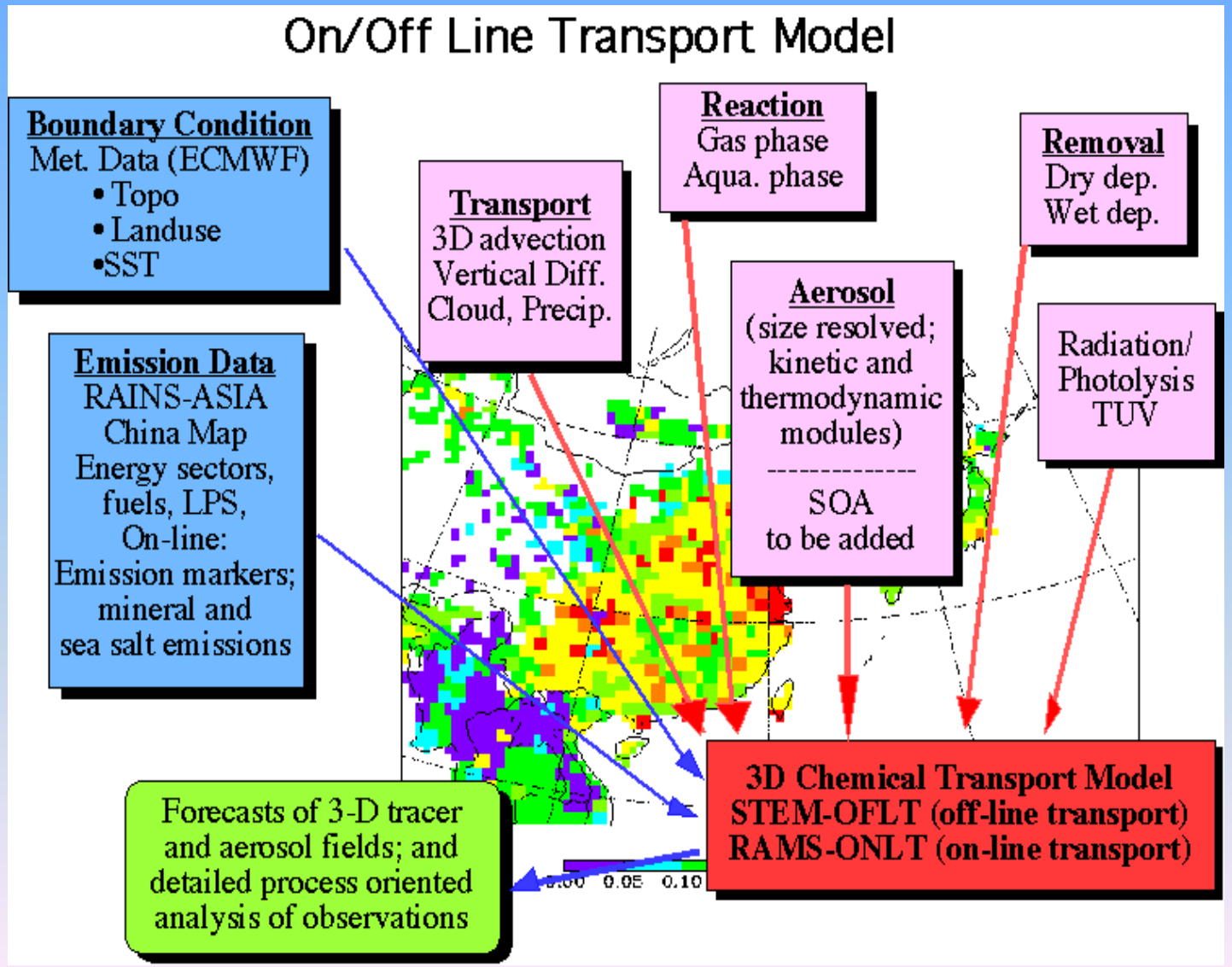
Saturated Carbonyl → **Higher Molecular Weight Unsaturated Carbonyl**

Impacts on RO2? On Organic aerosol mass?, others?

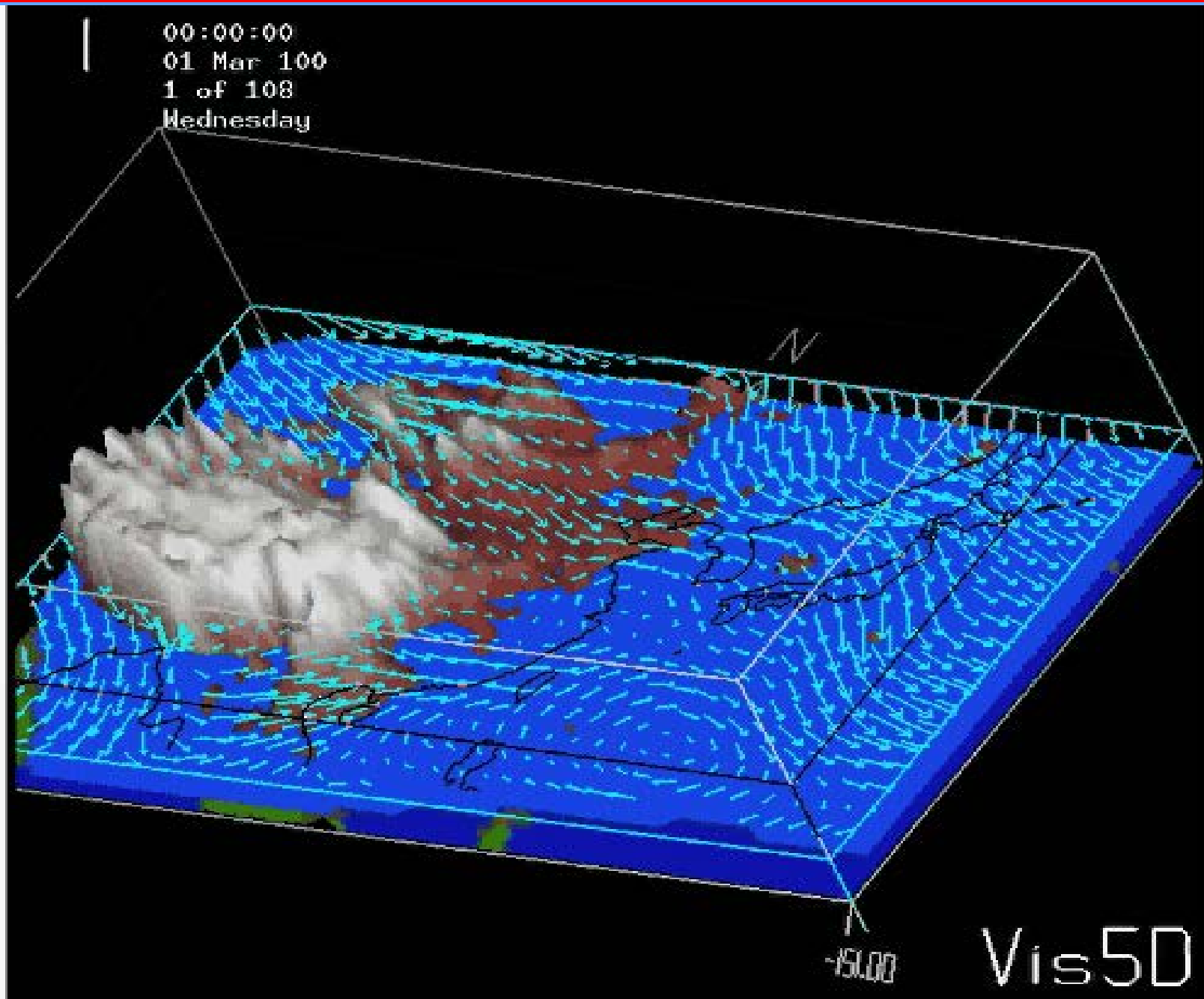
Summary of Combined Laboratory and Modeling Study

- 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)**

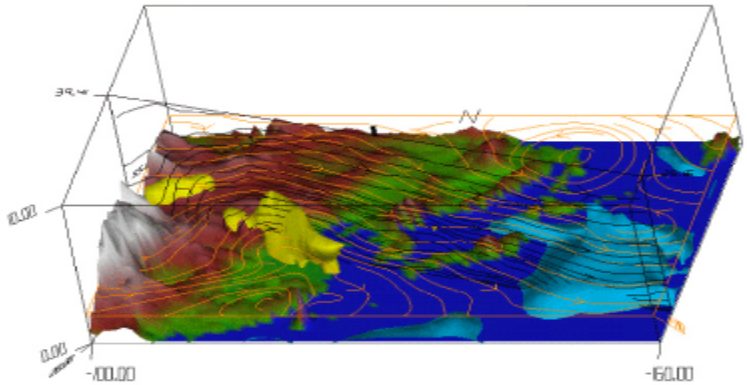
Three-Dimensional Combined Transport/Chemistry Analysis (STEM-III)

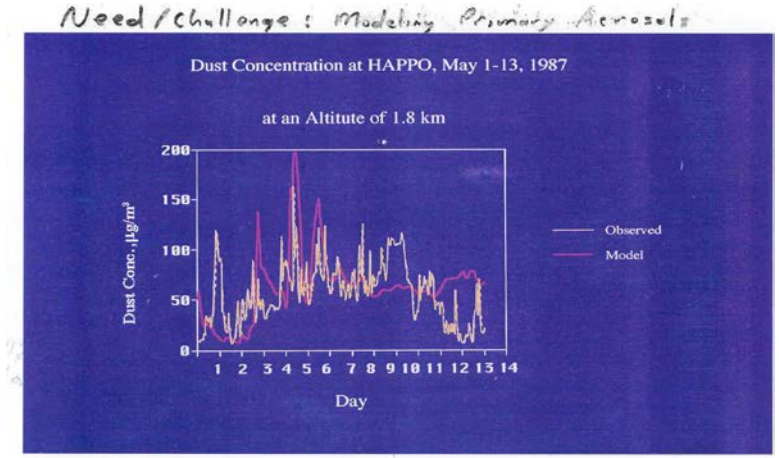


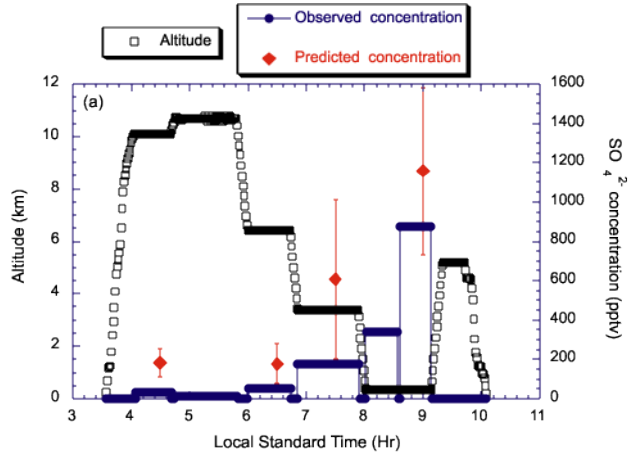
Dust, Sulfate, CO, Lightning NO_x during March-April 2000

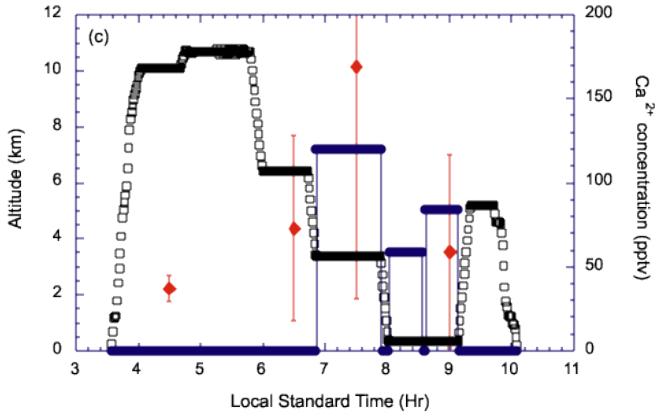


Model is Able to Capture Many Important Observed Features



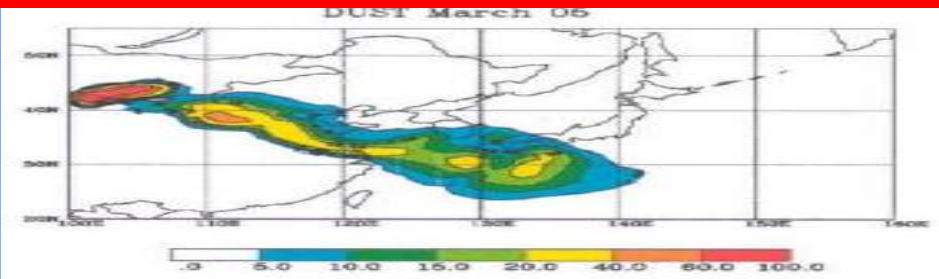


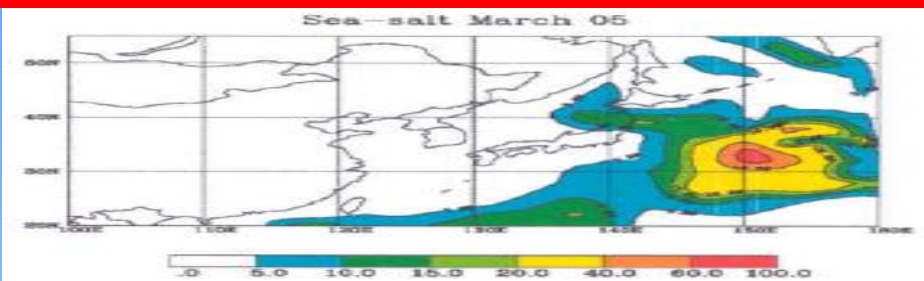


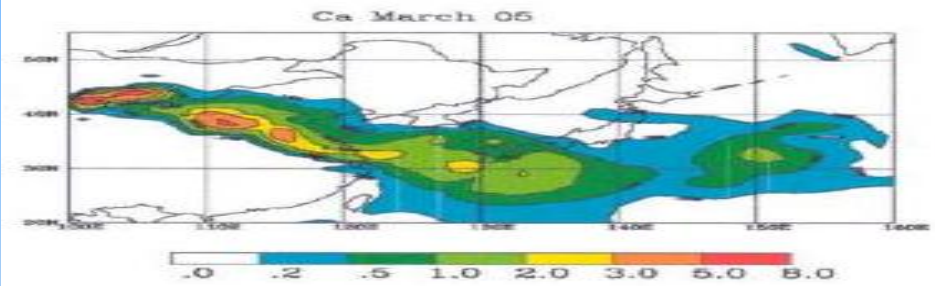


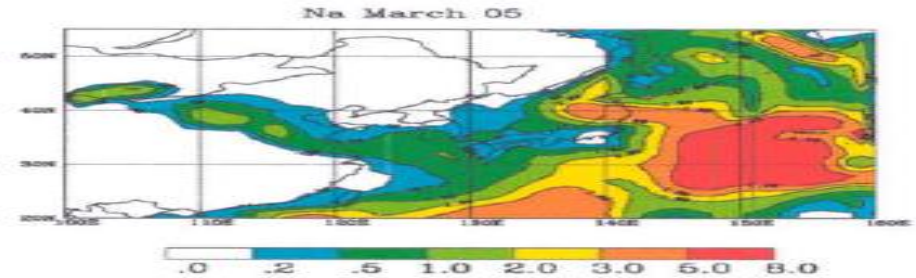
Phadnis et al., J. Atmos. Chem., 36:285 (2000); Song et al., JGR, in press (2001)

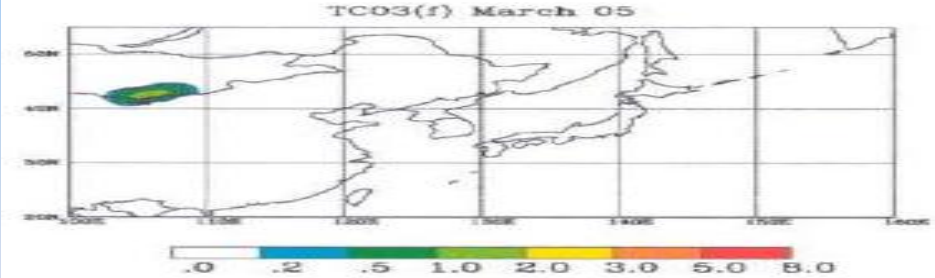
Calculated Fine and Coarse Mode Aerosol Distributions in the Boundary Layer; PEM-WEST B

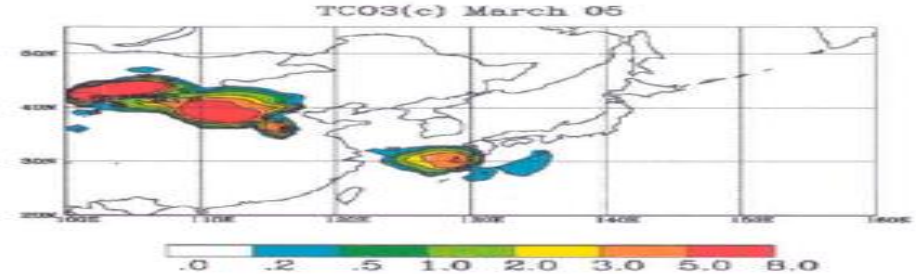


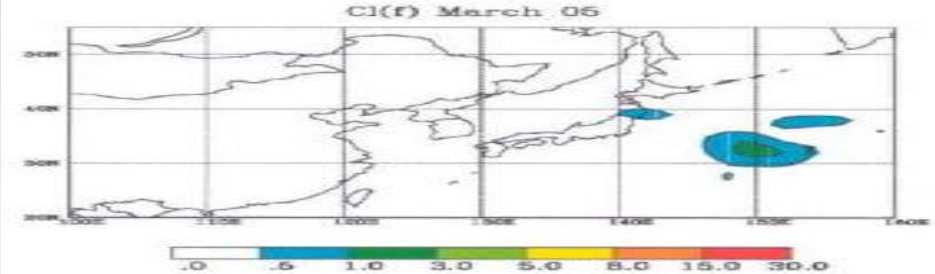


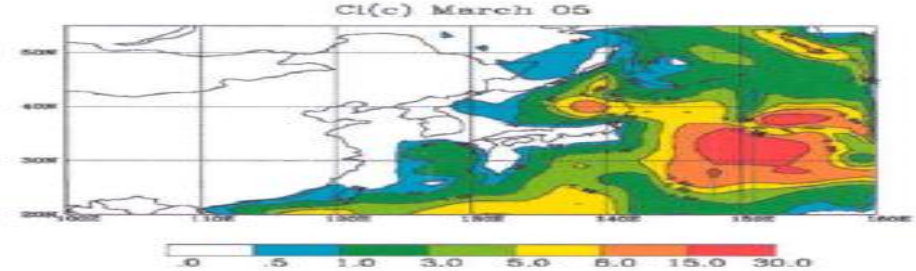




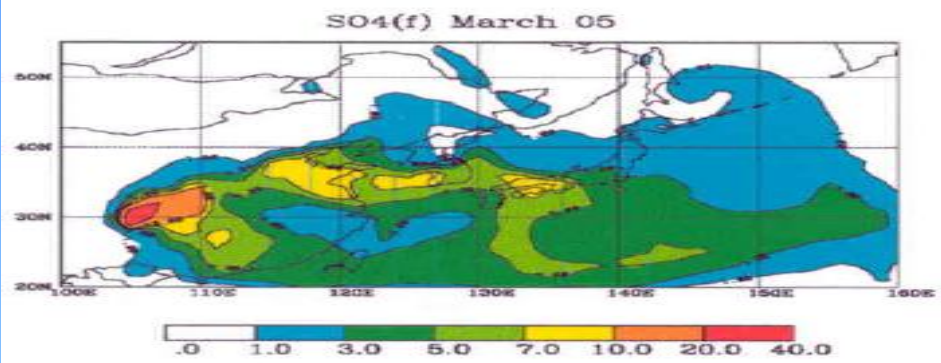


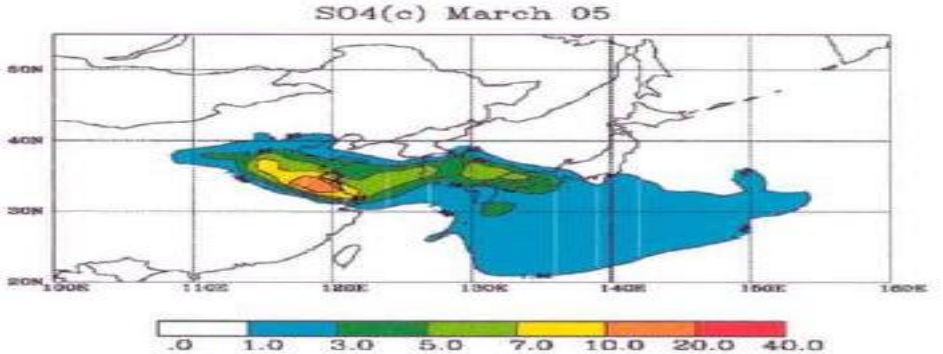


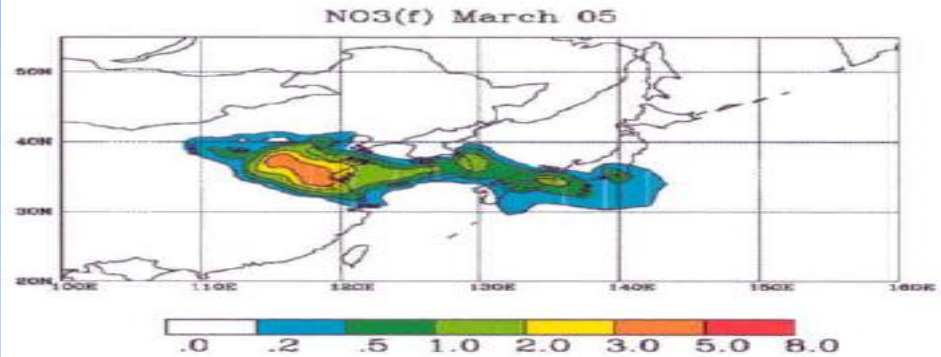


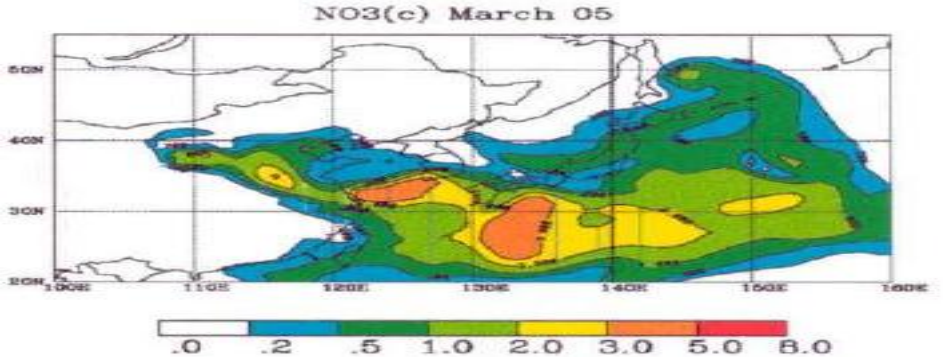


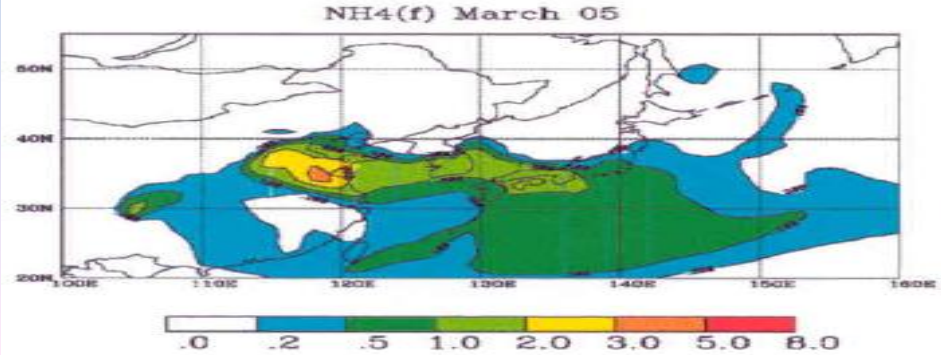
Calculated Fine and Coarse Mode Aerosol Distributions in the Boundary Layer; PEM-WEST B

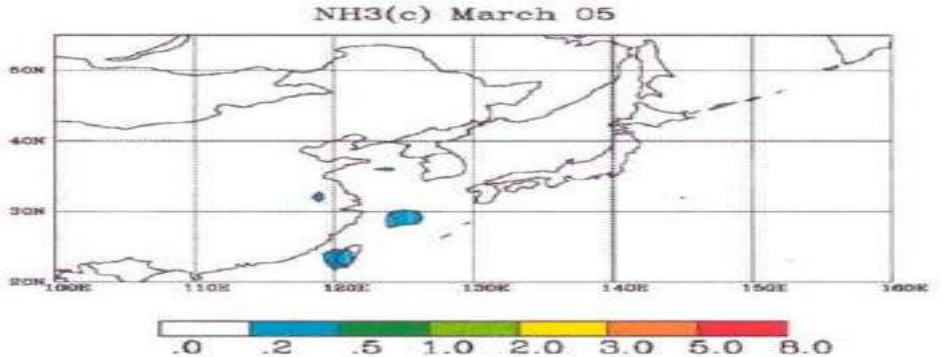




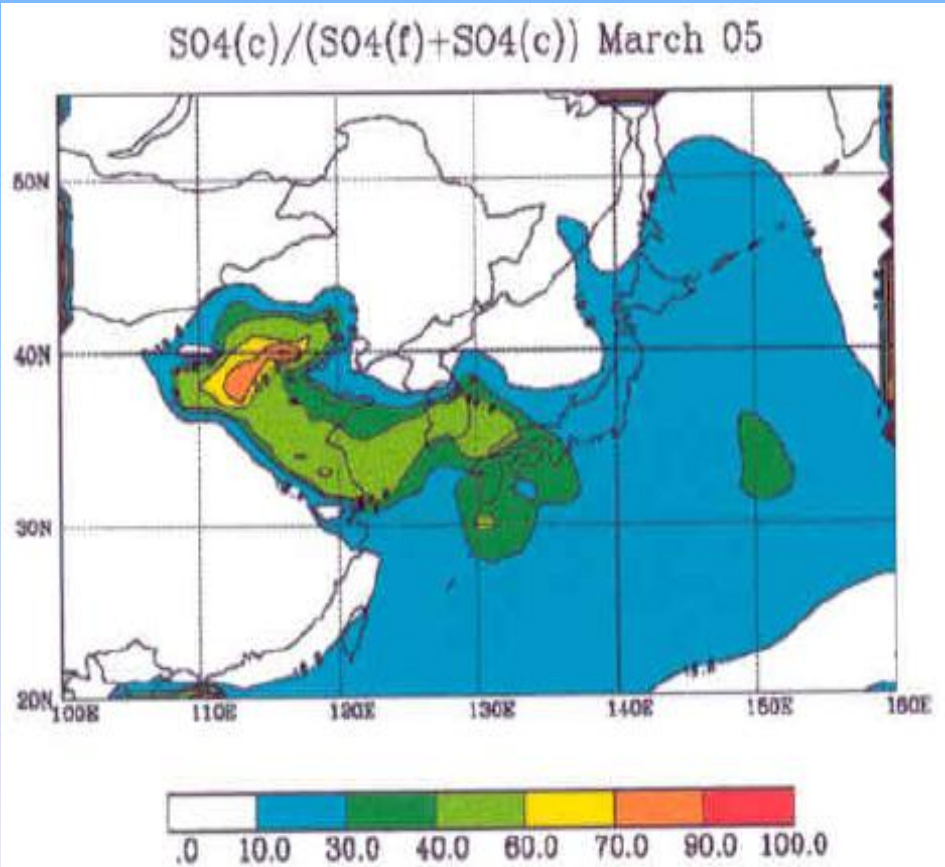


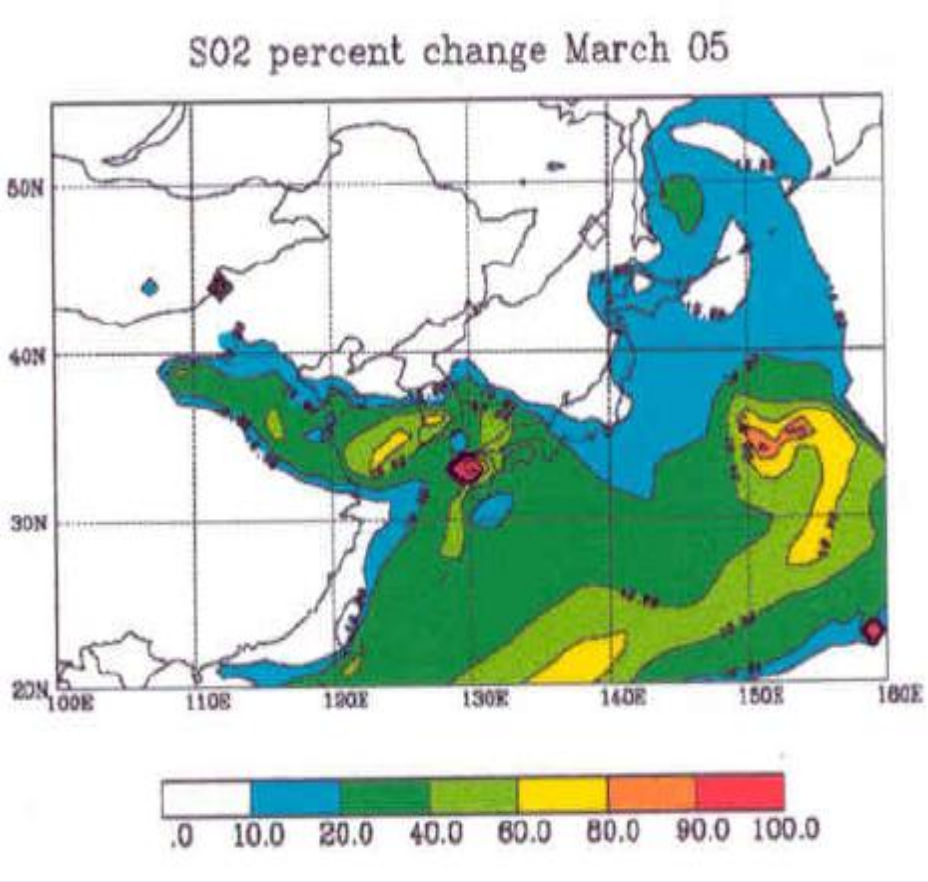






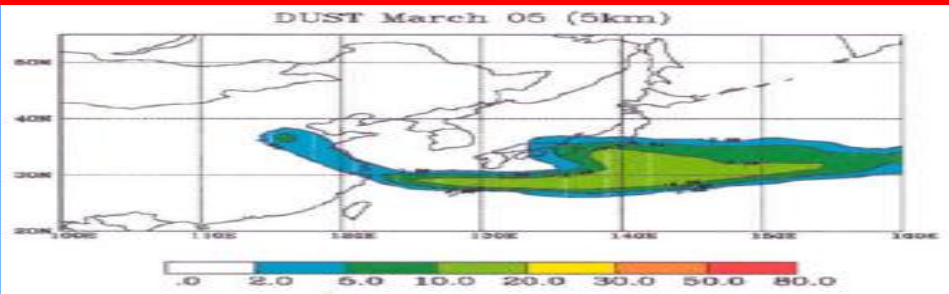
INTERACTIONS OF SO_2 WITH MINERAL AEROSOL CHANGE *SULFATE SIZE DISTRIBUTION* AS WELL AS THE CHEMICAL LIFETIMES OF SULFUR. THESE INTERACTIONS HAVE IMPLICATIONS FOR *RADIATIVE FORCING*

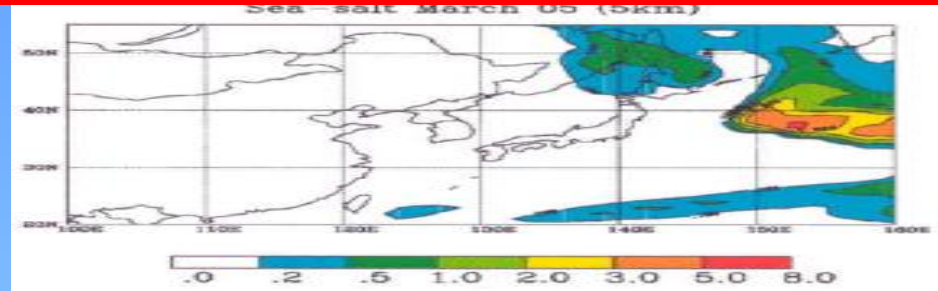


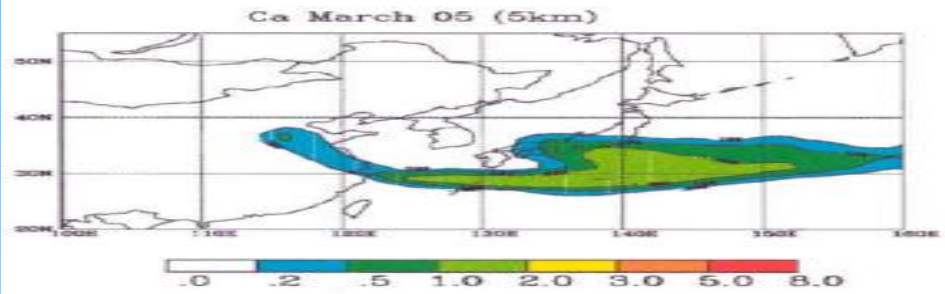


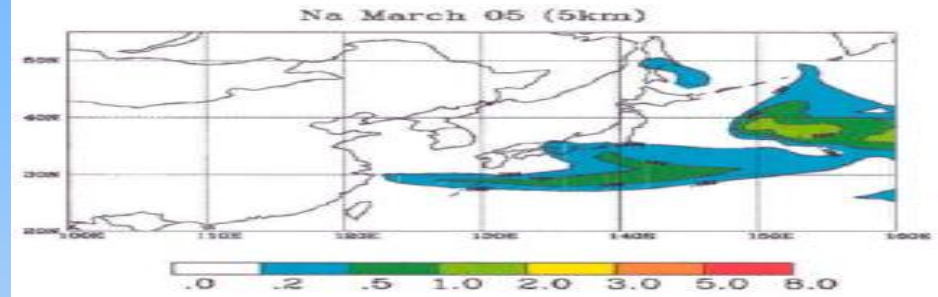
Song et al., JGR in press

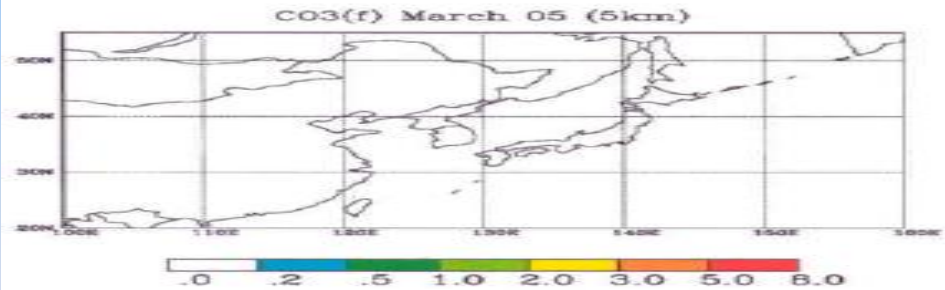
Calculated Fine and Coarse Mode Aerosol Distributions at 5 km; PEM-WEST B

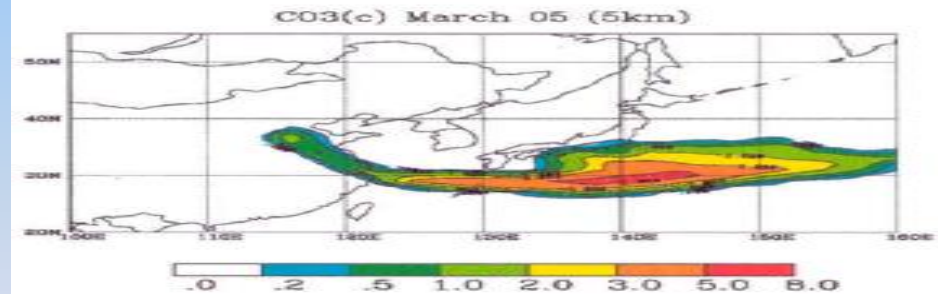


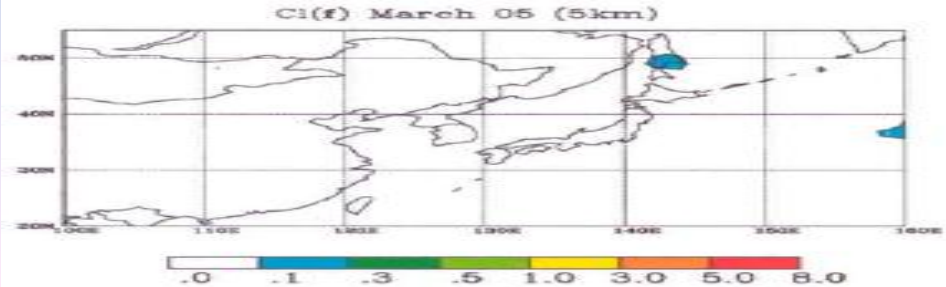


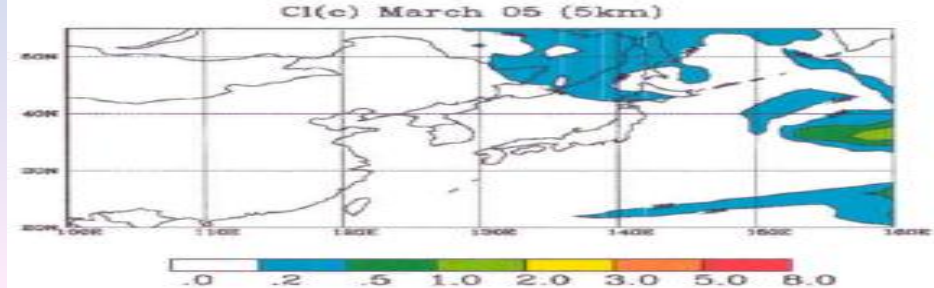




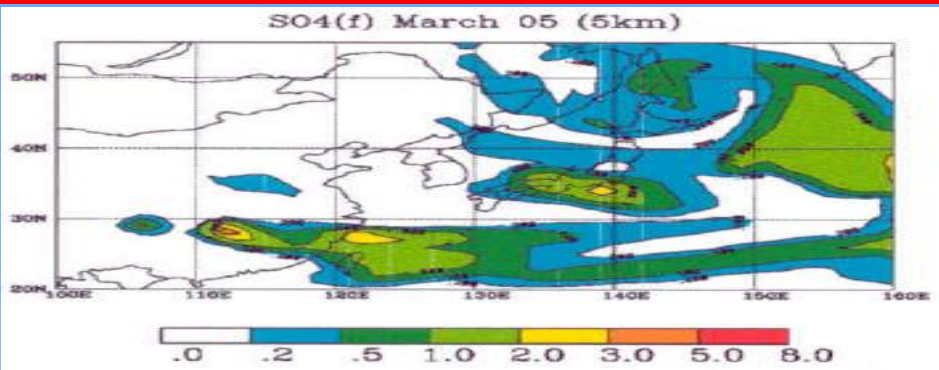


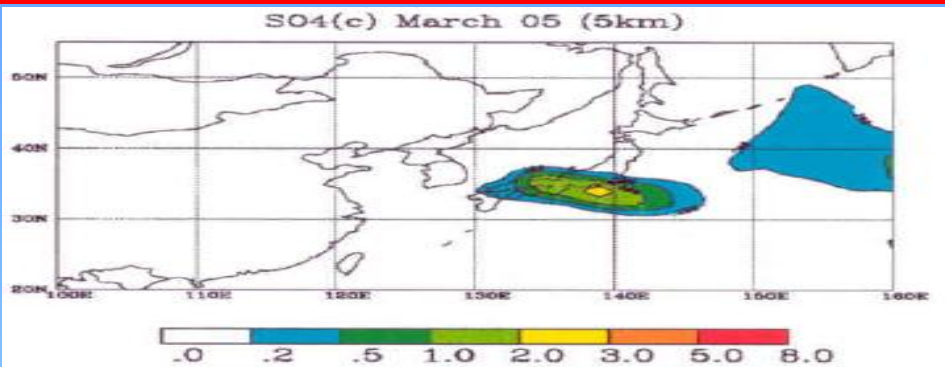


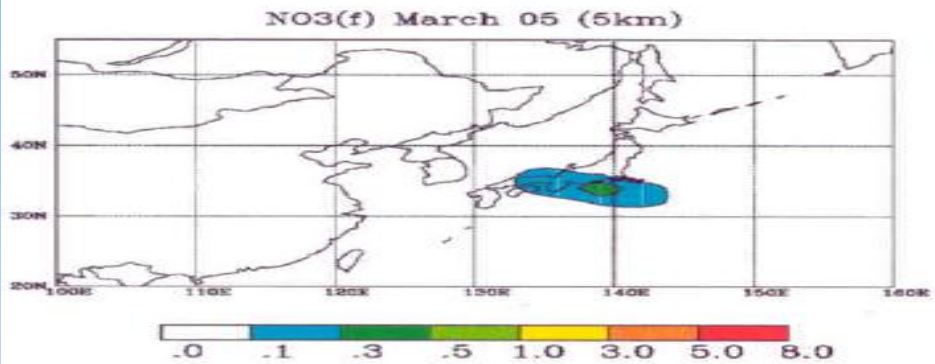


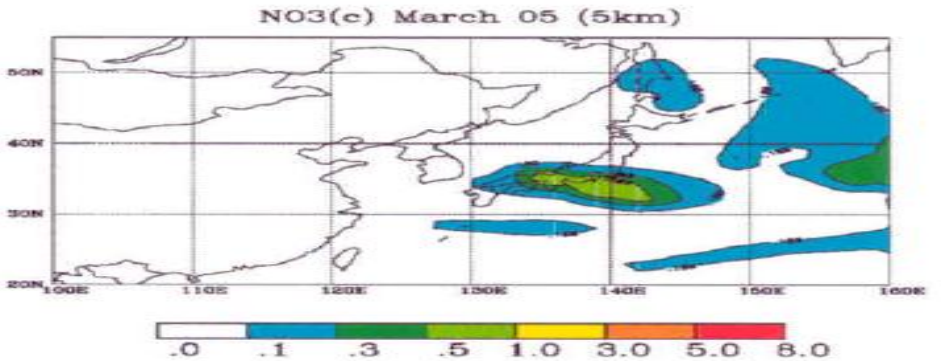


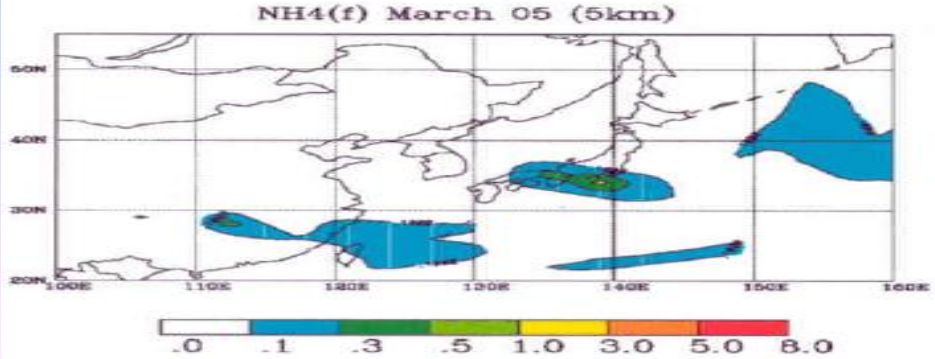
Calculated Fine and Coarse Mode Aerosol Distributions at 5 km; PEM-WEST B

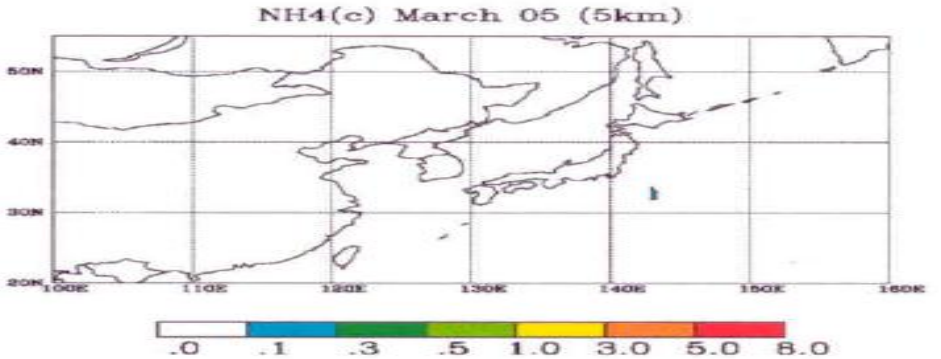




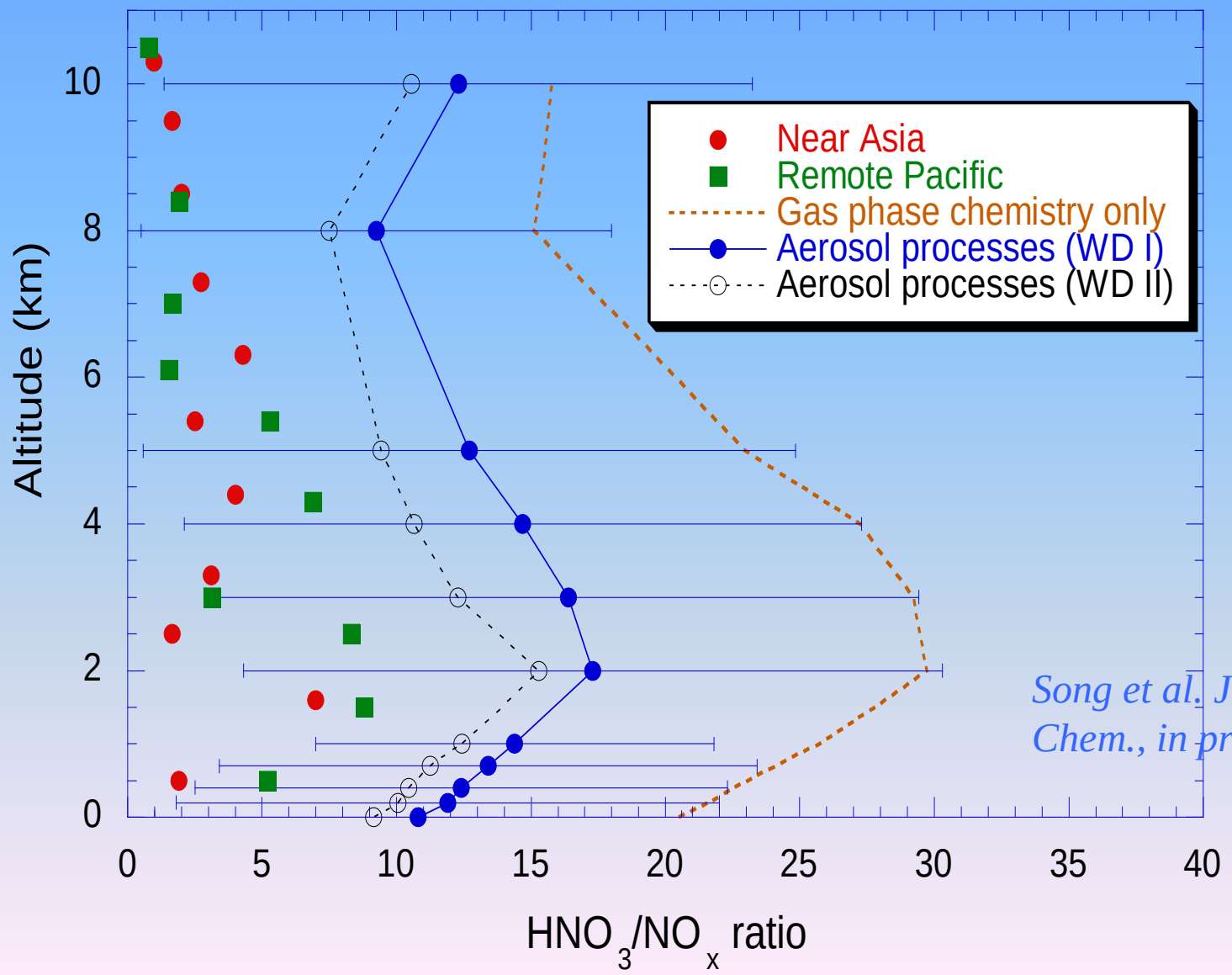






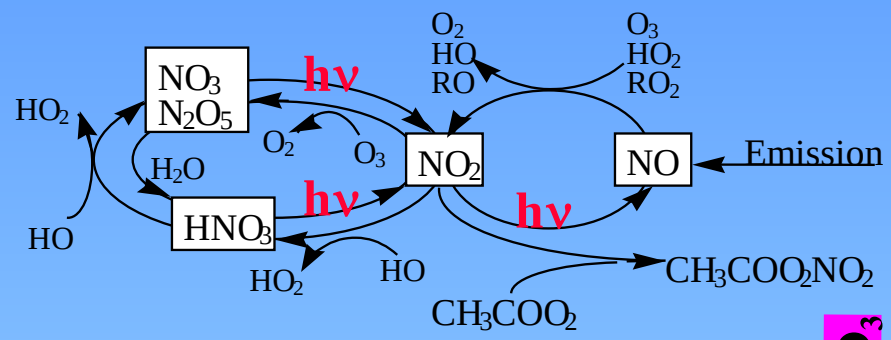


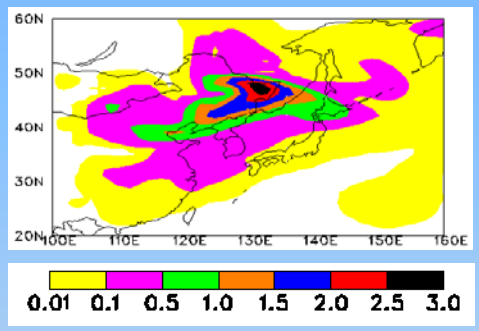
The Presence of Mineral Aerosol Alters the Partitioning of Nitric Acid



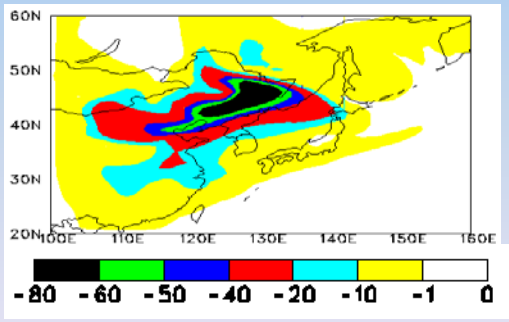
Song et al. J.Atmos. Chem., in press

Impact of Aerosols on the Photochemical Oxidant Cycle Through Photolysis Rates





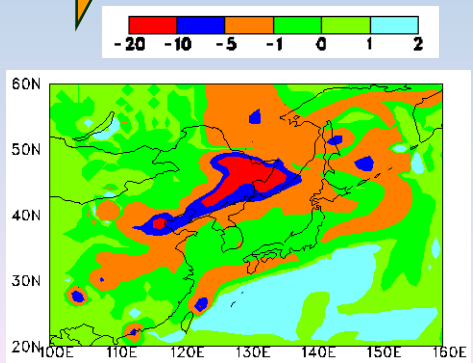
Mineral Optical Depth



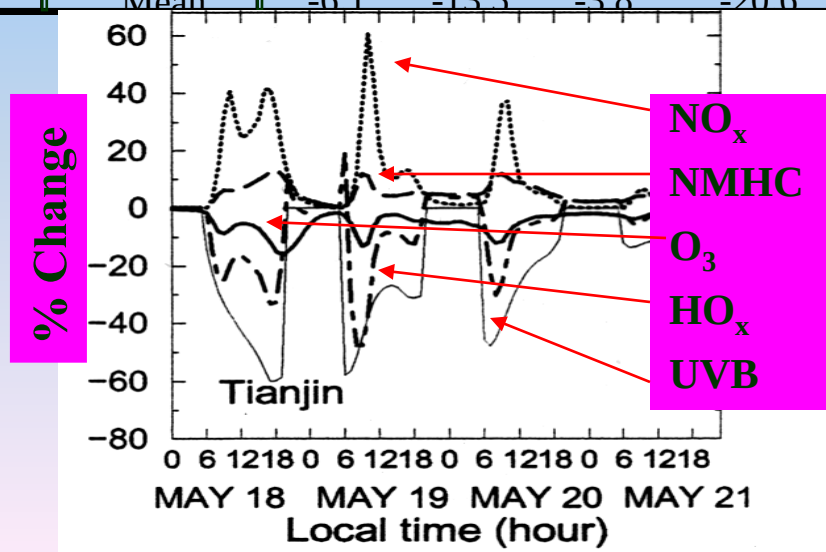
%Change NO_2 Photolysis

Dust Impact on O_3

Season	Aerosol impact on O_3	ΔSulfate %	ΔSoot %	ΔMineral %	ΔTSP %
Winter	+	\	\	\	\
	-	18.9	**48.6**	13.2	**58.7**
	Mean	-8.6	-25.8	-5.8	-33.0
Spring	+	**1.3**	\	\	\
	-	13.1	24.4	9.7	36.8
	Mean	-3.7	-11.1	-3.2	-16.8
Summer	+	**1.5**	\	\	\
	-	13.7	**25.4**	8.3	**37.6**
	Mean	-4.0	-12.9	-3.0	-19.1
Fall	+	\	\	\	\
	-	15.5	27.3	9.2	40.3
	Mean	-6.1	-13.5	-3.8	-20.6



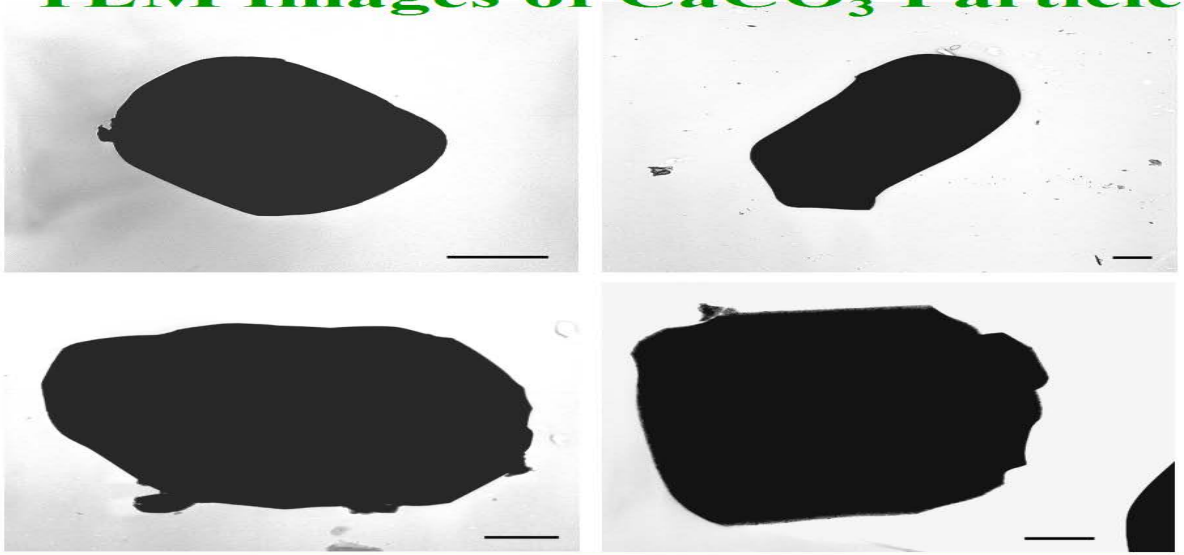
Ozone variations (%) due to dust



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

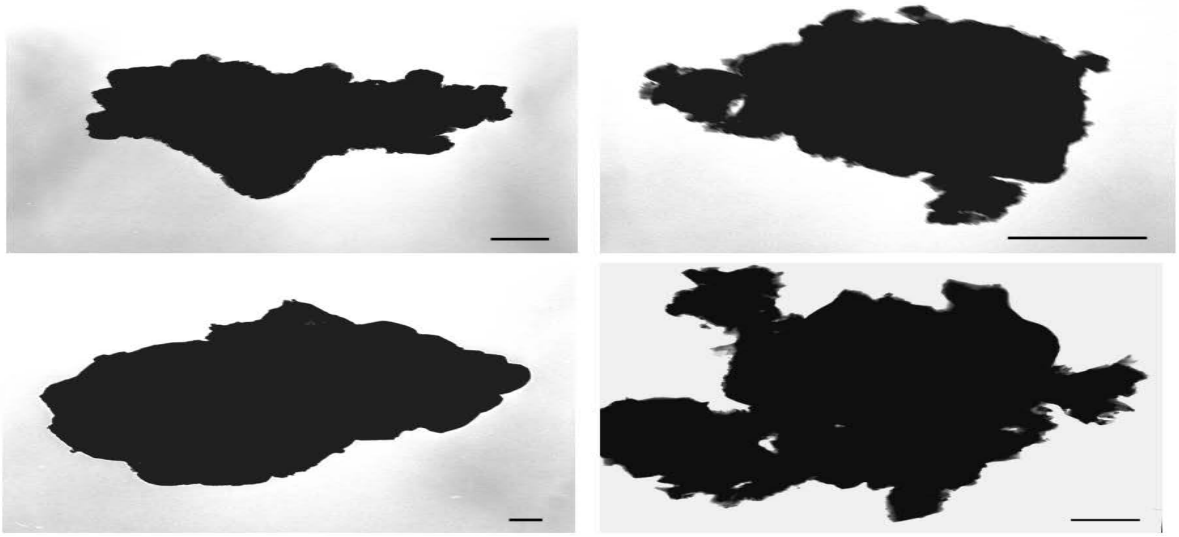
TEM Images of $CaCO_3$ Particles

$CaCO_3$



0.5 μ

HNO_3 Reacted $CaCO_3$

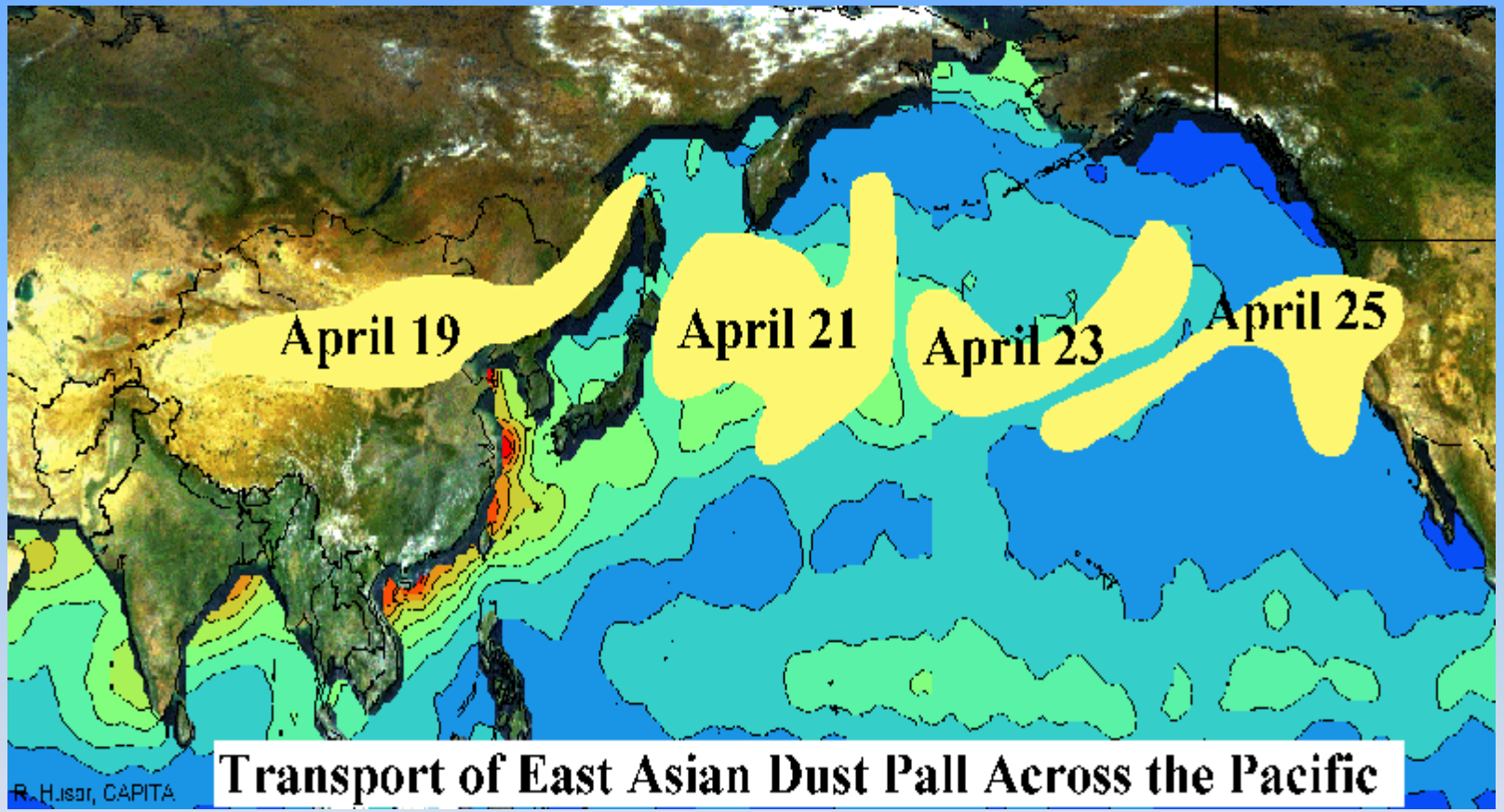


Goodman et al., JGR, 105:29053, 2000

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.

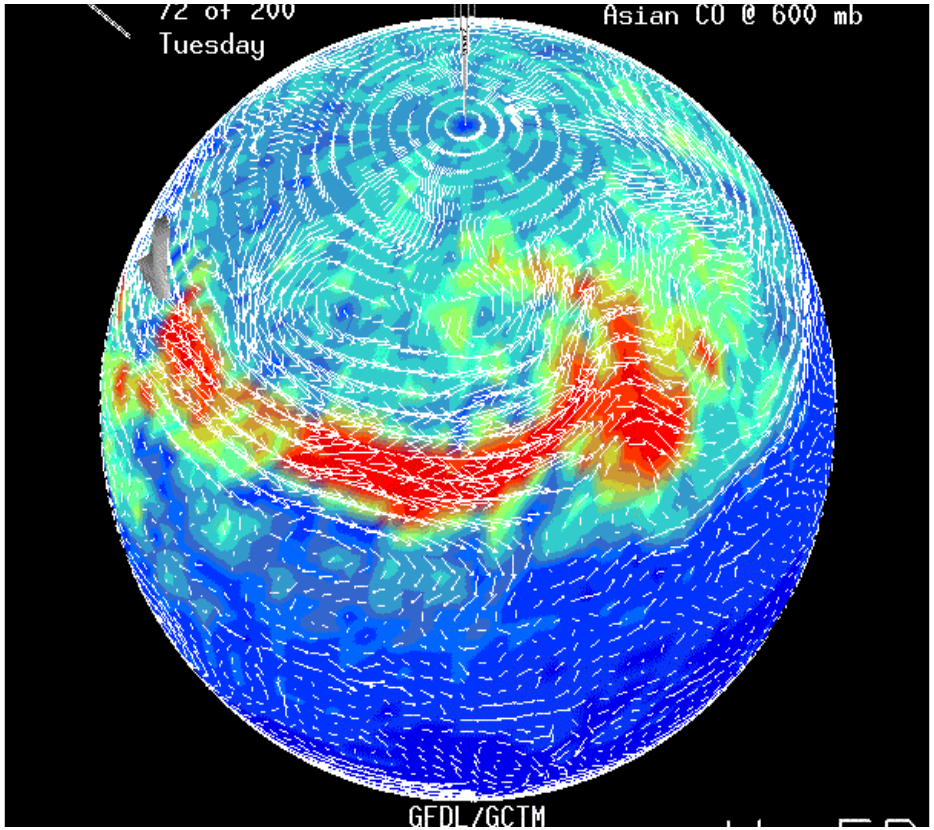
April 1998 Asian Dust Event

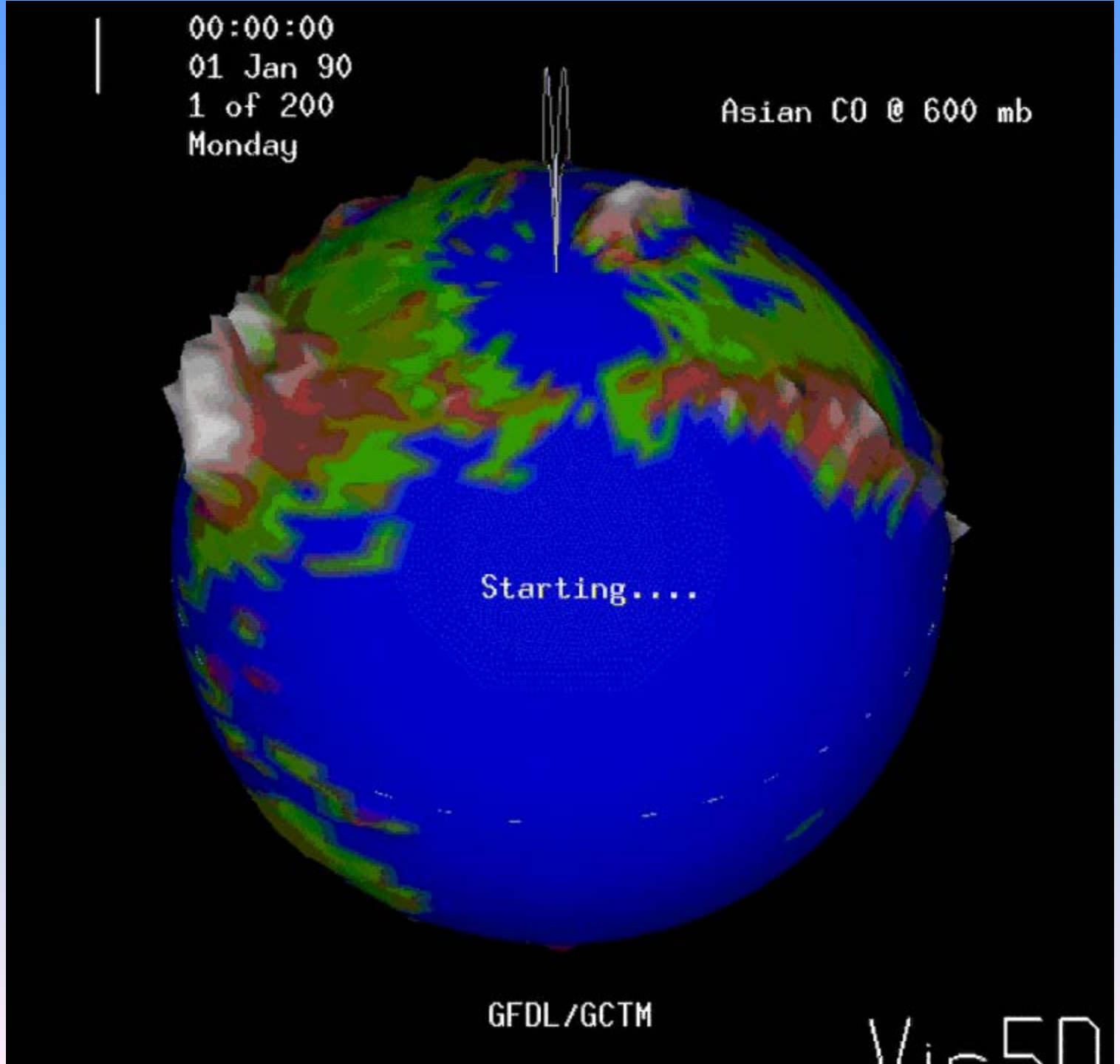


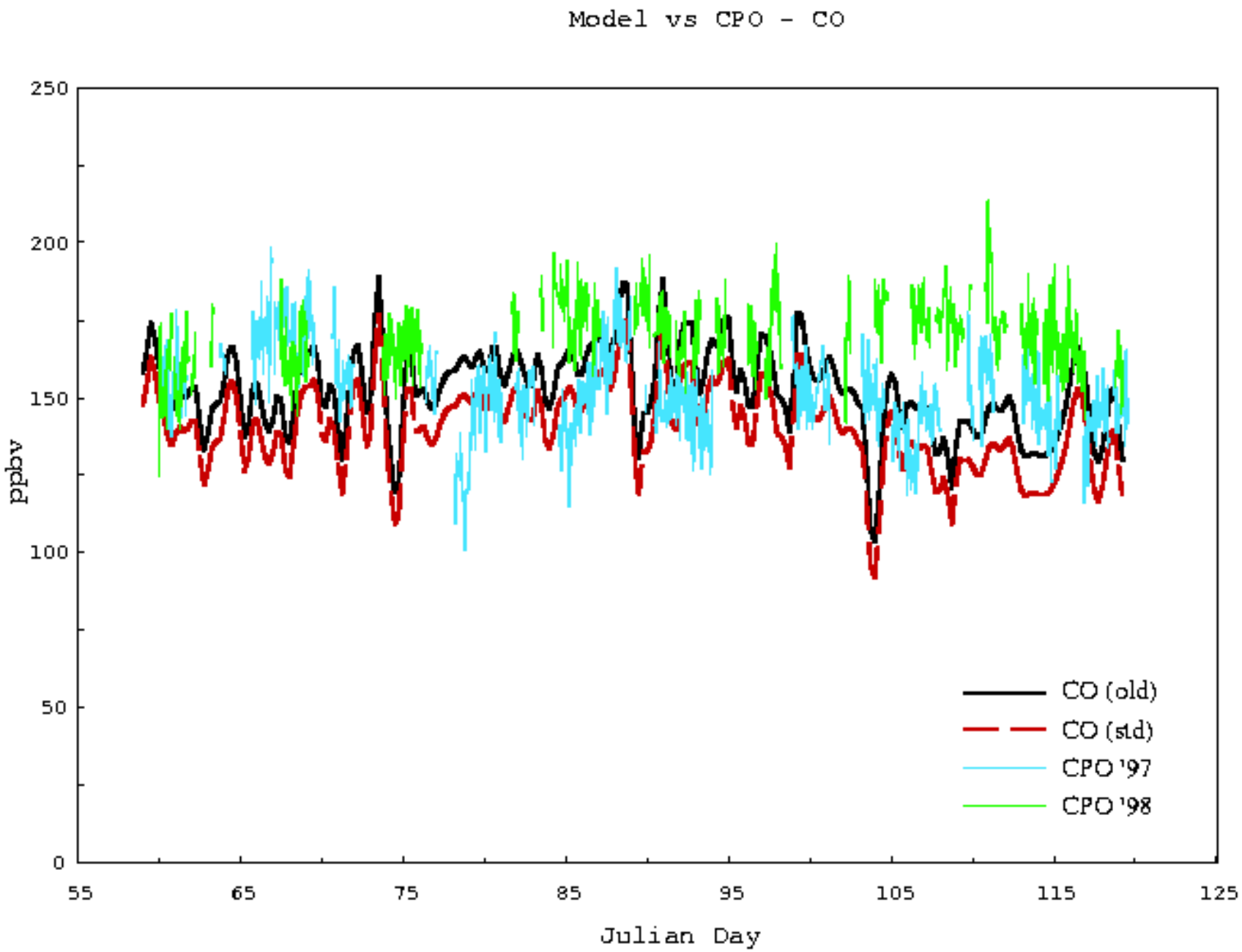
Courtesy of Rudolf Husar, Washington University, St. Louis: http://capita.wustl.edu/Asia-FarEast/

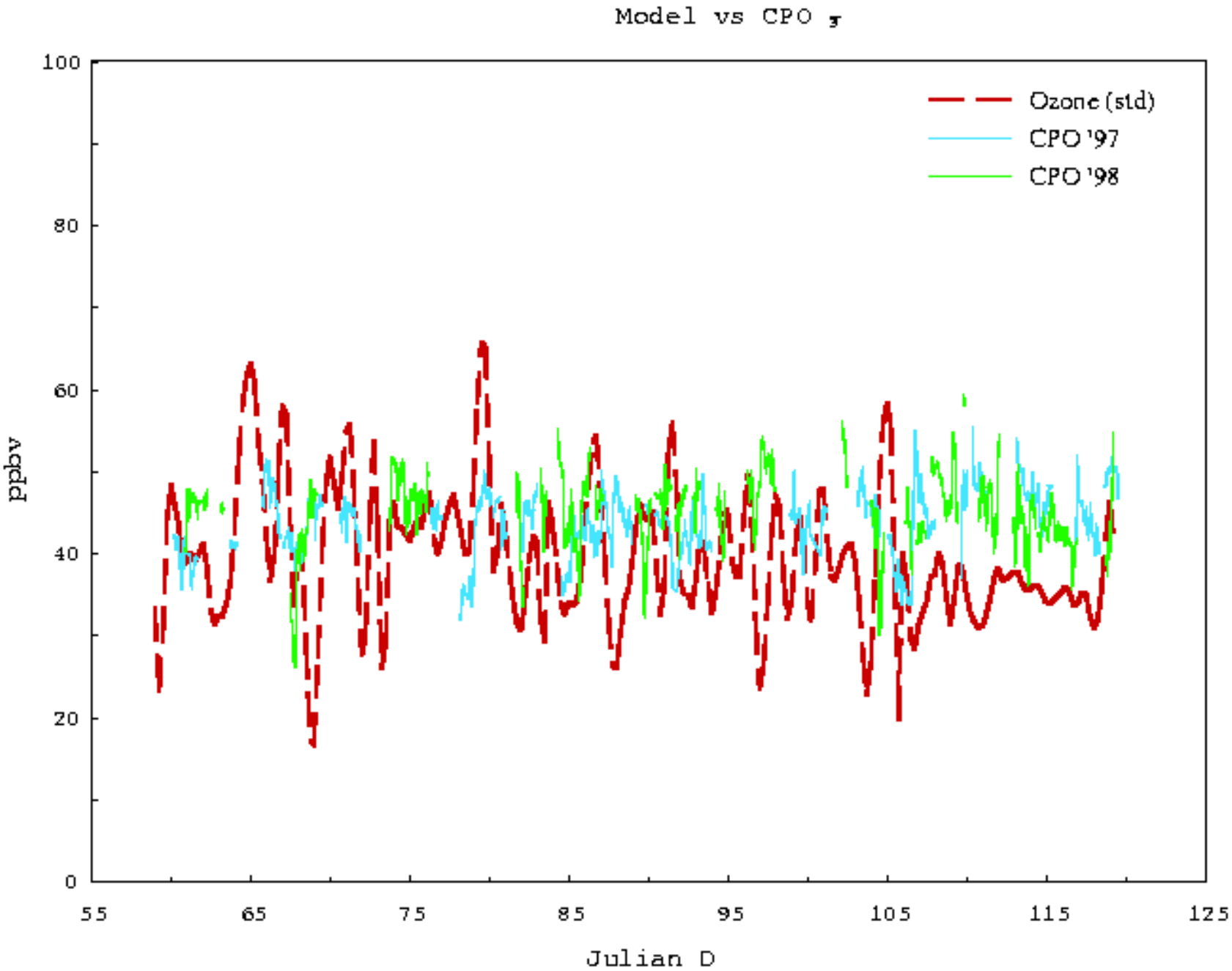
The Changing Air Quality of the Northern Hemisphere Pacific Basin

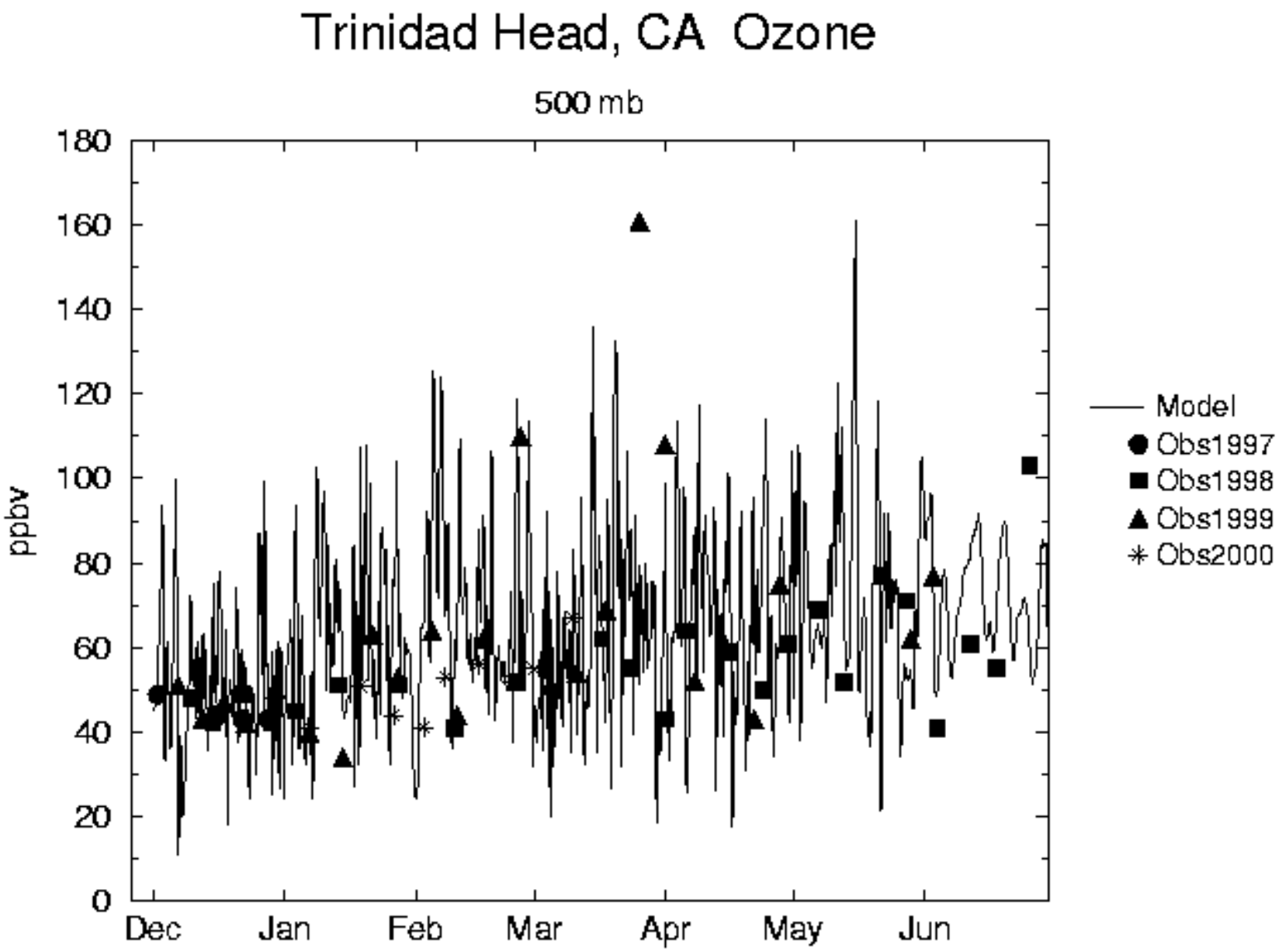
- Pressures are from the Pacific Rim Countries and beyond.
- Changing patterns and growth rates of energy use and resulting emissions are the primary forcing factors -- with East and West following different paths.
- Impacts are local, regional (East and West), basin-wide, and global.
- Complexities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




00:00:00
01 Jan 90
1 of 200
Monday
Asian CO @ 600 mb
Starting....
GFDL/GCTM


Model vs CPO - CO
250
200
150
100
50
0
ppbv
55
65
75
85
95
105
115
125
Julian Day
CO (old)
CO (std)
CPO '97
CPO '98


Model vs CPO 3
Ozone (std)
CPO '97
CPO '98
ppbv
100
80
60
40
20
0
55
65
75
85
95
105
115
125
Julian D


Trinidad Head, CA Ozone
500 mb
ppbv
180
160
140
120
100
80
60
40
20
0
Dec
Jan
Feb
Mar
Apr
May
Jun
Model
Obs1997
Obs1998
Obs1999
Obs2000

Contribution of Fossil Fuel Burning to Tropospheric Ozone

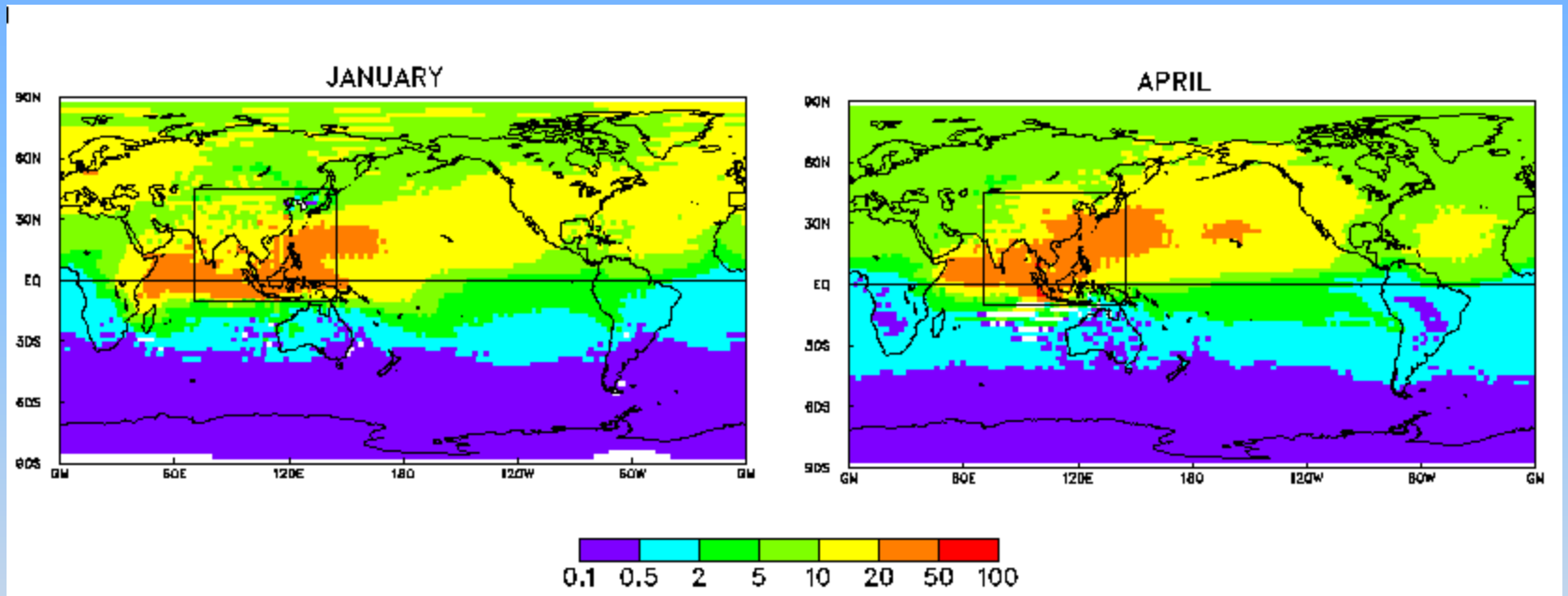
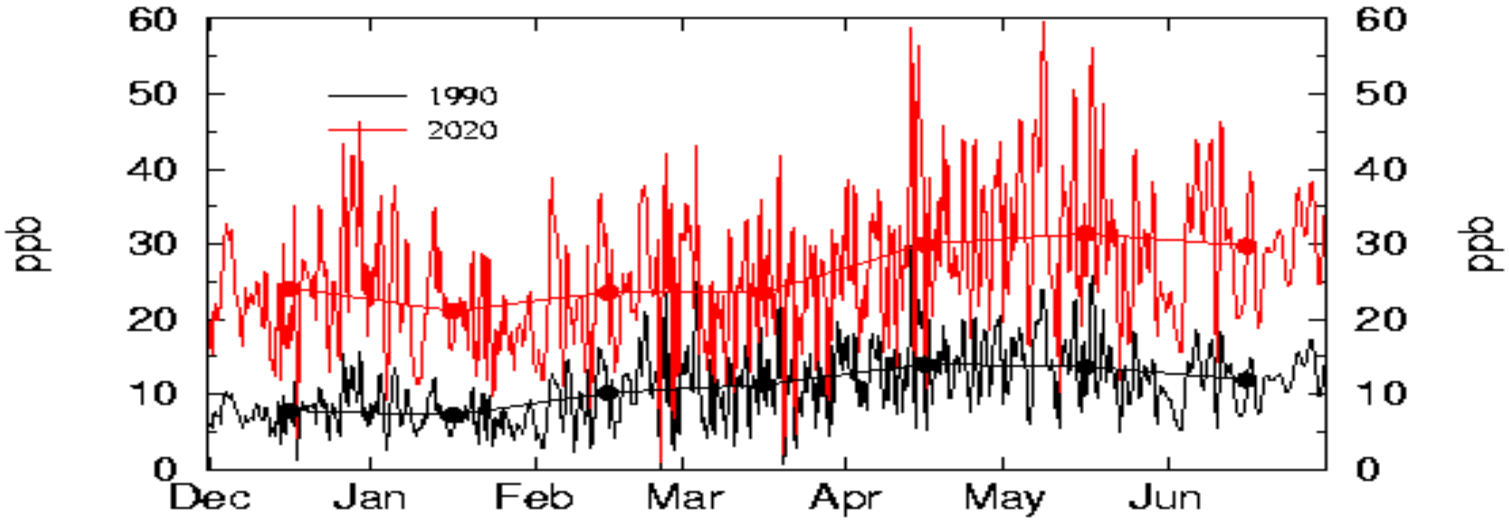


Figure 3.14. Percentage of total ozone accounted for by Asian fossil fuel NOx emission at 940 mb (sigma surface) for January, April

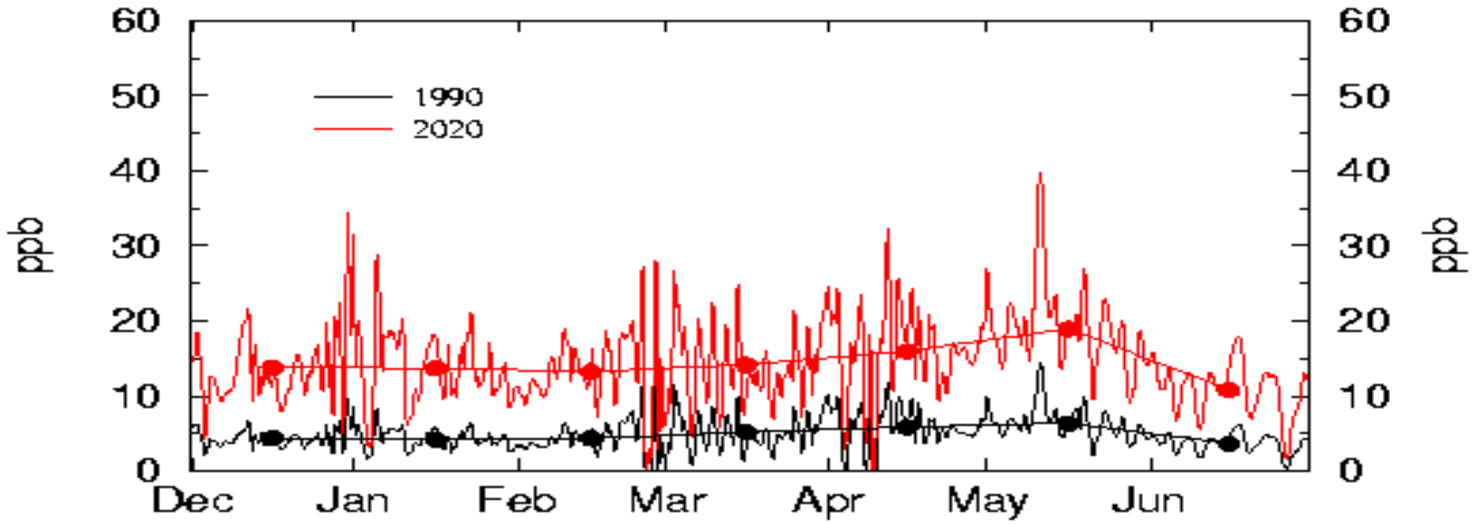
Yienger, et al, JGR 2000

Asian Contribution to Total Ambient Ozone over Central California

500mb (6hour averages)



940mb (6hour averages)



Yienger et al., JGR, 26931, 2000

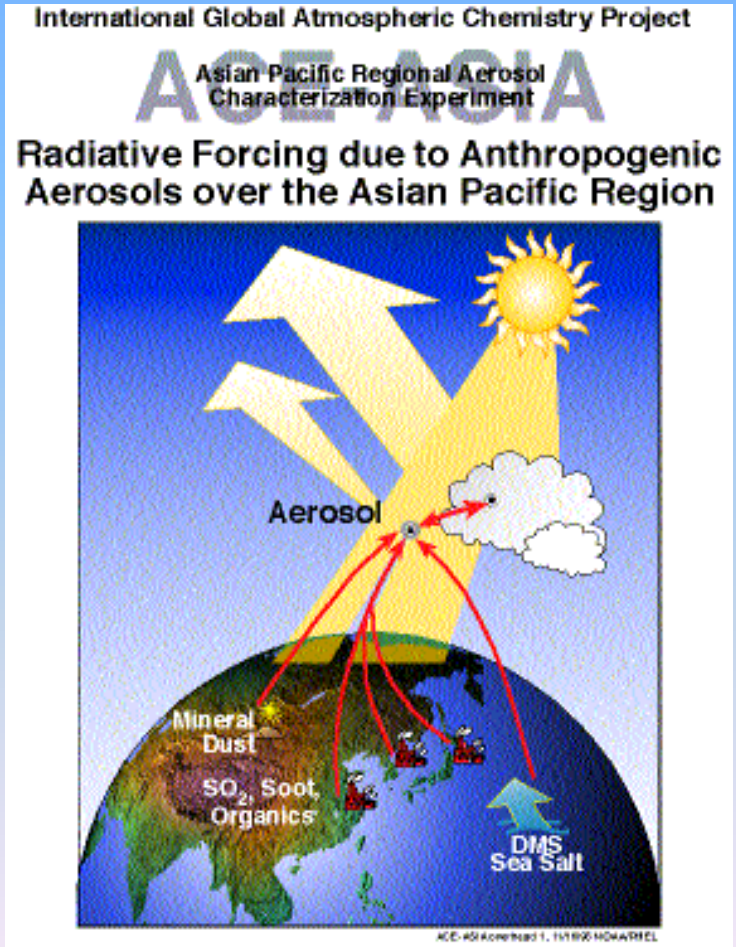
Further Quantification Requires

- *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



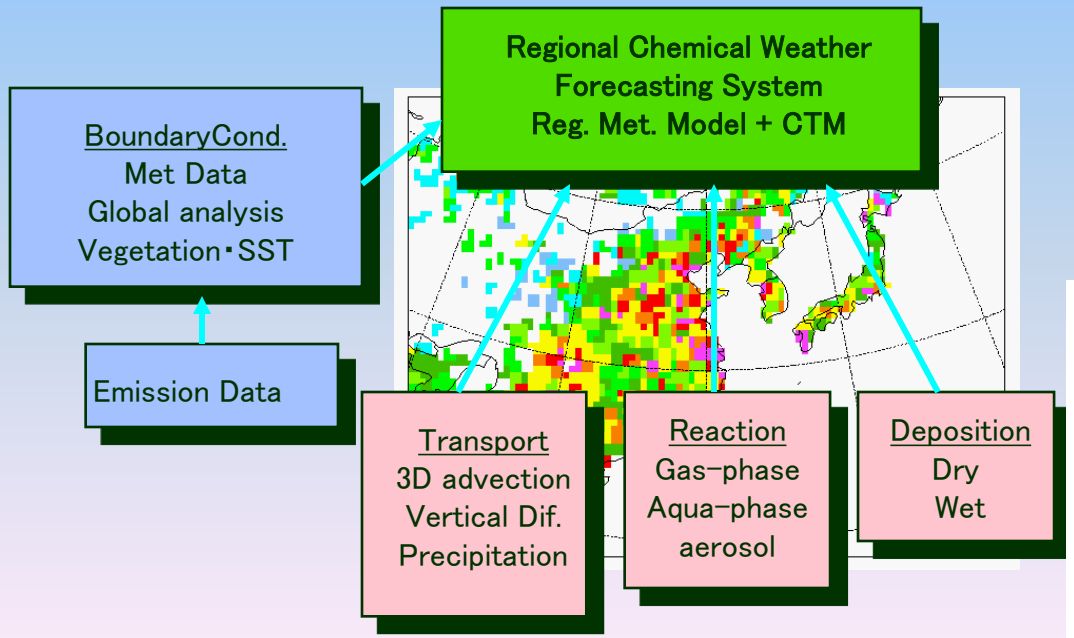
NASA/GTE DC-8

Development and Application of Chemical Weather Forecasting System over East Asia

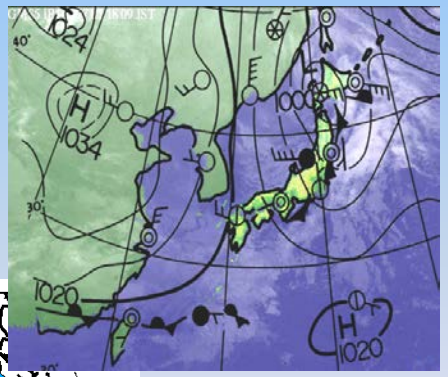
Itsushi Uno (RIAM/Kyushu-U), Gregory R. Carmichael (UI/CGRER)

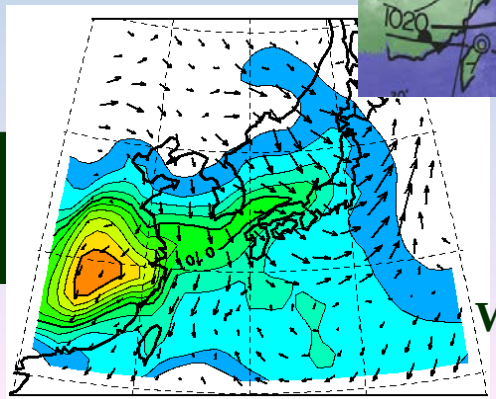
We are developing and applying an operational chemical weather forecasting system based on 3-D on-line regional scale chemical transport model fully coupled with RAMS (Regional Atmospheric Modeling System, Pielke *et al.*, 1992). This system consists of several important components; i) operational global 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.



Weather Map

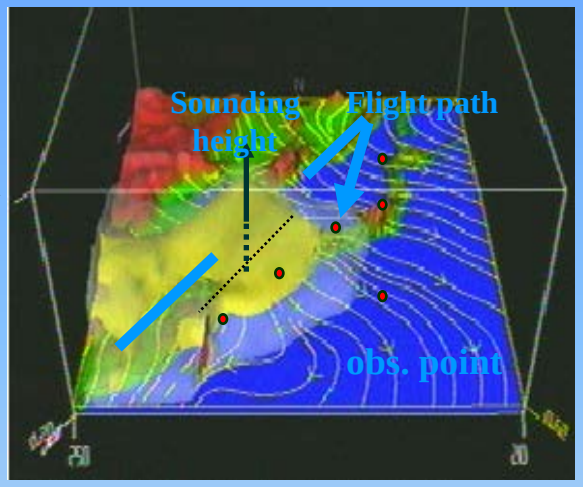




Chemical Weather Map

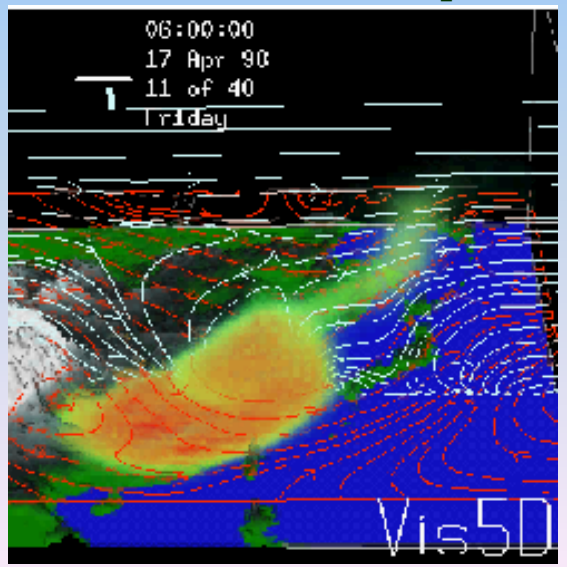
Application & Model Validation

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

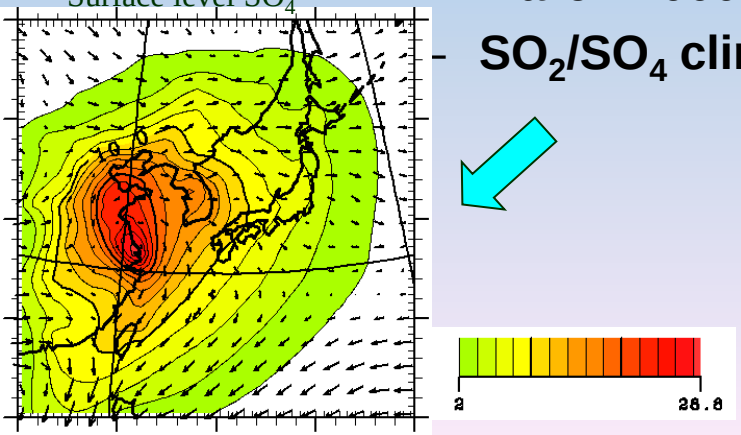


- Model Validation for Episodic Application
  - Yellow Sand (Kosa) April 1998 & March 2000
  - SO_2/SO_4 climate simulation

Simulated Kosa Onset April 98

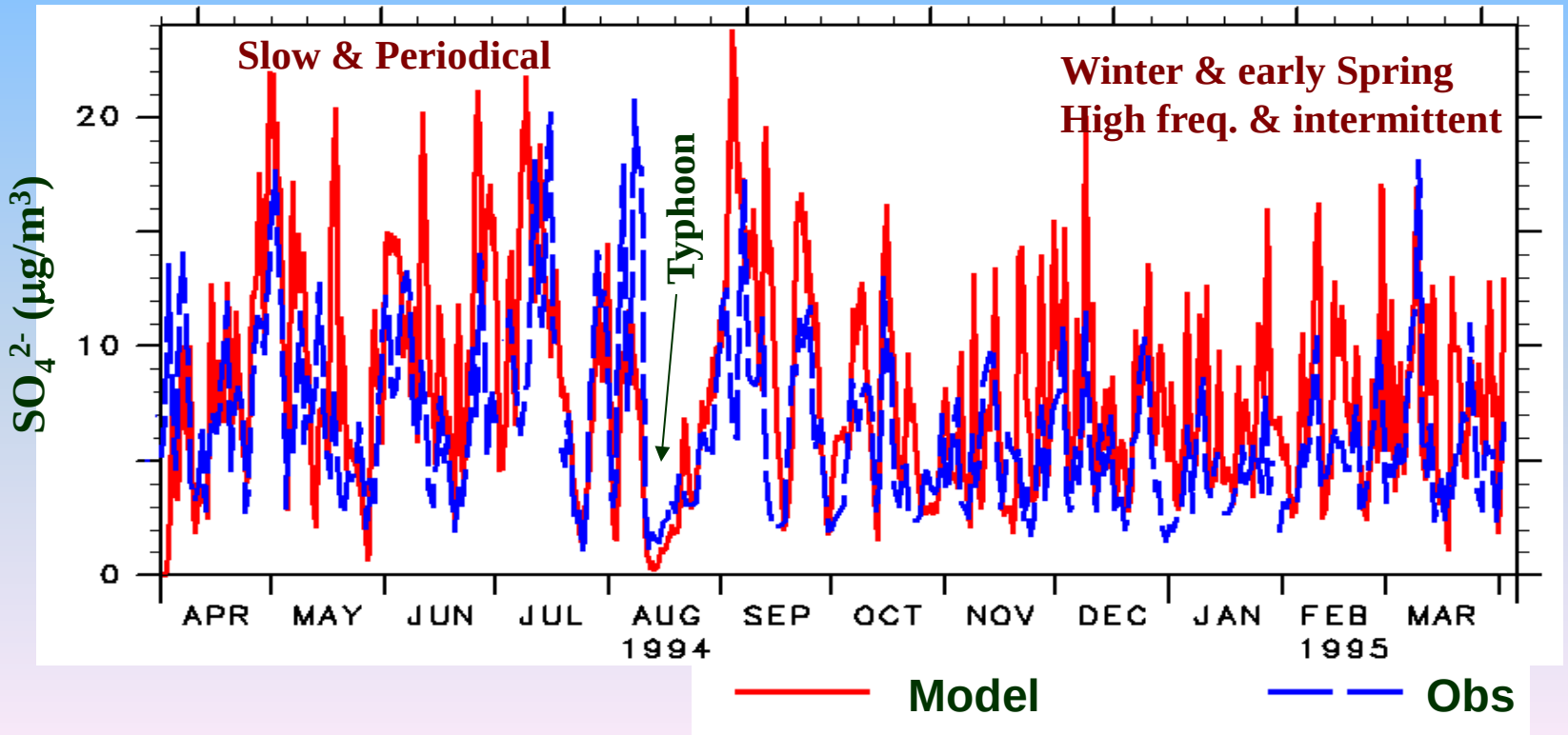


Annual averaged Surface level SO_4^{2-}



Observation at Osaka and Model Results

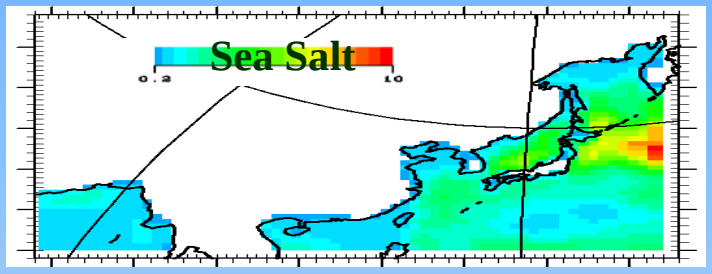
Observed sulfate concentration at Osaka compared with model results show a good agreement, and the intermittency during the winter season and the periodicity typical of spring/fall rainy seasons, when the alternance of high/low pressure systems characterize the meteorology of the region, is nicely reproduced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



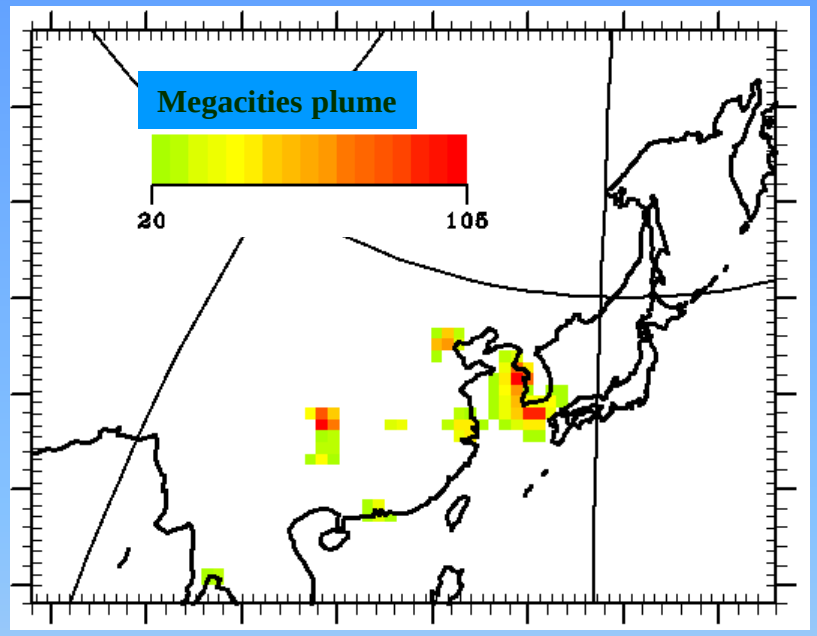
Modeled twelve hours averaged concentration (straight line) and daily averaged observation (dot line)

Tracer Species (2)

Several useful atmospheric tracer to
Understand the origin of air mass

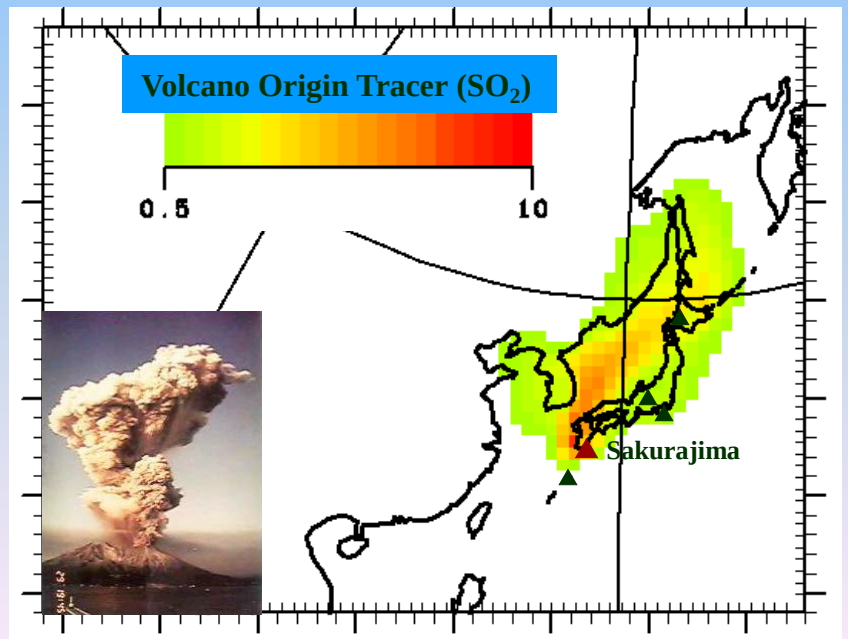


12 bin Sea Salt from Gong et al.
Production rate depends on wind speed

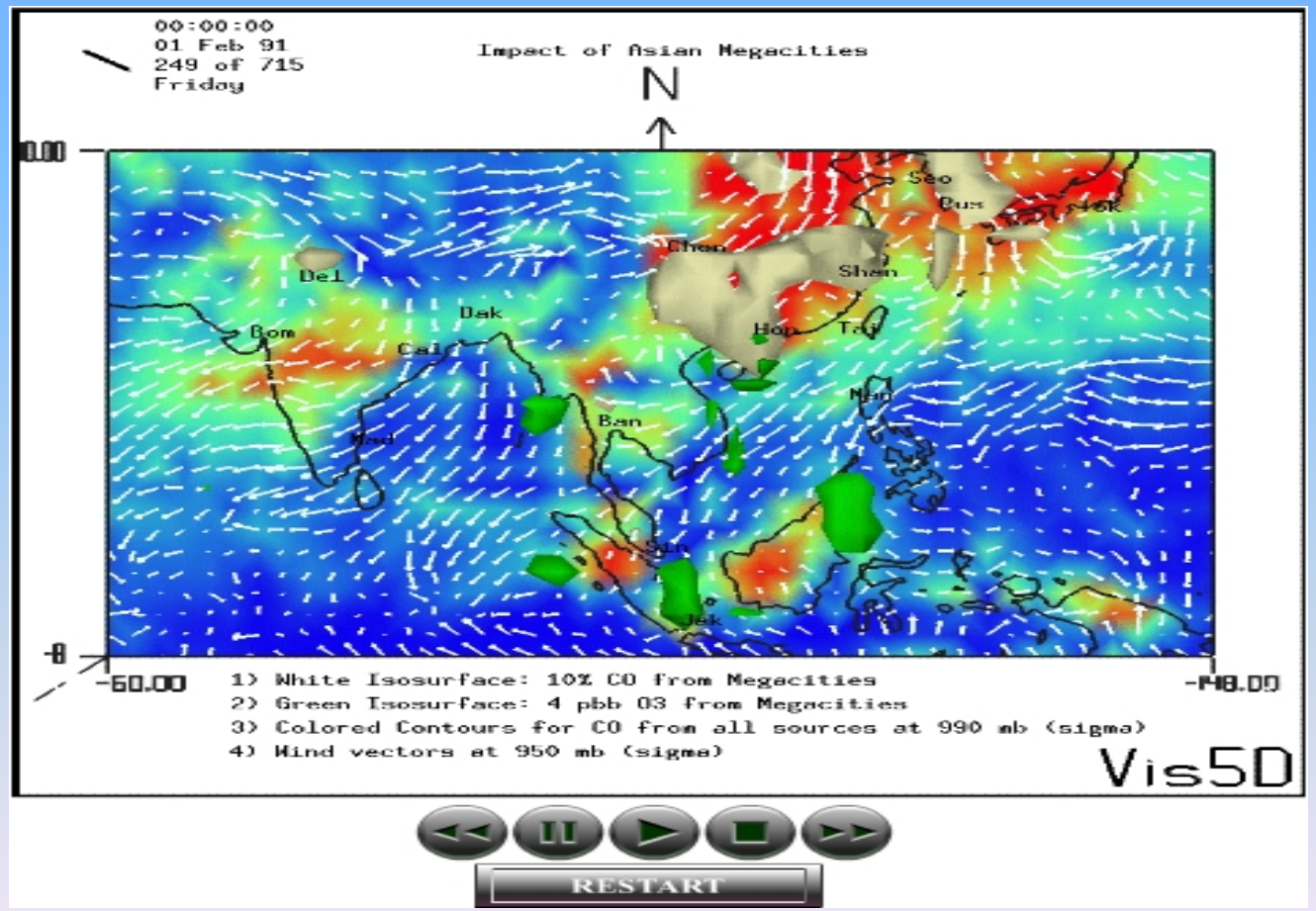


Tracers/Markers:

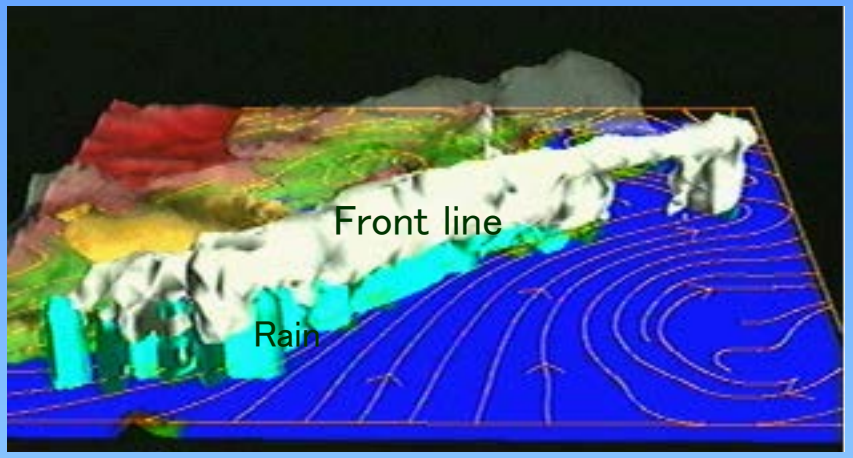
SO2/Sulfate	DMS
BC	OC
Volcanic	Megacities
CO fossil	CO-Biomass
Ethane	Ethene
Sea Salt	Radon
Lightning NOx	Dust 12 size bins

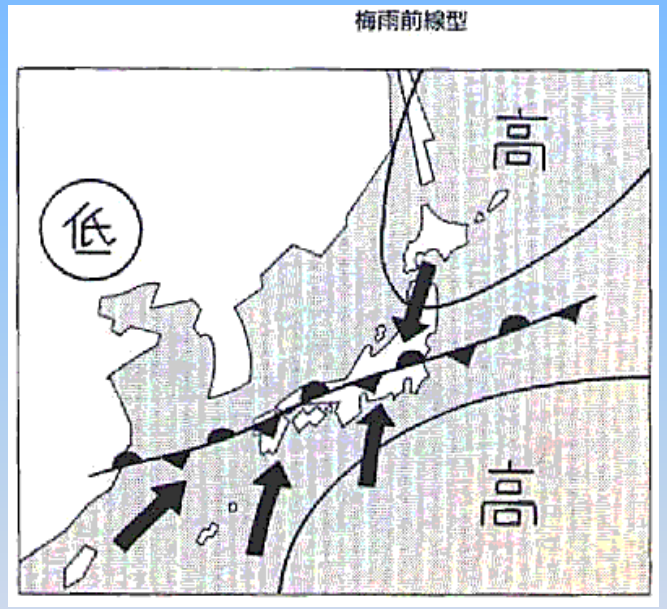


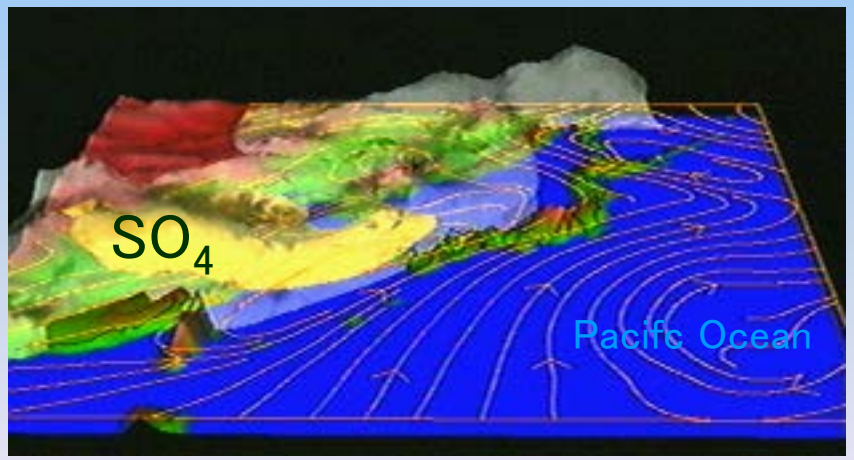
Asia Megacity Tracers



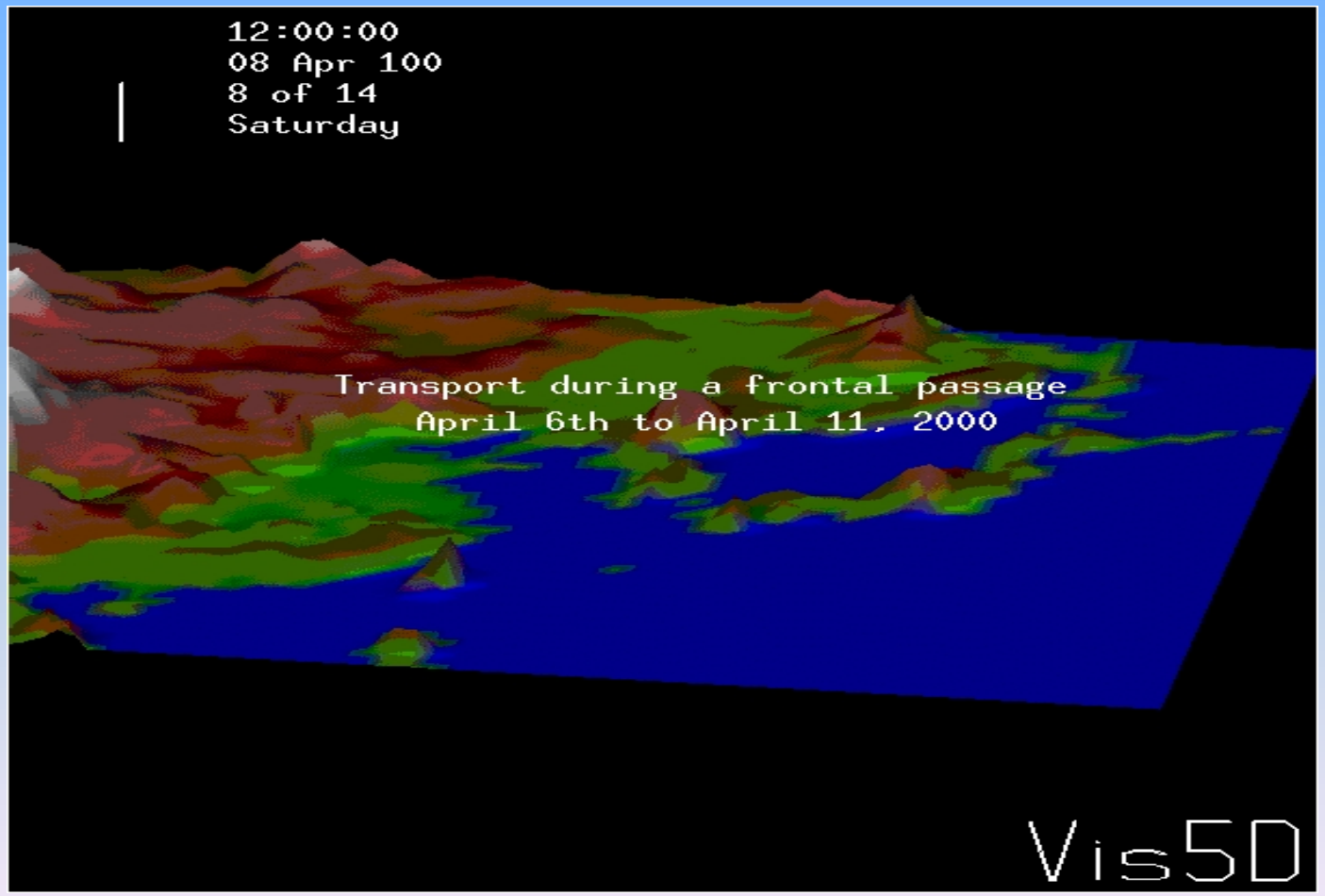
Rainy Meso Front devides air mass



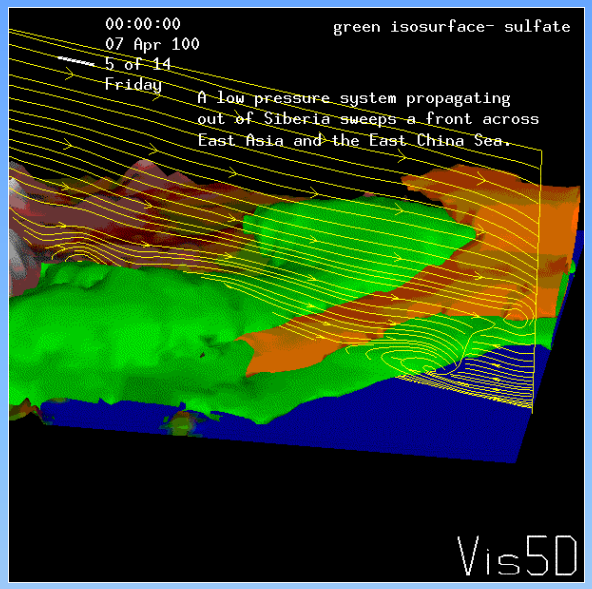


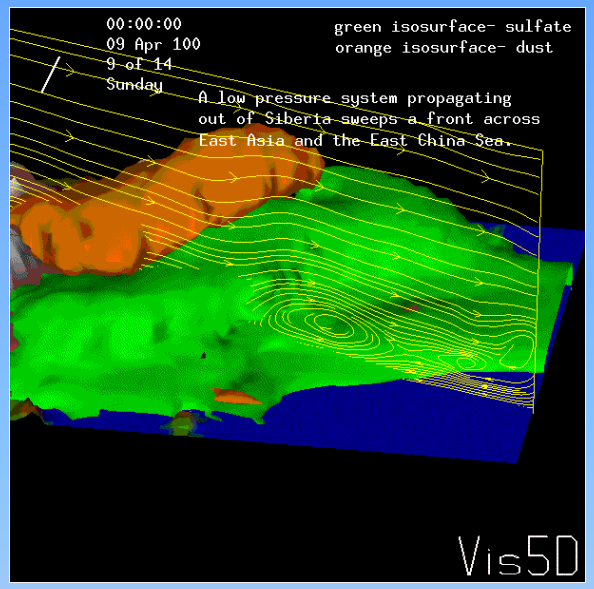


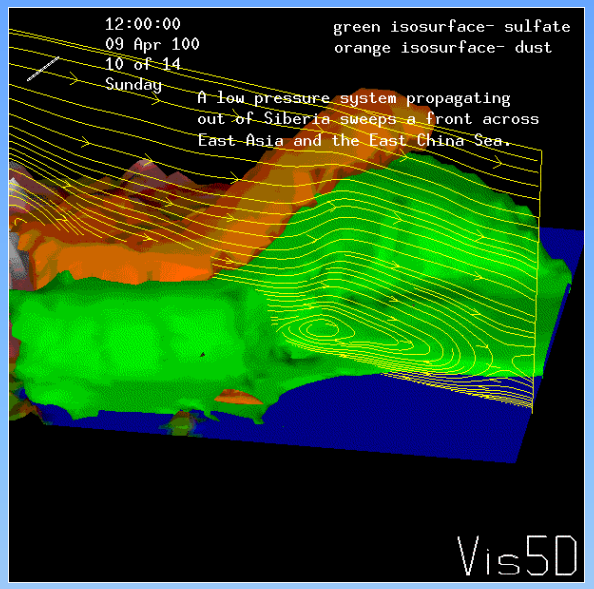
Frontal Outflow is the Key Transport Mechanism and needs to be Better Quantified

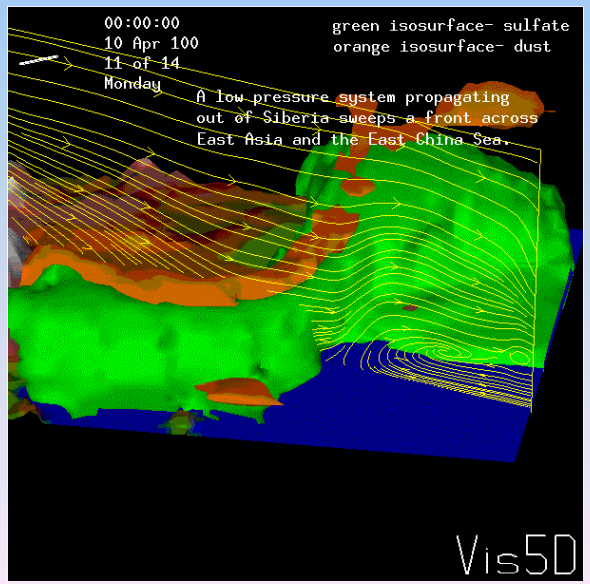


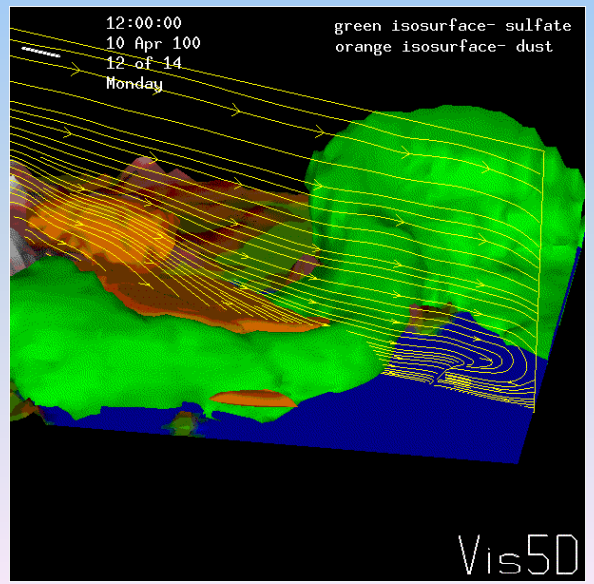
Frontal Outflow: Dust (brown) & Sulfate (green)

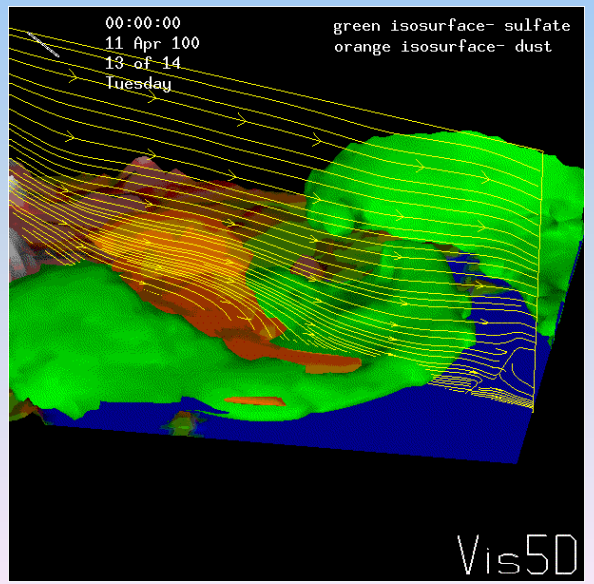








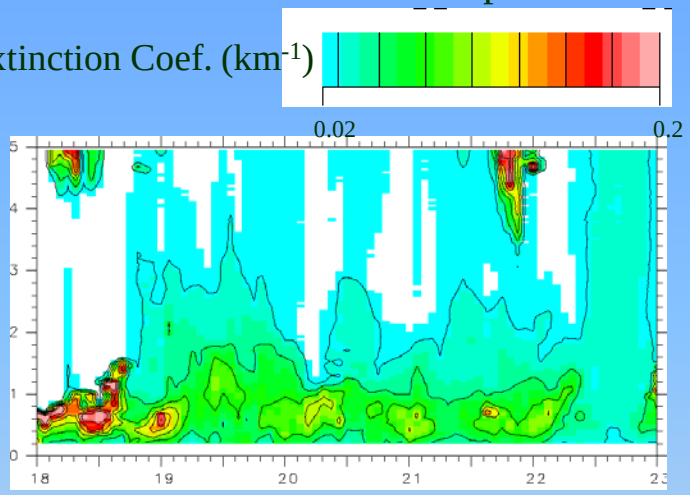




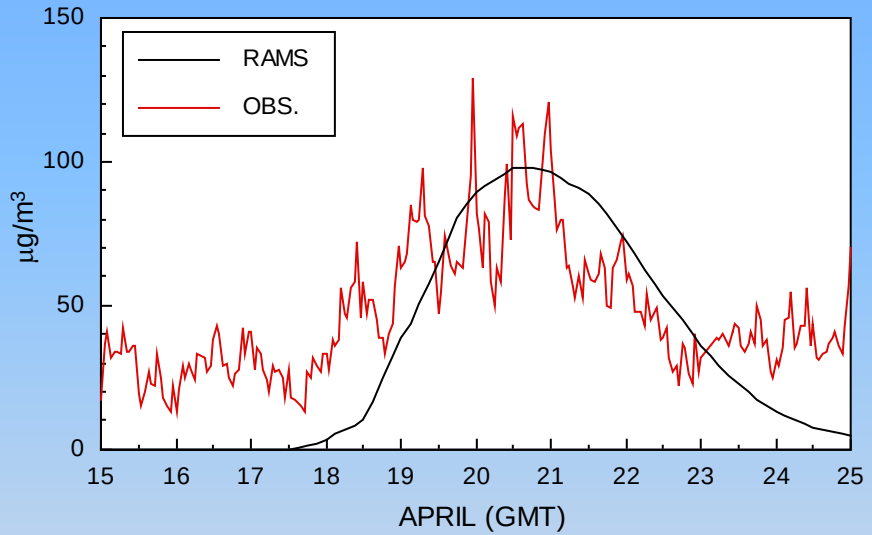
Tokyo & Tsukuba (observation and model results)

National Inst. Env. Studies Compact Mie Lidar

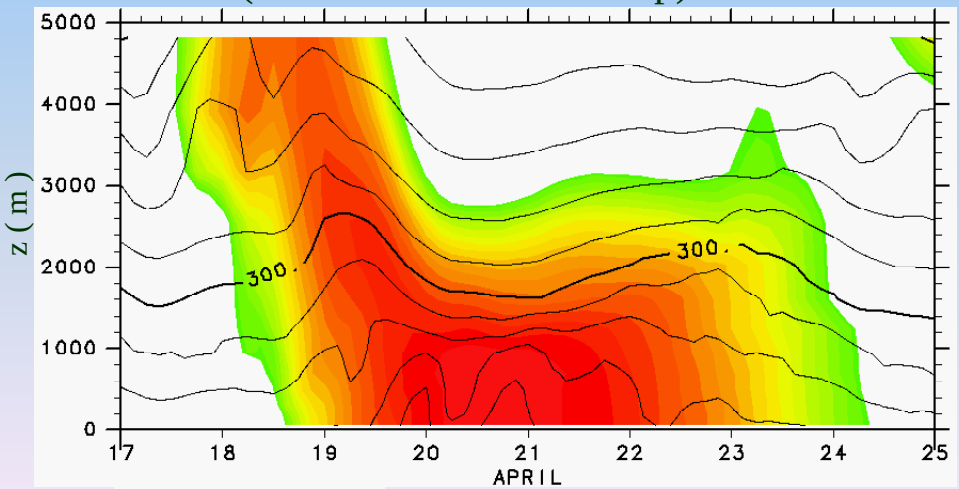
Extinction Coef. (km^{-1})



SPM (Observation and Model)at Tokyo



Model (Dust and Potential Temp)

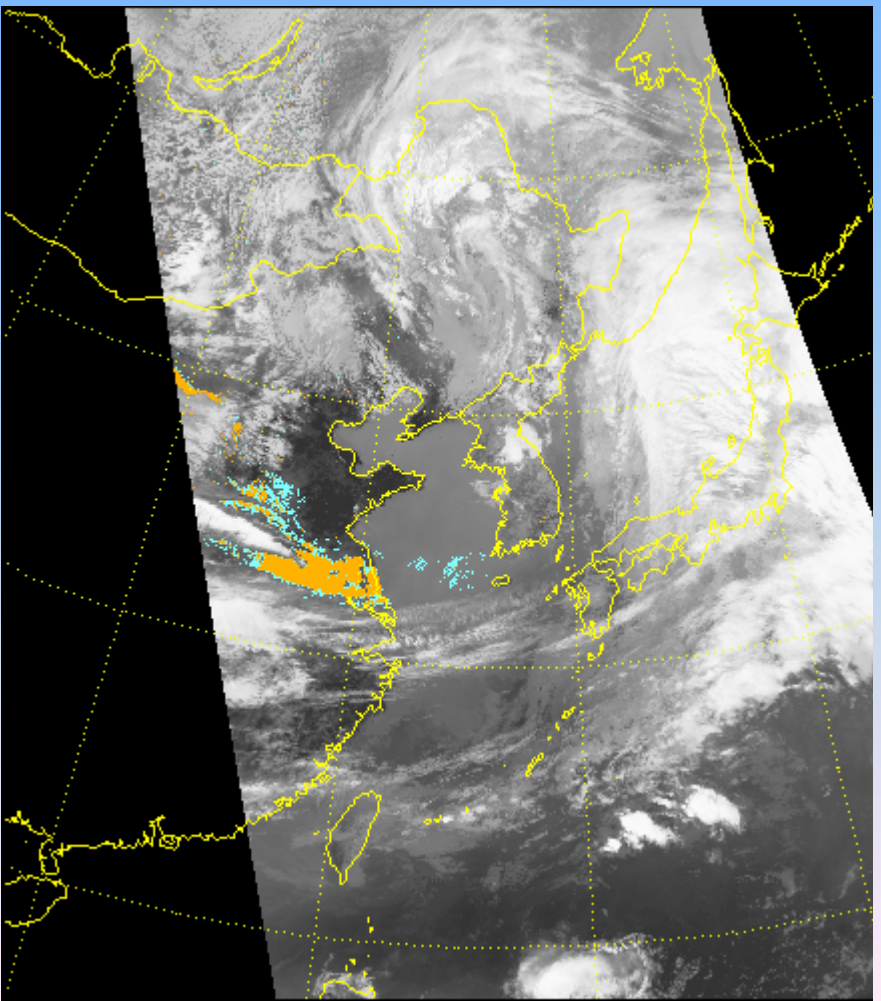


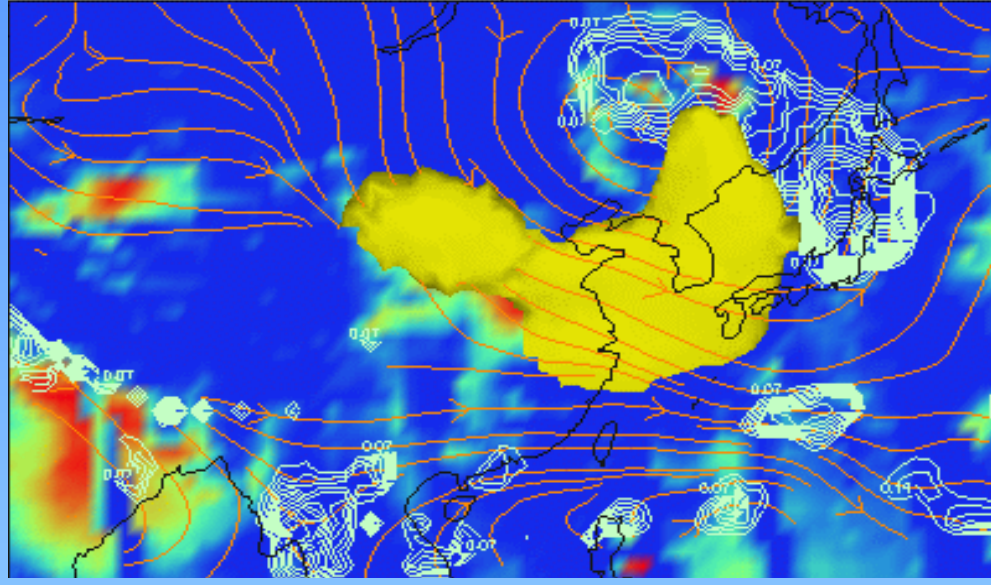
Conc unit log($\mu g/m^3$)

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)

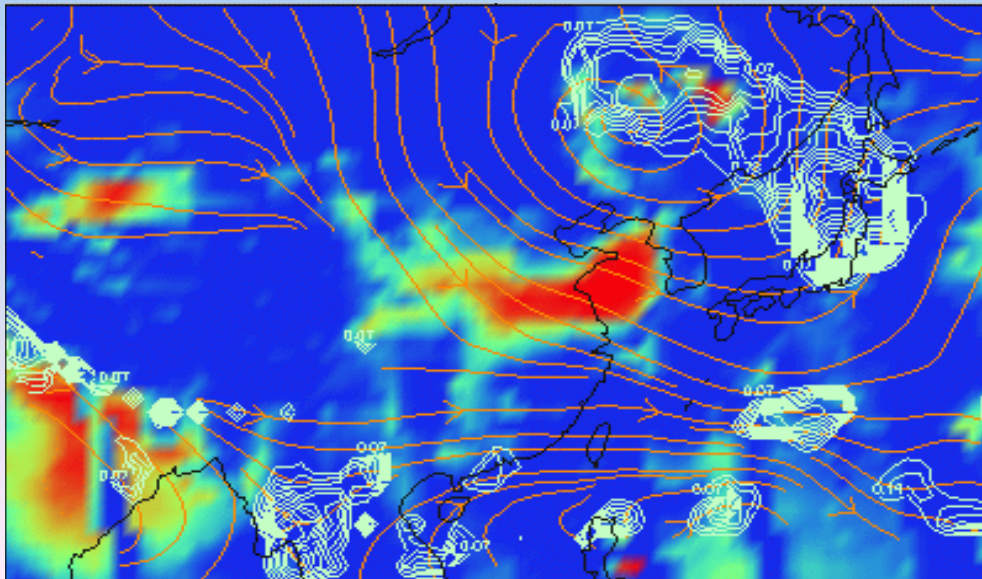


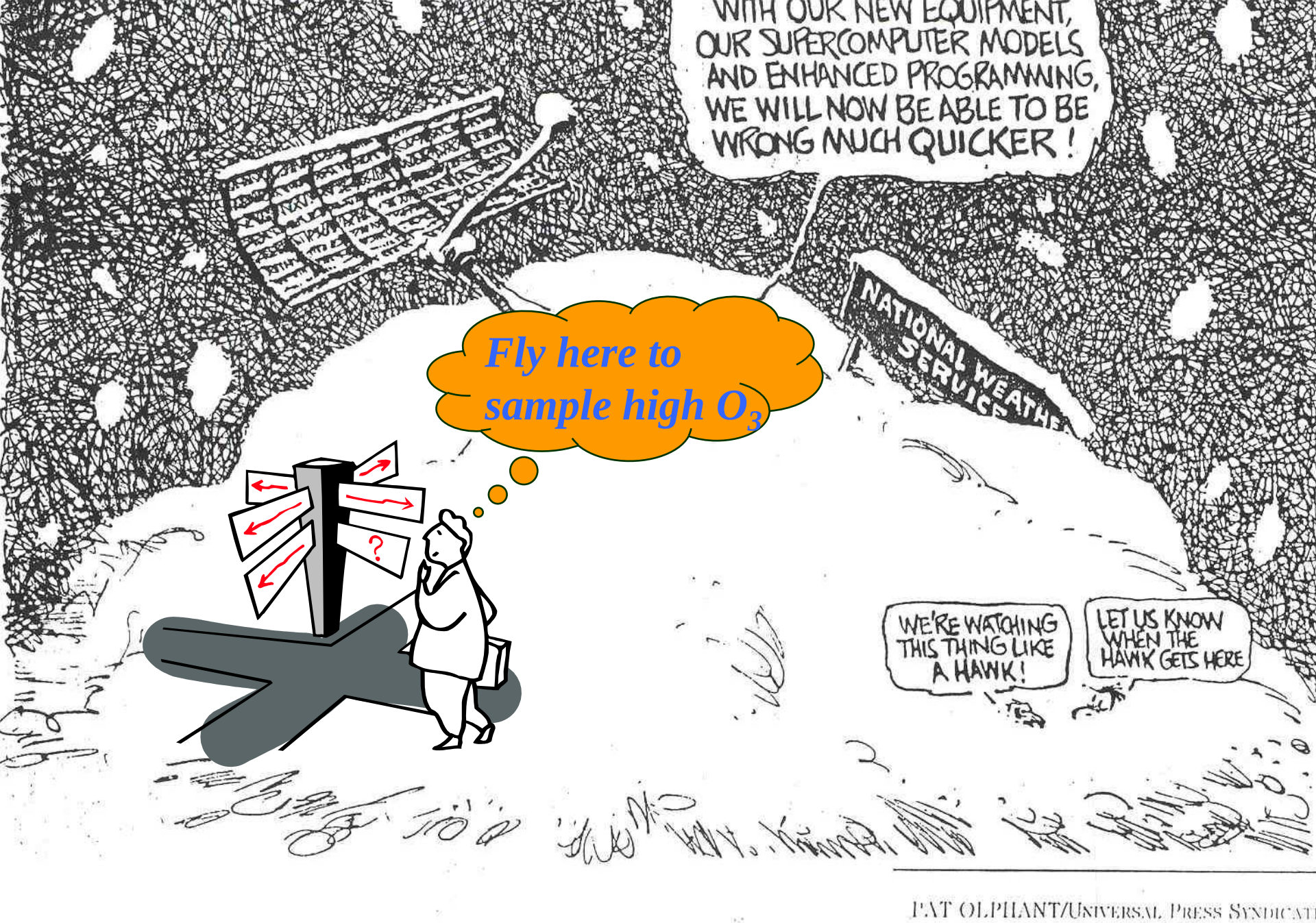


TOMS AI over laid Dust (Yellow Sand) and BC

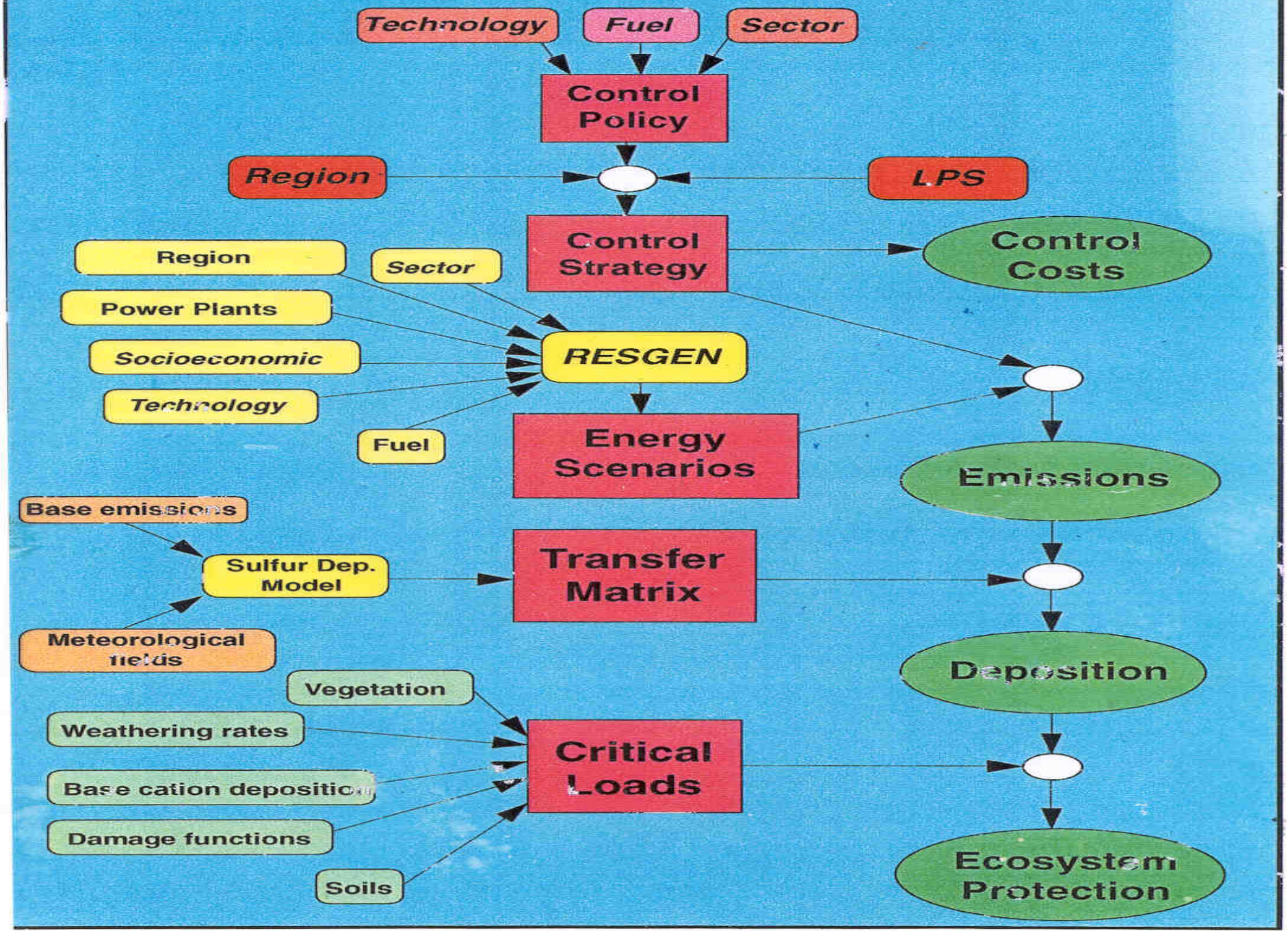
TOMS-IA (Color shading)
Dust Isosurface (Yellow)
BC Isosurface (Orange)
Precipitation (Contours)

TOMS AI and Precip

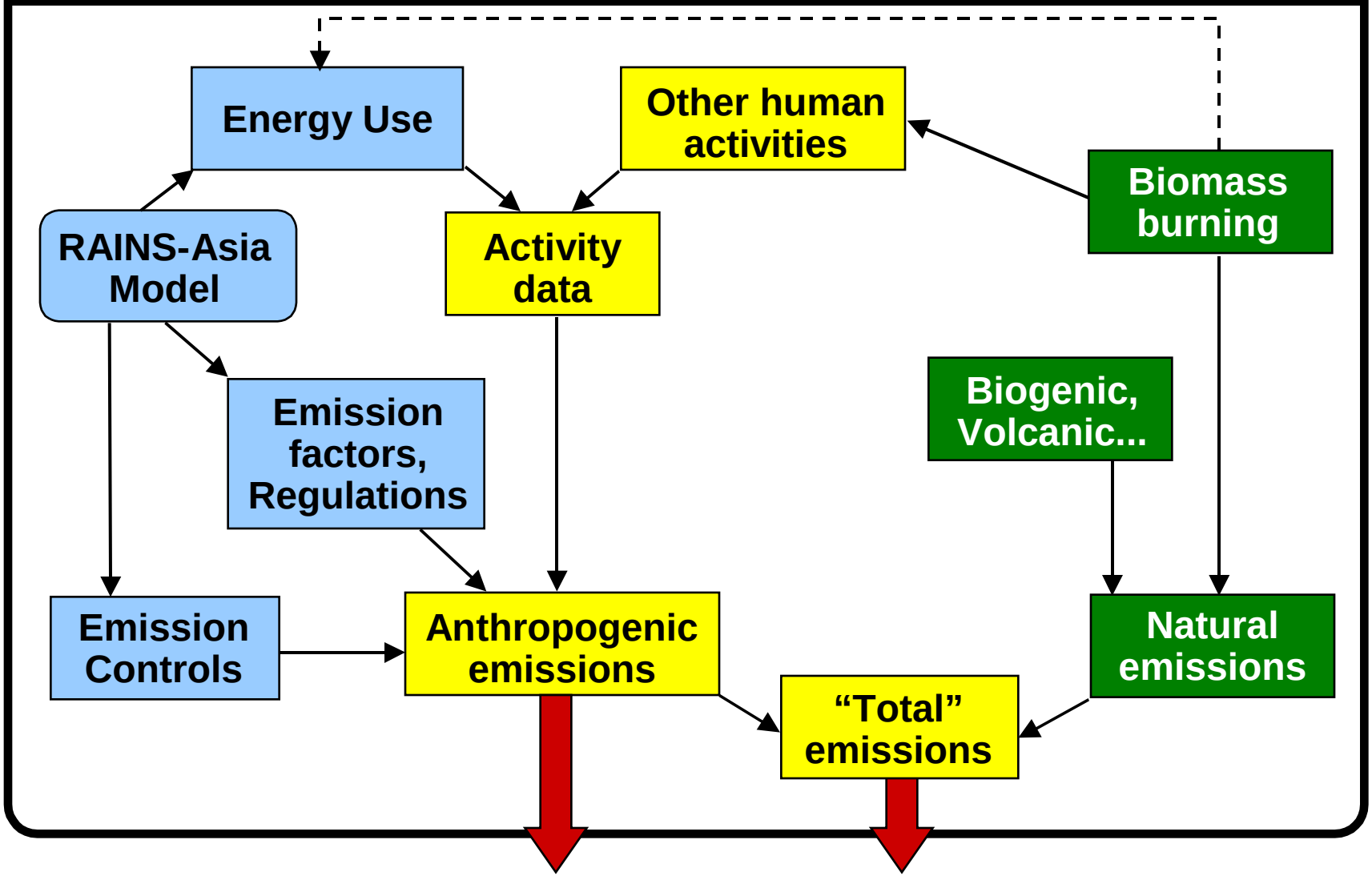



WITH OUR NEW EQUIPMENT, OUR SUPERCOMPUTER MODELS AND ENHANCED PROGRAMMING, WE WILL NOW BE ABLE TO BE WRONG MUCH QUICKER!
NATIONAL WEATHER SERVICE
Fly here to sample high O_3
WE'RE WATCHING THIS THING LIKE A HAWK!
LET US KNOW WHEN THE HAWK GETS HERE
PAT OLPHANT/UNIVERSAL PRESS SYNDICATE

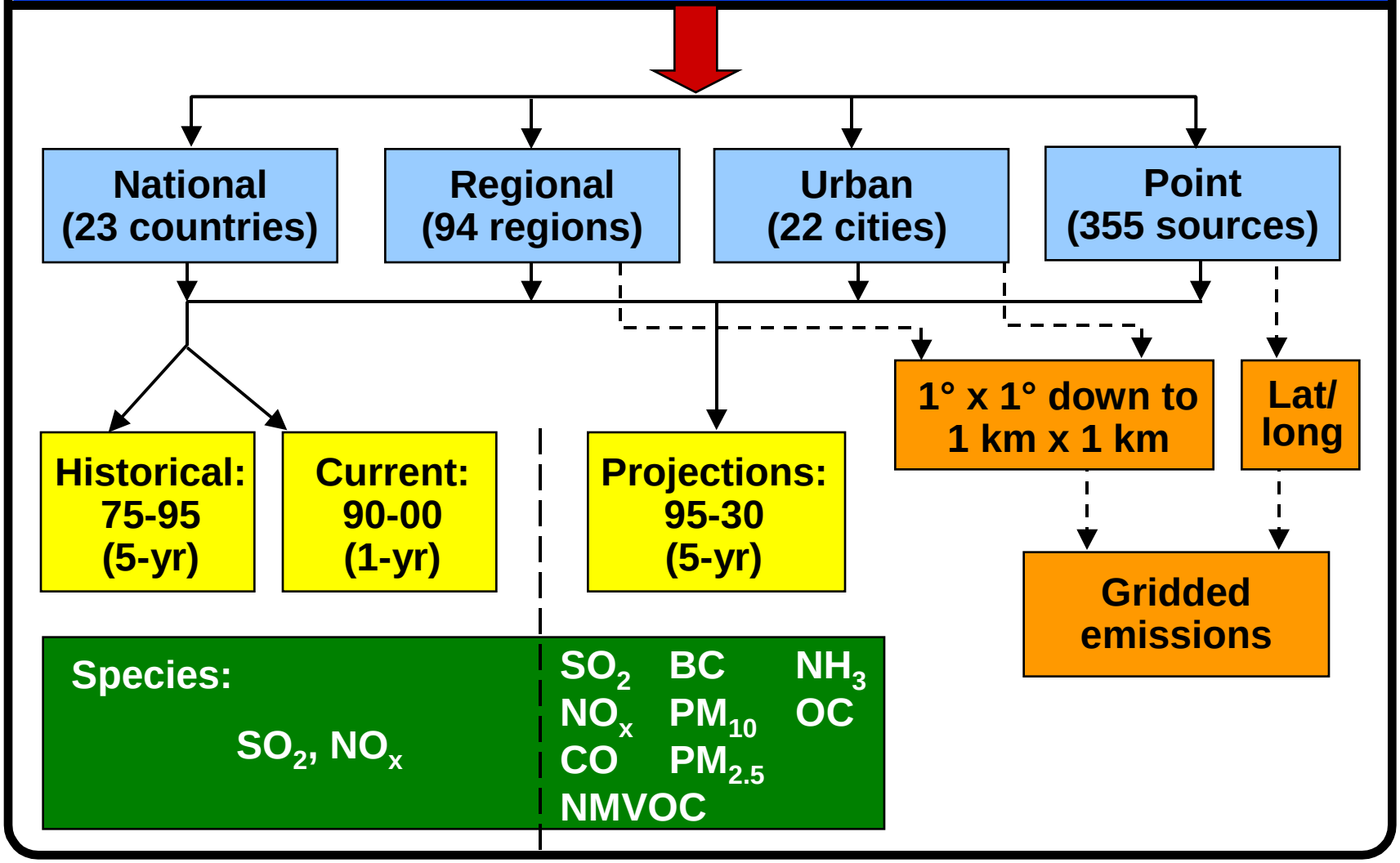
RAINS ASIA FLOW CHART



Methodology for Asian Emission Estimates



Organization of Emissions Data



ACESS-website(Emission Data)

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Bookmarks Location: http://www.cgrer.uiowa.edu/people/carmichael/ACESS/Emission-data_main.html What's Related

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ACE-Asia

Chemical Transport Model

Emission Data

Mission Planning

On-line Tracers

Relevent Publications

TRACE-P

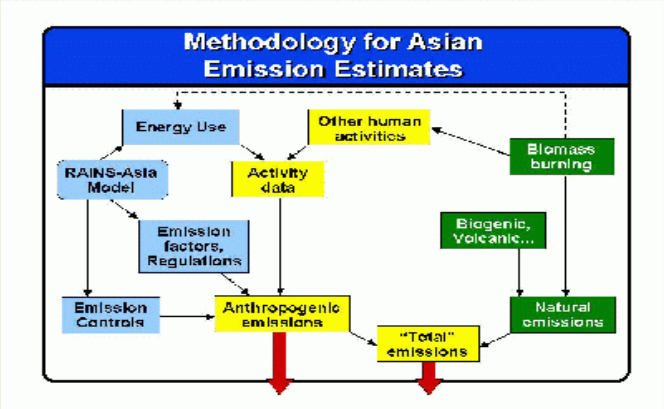
Weather Forecast Model

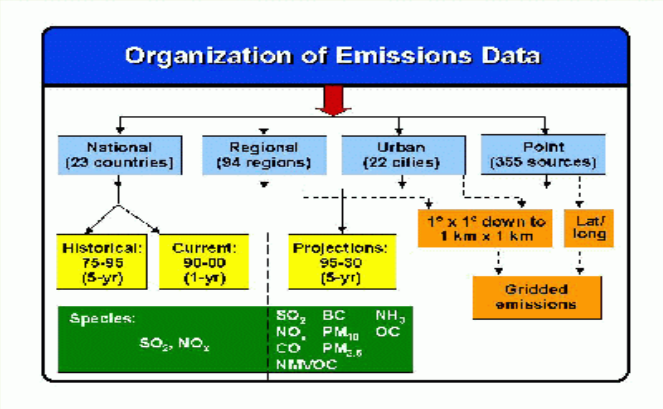
Back to GRC's Homepage

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.





Datasets

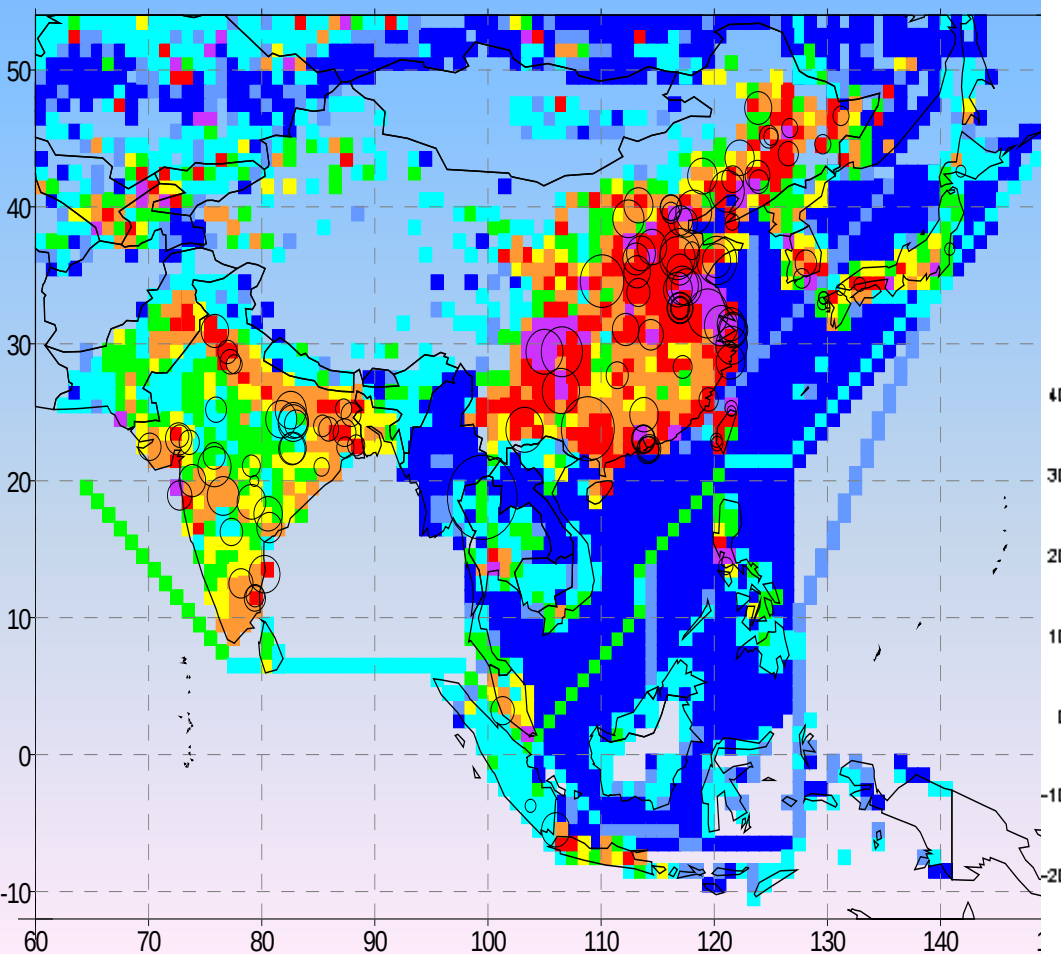
Level	Species
Regional	1) Black Carbon 2) CO 3) NMVOC 4) NOx 5) SO_2
1° x 1°	1) Black Carbon 2) CO 3) NMVOC 4) NOx 5) SO_2

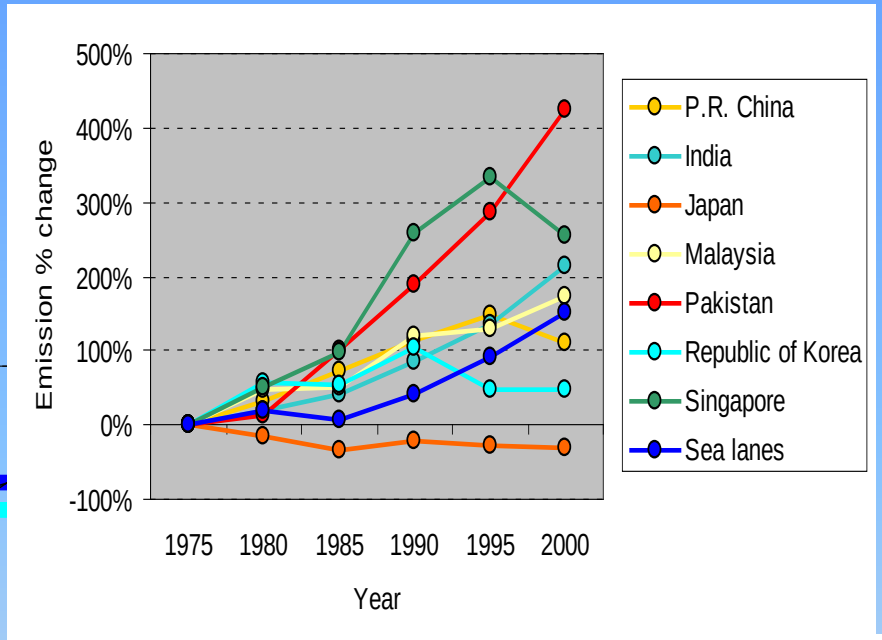
Gridding Capabilities

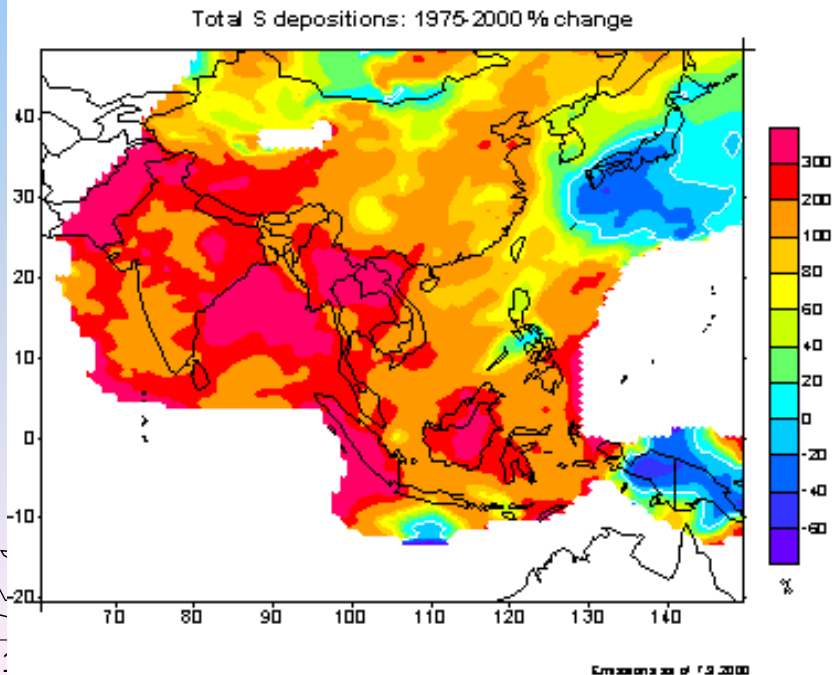
We will put procedures and examples about the gridding here.

- Gridding Procedure for Black Carbon

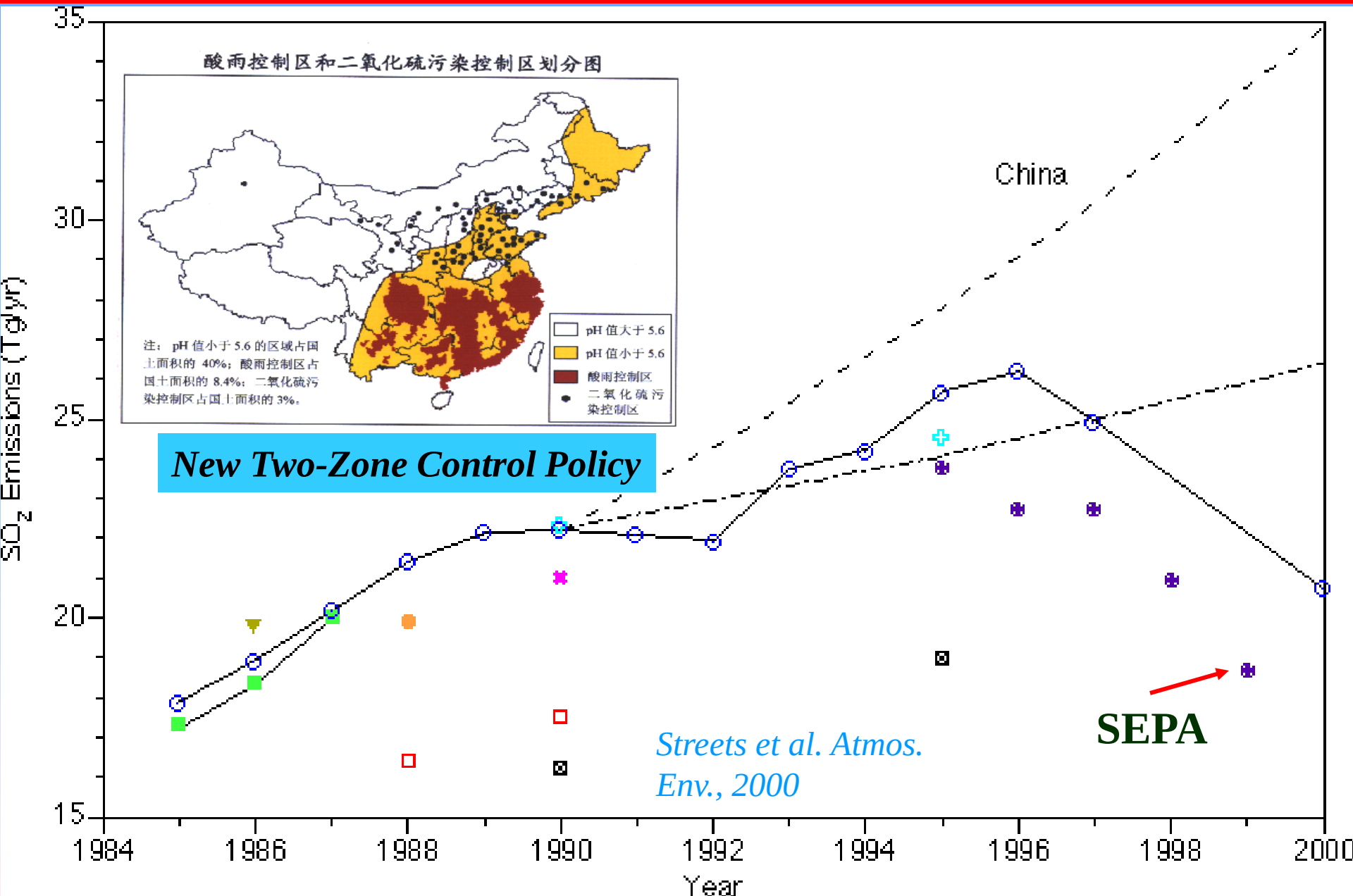
SULFUR EMISSIONS IN ASIA ARE CHANGING, REGION BY REGION, IN DIFFERENT WAYS





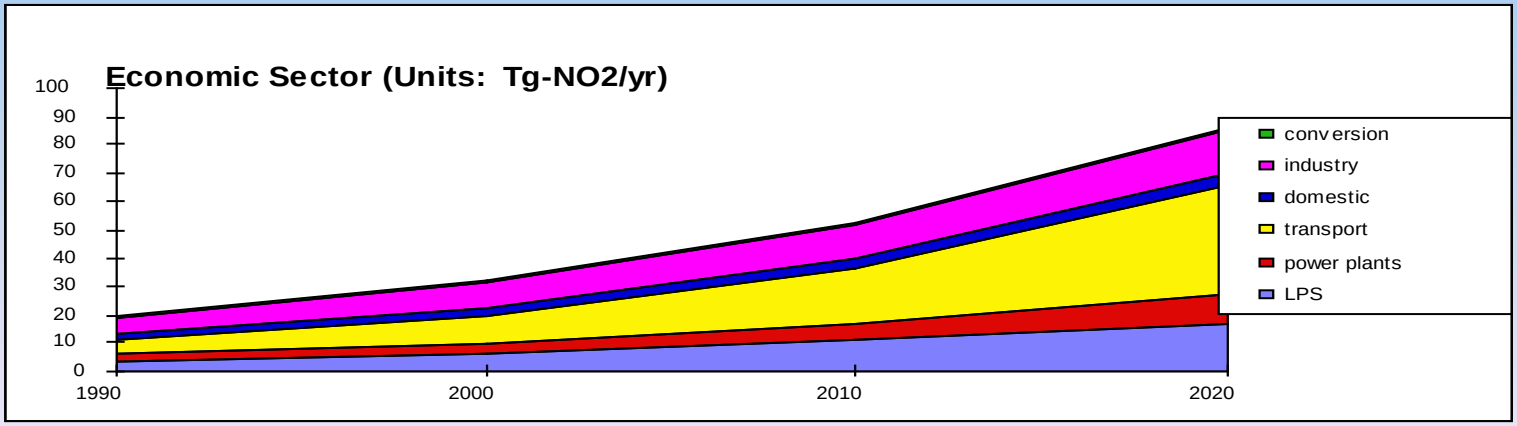


UNEXPECTED DECREASES IN CHINA'S SULFUR EMISSIONS
--- *CAN THESE REDUCTIONS BE SUSTAINED?*



GAS GUZZLING: A Bangkok tie-up symbolizes growing Third World energy demands, which hurt efforts to cut fossil-fuel use

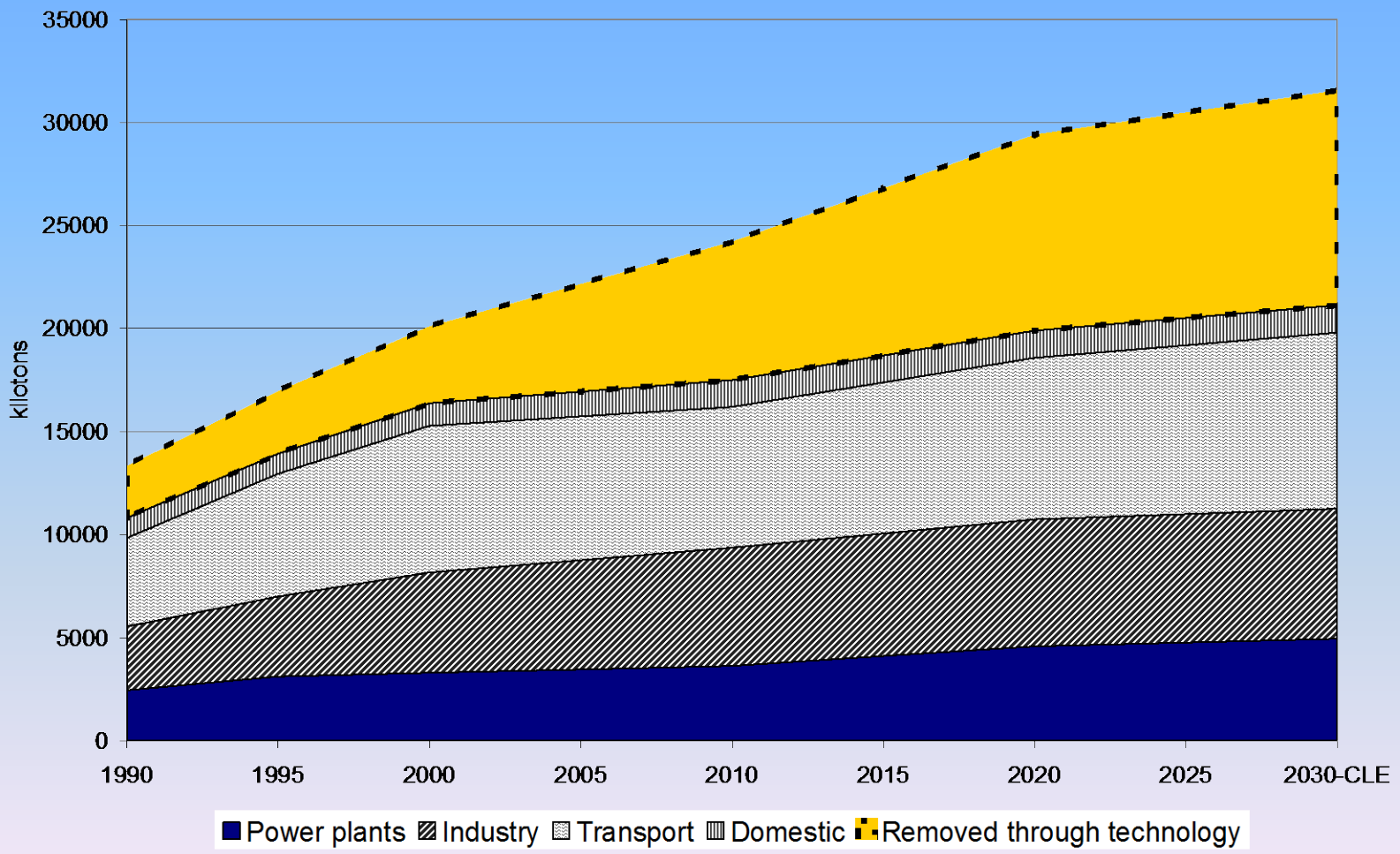
The Rising Role of the Transport Sector in Asia



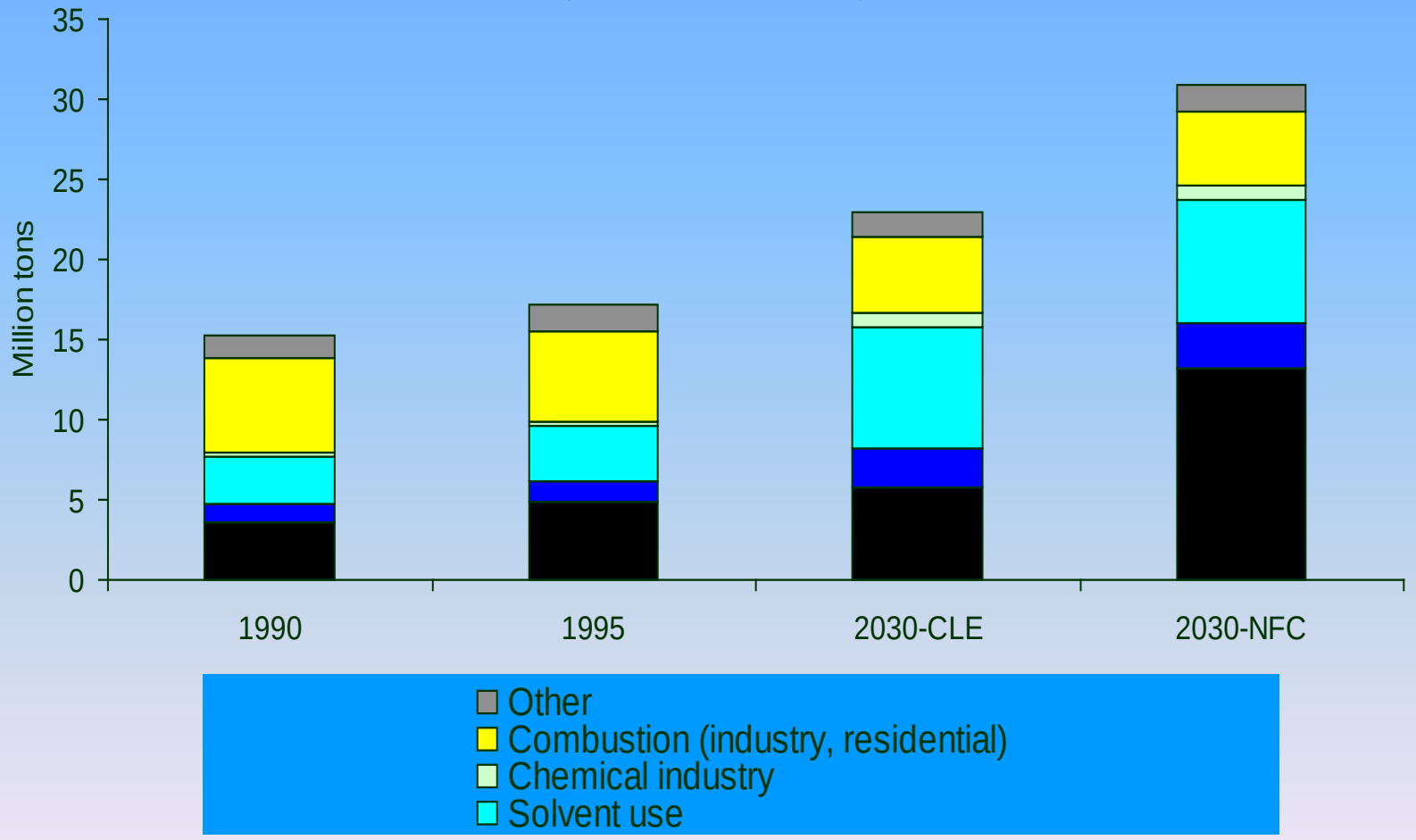
Van Aardenne, et al., Atmos. Env., 33: 633 (1999)

A NO_x projection for East Asia

Source: Klimont *et al*., IIASA

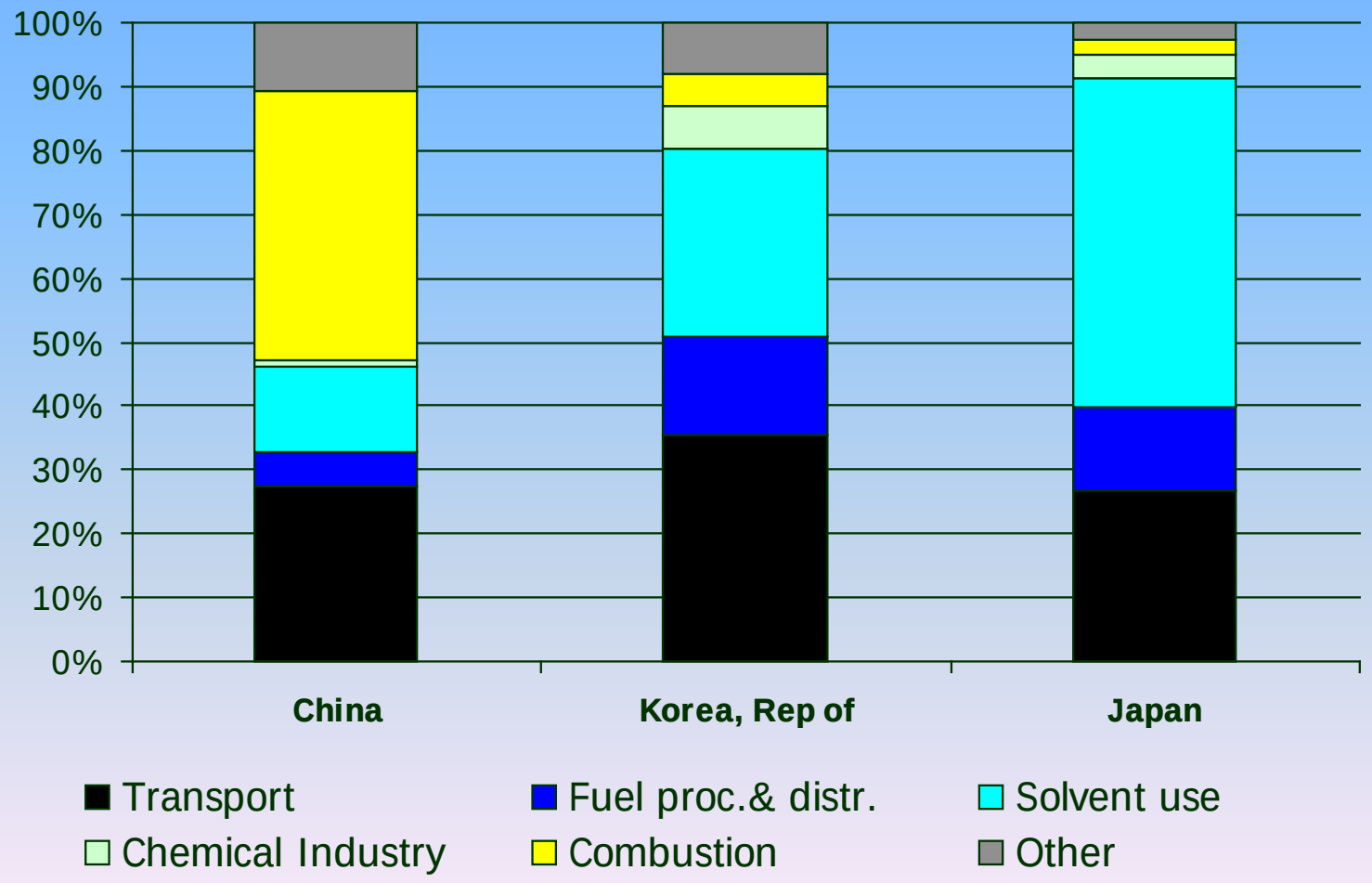


Emissions of VOC in East Asia
(Mt VOC)



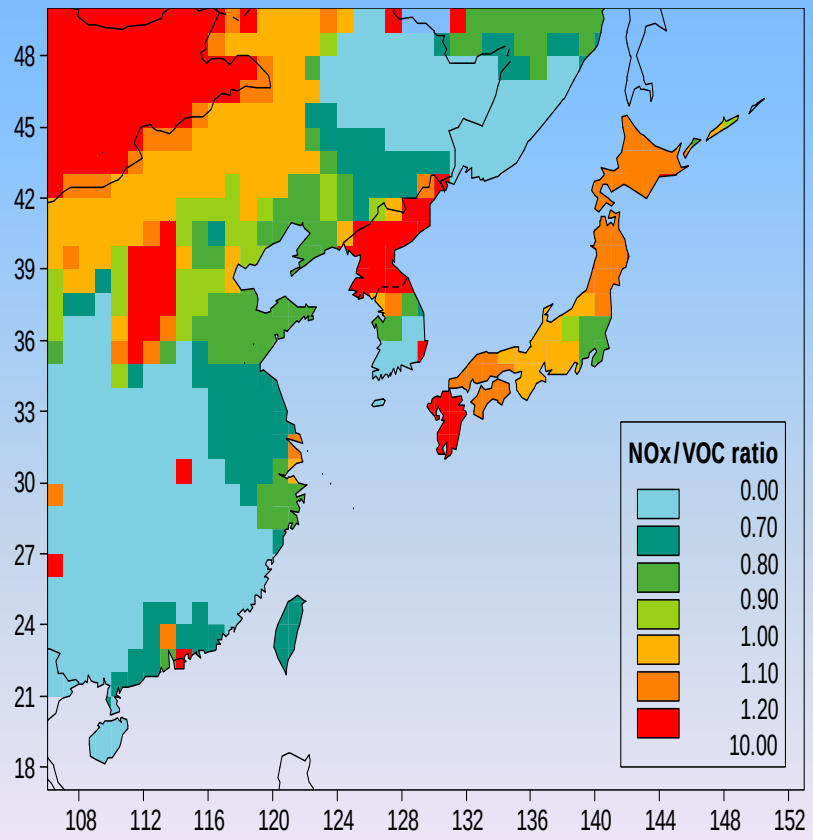
Structure of VOC emissions in 1995

for selected countries

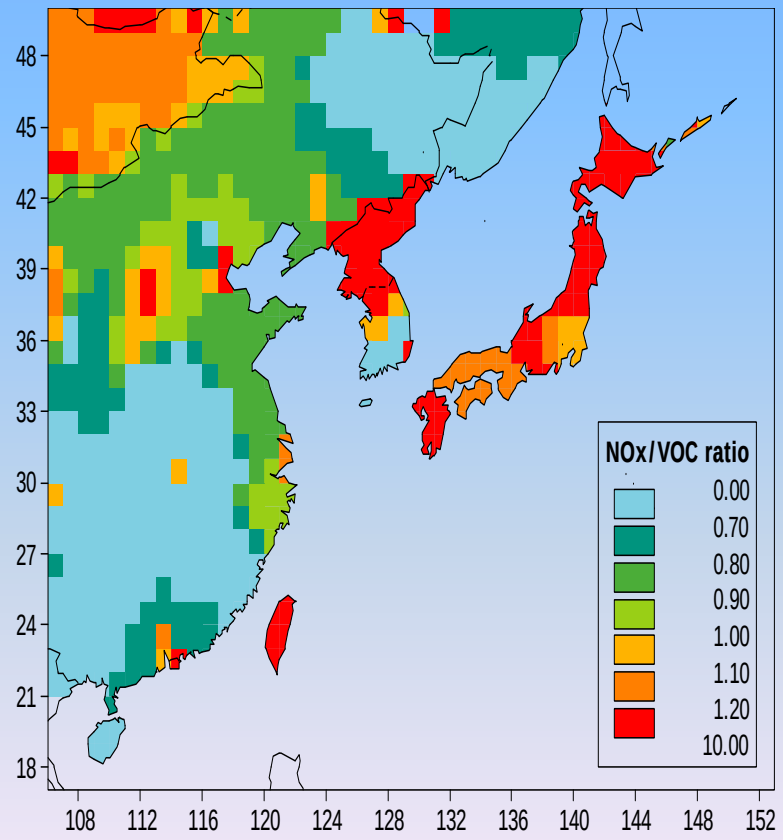


Ratio of NO_x to VOC emissions in East Asia

1995

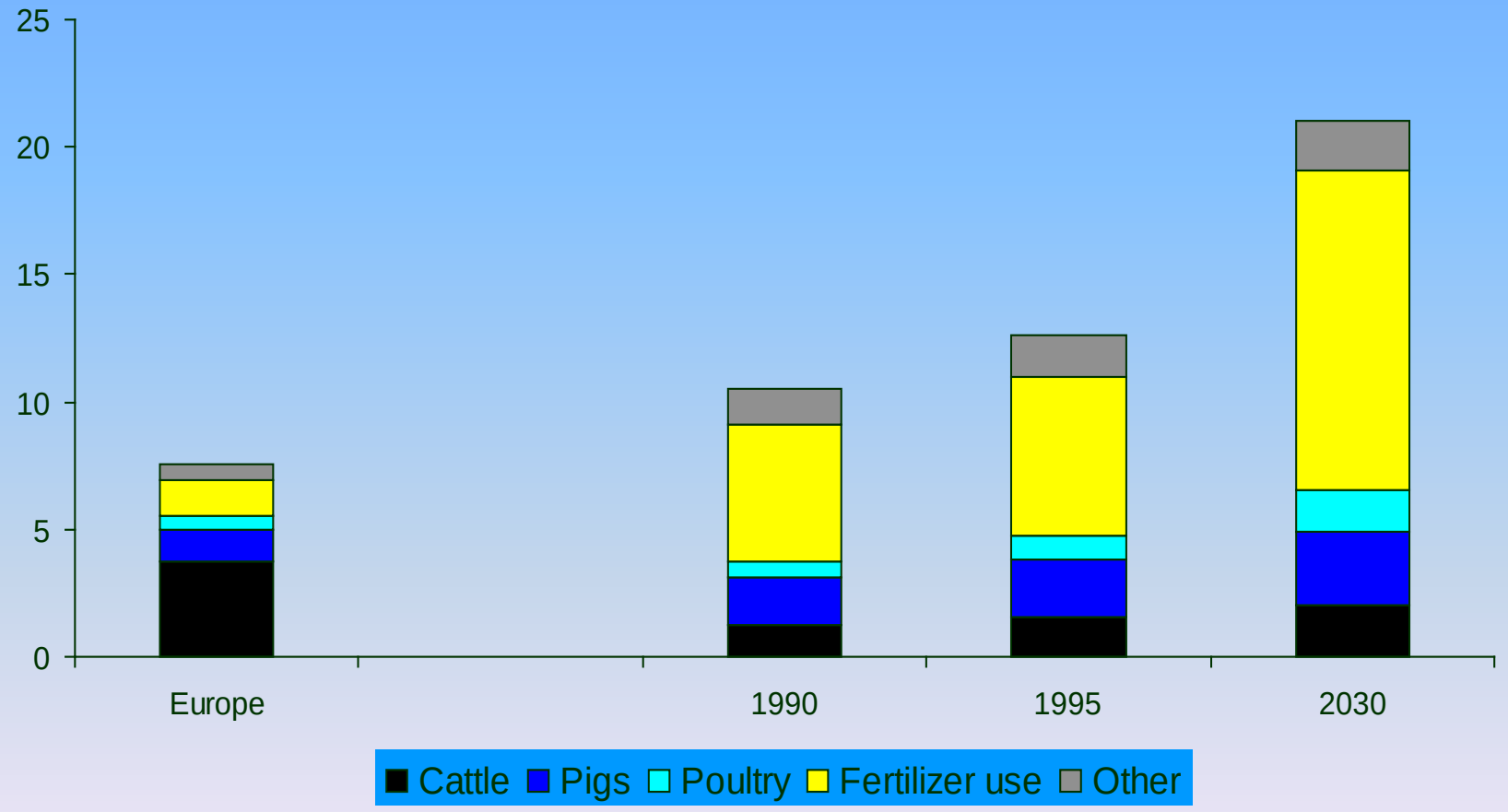


2030(CLE)

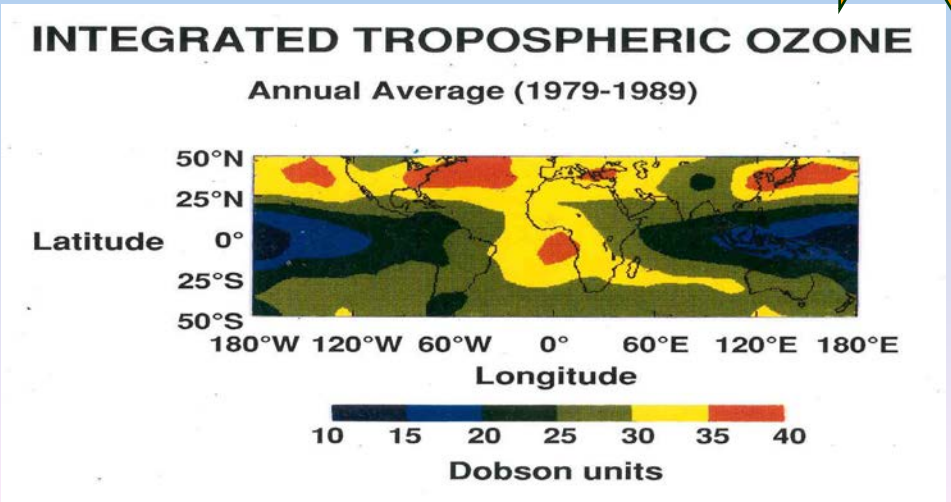


Emissions of ammonia in East Asia

(Mt NH_3)



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 retreval algorithms

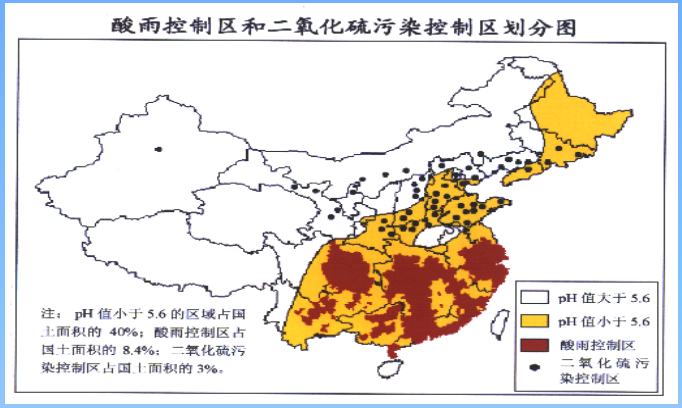
Utilities/tools for data mining and analysis for use by a broad community

Acknowledgements

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

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

Impacts Lead To Action



New Two-Zone Control Policy

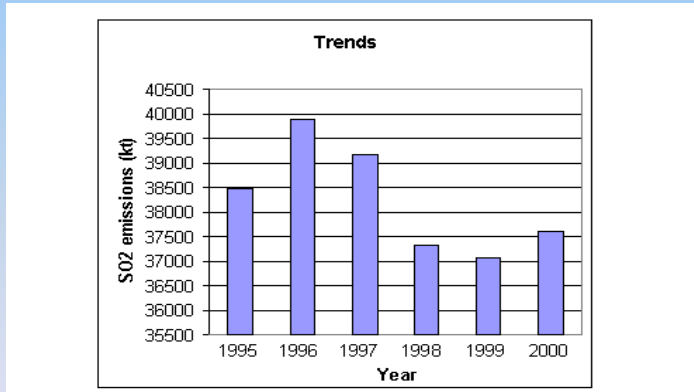


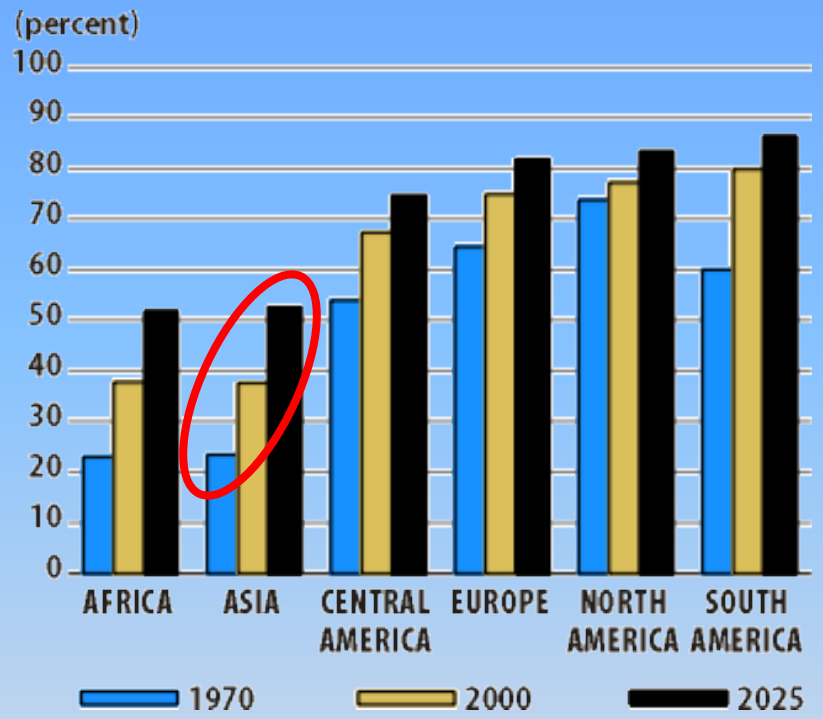
Figure 5. Estimated annual variation in Asian anthropogenic SO_2 emissions from 1995 – 2000.

China Emissions are changing in unanticipated ways!

Decreased Crop Yields

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

Urban Environments in Asia



- 4%/yr urban growth rate in Asia-Pacific region
- 23 megacities in 1995 (17 in developing countries) to 36 in 2025 (23 in Asia)

- 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 CO_2 every day (six times global per capita average)
- Indoor and Outdoor air pollution pose severe human health concerns