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

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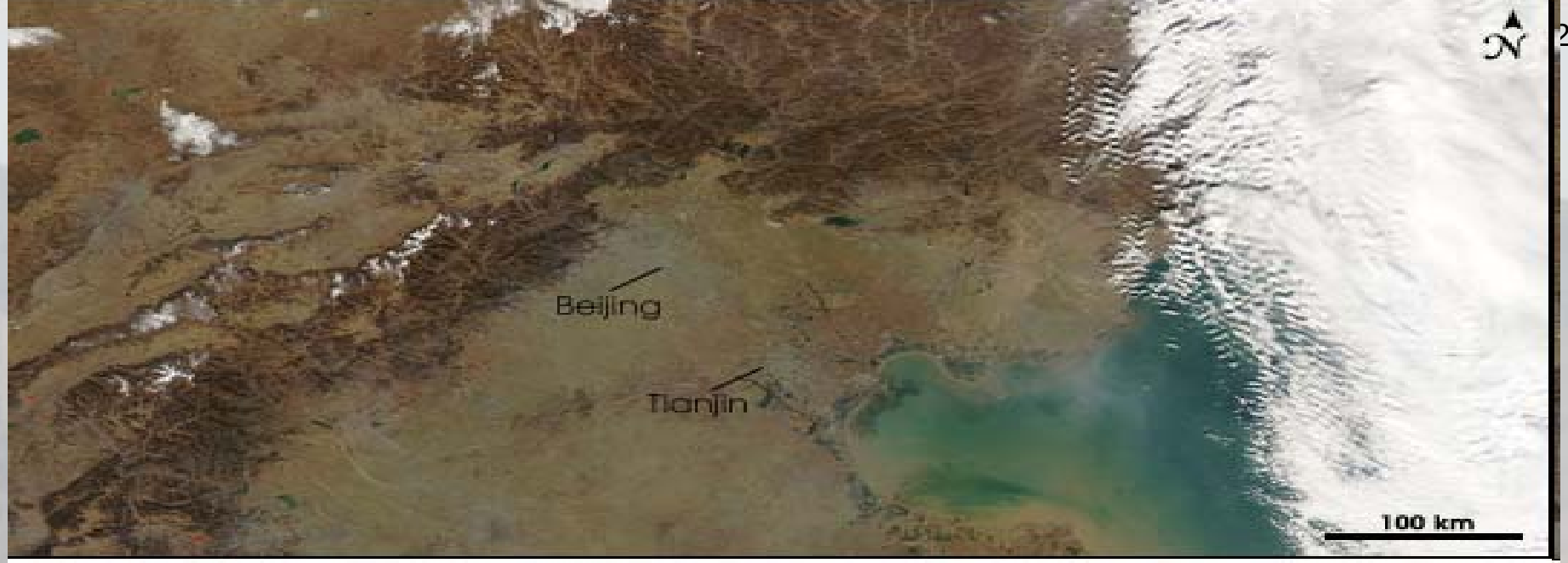
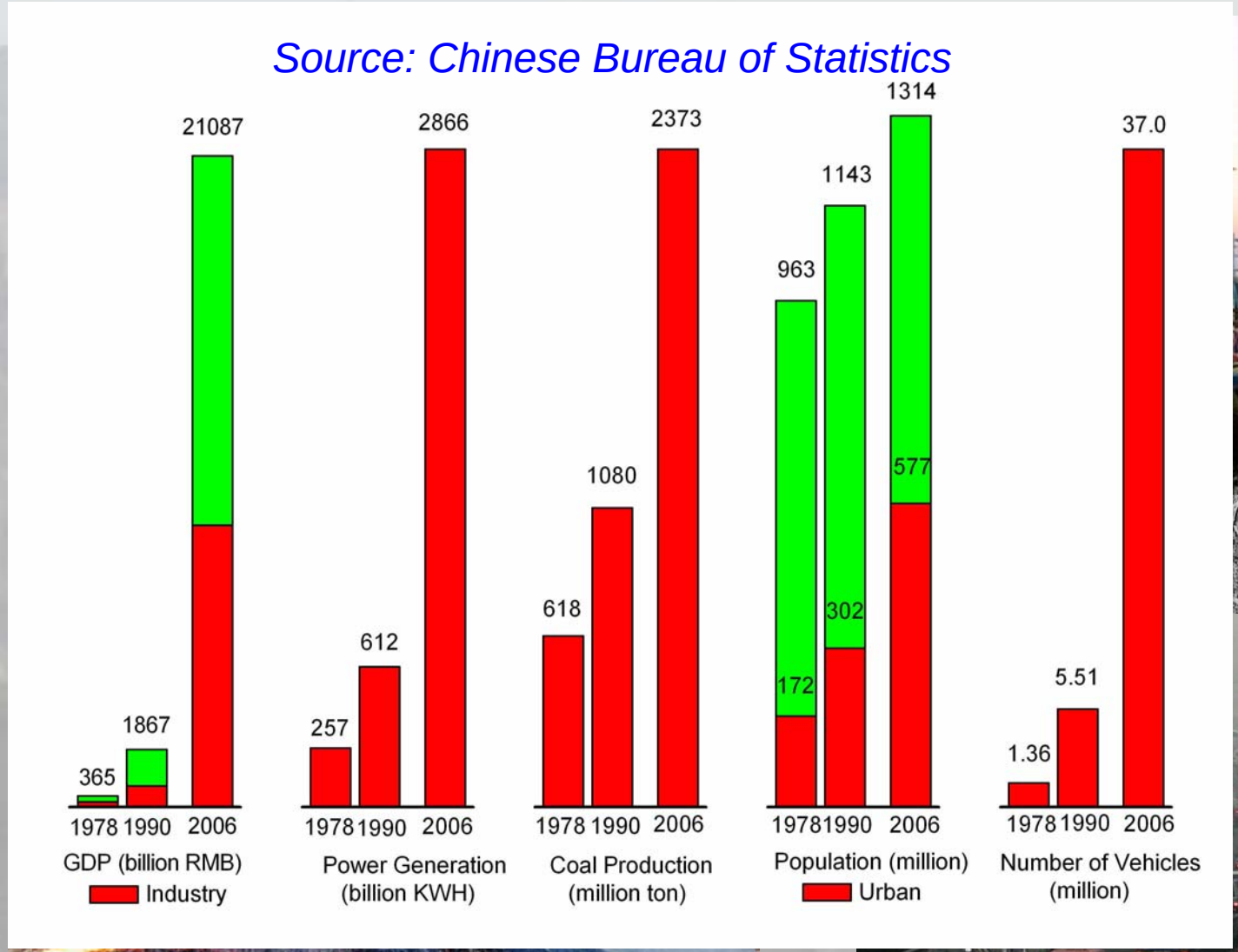
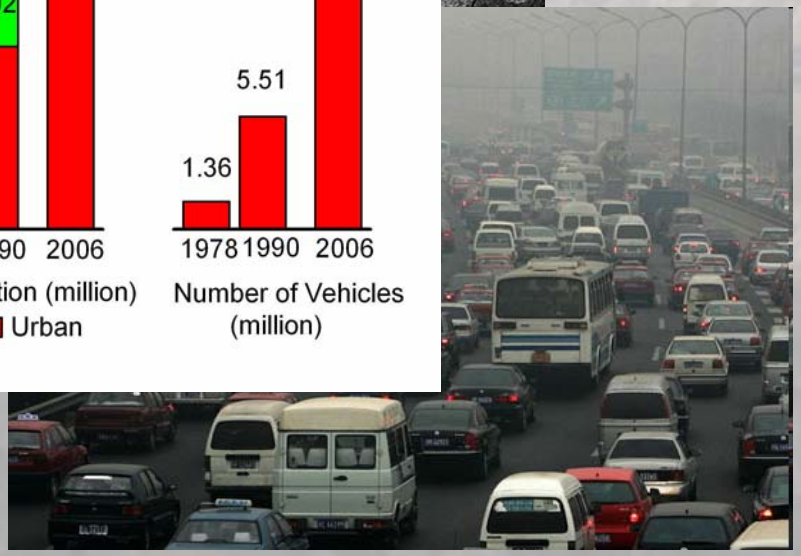


November 2, 2004 **Beijing AERONET AOD: 0.3 (440 nm)**

November 3, 2004 **Beijing AERONET AOD: 1.8 (440 nm)**

Industrialization and Urbanization





- 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?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Archive for Tuesday, March 06, 2007

Asian air pollution affects our weather

Scientific Questions and Approaches

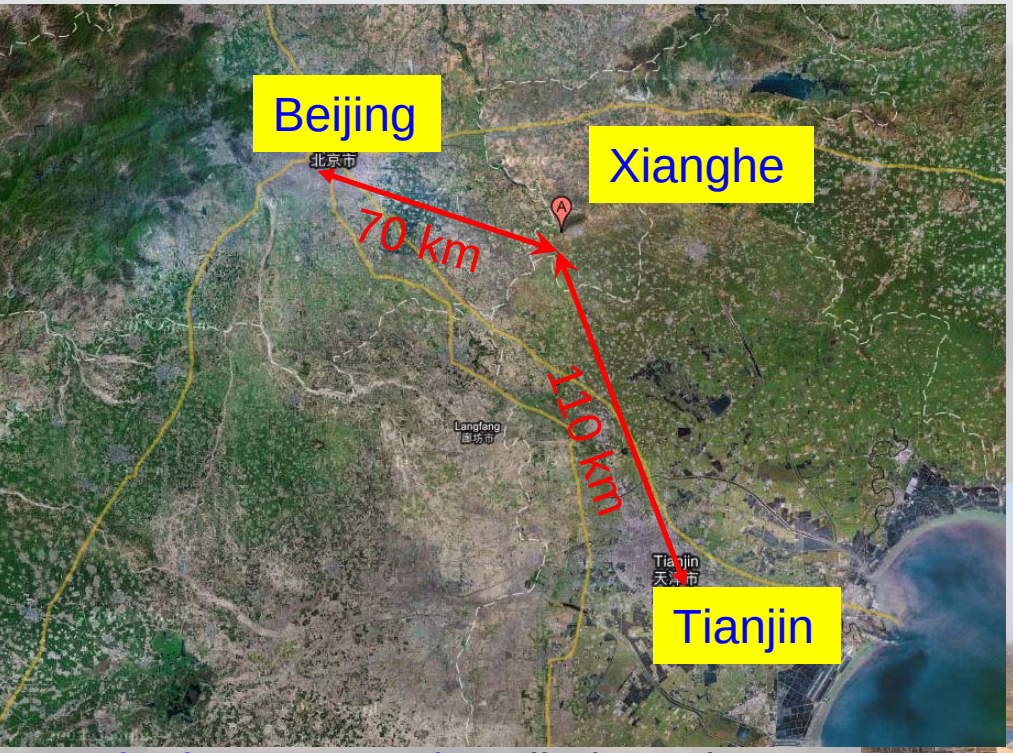
- Amount of pollutants emitted – emission inventories for China: how good are they? - Ground measurements near source regions

- 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.

Xianghe Ground Experiment



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)

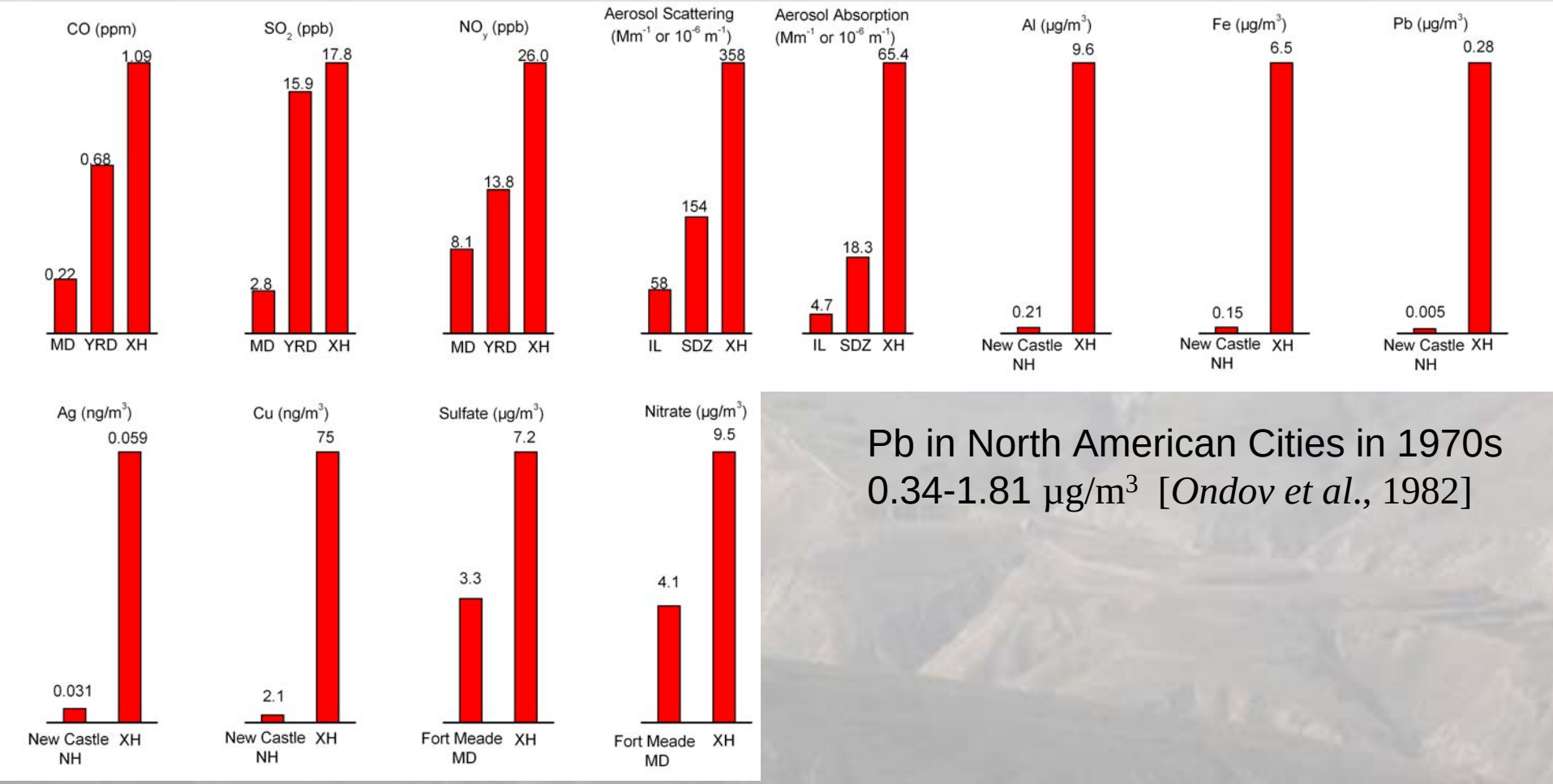
Mixed agricultural land, residence, and light industry

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

First intensive field campaign of EAST-AIRE (March, 2005): Trace gases (O_3, CO, SO_2, NO_y), aerosol scattering and absorption, aerosol chemical composition.


WEST
EAST
07.03.2005 08:05

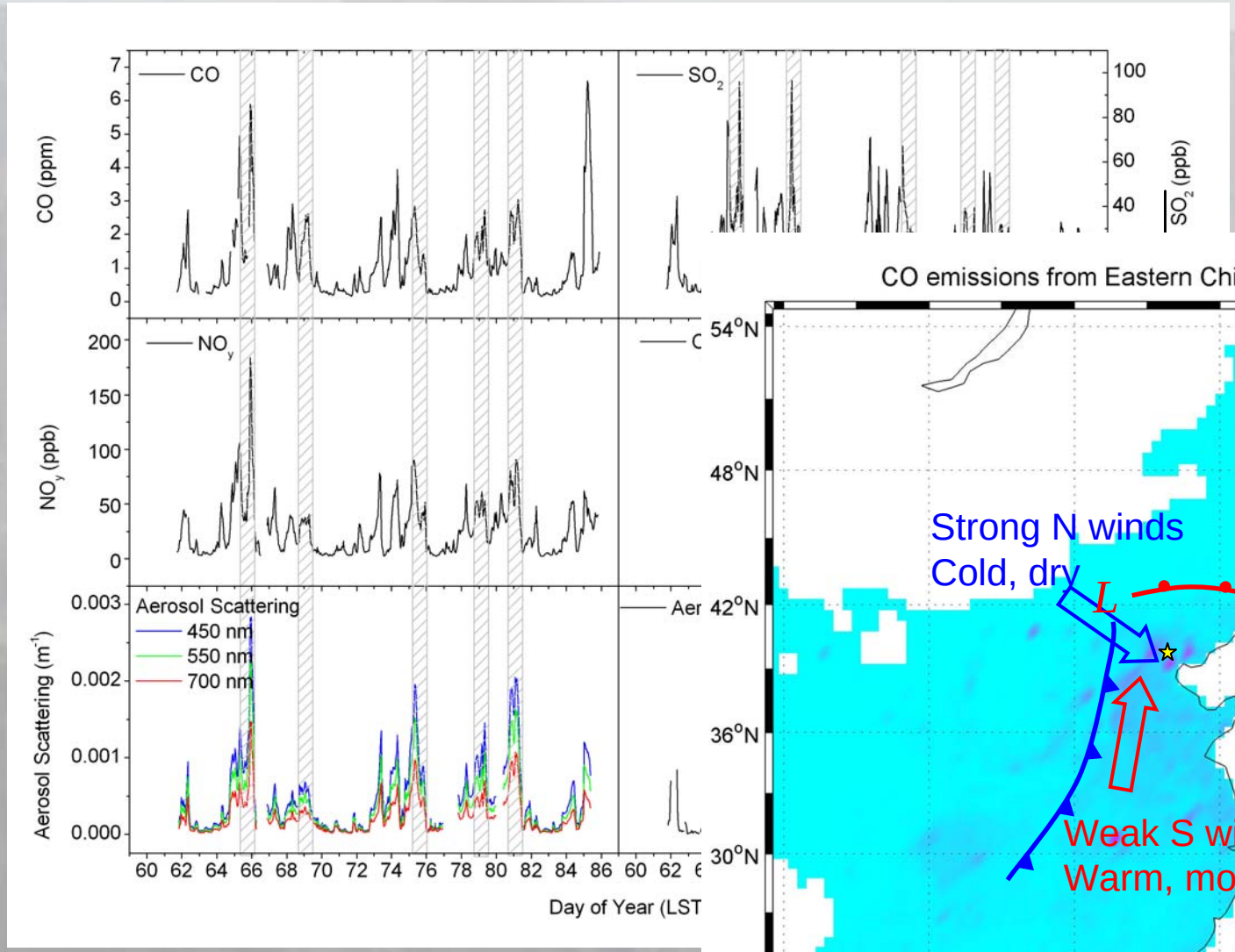
Overall, Polluted

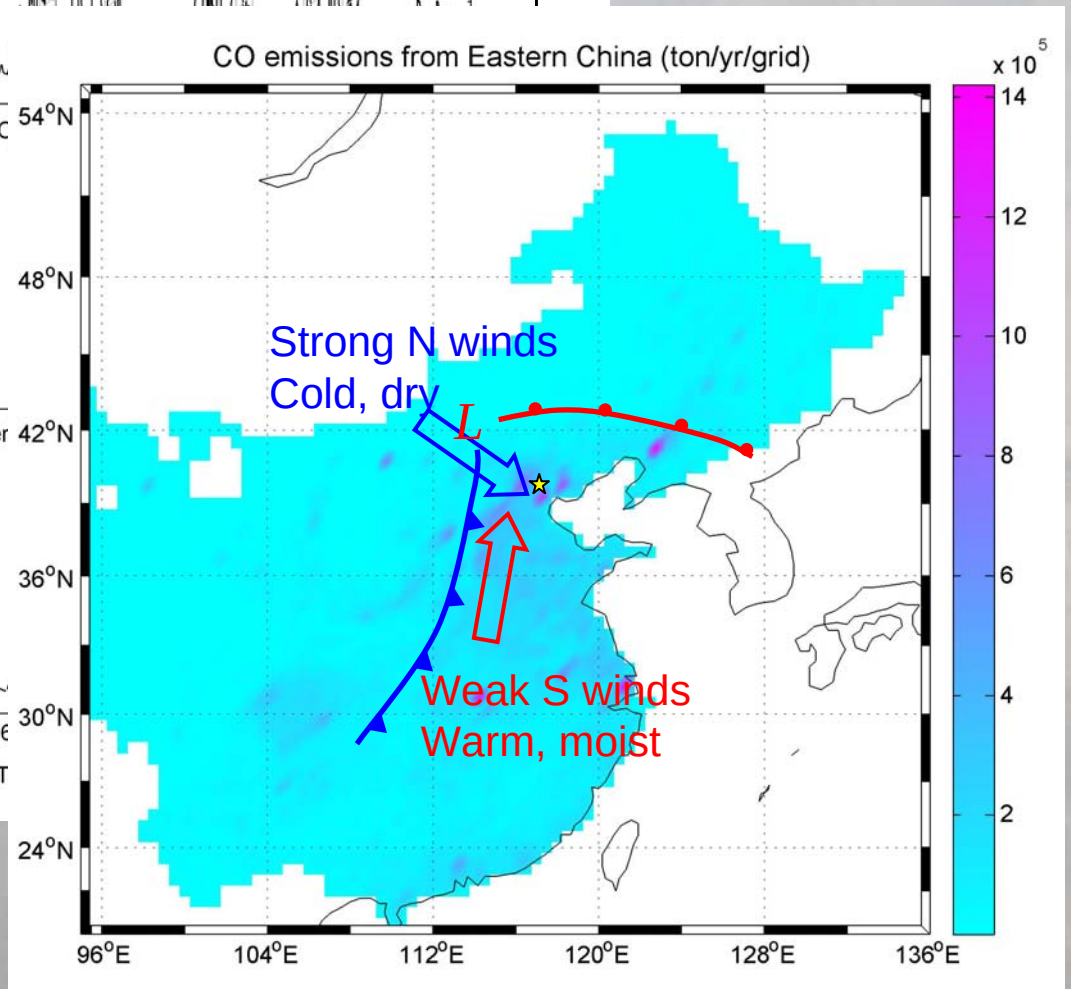


Pb in North American Cities in 1970s
0.34-1.81 $\mu g/m^3$ [*Ondov et al*., 1982]

"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)

	This study	*Wen et al.* [2008]	*Streets et al.* [2003]	*Streets et al.* [2006][a]	*X. Wang et al.* [2005][b]
CO/SO_2	35.8	72.1	13.0	17.5	7.7
CO/NO_x	27.2	25.4	16.7	22.7	8.6

[a] Use SO_2 and NO_x emission data from *Streets et al.* [2003].

[b] Emission inventory is regional.

Large uncertainties in emission inventories? CO underestimated? SO_2 overestimated? 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.

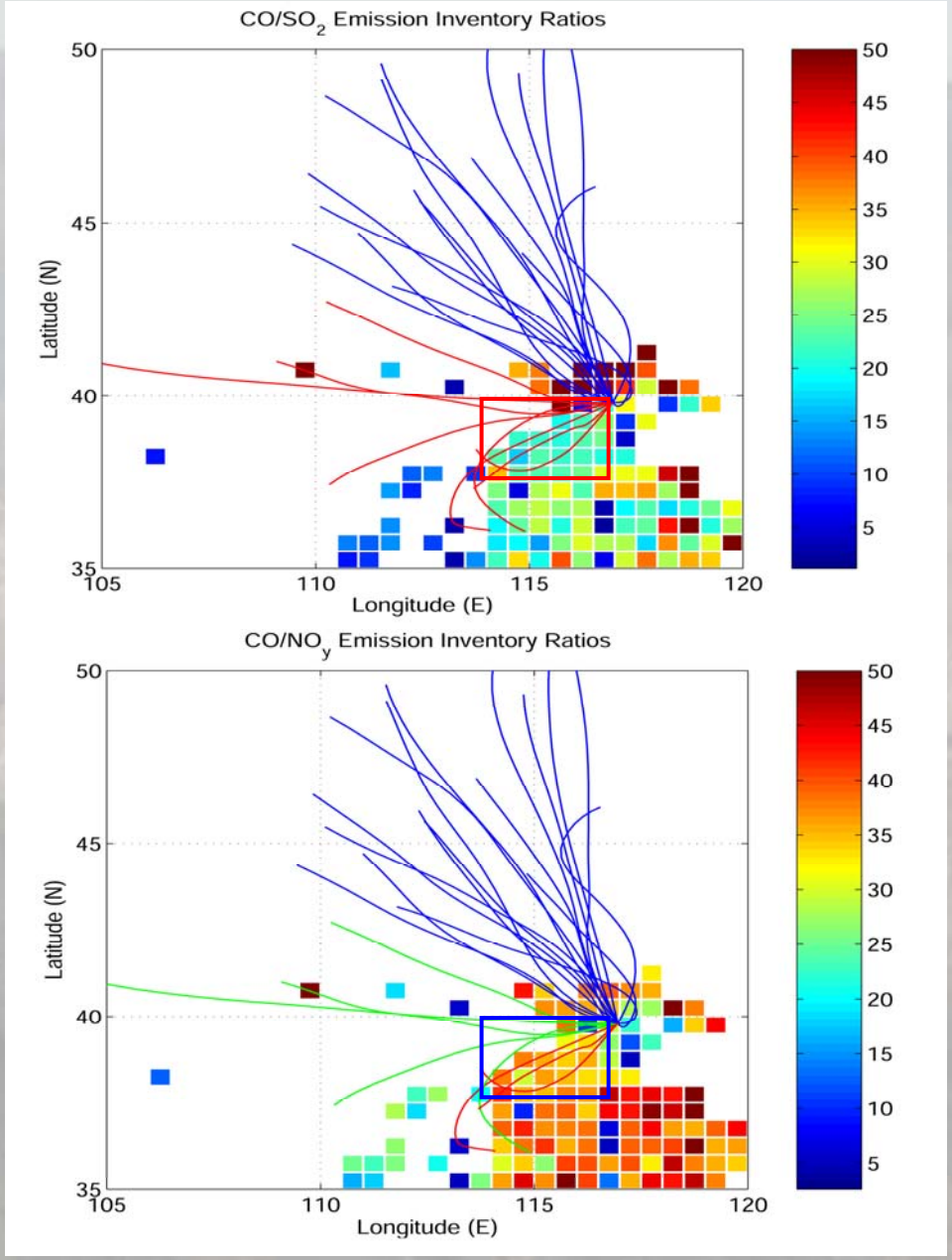
Spatial Distribution of Pollutant Ratio

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/SO_2: 37 vs. 10-17
CO/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…

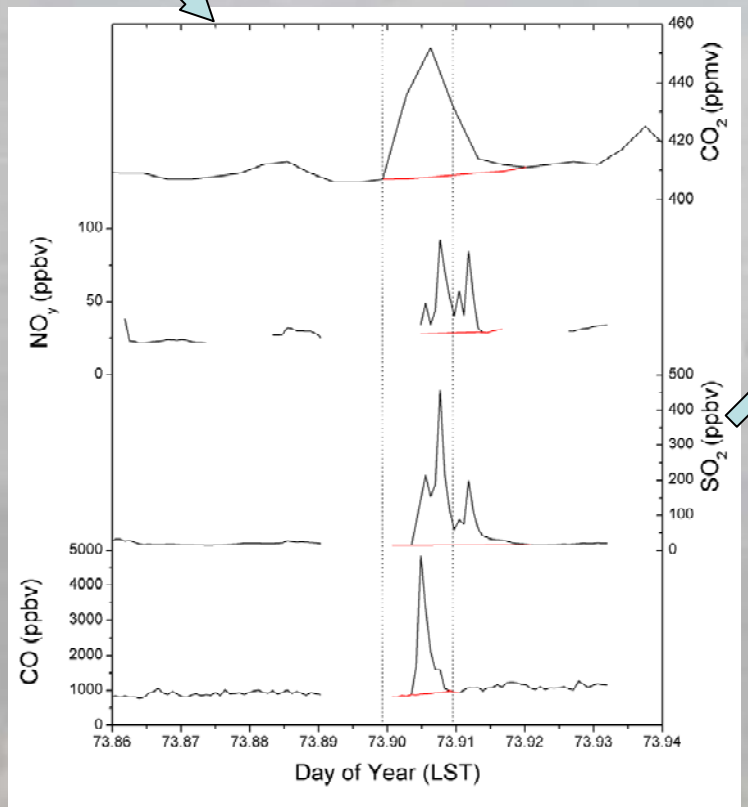
Small boilers? Traffic? Biofuel?



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 SO_2 concentration.



Correlation analysis derived emission factors and inventory values:

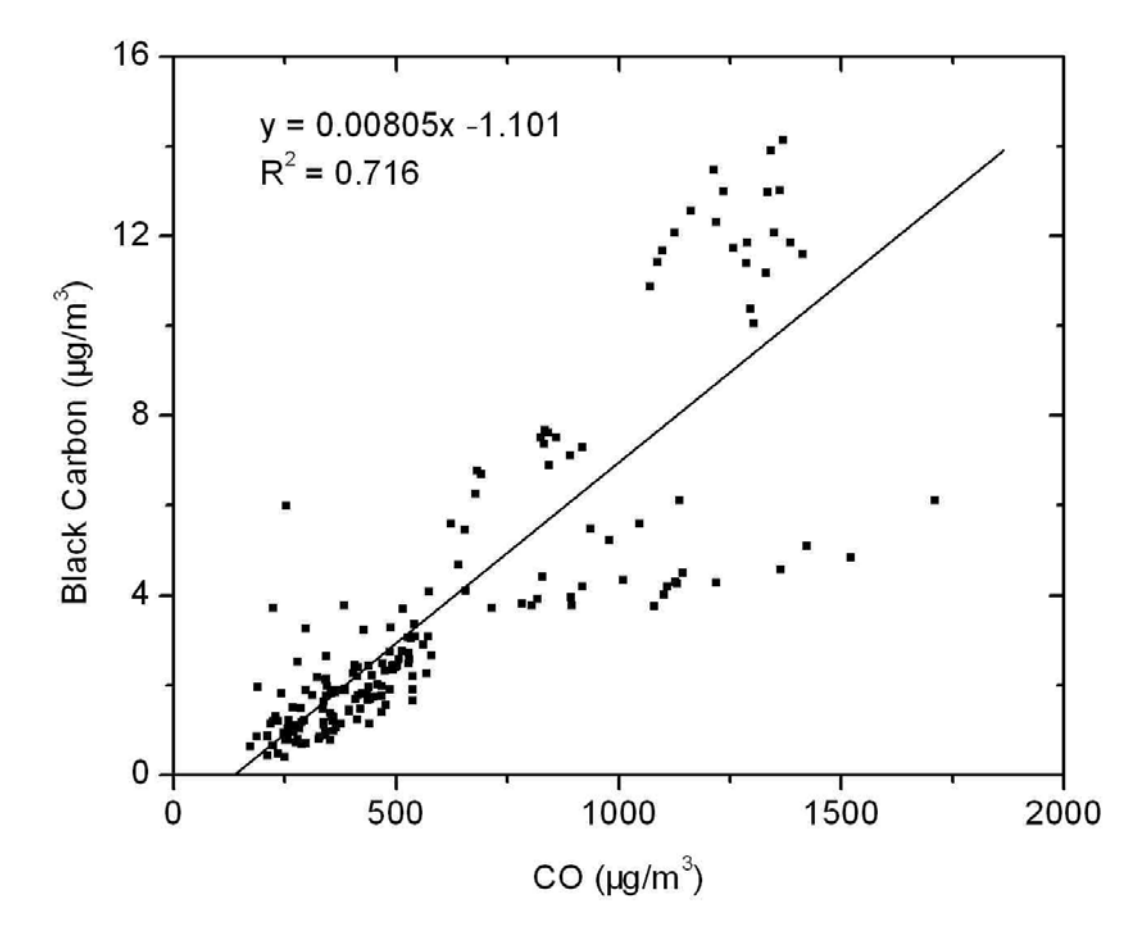
SO_2: 15.4 g SO_2/kg vs. 3.29-54.8 g SO_2/kg in inventory [*Streets et al.*, 2003];

NO_x: 1.8 g NO_2/kg vs. 1.19-2.24 g 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]

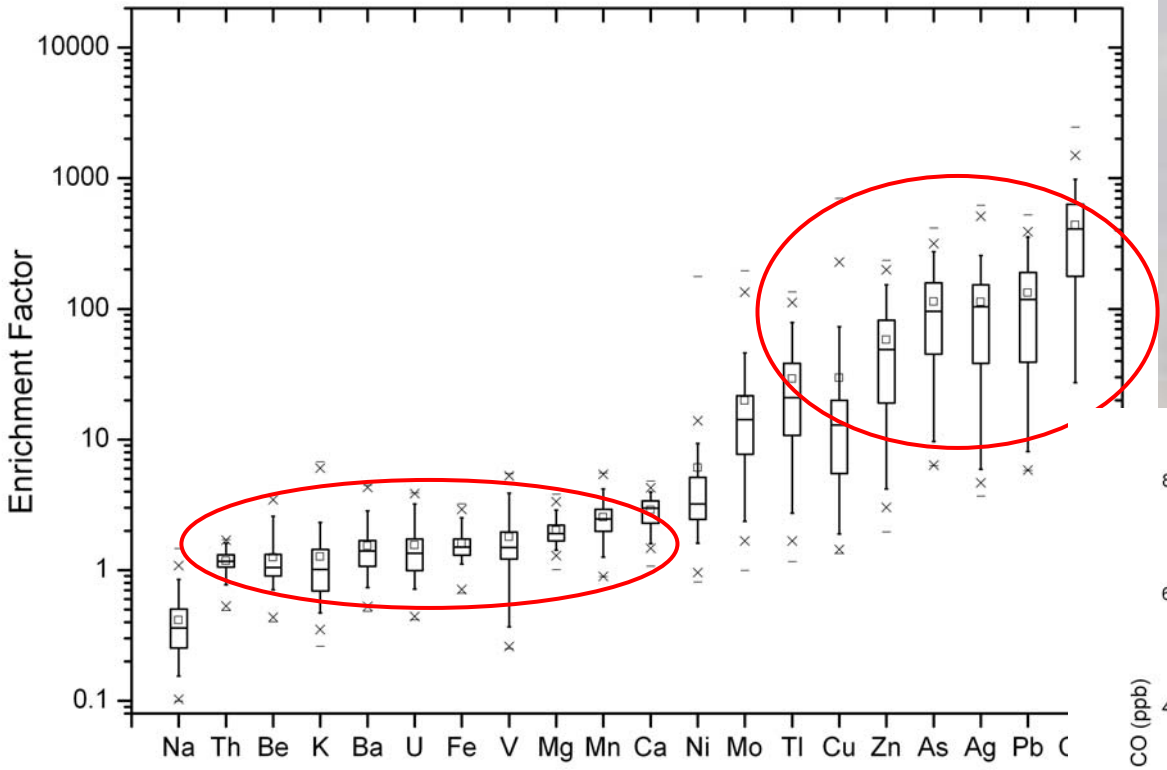
Better agreement compared to a similar study for India [*Dickerson et al.,* 2002]

Single Scattering Albedo (SSA): Morning: ~0.81, Afternoon: ~0.85

Source of Aerosols: Elements

Calculation of enrichment factor:

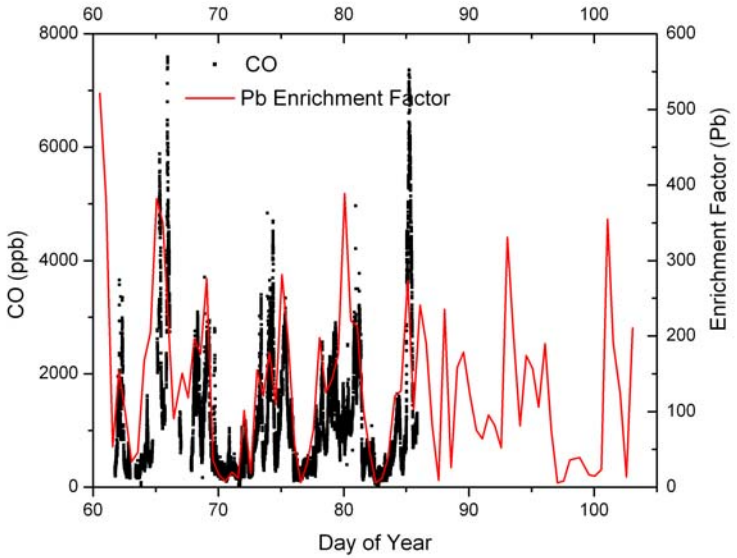
$$EF_X = \left(\frac{X}{Al}\right)_{Aerosol} \Big/ \left(\frac{X}{Al}\right)_{Crust}$$



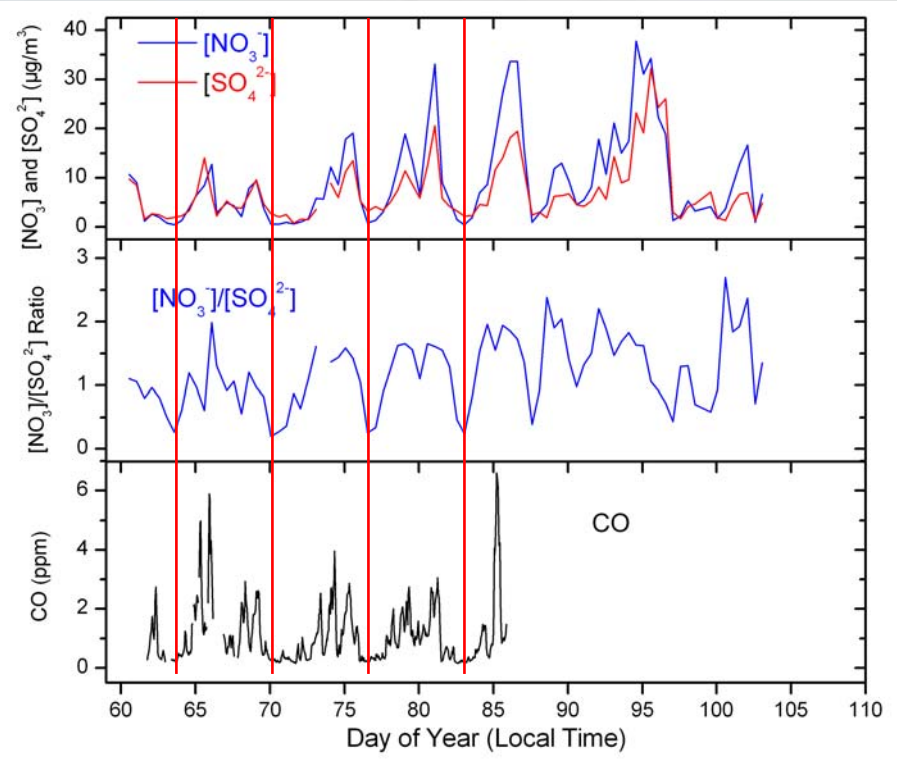
Abundant crustal elements: soil particles

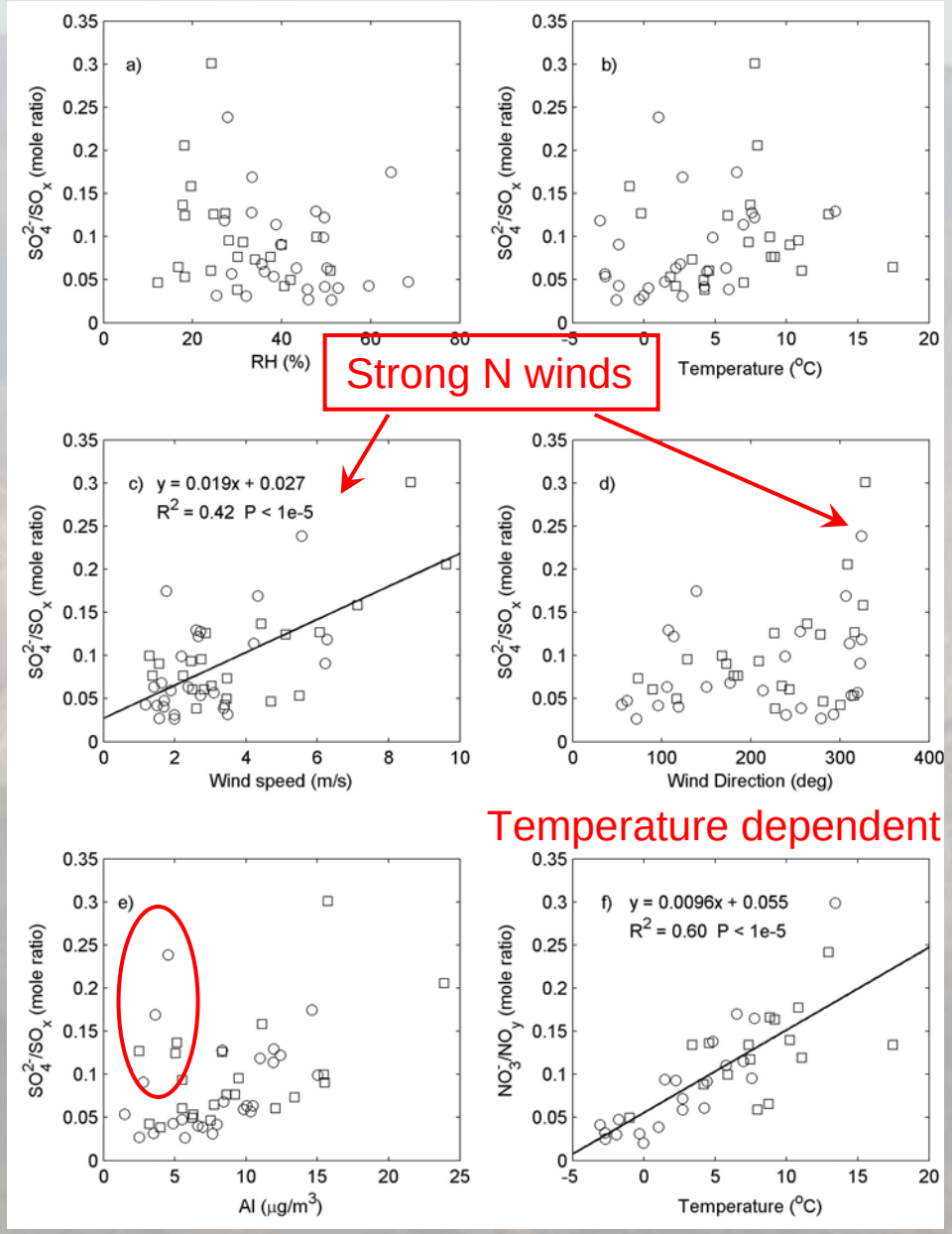
Enriched pollution tracer elements: anthropogenic emissions

Both soil and anthropogenic sources



Source of Aerosols: Secondary Species





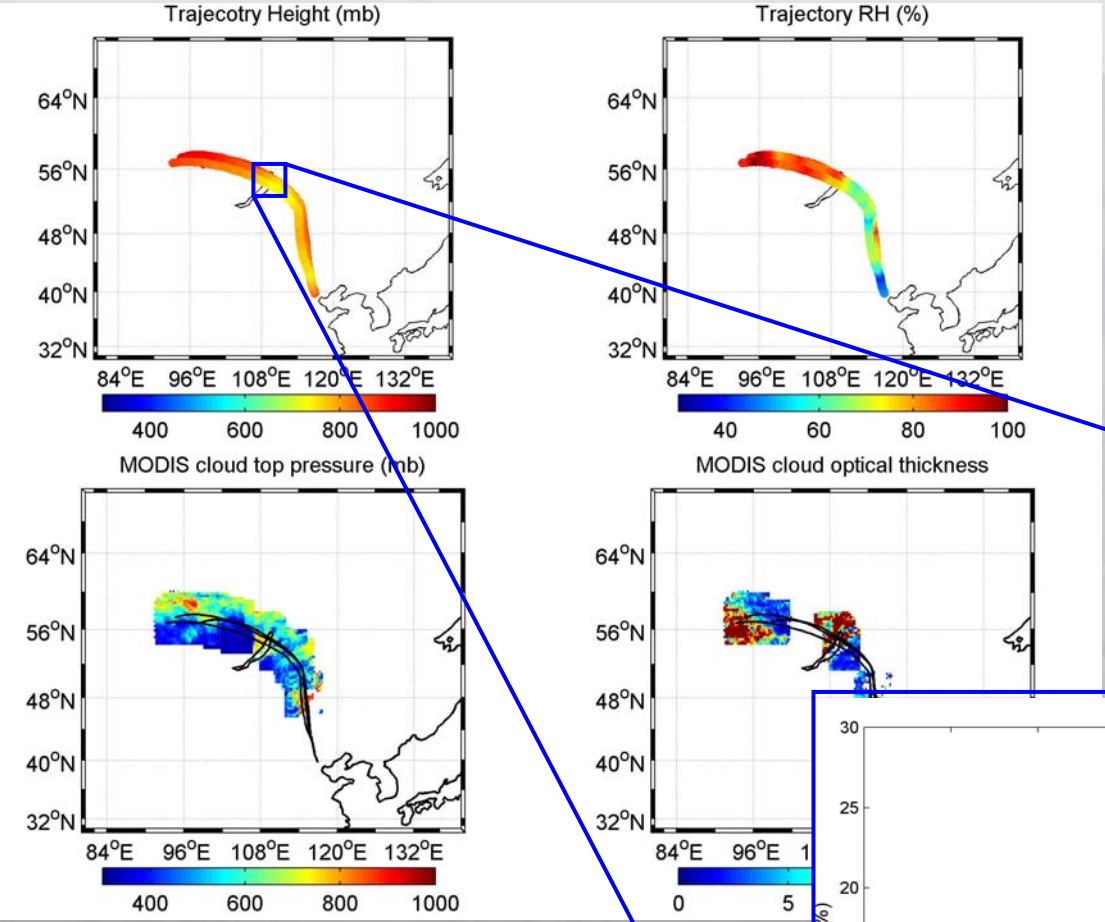
Synoptic change in nitrate/sulfate ratio

$SO_x = SO_4^{2-} + SO_2$, SO_4^{2-}/SO_x and NO_3^-/NO_y reflect the fraction of SO_2 and NO_x oxidized

NO_3^-/NO_y: mainly through gas-phase

SO_4^{2-}/SO_x: some other mechanism …

Source of Aerosols: Secondary Species

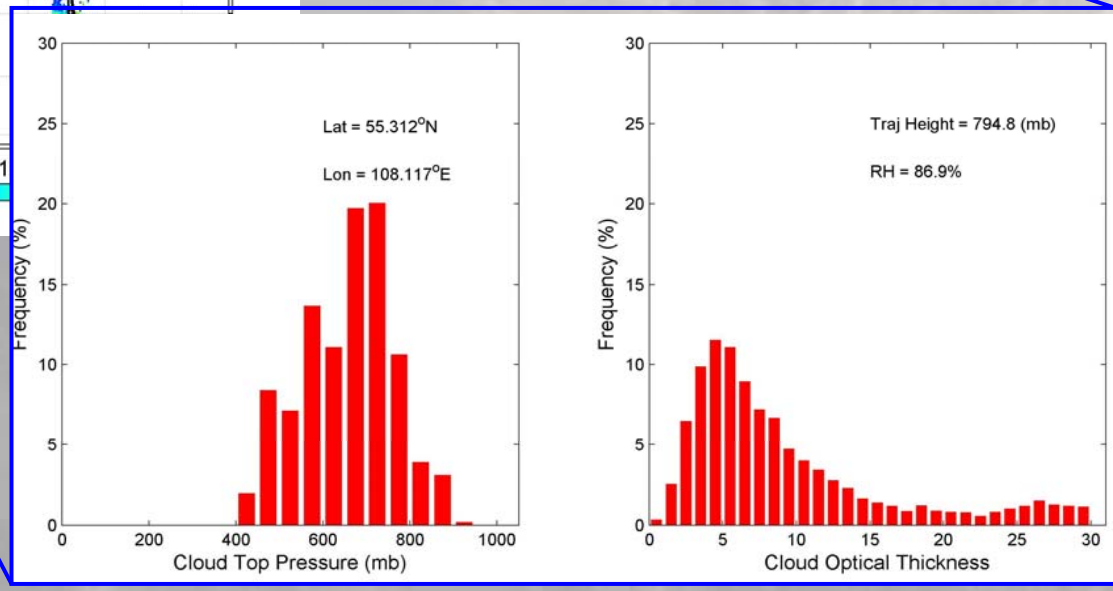


Backward trajectories (200, 500, and 1000 m above ground) and EOS/MODIS satellite cloud product synchronized in time and space (± 15 min and $\pm 2.5^\circ$ lat/lon)

Cloud interaction?

Trajectory RH $\geqslant$ 80%

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

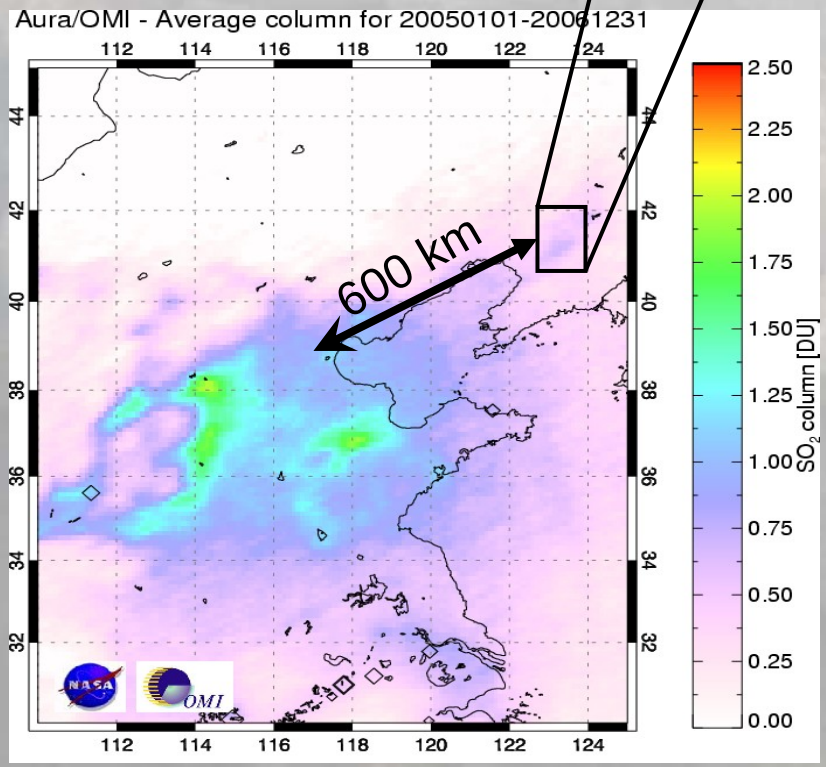


~70% of samples with relatively high SO_4^{2-}/SO_x (or low NO_3^-/SO_4^{2-}) are possibly related to cloud interaction. In-cloud processing of SO_2 could be important during transport behind cold fronts.

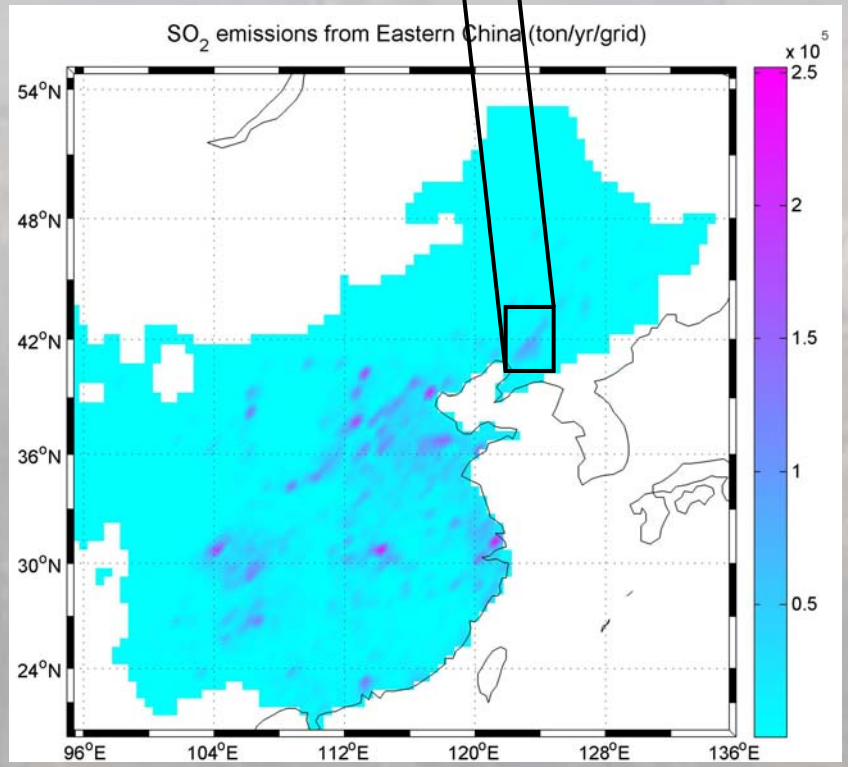
Aircraft campaign

Location: Shenyang;
~600 km NE of Beijing;
~6 million people;
Industrialized region;
April 1-12, 2005, 8 flights

DU: Dobson Unit: 2.69×10^{20} molecules/m^2

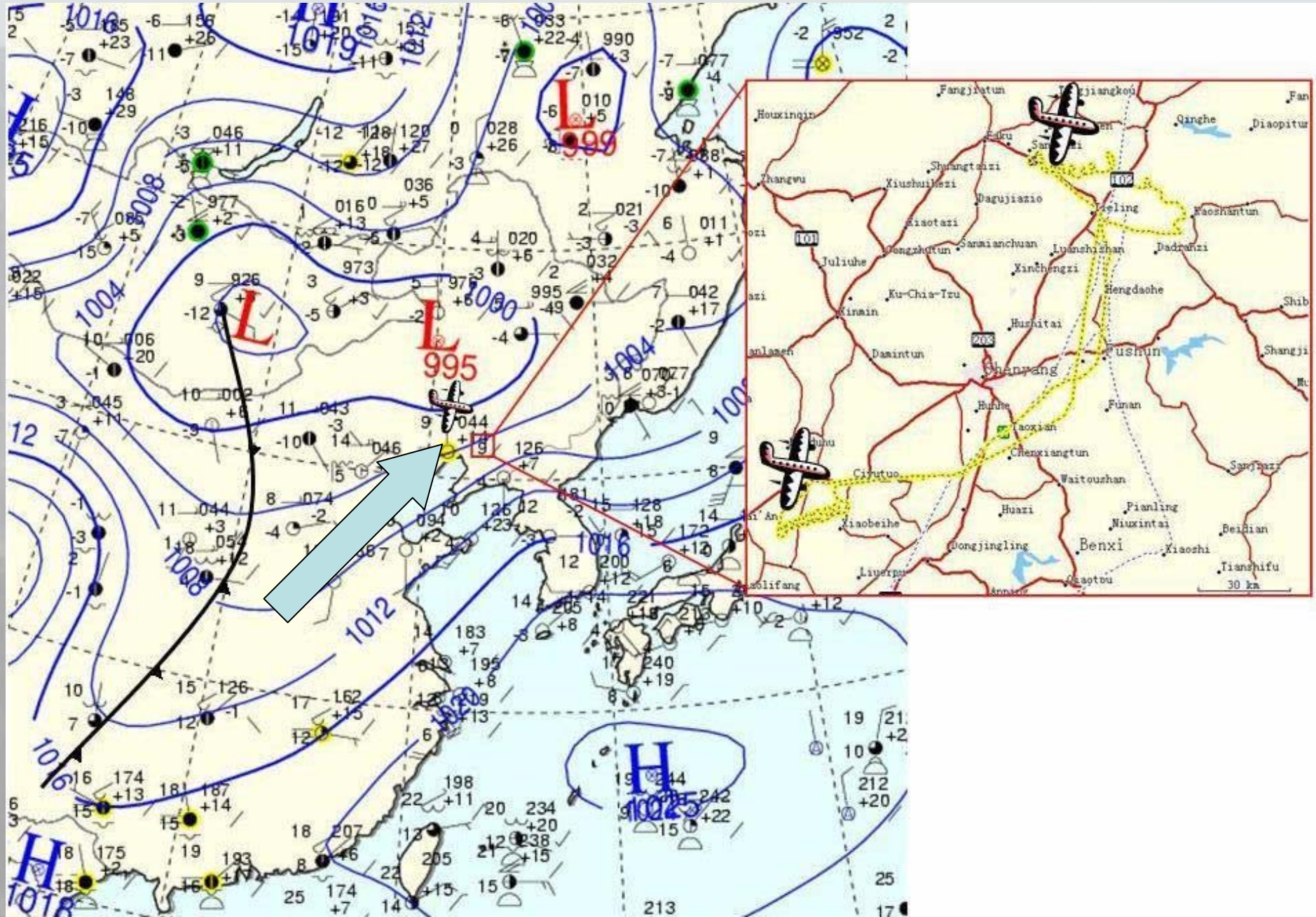


Aura/OMI SO_2 (2005-06) [*Krotkov et al.*, 2007]

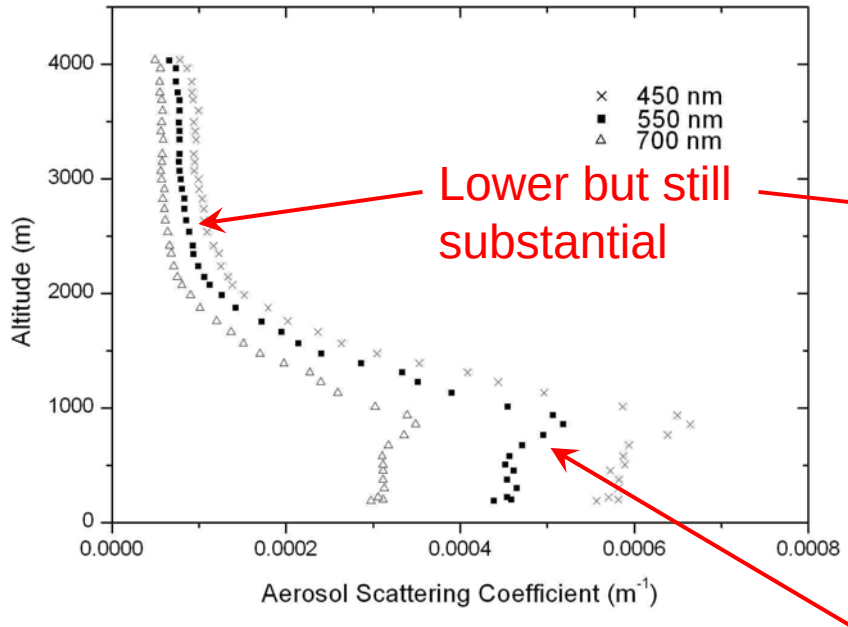


SO_2 emissions [*Streets et al.*, 2003]

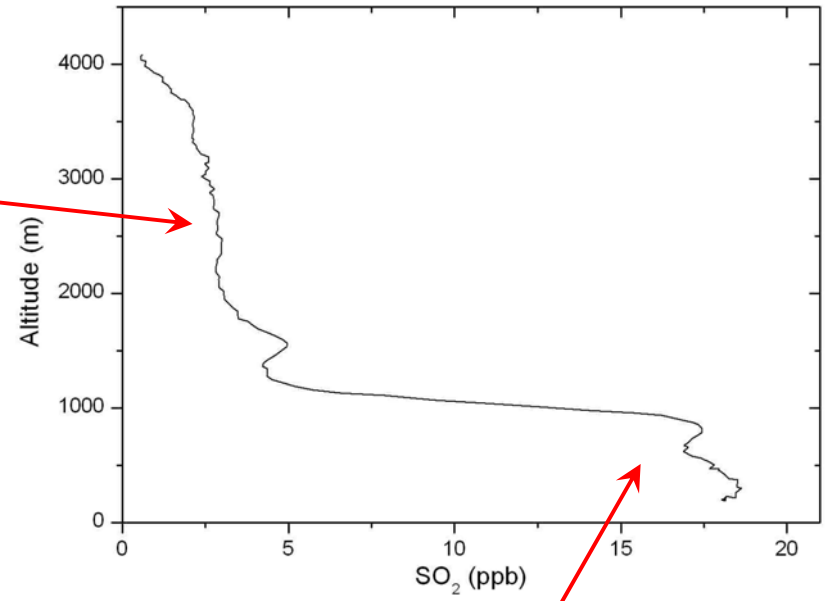
April 5, 2005: Flight Ahead of Cold Front

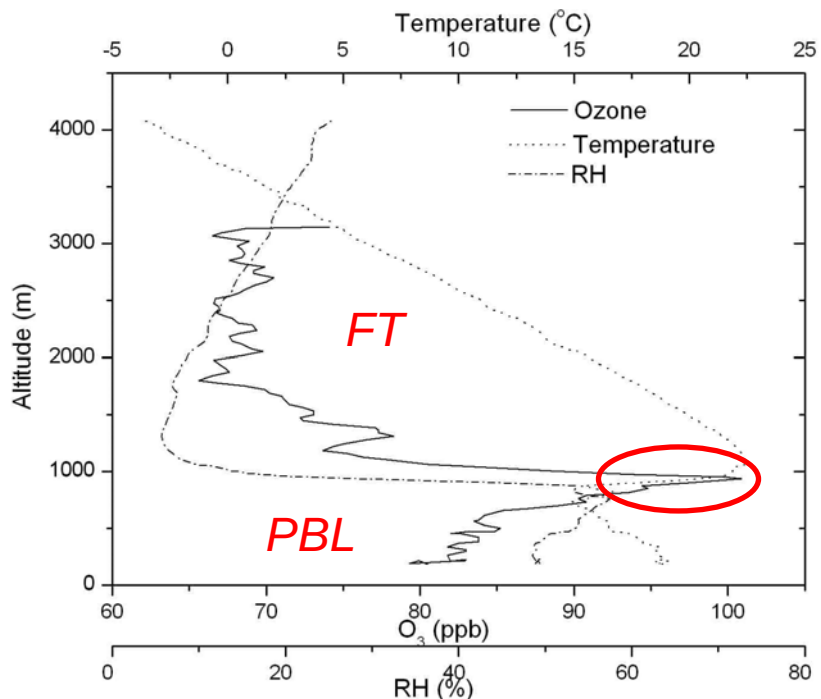


Vertical Profiles of Trace Gases and Aerosols



Lower but still substantial

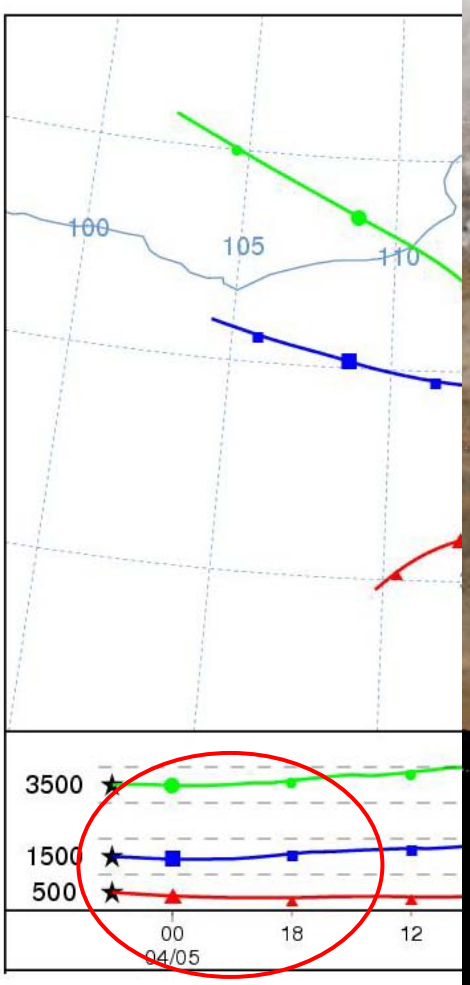


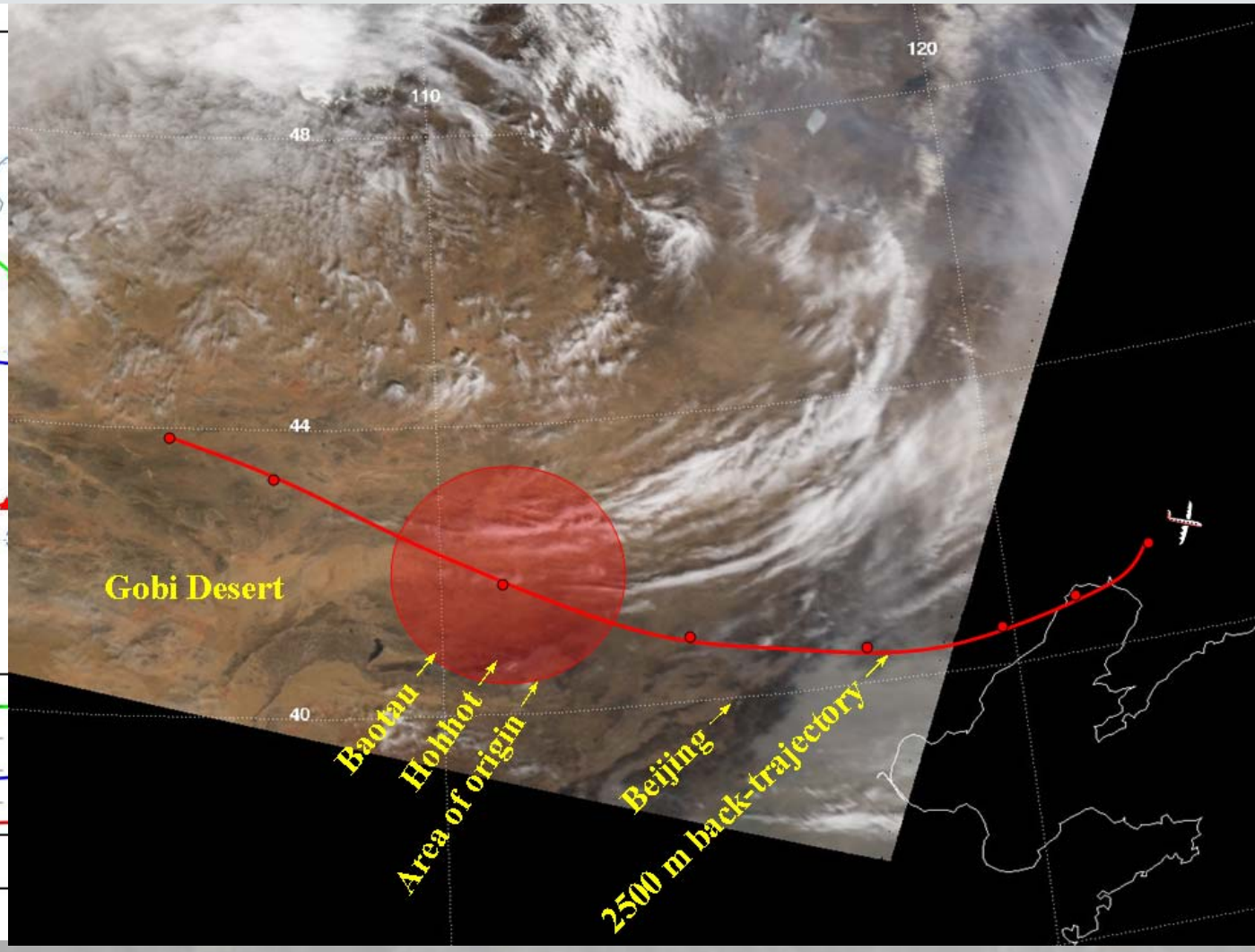


Polluted PBL: almost 20 ppb SO_2 and 500 Mm^{-1} aerosol scattering – SW flow from polluted region in the lower atmosphere, also dust emissions due to strong surface winds.

Pollutants in FT may travel far downwind: *the lofting mechanism ?*

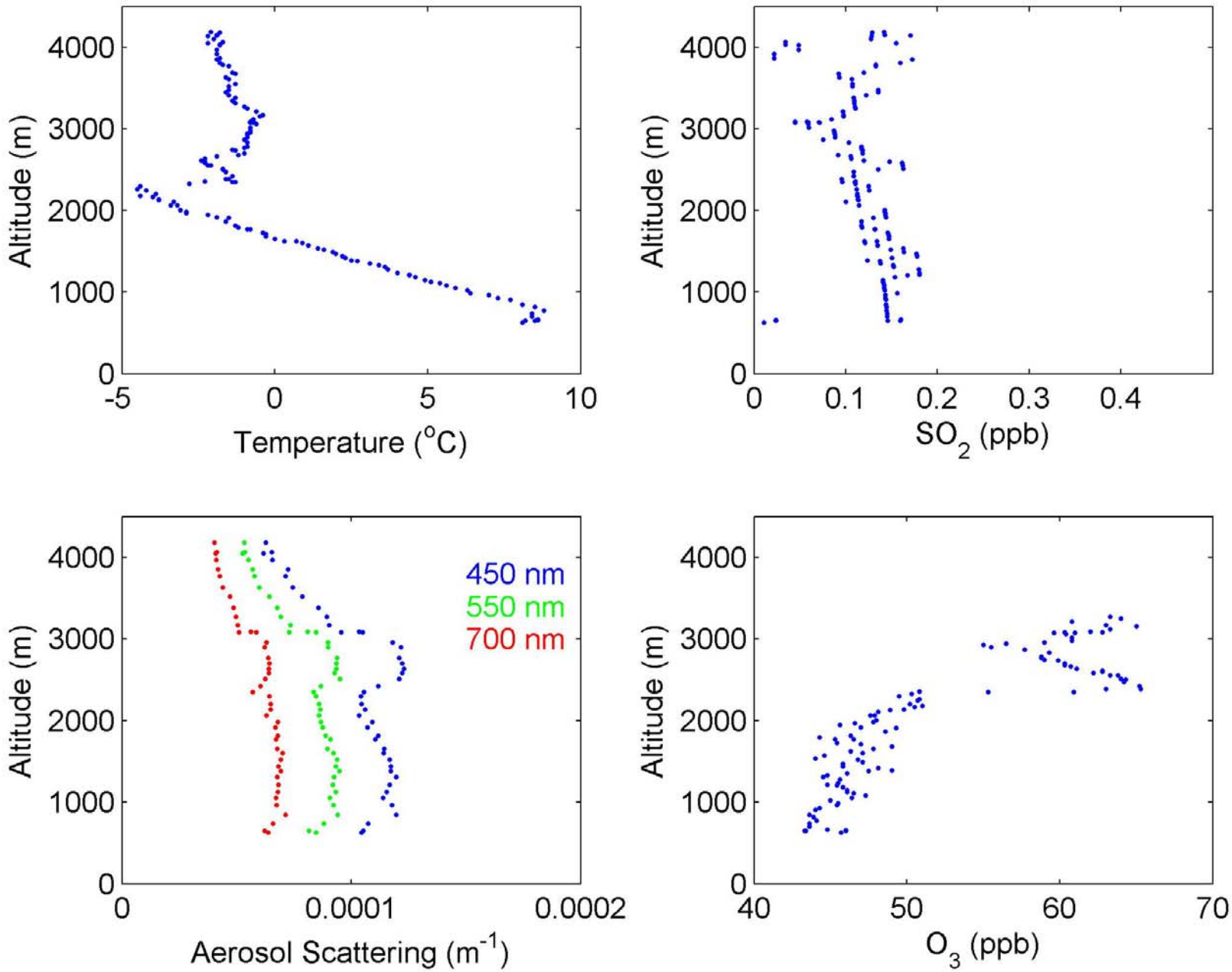
Lofting by Convective Clouds over Upwind Industrial Region

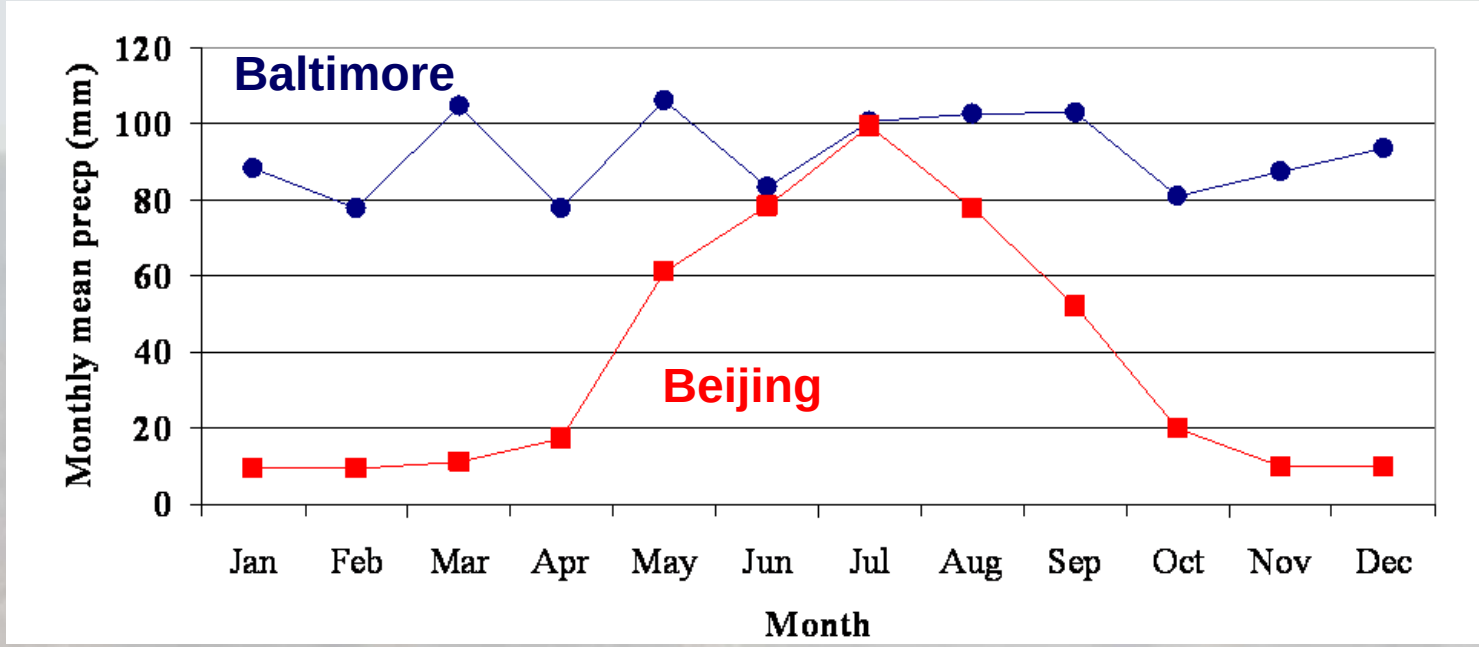




MODIS true color image for 04 UTC April 4, 2005 with 2500 m back trajectory superimposed (no precipitation observed on April 4 over the industrial region marked by red disk).

April 7, 2005: Behind Cold Front





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.

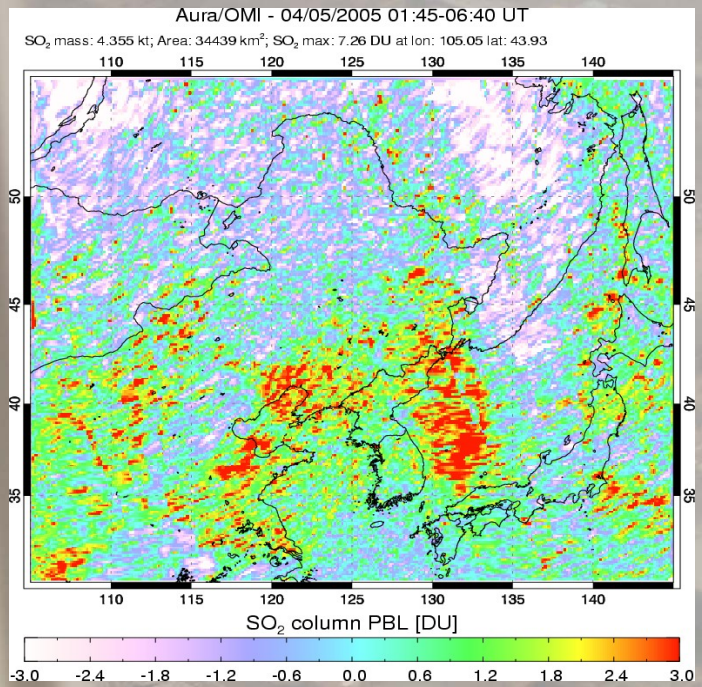
Dry convection can be very important in exporting pollutants from China, but needs more flights and numerical simulation.

Observe the April 5 episode at larger scale?

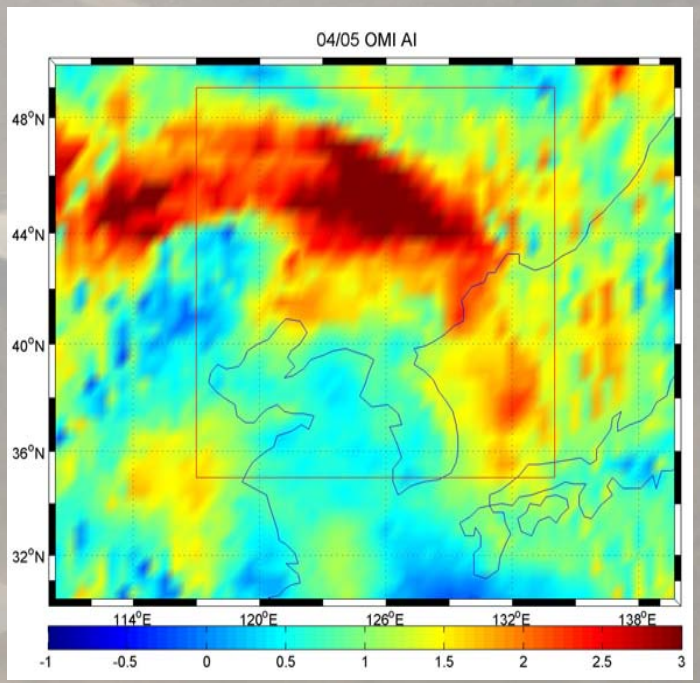
Where would the pollution plume travel to?

How would the plume evolve during transport?

From Space: Lots of Dust and SO_2, and over a Large Region



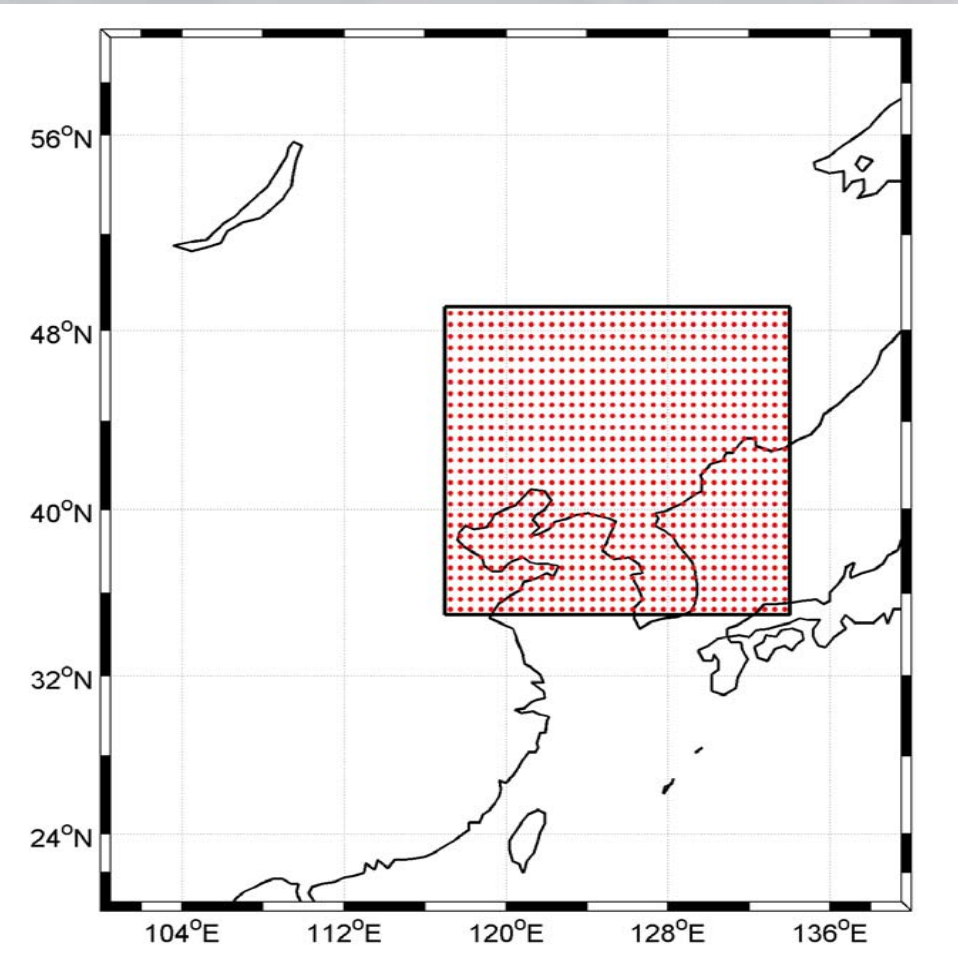
Aura/OMI Operational SO_2 April 5



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;

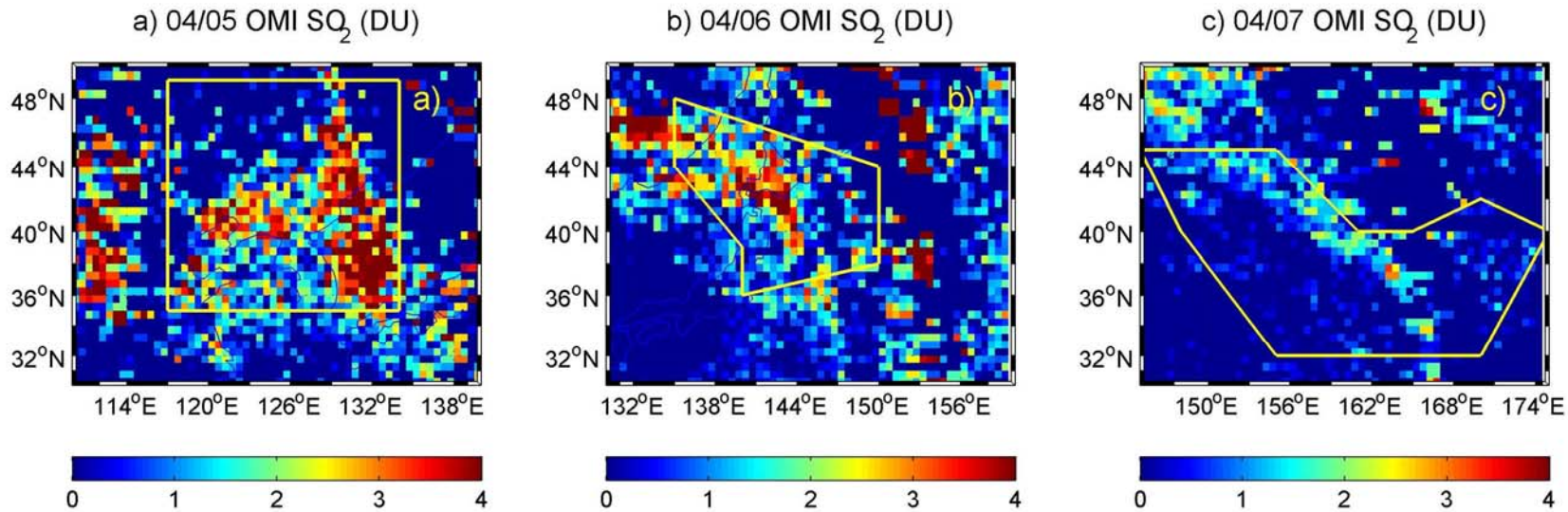
Region: 35-49N, 117-134E;

Forward trajectories initiated from the center of grids 0.5×0.5° in size;

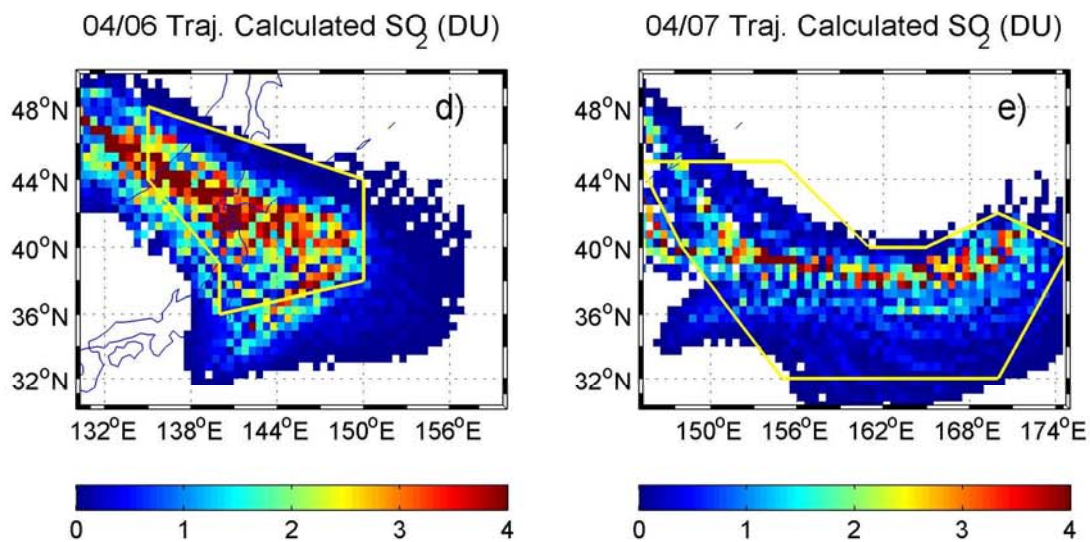
Eight Levels: every 500 m from surface to 4000 m AGL;

Each trajectory *tagged* with MODIS-retrieved AOD and OMI-retrieved SO_2, scaled with *aircraft-measured vertical profile*

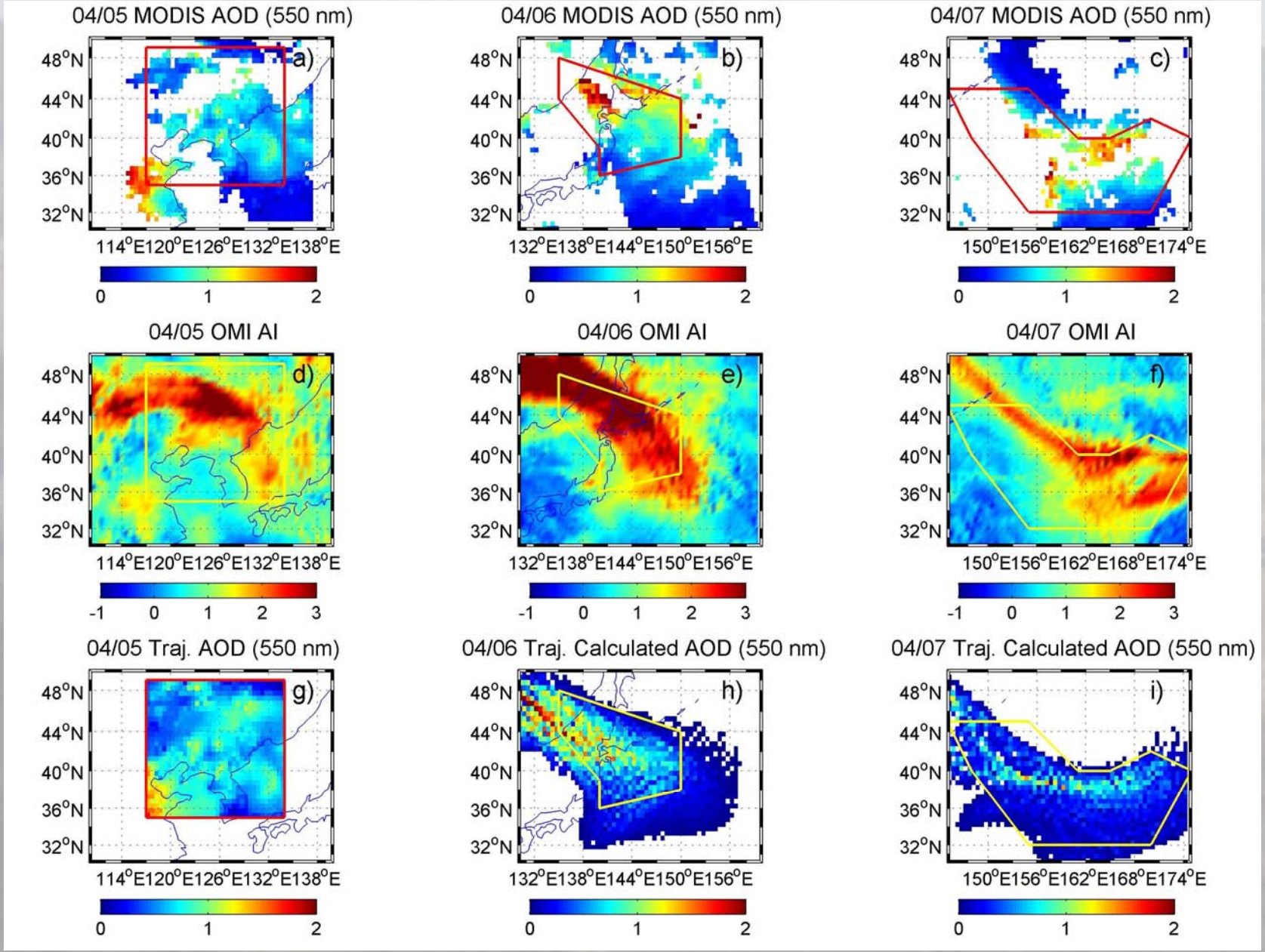
Snapshots of SO_2 Distribution by Aura/OMI



First-order correction for:
SO_2 vertical profile;
Aerosol and cloud interference;
Viewing geometry
[*Krotkov et al*., 2007]



Snapshots of Aerosol Distribution by Aqua/MODIS and Aura/OMI - Application of “A-Train”

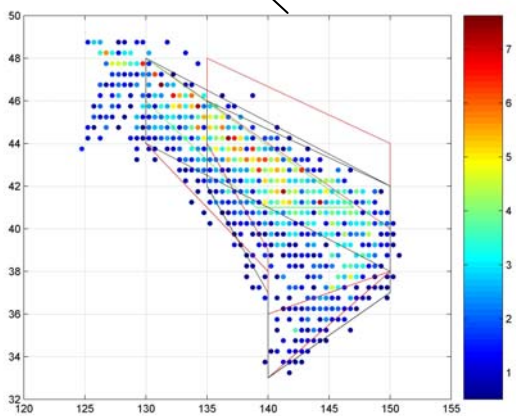


Detailed information for retrieval improvement

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

Estimate the fraction of the plume observed by satellite

Initial vertical distribution for tracer model



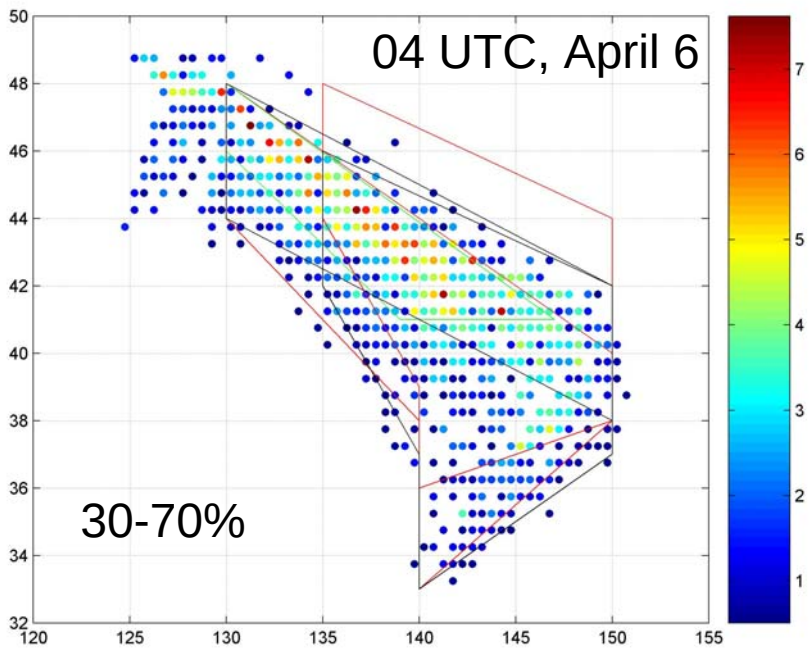
Measurements over small area; Detailed vertical information

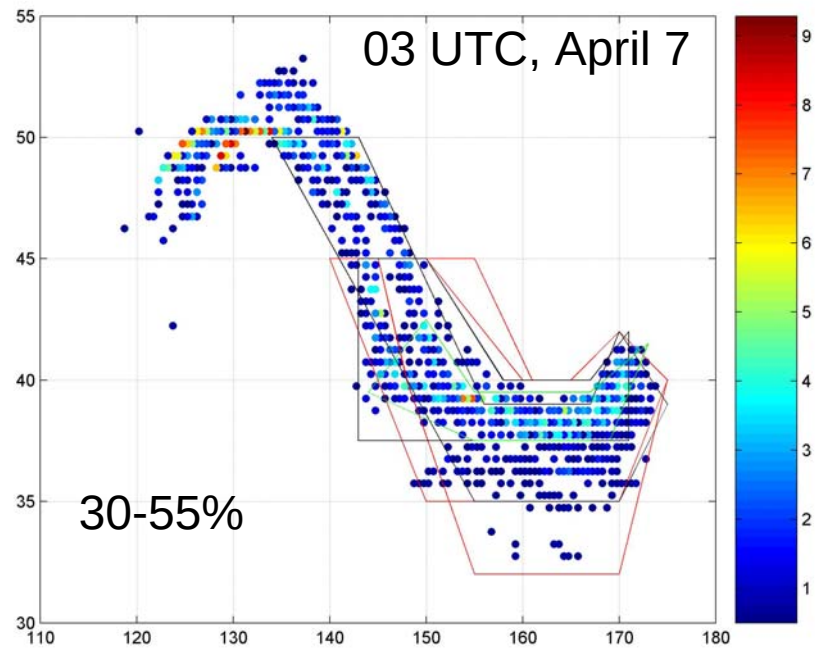
Assuming conserved tracers; Find the part of the plume with strongest signals.

Satellite observes the part of the plume with strongest signals;
Tracer Model estimates the fraction observed the by the satellite;
Aircraft provides information for trajectory and satellite

Observing Part of the Plume, Quantifying the Whole Plume

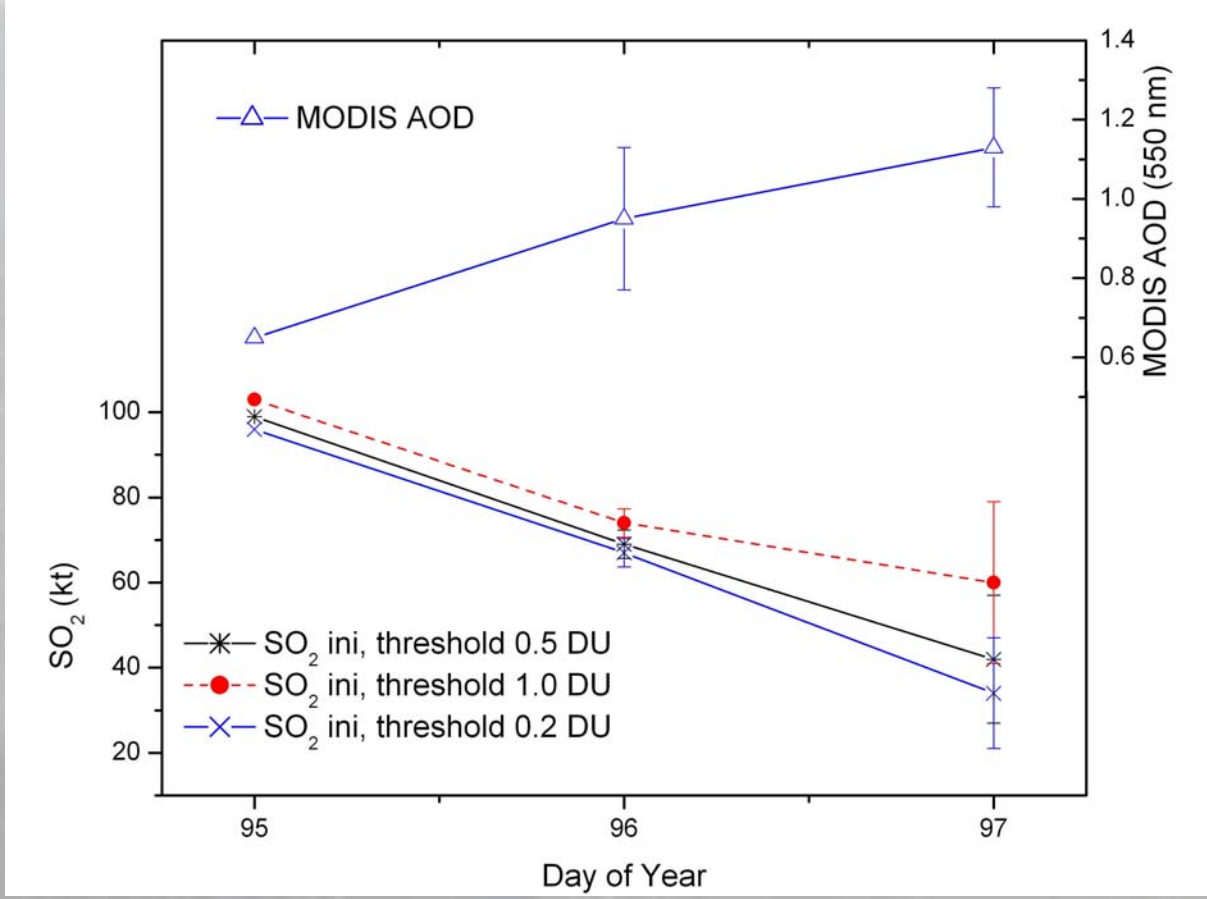
Select a polygon box (covers a good part of the trajectory-projected main body of the plume) →
Estimate the fraction of the plume covered by this box (using trajectory-projected SO_2 distribution, total trajectory SO_2 remains constant) →
Determine the SO_2 mass within the box (with satellite data, e.g., 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

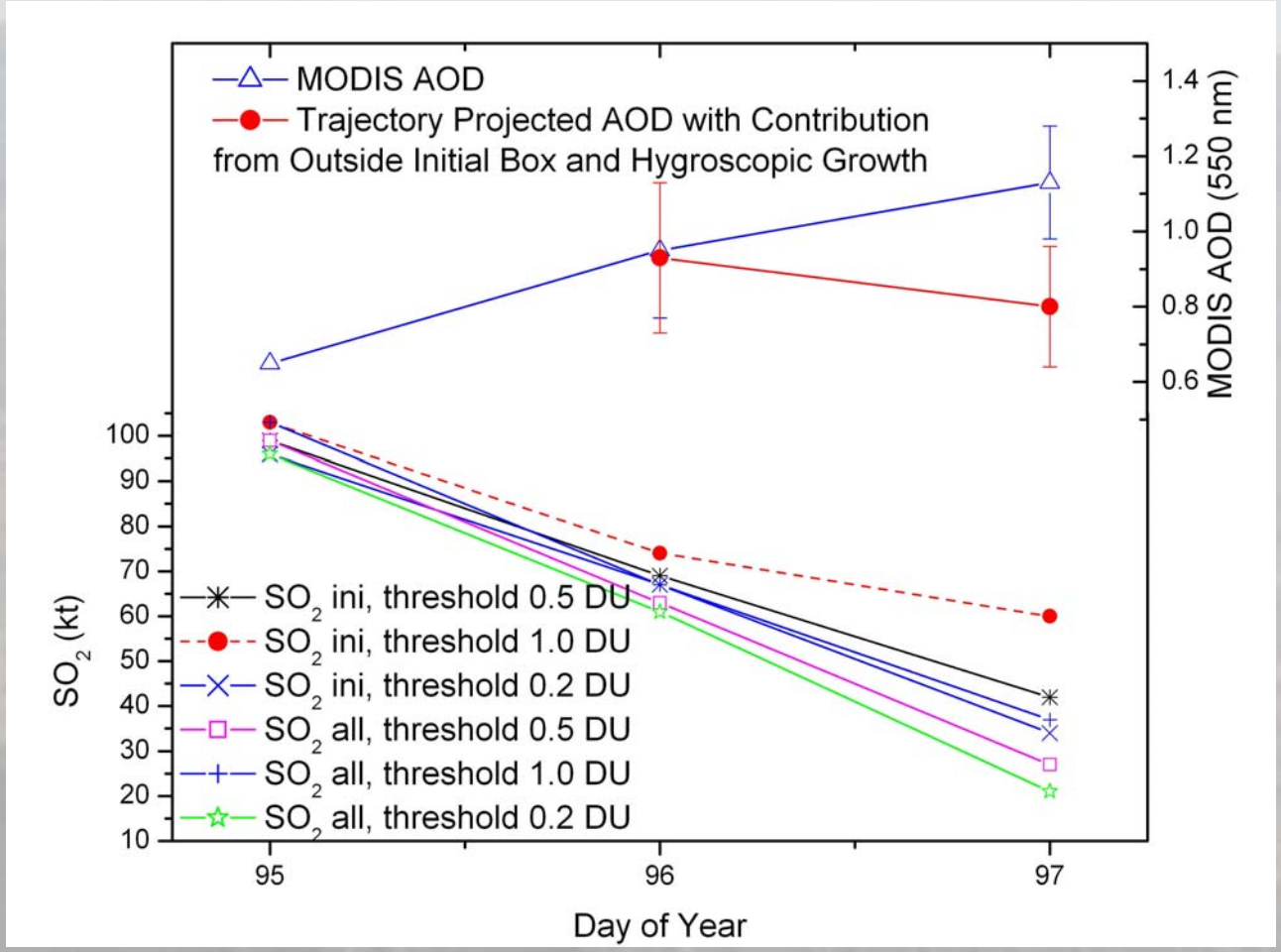


The satellite sensitivity to SO_2 may change during transport: different threshold values.

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*.

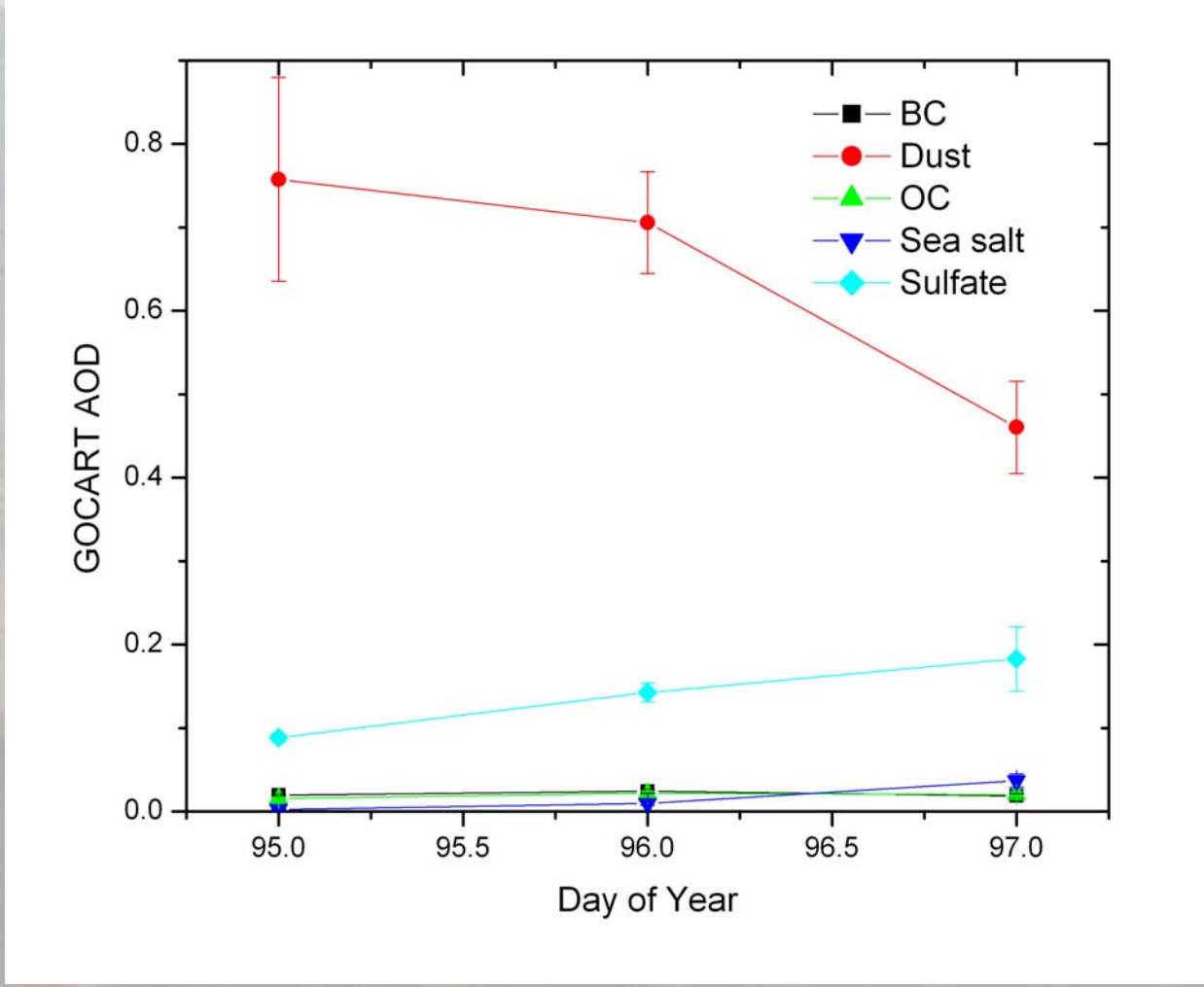
Hygroscopic growth of aerosols.

SO_2 Lifetime and SO_2 to Sulfate Conversion



SO_2 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 SO_2 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.

AOD of Different Aerosol Compounds near the Plume Core



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?