SOURCE APPORTIONMENT OF FINE ORGANIC AEROSOL USING CMAQ TRACERS

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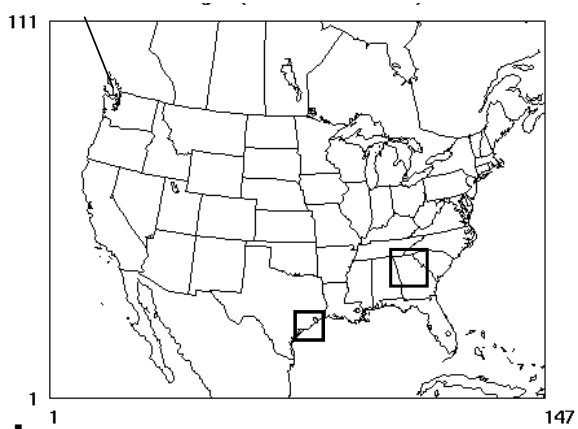
Objectives

- Implement and evaluate the reliability of using tracers for fine organic PM in CMAQ (CMAQ-TR)
- Apply CMAQ-TR to simulating source-specific impacts of regional emissions on ambient organic aerosol concentrations
- Comparing source apportionment results with receptor models

Modeling domain



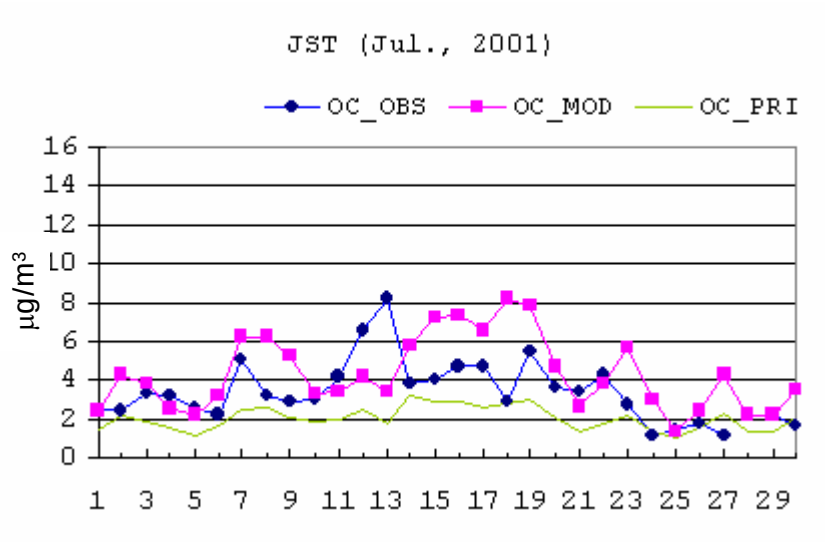
Air quality model

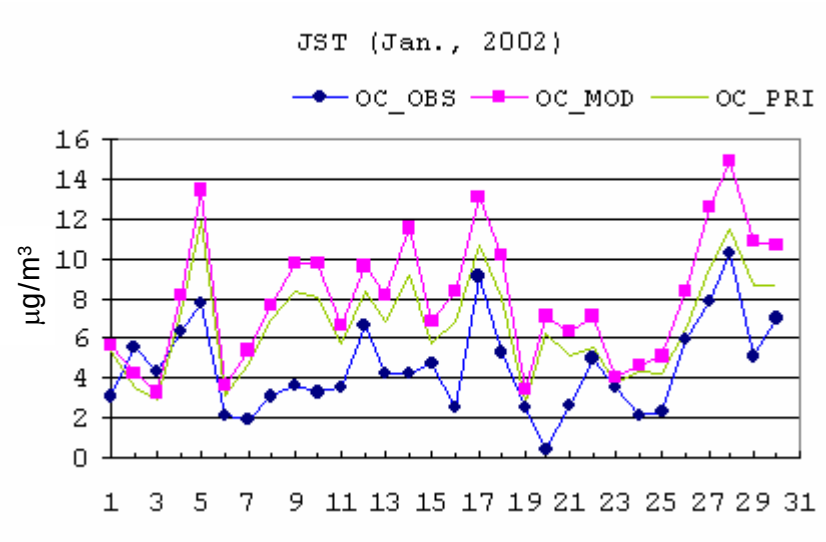
Model	CMAQ version 4.3
Grid size	36 km
Dimension	147 x 111
Periods	July 2001 and January 2002
Number of vertical layers	9
Top pressure of the model domain	100 hPa

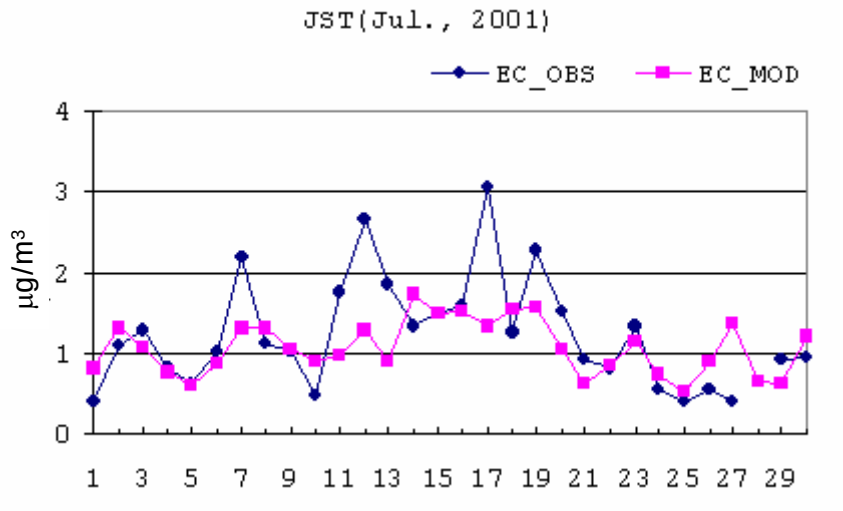
Emission inventories

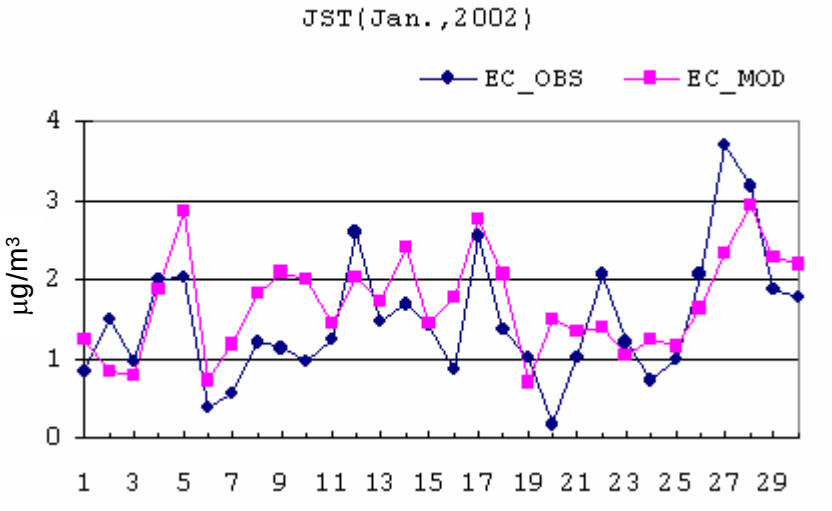
Model	SMOKE version 1.5 and 2.1
Base inventories	EPA 2001 inventories
Point sources in Georgia	EPA NEI 2002 (draft)
Forest fire, land clearing debris in 2002	VISTAS, 2005; Tian, 2005
Residential meat cooking	Emissions developed

Accuracy of CMAQ results – OC & EC





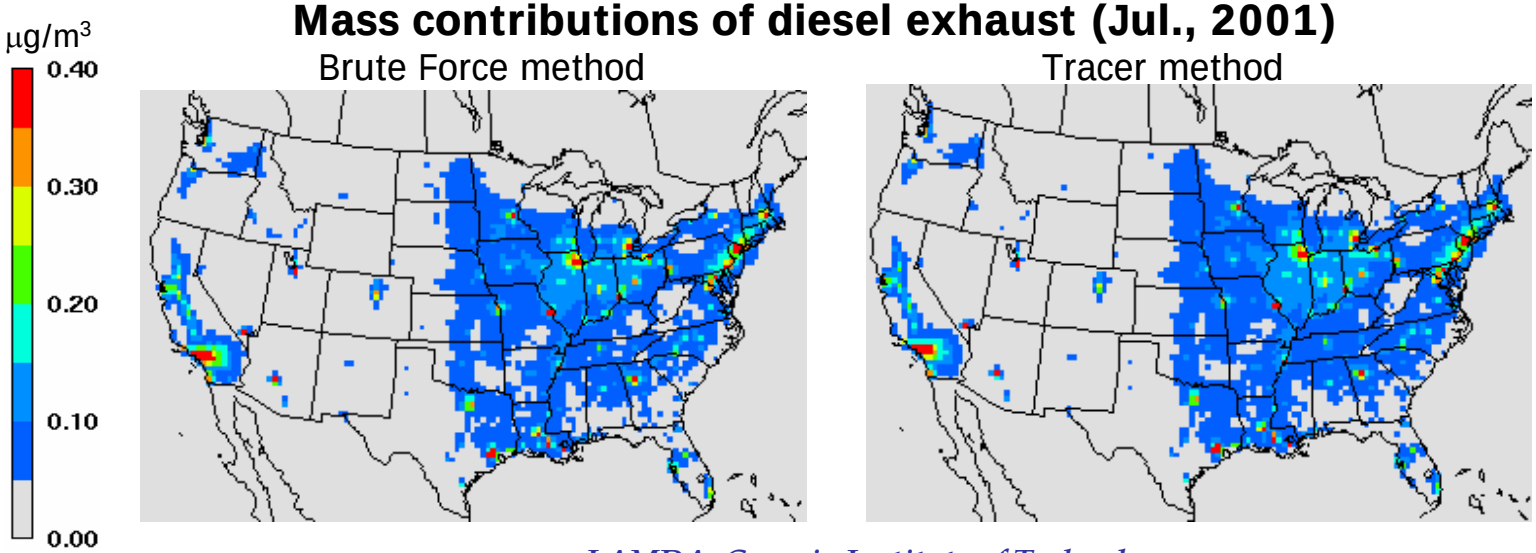




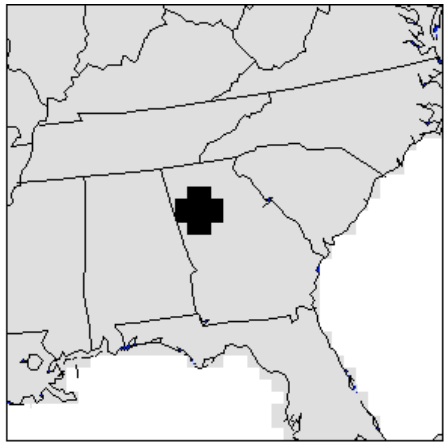
CMAQ-Tracer method

- Method
  - Add tracers for primary organic aerosols categorized into 34 sources, such as wild fires, fireplaces, natural gas combustion, etc.
- Reliability
  - Source apportionment results of 5 categories were compared with those using Brute Force
  - Mean fractional errors between two results were less than 5% with less than 3% of mean fractional bias for any source
- Usefulness
  - Detailed source apportionment of primary aerosols
  - Enhanced integrated emission-based/receptor model method

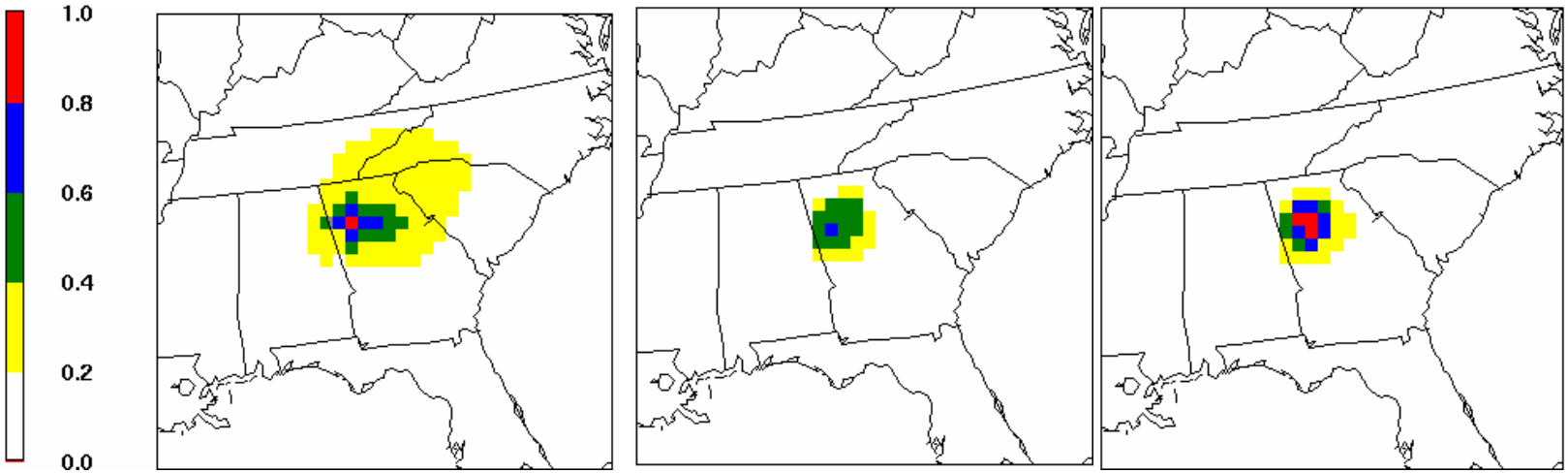
Mass contributions of diesel exhaust (Jul., 2001)



Regional impacts of emissions from the Atlanta area



- Changes in separate source categories had different trends both in spatial distribution and quantities



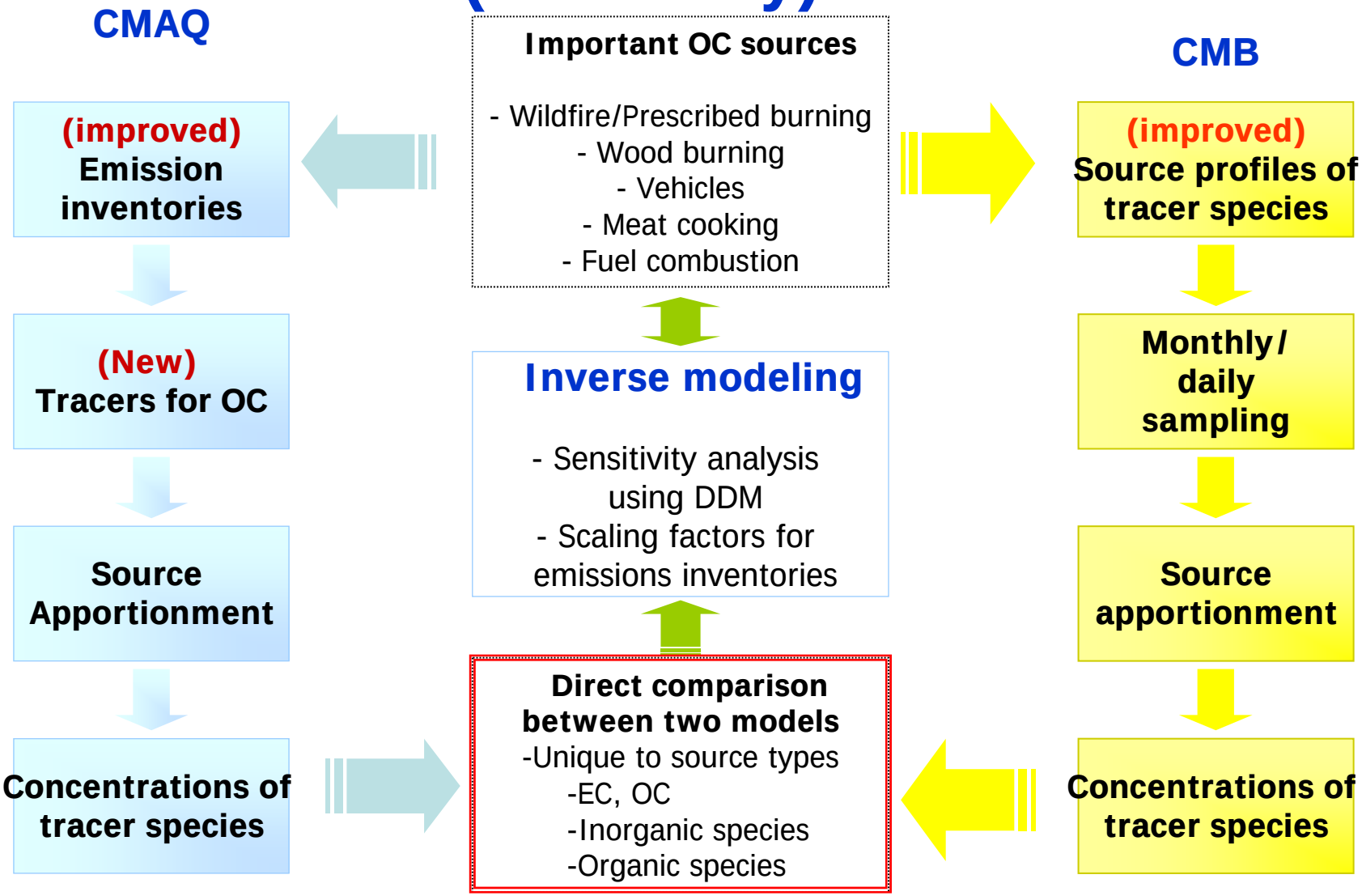
Industrial process – asphalt roofing

Wildland fire

Meat cooking

*1.0 – 100% of primary organic aerosol came from the Atlanta area

Integrated source/receptor based methods using CMAQ & CMB models for primary OC (Underway)



Comparison with receptor models

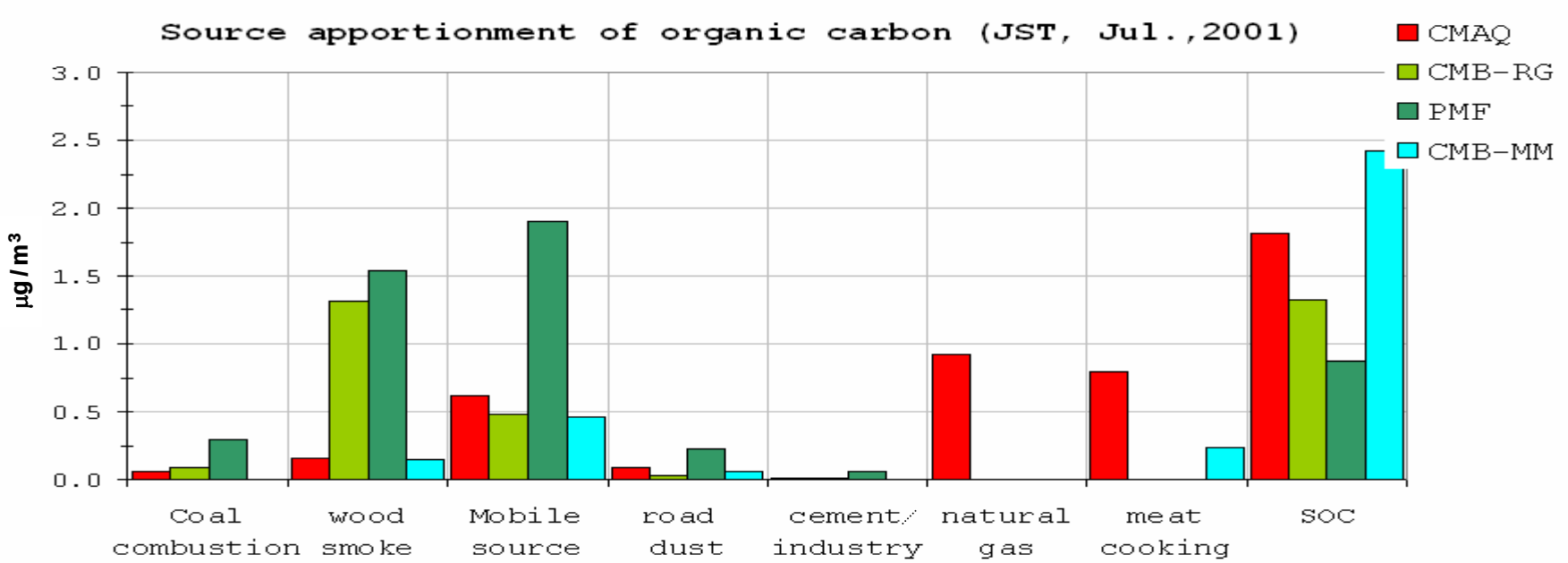
- Positive Matrix Factorization (PMF)
  - Ambient measurements and meteorology data
  - Source profiles are obtained by factor analysis

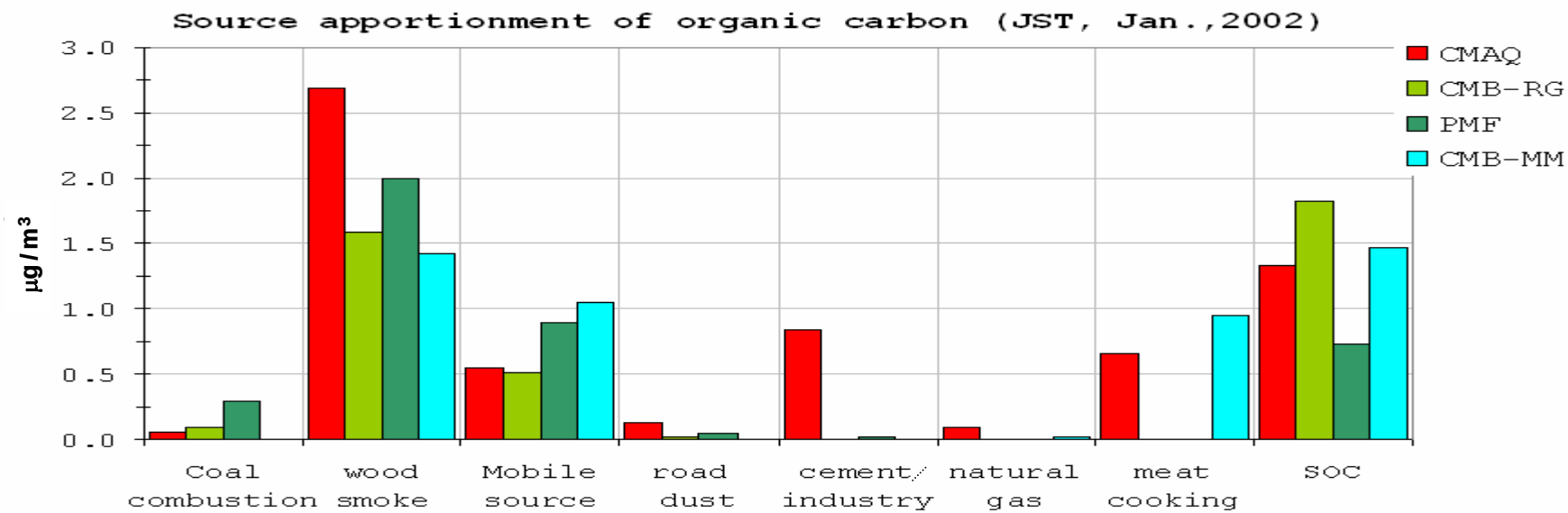
- Chemical Mass Balance-Regular (CMB-RG) model
  - Ambient measurements and source profiles
  - Source profiles are measured at emission sources
  - Inorganic and metal species as fitting species

- Chemical Mass Balance-Molecular marker (CMB-MM) model
  - Additional organic compounds are used as fitting species

* *W. Liu and S. Lee, 2005 (PMF, CMB-RG), B. Yan and M. Zheng, 2004 (CMB-MM)*

Comparison – Source apportionment





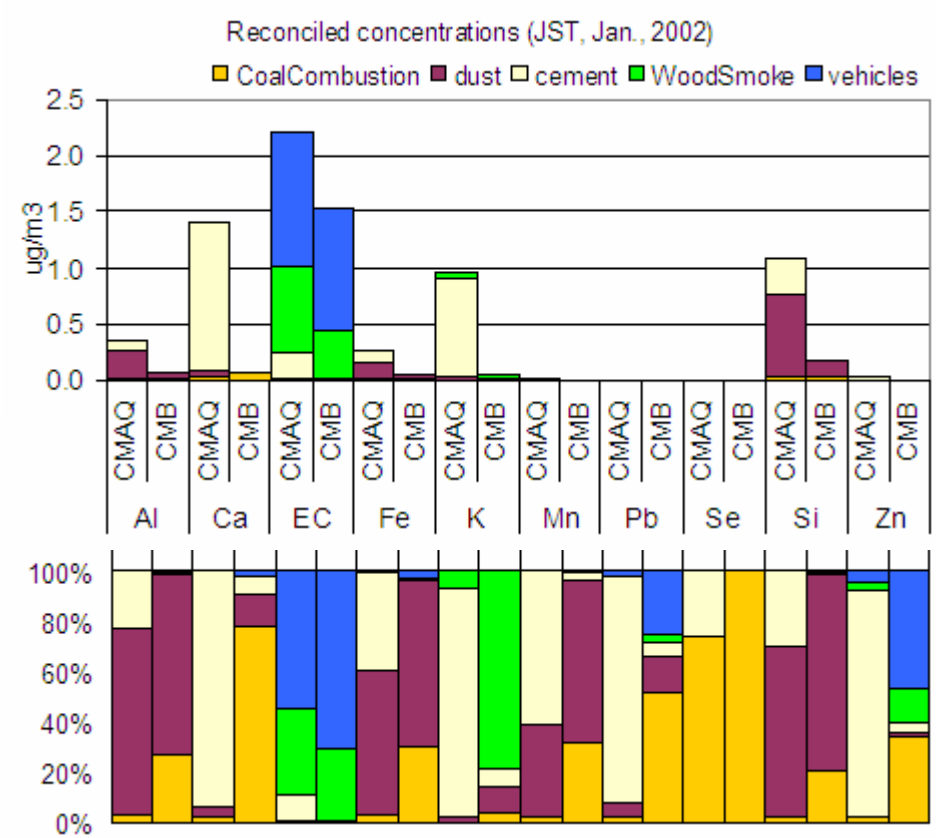
Why are they different?

- Difference in reconciled concentrations
- Differences between SMOKE speciation profiles and source profiles in receptor models
- Unaccounted sources in receptor models
- Accuracy of source profiles
- OM to OC conversion

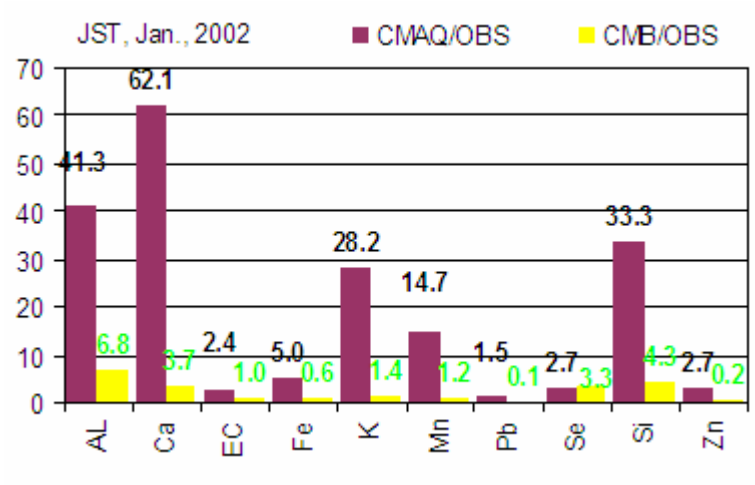
Difference in reconciled concentrations

- Simulated OC at JST was high especially in Jan., 2002
- Difference between reconciled concentrations is more related with contribution of each of source categories than total OC concentration

<Reconciled concentrations>



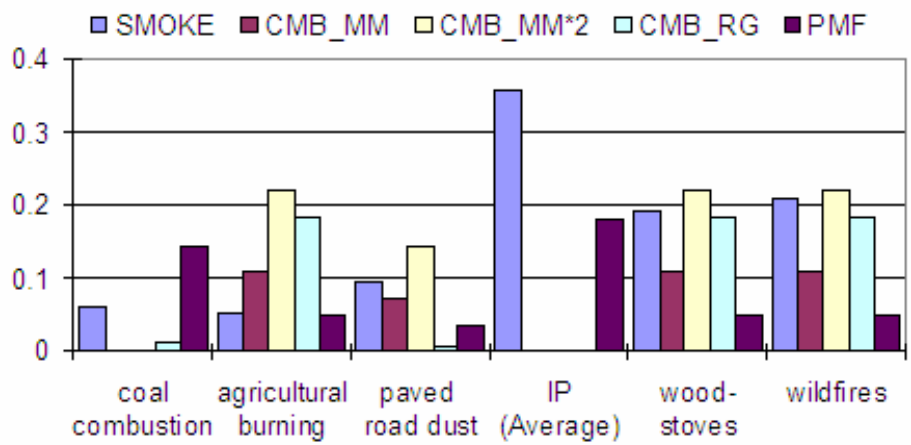
<Ratios of estimated concentrations to observations>

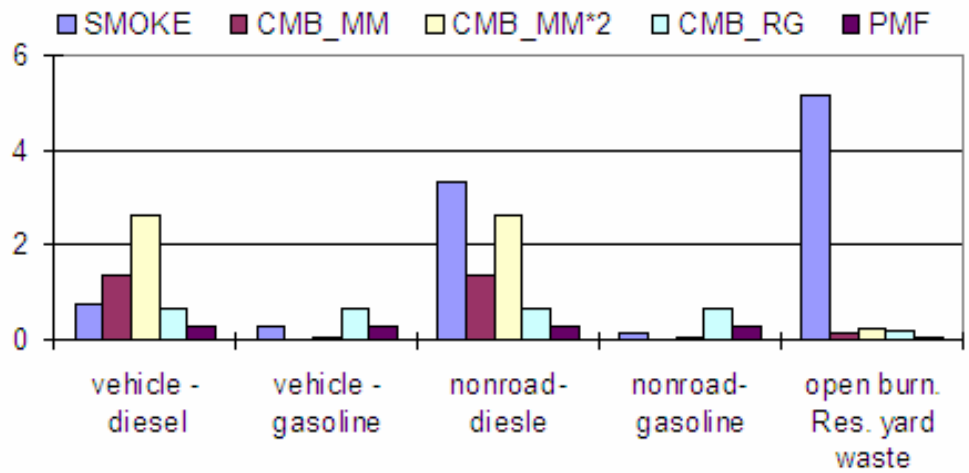


Different EC/OC ratios in source profiles

- EC to OC ratios play an important role in source apportionments of diesel exhaust and industrial process
- Source profiles in receptor models are mixtures of many sources (e.g., a wood burning category includes sub-categories such as forest fire, fireplace and leaf species burning)
- Sub-categories have different EC/OC ratios in SMOKE profiles

➢ Source profiles in receptor models should be site specific





Unaccounted sources in receptor models

- Important sources of primary organic aerosol
  - According to CMAQ simulations and the inventory, different sites have different dominant sources
  - If there are missing sources in receptor models, source apportionment results are in substantial errors (Christensen, 2004)

Jefferson St.

CMAQ results	CMB-RG / PMF		CMB-MM	
	Jul.2001	Jan. 2002	Jul., 2001	Jan., 2002
Total POA (mg/m^3)	1.85	6.18	1.85	6.18
Sum of POA from sources treated in receptor models	0.95	4.26	1.69	5.95
Sum of POA from sources not-treated in receptor models	0.9 (49%)	1.92 (31%)	0.16 (8%)	0.23 (4%)

Conversion from organic matter to organic carbon

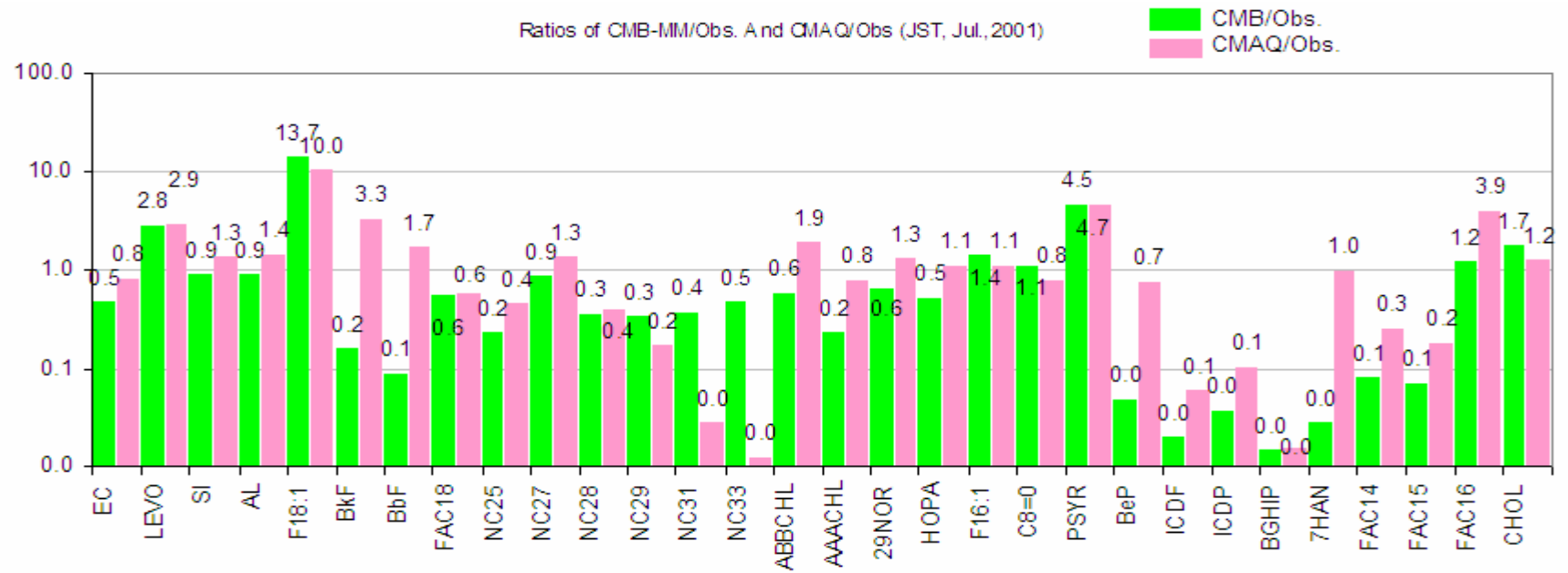
- CMAQ organic carbon concentrations
  - Simulation results of organic aerosol from CMAQ are concentrations of organic matter
  - A conversion factor from OM to OC is set as 1.4
  - Conversion factors are different from sources; factors are needed to be defined at each of sites (Turpin, 2000)

- Molecular weight per carbon weight (Rogge, 1993; Schauer, 1998)

Compound class	MWt/C Wt	Compound class	MWt/C Wt
n-Alkanes	1.2	Diterpenoid acids	1.3
n-Alkanoic acids	1.3-1.5	PAH	1.0-1.1
n-Alkenoic acids	1.3-1.5	Cholesterol	1.2
Ketocarboxylic acids	1.9-3.1	Levoglucosan	2.3

Accuracy of source profiles

- Some reconciled species are markedly overestimated in CMAQ
  - Ratios of simulated concentrations of Si to observations are 25 in Jul., 2001, and 33 in Jan., 2002
  - Al, Ca and K have ratios higher than 20 in Jan., 2002
  - Octadecenoic acid, benzo(k-,b-)fluoranthen, abietic acid were overestimated by a factor of 10



Further studies

- Improving CMAQ and receptor models
  - Inverse modeling using reconciled concentrations of species
  - Site specific OM to OC conversion factors
  - Modification of source profiles in receptor models and speciation profiles in CMAQ (SMOKE) model
  - Implementing results from recent monitoring studies (Prescribed burns, highway/rural monitoring)
  - Identifying unknown sources in receptor models
  - Further inter-comparison with receptor models
  - Further evaluation with chemically detailed observations
  - Identifying SOA tracers

Thank you for your attention!