Source Apportionment of Diesel Particulate Matter in the Southeastern United States Using Models3/CMAQ

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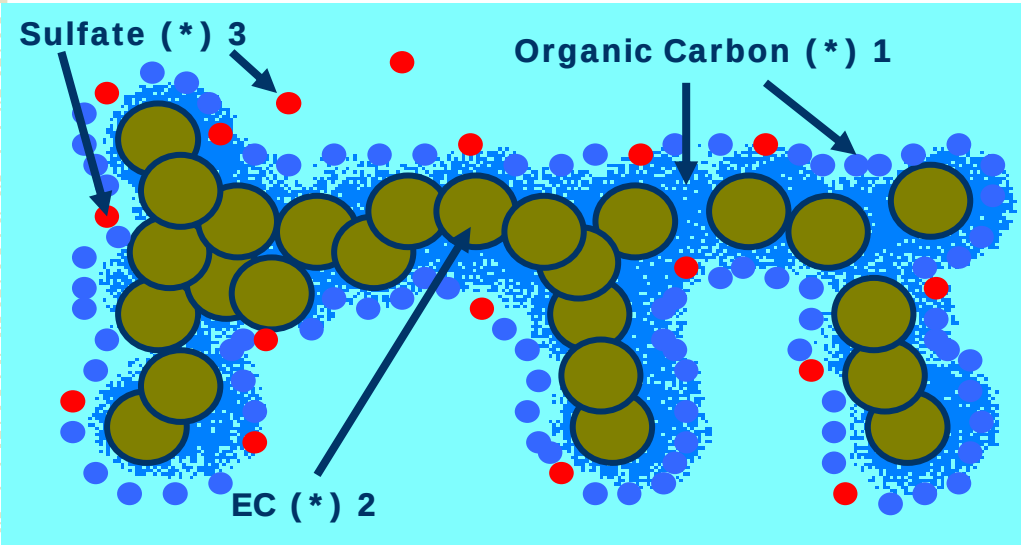
Outline

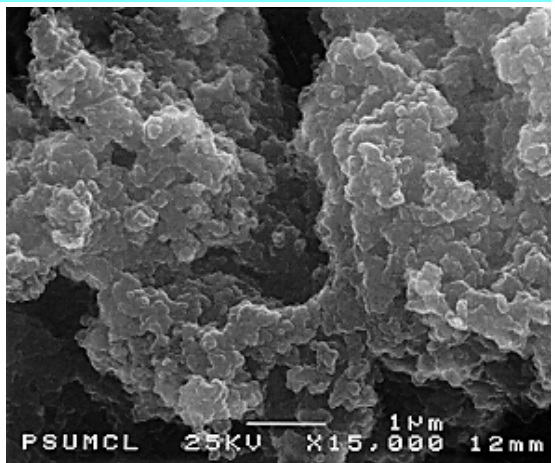
- Introduction & Background
- Methodology used in this study
- Results
- Conclusions

Introduction

- Diesel particulate matter (**DPM**) is emitted from a broad range of diesel engines; mainly on-road and non-road sources.
- **DPM** is in the category of fine and ultra fine particles.
- **DPM** is part of a complex agglomerated mixture composed mainly of elemental carbon (**EC**) with hundreds of adsorbed organic compounds, sulfates, nitrogen oxides, heavy metals, trace elements, and irritants (such as acrolein, ammonia, and acids).
- Specific toxic chemicals of concern include polycyclic aromatic hydrocarbons (**PAHs**) and **nitroarenes** (CAL EPA, 1998) which are concentrated in the particle phase.

Introduction (cont.)





DPM is emitted under a form that particles with up to 0.02 μm in diameter.

*1: unburned constituents of fuels and lubricants
*2: generated by imperfect combustion of fuels and lubricants
*3: sulfur in fuel is oxidized, and emitted as sulfate.

Introduction (cont.)

- Increased mortality and morbidity in communities with high DPM concentrations has been reported by a variety of studies (Conrad et al 2005, Fruin et al 2004, and Adonis et al 2003)
- Using the NEI 1999, Conrad estimated that DPM posed a cancer risk that was 7.5 times higher than the combined total cancer risk from all other air toxics in the U.S.
- DPM shortness the lives of nearly 21,000 people each year in U.S.

Background

1999 Primary PM2.5 Emissions

Source	%
Fuel Combustion	11.2
Industrial Processes	13.5
Transportation	9.5
Miscellaneus (fugitive dust, agricultural, etc.)	65.8

1999 PM2.5 Ambient Composition

Constituens	Easter US (%)	Western US (%)
Sulfate	56	33
Org. Carbon	27	36
Elem. Carbon	**5**	**6**
Nitrate	5	8
Crustal	7	17

Background (cont.)

- Diesel-Fueled Sources (DFS) are the major sources of the atmospheric EC (Schauer 2003)
- The EC contribution to ambient DPM has been ranged between 50 and 80%
- EC has been used as a marker to estimate the DPM concentrations and as subrogate for DPM (Shah et al 2004, Tamura et al 2003, Birch et al 1996, y Schauer 2003)
- However, EC is not a unique tracer for ambient DPM, which could generate significant bias.
- There is not available methods to measure ambient DPM.

Background (cont.)

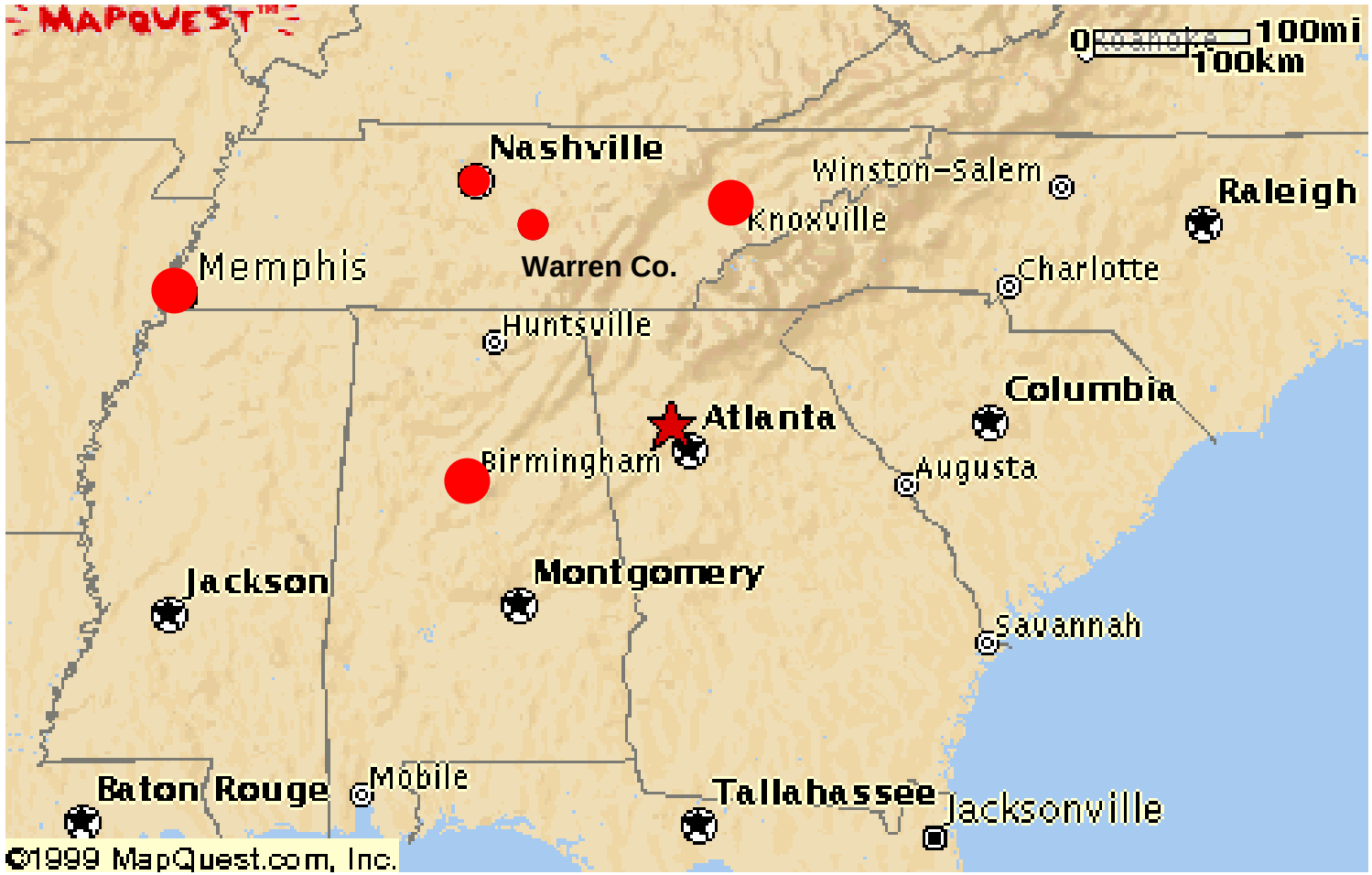
- Since the fine and ultra fine sizes of DPM are of greatest health concern, it requires identification and control of sources responsible for precursor emissions.
- The objective of this study is to estimate the contribution of major source categories to emissions and ambient DPM in the Southeastern region in the NEI 1999.
- Typically receptor models have been used for source apportionment purposes.

Background (cont.)

- Receptor models, however, do not fully take into account the chemical reactions involved in formation of secondary fine particles (NRC report, Vol. II, 1999)
- This study utilized an emissions/source-based model EPA's Models3/CMAQ modeling system over Five Urban areas and one Rural area from the Southeastern US.
- The uncertainty to use EC as a tracer was reduced, since DPM was estimated eliminating the DFS in the NEI 1999.

Background (cont.)

Southeastern DPM Research



Methodology

- Modeling Domain: 36 km domain
- Episode: Aug 30 –Sep 8, 1999
- Process Meteorology Inputs –MM5 version 3
- Process Inventory –SMOKE Version 2.0
  - Process average county based emissions to hourly gridded emissions
  - Speciation of emissions using Carbon-bond IV (CB-IV) mechanism
- CMAQ Version 4.3 Base case run
- CMAQ Version 4.3 Base case without DFS

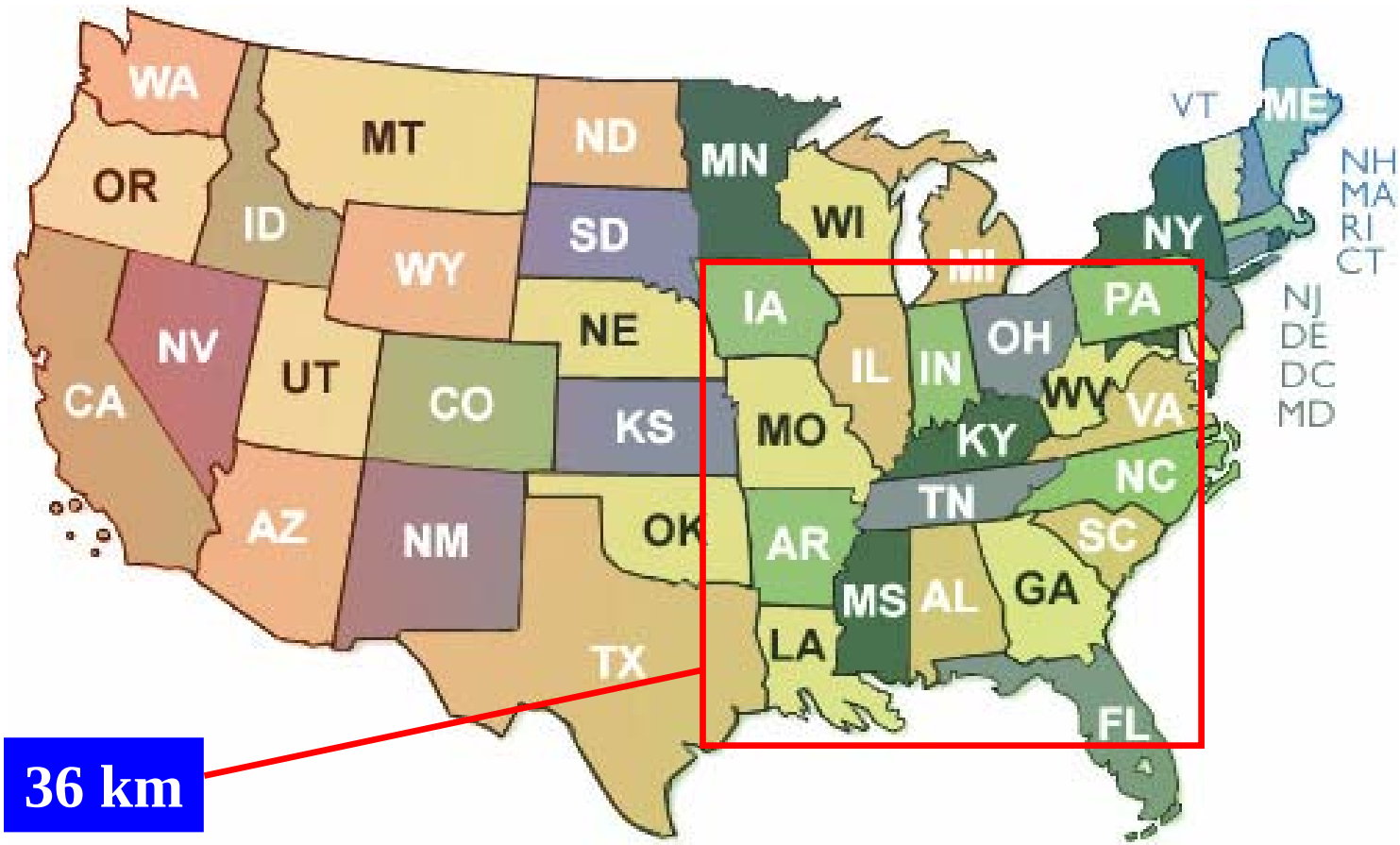
Methodology

Base Case with all sources

Base Case without diesel fueled sources

Diesel fueled sources contribution

Modeling Domain



Inventory Development

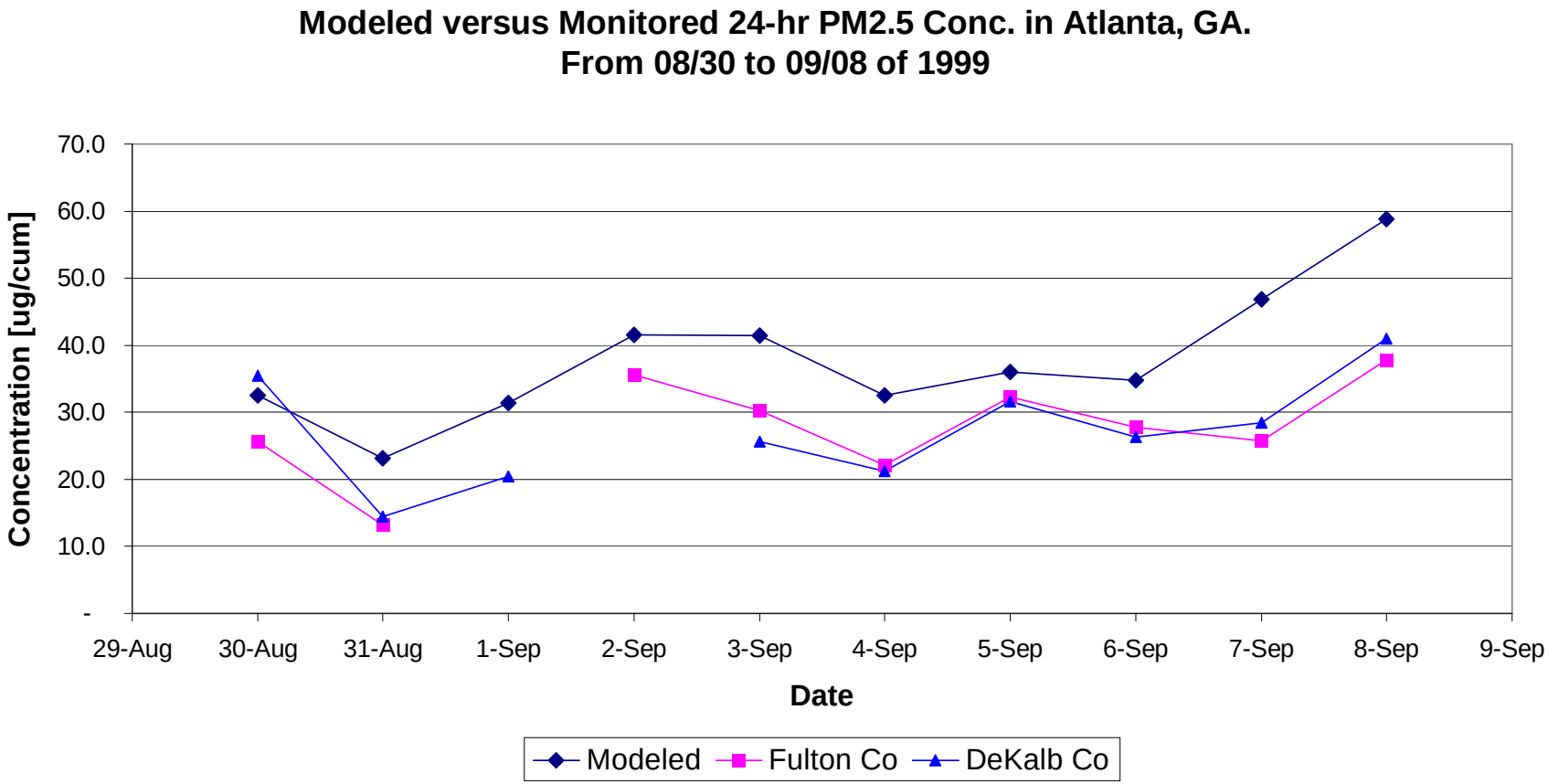
- On-road mobile source inventory and point source inventory for TN were developed by UT
- Ammonia emission inventory for TN was developed at UT
- NEI 1999 for TN Area source and Non-Road mobile source inventory (except ammonia)
- NEI 1999 inventory was used for all other states for all sources

CMAQ modeling

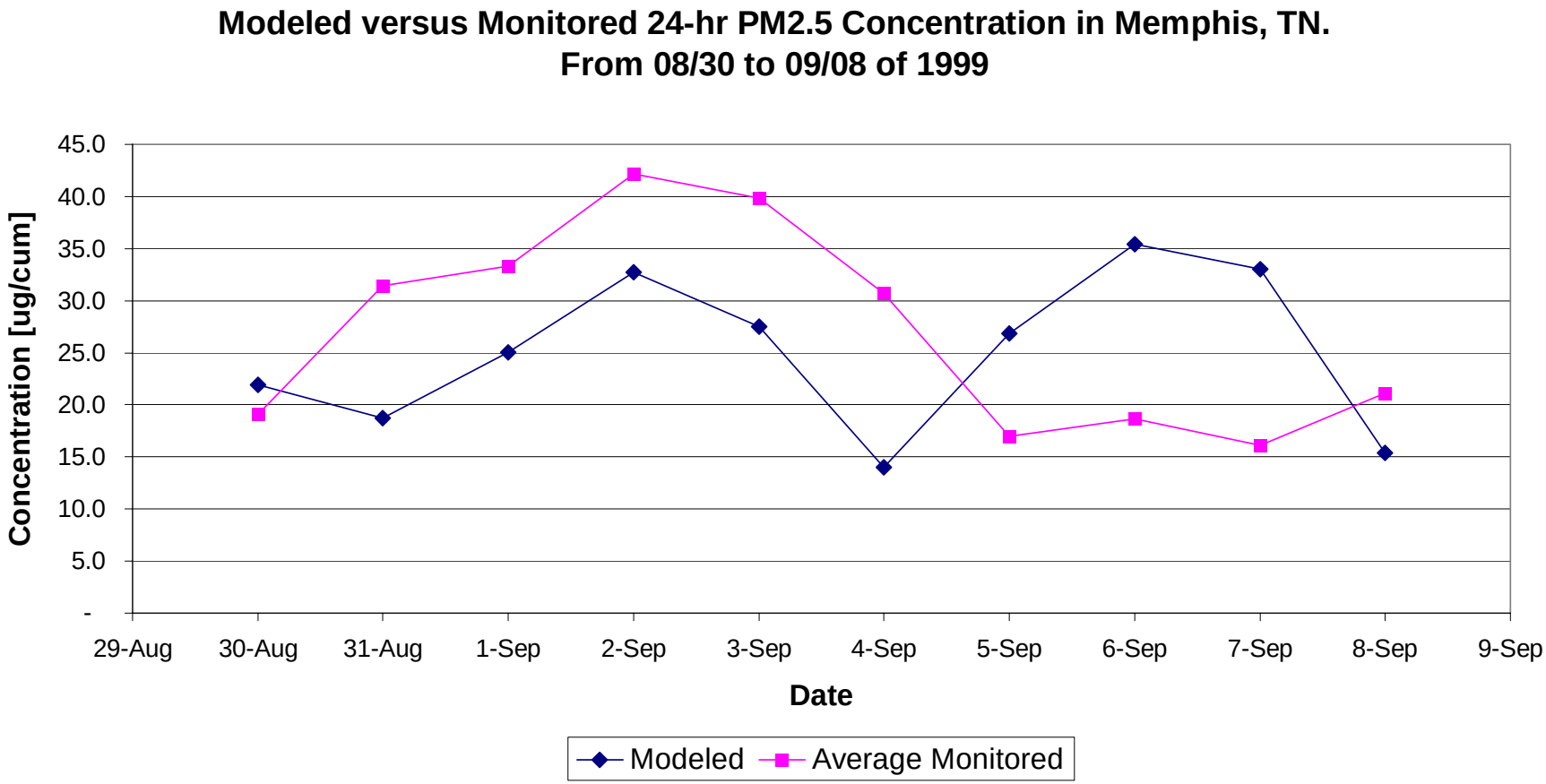
- **Base Case**
  - Use a default time invariant set of IC/BC for 36 km run
  - CMAQ run set to start three days prior to first day of model episode to allow for “spin-up” period and to avoid the influence of IC on model results

- **Base Case Without DFS**
  - The DPM was estimated eliminating DFS in the model.

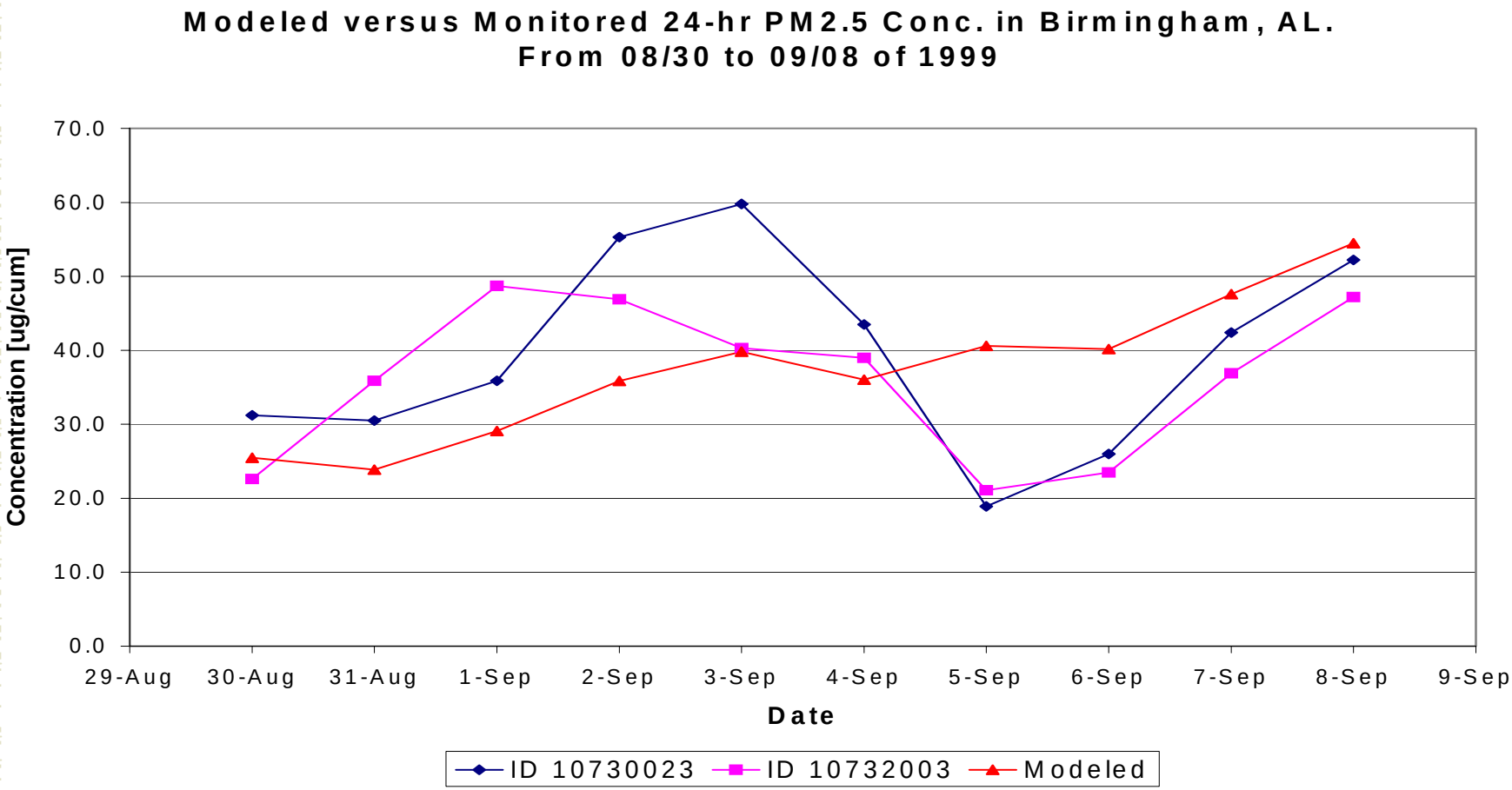
Results & Modeling Performance



Results & Modeling Performance



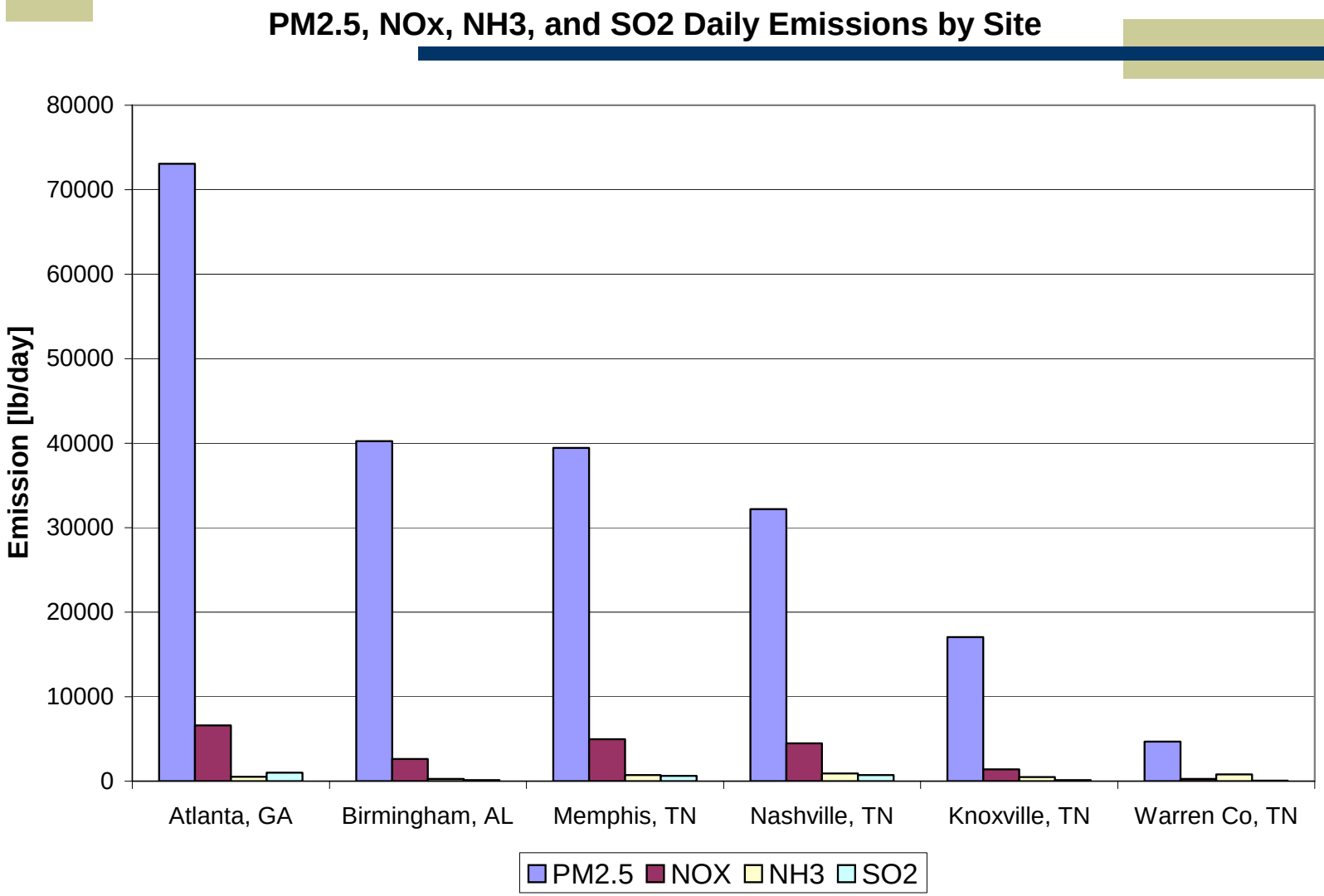
Results & Modeling Performance



Model Performance

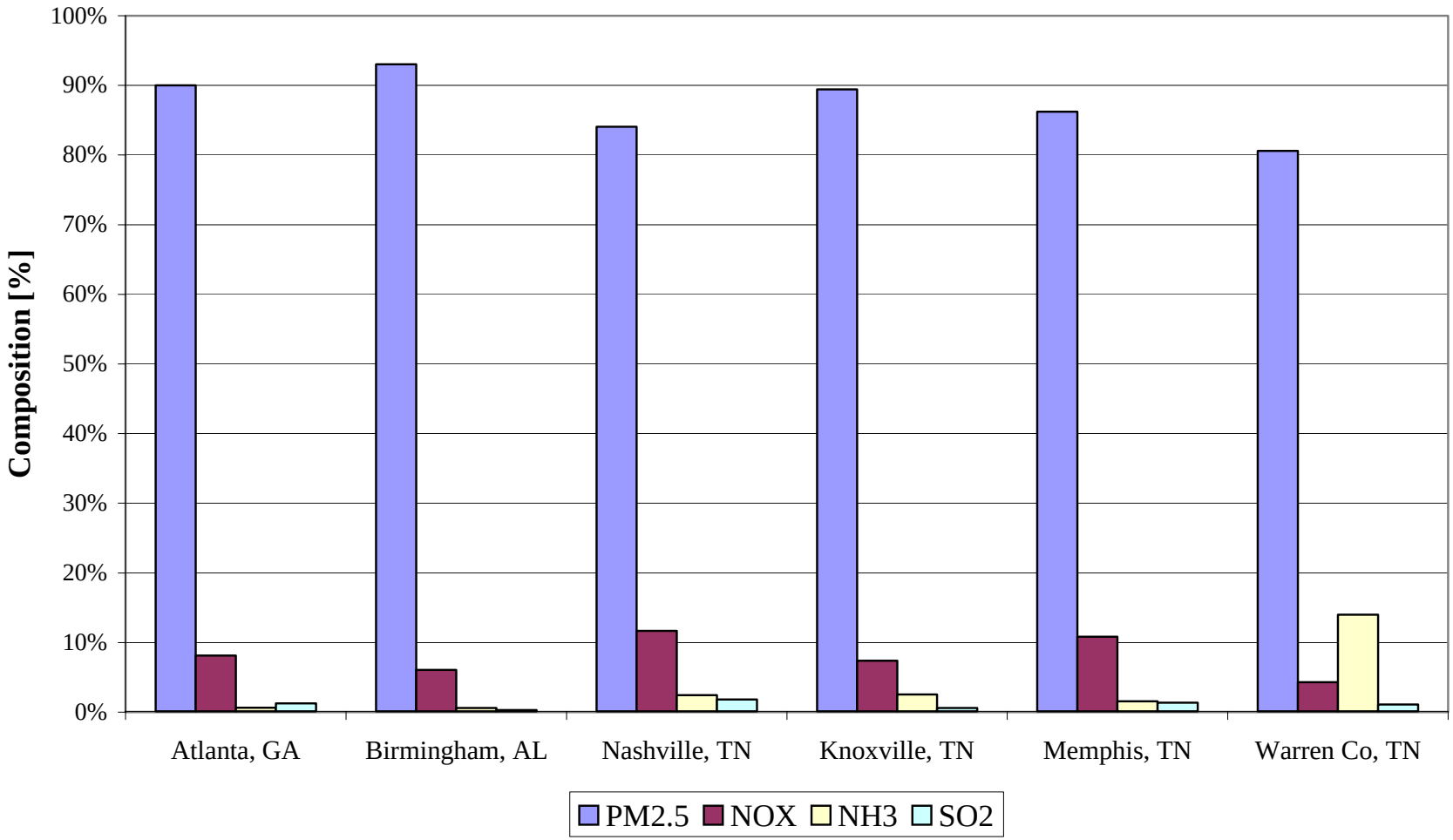
- Limited monitored data available for the episode
- Model over-predicted in Atlanta, GA, but it reproduced the trend during the whole episode of 1999.
- Model under-predicted prior to Sep 4, 1999 and over-predicted for days after Sep 5, 1999 in Memphis, TN
- Model reproduced the trend at least for part of the episode (for Memphis site -prior to Sep 4, 1999)
- Model under-predicted prior to Sep 4, 1999 and over-predicted for days after Sep 5, 1999 in Birmingham, AL
- Model reproduced the trend at least for part of the episode (for Birmingham sites -after Sep 5, 1999)
- Overall, the model fell within the range of current EPA guidance

Emissions Results for the BC



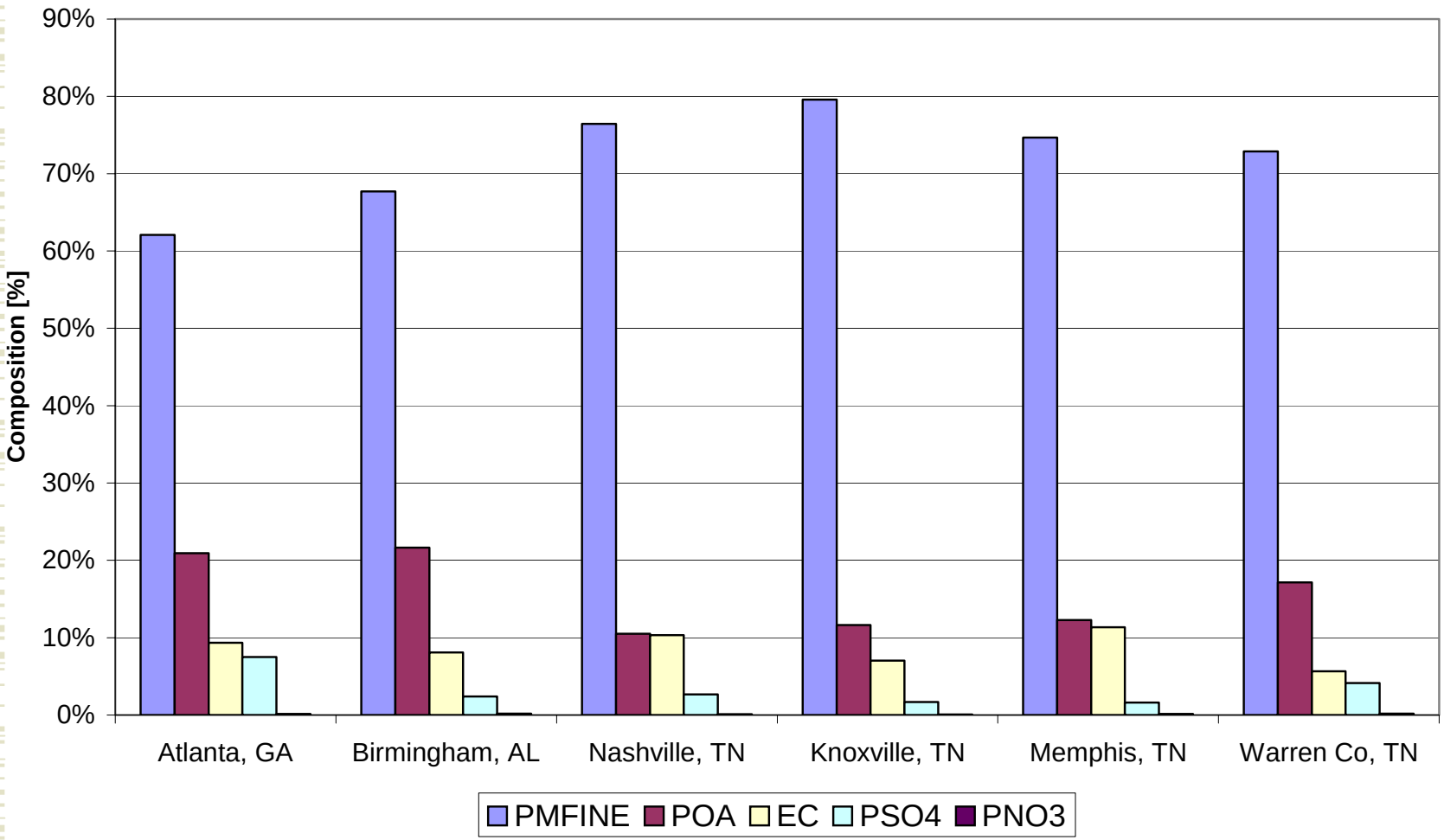
Emissions Results for the BC

PM2.5, NOx, NH3, and SO2 Emissions Composition by Site
From 08/30 to 09/08 of 1999



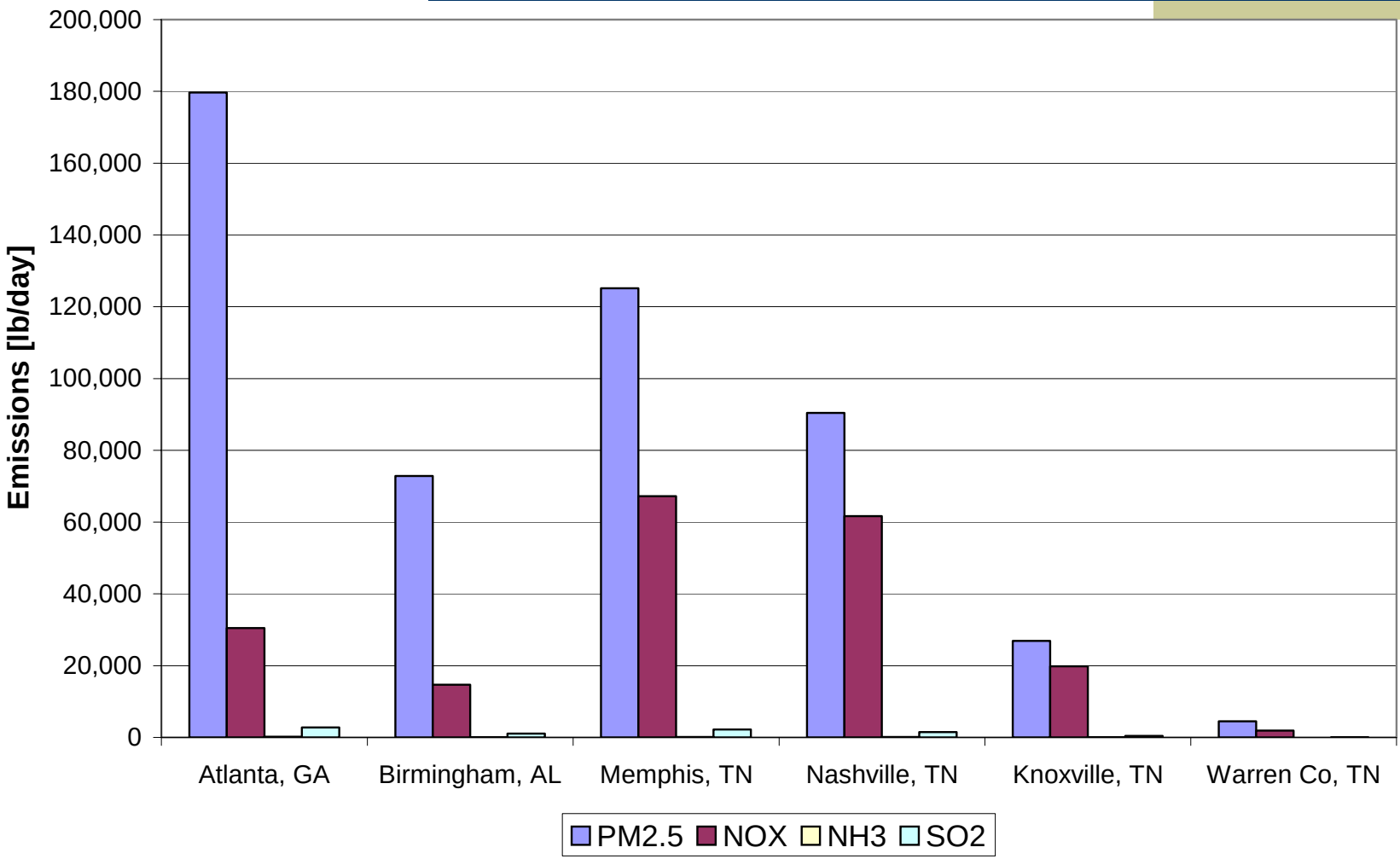
Emissions Results for the BC

Species Composition of PM2.5 Emissions by Site
From 08/30 to 09/08 of 1999



Emissions Results for DFS

Daily DFS Emissions from 08/30 to 09/08 1999



Emissions Results for DFS

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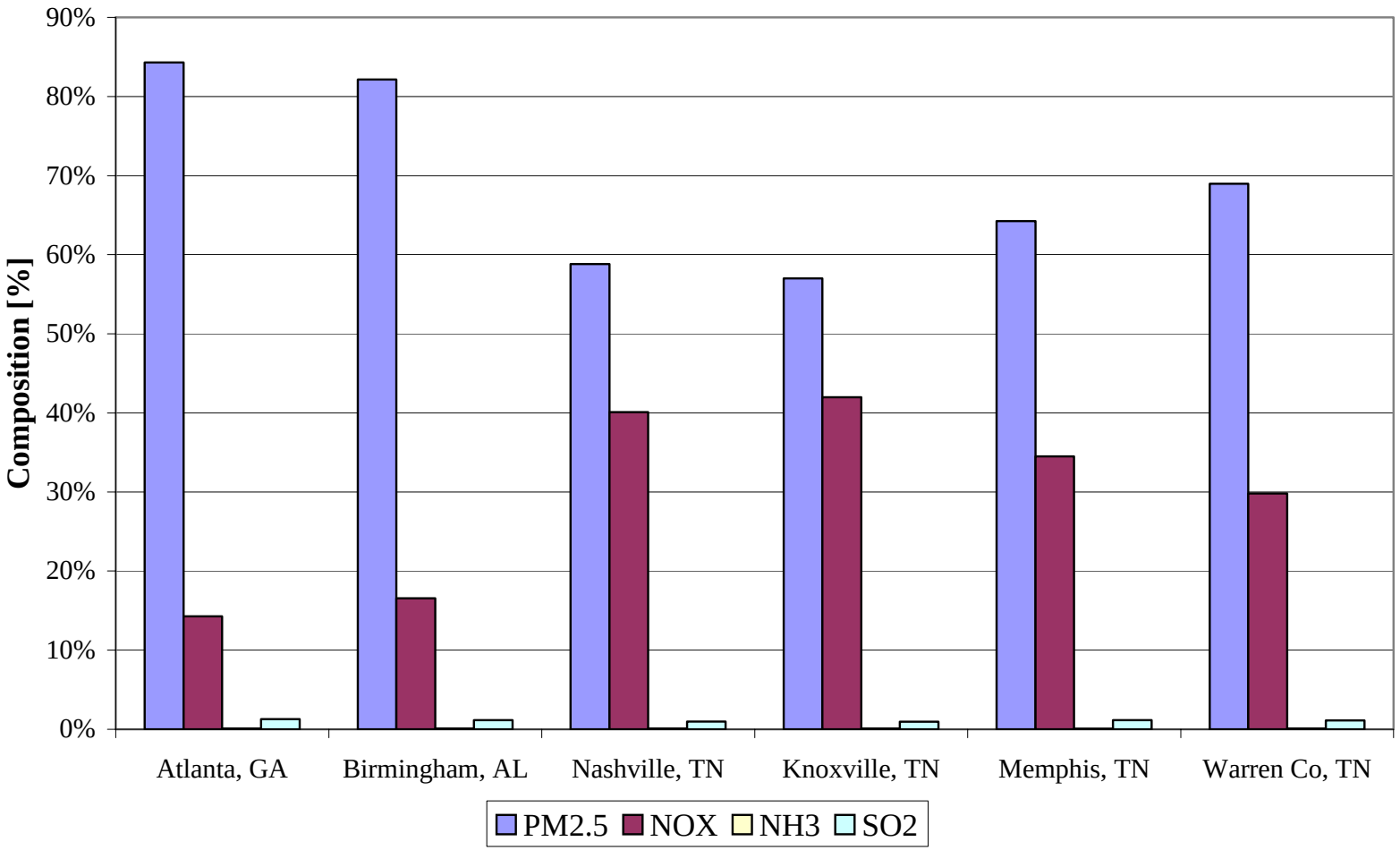
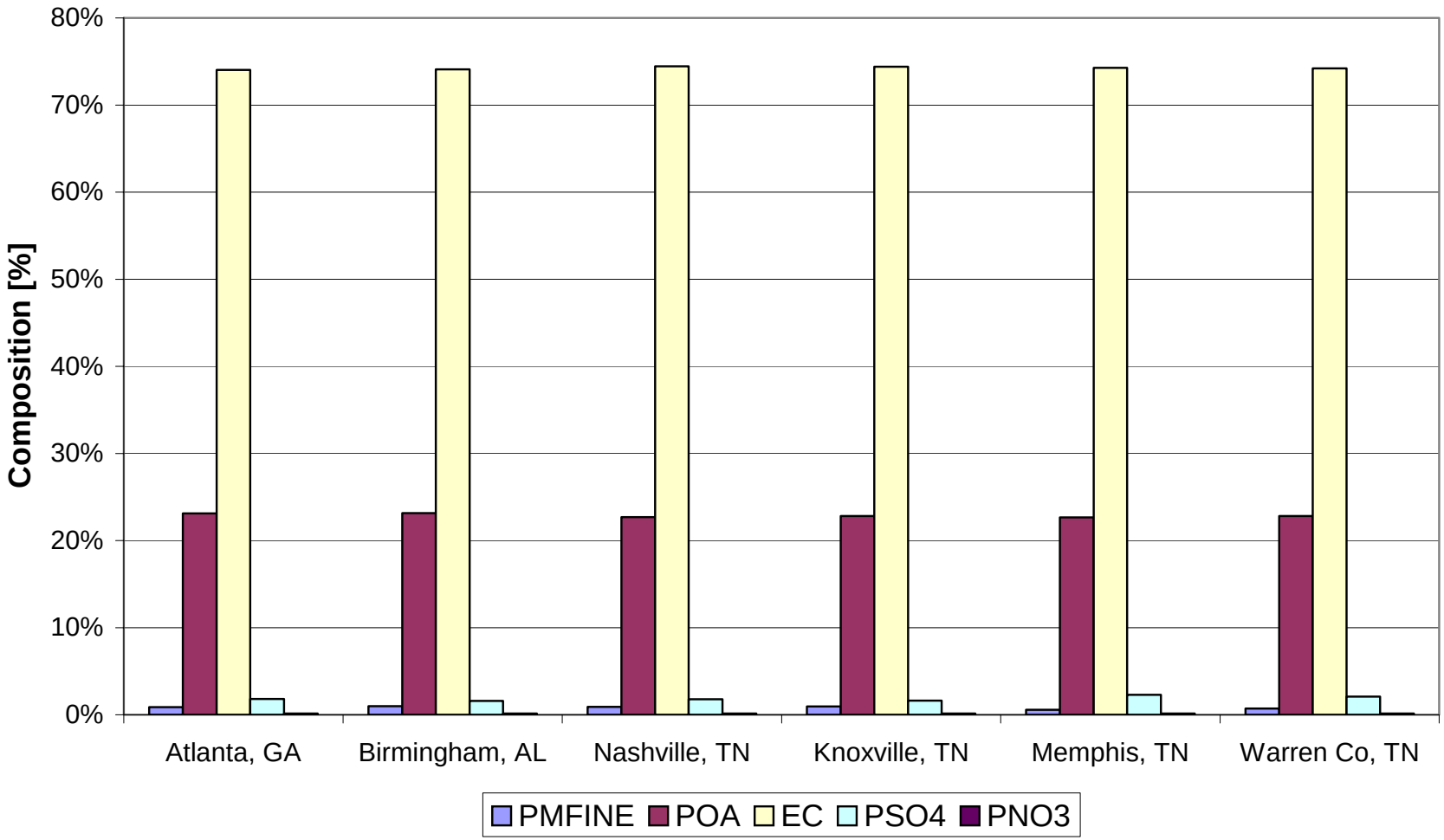


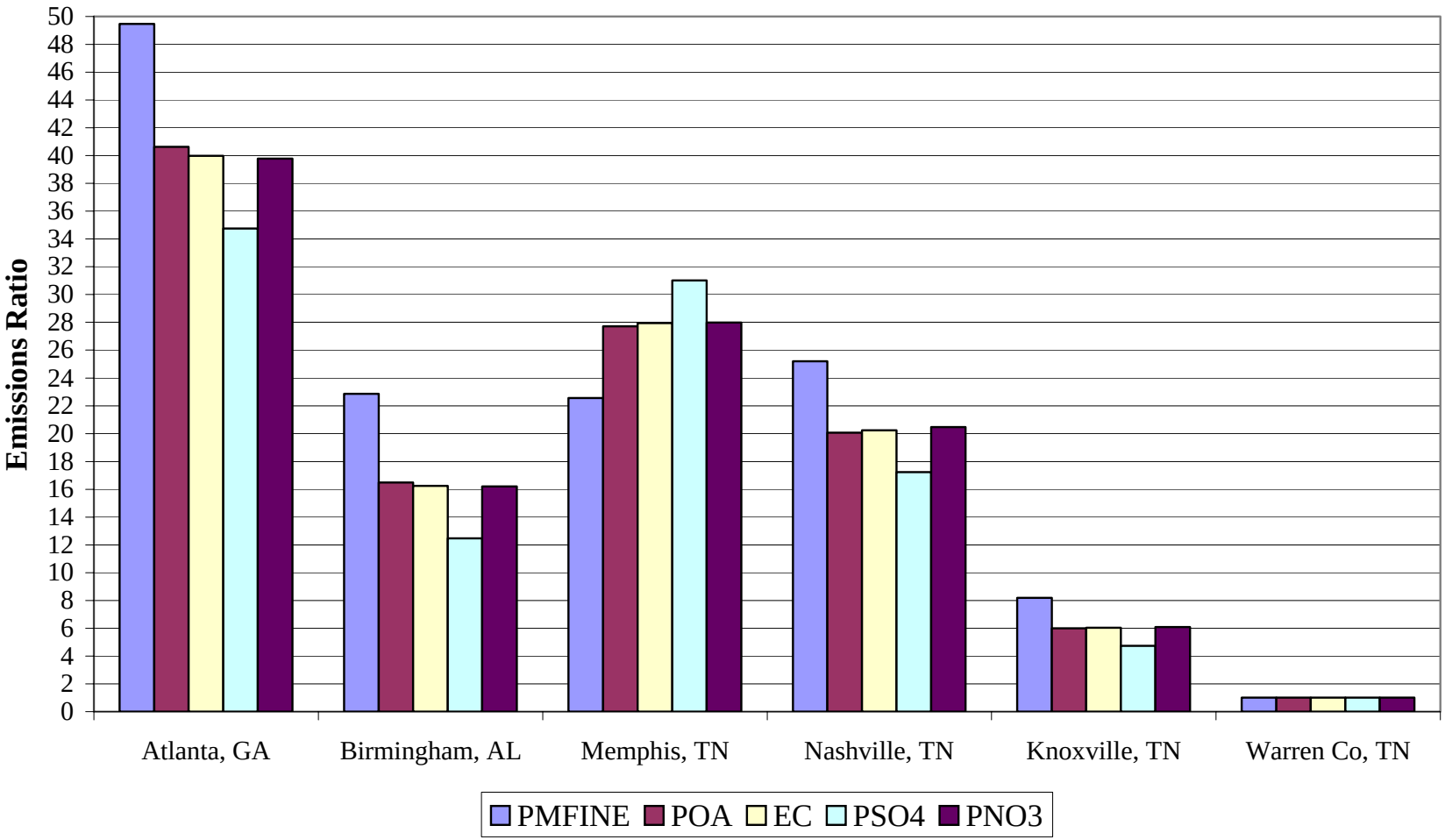
Emissions Results for DFS

Species Composition of DFS PM2.5 Emissions by Site
From 08/30 to 09/08 of 1999



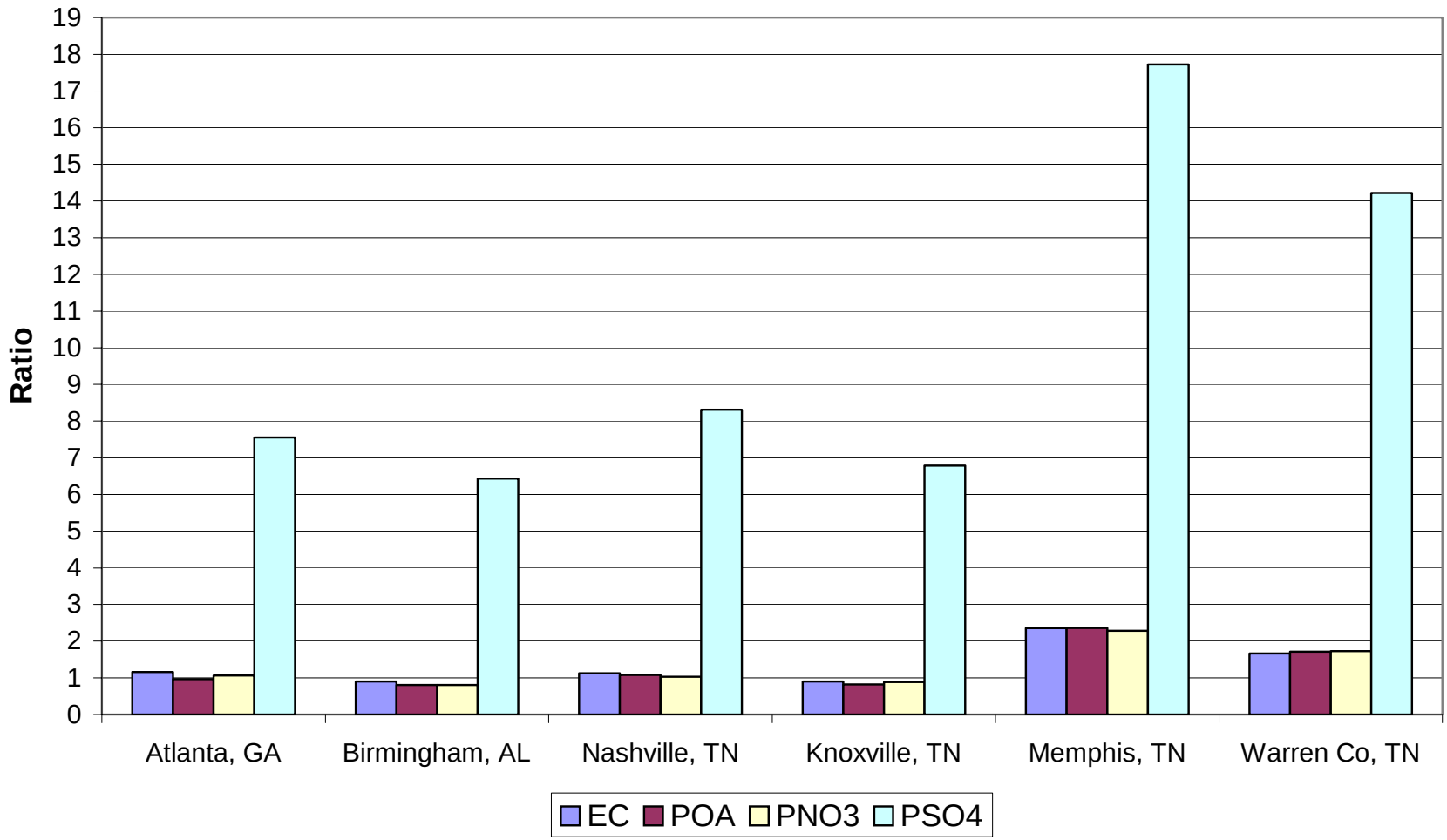
Emissions Results for DFS

Emissions Ratio to Compare PM2.5 Species from DFS by Site
08/30 - 09/08 of 1999

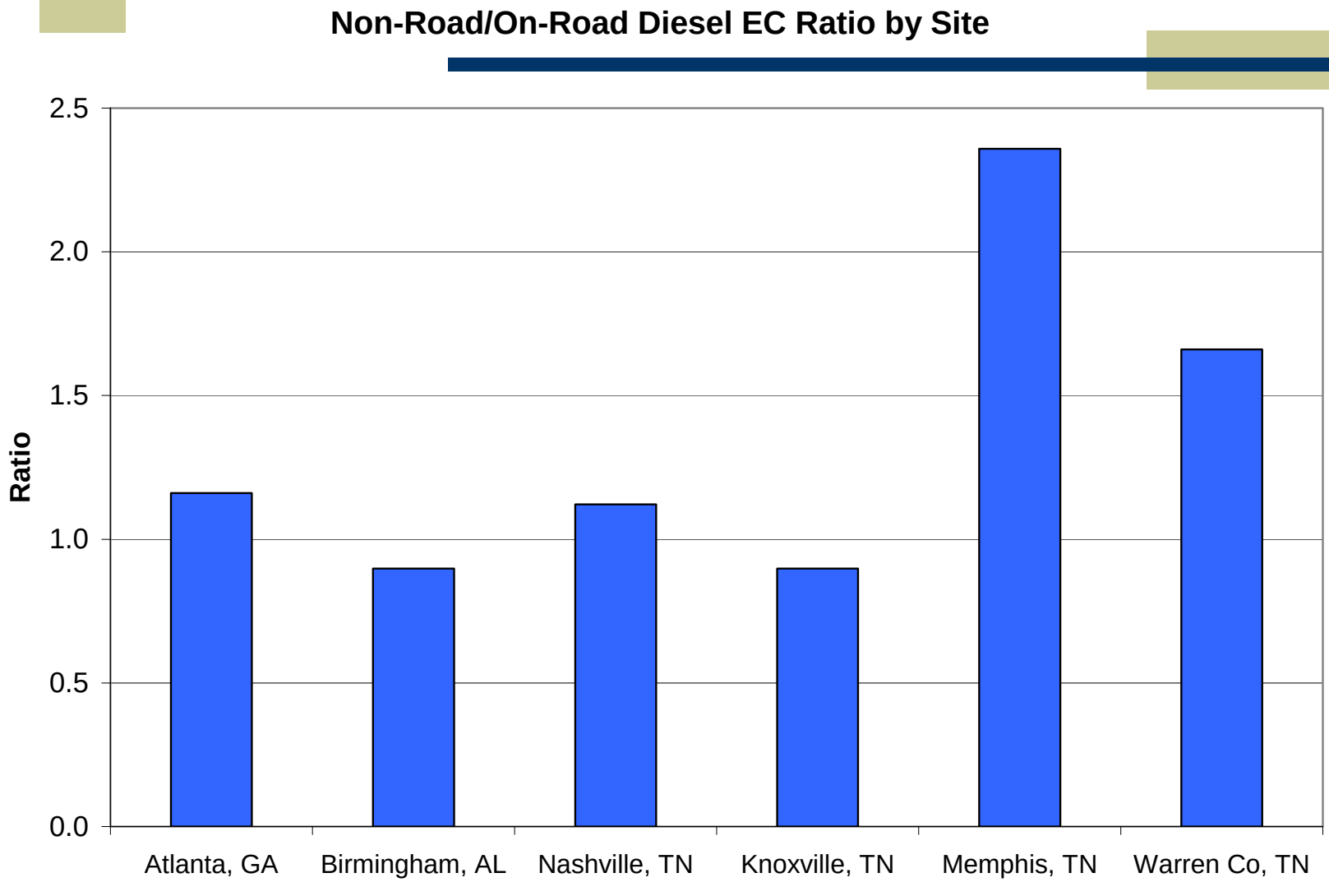


Emissions Results for DFS

Non-Road/On-Road Ratio of DFS Emissions by Site
From 08/30 to 09/08 1999

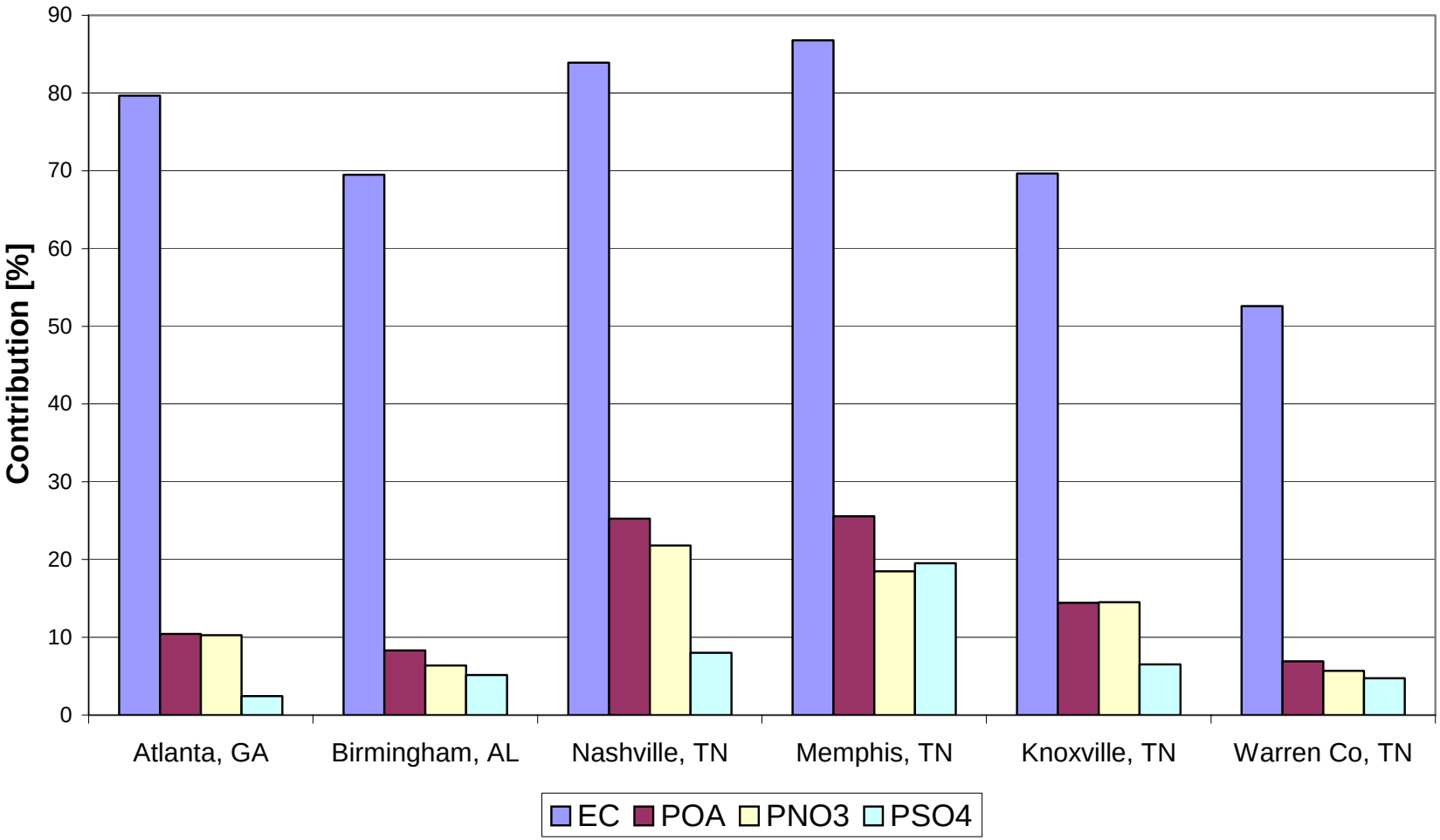


DFS EC Emissions



DFS Emissions Contribution

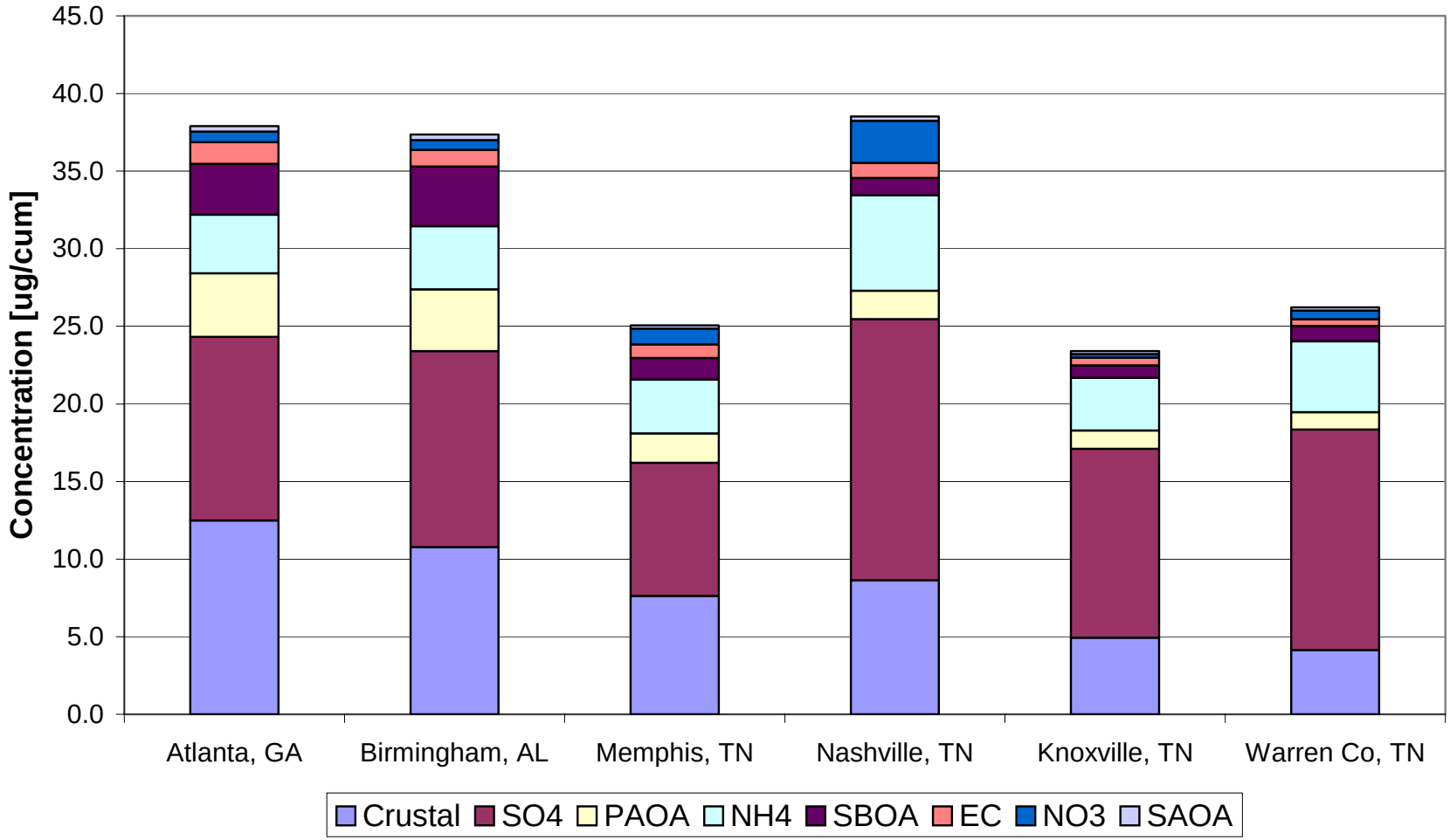
DFS Contribution to PM2.5 Species Emissions
From 08/30 to 09/08 1999



Concentrations

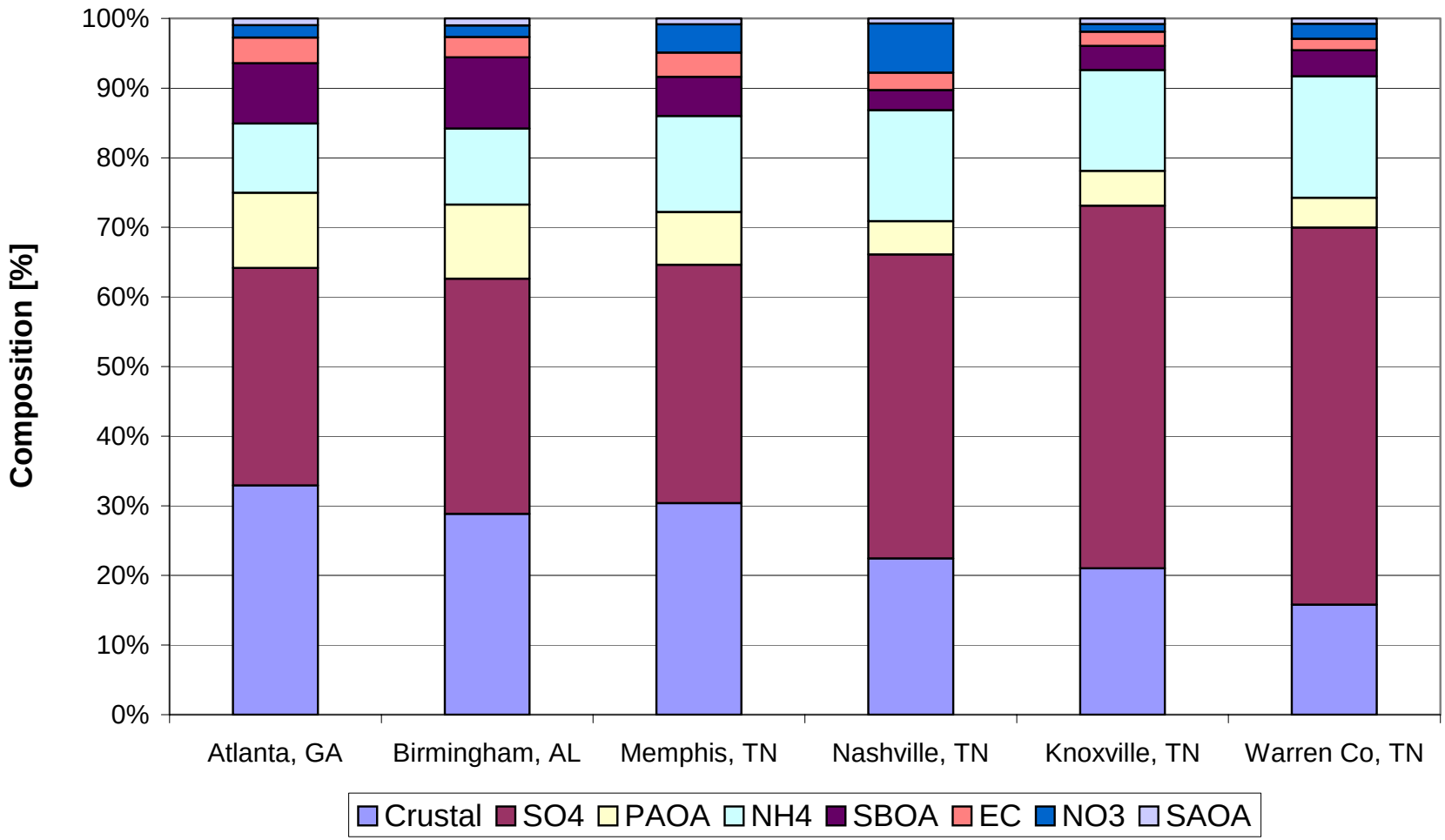
Concentrations for the BC

PM2.5 Aerosol Species by Site for the BC
From 08/30 to 09/08 1999

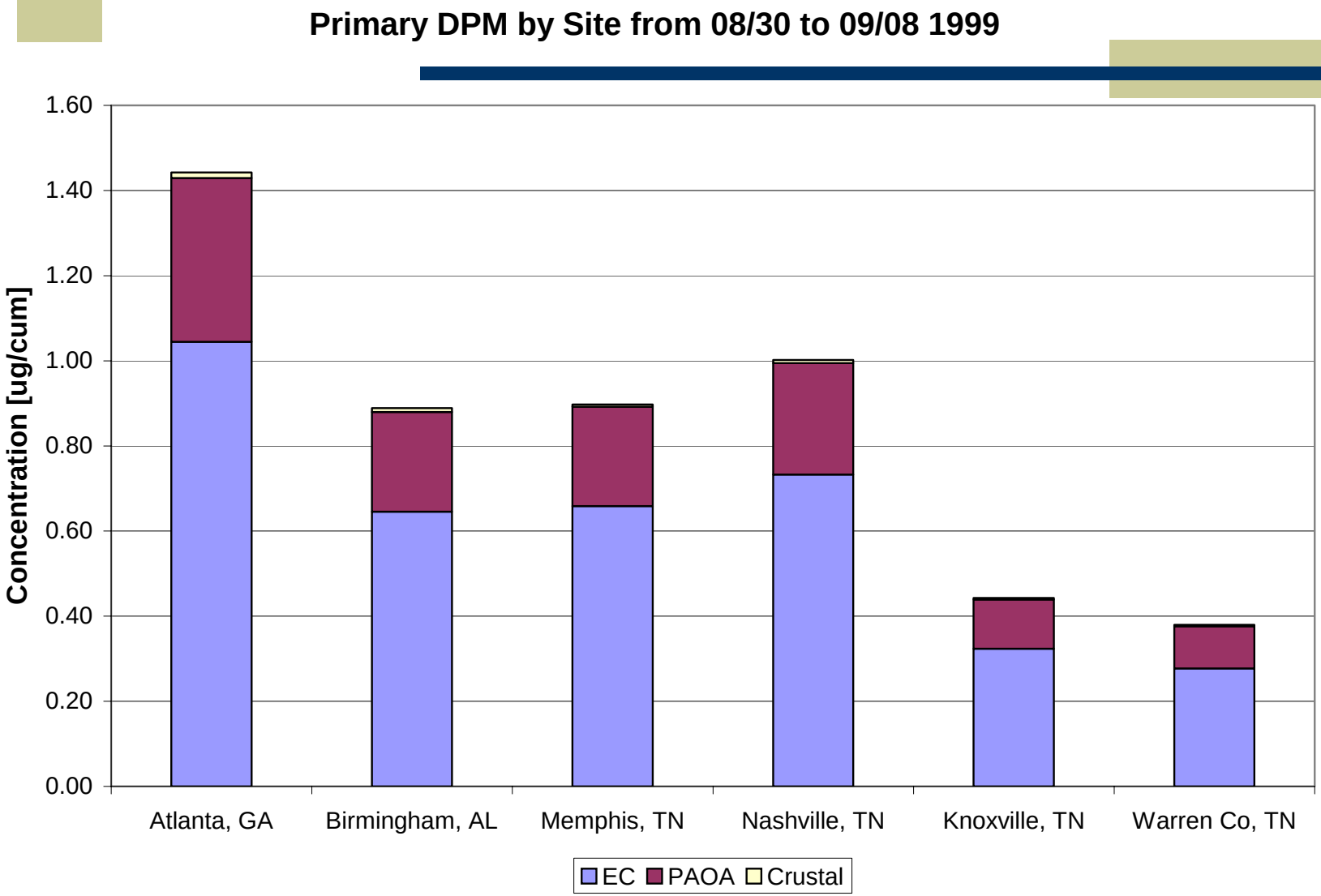


Compositions for the BC

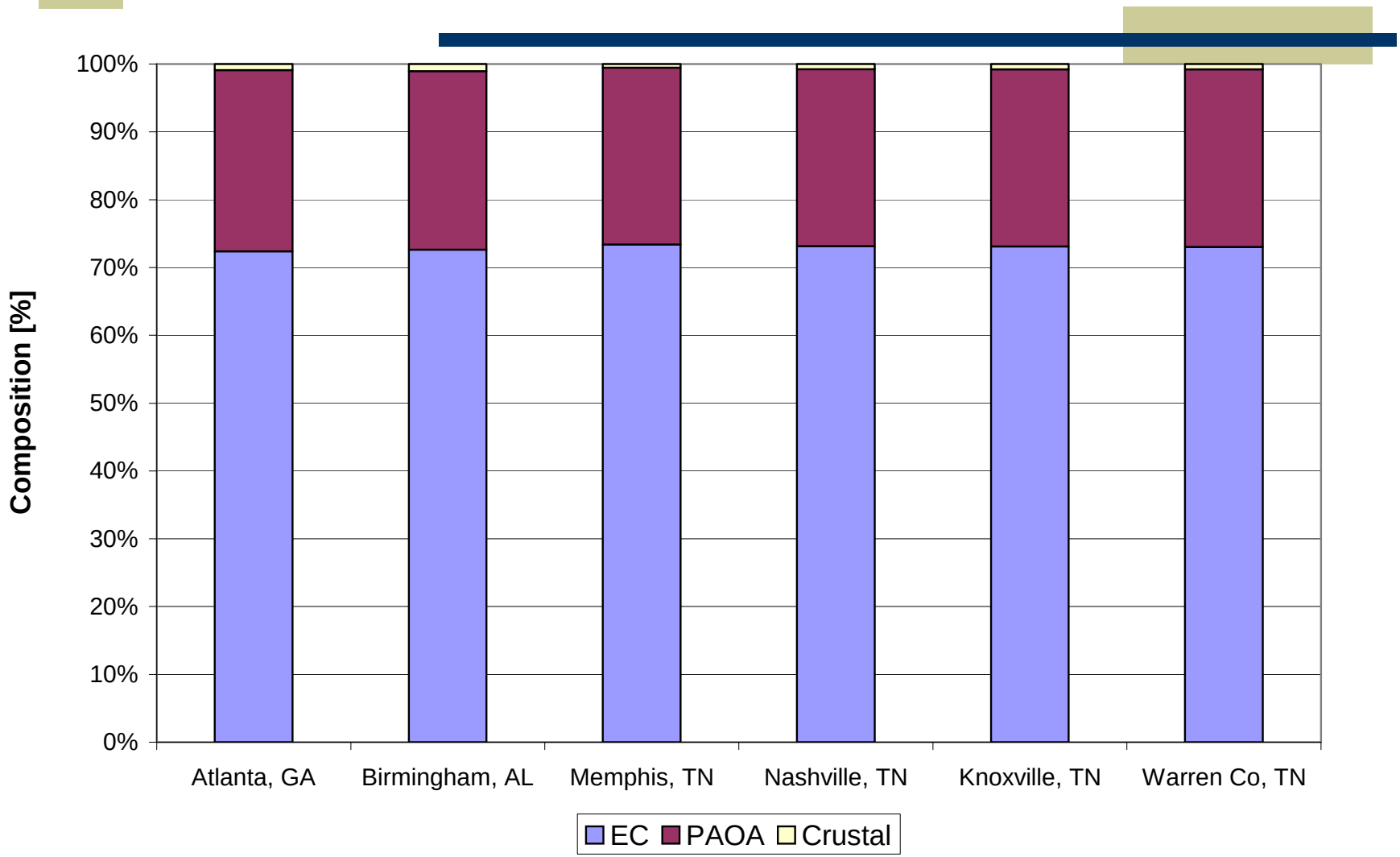
PM2.5 Composition by Site for the BC
From 08/30 to 09/08 1999



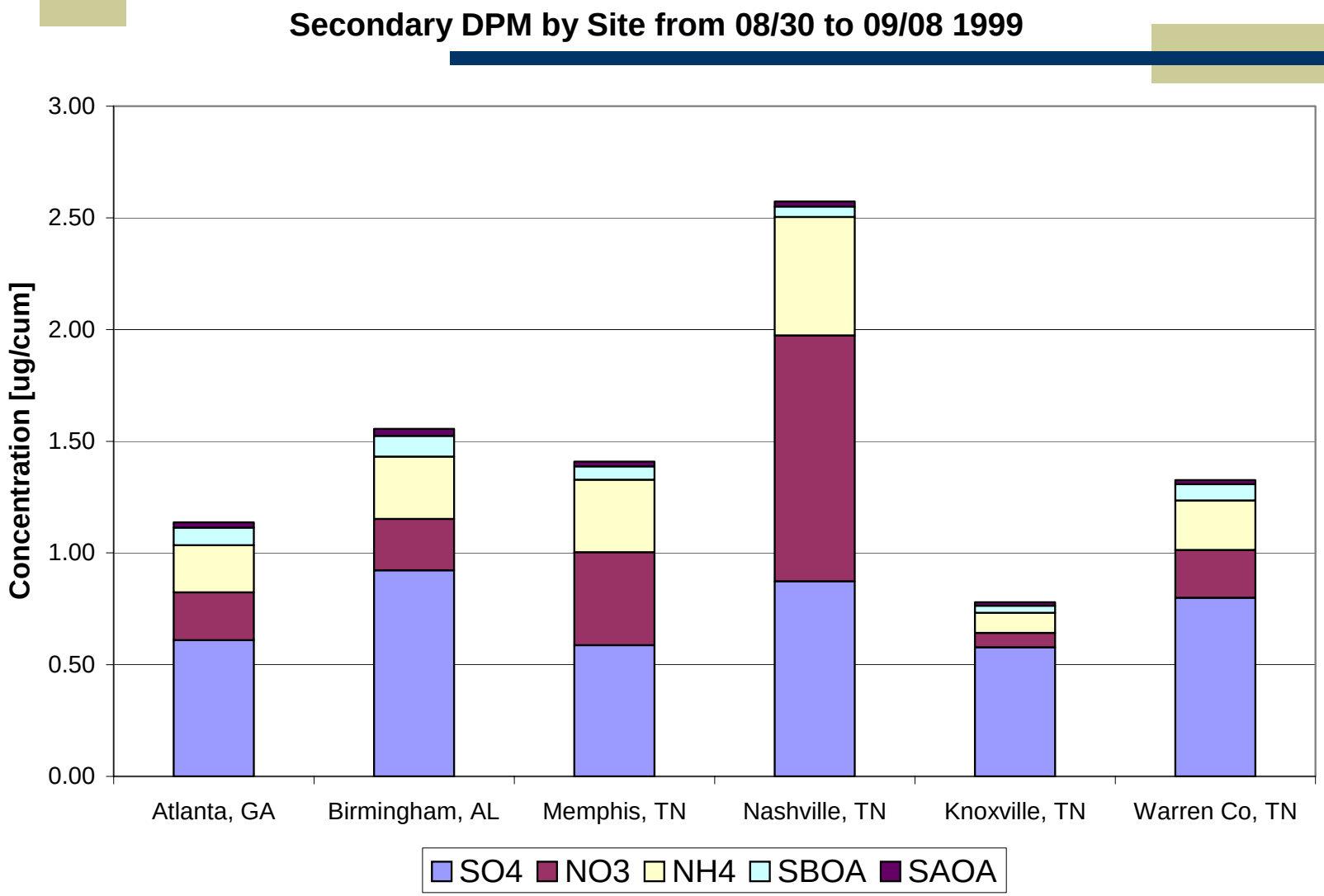
Primary DPM


Primary DPM by Site from 08/30 to 09/08 1999
Concentration [ug/cum]
1.60
1.40
1.20
1.00
0.80
0.60
0.40
0.20
0.00
Atlanta, GA
Birmingham, AL
Memphis, TN
Nashville, TN
Knoxville, TN
Warren Co, TN
EC
PAOA
Crustal

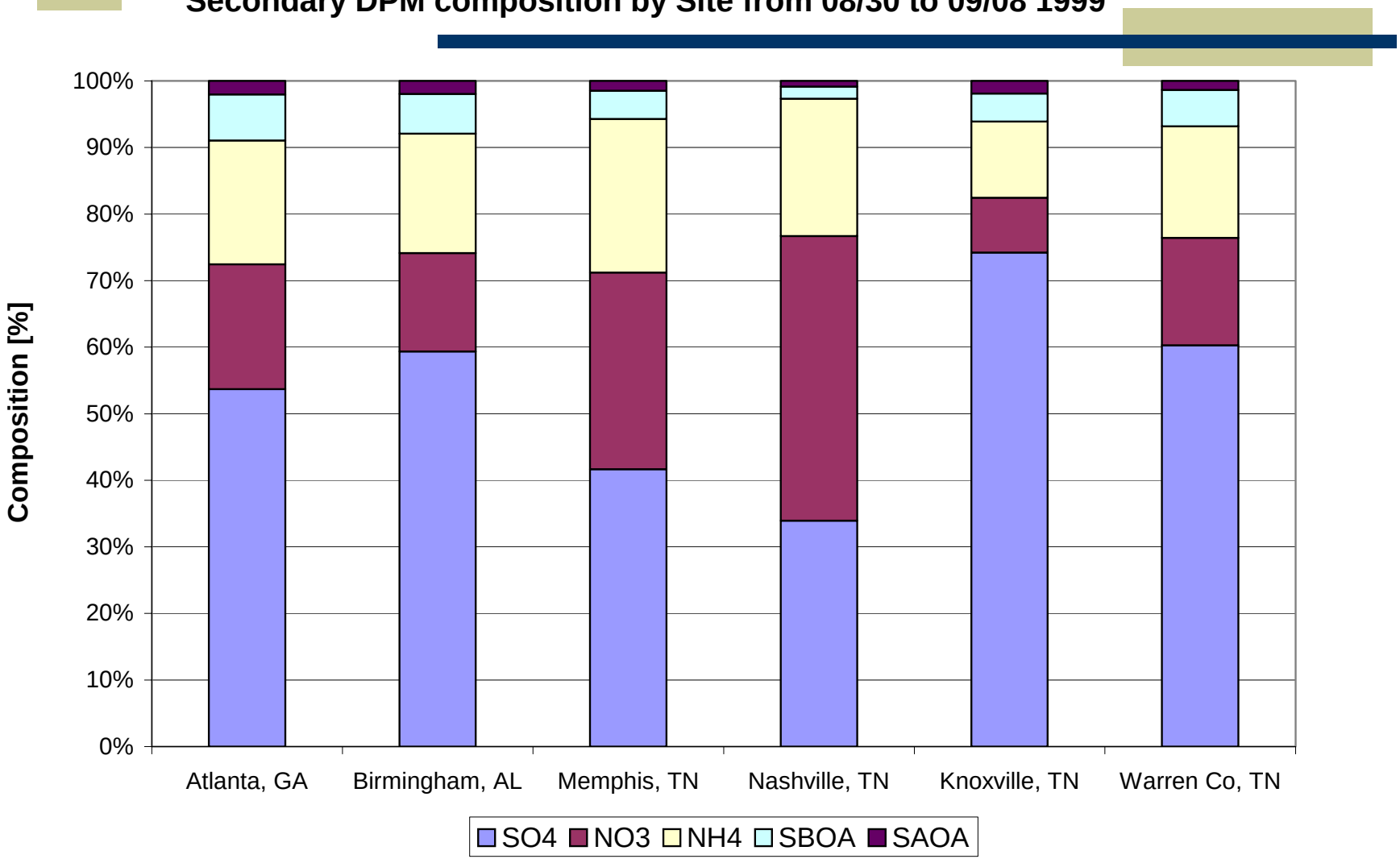
Primary DPM


Primary DPM composition by Site from 08/30 to 09/08 1999
Composition [%]
100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
Atlanta, GA
Birmingham, AL
Memphis, TN
Nashville, TN
Knoxville, TN
Warren Co, TN
EC
PAOA
Crustal

Secondary DPM

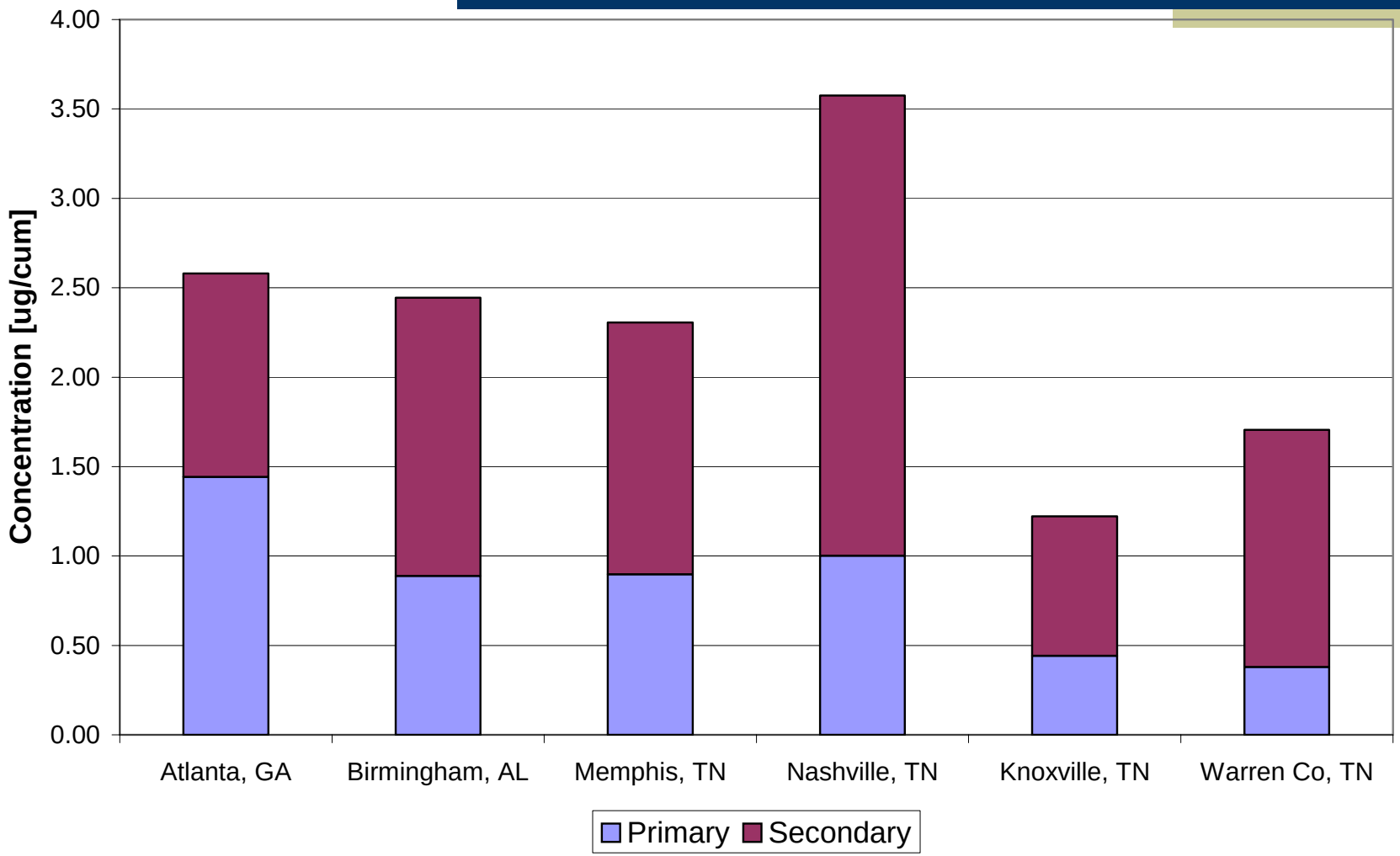


Secondary DPM



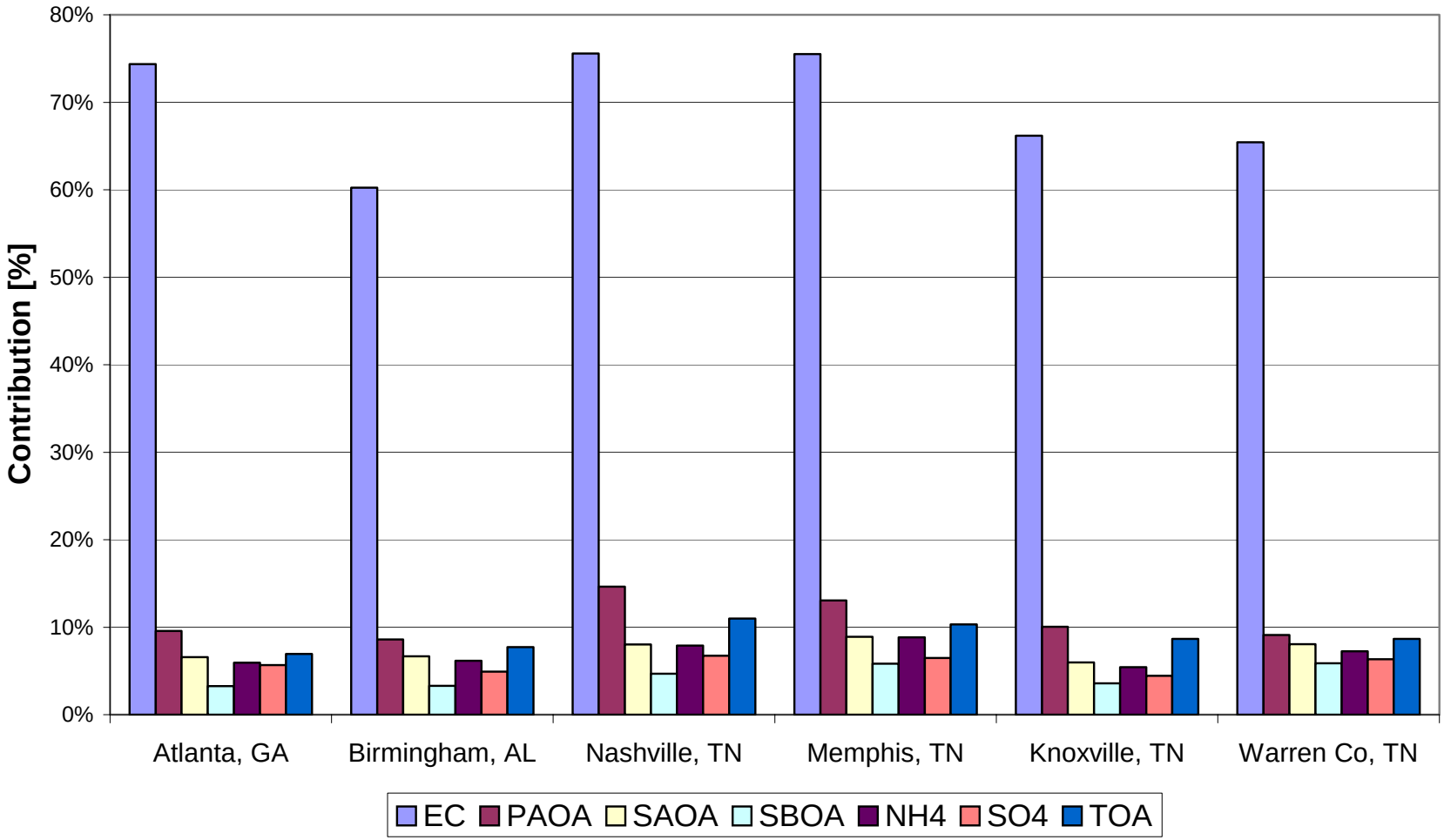
Primary & Secondary DPM

Primary and Secondary DPM by Site from 08/30 to 09/08 1999



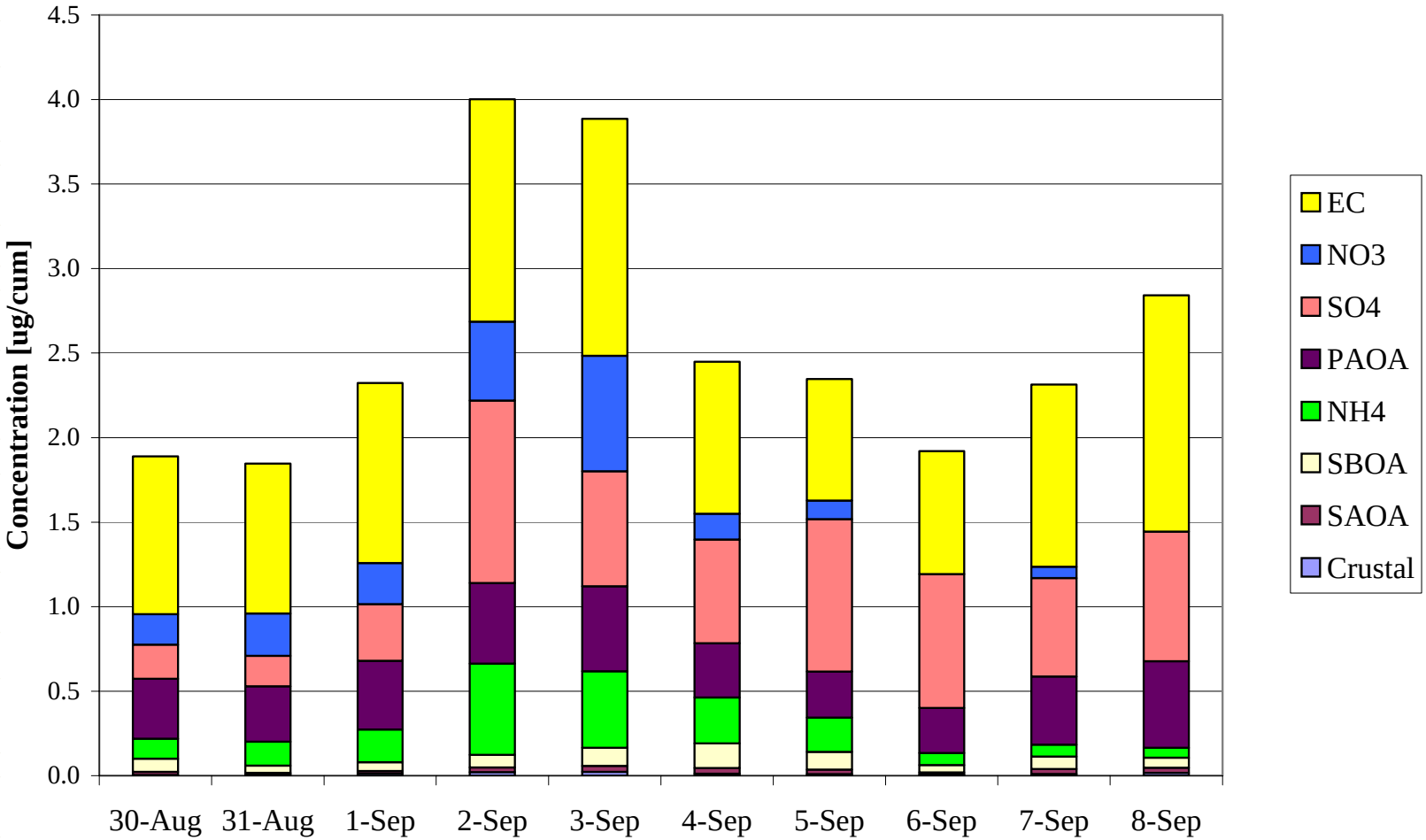
DFS Contribution

DFS Contribution to PM2.5 Aerosols by Site
From 08/30 to 09/08 1999

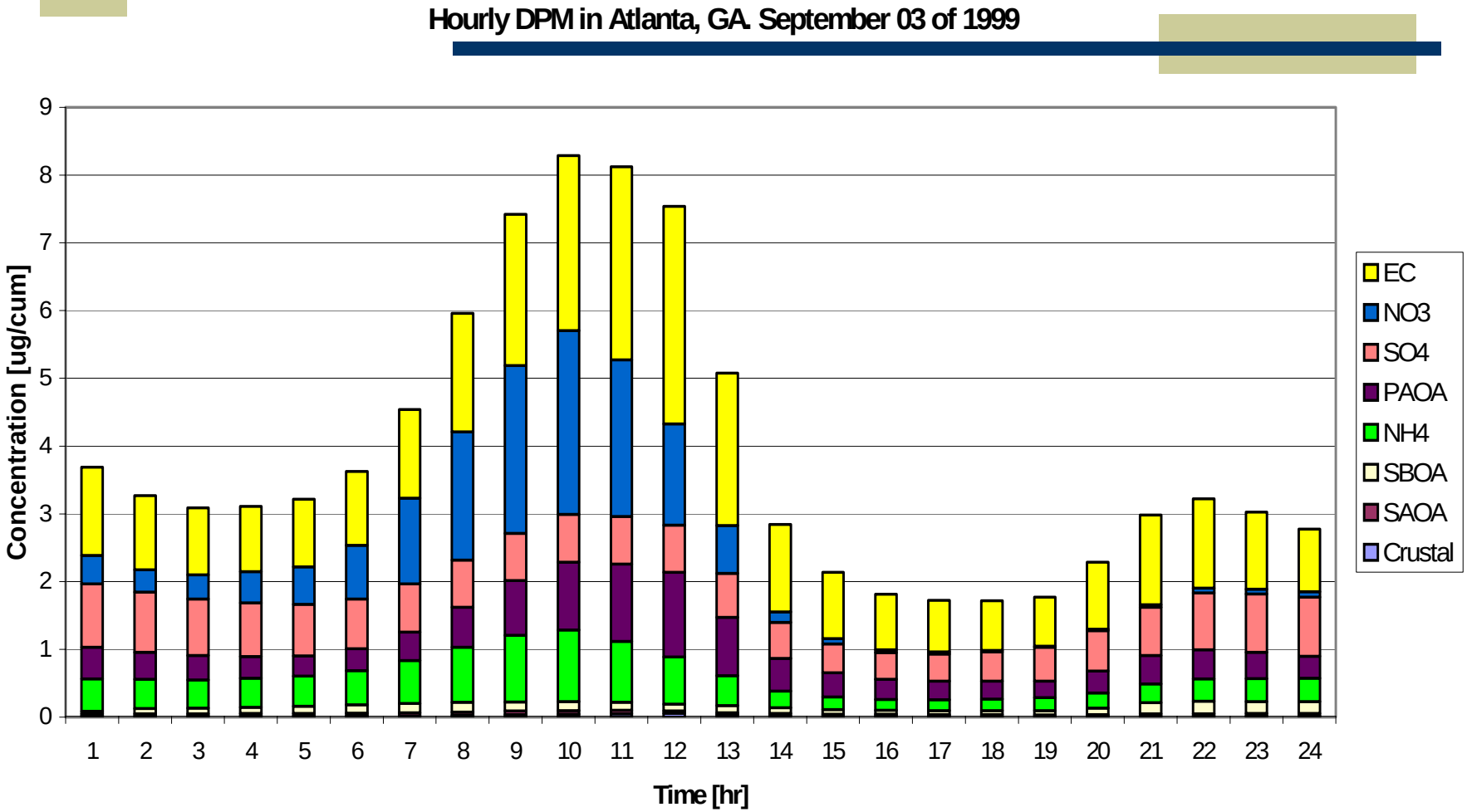


Predicted Daily DPM in Atlanta

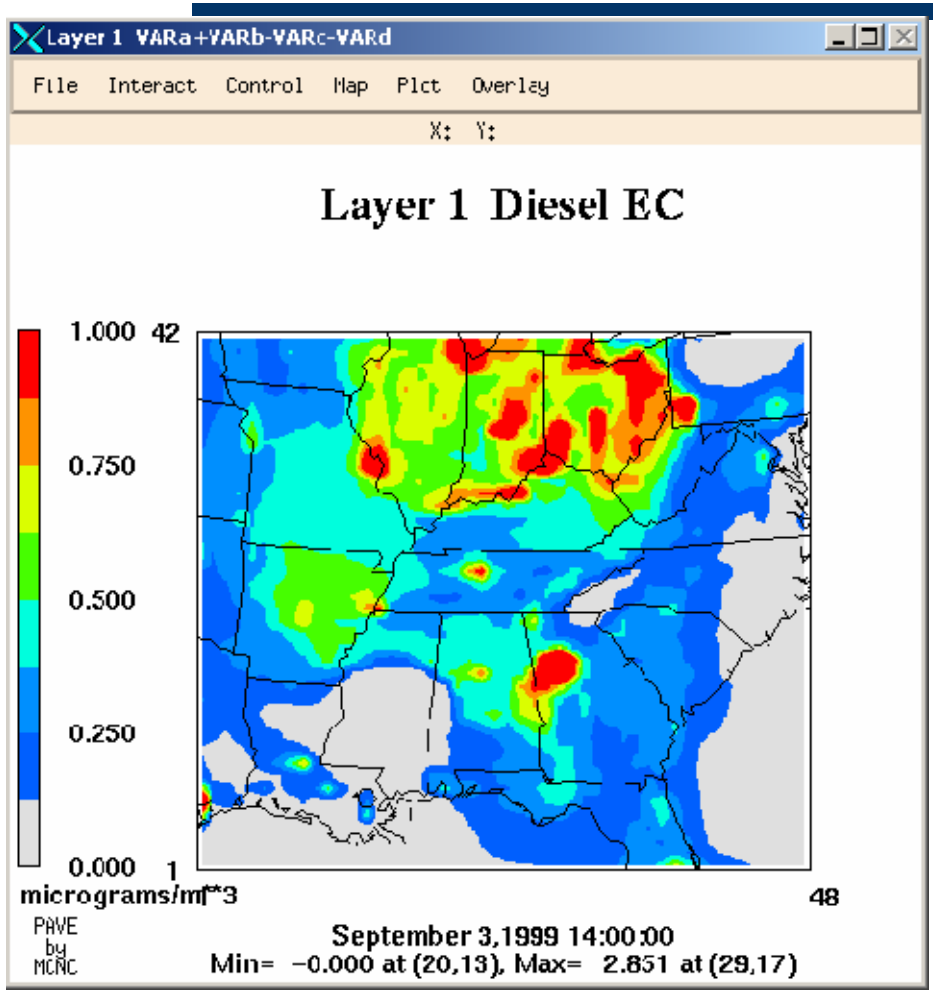
Daily Diesel PM2.5 Speciation in Atlanta, GA.



Predicted Hourly DPM in Atlanta



Diesel EC Concentrations



Conclusions

- The maximum PM2.5, NH3, and SO2 DFS emissions occurred in Atlanta, which were 40.1, 41.2, and 38.7 times respectively higher than those from the rural area Warren County TN on the studied episode.
- The maximum diesel EC and POA emissions occurred in Atlanta, which were 40.0 and 40.6 times higher than those from the rural area Warren County TN .
- DFS emissions contributed by (73.7 % ± 12.6) of EC, where the highest contribution of EC was allocated in Memphis TN, where there are more off-road sources than the other analyzed sites.

Conclusions

- TN sites showed more DFS contribution on EC mainly due to truck speed on urban interstates, which are higher than AL and GA.
- The maximum primary DPM concentrations occurred in Atlanta, Memphis, and Nashville, which were 3.8, 2.6, and 2.3 times higher than those from the rural area Warren County TN respectively.
- In average EC accounted by 72.9% of the Primary DPM followed by PAOA for each site.
- DFS contributed by (69.5% ± 6.5) of the total EC concentration.
- Significant geographic variability in the EC contribution from DFS
- The contribution over the secondary DPM aerosols was uncertain mainly on nitrate and sulfate species

Questions?