Interannual Comparison of Air Pollution Source Signal and Attribution in the Great Lakes Region

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Scientific Objectives

- Determine the sources influencing air quality measurements at a northern Michigan field site
- Compare the sources from two summers' observations at the same location
  - Source temporal signal
  - Source profile
  - Source characteristics
  - Source apportionment

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Measurements: PROPHET

- **Program for Research on Oxidants: Photochemistry, Emissions, and Transport**
- **Intensives: Spring, Summer, Fall, 1997; Winter, Summer, 1998**
- **Participants**
  - AES
  - University of Colorado – Boulder
  - California Institute of Technology
  - Carnegie Mellon University
  - ERIM
  - **Michigan Technological University**
  - NCAR
  - Ohio State University
  - Ohio University
  - Pennsylvania State University
  - Purdue University
  - Stanford University
  - SUNY – Old Westbury
  - University of Miami
  - University of Michigan
  - University of Virginia
  - Washington State University
  - Western Michigan University
  - York University

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Measurements: Particulate

- **Particulate mass concentration**
  - **Continuous $PM_{2.5}$: Rupprecht and Patashnik TEOM**
- **Particulate number concentration**
  - **Fine: TSI scanning mobility particle sizer**
  - **"Coarse": PMS laser spectrometer**
- **Particulate characterization**
  - **4 hour integrated filter samples**
  - **Scanning electron microscopy**
  - **MetOne meteorological station**
  - **Trace gases: O_3, CO, PAN, isoprene**

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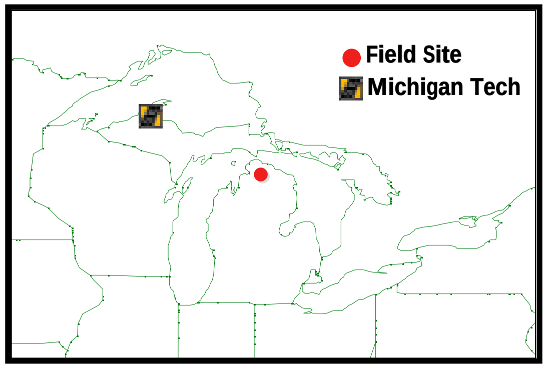
Measurements: Location



- Measurements
  - July 29 to August 17, 1997
  - Seven species
  - 2116 5-minute observation series

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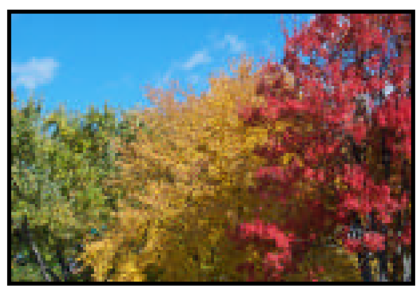
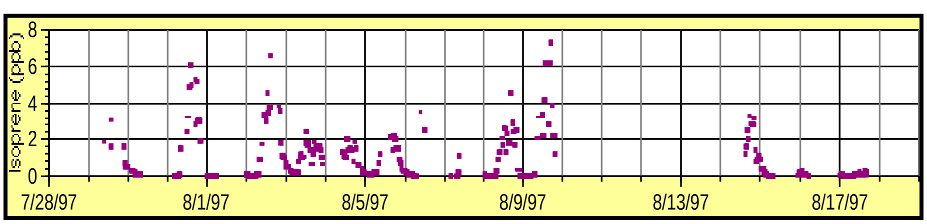
Data: Isoprene

- Sources
  - Deciduous trees
- Sinks
  - Reactions with OH•, O_3, and NO_3 (nighttime)
- Approximate lifetime
  - Hours





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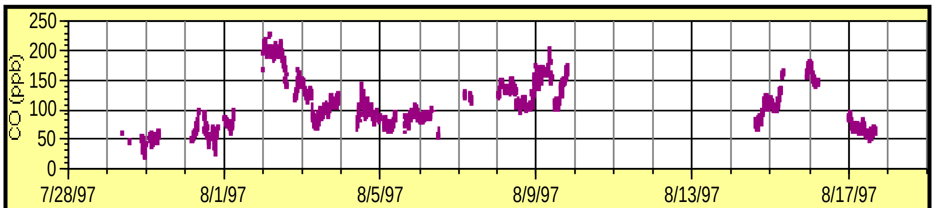
Data: Carbon Monoxide

- Sources
  - Combustion, industrial processes, oxidation of HC
- Sinks
  - Oxidation by OH•
- Approximate lifetime
  - Months



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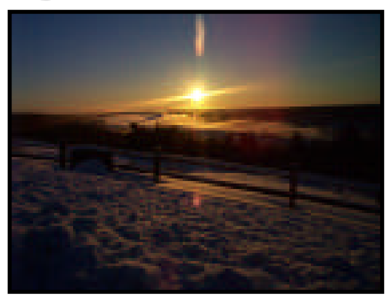
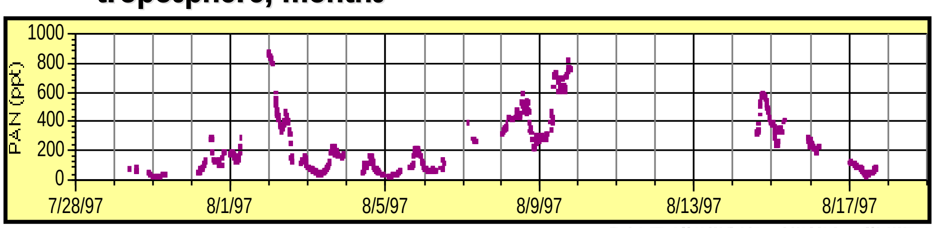
Data: Peroxyacetyl Nitrate

- Sources
  - Oxidation of HC with NO_X
- Sinks
  - Thermal decomposition
- Approximate lifetime
  - Temperature dependent, in free troposphere, months





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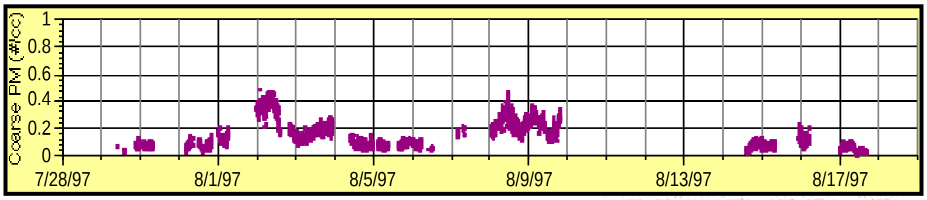
Data: “Coarse” Particulate

(D_p > 1.50 μm)

- Sources
  - Mechanical processes
- Sinks
  - Fallout (due to size)
- Approximate lifetime
  - Hours



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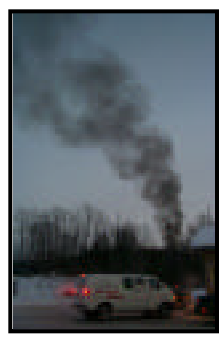
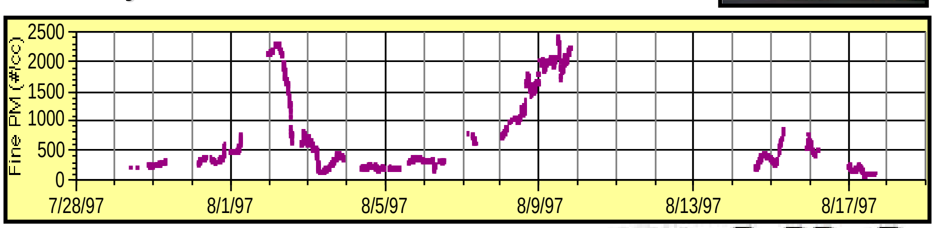
Data: Fine Particulate

(0.12 μm < D_p < 1.50 μm)

- Sources
  - Combustion (primary particles), coagulation, gas-to-particle conversion (second particles)
- Sinks
  - Washout, coagulation, and deposition
- Approximate lifetime
  - Days





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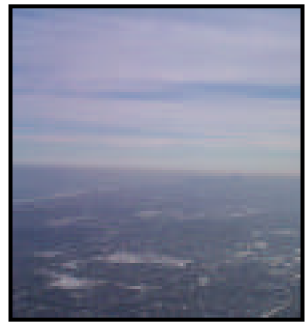
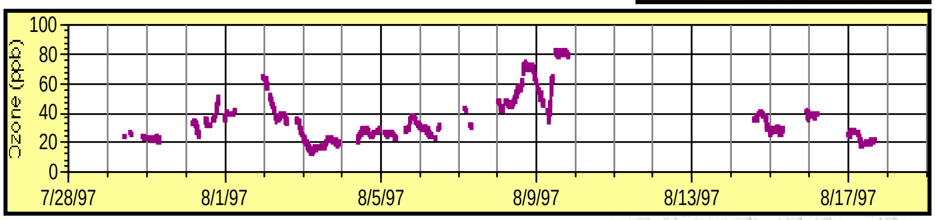
Data: Ozone

- Sources
  - Photochemical production via NO_X-HC-CO chemistry
- Sinks
  - Dry deposition, photolysis and other
- Approximate lifetime
  - Varies, hours to month





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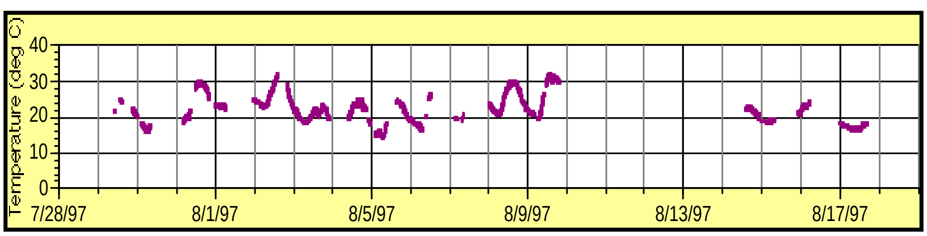
Data: Temperature



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Data: Methods

- Measurements and Modeling
  - Generate data necessary to assess conditions, dynamics, and transport
    - Integrate meteorological, atmospheric trace gas, and particulate measurements
    - Trajectory and residence time modeling
  - Data analysis of sources
    - Positive Matrix Factorization (PMF)
    - Correlations with potential influences

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Data: Analyses

- Source attribution
  - Positive matrix factorization
  - Particle distributions, dynamics, and composition
  - Composition and dynamics of sources
  - Transport modeling
- Meteorological correlations
  - Wind speed
  - Wind direction
  - Temperature
  - Humidity

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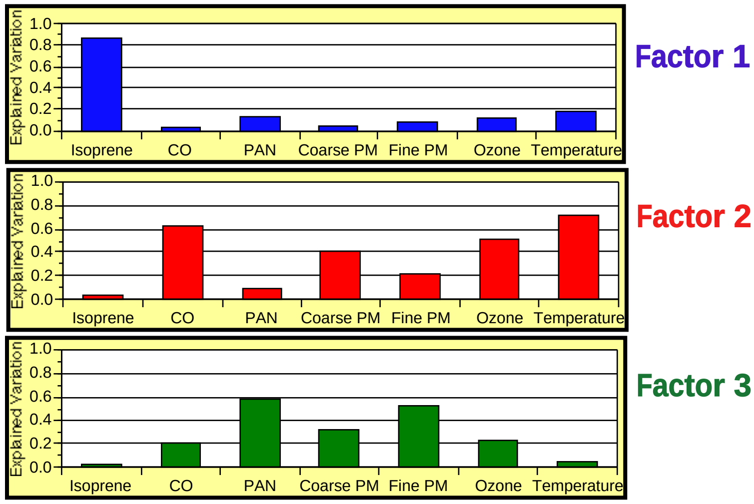
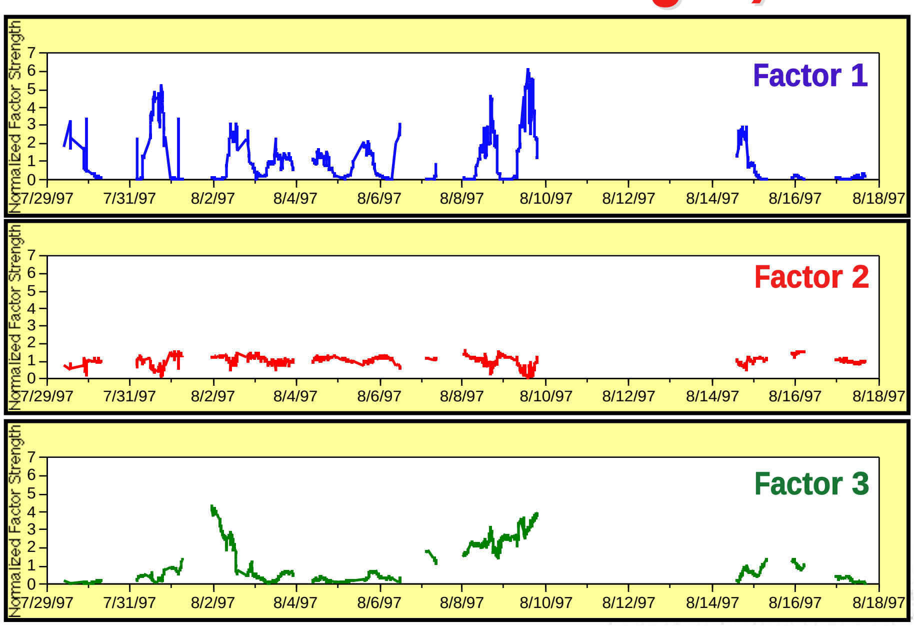
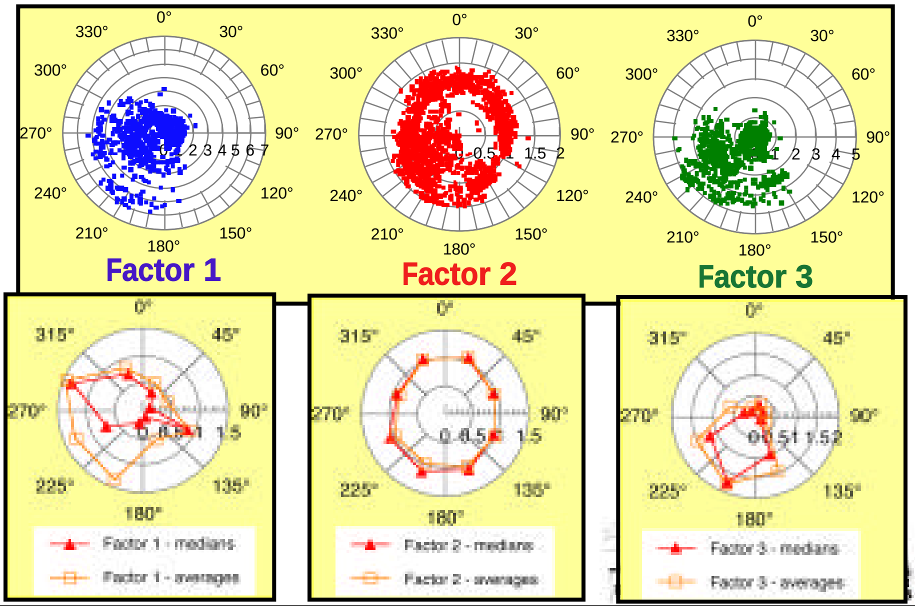
PMF: Source Profile, EV



PMF: Source Strength, G

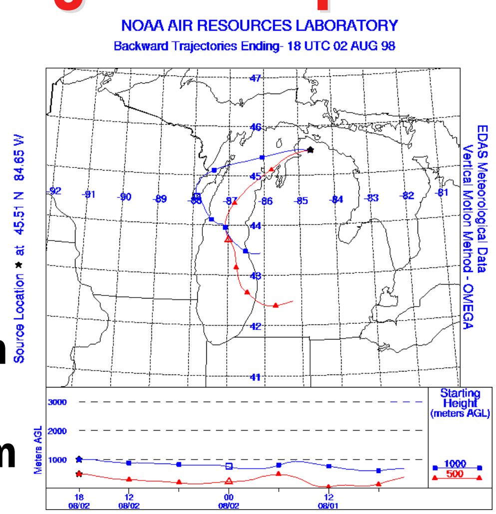


PMF: Wind Analysis



PMF: Long-Range Transport

- Assess Factor 3
  - G: +/- 1 std dev for dirty and clean events
  - Three-day back trajectories
  - Residence time
- Dirty air arrives from industrial Midwest
- Clean air arrives from central Canada



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PMF: Source Identification

- Three factor PMF solution
  - Factor 1 = *Biogenic Sources*
    - Diurnal signal
    - Unique to isoprene
  - Factor 2 = *Local Sources*
    - Insensitive to wind direction
    - Correlates with temperature, CO, coarse PM
  - Factor 3 = *Long-Range Transport*
    - Low frequency signal
    - Contains PAN, fine PM (aged air)

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PMF: Source Apportionment, F

	Isoprene (ppb)	CO (ppb)	PAN (ppt)	Coarse PM (#/cm^3)	Fine PM (#/cm^3)	Ozone (ppb)	Temp. (°C)
Factor 1 (biogenic)	1.128	4.0	25	0.006	43.2	5.4	5.0
Factor 2 (local)	0.003	71.0	11	0.053	87.7	18.2	16.0
Factor 3 (transport)	0.002	29.4	174	0.067	499.8	11.4	1.2
Sum of factor contributions	1.132	104.4	210	0.126	630.7	35.1	22.2
Measured mean	1.136	107.6	235	0.447	705.0	35.9	22.3
% difference	-0.3	-3.0	-10.4	-71.8	-10.5	-2.1	-0.5

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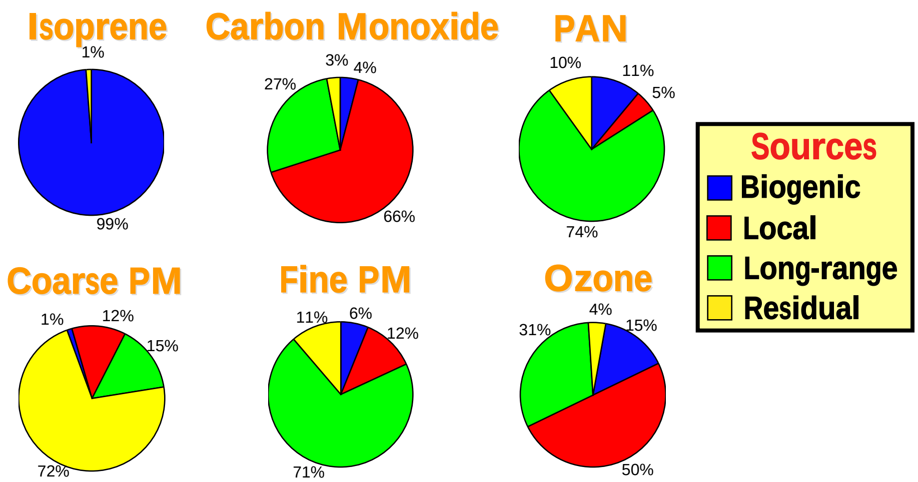
PMF: Source Apportionment



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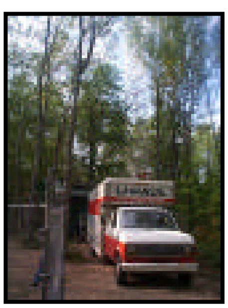
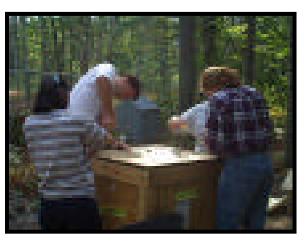
Measurements: Repeat

- Summer Campaign #2
  - PROPHET site
  - July 27 to August 16, 1998





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Measurements: Averages

Constituent	1997	1998	Change
Isoprene (ppb)	1.132	1.561	+37.9%
CO (ppb)	104.4	210.2	+101.7%
PAN (ppt)	210.0	219.8	+4.7%
Coarse PM (#/cm^3)	0.126	0.265	+110.3%
Fine PM (#/cm^3)	630.7	1019.5	+61.6%
Ozone (ppb)	35.1	46.7	+33.1%
Temperature (°C)	22.2	23.4	+5.4%

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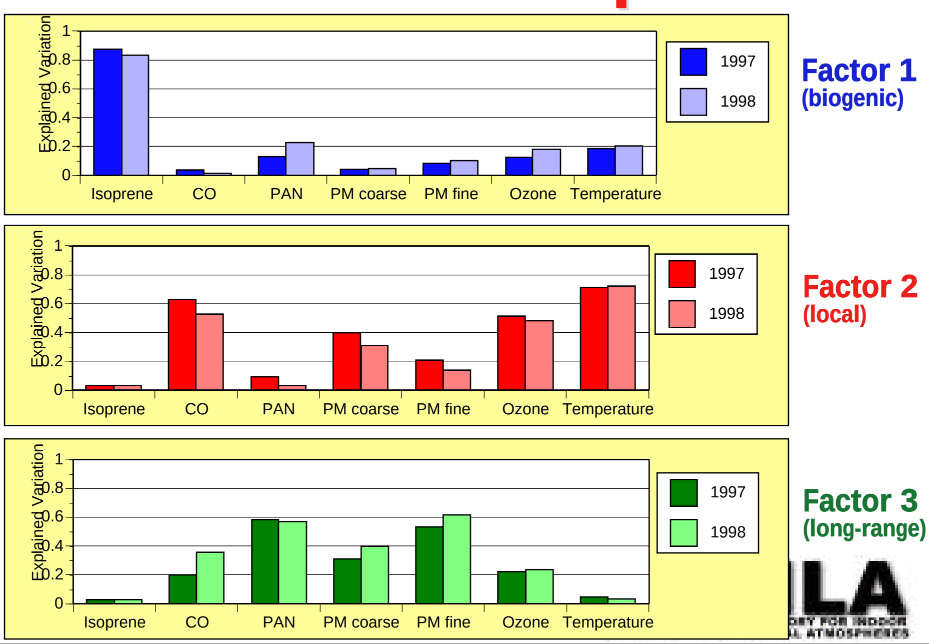
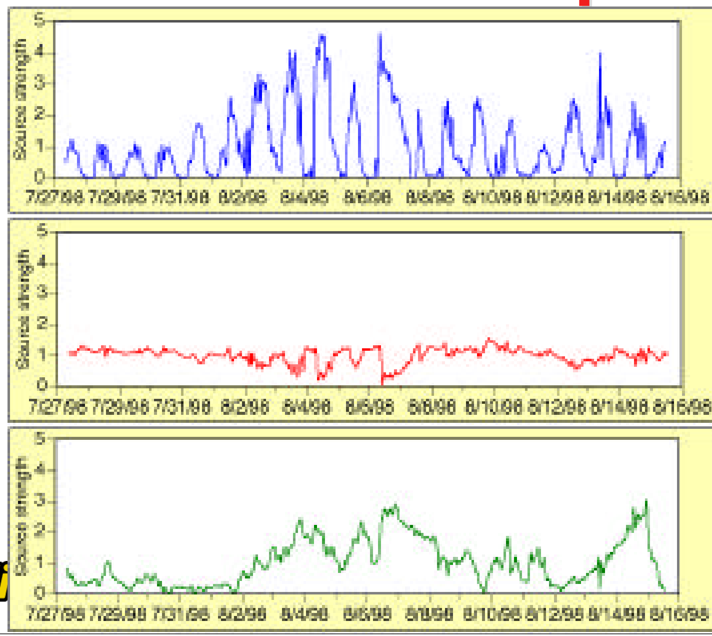
PMF: Source Comparison



PMF: Source Comparison



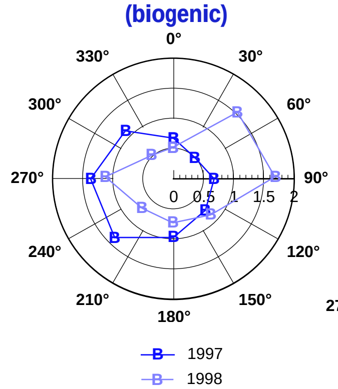
Factor 1
(biogenic)

Factor 2
(local)

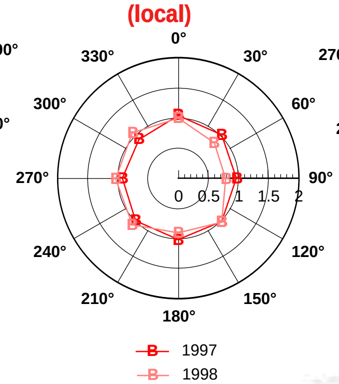
Factor 3
(long-range)

PMF: Source Comparison

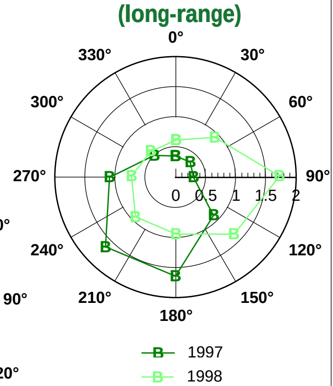
Factor 1
(biogenic)



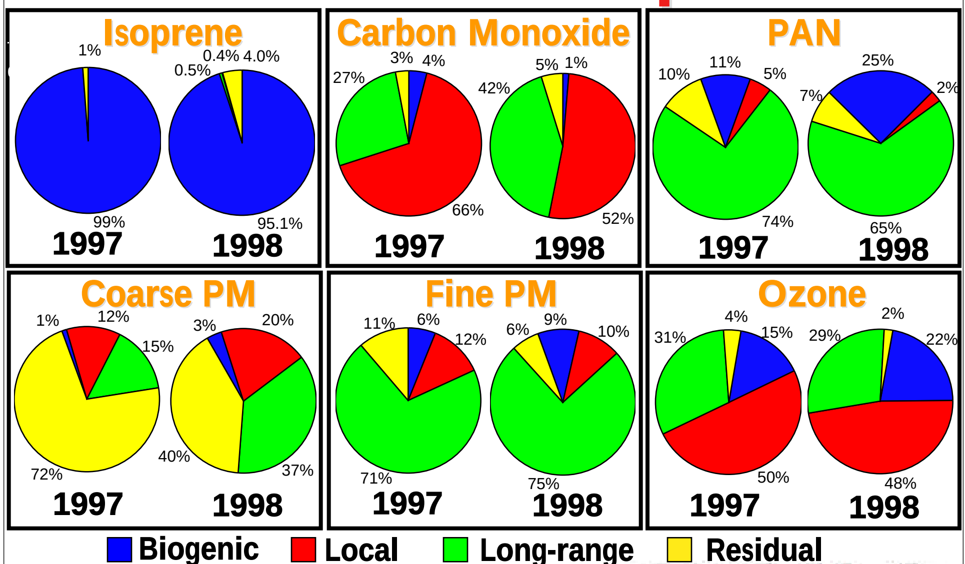
Factor 2
(local)



Factor 3
(long-range)



PMF: Source Comparison



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Conclusions

- Three sources (local, long-range, biogenic) effectively explain the measurements in summers 1997 and 1998
- Source temporal signals
  - Each source exhibited similar temporal characteristics (frequency and amplitude) both summers
- Source profile
  - Each source had a similar profile both summers
- Source characteristics
  - Different transport patterns in the two summers resulted in different source regions for the long-range and biogenic sources
- Source apportionment
  - In general, there were slight increases in long-range transport and decreases in local source contributions in the summer of 1998

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Next Steps

- Increase Number of Pollutants
  - PROPHET site, summer 1998
  - 17 distinct constituents
  - 6 sources
- Evaluate metrics for quantitative comparison of sources

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