Total Non-Methane Organic Carbon

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Motivation

- **Goals:** 1. Measure total non-methane organic compounds (TNMOC), and

- 2. *Determine the relationship between TNMOC and the sum of the speciated volatile organic compounds (VOC’s) measured by standard techniques (Gas chromatograph/flame ionization detector).*

- VOC’s are one of the key determinants of air quality and control strategies.

- Standard measurement methods are known to detect hydrocarbons and their oxidation products incompletely.
Possible Types of Excess TNMOC Compared to the Sum of Speciated VOC’s
(Standard VOC Measurement)

- Compounds that are lost in the inlet or column (polars, semi-volatiles).
- Compounds that are obscured in the GC baseline (hydrocarbons).
- Heteroatom compounds that have a reduced response in an FID.
Possible Sources of Excess TNMOC Compared to the Sum of Speciated VOC’s in Ambient Air

- Photochemical oxidation of hydrocarbons.
- Direct source emissions. These may be either oxygenates or semi-volatile hydrocarbons.
Method

• Trap VOC’s from 2 ambient air samples simultaneously in a cryogenically cooled trap. Allow CO, CO$_2$, and CH$_4$ to pass through.
• Desorb both VOC samples.

Speciated VOC’s: analyze directly with DB-1 Column, GC/FID. = Standard Measurement

TNMOC: oxidize CO$_2$, analyze as methane w/ GC/FID. = Total VOC’s

3. Compare TNMOC with the standard measurement of VOC’s.
Flow Schematic

Valve A:B: Sampling position
Valve E: non-oxidation position
Oxidation Catalyst

Valve C,D: Injection position

Valve A,B: Sampling position
Valve E: non-oxidation position
Oxidation Catalyst

Valve C,D: Injection position

Valve E: non-oxidation position
Oxidation Catalyst

Valve C,D: Injection position

Valve E: non-oxidation position
Oxidation Catalyst

Valve C,D: Injection position
Trap I Design

- Trap I Design
- Grooves for trap tubing: D: 6.2mm
- Grooves for thermocouples: D: 1mm
- O.D.: 6 mm, I.D.: 5.3 mm
- Quartz wool plug
- Fused silica Beads
- Liquid nitrogen vent
- Liquid nitrogen
- Screws
- 30 mm
- 140 mm
- 100 mm
- 30 mm
- 20 mm
- 60 mm
TNMOC = 620 ppbC

Sum of Speciated VOC’s = 450 ppbC
Trapping Efficiency vs. Temperature

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**Trapping efficiency (%)**

- CO$_2$
- 1-hexene (1)
- 1-pentene (1)
- 1-butene (1)
- propene (1)
- 1-hexene (2)
- 1-pentene (2)
- 1-butene (2)
- propene (2)
- ethanol
- acetaldehyde
- formaldehyde

**Equivalent Trapped CO$_2$ (ppbC)**

- Temperature of Trap I (°C)

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-140 -120 -100 -80 -60 -40 -20 0

0 20 40 60 80 100 120

0 200 400 600 800 1000 1200
Correction for Light Hydrocarbons

• Hydrocarbons with 4 or 5 carbon atoms trap with efficiencies between 5 and 80% and C₂ and C₃ hydrocarbons only minimally. Acetaldehyde, methanol, ethanol and acetone etc. also elute in this region, and are collected at 100%.

• Loss of C₃ - C₅ hydrocarbons was corrected as follows:

• 20 samples were trapped at –100 °C where 100% of C₄ and C₅’s trap and 64% of C₃’s trap. These were compared to the chromatograms collected at –60 °C immediately before and after.
Light Hydrocarbons

-60 C

-100 C

-60 C
Correction for Light Hydrocarbons

- $C_3$ concentrations were corrected using their trapping efficiencies.
- Peaks eluting between $C_3$ and $C_5$ were normalized to the sum of spec. VOC’s at $-100 \, ^\circ C$.
- The result was compared to the $-60 \, ^\circ C$ chromatograms, also normalized.
- The difference, equal to the lost $C_3$-$C_5$ hydrocarbons at $-60 \, ^\circ C$, averaged 12.5% of the sum of speciated VOC’s at $-60 \, ^\circ C$.
- 6.5% was added to account for untrapped $C_2$’s, based on SCOS-97 Azusa data (McCauley, 1999).
Correction for Light Hydrocarbons

• Because the lost light hydrocarbons are added to both the TNMOC and the Sum of Speciated VOC's, they have a minor effect on the ratio of the two:
• This correction shifts the average TNMOC/sum of speciated VOC’s ratio from 1.37, 1.77 and 1.1 to 1.30, 1.65 and 1.1 for UCLA 2000 summer clear, cloudy, and winter.
• A similar peak-by-peak correction for the Azusa data shifted the that average ratio from 1.29 to 1.25.
Linearity

**CO₂ Sample Volume (L)**

**VOC Concentration (ppbV)**

- Peak Area (Arbitrary Units)

**CO₂ Concentration (ppbC)**

- Peak Area (Arbitrary Units)

**Ambient CO₂**
- Acetaldehyde
- Methanol
- Acetone
- Hexane
- Dibutyl ether
- Benzene
Chamber Oxidation of m-Xylene

Oxidation products of this aromatic are not measured by standard GC/FID, but are measured well with the TNMOC channel.
# AQMD Intercomparison

<table>
<thead>
<tr>
<th>Sampling Times (PST)</th>
<th>TNMOC Instrument (ppmC)</th>
<th>SCAQMD PAMS GC</th>
<th>Total Speciated VOC’s (ppmC)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>TNMOC corrected for light hydrocarbons</td>
<td></td>
<td></td>
</tr>
<tr>
<td>10:00 - 11:00</td>
<td>1.4</td>
<td>1.55</td>
<td>1.7</td>
</tr>
<tr>
<td>11:30 - 12:30</td>
<td>1.7</td>
<td>1.7</td>
<td>1.53</td>
</tr>
<tr>
<td><strong>Average</strong></td>
<td><strong>1.55</strong></td>
<td><strong>1.7</strong></td>
<td></td>
</tr>
<tr>
<td>11:30 - 12:30*</td>
<td>0.5</td>
<td>0.7</td>
<td>0.63</td>
</tr>
<tr>
<td>13:00 - 14:00</td>
<td>0.9</td>
<td>0.7</td>
<td>0.63</td>
</tr>
<tr>
<td><strong>Average</strong></td>
<td><strong>0.7</strong></td>
<td><strong>0.75</strong></td>
<td></td>
</tr>
<tr>
<td>14:30 - 15:30*</td>
<td>1.8</td>
<td>0.75</td>
<td>3.22</td>
</tr>
<tr>
<td>16:00 - 17:00</td>
<td>5.2</td>
<td>3.8</td>
<td></td>
</tr>
<tr>
<td><strong>Average</strong></td>
<td><strong>3.5</strong></td>
<td><strong>3.8</strong></td>
<td></td>
</tr>
<tr>
<td>Period</td>
<td>Sample Volume (mL)</td>
<td>n</td>
<td>([\text{CO}_2]^*) Average ± SD (ppbC)</td>
</tr>
<tr>
<td>-----------------</td>
<td>--------------------</td>
<td>---</td>
<td>----------------------------------------</td>
</tr>
<tr>
<td>Sunny 09/12/00-10/09/00</td>
<td>1140 570</td>
<td>15</td>
<td>49±14</td>
</tr>
<tr>
<td>Cloudy 09/12/00-10/09/00</td>
<td>1140 570</td>
<td>10</td>
<td>54±8</td>
</tr>
<tr>
<td>11/30/00-12/20/00</td>
<td>1140 570</td>
<td>11</td>
<td>59±5</td>
</tr>
</tbody>
</table>
## Detection Limits and Uncertainties

<table>
<thead>
<tr>
<th>Data Type</th>
<th>Detection Limit</th>
<th>Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>TNMOC channel</td>
<td>35 ppbC</td>
<td>±10 –20 ppbC; ± 8% at concentrations over 200 ppbC, increasing below</td>
</tr>
<tr>
<td>Sum of Speciated VOC’s Channel</td>
<td>1 ppbC</td>
<td>+ 3-5%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- 10-30%</td>
</tr>
<tr>
<td>Individual VOC’s</td>
<td>10 pptC</td>
<td>± 3-20%, depending on separation</td>
</tr>
<tr>
<td>TNMOC/Sum of Speciated VOC’s Ratio</td>
<td>___</td>
<td>± 0.05 to ± 0.10 for ratios between 1 and 2, depending on concentration, - 0.1, + 0.5 for ratios above 2</td>
</tr>
</tbody>
</table>
Field Measurements

- Sources
- Ambient air
  - Azusa
  - Burbank
  - UCLA
    - Winter
    - Summer
## Diesel and Gasoline

<table>
<thead>
<tr>
<th>Source</th>
<th>TNMOC/Sum of Speciated VOC's</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gasoline Vapor</td>
<td>1.07-1.09</td>
</tr>
<tr>
<td>Diesel Fuel Vapor</td>
<td>1.39-1.44</td>
</tr>
<tr>
<td>Diesel Exhaust range</td>
<td>1.2 ± 0.2</td>
</tr>
<tr>
<td></td>
<td>1.0-1.6</td>
</tr>
</tbody>
</table>
Figure 1-7. Map showing the major freeways and highways in the South Coast Air Basin as well as the locations of the "core" monitoring sites used in many of the analyses in this report.
## Instrumentation

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Instrument</th>
<th>Sampling Period (min.)</th>
<th>Sample frequency (1/h)</th>
<th>Sample volume/flow rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>TNMOC And VOC’s</td>
<td>This work</td>
<td>10-20</td>
<td>1-2</td>
<td>500-1140 mL@50-57 mL/min</td>
</tr>
<tr>
<td>NO, NO₂, and NOₓ, O₃</td>
<td>Thermo Environmental Model 42</td>
<td>1</td>
<td>60</td>
<td>100 mL/min</td>
</tr>
<tr>
<td></td>
<td>Dasibi Model 1003-RS</td>
<td>1</td>
<td>60</td>
<td>2 L/min</td>
</tr>
<tr>
<td>Aerosols</td>
<td>Particle Measuring Systems LAS-X</td>
<td>1</td>
<td>60</td>
<td>200 mL/min</td>
</tr>
<tr>
<td>Windspeed, direction, Temperature</td>
<td>Davis Instruments Weather Wizard III</td>
<td>1</td>
<td>60</td>
<td>-</td>
</tr>
</tbody>
</table>
## Field Data Summary

<table>
<thead>
<tr>
<th>Site</th>
<th>Met.</th>
<th>TNMOC (ppbC)</th>
<th>Sum of Spec. VOC's (ppbC)</th>
<th>TNMOC/Sum of Speciated VOC's</th>
</tr>
</thead>
<tbody>
<tr>
<td>Burbank Summer</td>
<td>Clear</td>
<td>2300 ±250 740-4000</td>
<td>2070 ± 200 730-3000</td>
<td>1.11 ± 0.08 0.8-1.4</td>
</tr>
<tr>
<td>range 8/99</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>UCLA Summer</td>
<td>Clear</td>
<td>426 ± 65 150-954</td>
<td>314 ± 42 166-622</td>
<td>1.37 ± 0.12 0.8-2.2</td>
</tr>
<tr>
<td>range 8-9/99</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>UCLA Summer</td>
<td>Clear</td>
<td>377± 40 108-925</td>
<td>293 ± 31 85-713</td>
<td>1.30 ± 0.04 1.01-2.4</td>
</tr>
<tr>
<td>range 9-10/00</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>UCLA Summer</td>
<td>Cloudy</td>
<td>229 ± 29 75-700</td>
<td>145 ± 21 45-529</td>
<td>1.65 ± 0.08 1.10-3.05</td>
</tr>
<tr>
<td>range 9-10/00</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>UCLA Winter</td>
<td>Cloudy</td>
<td>317 ± 45 41-1047</td>
<td>295 ± 43 37-977</td>
<td>1.1 ± 0.03 1.0-1.96</td>
</tr>
<tr>
<td>range 11-12/00</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Azusa Summer</td>
<td>Clear</td>
<td>619 ± 37 201-1475</td>
<td>410 ± 26 242-966</td>
<td>1.26 ± 0.04 0.7-2.1</td>
</tr>
<tr>
<td>9-10/1997</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*All uncertainty ranges are $2\sigma_{\text{mean}}$*
Literature “TNMOC”

- Other measurements indicate there is a significant additional pool of VOC’s.
- Roberts et al. (1998) measured TNMOC/sum of speciated VOC’s ratios of 1.16 –1.36 in rural Nova Scotia using a related approach, in reasonable agreement with our results.
- Alastair et al. (2000) used 2-D GC to find hundreds of additional organics in the chromatogram baseline, with a T/S ratio of ~1.67 in Melbourne, Australia.
- Because their measurement used a GC column, it may have missed many compounds that we measure in our TNMOC channel.
- The cause for this discrepancy may be the selection of the speciated baseline.
- We set our baseline conservatively (low) to avoid overestimating the T/S ratio. With auto integrations, we get higher T/S ratios.
- Comparing TNMOC measurements to conventional GC data is tricky.
VOCs and T/S Ratio and Ozone at Azusa

Concentration (ppbC)

T/S ratio

[TNMOC] [Σ of Speciated]

Ozone (ppbV)

Time of Day

Azusa

09/04/97-09/29/97

00:00 04:00 08:00 12:00 16:00 20:00 24:00

1.0

1.5

2.0

2.5

3.0

0 20 40 60 80 100

Azusa

09/04/97-09/29/97

00:00 04:00 08:00 12:00 16:00 20:00 24:00

T/NMOC/Σ of Speciated VOC's

T/S ratio [Ozone]
VOCs and TNMOC/Sum of Speciated VOC’s at Burbank

Burbank
Average of [TC](ppmC) = 2.27 +/- 0.65
Average of [\(\Sigma SP\)](ppmC) = 2.06 +/- 0.51

Burbank(09/15/99-10/12/99)
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Clear</td>
<td>100</td>
<td><strong>377</strong></td>
<td><strong>293</strong></td>
<td>1.30</td>
<td>36</td>
<td>75</td>
</tr>
<tr>
<td></td>
<td>2σ$_{\text{mean}}$</td>
<td>40</td>
<td>31</td>
<td>0.04</td>
<td>σ = 22</td>
<td>σ = 16</td>
</tr>
<tr>
<td></td>
<td>range</td>
<td>108-925</td>
<td>85-713</td>
<td>1.01-2.4</td>
<td></td>
<td>45-100</td>
</tr>
<tr>
<td>Cloudy</td>
<td>90</td>
<td><strong>229</strong></td>
<td><strong>145</strong></td>
<td>1.65</td>
<td>42</td>
<td>61</td>
</tr>
<tr>
<td></td>
<td>2σ$_{\text{mean}}$</td>
<td>25</td>
<td>17</td>
<td>0.08</td>
<td>σ = 17</td>
<td>σ = 13</td>
</tr>
<tr>
<td></td>
<td>range</td>
<td>75-700</td>
<td>45-529</td>
<td>1.1-3.05</td>
<td></td>
<td>41-79</td>
</tr>
</tbody>
</table>
Atmospheric Temperature Structure
Summer

Clear

Cloudy

Early morning inversion, daytime shallow mixed layer

Deeper mixed layer, little diurnal temperature variation
UCLA Clear and Cloudy VOC Concentrations

**UCLA Clear**
09/12/00-10/08/00

**UCLA Cloudy**
09/12/00-10/08/00
TNMOC/Sum of Speciated VOC’s UCLA Clear and Cloudy

09/12/00-10/08/00

T/S ratio

[Ozone]
Atmospheric Temperature Structure
Winter

[Graph showing the atmospheric temperature structure with height and temperature axes, and data points indicating temperature at different heights for 5:00 AM and 5:00 PM.]
VOCs and T/S Ratio at UCLA-Winter

[Graph showing concentration of TNMOC and Σ of Speciated VOCs from 11/30/00 to 12/20/00 at UCLA]

[Graph showing T/S ratio and Ozone concentration from 11/30/00 to 12/20/00 at UCLA]
Correlations

TNMOC/Sum of Speciated VOC’s is not correlated with:

- $O_3$
- relative humidity
- wind speed or direction
- or for the most part time of day or day of week ("weekend effect").
- Weakly correlated with VOC concentration
High TNMOC/sum of speciated VOC’s are associated with low NO concentrations.
Relationship Between VOC Loading and T/S Ratio

![Graph showing the relationship between TNMOC and the sum of speciated VOCs for clear and cloudy conditions.]

- Clear condition: \(13 + 1.29x\), \(R^2 = 0.91\)
- Cloudy condition: \(41 + 1.35x\), \(R^2 = 0.85\)
## T/S Ratio and Ozone

<table>
<thead>
<tr>
<th>Site</th>
<th>R² for Correlation with Ozone</th>
</tr>
</thead>
<tbody>
<tr>
<td>Azusa 1997</td>
<td>N/C</td>
</tr>
<tr>
<td>UCLA Summer 1999</td>
<td>0.19</td>
</tr>
<tr>
<td>Burbank Summer 1999</td>
<td>N/C, Negative trend</td>
</tr>
<tr>
<td>UCLA Summer 2000 Clear</td>
<td>0.11</td>
</tr>
<tr>
<td>UCLA Summer 2000 Cloudy</td>
<td>N/C</td>
</tr>
<tr>
<td>UCLA Winter</td>
<td>N/C Negative trend</td>
</tr>
<tr>
<td>UCLA Summer 5:00-8:00AM</td>
<td>0.38</td>
</tr>
</tbody>
</table>

The graph shows the correlation between T/NMOC/Sum Sp. VOC’s and Ozone from 5:00-8:00 AM with an R² value of 0.38.
Photochemical Processing: Estimating Photochemical Age

- Pairs of aromatics with different lifetimes can be used as markers of photochemical processing.
- The best are aromatics that are emitted in constant ratios regardless of source.
- m,p-Xylene and ethylbenzene are typically correlated with $R^2 < 0.95$.

<table>
<thead>
<tr>
<th>Compound</th>
<th>$K_{OH}$</th>
<th>Lifetime (hrs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Benzene</td>
<td>1.23</td>
<td>75</td>
</tr>
<tr>
<td>Toluene</td>
<td>5.96</td>
<td>15</td>
</tr>
<tr>
<td>Ethylbenzene</td>
<td>7.1</td>
<td>13</td>
</tr>
<tr>
<td>m-Xylene</td>
<td>23.6</td>
<td>4</td>
</tr>
<tr>
<td>p-Xylene</td>
<td>14.3</td>
<td>6.5</td>
</tr>
<tr>
<td>o-Xylene</td>
<td>13.7</td>
<td>6.8</td>
</tr>
</tbody>
</table>
Air Mass Age

Aromatics indicate maximum average photochemical processing times of 2-4 hours.

UCLA(8/18/99-9/08/99)

ln([m,p-Xylene]/[Ethylbenzene]) = ln(3.7)
T/S Ratio and Photochemical Processing

• Assume:
  – Average speciated mixture of 100 hydrocarbons from the EPA for 29 cities at 6-9 AM survey (Jeffries 1995).
  – Rate constants for each hydrocarbon reacting with OH, and for alkenes with ozone (Atkinson, 1997).
  – 0.1 ppt OH ($2.5 \times 10^6$ molec cm$^3$) and 50 ppbv O$_3$ for 4 hours.

\[ A = A_0 \exp\left(-\left(k_{OH}[OH] + k_{O3}[O_3]\right)t\right) \]
Then:
- 30% of the hydrocarbons react once with either OH or ozone.
- The organics have an average of 7 carbons, and add ~1.5 functional groups (alcohol, carbonyl, or nitrate) per reaction.
- The total mix increases its heteroatom content relative to the carbon content by about 7%.
- The effect of this increase on the T/S ratio cannot be calculated precisely; heteroatoms either reduce the FID response or cause the compound to be lost or broadened in the column.
- The T/S ratio should have little dependence on the time of day or ozone.
UCLA Aromatic Correlations

<table>
<thead>
<tr>
<th>Compound</th>
<th>$K_{\text{OH}}$</th>
<th>Lifetime (hrs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Benzene</td>
<td>1.23</td>
<td>75</td>
</tr>
<tr>
<td>Toluene</td>
<td>5.96</td>
<td>15</td>
</tr>
<tr>
<td>Ethylbenzene</td>
<td>7.1</td>
<td>13</td>
</tr>
<tr>
<td>m-Xylene</td>
<td>23.6</td>
<td>4</td>
</tr>
<tr>
<td>p-Xylene</td>
<td>14.3</td>
<td>6.5</td>
</tr>
<tr>
<td>o-Xylene</td>
<td>13.7</td>
<td>6.8</td>
</tr>
</tbody>
</table>

Period UCLA2000

<table>
<thead>
<tr>
<th>Aromatic Ratio</th>
<th>$R^2$ w/</th>
<th>$R^2$ w/</th>
<th>$R^2$ w/</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>self O$_3$ T/S</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\ln(\text{Ethylbenzene}/\text{Benzene})$</td>
<td>0.48</td>
<td>0.13</td>
<td>0.40</td>
</tr>
<tr>
<td>$\ln(\text{Toluene}/\text{Benzene})$</td>
<td>0.46</td>
<td>0.19</td>
<td>0.36</td>
</tr>
<tr>
<td>$\ln(\text{o-Xylene}/\text{Benzene})$</td>
<td>0.48</td>
<td>0.23</td>
<td>0.34</td>
</tr>
<tr>
<td>$\ln(\text{m,p-Xylene}/\text{Ethylbenzene})$</td>
<td>0.99</td>
<td>0.49</td>
<td>0.03</td>
</tr>
</tbody>
</table>
## Contribution of Light Oxygenates: Azusa

<table>
<thead>
<tr>
<th>Oxygenate</th>
<th>Average Concentration (ppbC)</th>
<th>Percent of TNMOC</th>
<th>FID Response Factor (RF = 1 per C in NMHC’s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Acetaldehyde</td>
<td>8.4</td>
<td>1.8</td>
<td>1.65</td>
</tr>
<tr>
<td>Methanol</td>
<td>6</td>
<td>1.3</td>
<td>0.77</td>
</tr>
<tr>
<td>Ethanol</td>
<td>11</td>
<td>2.4</td>
<td>1.02</td>
</tr>
<tr>
<td>Sum</td>
<td>25</td>
<td>5.5</td>
<td></td>
</tr>
<tr>
<td>TNMOC meas. in Spec. channel (%)</td>
<td></td>
<td></td>
<td>3.7</td>
</tr>
</tbody>
</table>
Light Oxygenates at UCLA

Average propanal + acetone would result in a T/S ratio of ~1.08; observed ratio is 1.45 for clear and cloudy combined.
Conclusions

- Standard VOC measurement underestimates VOC level typically by 10-60%, total can be up to 3x higher than the sum of speciated VOC’s.
- Excess varies strongly with location, day of week and meteorology.
- At UCLA, large excess TNMOC is strongly associated with either high mixing heights (summer) or strong nighttime inversions coupled with less influence from sources.
- Reduced FID response of oxygenates does not account for much of the excess organics.
- Excess organics appear to be associated with photochemical activity and with mixing from aloft.
- Chemical identity and source of excess VOC’s is still to be determined.
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