Sources and Atmospheric Processing of Fine Particles from Asia and the Northwestern United States Measured During INTEX-B

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Outline/Methods

• INTEX-B C-130 PM$_1$ Results
  – Asian vs North American air masses
    • $SO_4^{2-}$ and WSOC average concentrations
    • WSOC/$SO_4^{2-}$ vertical profiles
  – Source for Free Trop. Organic Aerosol
    • INTEX-B Eg; WSOC in region of cloud detrainment
    • WSOC vs Biogenic/Anthrop. VOCs, multiple regression results

• Methods: PILS; PM$_1$ particles captured in water
  – Soluble anions and cations by IC ($SO_4^{2-}$)
  – Water-soluble Organic Carbon (WSOC) by TOC analysis
INTEX-B: NSF C-130 Flights

April 21 to May 15, 2006
Altitude Range: ~ 0.1 to 7.3 km asl.
10 Flights out of Everett WA
Asia vs N. American Influence Using Flexpart CO

E.g., Predominant North American Influence

E.g., Predominant Asian Influence

Flexpart CO: Emissions Only

- Asian CO: >75% Asian
  48%, n = 2931
- N. American CO: >75% NA.
  12%, n = 704
- Remainder: 40% not clear which dominated
  Asian and N.A. Flexpart CO>100ppbv ~ 95%
INTEX-B vs ICARTT

<table>
<thead>
<tr>
<th>Median Concentrations</th>
<th>All</th>
<th>Asia</th>
<th>NA</th>
<th>Median Concentrations</th>
<th>All</th>
</tr>
</thead>
<tbody>
<tr>
<td>WSOC (\mu gC/m^3)</td>
<td>0.3</td>
<td>0.2</td>
<td>0.9</td>
<td>WSOC (\mu gC/m^3)</td>
<td>1.6</td>
</tr>
<tr>
<td>SO4(=) (\mu g/m^3)</td>
<td>0.6</td>
<td>0.4</td>
<td>0.8</td>
<td>SO4(=) (\mu g/m^3)</td>
<td>4.0</td>
</tr>
</tbody>
</table>
In many regions, free trop. particulate OA is greater than SO$_4^{2-}$ mass.

Comparison of Fine OA/\(SO_4^=\) Alt. Profiles

In many regions, free trop. particulate OA is greater than \(SO_4^=\) mass.

In Asian air masses, recorded from C-130 during INTEX-B, free trop. depleted in OA wrt \(SO_4^=\).
Why is INTEX-B Asian Altitude Profile Different

**OA & SO₄²⁻ Lost in Precip. near Asia**

- SO₄²⁻ reformed in route, some SO₂ penetrates precip.
- OA not reformed
  - SOA precursors scavenged and/or depleted prior to precip., and no sig. source over ocean.

**Precipitation Scavenging**

**Secondary Production**

Observation by C130 research aircraft characterized by high sulfate relative to WSOC.

NORTH AMERICA

ASIA

PACIFIC OCEAN

~1 day

~6-8 days
**What is Source for FT Organic Aerosol?**

**INTEX-B:**
Northwestern U.S. free trop.
OA enhanced wrt $\text{SO}_4^{2-}$.

Region of high organic aerosol concentrations measured in FT over Northern Nevada (alt ~ 4 km)

![Graph showing INTEX-B and ICARTT/NEAQS regions with median WSOC values](image)

* From Heald et al. 2005
What is Source for N.A. FT Organic Aerosol?

- Possible California Central Valley & coastal urban areas
- WSOC somewhat correlated with CO
- $\Delta$WSOC/$\Delta$CO higher than other studies

Average urban Eastern U.S. clear sky SOA production $\sim 34 \, \mu\text{gC/m}^3/\text{ppmv}$ (de Gouw et al. 2007)
Enhanced Free Trop. WSOC Is In Region of Cloud Outflow

- Aircraft video shows cloudy region
- Back trajectories thru clouds
- WSOC - H$_2$O$_v$ correlated

**Graph:**
- Slope: $0.33 \pm 0.03$
- Int: $0.19 \pm 0.10$
- $r^2 = 0.80$

**MODIS (Aqua) AOD/COT observed between 21:25Z and 21:35Z**

Satellite images provided by Chieko Kittaka

- **Aircraft video** shows cloudy region
- **Back trajectories** thru clouds
- **WSOC - H$_2$O$_v$** correlated
WSOC Biogenic/Anthropogenic by Multivariate Analysis

\[ WSOC_{pred} = B_0 + B_1 x_1 + B_2 x_2 + \sum B_i x_i \]

Independent Variables \((x_i)\) as Source Tracers

<table>
<thead>
<tr>
<th>Fossil Fuel VOCs</th>
<th>Biogenic VOCs</th>
</tr>
</thead>
<tbody>
<tr>
<td>1,1,1-trichloroethane</td>
<td>Methyl chloride</td>
</tr>
<tr>
<td>Methylethylketone</td>
<td>Methanol</td>
</tr>
<tr>
<td>Isopentane</td>
<td>Hydroxyacetone</td>
</tr>
<tr>
<td>Pentane</td>
<td>Acetonitrile</td>
</tr>
<tr>
<td>Butane</td>
<td>Isoprene</td>
</tr>
<tr>
<td>Acetaldehyde</td>
<td>Acetone</td>
</tr>
<tr>
<td>Isobutane</td>
<td>Acetic Acid</td>
</tr>
<tr>
<td>Toluene</td>
<td></td>
</tr>
<tr>
<td>Benzene</td>
<td></td>
</tr>
<tr>
<td>Methyl tertiary butyl ether</td>
<td></td>
</tr>
<tr>
<td>Isopropyl nitrate</td>
<td></td>
</tr>
<tr>
<td>o-xylene</td>
<td></td>
</tr>
<tr>
<td>n-pentane</td>
<td></td>
</tr>
</tbody>
</table>

[VOC]'s normalized.
Allows direct comparisons between \(B_i\).

Biogenic vs Anthropogenic (FF) influence by sum of \(|B_a|\) versus \(|B_b|\)
WSOC Biogenic/Anthropogenic WSOC by Multivariate Analysis For INTEX-B Data

Percent observed WSOC variability explained by model

- Asian: WSOC mostly associated with anthro. VOCs, some BB
- N. American 50/50
- Cloud processed 2/3 biogenic VOCs
Summary: INTEX-B C-130

• In Asian air masses reaching N. America:
  – PM$_1$ SO$_4$$^-$ and OA concentrations are low < 1 µg/m$^3$
  – Role of precip. scavenging during transport results in?
    • PM$_1$ Free Trop. OA < SO$_4$$^-$
    • Unlike SO$_4$$^-$, anthrop. OA not regenerated far from source.

• North American (continental) air masses:
  – Free Trop. OA > SO$_4$$^-$
  – Evidence for cloud processing as a source for PM$_1$ FT OA, and mainly associated with biogenic VOCs

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