Predicting and Identifying Concentrations and Sources of Ultrafine Particulate Matter in California for Health Effect Studies

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• Collaborators:
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  – U.S. Environmental Protection Agency
  – California Air Resources Board
  – UC Atmospheric Aerosol and Health Grant
Evolution of Ambient Particulate Matter Standards

- TSP = total suspended particles
- NAAQS: National Ambient Air Quality Standards
- CAAQS: China Ambient Air Quality Standards

Timeline:
- 1971: TSP NAAQS
- 1987: PM$_{10}$ NAAQS
- 1997: PM$_{2.5}$ NAAQS
- 2006: Lower 24h PM$_{2.5}$ NAAQS
- 2012: Lower Annual PM$_{2.5}$ NAAQS

Standards:
- 1982: GB3095-82 TSP CAAQS
- 1996: GB3095-1996 PM$_{10}$ CAAQS
- 2012: GB3095-2012 PM$_{2.5}$ CAAQS
PM$_{2.5}$ Standards Based on Evidence from Epi. Studies
- consistent associations between outdoor PM concentrations and adverse health effects

An increase of 10 μg/m$^3$ in the PM$_{2.5}$ concentration was associated with a reduction in life expectancy of 0.46±0.22 (p=0.039) in 1979-1983, 0.37±0.20 (p=0.091) in 1997-2001, and 0.35±0.16 (p=0.033) in 2000-2007
The Injury Mechanisms of PM Remains Unknown

- **Health Effects Associated with PM Exposure**
  - Autonomic nervous system
  - Development: Low birth weight/preterm birth
  - Increase in asthma and other respiratory disease in children
  - Decrease in lung development and function in children
  - Cardiovascular disease including atherosclerosis in adults
  - Cancer

- **All airborne PM is toxic to some degree; potency is based on physical and chemical characteristics**
  - What PM sizes
  - What PM compositions
  - What PM sources

  are responsible for observed health effects?
Ultrafine Particles Have an Important Role in Toxicity

Oberdorster et al., EHP, 2005
Ultrafine Particles Have an Important Role in Toxicity

Redox Activity

When activity is expressed per mass, the results reflect the potency of the sample

Why No Ultrafine/PM0.1 Standards

• Because no consistent evidence from epidemiological studies
  – Due to limited scientific information about PM0.1 characteristics
    • PM0.1 number? Surface area? Mass?
    • PM0.1 chemical composition?
    • PM0.1 sources?
Ultrafines and Surface Area

• Ultrafine particles have the high surface area-to-volume ratio that can provide numerous sites for heterogeneous reactions. (Seaton and Macnee, 1995)

• Particle surface area may be most appropriate parameter to evaluate inflammatory potential and predict adverse effects of UFP (Stoeger, 2006)

• Most epidemiological studies have used particle number concentration as a surrogate for particle surface area
Challenges for Ultrafine Modeling

1. Sharp spatial gradient in particle number concentrations

2. Incomplete theory about particle nucleation, coagulation

3. Limited source emission profiles about UFP

• California is an ideal place for developing UFP modeling studies
  – Long study history of air quality modeling
  – The richest ambient UFP measurement dataset available for model validation
  – The most accurate emissions inventories
  – The most health effect study groups of any state in the United States
UFP Mass vs. UFP numbers

• Recent study (Marheit et al. 2006) indicated that PM$_{0.1}$ mass is closely aligned with particle surface area

UFP Modeling Study for California

• UCD_Primary air quality model system for modeling UFP mass concentrations, chemical compositions, and sources
  – Track ~900 primary sources
  – No nucleation, no gas-particle conversion
  – 4 km resolution
  – 7 years, 2000-2006

• WRF for meteorology fields
• In-house tools for emission processing
  – ~300 UFP source profiles (size, composition)
Air Quality Models

Figure courtesy of Prakash Bhave, US EPA.
UCD_P Air Quality Model

Observation Data

Model Gridded Data

Terrestrial Data

Meteorological Modeling by WRF

Emissions Inventory

Biogenic Emissions

Wildfire Emissions

Source Profiles from Emission Tests

Emissions Modeling UCD

UCD_P Air Quality Model

\[
\frac{\partial C_i}{\partial t} + \nabla \cdot uC_i = \nabla K \nabla C_i + E_i - S_i.
\]

Particulate Matter (EC, POA, K, Fe, Cu, Mn, etc.)

Particulate Size Distributions (PM$_{0.1}$, PM$_{2.5}$, PM$_{10}$)

Source Apportionment Information
Our Cluster
PM$_{2.5}$ EC

Model works better with longer averaging time (>1 month)

Model Predicts well for Some Chemical Components, but not All

<table>
<thead>
<tr>
<th>Species</th>
<th>Sacramento</th>
<th>San Jose1</th>
<th>San Jose2</th>
<th>Fresno</th>
<th>Bakersfield</th>
<th>Los Angeles</th>
<th>Riverside</th>
<th>El Cajon</th>
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<td>0.00</td>
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<td>-1.17</td>
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<tr>
<td>MG</td>
<td>-1.32</td>
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<td>-1.51</td>
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<td>-1.61</td>
<td>-1.65</td>
<td>-1.64</td>
<td>-1.88</td>
</tr>
</tbody>
</table>

- Mean Fractional Bias in numbers
- correlation coefficients in colors

Overall Model Has Good Performance on $\text{PM}_{0.1}$ mass and $\text{PM}_{0.1}$ EC, but Not Perfect

Sources of Primary PM\textsubscript{2.5} and PM\textsubscript{0.1}

**Los Angeles Primary PM\textsubscript{2.5} Mass**

- Non-constrained PM\textsubscript{2.5} sources
- Construction and mining gasoline
- Ships in-transit foreign motor
- Onroad cat. gasoline cold exhaust
- Offroad industrial diesel equipment
- Road construction and demolition
- Industrial natural gas combustion
- Construction and mining diesel
- Other constrained PM\textsubscript{2.5} sources
- Offroad trans refrigeration diesel
- Onroad diesel idle
- Paved road dust local streets
- Commercial bldg construction & demolition
- Paved road dust freeways
- Onroad cat. gasoline hot exhaust
- Onroad diesel exhaust

**Los Angeles Primary PM\textsubscript{0.1} mass**

- Non-constrained PM\textsubscript{0.1} sources
- Trains Metrolink
- Recreational gasoline boats 2-st
- Ships berthing foreign motor
- Ships in-transit foreign motor
- Lawn and garden gasoline 4-st
- Lawn and garden gasoline 2-st
- Residential wood fireplaces
- Other constrained PM\textsubscript{0.1} sources
- Ships maneuvering foreign motor
- Commercial LPG combustion
- Industrial gasoline equipment 4-st
- Light commercial gasoline equipment
- Construction and mining gasoline
- Onroad cat. gasoline cold exhaust
- Residential wood stoves
- Onroad diesel idle
- Onroad non-cat. gasoline hot exhaust
- Residential wood fireplaces
- Cat. gasoline buses
- Non-cat. gasoline cold exhaust
- Commercial cooking unspecified
- Offroad trans refrigeration diesel
- Onroad diesel exhaust

Regional Source Contributions of PM0.1

Air Pollution Exposure

• Strong spatial heterogeneity in concentrations and population

• Use one or a few monitor sites to represent an entire county/air basin could lead to exposure misclassification

• Population weighted concentrations (PWC)

\[ PWC = \frac{\sum_i C_i P_i}{\sum_i P_i} \]
Are We Under-Estimating the Health Effects of Particulate Matter?

Source: Manuscript in preparation.
Significant Difference Exists for PWC/CMC Ratios among Sources

### Hazard Rates for UFP Mass and Constituents Associated with Ischemic Heart Disease

Table 5. Hazard ratios (HR) and 95% confidence interval (CI) for association of UF with Ischemic Heart Disease Mortality.

<table>
<thead>
<tr>
<th>Pollutant</th>
<th>HR</th>
<th>Low CI</th>
<th>Upper CI</th>
<th>p-value</th>
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<td>Mass</td>
<td>1.10</td>
<td>1.02</td>
<td>1.18</td>
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<tr>
<td>Cu</td>
<td>1.06</td>
<td>1.03</td>
<td>1.09</td>
<td>&lt; 0.0001</td>
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<td>Fe</td>
<td>1.03</td>
<td>1.00</td>
<td>1.06</td>
<td>&lt; 0.05</td>
</tr>
<tr>
<td>Mn</td>
<td>1.00</td>
<td>0.99</td>
<td>1.01</td>
<td>0.62</td>
</tr>
<tr>
<td>Nitrate</td>
<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td>EC</td>
<td>1.15</td>
<td>1.06</td>
<td>1.26</td>
<td>&lt; 0.001</td>
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<tr>
<td>OC</td>
<td>1.08</td>
<td>1.01</td>
<td>1.15</td>
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<tr>
<td>Other Compounds</td>
<td>1.10</td>
<td>1.04</td>
<td>1.16</td>
<td>&lt; 0.001</td>
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<tr>
<td>Other Metals#</td>
<td>1.13</td>
<td>1.05</td>
<td>1.21</td>
<td>&lt; 0.01</td>
</tr>
<tr>
<td>SOA biogenic</td>
<td>1.10</td>
<td>1.02</td>
<td>1.19</td>
<td>&lt; 0.01</td>
</tr>
<tr>
<td>SOA anthropogenic</td>
<td>1.25</td>
<td>1.13</td>
<td>1.39</td>
<td>&lt; 0.001</td>
</tr>
<tr>
<td>S1: On-road gasoline</td>
<td>1.12</td>
<td>1.04</td>
<td>1.22</td>
<td>&lt; 0.01</td>
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<tr>
<td>S2: Off-road gasoline</td>
<td>1.14</td>
<td>1.04</td>
<td>1.24</td>
<td>&lt; 0.01</td>
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<tr>
<td>S3: On-road diesel</td>
<td>1.13</td>
<td>1.03</td>
<td>1.24</td>
<td>&lt; 0.01</td>
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<td>S4: Off-road diesel</td>
<td>1.14</td>
<td>1.05</td>
<td>1.23</td>
<td>&lt; 0.01</td>
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<tr>
<td>S5: Wood smoke</td>
<td>0.95</td>
<td>0.89</td>
<td>1.02</td>
<td>0.20</td>
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<tr>
<td>S6: Meat cooking</td>
<td>1.11</td>
<td>1.03</td>
<td>1.20</td>
<td>&lt; 0.01</td>
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<td>S7: High sulfur fuel combustion</td>
<td>1.08</td>
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<td>S8: Other anthropogenic</td>
<td>1.06</td>
<td>1.01</td>
<td>1.10</td>
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</table>

# Besides Cu, Fe, and Mn; S1-S8 indicate sources of primary particles

Source: Environ. Health Persp, 2015
Low Birth Weight (LBW, <2.5 kg) Is Associated With Air Pollution

• Odds Ratio of LBW per IQR associated with primary PM\textsubscript{2.5}
  – Mass = 1.025 (1.017, 1.033)
  – Gasoline = 1.027 (1.018, 1.036)
  – Wood Burning = 1.020 (1.009, 1.031)
  – Com. meat cooking = 1.019 (1.013, 1.024)

• Odds Ratio of LBW per IQR associated with primary PM\textsubscript{0.1}
  – Mass = 1.026 (1.018, 1.034)
  – Gasoline = 1.028 (1.019, 1.037)
  – Wood burning = 1.024 (1.013, 1.035)
  – Com. meat cooking = 1.019 (1.013, 1.024)

Source: Environmental Research, 2014
Summary

• 7-year UFP mass and sources modeling results for health effect studies in California

• Model generally captured concentrations of FP and UFP of certain compositions, but not all

• Model works better with longer averaging time

• Significant spatial heterogeneity of UFP among sources

• Large bias is expected if not considering the spatial heterogeneity

• UFP is found be associated with ischemic heart disease and low birth weight risks.
On-going and Future Studies

• Direct surface area modeling

• 1 km or less air quality modeling

• Full chemistry modeling
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- **2012**: Lower Annual PM$_{2.5}$ NAAQS
- **????**: PM$_{0.1}$ NAAQS?

- **1982**: GB3095-82 TSP CAAQS
- **1996**: GB3095-1996 PM$_{10}$ CAAQS
- **2012**: GB3095-2012 PM$_{2.5}$ CAAQS
- **GB3095-???? PM$_{0.1}$ CAAQS?**
Thank You!