Contribution of Nucleation Events to Ultrafine Particle Exposures

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Atmospheric Aerosol Size Distribution

ULTRAFINE PARTICLES
< 100 nm

Number
(dN/dlogDp), cm^-3 \times 10^3

Volume
(dV/dlogDp), \mu m^3/cm^3

Diameter (micrometers)

0.01 0.1 1 10

Nucleation Mode
Aitken Mode
Accumulation Mode
Condensation Submode
Droplet Submode
Coarse Mode
Ultrafine versus PM$_{2.5}$

Graph showing the relationship between ultrafine particles and PM$_{2.5}$ concentrations.

Stanier et al. AST 2004; Cabada et al. AE 2004
Sources of ultrafine particles

**Primary**
- Gasoline
- On-road Diesel
- Non-road Diesel
- Biomass Combustion
- Industrial Emissions
- Cooking

**Secondary -- Nucleation**
- Binary (H$_2$SO$_4$ + Water)
- Ternary (H$_2$SO$_4$ + NH$_3$ + Water)
- Ternary (H$_2$SO$_4$ + Org + Water)
- Ion Induced
Typical Urban PM Size Distribution

Schenley Park Distribution vs. Time 10-Aug-2001

Particle Number

log \( \frac{dN}{d \log D_p} \)
Ultrafine PM from Traffic

Urban background site in Pittsburgh
Nucleation and Growth

Schenley Park Distribution vs. Time 11-Aug-2001

Particle Number

\[ \log \frac{dN}{d \log D_p} \]
Regional Nucleation

Pittsburgh
Regional Nucleation: Pittsburgh & Philadelphia (250 mi apart)
Nucleation happens a lot
Westervelt et al. ACP 2013; Ma and Birmili Sci Tot Env 2015
Drivers – Photochemistry and lower background

Condensational Sink (cm$^{-2}$)

Fall
(9/01-11/01)

UV x SO$_2$

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Particle Growth Rate

Particle Size (nm)

00:00 06:00 12:00 18:00 24:00

10 nm/hr
Particle Growth Rate

Mass Fraction (10-60 nm Particles) Zhang et al. EST 2004
Ultrafine as core for accumulation mode

- Small, source-specific core of primary PM (<100 nm; ~10% of mass)
- Thick coating of secondary PM that condensed over several days (200-300 nm; ~90% mass)
Survival Probability

- Competition between
  - Condensational growth (sulfuric acid / organics)
  - Coagulational scavenging
- Small nuclei suffer compared to primary particles
  - Takes longer to grow
  - More diffusive $\rightarrow$ higher collision probability
Survival and Growth

- Survival: 2% (1 nm to 100 nm); 45% (30 nm to 100 nm)
- Size resolution matters in modeling CCN formation
Ultrafine Exposure in Leibnitz

5-100 nm

Fraction of Particle Number

Regional  Urban Bck  Roadside

26.0  14.0  51.9

Fresh Traffic

74.0  38.5  6.74

Nucleation

47.5  16.5  22.8

Aged traffic / Urban Sources

Regional Background

Ma and Birmili Sci Tot Env 2015
Ultrafine Exposure in Leibnitz

Total Number

5 - 20 nm

Fraction of Particle Number

Regional
Urban Bck
Roadside

Regional
Urban Bck
Roadside

Fresh Traffic

Nucleation

Aged traffic / Urban Sources

Regional Background
More nucleation in summer

Urban background site

Nucleation
Aged traffic / Urban Sources
Regional Background
Conclusions

• Nucleation important source of ultrafine at urban sites
  • ~15-30% of exposure at urban background sites
• Nucleation has distinctive temporal and seasonal patterns
• Ultrafine particles rapidly change size (condensation)
• Ultrafine particles are rapidly lost (coagulation)
• Both processes occur on timescale of ~hours
• Both processes challenge ability to apply linear statistical models (factors) to explain ultrafine concentrations, size, variability
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