Interdependence of particle number concentration and PM 2.5 in highly polluted urban atmospheres

Martin Seipenbusch,
Markus Kasper, Mingzhou Yu
The Problem:

Number size distributions for polluted and non polluted days in Beijing:

$p$ = polluted: PM10 $> 150 \mu g/m^3$

$N$ = nonpolluted

Characteristics of aerosol size distributions and new particle formation in the summer in Beijing

Shanghai 2003 (G. Kasper)
The Problem:

Measurements made with DiSCmini by Andreas Mayer in Beijing:

120,000 p/cm³ at reported PM2.5 > 300 µg/m³
200,000-500,000 p/cm³ at reported PM2.5 < 50 µg/m³

Apparent disconnect between number concentrations and PM concentrations in heavily polluted atmospheres

Shanghai 2003 (G. Kasper)
What is the origin of this disconnect?

- A aerosol chamber experiment:

Ultrafine particle source:

Temporal Evolution of Nanoparticle Aerosols in Workplace Exposure

M. SEIPENBUSCH*, A. BINDER and G. KASPER

Simulated accumulation and coarse modes

DEHS-droplets from Collison atomizer  
Silica microspheres
Coagulation of ultrafine aerosol with sudden addition of „accumulation mode“ particles
Coagulation of ultrafine aerosol with sudden addition of „accumulation mode“ particles
Number size distributions for various bimodal systems

- BG peak
- NP peak ($t=16$ min)
- DEHS

- $3 \times 10^5 \text{cm}^{-3}$
- $1 \times 10^5 \text{cm}^{-3}$
- $8 \times 10^4 \text{cm}^{-3}$

Silica

$\Delta N/\Delta \ln(x)$ #/cm³ vs. particle size $x$ /nm
Theory: heterogeneous coagulation

Smoluchowski:

\[
\frac{dN}{dt} = \beta \cdot N_i \cdot N_j
\]

\[
\beta = 4\pi(D_i + D_j)(a_i + a_j)
\]

Homogeneous coagulation:

\[
D_i = D_j \quad a_i = a_j
\]

Heterogeneous coagulation:

\[
D_i >> D_j \quad a_i << a_j
\]

Friedlander, Smoke, Dust, and Haze, 2000
More theory: an aerosol model

A model was developed to describe the interaction of a background aerosol and a continuous ultrafine particle source.

Case 1: size distribution from the polluted case presented in Yue et al..
Cases 2 and 3 are fictional but with identical number concentration as C1.
Temporal evolution of the total particle number concentration

Approx. a factor of two difference in number concentrations

Non-polluted case

Heavily polluted case
Conclusions

- Measured data gives evidence for a disconnect between mass based assessment of fine particles and number concentrations.

- Laboratory experiment and modelling results suggest binary coagulation between the ultrafine and coarser modes of the size distribution as likely explanation.

- PM 2.5 does not appear to be a monitoring metric for heavily polluted atmospheres.

- Separate limit values needed for PN and PM:
  - emissions -> Euro 5B / Euro VI -> in force
  - ambient -> ...?
More theory: an aerosol model

\[
\frac{\partial n(v, t)}{\partial t} = \frac{1}{2} \int_0^V \beta(v - v', v') n(v - v', t) n(v', t) dv' - n(v, t) \int_0^\infty \beta(v, v') n(v', t) dv' \\
\text{coagulation within SP mode}
\]

\[
- n(v, t) \int_0^\infty \gamma(v, v') p(v', t) dv' - \frac{F_{\text{out}}}{V} n(v, t) + \frac{f(v, t) F_{\text{in}}}{V}
\]

\text{SP mode attached to BP mode} \quad \text{ventilation} \quad \text{injection}

\[
\frac{\partial p(v, t)}{\partial t} = \frac{1}{2} \int_0^V \beta(v - v', v') p(v - v', t) p(v', t) dv' - p(v, t) \int_0^\infty \beta(v, v') p(v', t) dv' \\
\text{coagulation within BP mode}
\]

\[
+ \int_0^V \gamma(v - v', v') n(v - v', t) p(v', t) dv' - p(v, t) \int_0^\infty \gamma(v, v') n(v', t) dv' - \frac{F_{\text{out}}}{V} p(v, t)
\]

\text{interaction between SP and BP mode} \quad \text{ventilation}