PM versus PN

which Parameter describes the Toxic Air Contaminant emitted by CI and SI Engines better and should thus be used for Emissions Limits and AQ-Limits

Andreas C.R. Mayer
Which TOC correlates to Mortality?

6-Cities-Study
USA 1978-93
15,000 cases

Correlation with fine particles only

Source: Dockery NEJM 1993
Interpretation difficult: NOx or PNC?
Rosengarten Studie Imhoff SAE 2008-01-0336

NO2 was always measured in epidemiological studies PNC not – «maybe health effects attributed to NO2 in the past result from PNC» Neuberger, Vienna April 2014
Mortality due to PM 2.5 quantified

This does not mean that PM2.5 is the toxic substance, but only means, that PM2.5 contains a toxic substance → so we need to find the culprit to draw right conclusions
What is PM2.5 - Mass [mg/m³] of what? mix of unspecified substances – which is the toxic one? what represents the engine emission?

PM2.5 [μg/m³] identical Mass

But these 3 situations can definitely not represent same air pollution = toxicity
If we do not know which Size and Substance is the Toxic Element

- we can not identify the responsible sources
- we can not determine the countermeasures
- we can not justify to spend money
- we can not control the success

Best example is Berlin LEZ, where traffic emissions were reduced by 50 % but PM10 by < 5 %
### Which Substance in PM2.5?

Health Effect Equivalence Analysis Analysis (HEQ), a tool to answer this question. Simplified Example:

<table>
<thead>
<tr>
<th>Toxicity Parameters</th>
<th>Sulfates</th>
<th>Nitrates</th>
<th>Mineral Dust</th>
<th>Solid Nano-Particles</th>
</tr>
</thead>
<tbody>
<tr>
<td>invasive (mobility)</td>
<td>☹️ 1</td>
<td>☺ 0.01</td>
<td>😞 &lt; 0.1</td>
<td>😞 1</td>
</tr>
<tr>
<td>penetrate membranes?</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Insoluble Solids?</td>
<td>☺️ 0.01</td>
<td>😞 1</td>
<td>☹️ 1</td>
<td>☹️ 1</td>
</tr>
<tr>
<td>persistent collected and stored?</td>
<td>☺️ 0.01</td>
<td>😞 1</td>
<td>☹️ 1</td>
<td>☹️ 1</td>
</tr>
<tr>
<td>carcinogen mutagene, genotoxic?</td>
<td>☺️ 0.01</td>
<td>☺️ 0.01</td>
<td>☹️ 1</td>
<td>☹️ 1</td>
</tr>
</tbody>
</table>
The Lung is an open door for ultrafine particles
Electron microscopic analyses revealed the dominance of retained soot and a surfeit of other particle types. A variety of metal-bearing particle types were found in all compartments, but Pb, Zn, and SnZn types appeared the least biopersistent. The results support the acute toxicologic importance of ultrafine carbonaceous and metal PM. *Key words: 1952 London smog, autopsy, lung*
Particle Size Penetrating Membranes

1000 nm Polystyrene Particles

78 nm Polystyrene Particles

Laser Scanning Microscopy

B. Rothen-Rutishauser, University Berne
Cerium Oxide FBC on Soot Particles

source: Rhodia
Partikel Emission of ICE

Diesel
Russpeak: 80 nm; $10^6$
Aschepeak: 10 nm; $10^7$

Petrol
Russpeak: 40 nm; $10^5$
Aschepeak: 10 nm; $10^7$

Soot and Ash Peaks
Aerosol Number/Size – Distribution
City (Zürich) and Country (Zürcher Oberland)

Urban Area: (Downtown Zürich)
- Day (SMPS)
- Night (SMPS)
- Day (OPC)
- Night (OPC)

Rural Region: (Zürcher Oberland)
- Day (SMPS)
- Night (SMPS)
- Day (OPC)
- Night (OPC)

Bukowiecki et al., Atmospheric Environment, 2002

6 November 2001
# Health Effect for PNC and Mass PM 2.5

Short Term Cardiovascular Mortality (CVD) – Katsuyanni ETH-NPC 2012

Original Data

<table>
<thead>
<tr>
<th>Study</th>
<th>City, Year</th>
<th>CVD [%] per PN P/cm³</th>
<th>CVD - PM 2.5 per 10 μg/m³</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atkinson</td>
<td>London 2010</td>
<td>2.2/10166</td>
<td>0 - 0.5 %</td>
</tr>
<tr>
<td>Stolzel</td>
<td>Erfurt 2007</td>
<td>3.1/9748</td>
<td>0 - 1.5 %</td>
</tr>
<tr>
<td>Breitner</td>
<td>Beijing 2011</td>
<td>7.3 / 6250</td>
<td>NA</td>
</tr>
<tr>
<td>Branis</td>
<td>Prag 2010</td>
<td>1.1/1000</td>
<td>0 - 0.4</td>
</tr>
<tr>
<td>Forastiere</td>
<td>Rom 2006</td>
<td>7.6/27790</td>
<td>0.1- 3.1 %</td>
</tr>
<tr>
<td>Kettunen</td>
<td>Helsinki 2012</td>
<td>8.5/4979</td>
<td>2.1 - 23 %</td>
</tr>
<tr>
<td>Average</td>
<td></td>
<td></td>
<td>3.1 %</td>
</tr>
</tbody>
</table>
# Health Effect for PNC and Mass PM 2.5

Short Term Cardiovascular Mortality (CVD) – Katsuyanni ETH-NPC 2012

normalized to 10’000 P/cc

<table>
<thead>
<tr>
<th>Study</th>
<th>City, Year</th>
<th>CVD - PNC per 10‘000 P/cm³</th>
<th>CVD - PM 2.5 per 10 μg/m³</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atkinson</td>
<td>London 2010</td>
<td>2.2 %</td>
<td>0 - 0.5 %</td>
</tr>
<tr>
<td>Stolzel</td>
<td>Erfurt 2007</td>
<td>3.2 %</td>
<td>0 - 1.5 %</td>
</tr>
<tr>
<td>Breitner</td>
<td>Beijing 2011</td>
<td>11.7 %</td>
<td>NA</td>
</tr>
<tr>
<td>Branis</td>
<td>Prag 2010</td>
<td>11 %</td>
<td>0 - 0.4</td>
</tr>
<tr>
<td>Forastiere</td>
<td>Rom 2006</td>
<td>2.7 %</td>
<td>0.1- 3.1 %</td>
</tr>
<tr>
<td>Kettunen</td>
<td>Helsinki 2012</td>
<td>17.%</td>
<td>2.1 - 23 %</td>
</tr>
<tr>
<td><strong>Average</strong></td>
<td>****</td>
<td><strong>7.9 %</strong></td>
<td><strong>3.1 %</strong></td>
</tr>
</tbody>
</table>
Calculate Particle Mass from N and d following the Maricq-Algorithm, respecting size statistics, fractal dimension and density.

- PMP method removes nuclei particles
- Remaining solid particles follow lognormal distribution with 2 free parameters
  - Number
  - Mean diameter
- Mean diameter between ~40 – 80 nm
- To fulfill number standard of $5 \times 10^{11}$ #/km $\to$ PM mass must be $< 1$ mg/km

Mass = $N \pi / 6 \rho_0 \mu_g df \exp(df^2 (\ln \sigma_g)^2 / 2)$
# Health Effect for PNC and Mass PM 2.5

Short Term Cardiovascular Mortality (CVD) – Katsuyanni 2012 comparing mass (PNC) to mass (PM2.5)

<table>
<thead>
<tr>
<th>Study</th>
<th>City, Year</th>
<th>CVD -PNC per 10 μg/m³</th>
<th>CVD - PM 2.5 per 10 μg/m³</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atkinson</td>
<td>London 2010</td>
<td>6.8 %</td>
<td>0 - 0.5 %</td>
</tr>
<tr>
<td>Stolzel</td>
<td>Erfurt 2007</td>
<td>9.9 %</td>
<td>0 - 1.5 %</td>
</tr>
<tr>
<td>Breitner</td>
<td>Beijing 2011</td>
<td>36.5 %</td>
<td>NA</td>
</tr>
<tr>
<td>Branis</td>
<td>Prag 2010</td>
<td>34.1 %</td>
<td>0 - 0.4</td>
</tr>
<tr>
<td>Forastiere</td>
<td>Rom, 2006</td>
<td>8.4 %</td>
<td>0.1- 3.1 %</td>
</tr>
<tr>
<td>Kettunen</td>
<td>Helsinki 2012</td>
<td>52.7 %</td>
<td>2.1 - 23 %</td>
</tr>
<tr>
<td>Average</td>
<td></td>
<td>24.7 %</td>
<td>3.1 %</td>
</tr>
</tbody>
</table>

Assumption: Particles 70 nm, Density:1, mass $3.2 \times 10^{-16}$ g/P / 10’000 P/cm³ = 3.2 μg/m³
Average CH-Compositions in Winter

Reiden (February)

Payerne (January)

Massongex (December)

Roveredo (December)

Zürich (January)

Black Carbon
Organic mass
Nitrate
Sulfate
Ammonium
Chloride
What does this mean for Monetary Health Impact MHI?

• assuming MHI is 400 €/kg PM10 (Swiss Data)
• assuming exhaust soot content of PM2.5 is 15 %
• concluding soot particles are the main toxic

→ MHI of soot is 3’200 €/kg soot

→ Benefit/Cost-Ratio of an emission measure eliminating soot will be > 20
→ Health Cost and B/C-Ratio must be based on PNC
VERT 1996

Based on this physiological and toxicological findings (mostly from occupational health, see Johannesburg convention 1952) a first definition was proposed

„Solid, insoluble particles in the mobility size range of 10-500 nm“

➔ development of new instrumentation
➔ BAT-particle filters
➔ start of the ETH-NPC
Conclusion on European Level
EU CO-Decision (Art.12, Rec.15 - 2008)

• In order to achieve these environmental objectives it is appropriate to indicate that particle number limits are likely to reflect the highest level of performance with particle filters using best available technology.

• .. the commission shall introduce particle number based limit values at a level appropriate to the technologies actually being used.
BAT is Filtration downstream Engine

Filtration achieves 99.99 % on every engine as long as SV is below the limit. One VERT test is sufficient Duplication avoided

99.99 % means 0.001 mg/kWh
### Directive 1999/30/CE for AQ

These values are those being elaborated in 1997 by a WHO working group and well-known as the WHO-AQG (Air Quality Guidelines of World Health Organization).

<table>
<thead>
<tr>
<th>EU limit values for PM10 and NO2</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>averaging period</strong></td>
<td><strong>limit value</strong></td>
</tr>
<tr>
<td>24 h</td>
<td>50 μg/m³ PM$_{10}$</td>
</tr>
<tr>
<td></td>
<td>35 exceedances/year</td>
</tr>
<tr>
<td>1 year</td>
<td>40 μg/m³ PM$_{10}$</td>
</tr>
<tr>
<td>24 h</td>
<td>50 μg/m³ PM$_{10}$</td>
</tr>
<tr>
<td></td>
<td>7 exceedances/year</td>
</tr>
<tr>
<td>1 year</td>
<td>20 μg/m³ PM$_{10}$</td>
</tr>
<tr>
<td>1 h</td>
<td>200 μg/m³ NO$_2$</td>
</tr>
<tr>
<td></td>
<td>18 exceedances/year</td>
</tr>
<tr>
<td>1 year</td>
<td>40 μg/m³ NO$_2$</td>
</tr>
</tbody>
</table>

*indicative limit values, to be reviewed by the EU Commission*

**Switzerland 1998:**

- PM 10 24h: 50 μg(m3 / 1 x pa
  - 1 year 20 μg/m3
- NO2: 1h 100 μg/m3
  - 1 year 30 μg/m3
Ambient Air Limit Values
Monitoring and Control

- Ambient Air Limit Values still PM10 resp. PM2.5
- not respecting impact of size or substance

→ Cleaning car exhaust not reflected by ambient air metrics
→ Toxicity is not correctly reflected by ambient air metrics
→ Epidemiologic conclusions are misleading
→ Policies based on mass (PM10 or PM2.5) will fail
Conclusions

- It would be possible to use the results of studies such as Atkinson et al. (2010) and Stolzel et al. (2007) to set air quality standards for (traffic generated) particles by number.
Messages and Conclusions

1. PM is not sufficient to address health effects
2. PM is not sufficient to define BAT emission control
3. PM criteria are misleading for filter selection
4. PN instrumentation is available
5. PN is indispensible to link emission to air quality
6. AQ must replace or complement PM by PN
7. Metrics in Emission and AQ must be coherent