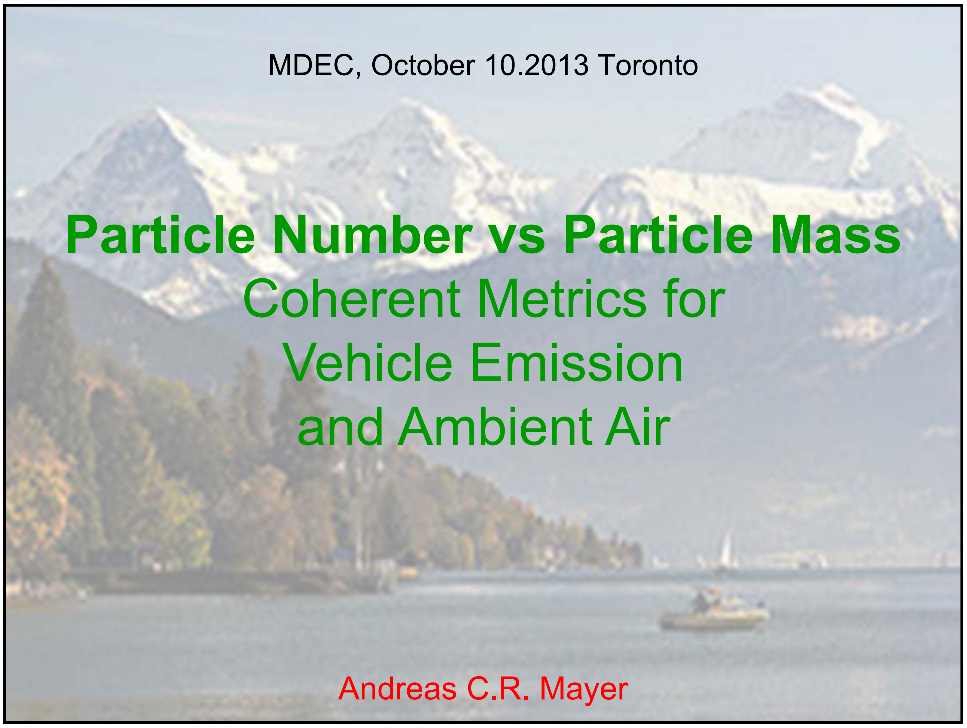


MDEC, October 10.2013 Toronto

Particle Number vs Particle Mass
Coherent Metrics for Vehicle Emission and Ambient Air

Andreas C.R. Mayer

Four Strong Reasons

why Particle Mass PM should be complemented by Solid Particle Number PN

- *address health effects in exhaust proximity,*
- *limit engine emission on BAT level*
- *characterize particle filter substrates properly*
- *correlate particle emissions to air quality*

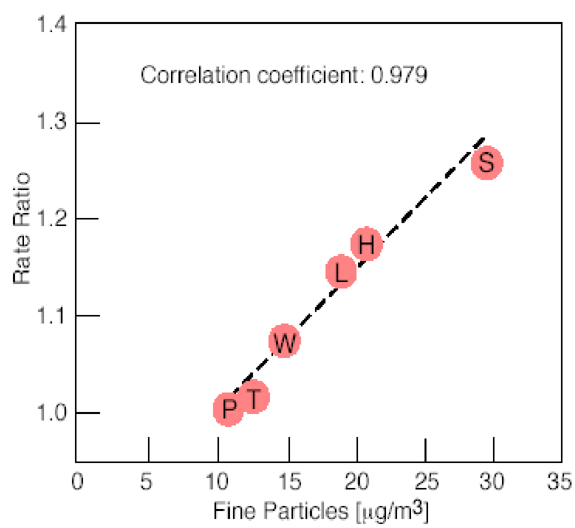
1.Address Health Effects

- why does PN better reflect health impact
- which size range is important ?
- which substances have priority ?

Long Term Effects

Mortality due to PM 2.5 quantified

6-Cities-Study
USA 1978-93
15'000 cases
correlation with
fine particles only
Dockery 1993

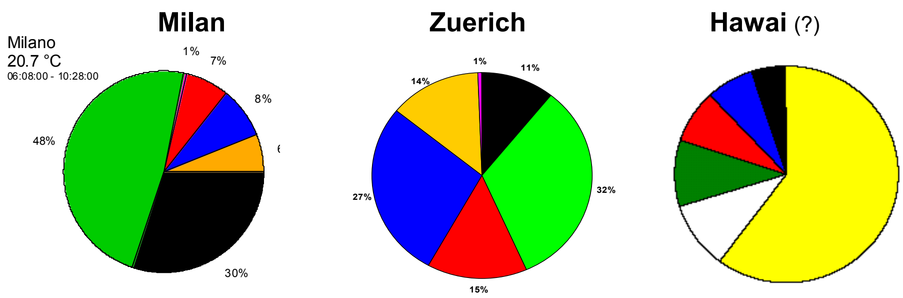


This does not mean that PM2.5 <u>is the toxic substance</u>,
but only means, that PM2.5 <u>contains a toxic substance</u>
<u>→ so we need to find the culprit</u>

What is PM2.5 - Mass [mg/m3] ?

mix of unspecified substances – which is the toxic one ?

what represents the engine emission ?



PM2.5 [µg/m^3] **identical**

But these 3 situations can definitely not respresent same air pollution = toxicity

Black Carbon
Organic mass
Nitrate
Sulfate
Ammonium
Chloride

If we do not know the Substance of the Toxic Air Contaminant (TAC)

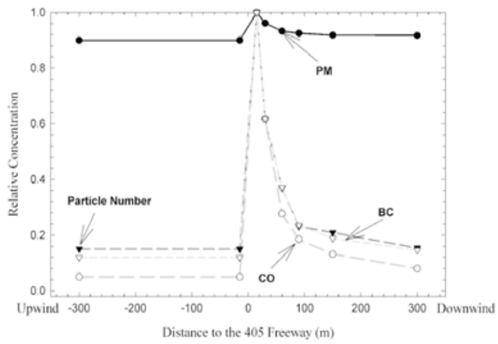
- *we can not identify the responsible sources*
- *we can not determine the countermeasures*
- *we can not justify to spend money*
- *we can not control the succes*

Best example is Berlin LEZ, where emissions were reduced by 50 % but PM10 war hardly touched

UFP-Concentration and Health Impact

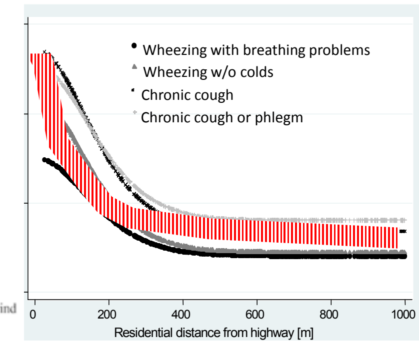
depend strongly on distance to high traffic roads

Particle Number Distribution near Roads



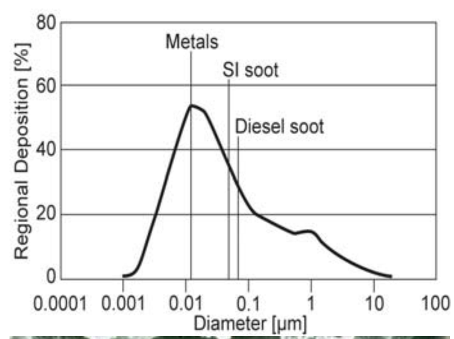
Hinds, Zhu et al
University of California, L.A.
Size distribution of UFP neas Los Angeles 405
Air & waste management Sept. 2002

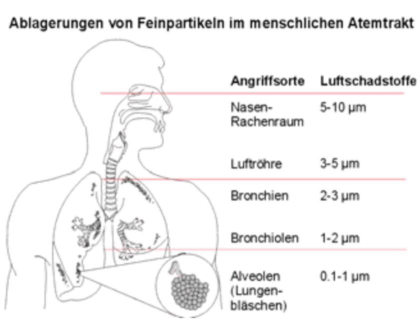
Respiratory Health Impact near Roads

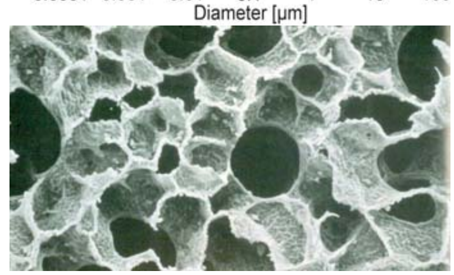


Hazenkamp, Künzli et al
Swiss Tropical and Public Health Institute, TPH
Impact of highway traffic on repiratory health in adults
Environmental Health 10/2011

Why is particle size decisive for health risk considerations



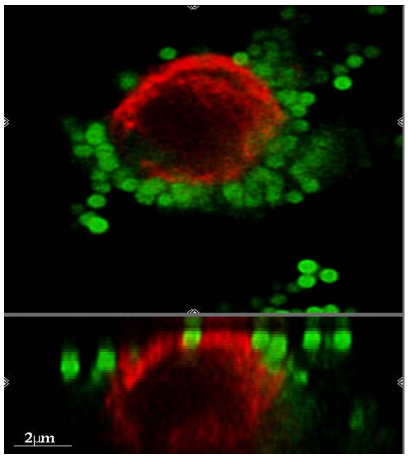




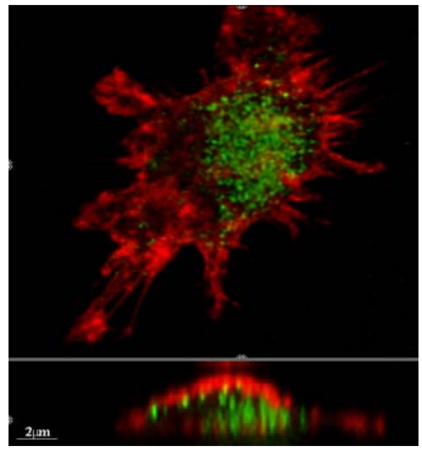
Ultrathin alveoli tissue permits penetration of gases and UFP into blood vessels

Macrophages *in vitro*

1000 nm Polystyrene Particles



78 nm Polystyrene Particles

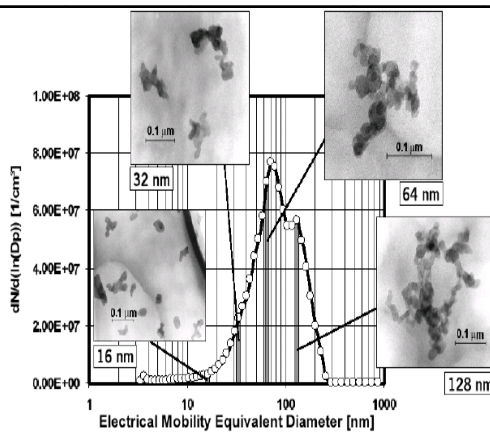


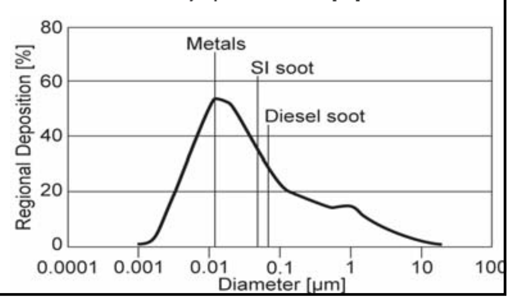
Laser Scanning Microscopy

B. Rothen-Rutishauser, University Berne

The weakest size range of the Lungs is the strongest emission range of the Engines and the weakest size range of Filters

The Lung is an open door for engine emitted particles





Based on this physiological and toxicological findings (mostly from occupational health, see Johannesburg convention 1952) a first definition was proposed in Y 1998 by VERT

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

→ leading to development of new instrumentation and the selection of BAT-particle filters
→ start of the ETH-NPC
→ research started at many places and now the first PN-epidemiological studies are available

Health Effect for PNC and Mass PM 2.5

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

Original Data

Study	City, Year	**CVD [%] per PN P/cm3**		CVD - PM 2.5 **per 10 µg/m3**
Atkinson	London 2010	2.2/10166		0 - 0.5 %
Stolzel	Erfurt 2007	3.1/9748		0 - 1.5 %
Breitner	Beijing 2011	7.3 / 6250		NA
Branis	Prag 2010	1.1/1000		0 - 0.4
Forastiere	Rom 2006	7.6/27790		0.1- 3.1 %
Kettunen	Helsinki 2012	8.5/4979		2.1 - 23 %
Average				3.1 %

Health Effect for PNC and Mass PM 2.5

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

normalized (simplified linearly) to 10'000 P/cc

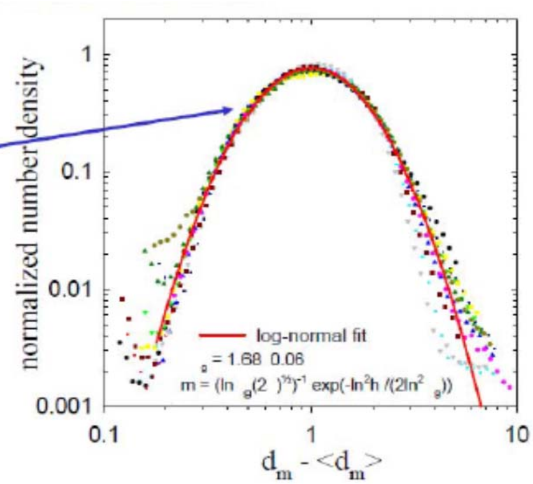
Study	City, Year		CVD - PNC per **10'000 P/cm3**	CVD - PM 2.5 **per 10 µg/m3**
Atkinson	London 2010		2.2 %	0 - 0.5 %
Stolzel	Erfurt 2007		3.2 %	0 - 1.5 %
Breitner	Beijing 2011		11.7 %	NA
Branis	Prag 2010		11 %	0 - 0.4
Forastiere	Rom 2006		2.7 %	0.1- 3.1 %
Kettunen	Helsinki 2012		17.%	2.1 - 23 %
Average			7.9 %	3.1 %

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 $5x10^{11}$ #/km → PM mass must be < 1 mg/km



$$\text{Mass} = N \, \pi/6 \, \rho_0 \, d_0^{(3-df)} \, \mu_g^{df} \exp(df^2 (\ln\sigma_g)^2/2)$$

Health Effect for PNC and Mass PM 2.5

Short Term Cardiovacular Mortality (CVD) – Katsuyanni 2012

PNC converted to mass

Study	City, Year	CVD - PNC per **10‘000 P/cm3**	CVD -PNC converted to mass -per **10 µg/m3**
Atkinson	London 2010	2.2 %	6.8 %
Stolzel	Erfurt 2007	3.2 %	9.9 %
Breitner	Beijing 2011	11.7 %	36.5 %
Branis	Prag 2010	11 %	34.1 %
Forastiere	Rom, 2006	2.7 %	8.4 %
Kettunen	Helsinki 2012	17 %	52.7 %
Average		7.9 %	24.7 %

Assumption: Particles 70 nm, Density:1, mass 3.2×10^{-16} g/P / 10‘000 P/cm3 = 3.2 µg/m3

Health Effect for PNC and Mass PM 2.5

Short Term Cardiovacular Mortality (CVD) – Katsuyanni 2012

comparing mass (PNC) to mass (PM2.5)

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

Assumption: Particles 70 nm, Density:1, mass 3.2×10^{-16} g/P / 10‘000 P/cm3 = 3.2 µg/m3

What does this mean for the Health Impact Analysis

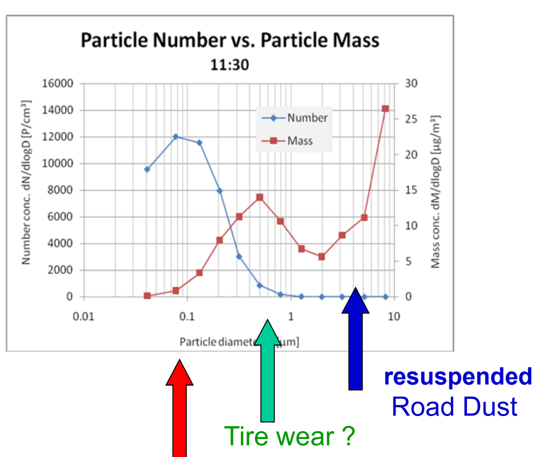
- the health impact of 1 g PM consisting of solid nanoparticles is 8 times the health impact of 1 g PM2.5 consisting of a varying mix of substances
- This 1 g PM2.5 may contain 15 % traffic particle mass (BC and metal oxides)

→ These 15 % evidently represent most of the toxicity contained in this kg PM2.5

→ Is this 15 % the BC content ?

Size Distribution at Curbside Zürich

would BC be the right parameter ?



Engine Exhaust

Tire wear ?
Break wear ?
Wall deposited particles

- Engine emitted (EC+metal oxides) particles have very little mass but high numbers
- Tire wear (BC) has low numbers but higher mass
- Resuspended material (also BC ?) has even higher mass

What does this mean for Monetary Health Impact MHI ?

- assuming MHI is 400 €/kg PM2.5 (Swiss Data)
- assuming exhaust soot content of PM2.5 is 15 %
- knowing soot particles are the main health impact

→**MHI of soot is 3’200 €/kg soot**

→ Benefit ratio of a emission measure eliminating soot will be >10 times higher than actual cost

2. Limit Engine Emission on BAT Level

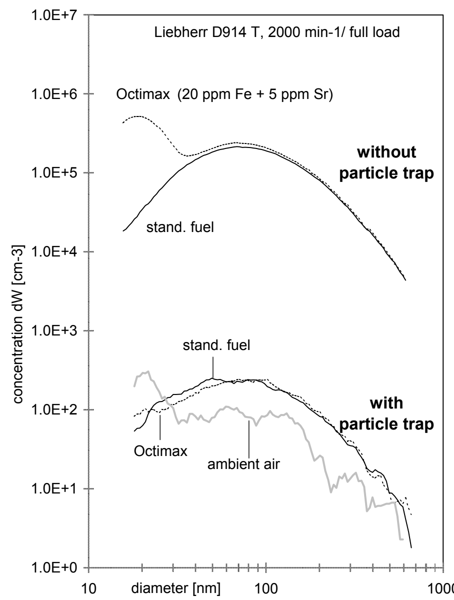
with respect to carcinogenic Particles

- *what is best available technology BAT*
- *how to measure particle emission*
- *what is Detection Limit of PM versus PN*
- *how to define the limit values*

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



Detection Limits

DT-Limit of PM-Measurement *(analog)*
acc. to PMP-research is commonly 3 mg/kWh
1 mg/kWh might be in reach with large scatter.
Size specific analysis is impossible with PM

DT-Limit of PN-Measurement is 1 Particle *(digital)*
realistically 100 P/cc = 10^9P/kWh = 0.0002 mg/kWh
PN-Measurement is dynamic and size-specific

→ PN metrology is at least 1000 x more sensitive
→ PN metrology permits to enforce lower limits
→ PN-Metrology permits real life dynamic testing and supports combustion development

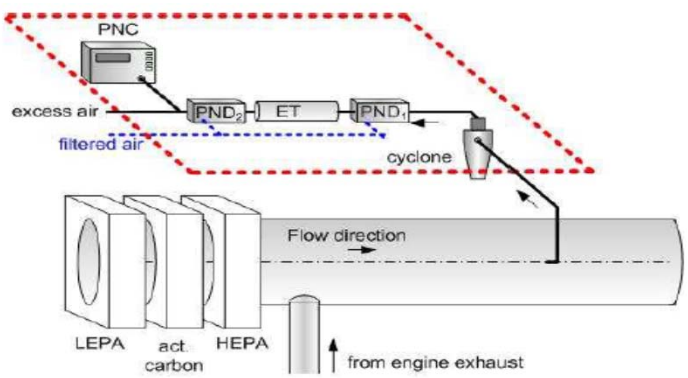
Conclusion on European Level

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.

PMP-Set-up for solid particle counting

PNC 23-2500 nm; ECE/324/ Add.48, R49 – July 2011



- Sensitivity PM: 1 mg/kWh
- Sensitivity PN : < 0.001 mg/kWh

Compare Regulation in USA and EU

Standard	PM requ.	PN eff.	PN requ	PM eff	Comment
Euro-I	700	$3x10^{14}$			No real progress
Euro-II	150	$2x10^{14}$			No real progress
Euro-III	100	$1x10^{14}$			No real progress
Euro-III DPF	-	$1x10^{10}$	-	0.02	Retrofit 99.99%
Euro-V	20	$6x10^{13}$			No real progress
EPA 2010	10	$3x10^{13}$			DPF not required PFF suficient
„Euro VI" w/o filter	10	$3x10^{13}$			Scania 2007
Euro VI (2013)	10	$3x10^{13}$	$6x10^{11}$	0.2	50x below EPA DPF required

PM [mg/kWh] CVS Particles < 2.5 µm
PN [PNC/kWh] CVS-PMP 23-2500 nm
PN converted to PM with particle diameter 70 nm

What does this mean for the comparison of EU and US HD-Emissions ?

1. Today EU and US require the same PM-Emission – seem to be equally clean

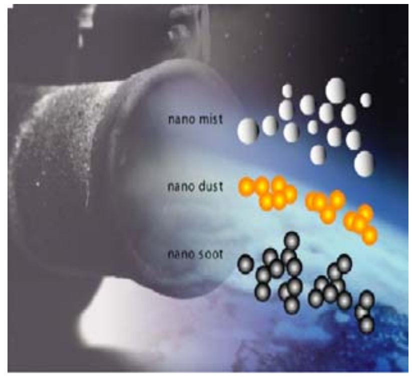
2. However, since EU requires in addition PN-limit the effective PM requirement of EU is 50 x stricter than US

3. US-Manufacturer could today reach the existing PM-limit without filters and they may do so – de-install filters.

4.In this case there effective PM emission will be 50 times higher than EU and US can hardly lower it because of PM metrology

What about metal oxide particles ?

- Gases: CO, HC, NOx, SOx
- Solid soot particles
- Metal oxide particles
- Volatile particles

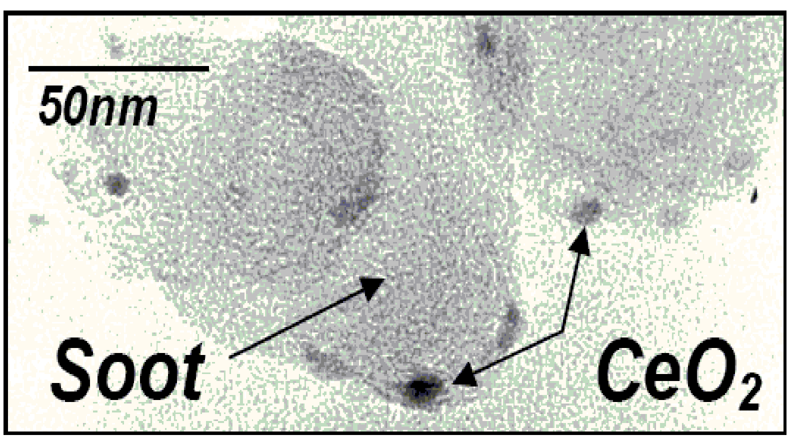


Metal oxides might be the most toxic part. They are very small and their concentration is very high in the exhaust gas of Diesel and Petrol engines

We can not measure them by mass but we can filter them

Cerium Oxide FBC on Soot Particles

source:Rhodia



Sources of Metals from ICE

Engine Wear: Fe, Ni, Cr, Al, Si
Bearing Wear: Cu, Sn
Lube Oil: Zn, Ca, P
Cat.Coatings: Pt, Pd, V, Cu, Al, Ce,
FBC: Fe, Ce, Pt, Cu

VERT-DPF-certification protocol
looks at metal emissions size-specific – part of the secondary emissions test VSET

BAT-DPF's
remove 99.9 % fo solid insoluble metal oxid particles from Diesel and Petrol engines

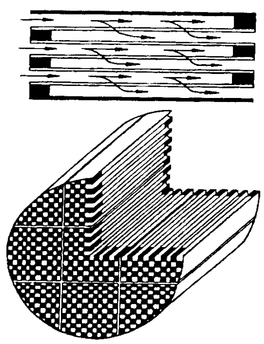
3. Particle Filter Qualtiy

- depends mainly on particle size

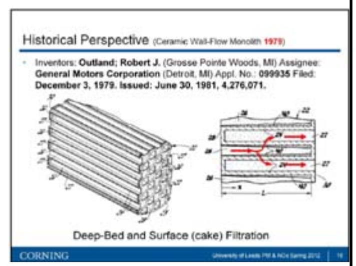
Filter für Diesel-Exhaust 1982

now over 30 Mio successful on the road

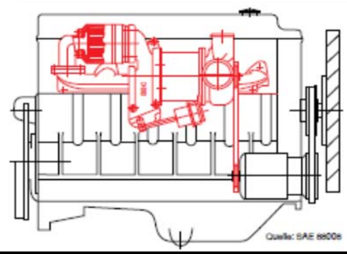
1982
Corning



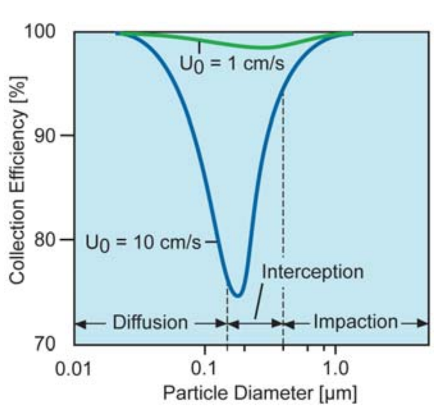
1979
GM

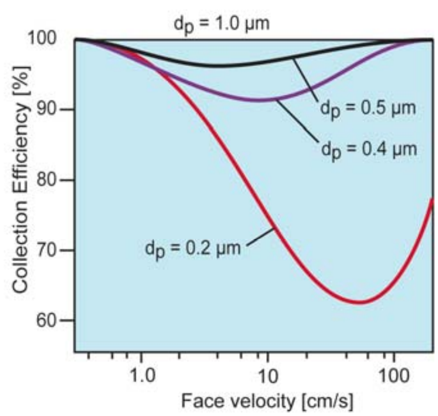


1985
BBC
DB



Many Filters are not perfect





→ Careful Filter Verification is required
→ VERT-Standard based on PN
→ SN 277206

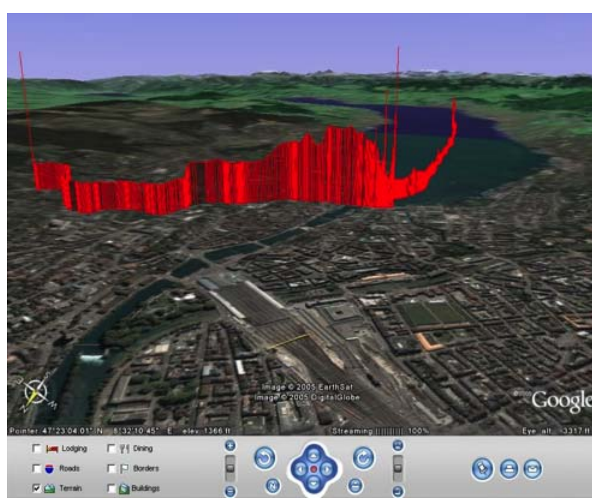
4. Ambient Air Monitoring and Control

- Should be coherent to emission
- Metrology can be identical
- Instrumentation is available

Introduce Portable Particle Counters

providing PNC+Size, Surface and Mass calculated

Swiss Ordinance Aug.2012



4

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 filter selection
4. PN instrumentation is available
   for emission control and ambient monitoring
5. PN is undispensible to link emission to air quality

But even PN may not be sufficient in the long run and should be complemented by information on substance (metals ?)

Black Carbon can not substitute PM nor PN because non-carbon toxics might play a very important role

Dose/Effect by PM2.5 or Soot ?