Lung-deposited surface area measurements in Zürich

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Over the last years, a number of epidemiological studies have shown detrimental health effects in people living close to a busy road. For example, children living closer to highways have smaller lungs, an increased risk for cardiovascular diseases, heart attacks and asthma. These increased risks cannot be explained in terms of PM10, which is usually quite uniform in a given region – the ultrafine particles from traffic hardly contribute to PM10. However, it is hard to disentangle the various pollutants emitted by traffic from each other, so it is not clear whether the ultrafine particles are responsible for these health effects. If they are, we must use a different metric than PM10 to measure them to explain why the proximity to roads is unhealthy. From a toxicological point of view, there is evidence linking particle surface area with cellular responses; for example, the higher mass-based toxicity of smaller particles has been explained with their increased surface area (Oberdörster et al, 2005), and a number of mechanisms for interaction of nanoparticles with biological tissue have been proposed which take place on the particle surface (Nel et al, 2006). Particle surface area might thus be a more appropriate metric to use than particle mass, at least for biopersistent particles. However, for health effects, only particles that actually end up in the lung will count, i.e. we should not be measuring total particle surface area in the ambient air, but rather the lung-deposited surface area (LDSA), so the size-dependent deposition in the lung must be taken into account. The obvious way of measuring the lung-deposited surface area is to (1) measure the particle size distribution with an SMPS, (2) to convert the number size distribution into a surface area size distribution (under the assumption of spherical particles) and (3) to multiply this with the size-dependent alveolar deposition fraction as calculated from the ICRP model. This approach needs an SMPS, and is therefore rather complex. By a lucky coincidence, one of the simplest aerosol instruments, the diffusion charger (DC), produces an instrument response that is very close to LDSA (Wilson et al, 2004). Diffusion chargers can thus be used to give an approximate value for LDSA, at least for compact particles. From this entire argument, we conclude that DC chargers appear to be more appropriate for measuring urban air quality than other instruments which measure particle number or particle mass per volume, because they (1) measure a more appropriate metric, and (2) the lung deposition is automatically taken into account, unlike in traditional measurements (e.g. particle number in 1/ccm, or mass in $\mu g/m^3$).

There are a number of instruments that can be used to measure LDSA; for example, TSI has two dedicated DCs in their program which are calibrated to report LDSA; and the Matter Aerosol DiSCmini can also report LDSA. Furthermore, any instrument relying on an internal diffusion charge could be calibrated appropriately (for example: Philips Aerasense NanoTracer, Grimm nanoCheck, Pegasor PPS, Dekati ELPI+, EcoChem DC). What makes those instruments attractive in general is that they are simple and can be built in compact and cost-effective packages. At our institute we are currently developing a miniaturized DC which should fit into a shirt pocket (272 cm3), which will hopefully be useful for simple and cost-effective personal exposure monitoring. Clearly, the particle composition is also highly relevant, but it is much harder to measure, and cannot be done in a shirt pocket. In this sense, we believe a miniature DC to be a good compromise for personal exposure monitoring between relevant information on one hand and ease-of-use on the other.

During the first week of September 2009, we performed a one-week measurement campaign in Zürich with 6 miniature diffusion size classifiers (Fierz et al, 2011). These instruments measure particle number concentration, average particle diameter and LDSA simultaneously. Two of the instruments were co-located with traditional instruments (CPC, SMPS). Measured data was averaged to hourly values for comparison with PM10 measurements. The table below gives an overview of the averages of the individual instruments, and the correlations between different measures.

Station	**<N> 1/cm^3**	**<LDSA> mm^2/cm^3**	**R^2 N-PM10**	**R^2 LDSA-PM10**	**R^2 N-LDSA**
Schwamendingen A1 (Highway)	16400	40	0.21	0.32	0.93
Bellevue (busy city road)	25600	63	0.34	0.44	0.95
Neumühlequai (busy city road)	31400	63	0.33	0.44	0.93
Walchestr. (inner city, little traffic)	7300	19	0.59	0.72	0.92
Stampfenbachstr. (average city road)	12600	28	0.21	0.32	0.92
Heubeeribüel (no traffic)	4500	11	0.23	0.46	0.84

The highest values of LDSA were found at sites with heavy traffic, while the lowest average value was nearly 6 times lower. This difference is very close to the highest difference in particle number concentrations, a factor 7. Checking the correlations between the different instruments, we note:

- For the two instruments co-located with SMPS and CPC, the correlation of the minidisc number concentration with traditional instrumentation was excellent (R^2~0.95).
- The correlation of the particle number concentration with lung-deposited surface area is typically also very high ($R^2 > 0.9$ except for the station with the least traffic).
- LDSA correlates better with PM10 than particle number at all sites, but both LDSA and particle number only correlate weakly with PM10.

From these measurements, we conclude that LDSA is a sensible or maybe even better alternative to traditional number and mass measurements. What makes LDSA particularly interesting is that it can easily be measured with existing instruments, and that these instruments offer further potential for miniaturization and lower cost. Furthermore, DC sensors offer long service intervals, which is important for long-term monitoring applications.

References:

Günter Oberdörster et al.: Nanotoxicology: An Emerging Discipline Evolving from Studies of Ultrafine Particles. Environ Health Perspect 113:823-839 (2005)

Andre Nel et al.: Toxic Potential of Materials at the Nanolevel. Science 311, 622-627 (2006)

Wilson W.E. *et al.*, "Use of the Electrical Aerosol Detector as an Indicator for the Total Particle Surface Area Deposited in the Lung," *Proceedings of 2004 A&WMA*, paper #37 (2004).

M. Fierz et al. "Design, Calibration and Field Performance of a Miniature Diffusion Size Classifier", Aerosol Science and Technology, 45, 1-10 (2011).

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The Mayer Question

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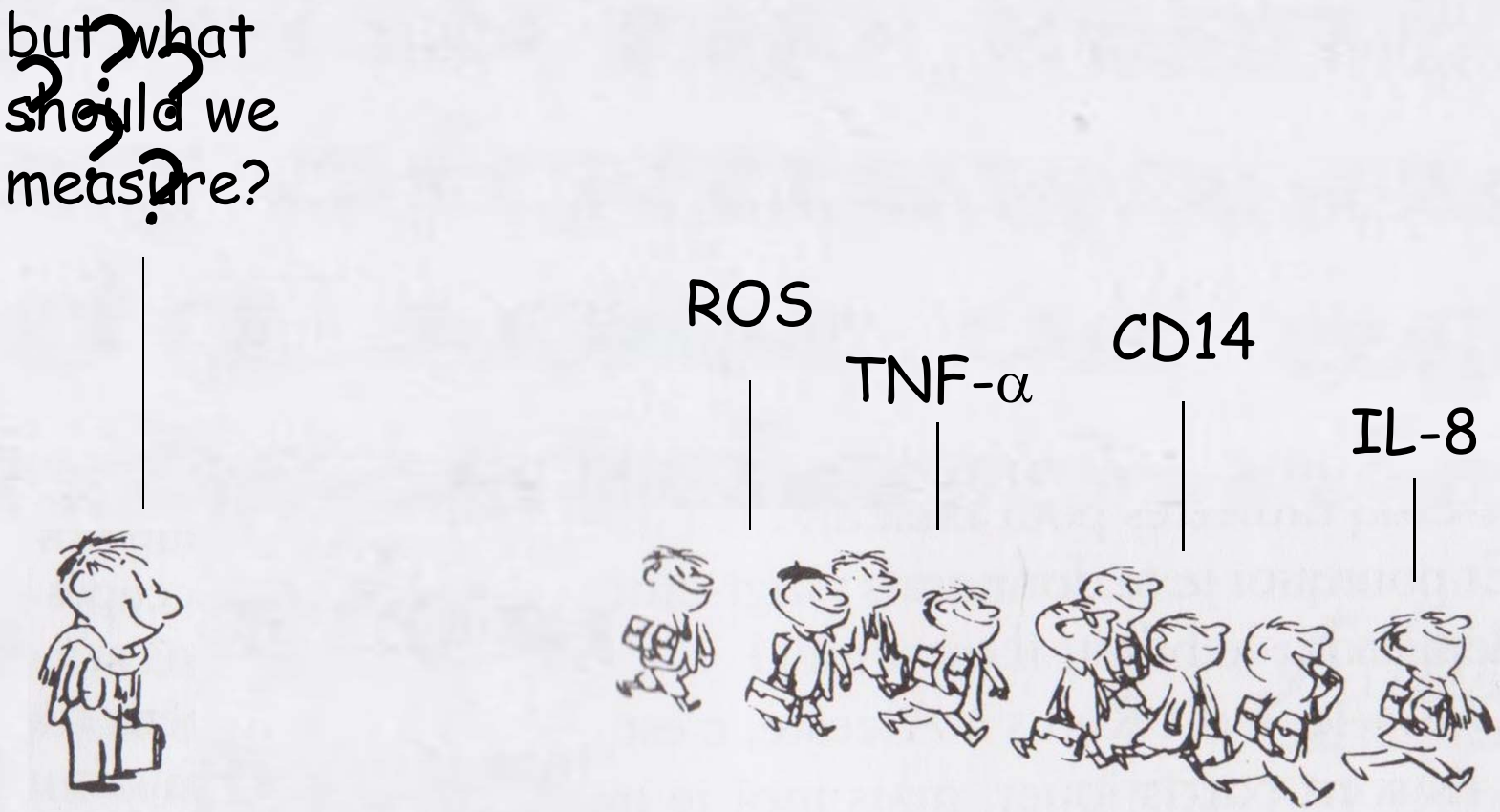


Quotes from ETH-conferences

- Children living closer to highways have smaller lungs and an increased risk for cardiovascular diseases and heart attacks. This risk is independent and additive to general background (PM10)

  -- R.Rapp, 2007

- Asthma risk is higher for children living close to a busy street

  -- N.Kuenzli, 2009

- Association between exposure to traffic and the onset of acute myocardial infarction

  -- R.Duffin, this morning

Quotes from ETH-conferences

- Current legislation, which is based on mass of PM emission might not adequately reflect health effects of environmental particles

  -- R.P.Verbeek, 2009

- Benefits from LEZ might be more far-reaching than currently shown by routine measurements (PM10)

  -- A.Peters, 2008

Quotes from ETH-conferences

- Mass sucks
  -- A.C. Pope, 2005

- We need to measure particle number concentration, not particle mass
  -- O.Brändli, yesterday

- Maybe particle surface area is a better predictor of toxic effects than particle mass
  -- F. Cassee, this morning

Surface area

Oberdörster et al.

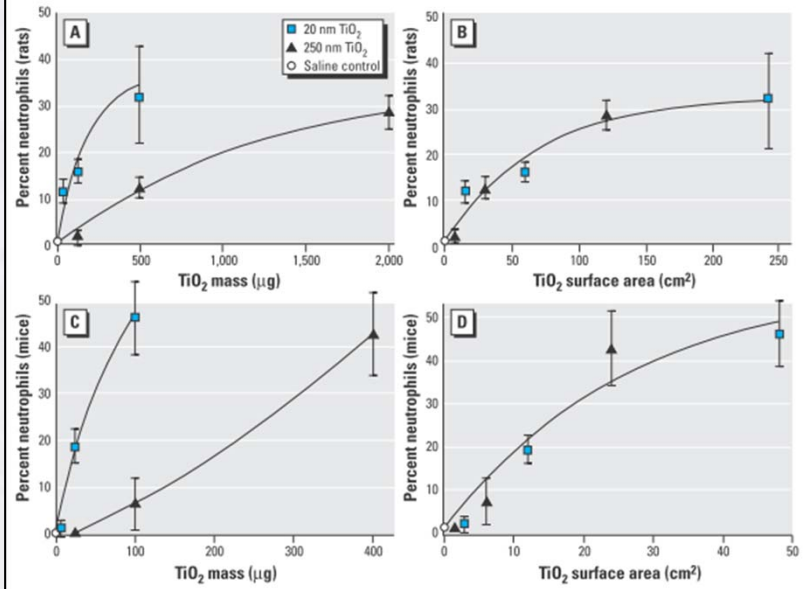


Figure 4. Percentage of neutrophils in lung lavage of rats (*A,B*) and mice (*C,D*) as indicators of inflammation 24 hr after intratracheal instillation of different mass doses of 20-nm and 250-nm TiO_2 particles in rats and mice. (*A,C*) The steeper dose response of nanosized TiO_2 is obvious when the dose is expressed as mass. (*B,D*) The same dose response relationship as in (*A,C*) but with dose expressed as particle surface area; this indicates that particle surface area seems to be a more appropriate dosemetric for comparing effects of different-sized particles, provided they are of the same chemical structure (anatase TiO_2 in this case). Data show mean ± SD.

Nel et al.

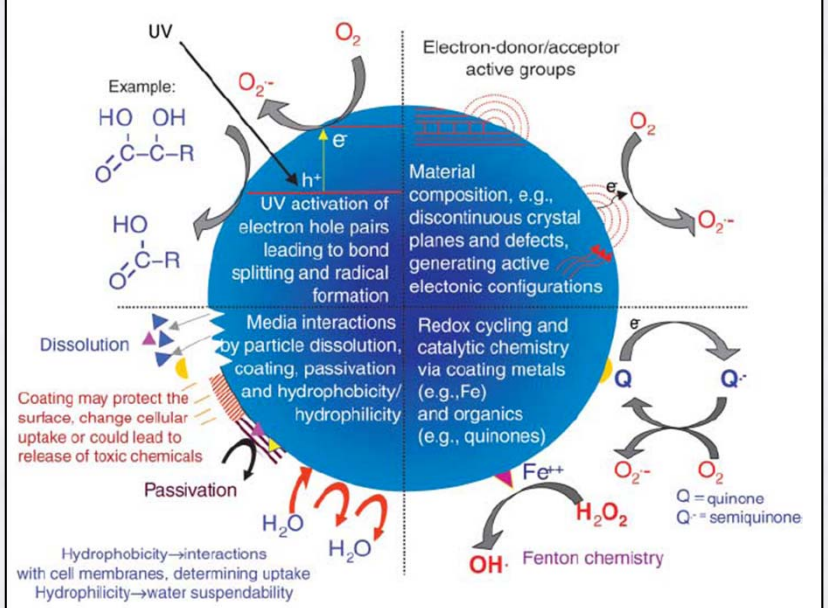


Fig. 2. Possible mechanisms by which nanomaterials interact with biological tissue. Examples illustrate the importance of material composition, electronic structure, bonded surface species (e.g., metal-containing), surface coatings (active or passive), and solubility, including the contribution of surface species and coatings and interactions with other environmental factors (e.g., UV activation).

Nanotoxicology: An Emerging Discipline Evolving from Studies of Ultrafine Particles. Günter Oberdörster et al. , Environ Health Perspect 113:823-839 (2005)

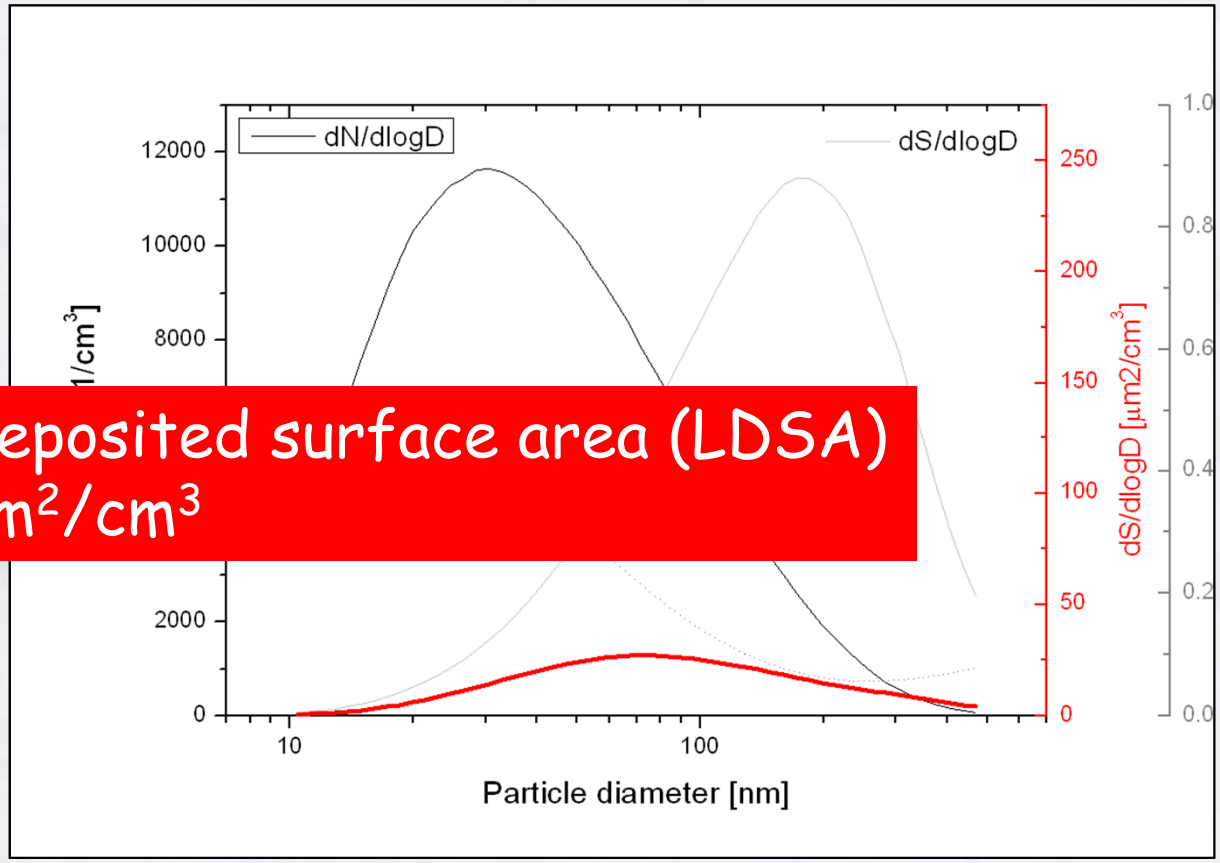
Toxic Potential of Materials at the Nanolevel. Andre Nel et al., Science 311, 622-627 (2006)

My personal conclusion

- There are traffic-related health effects that cannot be explained by PM10
- Legislators are fighting a losing battle to convince the public that LEZs are beneficial (a few % reduction in PM10 at best).
- If you want to disregard chemistry, there is evidence implying that particle surface area is the most sensible physical metric
- I know of no such evidence for particle number (PMP!)

Surface area in Zürich

- Average SMPS data for 2008
  (Thx to C.Hüglin, H.Herich, Swiss air pollution monitoring network)



Lung-deposited surface area (LDSA)
unit: $\mu m^2/cm^3$

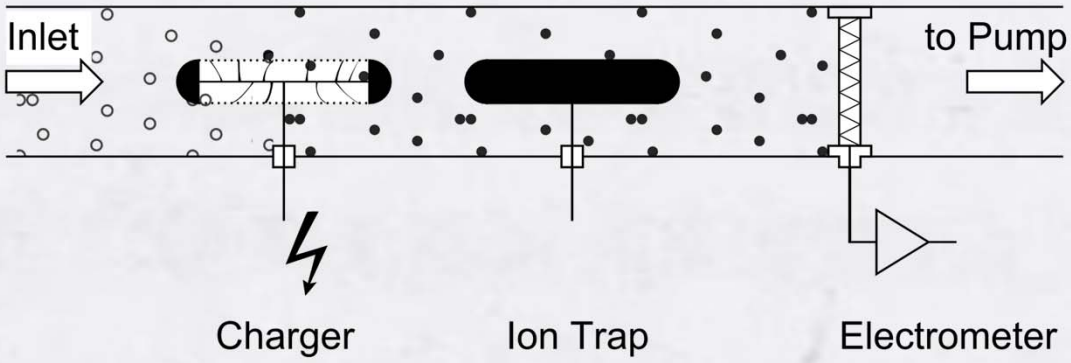
Lung-deposited surface area

- Can be measured by
  (1) measuring size distriution (e.g. SMPS) &
  (2) multiplying dS/dlogD by corresponding lung deposition probability (e.g. ICRP model)
- By a lucky coincidence, diffusion charging (DC) of aerosols produces an instrument response that is very close to LDSA!
- This is not a new observation – it was first made by W.E. Wilson of NIOSH – and implemented in the TSI NSAM, but it hasn't really caught on

Wilson W.E. *et al.*, "Use of the Electrical Aerosol Detector as an Indicator for the Total Particle Surface Area Deposited in the Lung," *Proceedings of 2004 A&WMA*, paper #37 (2004).

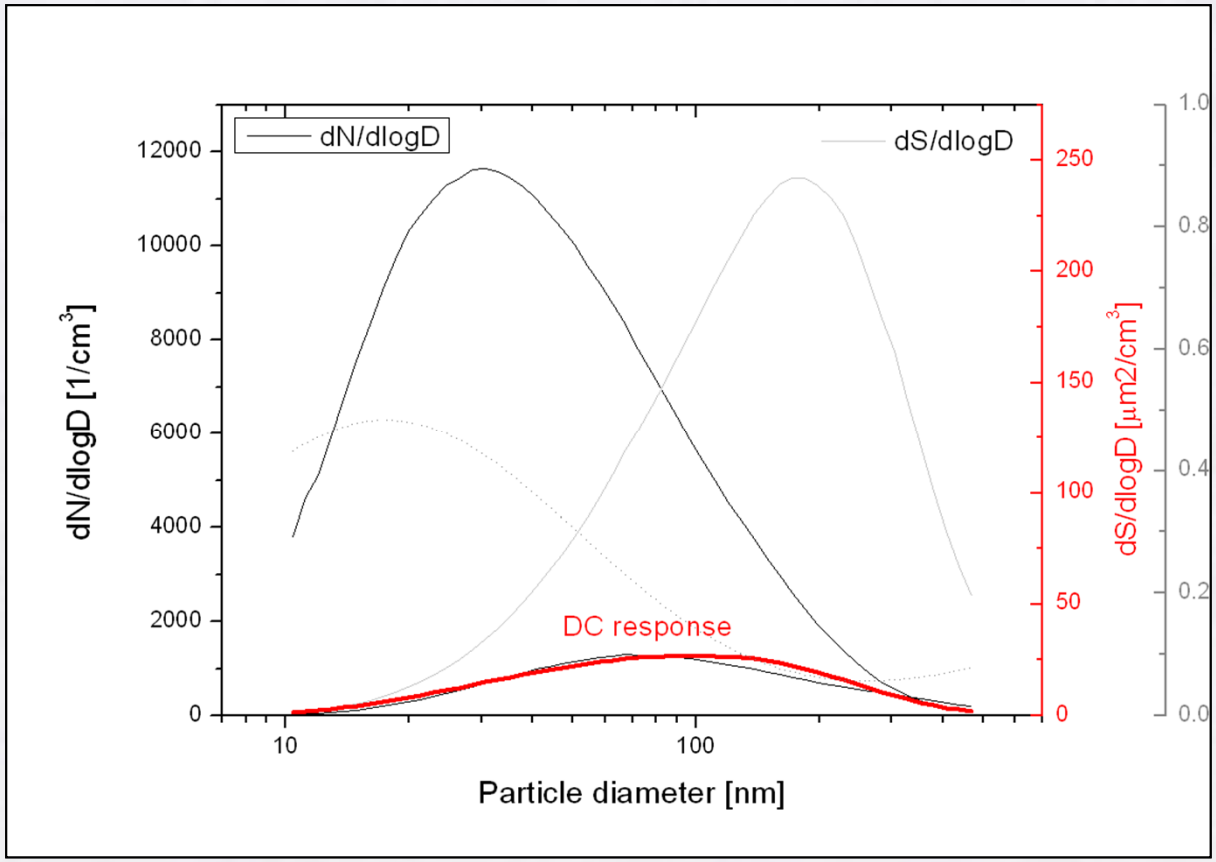
The diffusion charger (DC)

- Mix ions with particles = diffusion charging, remove ions, measure current in electrometer
- Measures current, but can be calibrated to LDSA



LDSA vs DC-signal

LDSA response and DC response to average Zürich aerosol



LDSA-capable instruments

- SMPS
- TSI NSAM, Aerotrak
- Matter DiSCmini
- but also any instrument with a DC inside: EcoChem DC, Pegasor PPS, Philips nanoTracer, Grimm nanoCheck, ELPI etc. – you just need to calibrate it accordingly.

DC "Dose meter"

- under development at FHNW
- shirt-pocket-sized (prototype = 272 cm^3)
- this instrument is my answer to The Mayer Question

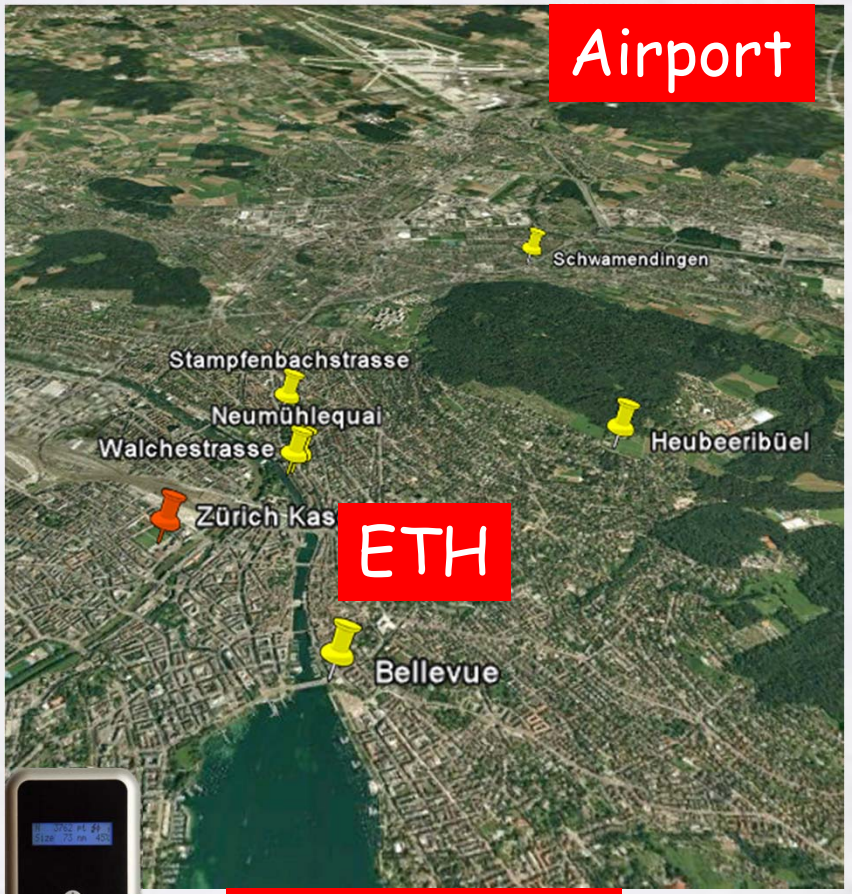
final thoughts on LDSA

- If you are looking at number, surface or mass, this is true
- The beauty of LDSA is that deposition is integrated in the DC measurement! size distribution does not have to be known!

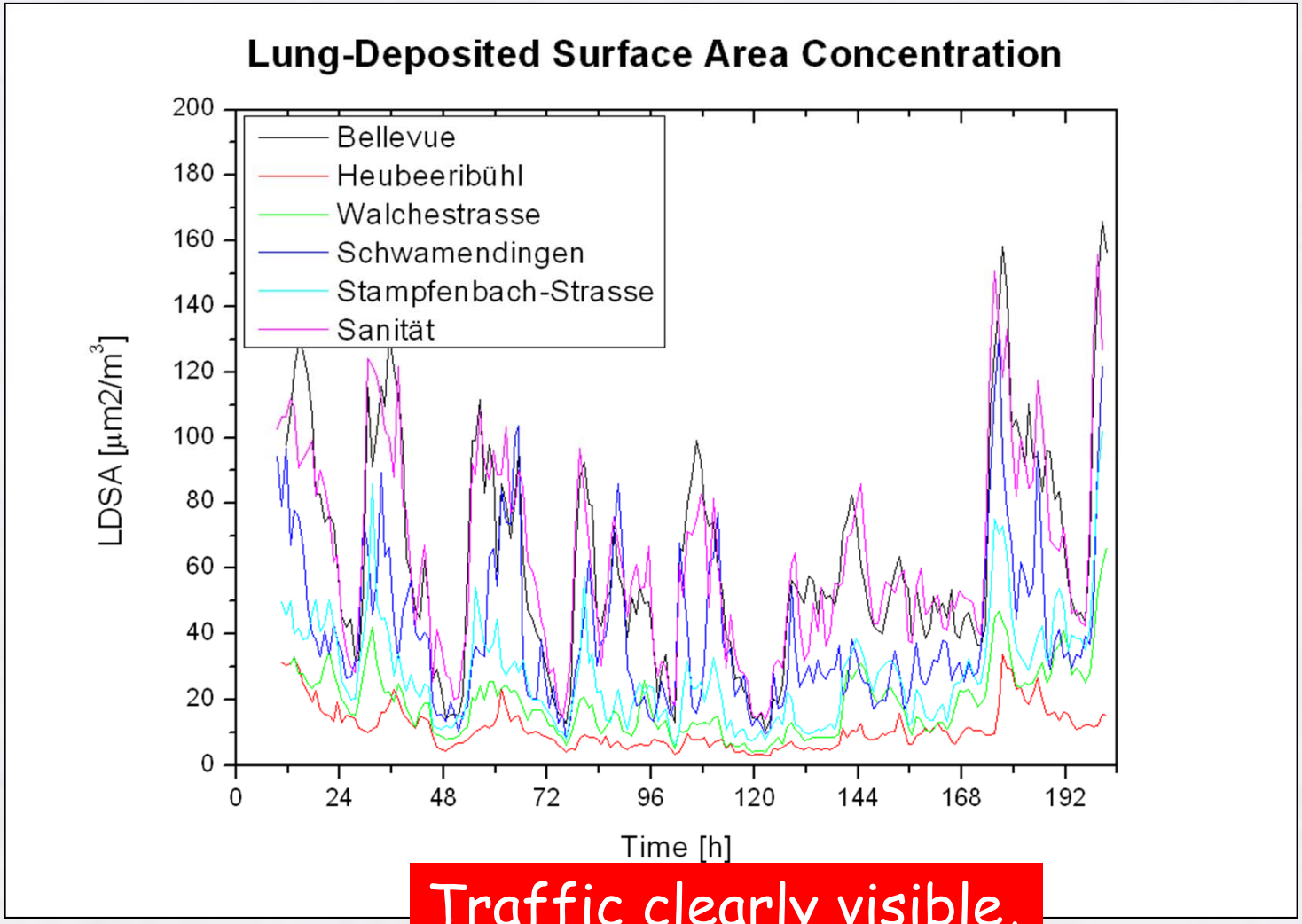
we need full information!
we need to measure the size distribution!

Measurements in Zürich

- 1st week of 9/2009
- 6 miniDiSCs (yellow)
- 1 co-located with 3775 CPC
- 1 co-located with UGZ home-built SMPS
- PM10 from NABEL (red)



LDSA time series



Traffic clearly visible, unlike with PM10

Aver…tions

Weak correlation for both PN/LDSA with PM10, but always better for LDSA

Station	<N> 1/ccm	<LDSA> µm²/cm³	R^2 N-PM10	R^2 LDSA-PM10	R^2 N-LDSA
Schwamendingen A1 (Highway)	16400	40	0.21	0.32	0.93
Bellevue (busy city road)	25600	63	0.34	0.44	0.95
Neumühlequai (busy city road)	31400	63	0.33	0.44	0.93
Walchest… (inner cit… traffic)			0.59	0.72	0.92
Stampfen… (average city road)			0.21	0.32	0.92
Heubeeribüel (no traffic)	4500	11	0.23	0.46	0.84

Factor 6 difference between heavy traffic and little traffic

Excellent correlation for PN - LDSA

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Conclusions

- Whatever we measure, the lung-deposited fraction is what is interesting ("per breath" instead of "per volume" metric) – in contrast to what is being done today!
- From collective work of toxicologists + biologists, I believe that LDSA is the most relevant physical parameter to measure – and by a lucky coincidence, it is easy to do so
- In traffic-dominated sites, LDSA correlates very well (R^2 ~ 0.95) with particle number, and you could measure either LDSA or PN – probably both would correlate with traffic-induced health effects
- Integrating LDSA-instrumentation in monitoring networks, especially in LEZ settings, would allow traffic-related emissions to be seen much more clearly (BC would serve a similar purpose)

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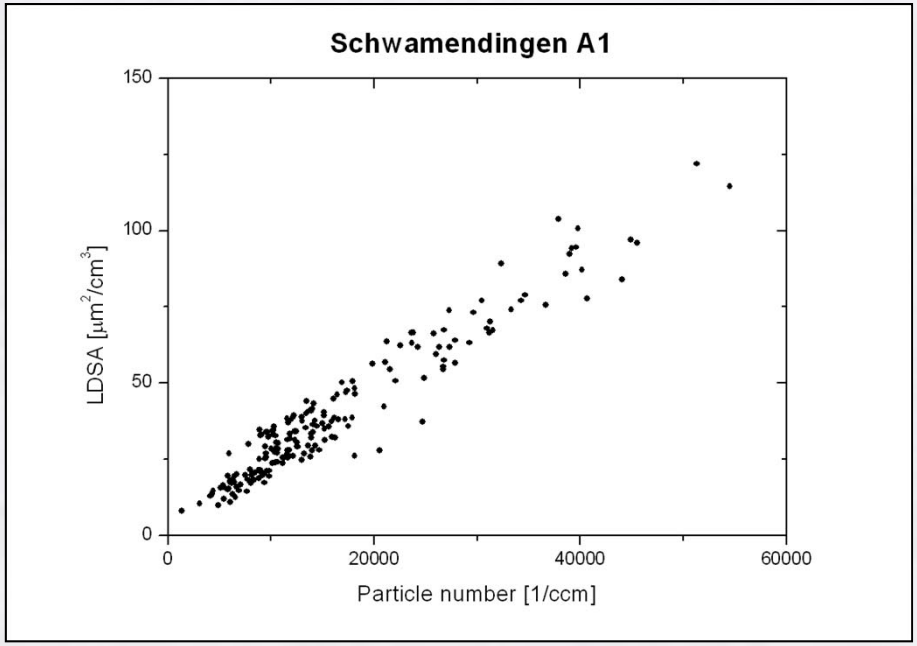
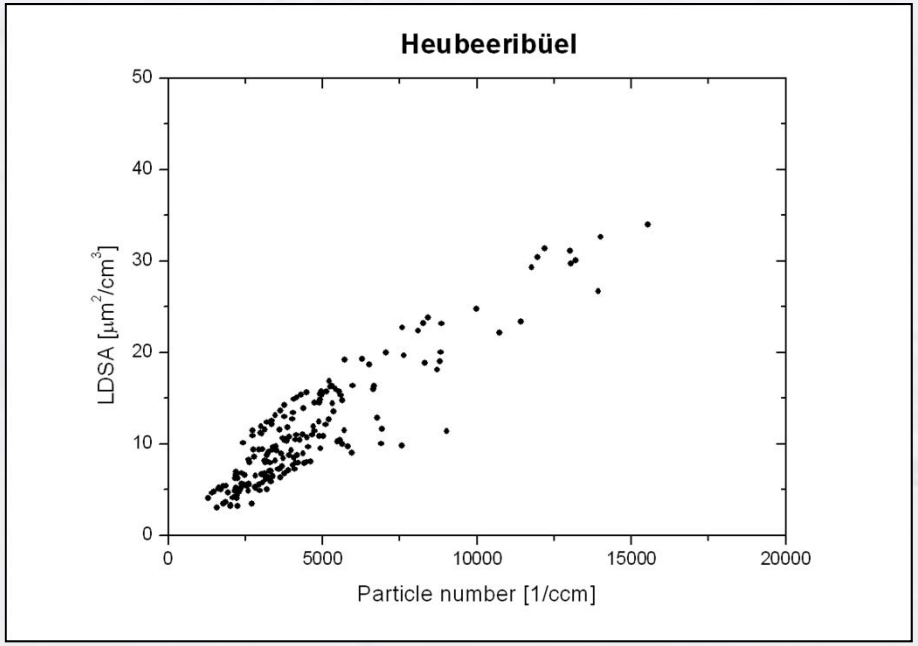
LDSA vs PN

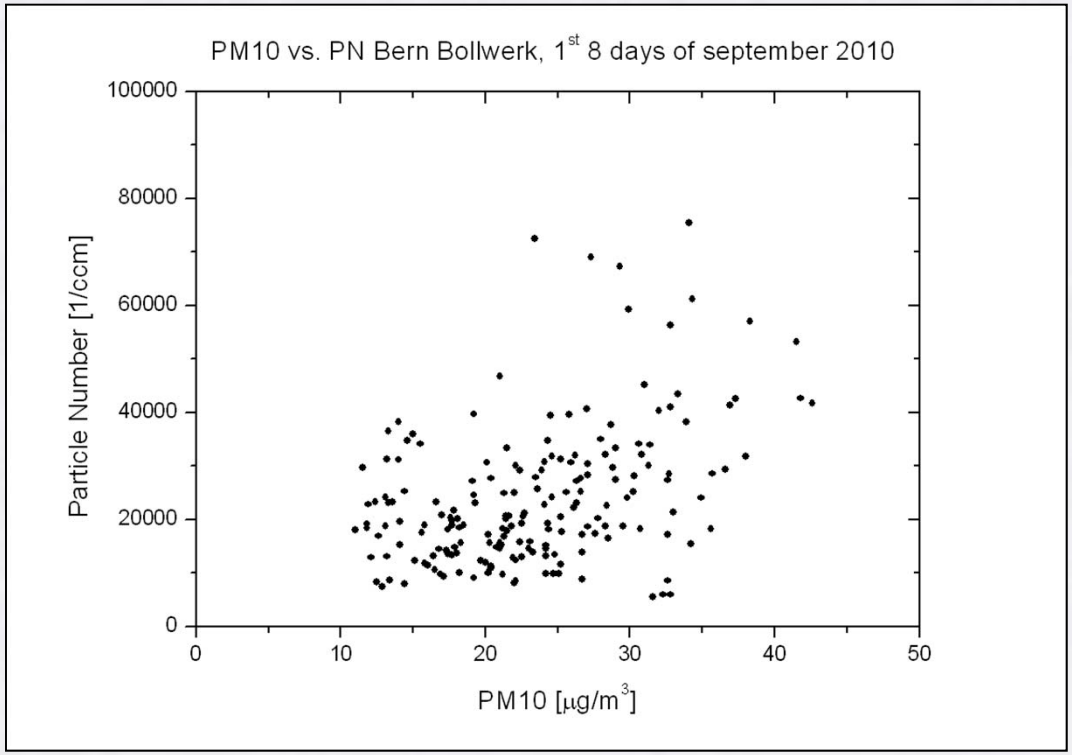
- better correlation PN/LDSA for sites with heavy traffic
- simple explanation: both PN/LDSA are dominated by traffic (>> background)





PM10 and PN at bollwerk

- PM10 vs PN/LDSA correlation not valid because not measured at same place?
- Perhaps, but look at Bern-Bollwerk NABEL station: (R^2 = 0.18)



Verification Results

- Verification for number concentration
  CPC vs miniDiSC: R^2 = 0.94, slope 0.89
  SMPS vs miniDiSC: R^2 = 0.95, slope 0.83
  (Thx to Jürg Brunner + Susanne Schlatter UGZ)