Diffusion charging for Easy Monitoring of Integral Particle Metrics

(Average Diameter, Number and Surface Concentration)

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Introduction

- miniature, simple and comparatively cheap instruments for UFP measurement based on diffusion charging have become available recently

DiSCmini

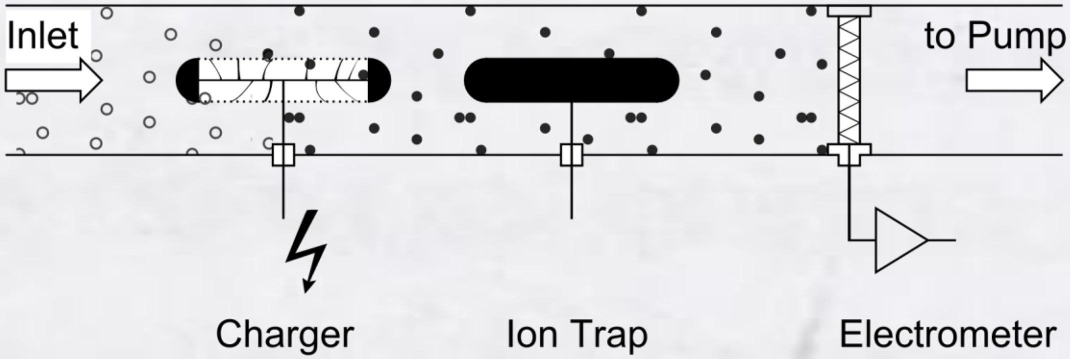
nanoTracer

nanoCheck

1. Diffusion charging basics and lung-deposited surface area (LDSA)

Diffusion charging basics

- label particles with electrical charges
- Detect currents at fA levels
- Simplest version: measure total current (Diffusion charging sensor or DC or DCS)



Diffusion charging result

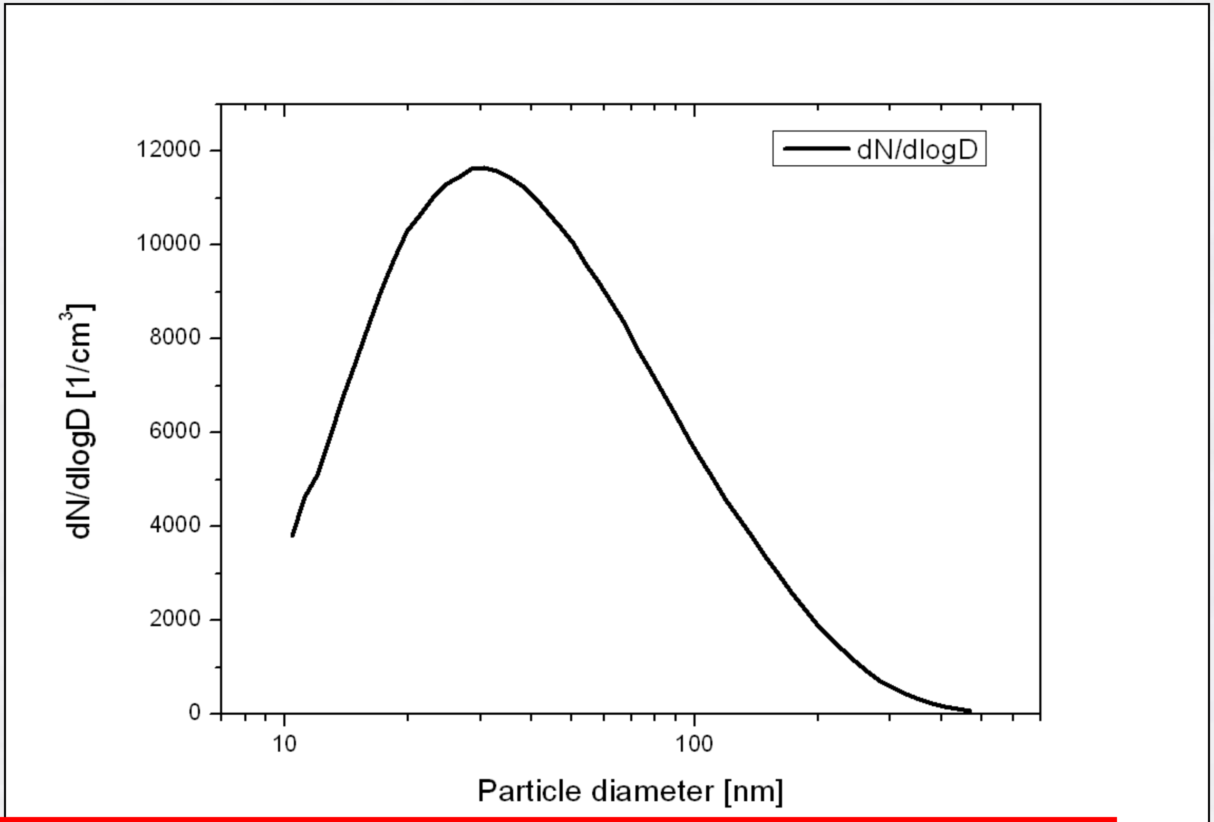
- Particles acquire a charge q that can be approximated by a power law:

  $q = a * d^b$ (d: diameter, a,b: constants, $b \approx 1.1$)

- What does the DC signal mean? It can be interpreted as lung-deposited surface area!

Surface area in Zürich

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



particle size distribution, number-weighted

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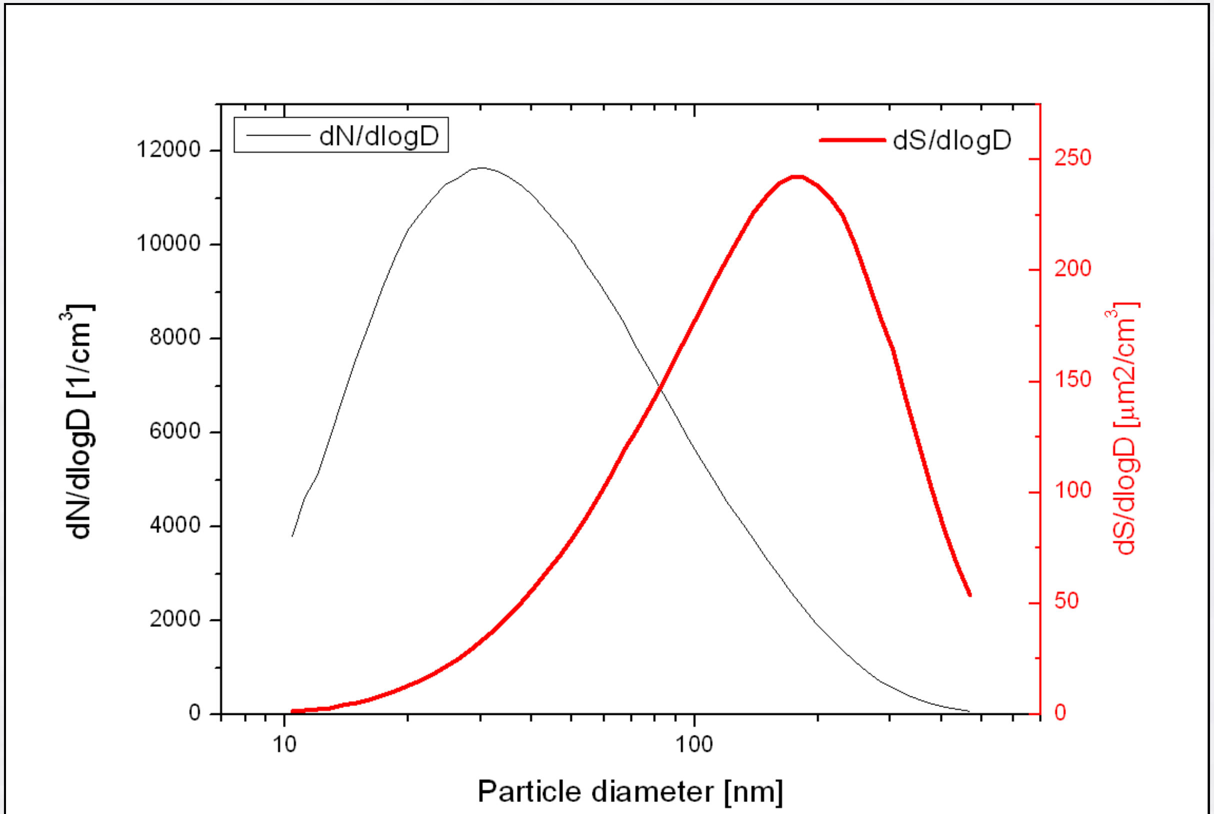
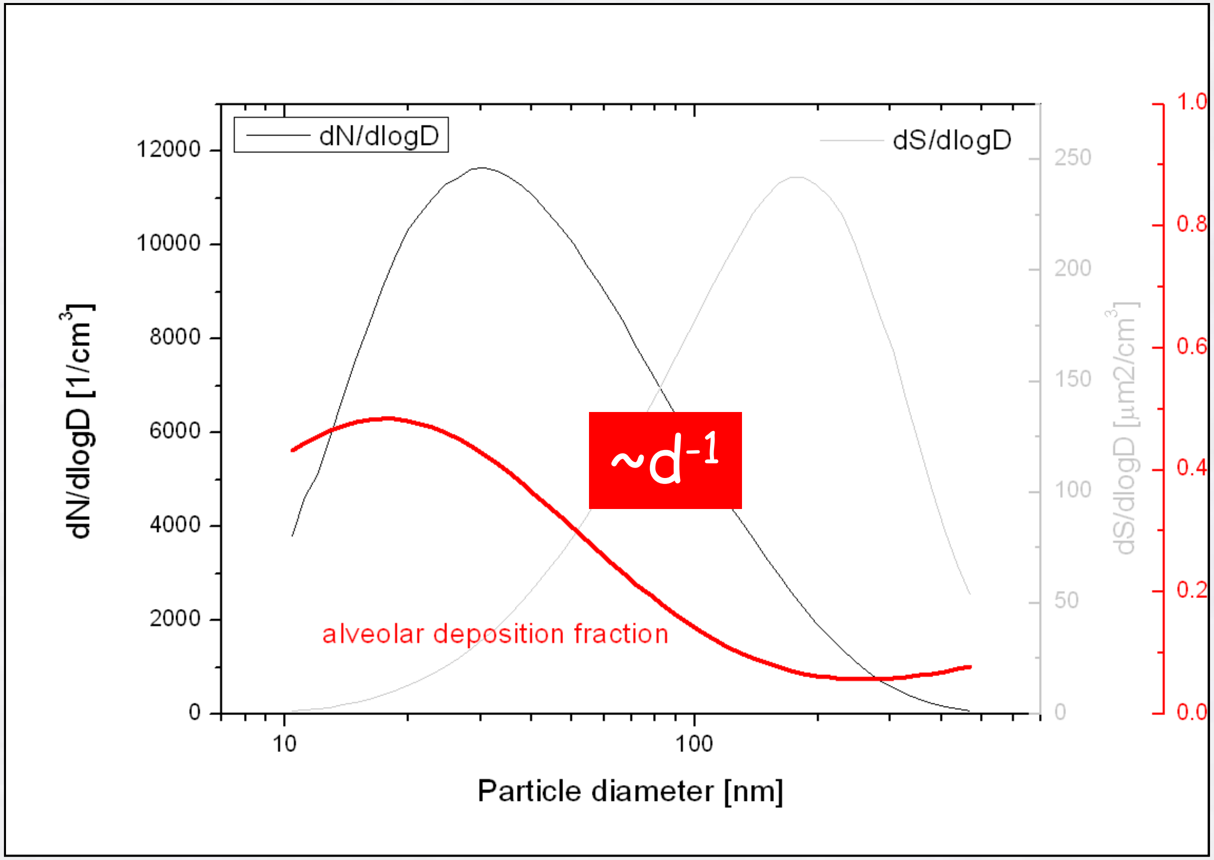


particle size distribution, surface-weighted

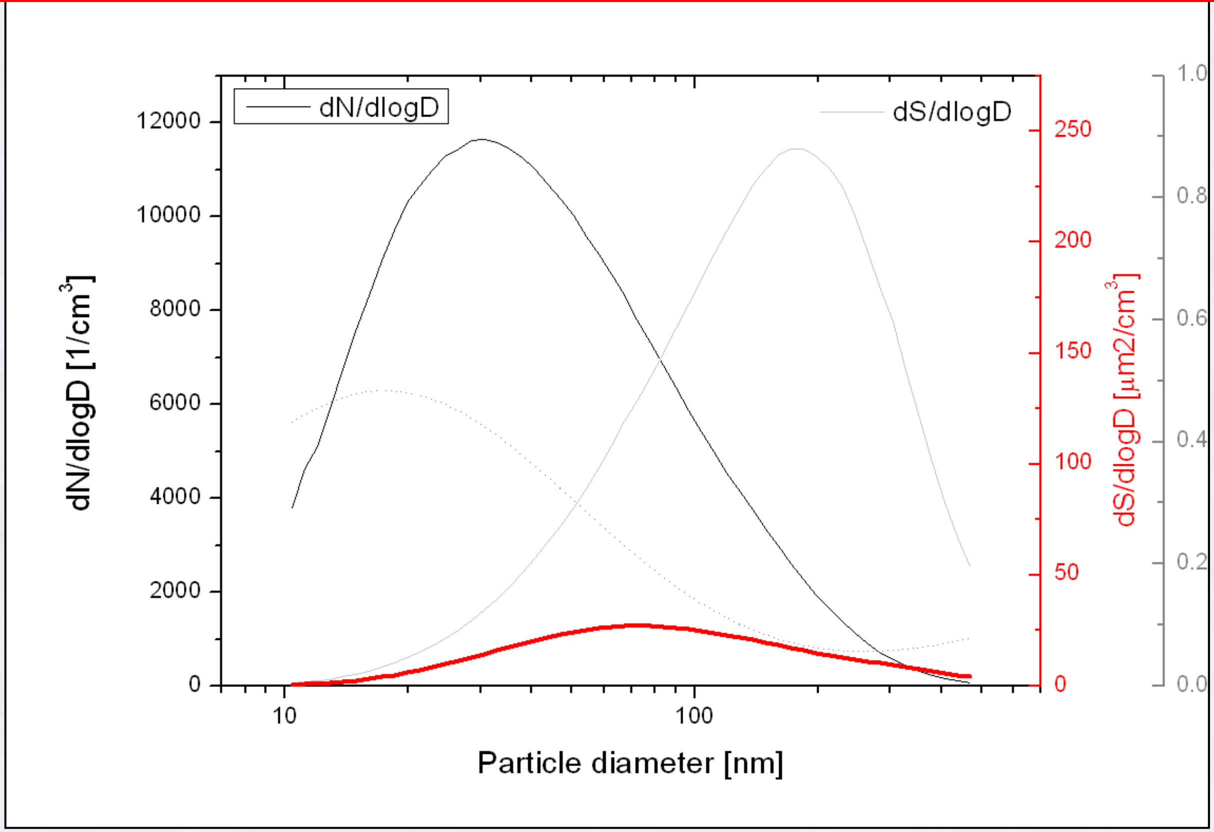
Surface area in Zürich

for health effects, we want to know what ends up in the body, so we multiply with the (alveolar) deposition fraction



Surface area in Zürich

This gives us the lung-deposited surface area distribution
Note how it is quite different from the original surface area distribution!



Surface area in Zürich

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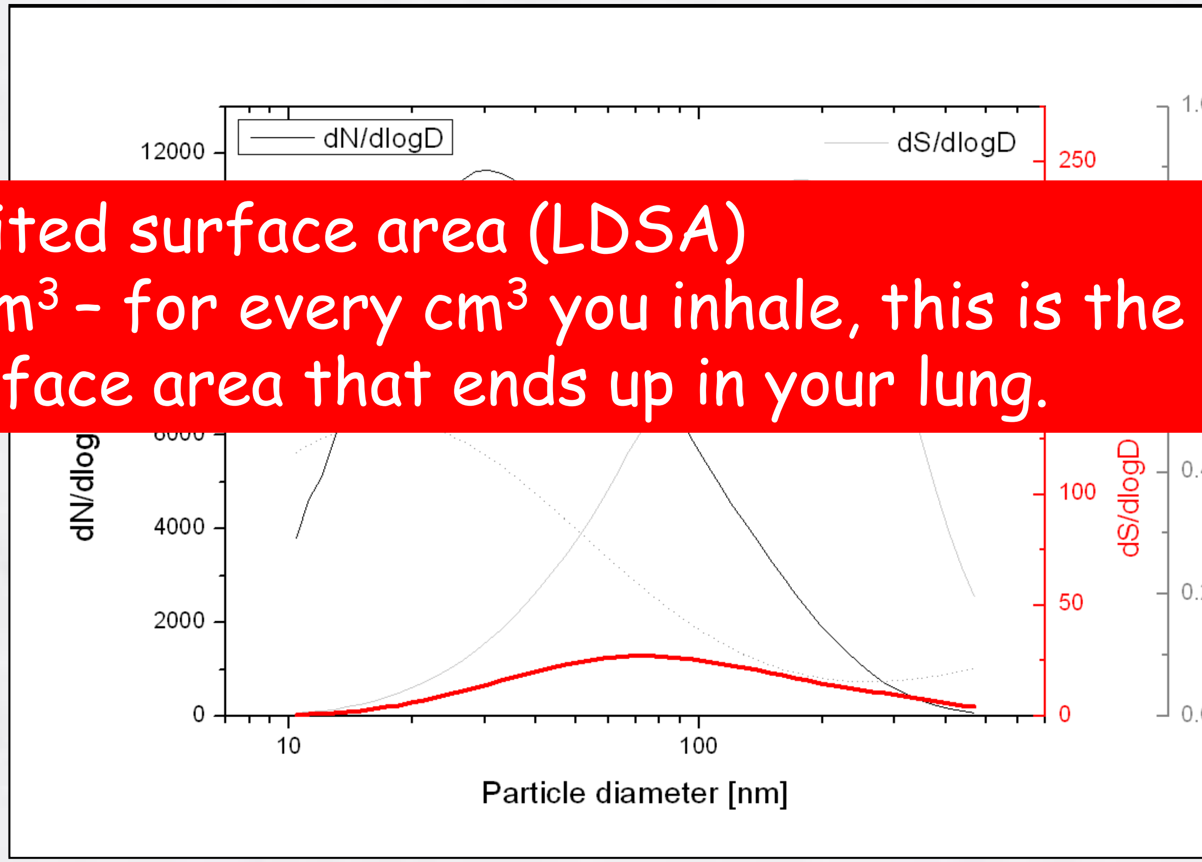


Lung-deposited surface area (LDSA)
unit: $\mu m^2/cm^3$ – for every cm^3 you inhale, this is the amount of particle surface area that ends up in your lung.

Lung-deposited surface area

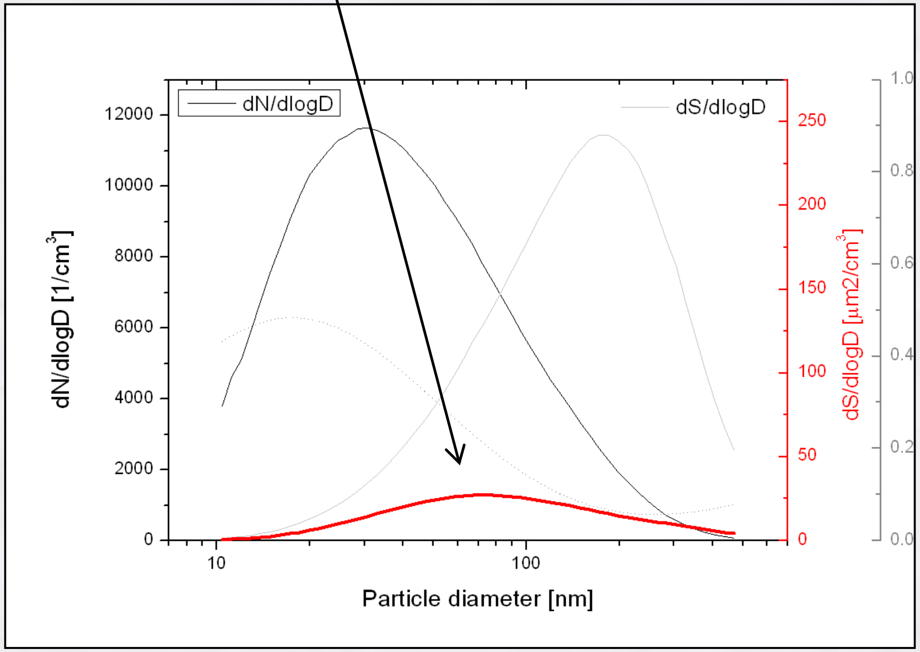
- Can be measured by
(1) measuring size distribution (e.g. SMPS) &
(2) multiplying dS/dlogD by corresponding lung deposition probability (e.g. ICRP model)
- By a lucky coincidence, diffusion charging (DC) of aerosols produces an instrument response that is very close to LDSA! (LDSA: $d^2 * d^{-1} \approx d^1$: DC)
- 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

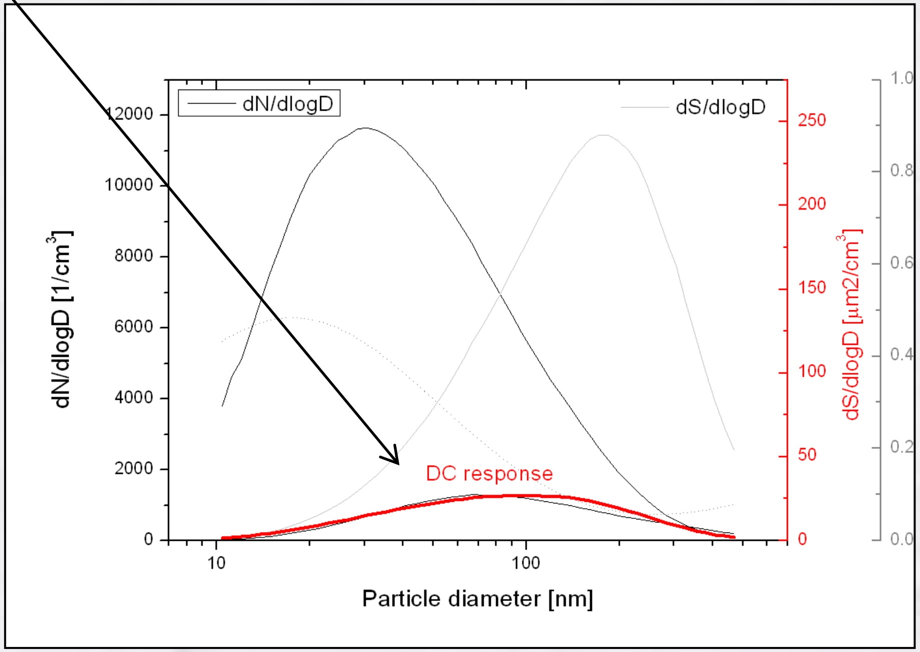
Wilson W.E. *et al.*, "Use of the Electrical Aerosol Detector as an Indicator for the Total Particle Surface Area Deposited in the Lung," *Proceedings of 2004 A&WMA*, paper #37 (2004).

LDSA vs DC-signal

LDSA response and DC response to average Zürich aerosol

They are not quite identical, but very similar





Conclusion on diffusion charging

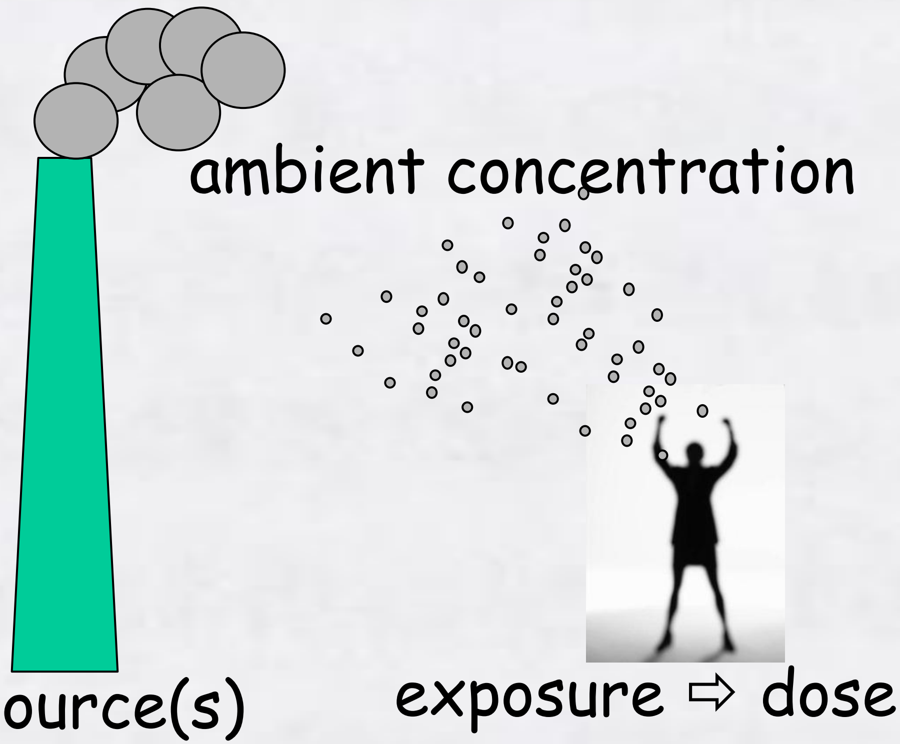
- No radioactive source needed as in SMPS
- No working fluid necessary as in CPCs
- Sensitive for nanoparticles (down to 10nm) unlike optical instruments
- Material-independent unlike optical instruments
- Simple, reliable, works in any orientation and can be miniaturized ⇨ personal exposure monitoring
- Measures LDSA, which is probably health-relevant
- can be extended to measure/estimate particle number and average particle diameter

2. Health relevance

health relevance

air pollution monitoring is done not for its own sake, but because pollution leads to health effects

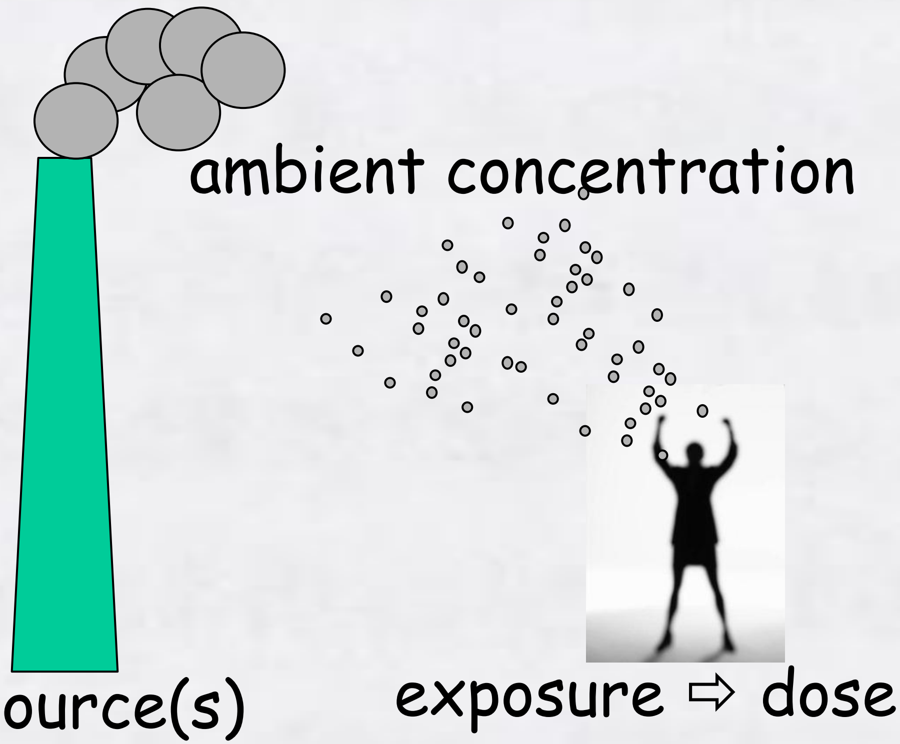
pollution sources lead to ambient concentration which leads to exposure which leads to a particle dose which leads to effects



dose	⇨ acute effects
retained dose	⇨ chronic effects

health relevance

exposure and dose are NOT the same; they are related by the particle size dependent uptake of particles in the body (lung deposition efficiency)



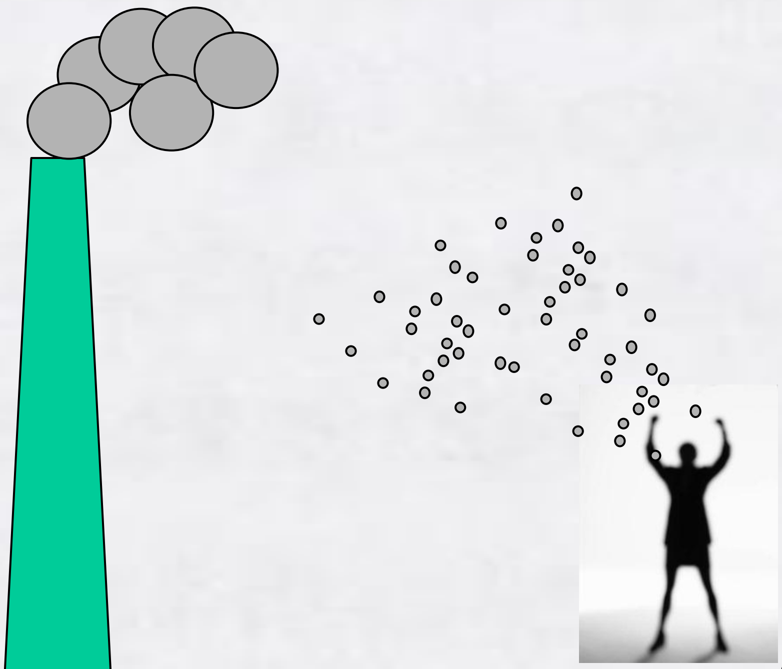
dose ⇨ acute effects

retained dose ⇨ chronic effects

health relevance

we would like to measure health effects directly, but can't

ambient concentration



source(s)

exposure ⇨ dose

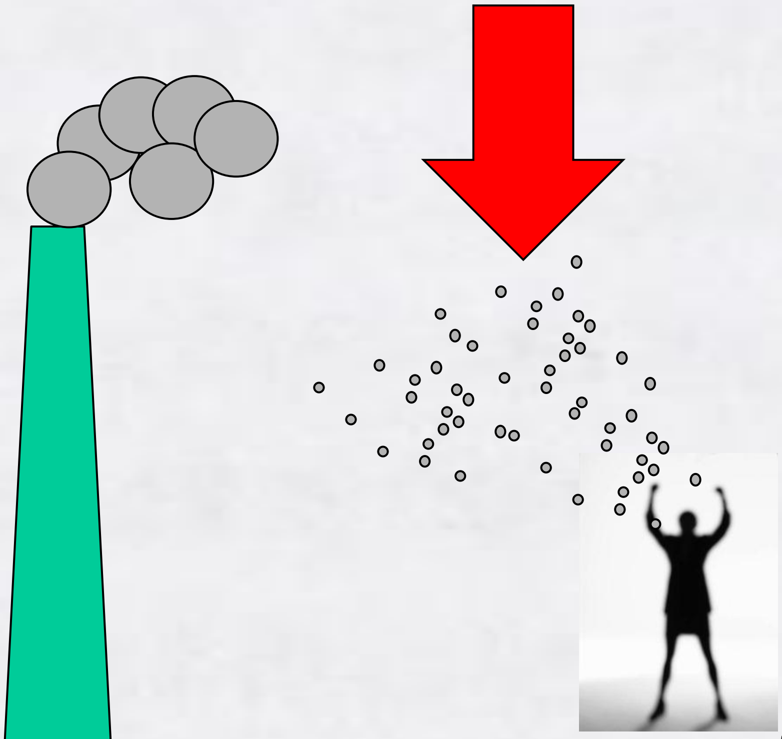
dose ⇨ acute effects

retained dose ⇨ chronic effects

health relevance

instead, today we measure ambient concentration (exposure) which is "far away" of health effects

ambient concentration



source(s)

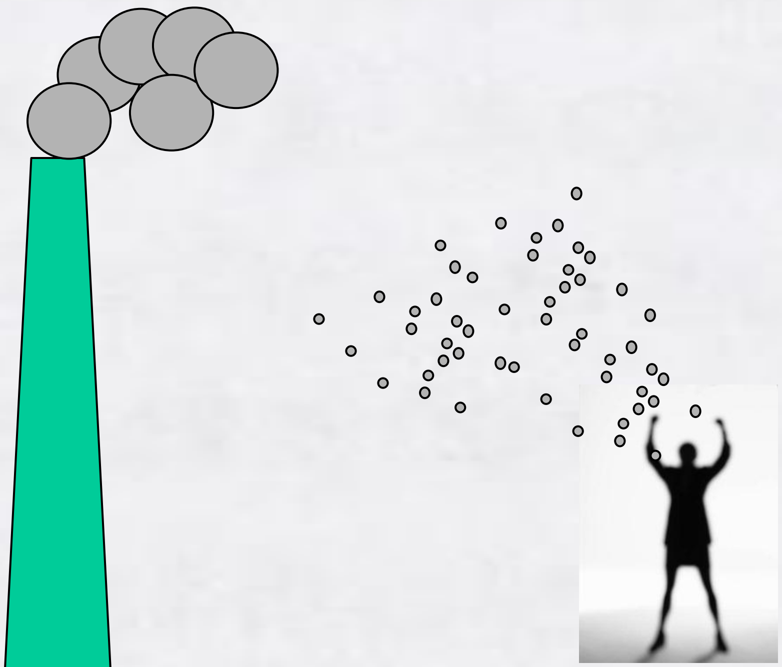
exposure ⇨ dose

dose ⇨ acute effects

retained dose ⇨ chronic effects

LDSA measurement brings us to a dose, one step closer to what we actually want to measure

ambient concentration



source(s)

exposure ⇨ dose

dose ⇨ acute effects

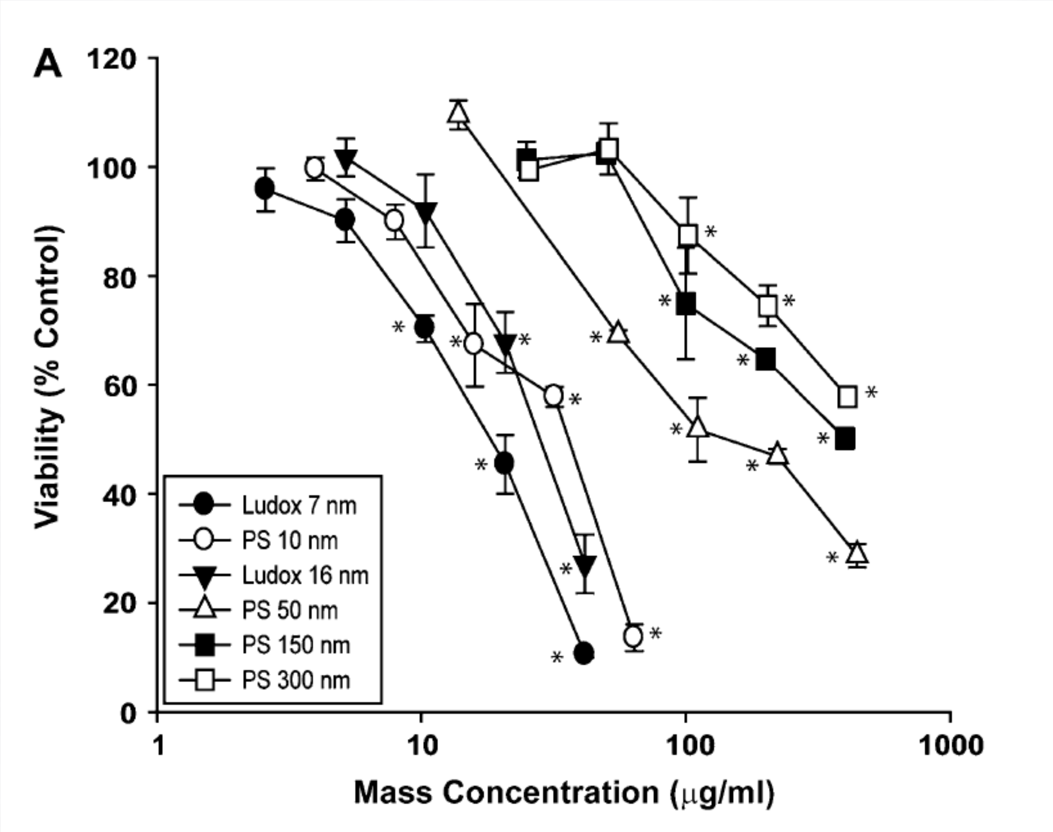
retained dose ⇨ chronic effects

Toxicologists vote for surface area

- Next slides show 3 plots from K.M.Waters et al. *Tox Sci* **107(2)**, 553-569 (2009)
- Macrophages exposed *in vitro* to amorphous silica particles from 7 – 500nm diameter, measured biological endpoint is macrophage cytotoxicity
- There are many similar examples in the literature, e.g. for other particle types, and for in-vivo experiments (mice, rats)

Toxicologists vote for surface area

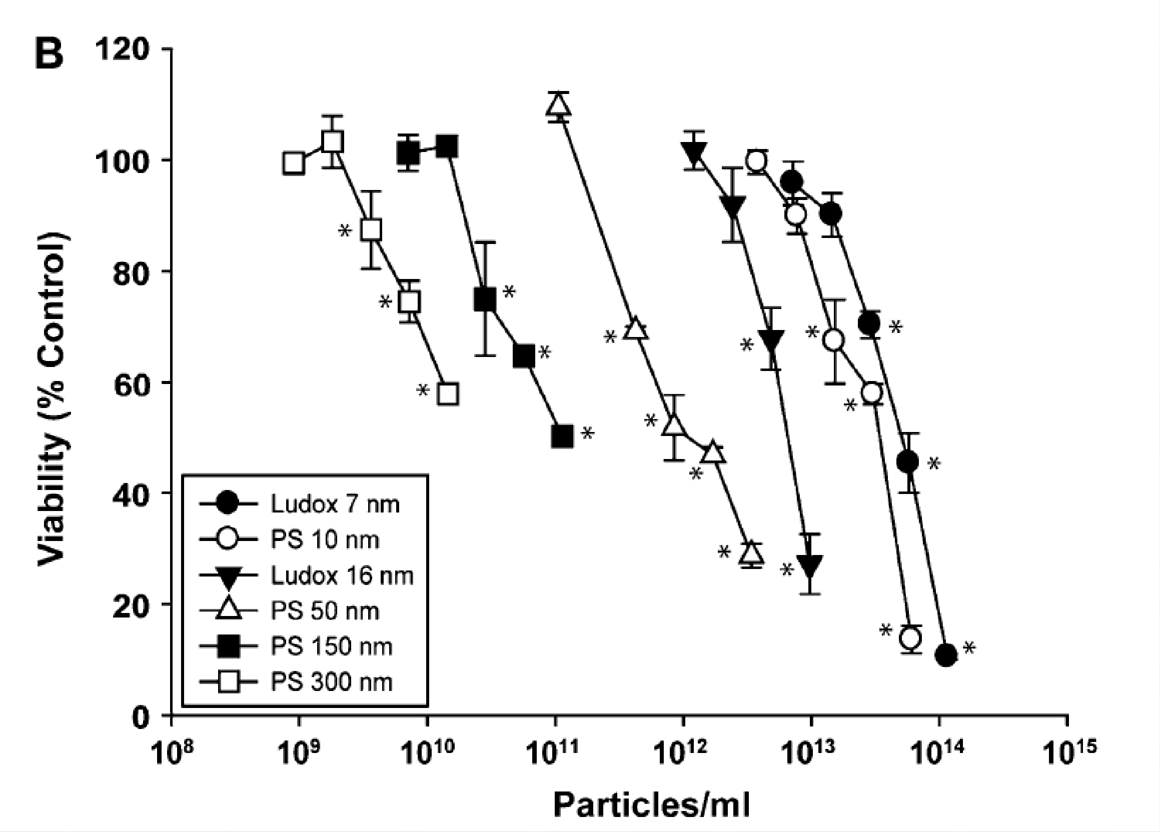
- Toxicity as function of particle mass: small particles are more toxic



K.M.Waters et al. *Tox Sci* **107(2)**, 553-569 (2009)

Toxicologists vote for surface area

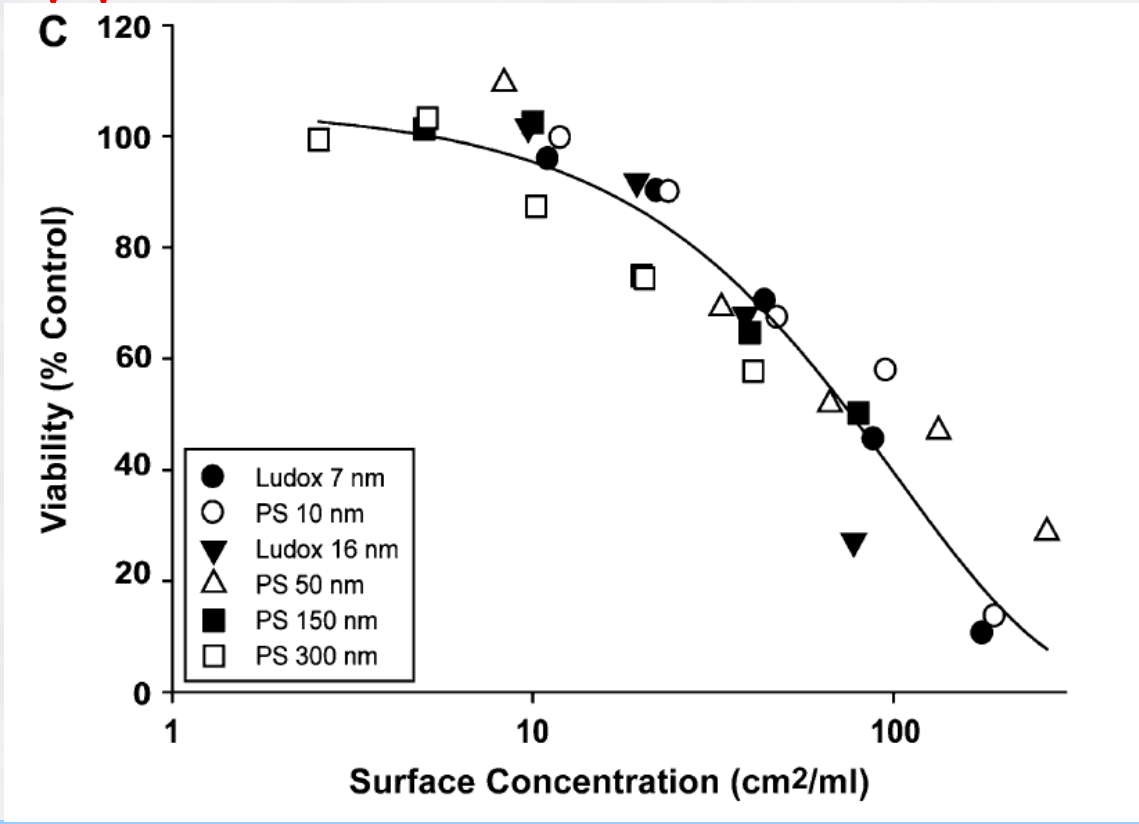
- Toxicity as function of particle number: large particles are more toxic



K.M.Waters et al. *Tox Sci* **107(2)**, 553-569 (2009)

Toxicologists vote for surface area

- Toxicity as function of particle surface: all particles fall on one line, i.e. the toxicity is driven by particle surface area

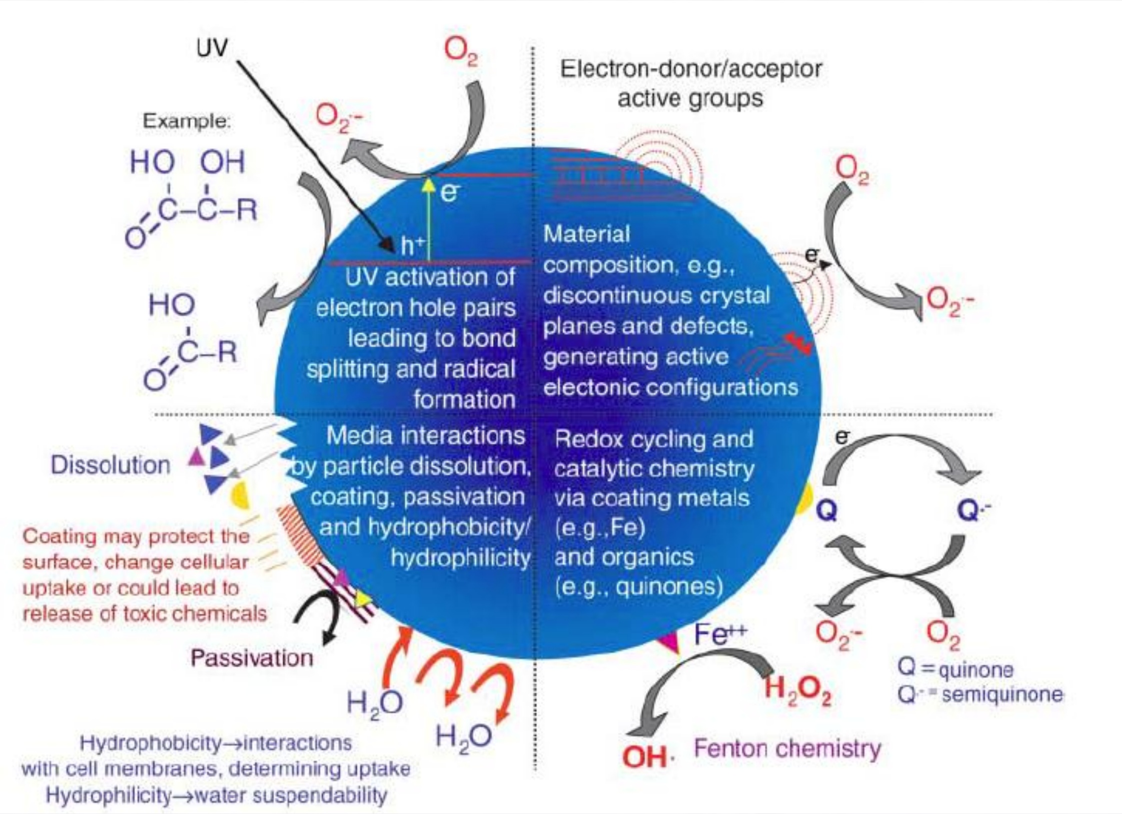


If this does not convince you, then probably nothing will...

K.M.Waters et al. *Tox Sci* **107(2)**, 553-569 (2009)

Toxic Potential of Materials at the Nanolevel

we also have a mechanistic understanding why particle surface area is important – e.g. by producing ROS



Andre Nel, et al. Science 311, 622 (2006)

My personal take on health effects

- Epidemiology tells us that there are traffic-related health effects that cannot be explained by PM10 (e.g. asthma, heart attacks increased when living close to busy streets)
- To explain these effects in an epidemiological context, we need a variable that is independent (not correlated to) of PM10 ("orthogonal base vector" in linear algebra)
- There is toxicological (not epidemiological) evidence that particle surface area is the most sensible physical (disregarding chemistry) metric
- LDSA is a great candidate for the "missing" orthogonal base vector (of course there are also other candidates, such as BC, ROS activity etc.), in particular because it is already a dose, and not an exposure (which is what you get when you measure BC).

3. Simple instruments based on DC

Instruments

bias warning: I was involved in the development of 3 of these instruments (but all 6 are from different manufacturers)

Further information on the web

Diffusion chargers:

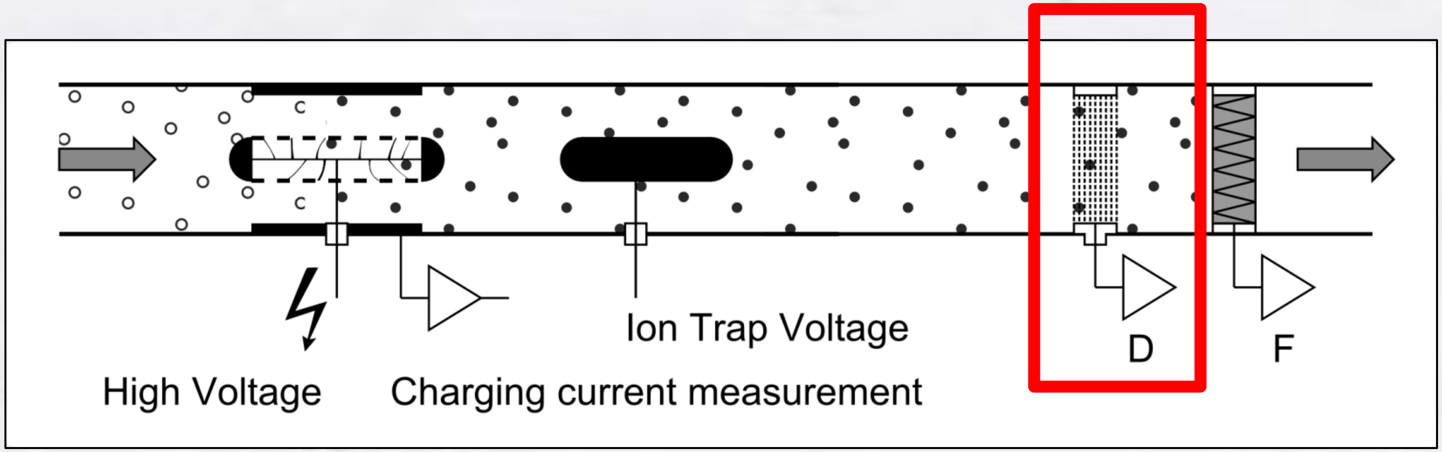
- www.tsi.com (NSAM)
- www.naneos.ch (Partector)
- mailto: antti.rostedt (at) tut.fi (nanoDevice)

Extended to measure number & average size:

- www.matter-aerosol.com (DiSCmini)
- www.aerasense.com (nanoTracer)
- www.grimm.com (nanoCheck)

miniDiSC / DiSCmini

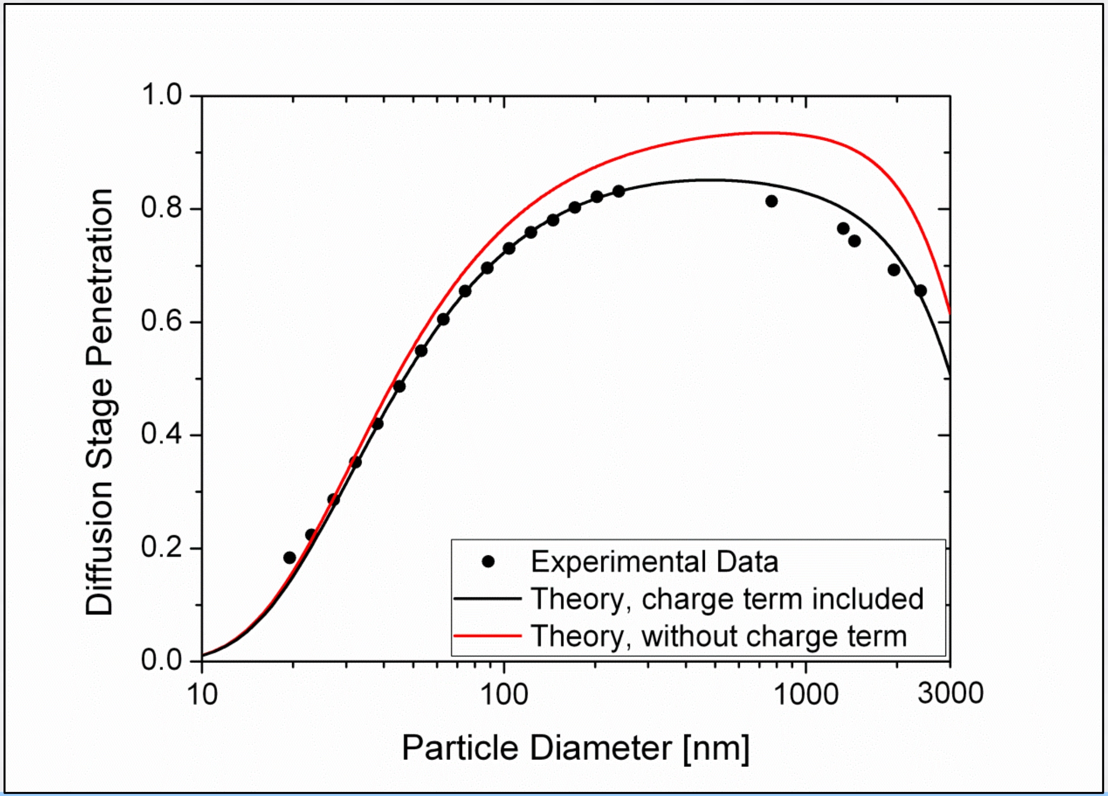
- add a stage consisting of stainless steel grids where particles are deposited by diffusion



- measure 2 currents simultaneously, D and F (on diffusion and filter stage) with 1s time resolution
- Calculate number concentration and average particle diameter from F and D

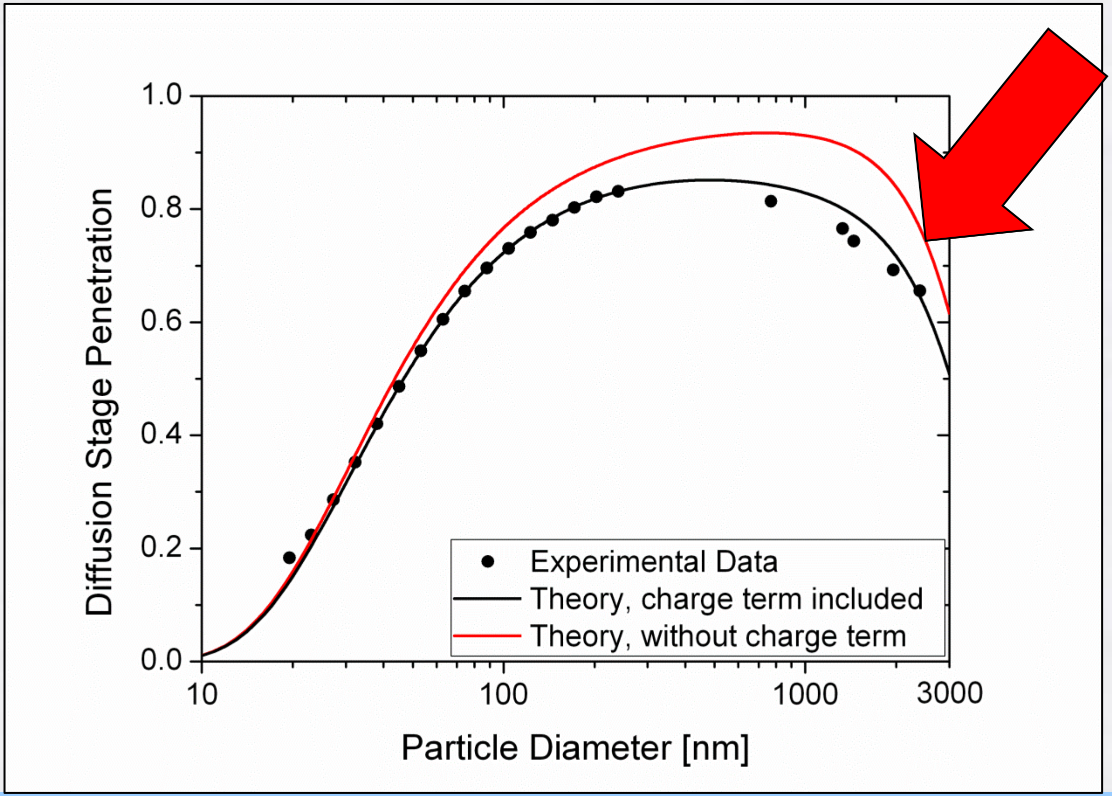
miniDiSC "data inversion"

- measure penetration through diffusion stage
  P = F / (F+D) for monodisperse particles
- compare measured P with calibration value



miniDiSC "data inversion"

- Because P is not a monotonic function of diameter, this only works if the number of coarse particles is low (this is mostly the case), or if you remove them with pre-separator

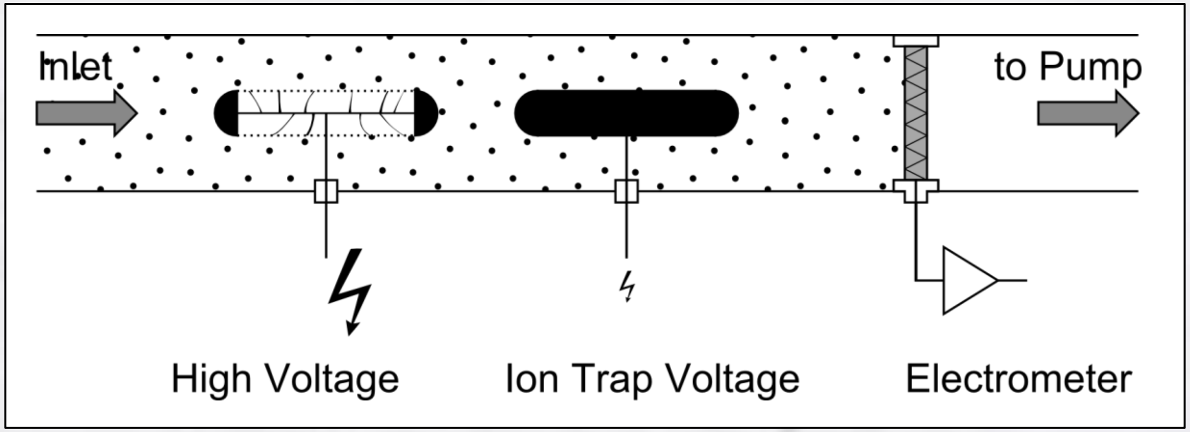


Calibration for polydisperse aerosol

- Real aerosols are not monodisperse
- Larger particles carry more charge, and thus more weight in the signal
- To calculate the correct particle diameter, and from this the correct particle number, we need to make an assumption on the particle size distribution
- assumption: lognormal with GSD 1.9
- As long as this is approximately fulfilled, results are approximately correct

nanoTracer / nanoCheck

- adjustable voltage on ion trap high/low/high/low (essentially no change necessary, very nice)
- 2 Signals (DC Signal I_{total} and loss ΔI at higher trap voltage) ⇨ use exactly the same ideas for data inversion as in miniDiSC



nanoTracer / nanoCheck

- Lower time resolution due to sequential measurement (6s time resolution for nanoCheck, 16s time resolution for nanoTracer)
  ⇨ Problems with rapidly changing aerosols
- Asbach et al. "comparability of portable nanoparticle exposure monitors", *Annals of occupational hygiene*, in press: miniDiSC performed better than nanoTracer / nanoCheck
- In principle, there is no reason why this should be the case.

Conclusion on sizing/counting instruments

- Sizing+counting instruments (DiSCmini / nanoCheck / nanoTracer) are limited by unknown particle size distribution
- By choosing a "sensible" particle size distribution in the calibration process, errors are limited to about ± 30%
- This error cannot be avoided (no matter what other instrument manufacturers tell you)
- Large particles will confuse sizing/counting instruments! Pre-separators necessary for ~500nm (but unreliable and/or problematic)

- Compared to CPCs, the miniDiSC seems more accurate than pTrak (which has a very low counting efficiency for small particles), but less than 3007 CPC; the miniDiSC has a higher upper limit (10^6 pt/ccm) than the 3007 (10^5 pt/ccm), which can be useful in personal exposure monitoring

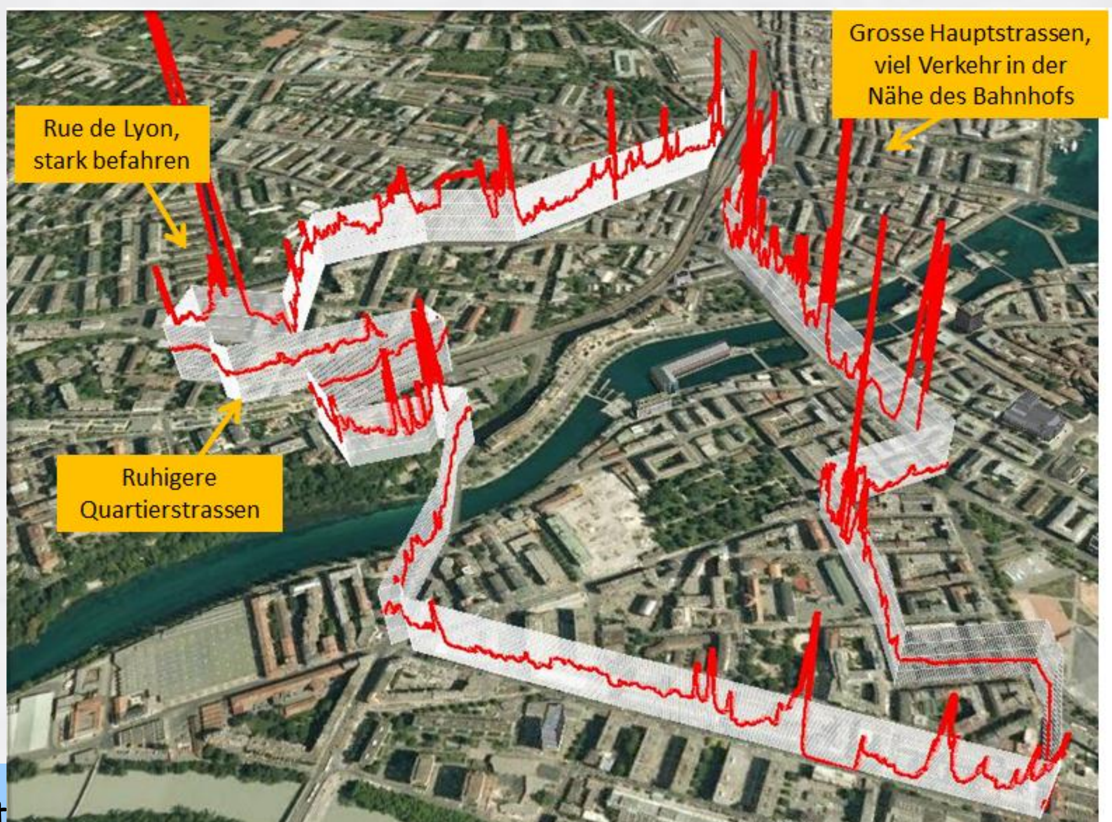
Miniaturization and urban monitoring

- Most instruments were designed with miniaturization in mind – this leads to a number of tradeoffs, in particular for reliability.
- None of these instruments is really well suited for 24/7 monitoring (perhaps with exception of TSI NSAM) ⇨ service necessary all 2-3 months (?)
- However, it would be easy to improve instruments in this respect if interest is here (e.g. with better pumps)
- DC instruments are comparatively cheap ⇨ sensor networks with high spatial resolution possible

4. Three applications

Personal exposure

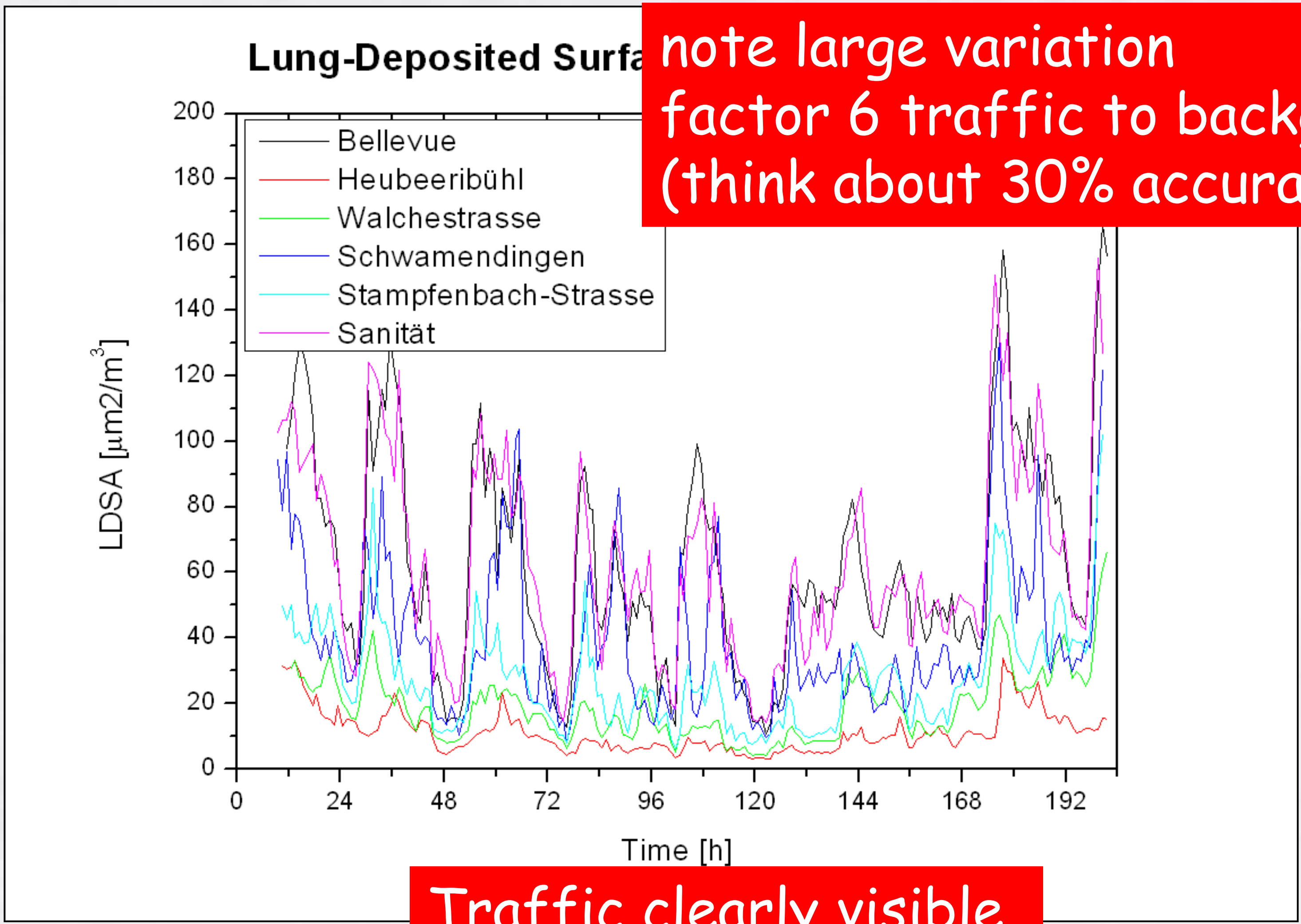
- nearly everyone has a smartphone with GPS
- alternatively, miniDiSC with built-in GPS and wireless data transmission to server available



LDSA 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



note large variation
factor 6 traffic to background
(think about 30% accuracy)

Traffic clearly visible,
unlike with PM10

Averages & Correlations

low correlation to PM10 background

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
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

Excellent correlation for PN – LDSA because both are dominated by the same source, traffic

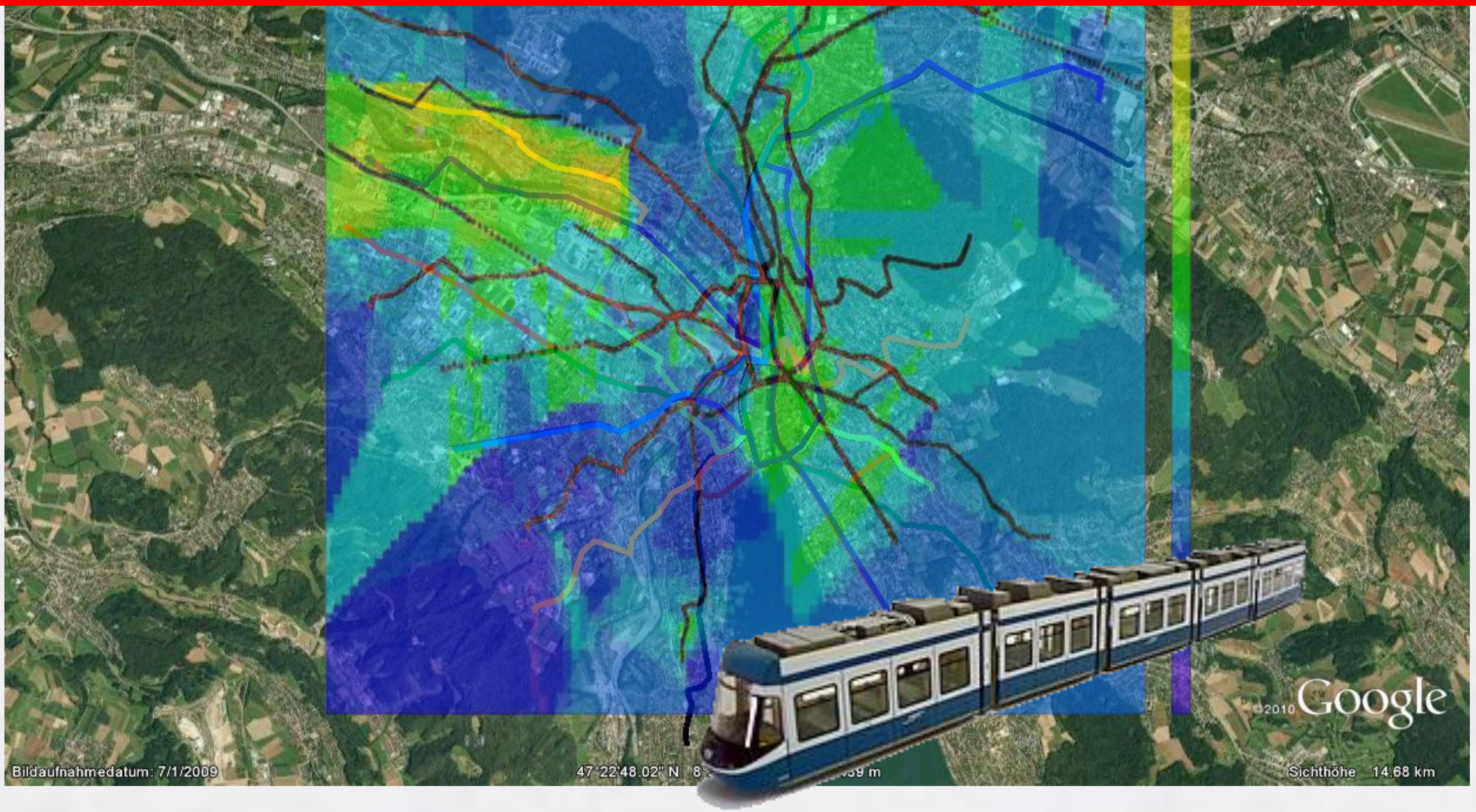
Tram project in Zürich

10 trams are being equipped with GPS, data transmission, gas sensors (all ETH Zürich) and miniDiSC (FHNW)

Measurements must run for months unattended! DC-based instruments can do this easily

Tram project: Kriging

Zürich has a very dense tram network. We will produce pollution maps with Kriging interpolation, or land use models



Conclusions

- New small / simple / cheap sensors based on diffusion charging available that measure number / diameter / LDSA
- From work of toxicologists, I believe that LDSA is the most relevant physical parameter to measure – and by a lucky coincidence, it is easy & cheap to do so
- Integrating (cheap) 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) and should be seriously considered.
- Remember yesterday's talk of Mrs. Katsouyanni: we need good data on an air pollutant first in order to generate epidemiological evidence!

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