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Legal Aspects of Particle Measurements

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Fourth International Workshop on Nanoparticle Measurement, August 7 / 8, 2000

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1 The Amount of Particles

1.1 What is a measurable quantity?

The term "legal aspects" in the title implies the reflections and considerations concerning which "quantity" has to be measured and how it is guaranteed, that the aimed quantity can be measured with the appropriate precision and with a known uncertainty.

Definition of quantity according VIM [1)]: *The (measurable) quantity signifies the attribute of a phenomenon, body or substance that may be distinguished qualitatively an determined quantitatively*.

The quantity length of a rod for instance can be defined differently (figure 1): 1. Microscopic distance on the surface between two edges. 2. Length of a longest straight line through the rod. 3. Length of the straight line between the points "in the middle" of the two ends.

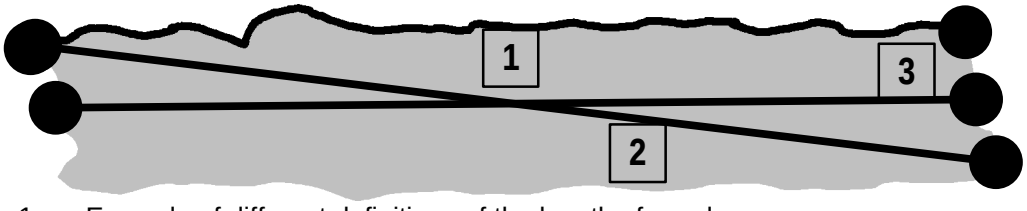


Figure 1: Example of different definitions of the length of a rod

Each definition is useful to answer the different characteristics of the rod. This example show that the purpose of the measurement must be known in advance. Therefore metrology must be engaged in defining quantities in order to be able to establish the requirements for measuring instruments.

1.2 Which Quantities [2)] describe Particles?

The question about the suitable quantity is essential in the field of particle measurement:

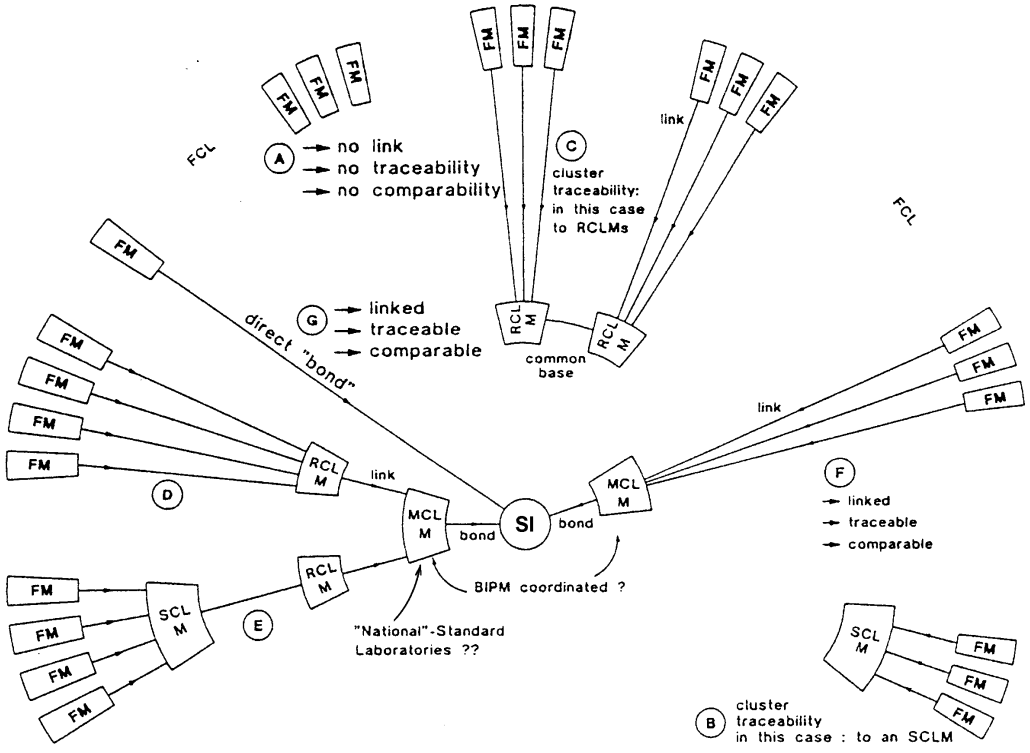
- The amount of substance is required for the investigation of combustion processes, toxicity of soot etc.
- The number of particles of a specific size seems to be a quantity for the mechanical irritation in the lungs and therefore a quantity for the health hazards [3)]. This quantity shall investigated closer.

- The optical characteristics indicate the interference with visibility and are important for the road safety.
- The photoemission correlates with surface effects and shows the potential of binding adsorbates.
- The mobility determines the diffusion coefficient and the mechanism of deposition of particles in the lungs [4)]. Together with the number of particles this quantity must be taken into account.
- The aerodynamic diameter is a quantity for the deposition, if the inertia of the mass is dominating. This is important for particles diameters above about 10 µm.

These examples show that measurement is a tool in order to get suitable answers. Before a measurement it must be clear which qualitative characteristic shall be compared or quantified.

2 Concept of Traceability

Assume the measurable quantity is sufficiently defined. The results of measurements from different laboratories are only equivalent, if all laboratories have established an unbroken chain of traceability to a common unit (e.g. SI).



Figur 1 : Scheme of different grades of traceability according P. De Bièvre and P. Taylor, Institute for Reference Material and Measurement, Belgium. FM = Field Measurement from field chemical laboratories; SCLM = Measurements of sectorial chemical laboratories; RCLM = Measurements of metrological chemical laboratories; BIPM = Bureau international des poids et mesures.

In figure 1 the situation is expressed for traceability in the field of analytic chemistry schematically:

- Cluster A shows the situation, where independent laboratories carry out field measurements (FM). Their results may be consistent within their own laboratory, but often differ from one to another laboratory. There is no way to determine the correct value.
- In cluster B and C different laboratories get the reference material (materialized standards for calibration) from sectorial chemical laboratories (SCL or as specialized laboratory: RCL). Their results are comparable. But the deviation of the measurement from the unit system SI - the error of measurement [1)] - is not known. The results are not comparable to results of laboratories outside the cluster.
- In cluster D to G the measurements are linked to the unit system SI. They are traceable and therefore comparable.

The measurement of the number of particles and their sizes is nowadays situated in cluster A of figure 1.

3 Need for Traceability

During the last ETH-Workshop a slight skepticism of the experts could be noticed regarding the results of other experts. This skepticism is necessary in order to detect errors and prevent them in the own measurements. But it must be reduced, before a measuring method can leave the field of fundamental research and enter trade, health security, or environment protection.

The main reasons for realization of traceability are:

- Improvement of comparability between different research groups
- Improvement of comparability between different constructions of instruments developed to measure the same quantity.
- Acceptance of the measuring method by non specialists
- Ultimate condition for the application of a measuring method in the legal field (laws, directives on national or international level, standards)

4 Realization of the Traceability

The realization of the traceability consists of several steps. Central elements are: Modeling of the measurement, the calibration of all relevant components, estimation of the uncertainties and the validation of the instruments by means of a round-robin-test.

4.1 Modeling the Measurement

The characteristics and the performance of the instrument shall be determined. Therefore the relations between all sources of influence and the result of measurement must be identified and quantified. This is the most demanding process of the modeling. In the ideal case it is possible to find a functional equation between the measurand or influence parameter and the result.

In the following example the relations are shown for the selection of particle mobility *Zp* in the differential mobility anlayzer (DMA). The distribution of particle mobility at the probe shall be called *N*(*Zp*). Following parameters determine the separation of particles in the DMA:

- In the probe the deposition *n(geometry, time)* of particles depends on the geometry of the probe and delay time:
$$N'(Zp) = n(geometry, time) \cdot N(Zp) \quad (1)$$
- The transformation in the probe can be selective due to the mobility or aerodynamic diameter of particles (coagulation, adsorption etc.):
$$N'(Zp) = \int n(Zp) \cdot N(Zp) \quad (2)$$
- The geometry of the DMA (r_1 = inner radius, r_2 = outer radius, L = length), the voltage on the central electrode *U* an the selected fluxes (Q_c = inlet sheath air, Q_m = outlet excess air, Q_a = inlet aerosol, Q_s = outlet monodisperse aerosol) determine the medium mobility of the selected particles.
$$Z_p = \frac{Q_c + Q_m}{4 \cdot \pi \cdot U \cdot L} \cdot \ln(r_2 / r_1) \quad (3)$$
and the width *DZ* of the separation.
$$\Delta Z_p = \frac{Q_a + Q_s}{2 \cdot \pi \cdot V \cdot L} \cdot \ln(r_2 / r_1) \quad (4)$$
- The gas temperature *T* and the pressure *p* of the air in the DMA influence the measurement and regulation of the fluxes:
$$Q_{eff} = Q_{cal} \cdot \frac{T \cdot p_{cal}}{T_{cal} \cdot p} \quad (5)$$
- The viscosity μ of the air is a function of the gas parameters and the chemical composition. It affects directly the mobility:
$$Zp_{eff} = Zp \cdot \frac{\mu_{eff}}{\mu} \quad (6)$$
- Using the SMPS (Scanning mobility particle sizer, TSI Incorporated) also the adjustments and algorithms in the software are essential. For example according the choice of the fluxes and the scan time the program proposes a delay time. The effective delay time deviates from this default value by 1,6 s, if the length of the tube between DMA and CPC differs by only 10 cm from the default value. In the mobility spectrum this deviation causes a shift of more than 4 %. Higher accuracy can be achieved, if instead of the automatic scan mode the voltages are scanned stepwise.
- The manufacturer indicates the accuracy of the CPC with 10 %. This value must be verified for each instrument.

4.2 Calibration and Estimation of Uncertainties

The modeling of the measurement leads to the relevant quantities and the magnitude of their influence. They allow the calculation [5)] of the uncertainty for the mobility, if the uncertainties of ***all*** quantities are known.

For DMA - but without the probe - an investigation of NIST [6] shows an estimation of the overall uncertainty of 3 % (table 1). It must be stressed, that the condition for this uncertainty is the calibration and adjustment of all components with the indicated uncertainty. Unfortunately this paper gives no indication on the parameters for the computation of the mobility spectrum.

In the summary of the influence parameters two type of parameters can be distinguished: The first type includes parameters, which are fixed at the fabrication of the instrument and do not change. These parameters like diameter or length of DMA, don't change by normal use and normal maintenance. They contribute almost 50 % to the overall uncertainty (according the paper). The second type covers parameters, which depend on the application and which may drift in time. The ambient conditions determine for example the temperature, the pressure but also the response time (due to tubing of the instrument). Drifting parameters are flow controllers, voltage etc.. These parameters must be periodically calibrated and adjusted. They contribute more than 50 % to the overall uncertainty.

Variable	Uncertainty in variable	Resulting uncertainty in diameter
Q_c = sheath air flowrate	1.0%	0.6%
Q_m = excess air flowrate	1.0%	0.6%
r_2 = outer radius	0.3%	0.26%
r_1 = inner radius	0.2%	0.16%
L = length	0.5%	0.30%
V = center rod voltage	0.45%	0.26%
e = elementary unit of charge	negligible	0.025%
μ = viscosity of air	0.04%	0.025%
C = slip correction	0.9%	0.5%
T = temperature	0.2%	0.01%
P = pressure	0.4%	0.16%
Worst case estimate from eqs (1) and (12)		±2.4%
Random error, R		±0.1%
Residual uncertainty associated with effect of aerosol flowrate on apparent size		±0.5%
Total uncertainty associated with classifier		±3.0%
Impurities related uncertainty		+0%/−0.3%
Total uncertainty—classifier + residue layer		+3.0%/−3.3%

Table 1: Summary of uncertainty calculation for DMA (Kinney et al., 1991)

Considering the uncertainties of the variables used for the evaluation of the mobility distribution (for SMPS e.g. delay time, scan time etc.), the uncertainties of particle counting (manufacturer indication: 10 %) [7] and of the transformation during sampling in the probe the overall uncertainty may rise to a multiple of the stated value.

4.3 Comparison between laboratories

Round-robin-tests are an important tool in metrology in order to prove the comparability. Comparisons intend to

- exchange experience
- recover systematic errors
- gain confidence in results of other laboratories
- improve competence of all participants

If a stable standard (e.g. stable gas mixture, standard of mass) exists, this standard can travel around and be measured by each laboratory. In the field of particles, the stability of the available combustion aerosol standards is not yet satisfying[8)].

5 Conclusions

- Particles can be described by different quantities.
- The goal of the measurement defines the quantity of interest.
- Legal application claims for traceability
- Traceability is based on
  - estimation of uncertainties
  - calibration of components
  - comparison between laboratories

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1) International vocabulary of basic and general terms in metrology (VIM). BIPM/IEC/OIML/IUPAC/ IUPAP, second edition (1993)

2) Heinz Burtscher (1992) Measurement and characteristics of combustion aerosols with special consideration of photoelectric charging an charging by flame ions, J*. Aerosol Sci*., Vol 23, No 6 pp 549-595

3) John McAughey (1999) Particle metrology and the assessment of health effects. In *Third international ETH-Workshop on Nanoparticle Measurement*, 9./10. August 1999.
Maria G. Costantini (1999) Particle Emissions Characterization and Health Effects Research. In *Third international ETH-Workshop on Nanoparticle Measurement*, 9./10. August 1999.

4) Hinds, W.C. (1982) Aerosol Technology, John Wiley, New York

5) Guide to the expression of uncertainty in measurement, ISO, 1993 (ISBN 92-67-10188-9)

6) Patrick D. Kinney, David Y.H. Pui, George W. Mulholland and Nelson P. Bryner (1991) Use of the Electrostatic Classification Method to Size 0.1 um SRM Particles - A Feasibility Study. *J. Res. Natl. Inst. Stand. Technol.* **96** (2) 147-176

7) B. Sachweh et al. (1990) Calibration of Optical Particle Counters: comparison between theoretically and experimentally derived results, *J. Aerosol Sci. Suppl. 1* **21**(1), S521-S525
H.Y. Wen and G. Kasper (1986) Counting efficiencies of six commercial particle counters. *J. Aerosol Sci.* **17**(6), 947-967
H. Bartz et al (1985) Response characteristics for four different condensation nucleus counters to particle in the 3-50 nm diameter range, *J. Arosol Sci.* **16**(5), 443-456

8) First experience with a new generator for a combustion aerosol standard (CAST) will be collected soon. See also contribution of Lianpeng Jing at this conference: Properties of soot particles produced by the soot generator CAST.