

Traceable size measurement of polystyrene particles for calibration of particle counters using SEM in transmission mode

I. Motivation

Vehicle emission standard Euro 6 and PTI

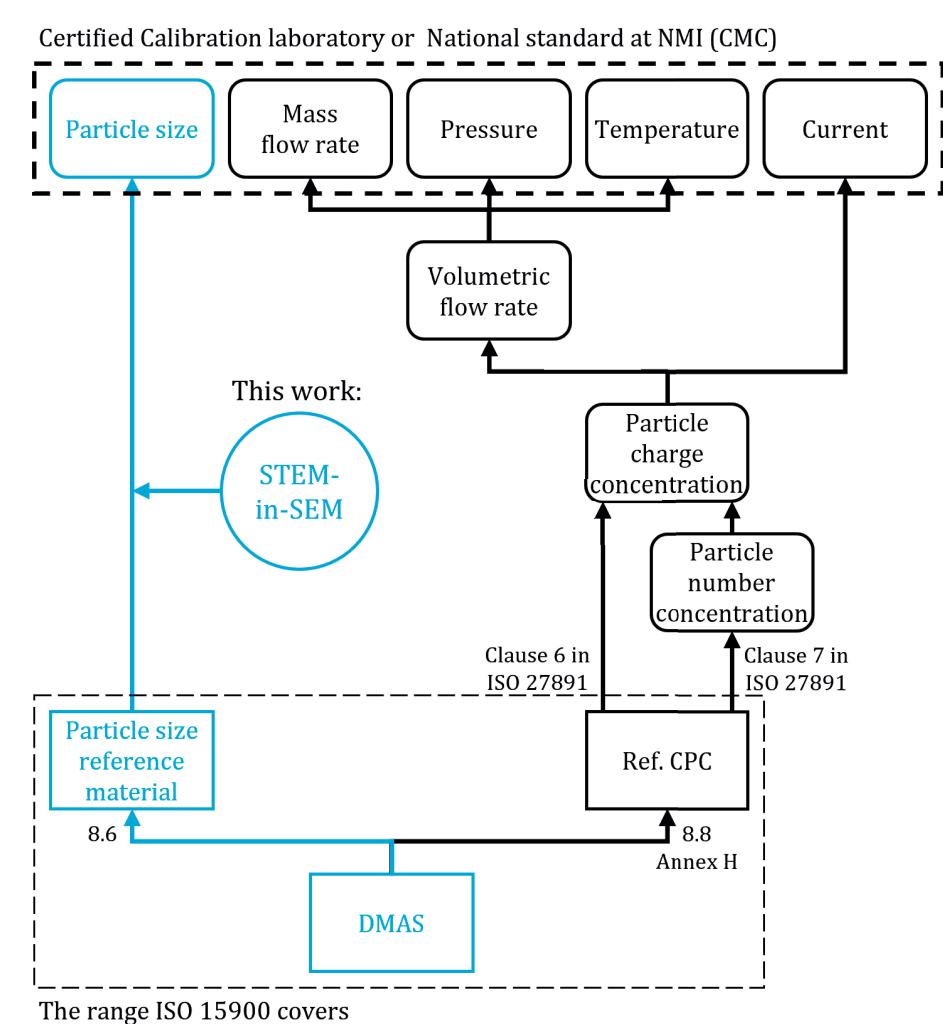
- Legal limits for the number of emitted particles per km
- Reference particle counters need to be calibrated
- These measurements must be traceable to SI units [1]

Need

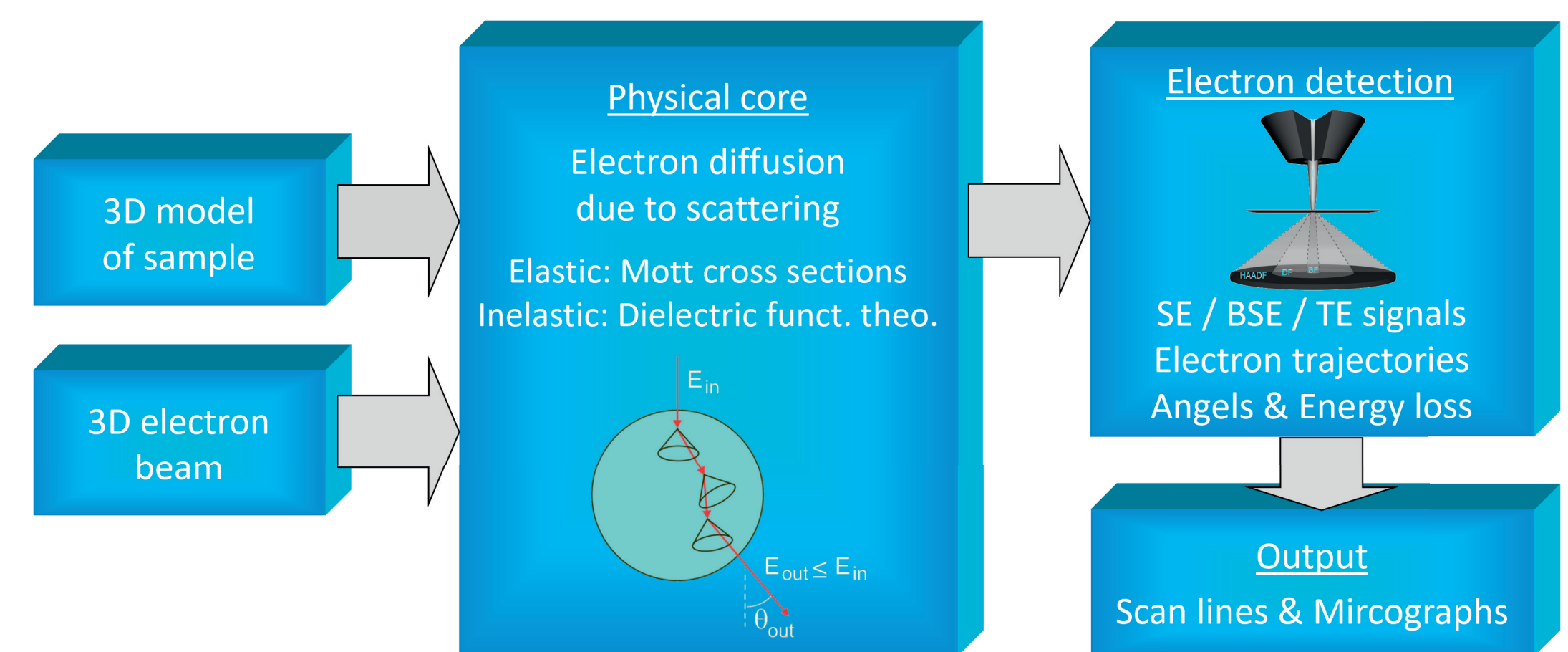
- Traceable determination of diameter of nanoparticles

Method

- STEM-in-SEM: Scanning electron microscopy in transmission mode
- Image analysis routine based on simulation of signals



IV. Monte-Carlo Simulation: Geant4SEM

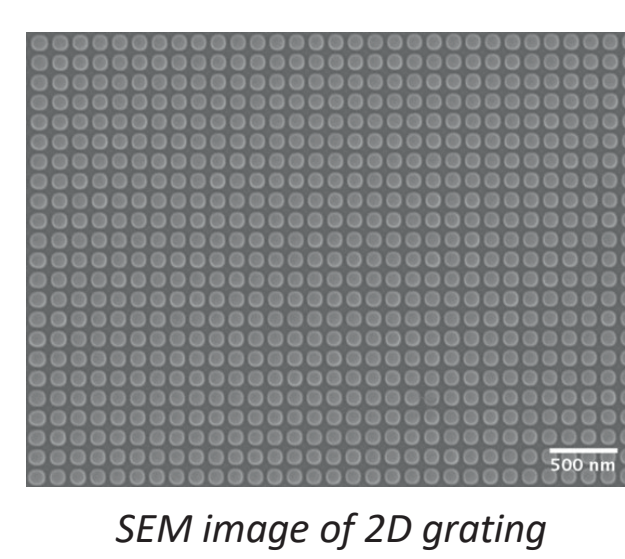


- Geant4SEM: Customized physics modules for powerful framework Geant4 [2]
- Elastic scattering: Mott cross sections calculated using ELSEPA [3] with muffin tin radii
- Inelastic scattering: Phenomenological model approximating the dielectric function

II. Traceability to SI unit meter

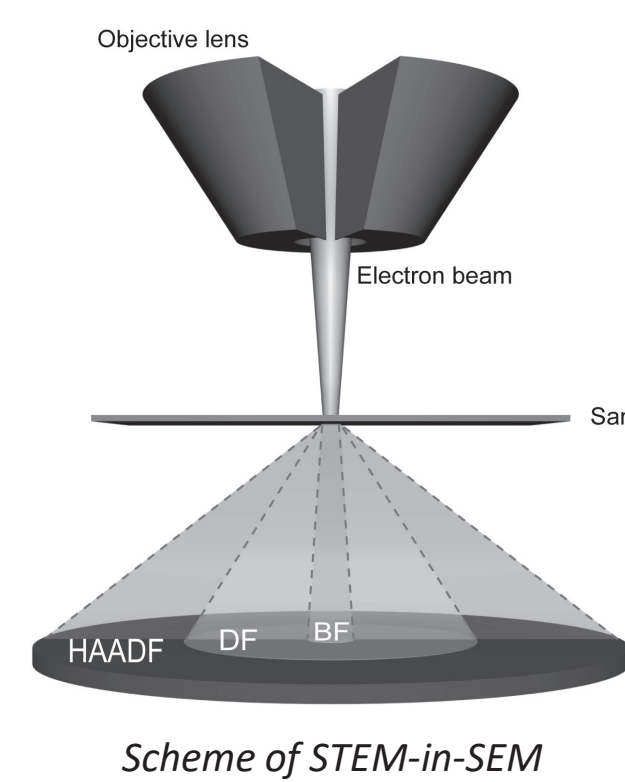
1. Traceable pitch of 2D grating

- HeNe laser as realization of the SI unit meter ...
- ... used in a diffractometer ...
- ... to determine the pitch a_{optical} of the 2D grating



2. Traceable pixel size of SEM

- Micrographs of 2D grating taken in SEM ...
- ... to determine the pitch a_{SEM} in pixel ...
- ... to calculate the pixel size as $p = a_{\text{optical}} / a_{\text{SEM}}$

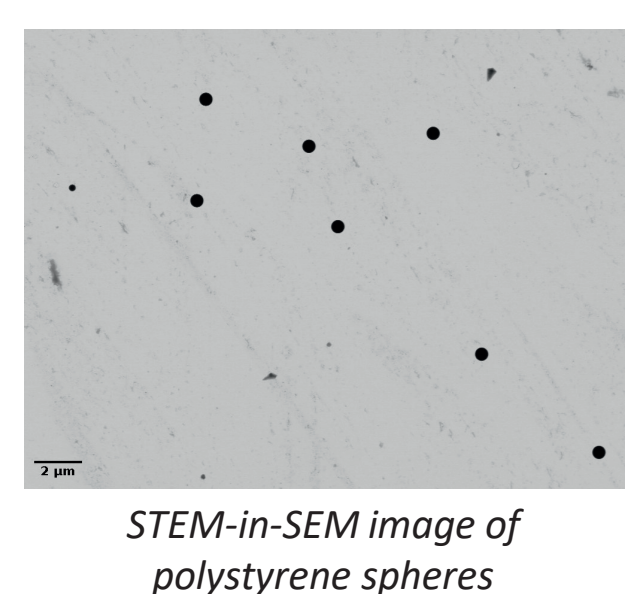


3. Traceable nanoparticle size

- Micrographs of nanoparticles using STEM-in-SEM
- Determination of preliminary size (see III)
- Adjustment of size based on simulation of STEM-in-SEM signal (see IV)

4. Traceable DMA measurements

- Using traceably measured nanoparticle sample ...
- ... together with traceable sheath air flow ...
- ... and traceable high voltage



III. Image analysis

Aim:

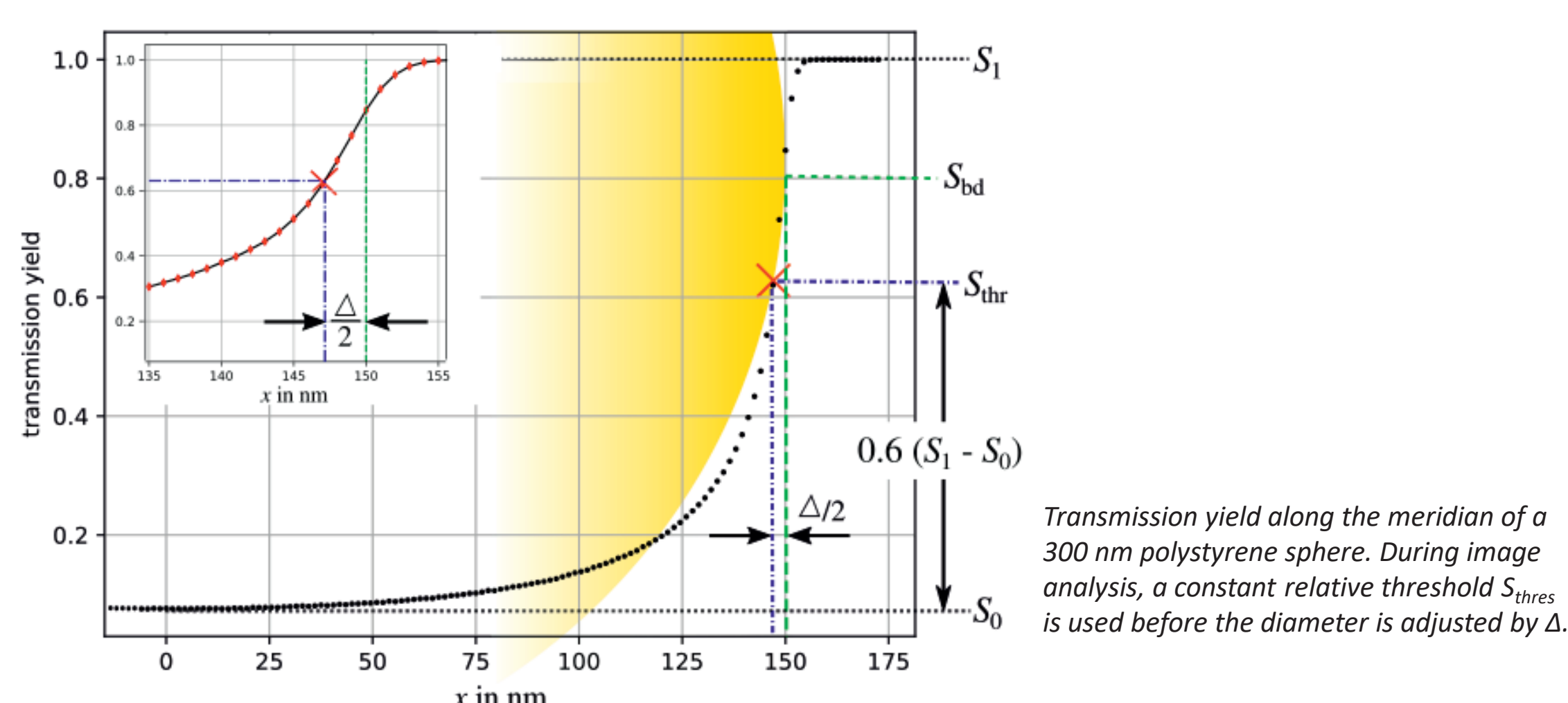
- Separation of background pixels from particle
- ... with sub-pixel accuracy

Obstacles:

- Size-dependent threshold
- Non-linear signal profile at particle boundary

Two-step approach:

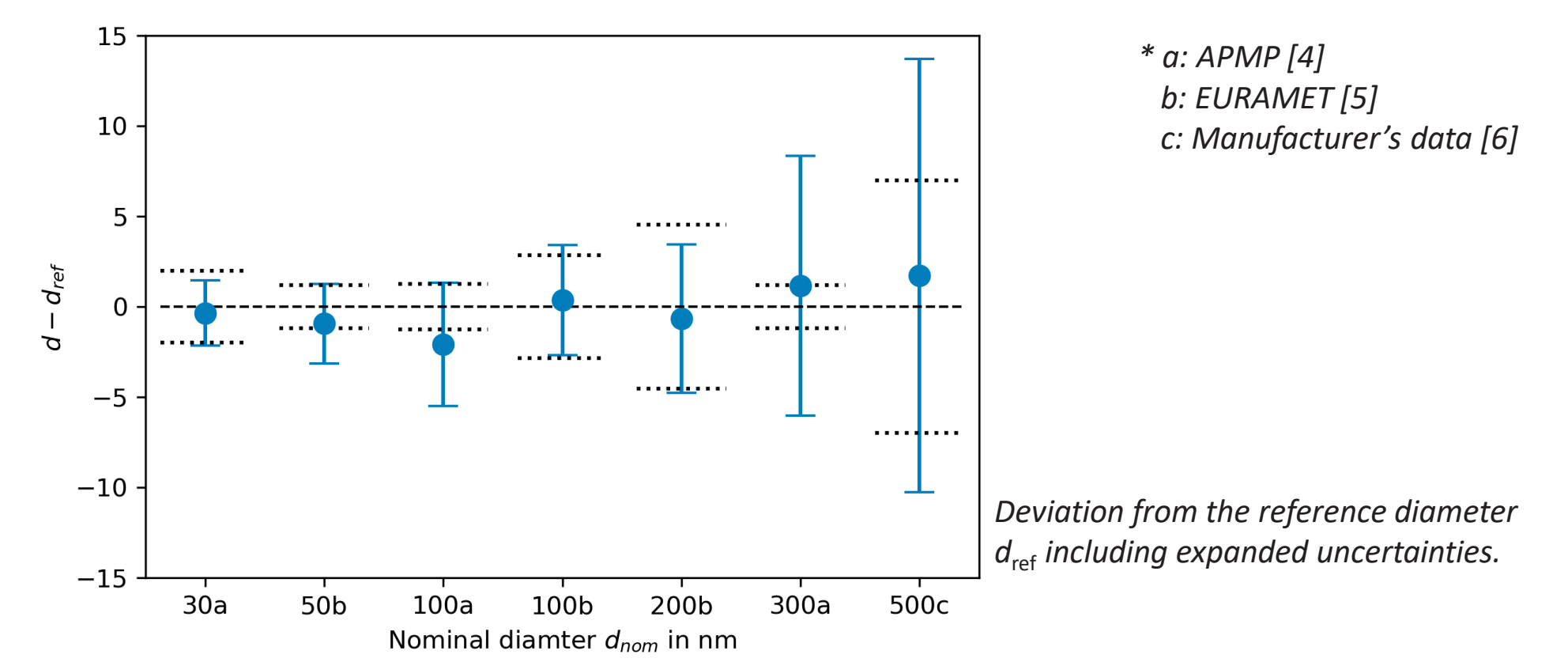
- Preliminary size at fixed threshold at linear region of signal
- Adjustment of size based on simulation results



Transmission yield along the meridian of a 300 nm polystyrene sphere. During image analysis, a constant relative threshold S_{thr} is used before the diameter is adjusted by Δ .

V. Results and intercomparisons

- We participated in two international intercomparisons*
- Our results are in blue, including expanded uncertainties
- Black dotted lines: Expanded reference uncertainties...
 - ... calculated from results of all participants



* a: APMP [4]
b: EURAMET [5]
c: Manufacturer's data [6]

Deviation from the reference diameter d_{ref} including expanded uncertainties.

VI. Conclusion

- Very good agreement over the entire size range: 30 nm to 500 nm
- Measurement uncertainty contains size-dependent effects
- Expanded uncertainty in the order of the pixel size

Literature

- This work: D. Hüser et al., Meas. Sci. Technol. **23**, 085016 (2023)
- ISO 15900:2020
 - S Agostinelli et al., Nucl. Instrum. Methods Phys. Res. **506**, 250 (2003)
 - F. Salvat et al., Comput. Phys. Commun. **165**, 157 (2005)
 - H.-L. Lin et al., Metrologia **56**, 04004 (2019)
 - F. Meli et al., Meas. Sci. Technol. **23**, 125005 (2012)
 - Data sheet: Nanosphere Size Standards, Thermo Fisher Sc., Fremont, CA, USA

Summary

Introduction: Nanoparticle emissions from cars are of considerable importance for European emission regulations such as EU6 and PTI. These nanoparticles are counted by different types of particle number instruments. Therefore, reference particle counters need to be calibrated by national authorities. These measurements must be traceable to SI units [1], including the traceable determination of geometric diameter for monodisperse nanoparticle samples. This is achieved by imaging the particles using a scanning electron microscope in transmission mode (STEM-in-SEM), followed by an image analysis routine that is based on the results of Monte Carlo simulations of the STEM-in-SEM signals.

Image analysis: For size determination, the image pixels belonging to a particle must be separated from the background pixels. To achieve highly accurate measurements, the signal profiles are linearly interpolated and an individual threshold dependent on the particle size is used. For relatively large polystyrene particles, the threshold at the particle boundary lies in the outer, non-linear part of the signal profile. Therefore, a two-step approach has been chosen. First, a preliminary size value is determined in the linear region of the signal profile at 60%. Subsequently, the final size is obtained by adding a tabulated value determined by simulations.

Monte Carlo simulation: To simulate the imaging process we developed Geant4SEM by adding customized physics modules for single electron scattering processes to the powerful Monte Carlo framework Geant4 [2].

Results & Conclusion: We measured polystyrene particles with nominal sizes ranging from 30 nm up to 500 nm in comparison to the reference values of two international intercomparisons [4-6]. The results show very good agreement over the entire size range.