Optimization of silver particle number size distributions from a homogenous nucleation furnace

- Needs for EECPC calibration
- Criteria of calibration aerosol
- Nucleation furnaces
- Results

WG 3.23: Aerosol and Particle Diagnostics

Andreas Nowak, Johannes Rosahl, Detlef Bergmann
Arne Kuntze, Margit Hildebrandt, Egbert Buhr, Volker Ebert

EECPC calibration needs

- Manufacture independent EECPC calibration
  → Preferably with traceable calibration chain
- Primary and secondary PN standard
- A suitable aerosol EECPC calibration
  → Comparable to engine exhaust
  → Metrological Uncertainties
- Previous work@PTB: EMRP-project, ENV02, PartEmission, 2011-2014)

  Goals: → Determination and Validation of primary and secondary method for PN
  → Development of suitable calibration aerosol/setup

ENV02-Project Results

- Aerosol criteria:
  - highly monodisperse
  - single charge
  - spherical morphology
  - tunable diameter size between 10 to 100 nm
  - sufficient particle number concentration, up to 10^5 cm^{-3}
  - highly thermal stability, up to 350 °C

- Criteria according to ISO 27891
- Promising aerosol candidate: silver nanoparticles

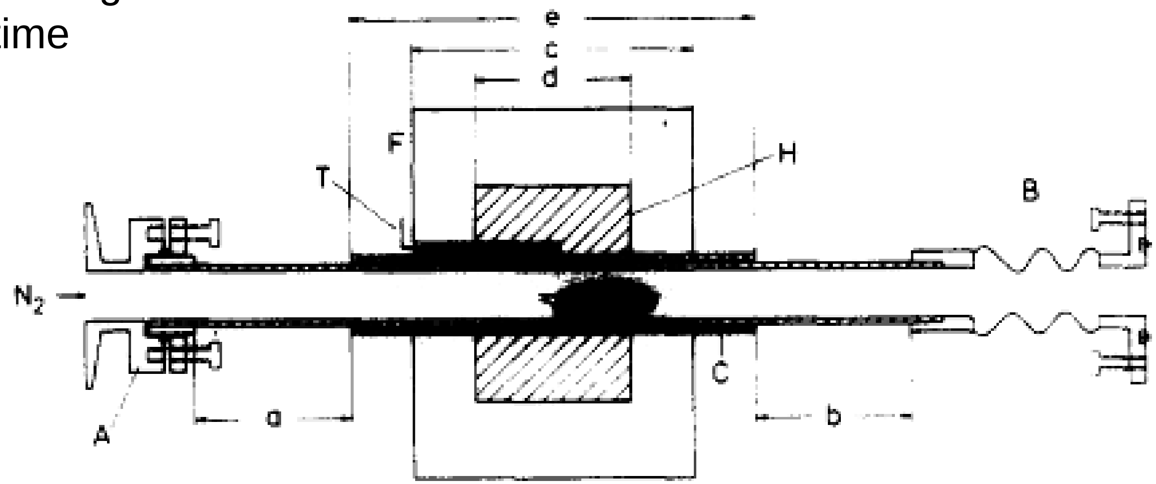
→ Homogenous nucleation process

Nucleation furnace: History

- simple tube furnace to generate silver particles (Scheibel and Porstendörfer*)

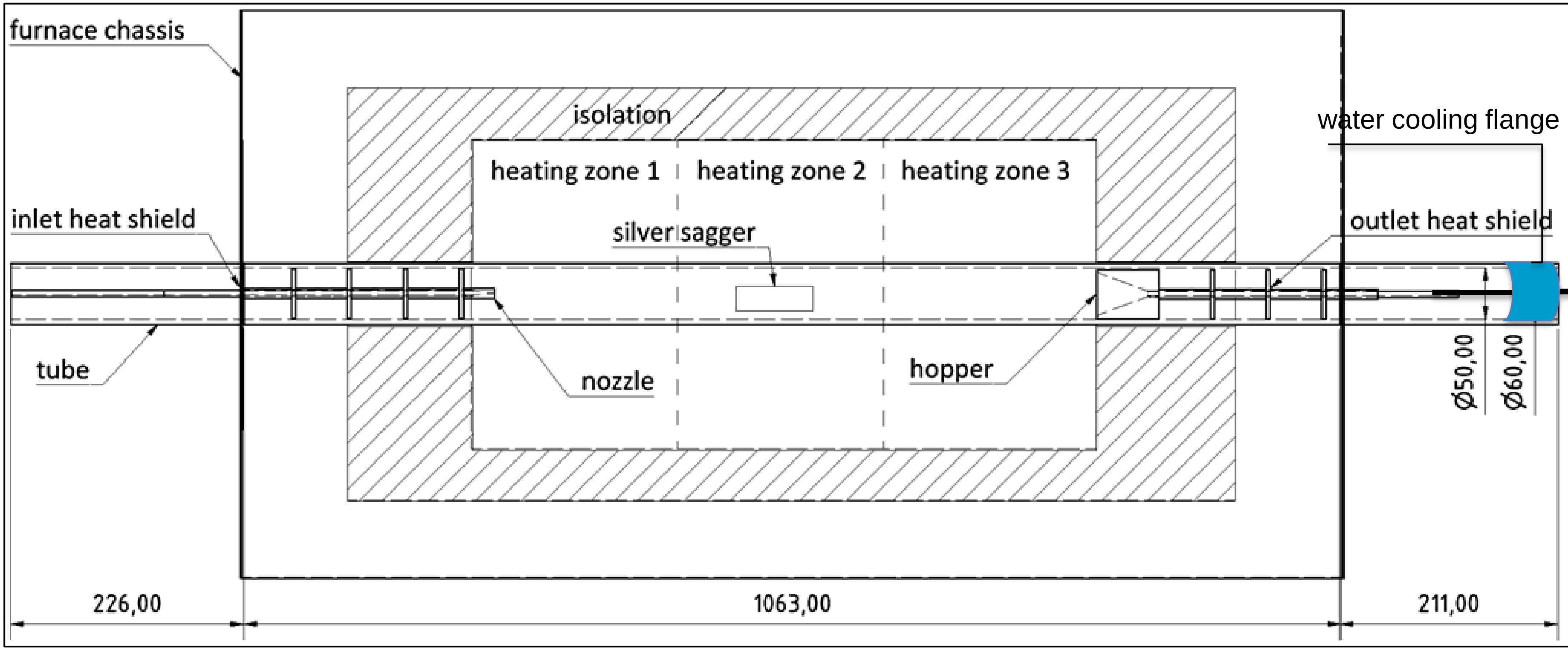
***Journal of Aerosol Science, Volume 14, Issue 2, Pages 113-126 (1983)**

– only one heating zone (max. 1300°C)
– short high temperature region
→short residence time



- Findings:
  - condensation dominates nucleation process in colder part of furnace
  - particles < 50 nm with high concentration (10^7), size down to 2 nm
  - monomodal size distributions with narrow distributions (σ~1.3-2.0)
  - particles above 30 nm: have tendency to be non-spherical particle

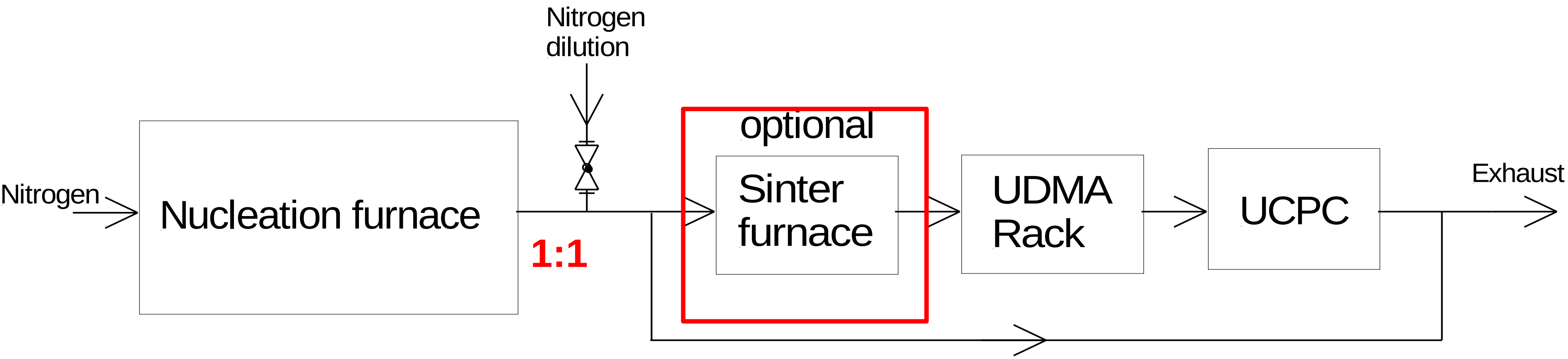
Nucleation furnace: Optimization at PTB



Goals:

- larger particles → increase of residence time → larger furnace with 3 heating zones
- optimization flow scheme and T gradient → implementation of heating shields:
  - – inlet shield with nozzle (different sizes)
  - – outlet shield with hoper
- minimization of conglomerates → water cooling flange at end of tube → shock cooling

PNSD measurements: Experimental setup

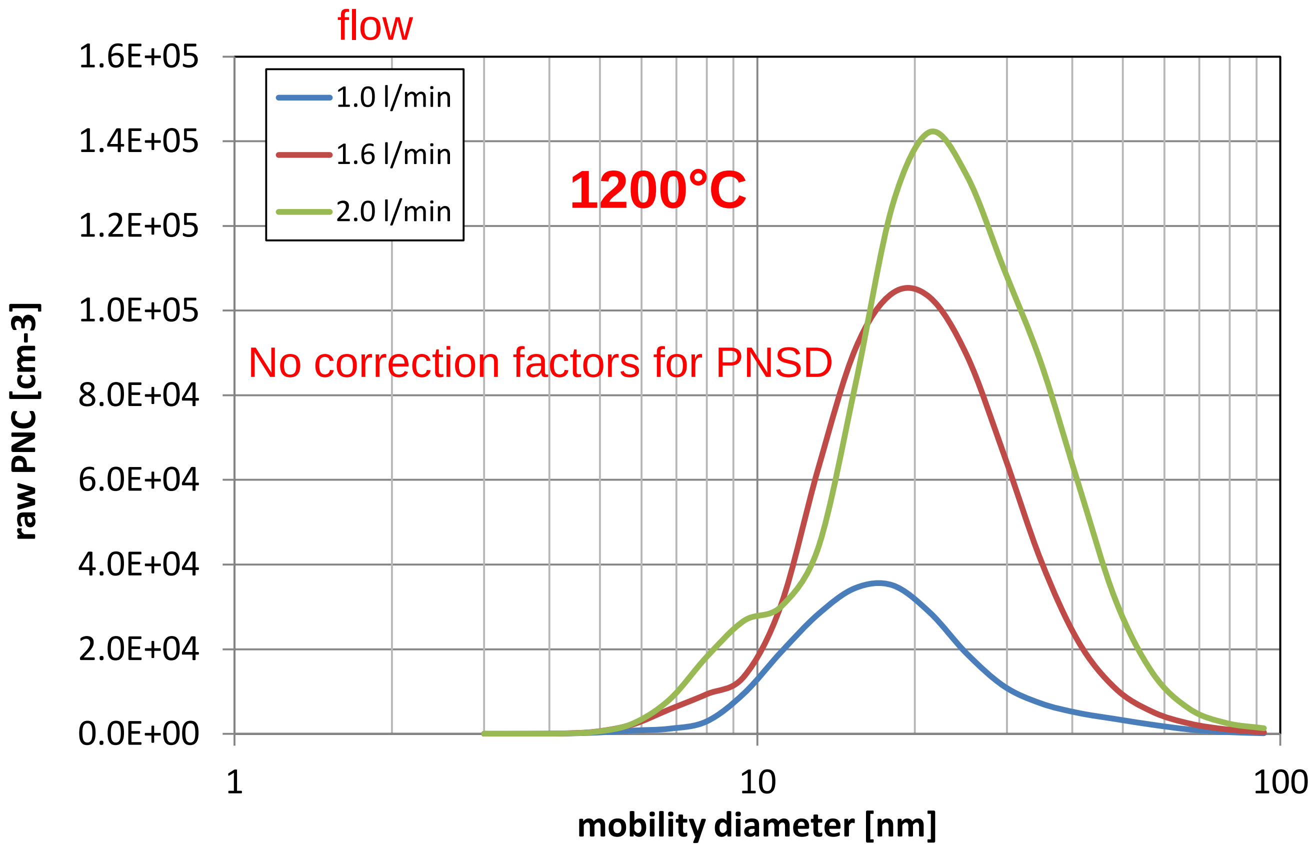


- Carrier flow: 0.8 – 2.0 l/min (N_2)
- Dilution flow: 0.8 – 2.0 l/min (N_2)
- UDMA (hauke short) flow: 1.5 : 15 l/min
- Scan range: 3 – 93 nm, stepwise
- UCPC (TSI 3776) at high flow mode

→ Tests for different flow ratios and configurations

PNSD results I: Flow effects

- Constant nozzle (1mm) and same silver loading at one temperature

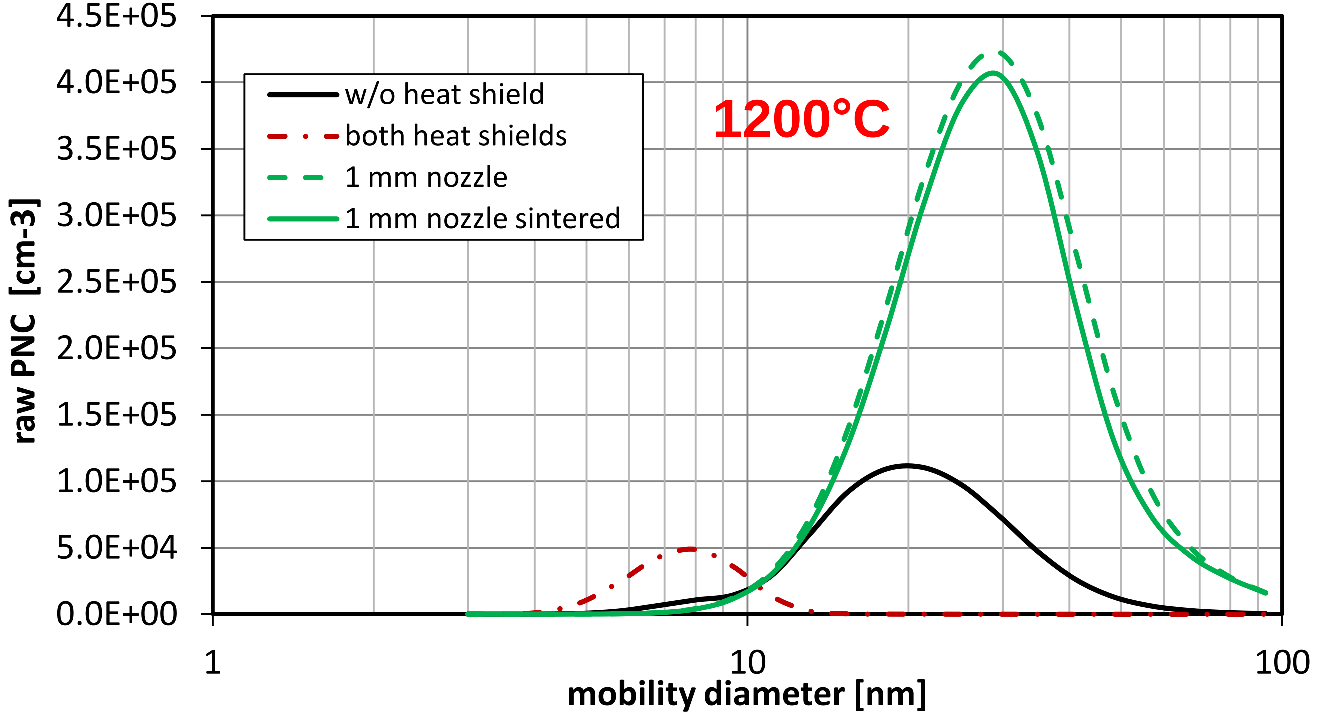


→Increasing PNC with higher flows → Indication for an optimal flow scheme

→ monomodal size distribution @ 1.6 l/min

PNSD results II: Different shield configurations

- Constant nozzle (1mm) at one temperature and one flow ratio for carrier and dilution flow (1:1)



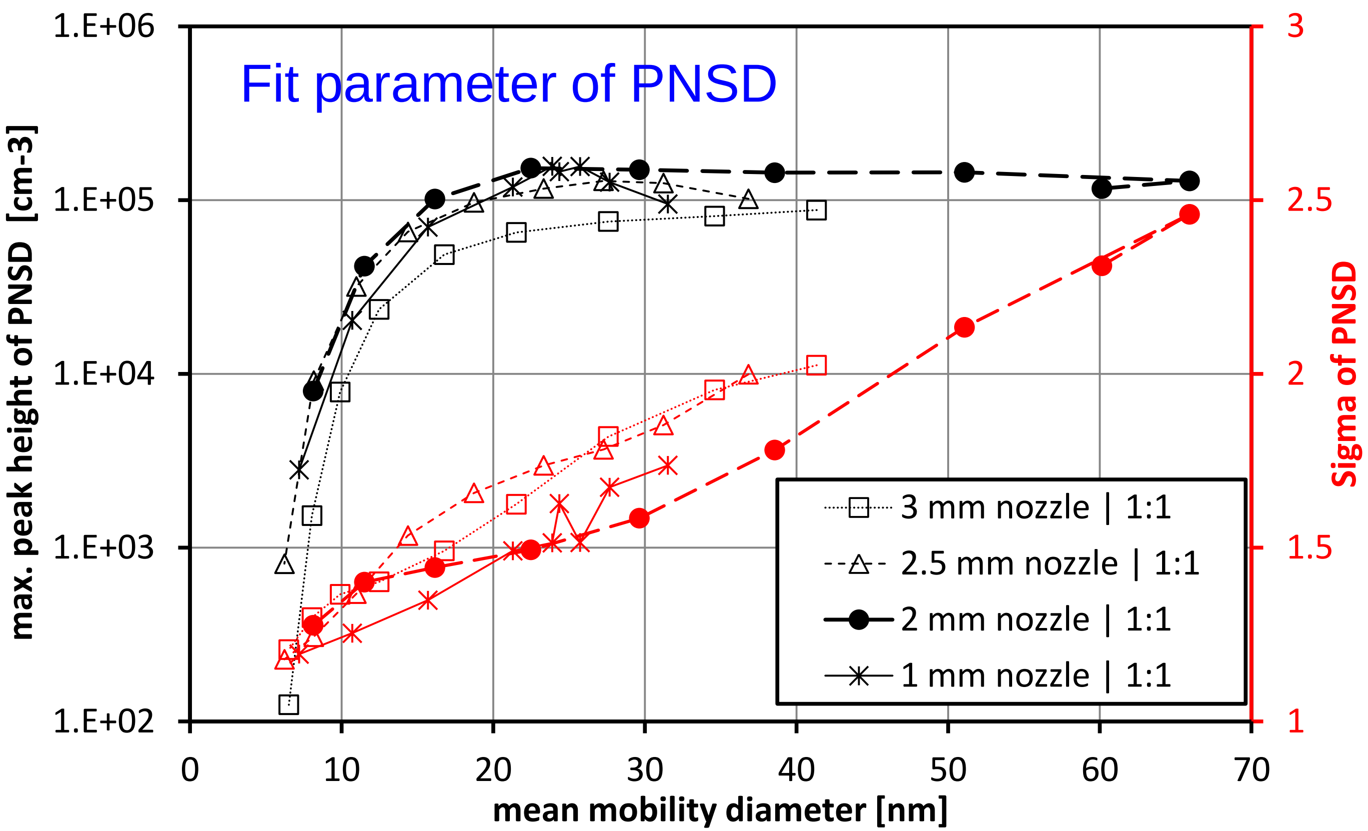
- w/o heat shield → less PNC
- Both heat shields → smaller silver particles, sharp peak in lower size range
- Only inlet heat shield with nozzle → higher PNC

➢ Sintering up to 650 °C → no influence for PNSD

➢ For further studies → only inlet heating shield with different nozzle sizes

PNSD results: Nozzle sizes

- For different nozzle sizes at one flow ratio (1:1) from 960 °C up to 1300 °C



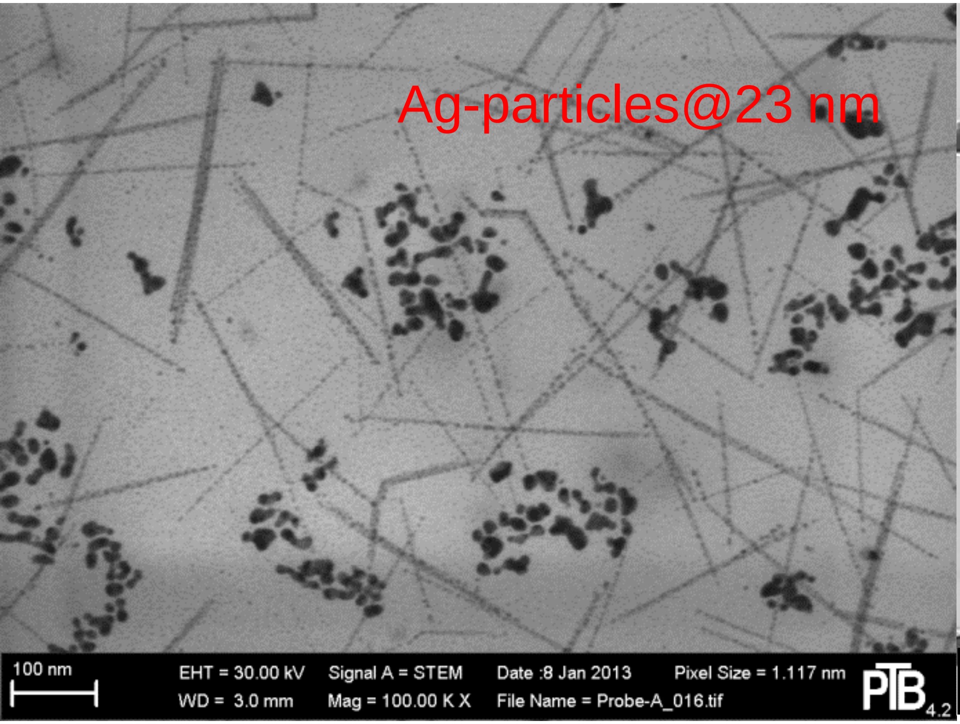
- High PNC (> 10^5 cm^{-3}) above 15 nm
- 2 mm nozzle: mean mobility diameter up to 65 nm, but broad size distribution

➢ 2 mm nozzle well suited for further experiments

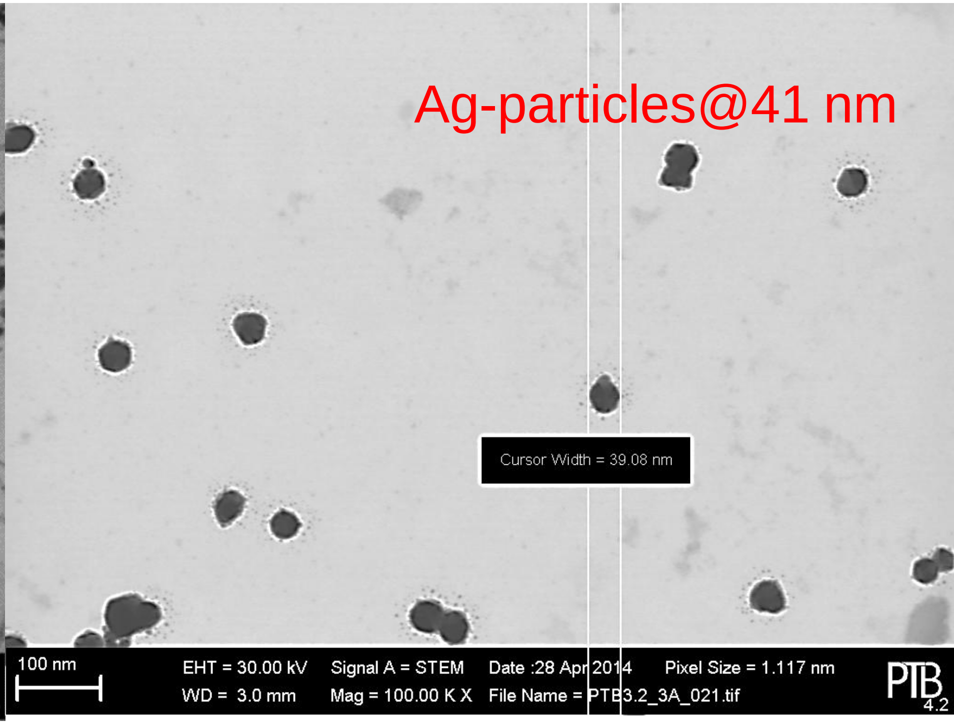
Results IV: Particle Shape

- STEM pictures for different types of tube furnaces

Old furnace construction comparable to Scheibel and Porstendörfer



New tube furnace at PTB



➢ Without sintering mostly spherical particles

Conclusion

- Optimization of tube furnace and homogenous nucleation through heath shields, different nozzles and flow control

  → larger particle sizes and suitable PNC for Ag-NP

- Only spherical particle were generated

  → Without required sintering for Ag-NP

- For ISO 27891

  → Entire particle size range

  → Sufficient PNC

Outlook

- Further increasing of size range to larger sizes above 100 nm

  → Heterogeneous silver nucleation with two furnaces

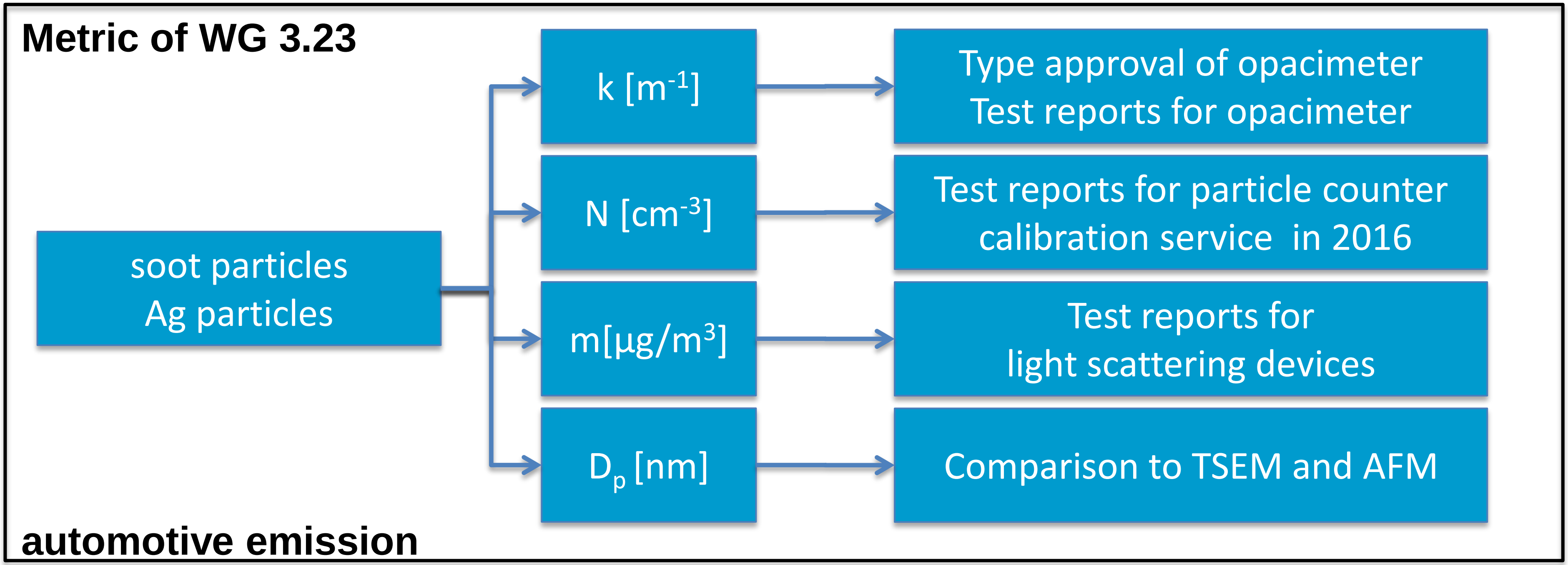
- Minimizing charge correction factors above 60 nm

  → 2^{nd} UDMPS with unipolar charging

  → Increasing of PNC for monodisperse fraction

➔ Communicate results (ENV02) into the legislative community (PMP/ISO)

➔ Establish EECPC calibration service at PTB in 2016



Thank you very much for your attention!

→ Questions?

Physikalisch-Technische Bundesanstalt
Braunschweig and Berlin
Bundesallee 100
38116 Braunschweig

Dr. Andreas Nowak
Email: andreas.nowak@ptb.de
www.ptb.de