Combination of Optical and Mobility based Number Size Distribution Measurements of Combustion Generated Particles

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Aerosol particles in smoke, produced by combustion processes, consist of black carbon, organic carbon and some inorganic material as well. These particles are often dominated by black carbon, which forms usually non-spherical, fractal-like particles through aggregation. Fractal aggregates are scale invariant, which means that within limits they appear the same when viewed over a range of scales.

The size is probably the most fundamental parameter describing an aerosol particle, it influences among others its scattering and absorption coefficient, its ability to take up water or activate as a cloud droplet. Due to this reason one of the most frequently performed aerosol measurement is the number size distribution measurement. This can be done by many different methods such as e.g. optical sizing, electrical mobility differentiation, microscopy, aerodynamic sizing; these different methods have usually different time resolution, are based on different type of diameter measurements and cover different size ranges.

If number size distribution measurement over a bigger diameter range is required, often combined measurement techniques have to be applied. Combining two size distributions based on different types of diameter measurement for non-spherical particles is a challenging task. Here, we present size distribution measurements of different combustion aerosol particles done by a scanning mobility particle sizer (SMPS, TSI) and a white-light optical particle counter (WELAS digital 2000, Palas). We have performed the following types of combustion experiments: flaming and smoldering wood burning, smoldering cotton wick, flaming heptane/toluene mixture, flaming decaline and flaming polyurethane.

The SMPS measured the number size distribution as function of the electrical mobility diameter in the range of 16-570 nm, one scan lasted 70 seconds whereas the WELAS measured the same as function of the optical diameter in the range of 150nm-10µm with 1 second time resolution. For fast changing aerosols the 70 seconds duration might be too long to obtain a correct SMPS size distribution, therefore we used an independent measure of the number concentration with sufficient time resolution, the WELAS integrated number concentration, to correct the measured SMPS number size distribution compared to a reference time point which was chosen to be the middle time point of the scan. It was assumed that during the 70 seconds the shape of the aerosol number size distribution remains unchanged.

The correction for the multiple charged particles in the SMPS is done by the estimation and subtraction of the multiple charged particles at every diameter bin from the highest towards the lowest diameters. For some fire types, due to the high modal diameter of the aerosol significant part of the size distribution is over the upper limit (570 nm, in our case) of the SMPS diameter range and therefore not captured. In these cases the multiple charge correction fails (see Figure 1.). To overcome this problem the single charge corrected number size distribution was fitted with a lognormal distribution and the fit was used to reconstruct its tail. This reconstructed, single charge corrected number size distribution was then used to recalculate the number of multiply charged particles and do the multiple charge correction (green line in Figure 1).

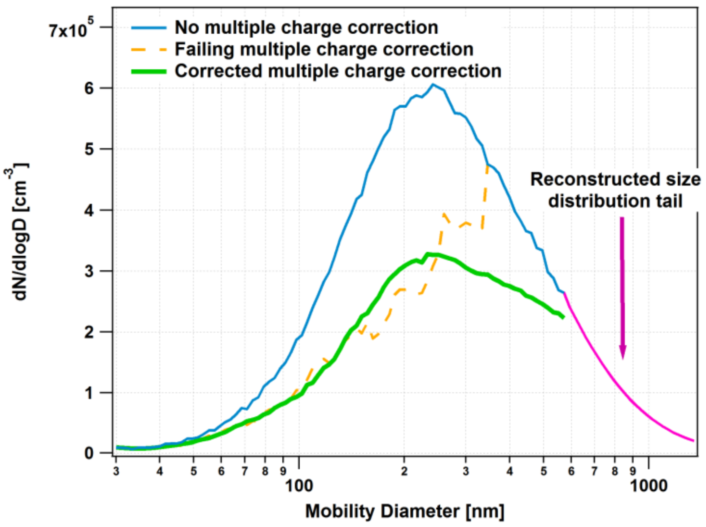

Figure 1. An example number size distribution of flaming decaline aerosol particles showing the original multiple charge corrected (dashed yellow line) and the modified multiple charge corrected (green line) number size distribution. For the modified multiple charge correction the reconstructed size distribution tail (violet line) was used.

The electrical mobility diameter and the optical diameter can differ substantially due to the non-sphericity and unknown refractive index of the particles. To combine the two size distribution measurements of different kinds, the mobility- optical diameter relation has to be known or guessed. We used the overlapping diameter range to empirically transform the WELAS optical diameter data into mobility diameter. It was assumed that a 2nd degree polynomial can describe the transformation between the mobility- and the optical diameter and the polynomial factors were searched by minimizing the deviation between the two different size distributions at the overlapping diameter range. Figure 2 represents an example for a smoldering cotton aerosol experiment.

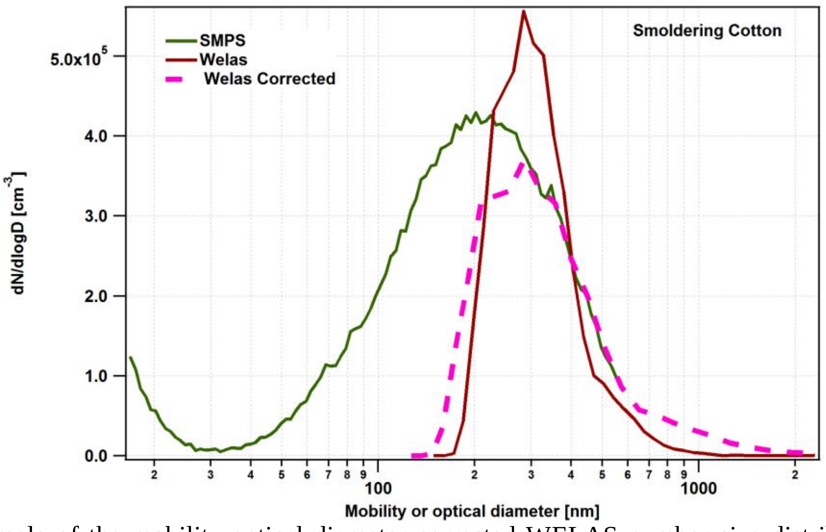

Figure 2. An example of the mobility-optical diameter corrected WELAS number size distribution scan during a smoldering cotton combustion experiment

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Introduction

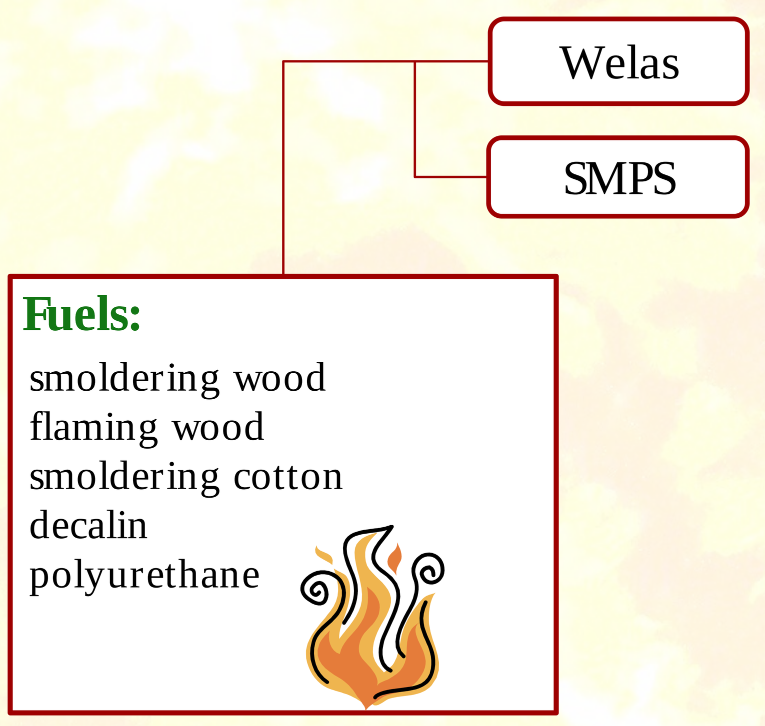

Figure 1 Schematic view of the measurement setup

Size distribution measurement

Welas: white-light optical particle counter
optical diameter: 200-10000 nm
time resolution: 1 sec
SMPS: scanning mobility particle sizer
mobility diameter: 16-750 nm
time resolution: ~70 sec

Problems to Solve

1 fast changing aerosol: skewed SMPS size distribution

2. no impactor before the SMPS: failing multiple charge correction for size distributions with high modal diameter

3. not spherical particles: no agreement between the optical and mobility diameter

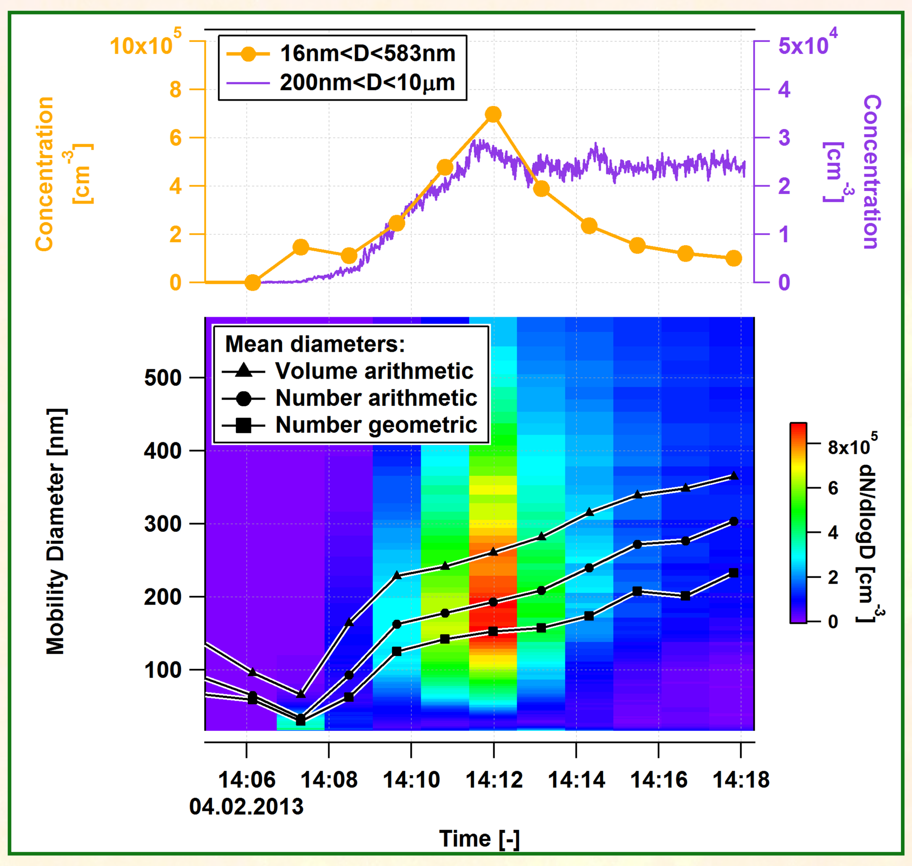

Figure 2. The time evolution of a flaming wood combustion experiment

Good to know

number size distribution

$$n(t) = \frac{dN(t)}{d\log D}$$

number concentration

$$N(t) = \int_{D_{min}}^{D_{max}} n(t)\cdot d\log D$$

1 Changing concentration during one SMPS scan

One SMPS scan lasted ~70 seconds and due to the fast changing aerosol it cannot be assumed that the aerosol number concentration stays stable during this time period.

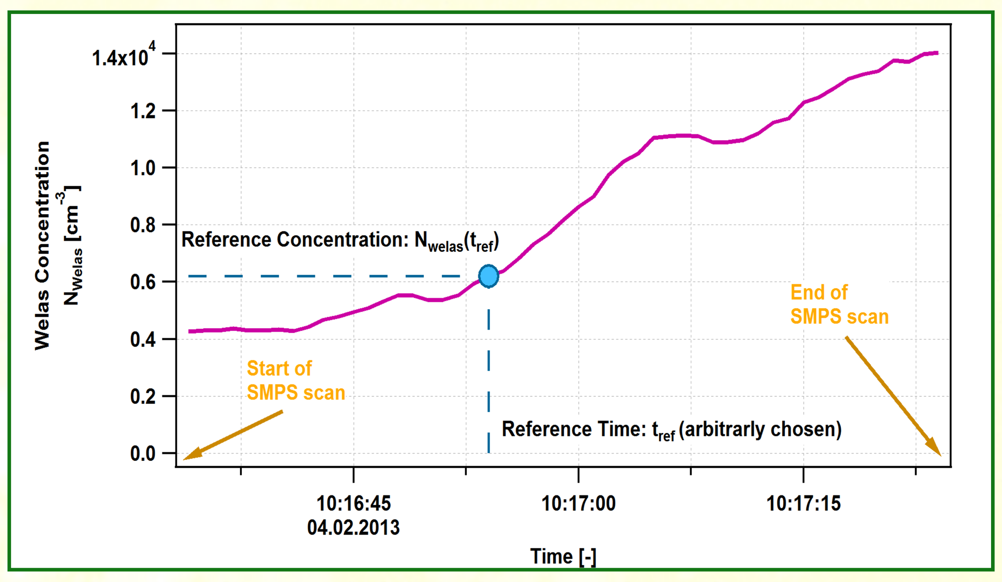

Figure 3. Evolution of the Welas aerosol number concentration (D>~200nm) with 1 second time resolution for a flaming wood experiment

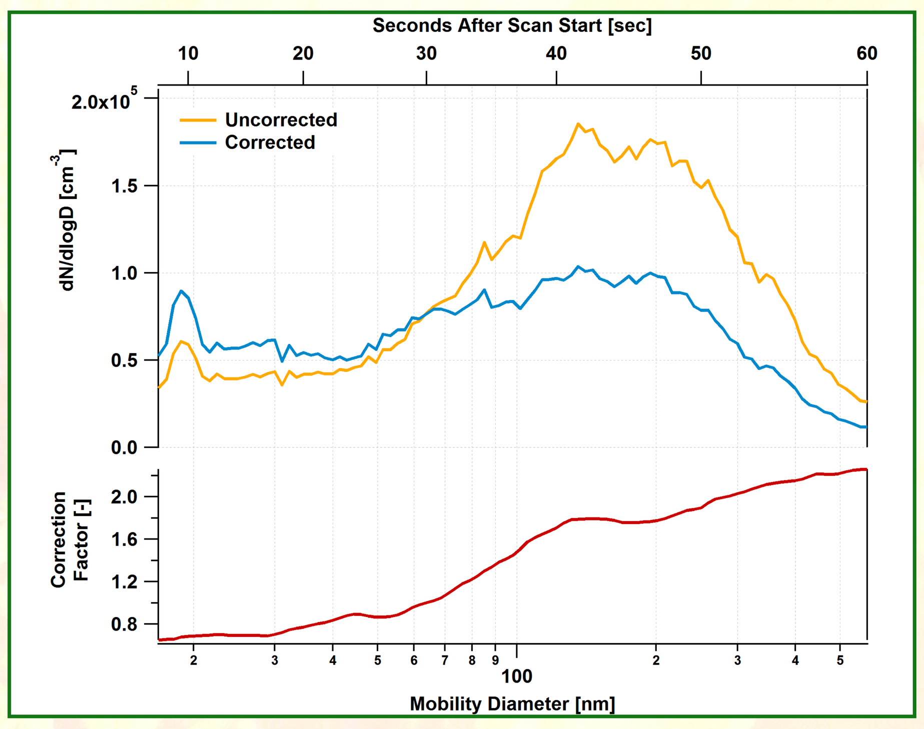

Figure 4. Process of the correction due to the fast concentration change for the same experiment and time interval as on Figure 3.

Needed for this correction: a measure which is proportional to the aerosol number concentration with good time resolution

We used the 1 second Welas number concentration:

$$N_{welas}(t) = N_{D>\sim 200nm}$$

Assumption: the size distribution shape is not changing significantly during one SMPS scan

Correction factor:

$$C_{corr}(D) = \frac{N_{welas}(D(t))}{N_{welas}(D(t_{ref}))}$$

Corrected number size distribution:

$$n_{corr}(D) = \frac{n_{uncorr}(D)}{C_{corr}(D)}$$

2. Manual multiple charge correction

For the experiments with high modal mobility diameter the size range of the SMPS does not cover a significant part of the size distribution. Here, since the particle concentration is not known, the concentration of the multiply charged particles (which appear at smaller sizes) cannot be calculated either.

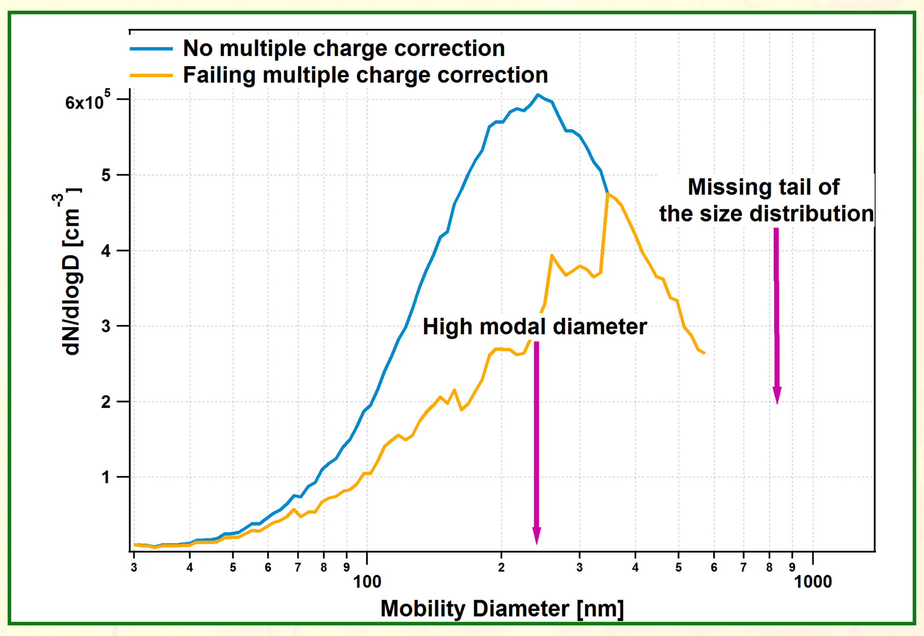

Figure 5. One example of a flaming decaline aerosol number size distribution with the default multiple charge correction by the TSI AIM software

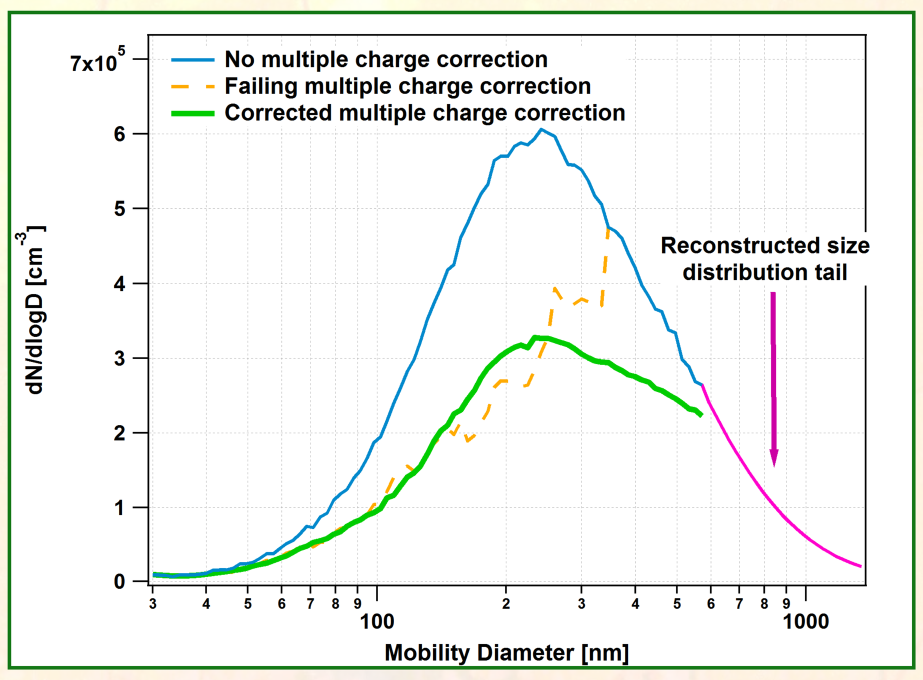

Figure 6. The same example number size distribution as on Figure 5 showing the multiple charge correction with the reconstructed size distribution tail

Solution: the tail of the size distribution is reconstructed before the multiple charge correction

How: fitting a lognormal distribution to the single charge corrected (SCC) size distribution (blue lines in Fig. 5 and 6) and extrapolate SCC size distribution with the fit

The multiple charge correction: perform the multiple charge correction (custom-written) on the extrapolated SCC size distribution

3. Optical and mobility diameter matching

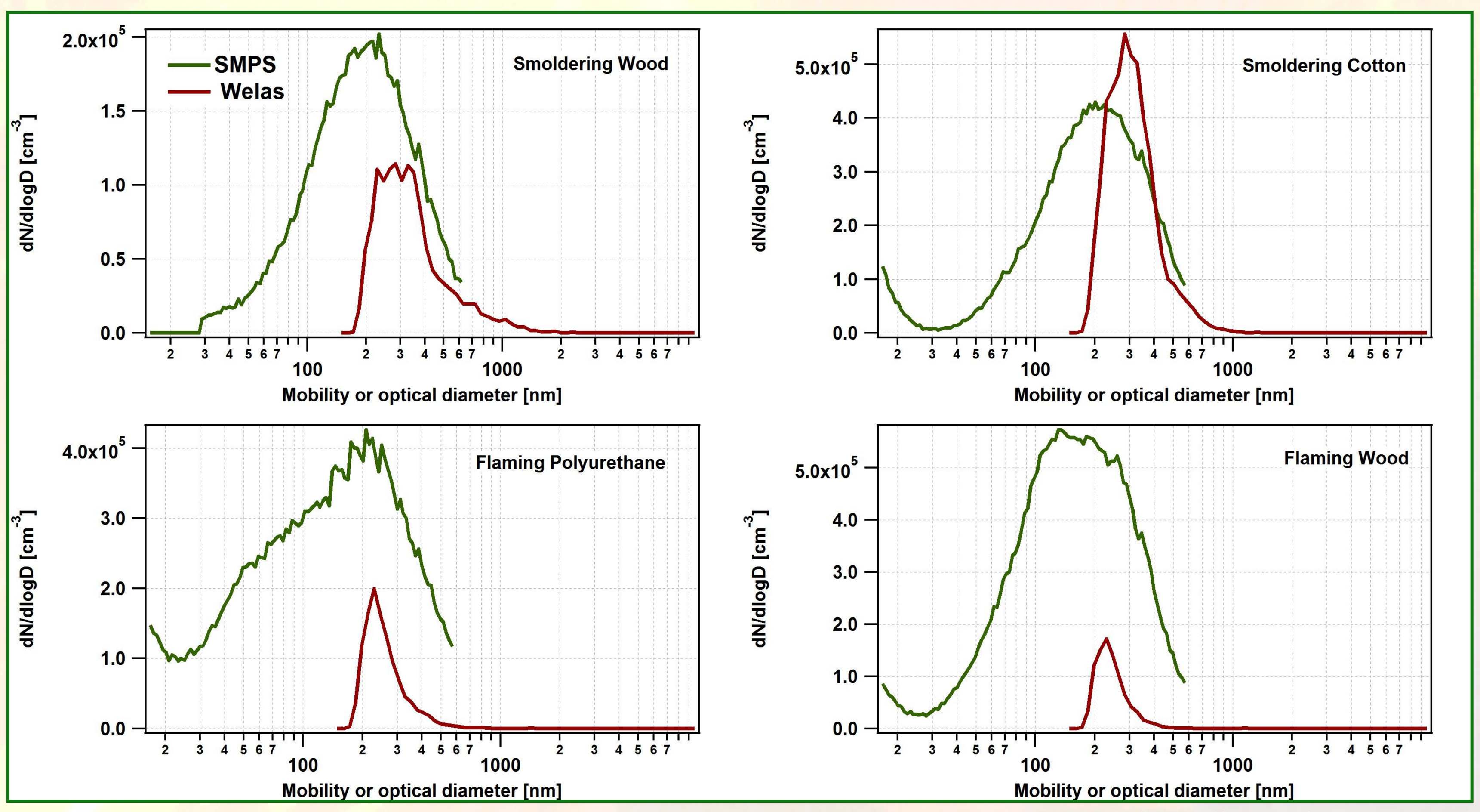

Figure 7. Mobility (SMPS) and optical (Welas) diameter based number size distributions for different fire types

Optical and mobility diameter: agreeing if the particles are spherical and their refractive index agrees with the calibration particles used for the Welas size calibration (usually PSL)

Combustion particles: usually fractal like shape and unknown refractive index

Empirical correction:

$$D_{mob} = f(D_{opt})$$

assuming that $f(D)$ can be expressed as a polynomial function

Determination of the polinomial factors using the overlapping diameter range

Using a 2nd degree polynomial:

$$D_{mob} = A_0 + A_1 D_{opt} + A_2 D_{opt}^2$$

The mobility based Welas size distribution:

$$n_{WELAS}(D_{mob}) = \frac{dN_{Welas}(f(D_{opt}))}{d\log(f(D_{opt}))}$$

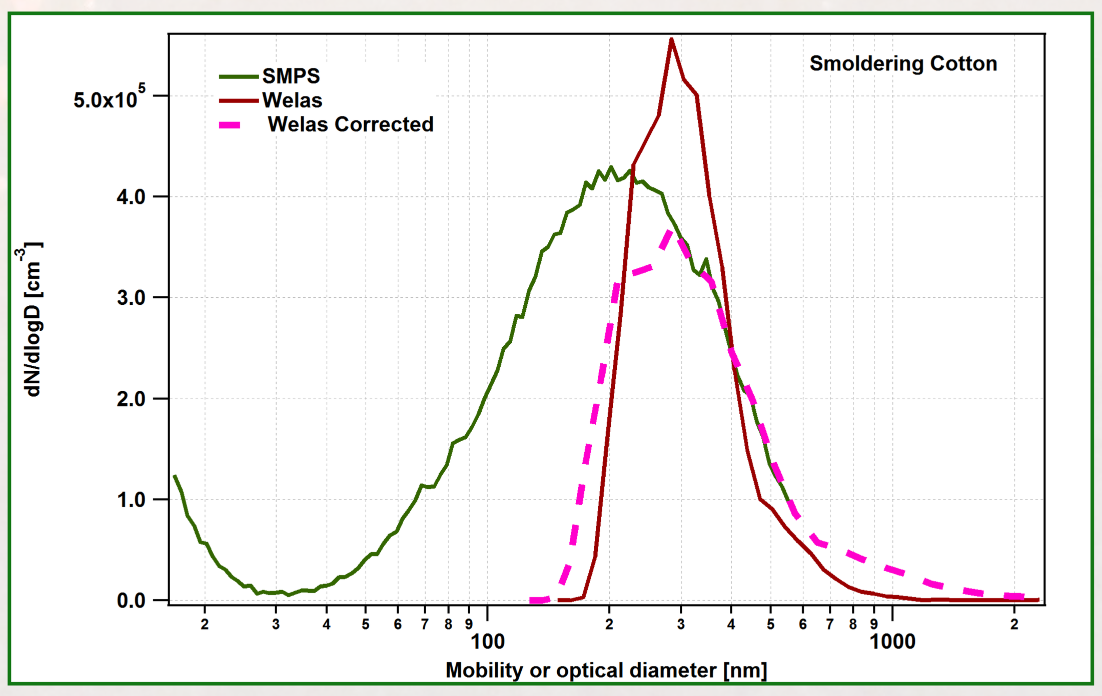

Figure 8. A mobility-optical diameter corrected Welas number size distribution scan during a smoldering cotton combustion experiment

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