Deliverable D3.19: Quality assurance procedures and uncertainties for measurements of coarse mode particle number size distributions using aerodynamic and optical particle size spectrometers in long-term observation programmes

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Comments	This deliverable focuses mainly on quality assurance for concentration and the various options to determined detection efficiency in the sub and lower super micron range.

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Quality assurance procedures and uncertainties for measurements of coarse mode particle number size distributions using aerodynamic and optical particle size spectrometers in long-term observation programs

As stated in D27, during the voluntary ACTRIS intercomparison workshop for aerodynamic and optical particle size spectrometers, a mismatch between the different instruments was determined.

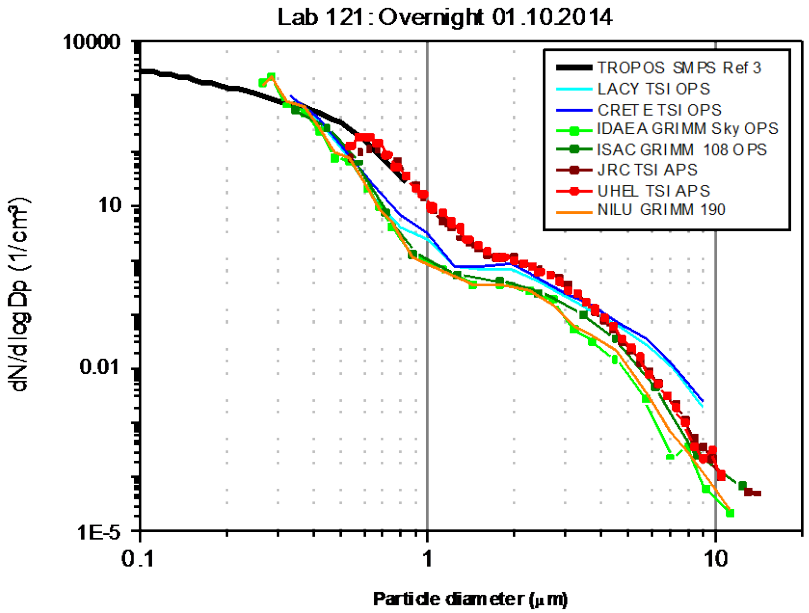

Figure 1: measured PNSD, recalculated to volume equivalent size, of ambient aerosol for three types of instruments (TSI-APSS, TSI-OPSS, Grimm-OPSS)

As shown in Figure 1, the deviations between for the same instrument types (TSI-APSS, TSI-OPSS, Grimm-OPSS) is much lower the deviation between each other. For instruments, calibrated by the manufacturer with PSL (TSI-APSS, TSI-OPSS), the deviation in sizing using again PSL was moderate, less equal 10%. The deviations between the instrument types are rather unsystematic, for certain size ranges higher and for others lower. It can therefore be deduced that this is not primarily an effect of counting efficiency.

The major reason for the deviation is that the measured ambient aerosol PNSD are based on equivalent particle diameters. This means that the deviations between the different instrument types are direct results of an insufficient and or faulty recalculation to a reference size.

One should split between the two aspects:

1. Analysing the performance of the individual instrument with respect to the specific instrument class
   a. Analysing the quality of sizing with respect to the internal calibration
   b. Analysing the measured concentration or detection efficiency
2. Convert the PNSD with their specific equivalent particle diameter (aerodynamic or optical) to a reference particle diameter (e.g. volume equivalent particle diameter)

The second aspect is an enormous scientific challenge with large field of activity. The more complex the aerosol is, with partly unknown internal composition or material properties (density, refractive index, and shape), the more complicated it becomes to convert to a volume equivalent diameter. The situation becomes even more complicated if it is a complex externally mixed aerosol. In this sense, a particle number size distribution for ambient aerosol becomes an inappropriate basis for quality assurance.

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Therefore, to analyze and establish quality assurance procedures for coarse mode particle size spectrometers, it is an essential prerequisite that one differentiate between the two points, only focusing on the first one (1a and 1b).

Analysing the quality of sizing with respect to the internal calibration

It is a well estebalished method to check the sizing, meaning the quality of the calibration, using PSL spheres as a traceable reference for the particle size. The TSI-APSSS and TSI-OPSS are size calibrated by the manufacturer using these particles. In general, differences in the quality of each instrument type are to be expected, e.g. difference in size resolution. In addition, due to measuring principles, different quality for different sizes must be expected, e.g. uncertainty in the Mie-resonance regime of an OPSS. Therefore, the quality standards have to be individually defined. The quality assurance for sizing should performed with different PSL sizes in the specific measuring range of the instrument, e.g. TSI-APSS or TSI-OPSS with approx. 6 sizes for the default measuring range. Examples can be found in the D27.

Analysing the measured concentration or detection efficiency

In general, all coarse-mode particle size spectrometer are effected by a specific detection efficiency.

To determine this size-depended effect, a reference for concentration is needed. To make the procedure applicable in the framework of quality assurance the method should fulfil certain requisitions:

- the reference has to be a SI-traceable method
- the procedure should be carried out in a reasonable time
- the procedure should be applied to several instruments in parallel/simultaneously

Tracer/Filter-Method

Tracer method with filter sampling analysis (gravimetric or SEM) is a well-established method as reference (Armandariz & Leith, 2002; Peters & Leith, 2003; Volckens & Peters, 2005). However, the filter analysis technique is relative time consuming. It is not possible to obtain results immediately or get prompt responses. Therefore the approach was not considered to be used in an operational calibration facility..

Generation of a traceable particle number concentration

Another idea was the generation of a traceable particle number concentration using a vibrating orifice aerosol generator (VOAG). Using this generator, the cconcentration is traceable by dilution flow and the piezo frequency. For a detailed explanation, a complete description can be found in the operation manual of VOAG). With this method, the particles are highly charged and particle losses to instruments have to be completely avoided. This aspect can‘t be guaranteed, therefore this method is also not considered to be used in an operational calibration facility. Nevertheless, the VOAG is a useful generator to produce well defined aerosol particles in the coarse mode size range.

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CPC

Using a CPC as reference for the particle number concentration is a well-established method as reference in the sub-micron range (Karg, Dua & Thiersch ,1991). The instruments are calibrated in the lower submicron range using an Faraday cup aerosol electrometer (FCAE) as SI-traceable reference. Unfortunately, the internal losses are due to impaction and sedimentation. For this approach, a DMA is needed to select monodisperse PSL particles and filter out any smaller (water residuals, coating, debris) or bigger particles (agglomerate). Using a long DMA, it is possible to use this method to compare the particle number concentration of more than one candidate to the upper useful detection limit of a reference CPC of 3 µm in parallel.

In Figure 2, the results from the determined detection efficiencies of three different TSI-APSS and one TSI-OPSS is shown. One can clearly see that the measured particle number concentration of the TSI-OPSS is in a good agreement with the reference CPC with deviations smaller than 5%. In contrast to this, for the TSI-APSS the counting efficiency is significantly decreasing for particles smaller than 0.7 µm. In the particle size range from 0.7 µm to 1.6 µm, all instruments are in a good agreement. This result confirms once again that the deviations for ambient aerosol from Figure 1 is not based on detection efficiency effects, but on a non-comparability of the optical and aerodynamic PNSD.

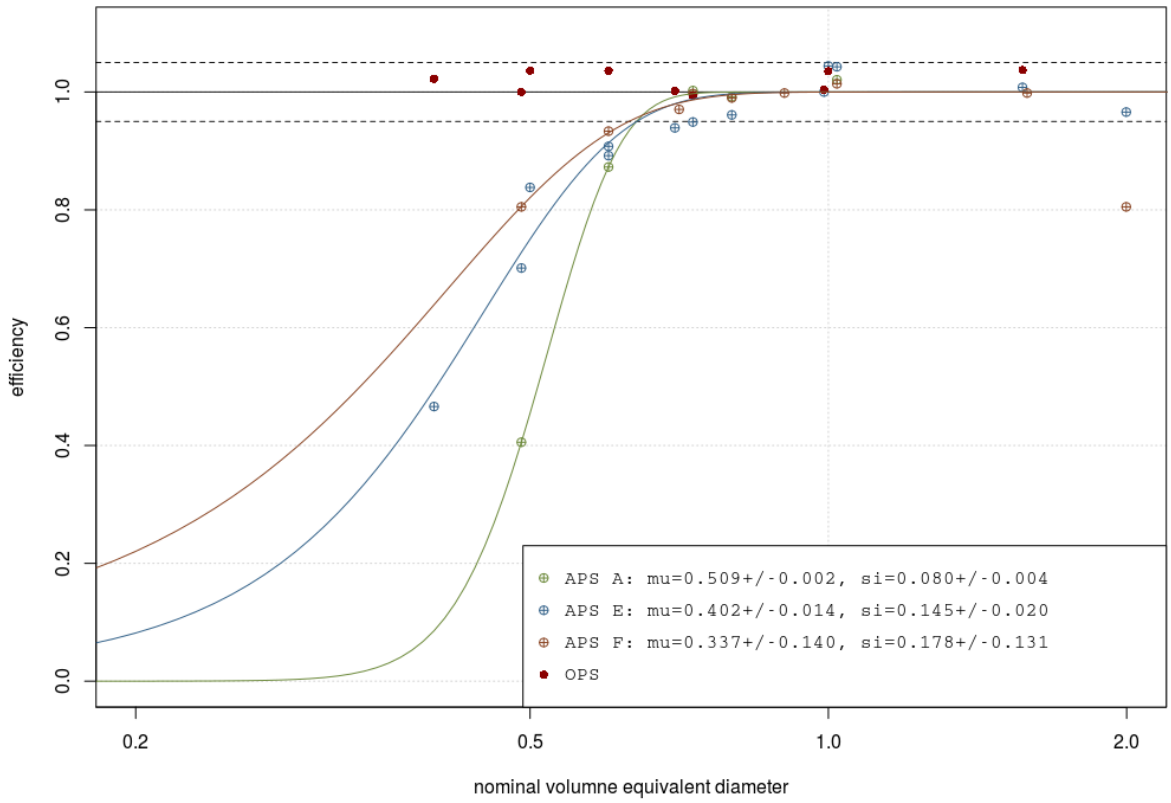

Figure 2: Resulting detection efficiency curve for three TSI-APSS and one TSI-OPSS using a CPC as reference downstream of a DMA

Beside of the upper useful detection range of the CPC, due to large particle losses inside the DMA, it is complicate to generate a moderate particle number concentration for particles larger than 3 using a standard nebulizer. Nevertheless, this method is applicable up 3 µm and covers already a wide range of the atmospheric relevant size range.

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Modified CPC

To improve the method in a further step, there is the possibility to improve the CPC for coarse mode particles. For this approach, the internal pipe layout has to be optimized with a strict vertical arrangement and a top inlet. This modified CPC have to be still calibrated in the lower submicrometer particle size range with FCAE. Traceability is ensured, because all potential particle for super-micron particles can be excluded. It has to be checked whether the instrument would be still operational with a vertical arrangement.

OPSS or another master device

From Figure 2, it is already noticeable that the performance of a TSI-OPSS is better than a TSI-APSS. Compared to a CPC, the instrument is designed for coarse mode particles with a top inlet and a straight pipe layout. The instrument has a weak aerodynamic focussing with a sheath flow. In comparison with the TSI-APSS, particle losses inside of the nozzle are not expected.
It should be checked whether a well-characterized TSI-OPSS or another particle spectrometer could be used as a master device. In such case, no DMA is needed, which then results in higher concentrations especially for particles >3 µm.
Furthermore, it is possible to perform this quality assurance procedure simultaneously with quality checks for sizing, even for PSL mixtures, which will significantly speed up the whole procedure.

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