A calibration procedure adapted to measure in real-time droplet size distribution of e-cigarette aerosols

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Introduction and Objectives

The measurement of aerosol physical properties is key to evaluate the dose exposure of inhalable aerosols. Indeed, depending on the aerosol size distribution, the fraction of the inhaled dose is transported further to the alveolar region (Bernstein, 2004; Kane, 2010). It is generally accepted that an aerosol is considered respirable when its related Mass Median Aerodynamic Diameter (MMAD) is below 2.5 micrometers as more than 80% of the aerosol in mass reaches the alveolar region.

Common practice for determination of particle size in aerosols is to use an impactor technique as it classifies gravimetrically aerosol droplets in distinct size classes and mimics to some extent the deposition behavior in lungs. However, due to the labor-intensive nature of the multi-stage cascade impactor, other techniques such as optical methodologies are usually preferred for high throughput measurement of aerosol size distributions. For this purpose, a commercial TSI Laser Aerosol Spectrometer (LAS) operating at a wavelength of 633 nm was assessed and validated for e-cigarettes.

Methods & Equipment

The LAS signal response was measured using particles/droplets with different indexes of refraction to build calibration curves. Polystyrene Latex (PSL) spherical particles and Di-Ethyl-Hexyl-Sebacat (DEHS) oil droplets were used. Practically, calibrated PSL particles of known sizes were conveyed to the LAS and the accuracy of the response was verified and corrected, if needed. The same procedure was applied for DEHS droplets generated with a Condensation Monodisperse Aerosol Generator (CMAG) and the sizes were checked by means of a Process Aerosol Monitor (PAM). In Figure 1, the LAS signal response (G3) is plotted as a function of selected sizes (D).

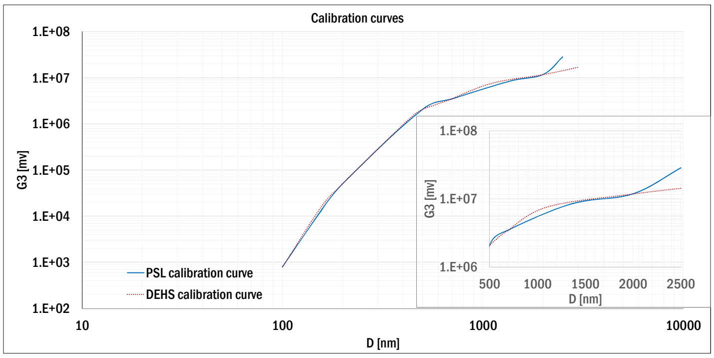

Figure 1: Calibration curves for two aerosol materials.

The instrument responses were significantly different on certain sizes, when comparing PSL particles and DEHS droplets. Typical LAS size responses are displayed in Table 1 for DEHS droplets of known size for both PSL and DEHS calibration curves.

- A good match was found when using the DEHS calibration curve (±10%).
- For a selected DEHS droplet size of 600 nm, according to PAM, the use of the PSL and DEHS calibration curves allowed us to obtain size responses of 499 and 595 nm respectively.
- An underestimation of approximately 15-20% was obtained when using the PSL calibration curve. This underestimation can most likely be attributed to the different indexes of refraction of the test aerosol in comparison to PSL particles. Consequently, it is important to know the refractive index of the particles prior to the measurement.

Table 1: LAS size response for DEHS droplets using two different calibration curves.

DEHS droplet size [nm] generated and measured with a PAM	LAS size response [nm] with PSL calibration	LAS size response [nm] with DEHS calibration
600	499	595
1050	822	924
2000	1705	1930

Experimental Set-Up

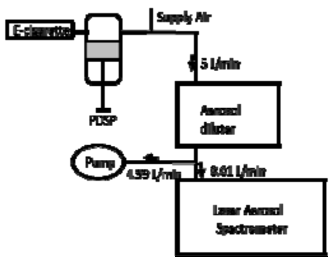

Figure 2: Set-up for e-cigarettes measurement with the LAS

- Measurements were performed on disposable cartridge e-cigarettes by means of a programmable Dual Syringe Pump (PDSP) under the CORESTA puffing regimen (3 seconds bloc puff, 55ml, with 30 seconds puff interval).
- A dilution by 10000 prior to measurement was necessary due to the limit of concentration detection of the LAS (Figure 2).
- Measurements were performed with the PSL calibration curve, due to the expected small size distribution and the higher resolution of the PSL calibration curve in this region.

Results

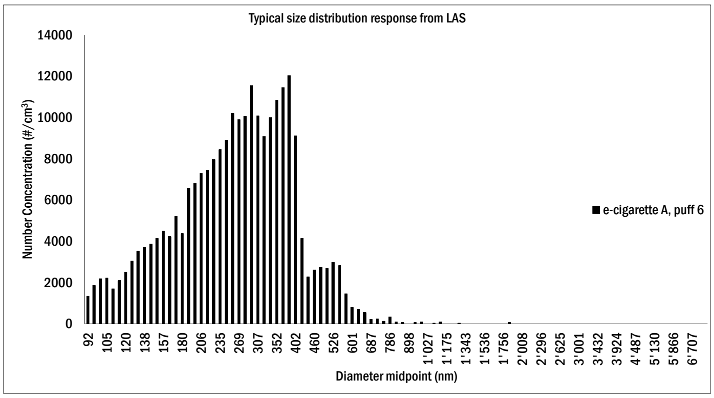

Figure 3: Typical LAS size distribution response for a selected e-cigarette puff

Figure 3 shows a size distribution response. The LAS counts each particle going through the detection chamber and classifies them in different size bins according to their scattered properties.

From the measured size distributions, the related Count Median Diameter (CMD) and the Geometric Standard Deviation (GSD) were calculated by means of an embedded Excel macro provided by the supplier, while the Mass Median Diameter (MMD) was evaluated using the Hatch-Choate equation:

$$MMD = CMD \times \exp(3 \times ln^2(GSD))$$

The calculated MMD presented in Table 2 for three disposable cartridges e-cigarette brands, are in good agreement with the literature.

Table 2: LAS size response for e-cigarette measurements

E-cigarette brands	Count Median Diameter (CMD) [nm]	Geometric Standard Deviation (GSD)	Associated Mass Median Diameter (MMD) [nm]
E-cigarette A	280	1.54	492
E-cigarette B	265	1.57	492
E-cigarette C	263	1.57	487

Due to the nature of the aerosol and the dilution applied prior to measurement, evaporation has to be taken into account, therefore, the actual droplet size could be slightly higher.

Conclusions and Discussion

The LAS instrument calibrated following the standard procedure is fully suitable in cases where the refractive indexes of the test aerosol is close to PSL particles. Otherwise, the calibration should be modified or alternatively, a bias correction factor could be applied. The indexes of refraction of tested e-liquids were found to be close to those of DEHS.

- The LAS size response provides an underestimated size response of 15-20 % for e-cigarette aerosols, when the PSL calibration curve is used.
- From measurements performed on three commercial e-cigarette brands, the Count Median Diameter (CMD) values were found to be from 263 to 280 nm (no bias correction applied) in agreement with literature values (Ingebrethsen, 2012).
- Measurement of the index of refraction of an e-liquid is required in order to determine whether a correction factor should be applied.
- Measurements of aerosols with a totally different chemical nature like iron or soot particles, with a complex index of refraction, would probably fail to provide accurate and linear size responses when PSL or DEHS calibration curves are used.

Perspectives

- Assess the influence of e-liquid's composition on LAS response. Build calibration curves with model e-liquid solutions (glycerin, propylene glycol).
- Compare measurement responses between various instruments (SMPS, FMPS, APS, Spraytech, PIXE, ELPI,...) to determine bias attributed to the nature of the aerosol. Define application domains per instrument and sample type.

References

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