

The manuscript describes the evaluation of a novel thermal precipitation based aerosol electrometer (TPAE) that simultaneously collects particles for subsequent analyses while measuring the current stemming from the deposition of charged particles. If the charge level of the particles is known, e.g. when used downstream of a DMA, the number concentration of particles can be determined. I find the device very interesting and the evaluation has thoroughly been carried out. However, I have a several major and a few minor concerns that I believe the authors should address in a revision:

Major concerns:

It does not become very clear, what is main field of application for the device. Is it mainly to be used downstream of a DMA? This would guarantee that particles are mostly singly charged and that the number concentration can be derived from the current measurement. But then is the main purpose to collect particles for subsequent analyses or to measure particle number concentrations and/or size distributions?

Reply: Thanks for the question. Like existing aerosol electrometers (equipped with HEPA filters), TPAE requires working with charged particles. An aerosol charger is required to charge particles prior to TPAE. The total number concentration of particles can be measured if the average charge status of particles after the charger is known. The assembly of charger-electrometer has also been applied for the particle surface and mass concentration measurement. In the above cases, empirical calibration of assembled devices using typical particles is required and the calibration curves may be varied when measuring particles in the chemical composition different from that of calibration particles. When an electrical mobility classifier/analyzer (DMC/DMA) is included between the charger and aerosol electrometer, the size distribution of particles can be obtained after the correct of multiple charges. The new feature of TPAE is the soft collection of sub micrometer particles, enabling the offline characterization of particle morphology and chemical composition. The above feature is desired when measuring particle agglomerates. The other advantage of the TPAE is without the requirement of periodically changing filters.

When size distributions are measured, how much does the “smearing effect” of the particle deposition inside the device due to the residence time of the particles in the gap space between the two plates before being deposited affect the accuracy? This effect is also likely the reason for the differences observed in the dynamic responses in Figure 4.

Reply: Thanks for the question. Working with a DMA (differential mobility analyzer), particle smearing occurs during the transportation of classified particles from the DMA exit to the collection of particles in the TPAE. The transportation time in TPAE can be calculated in the following: Taking Q as the flowrate of sample flow, h as the distance between two plates, x as the distance between particle position and center axis of disk, t is the residence time of particle in the gap space, it could be calculated that

$$\frac{Q}{2\pi x h} \cdot dt = dx$$

$$dt = \frac{2\pi xh}{Q} \cdot dx$$

$$t = \frac{\pi h}{Q} x^2 \Big|_{x=60\text{mm}} - \frac{\pi h}{Q} x^2 \Big|_{x=0\text{mm}} = \frac{3.14 \times 0.5\text{mm} \times 60^2\text{mm}^2}{0.3 \times \frac{10^6\text{mm}^3}{\text{min}}} = 0.019\text{min} = 1.13\text{s}$$

When Q is 0.3L/min, the maximal particle residence time in TP AE is 1.13s.

Figure 4 shows the overall response time of TP AE including the particle residence time in TP AE and the response time of micro-current measurement circuit.

Note that the particle smearing effect can be minimized when the DMA is operated at the voltage stepping mode instead of voltage scanning mode.

It is not mentioned what size range the instrument is designed for. I assume, only for particles smaller than approximately 300 nm, for which nearly size-independent thermophoretic deposition can be expected. It is also not discussed that the assumption of size-independence only holds in the free molecular and near free-molecular regime, where the thermophoretic force (see L. Waldmann and K. H. Schmitt, *Aerosol Science*, edited by C. N. Davies, Academic, New York, 1966) and the opposing drag force share the same size dependence.

Reply: Thanks for the comment. The TP AE is designed for particles in sizes smaller than 200 nm. Accordingly, the size range description has been included in the revised manuscript (Line 17).

When the TP AE is used downstream of a DMA, the size-independence of the deposition only holds for a fixed DMA-voltage. When the voltage is ramped during size distribution measurements, the concentration and thus the representativeness of the collected sample is dependent on the particle size due to the size dependent particle charging probability.

Reply: Thanks for the comment. Indeed, for the offline characterization of particles, the DMA voltage requires to be fixed. When the DMA+TP AE is used for the size distribution measurement, the smearing of classified particles can be prevented by operating the DMA at the voltage stepping mode instead at the voltage ramping mode.

Why was a radial preferred over a rectangular design? While the deposition efficiency is nearly independent of particle size (with the limitations mentioned above), the radial design introduces a spatial dependence on the particle deposition, due to the decreasing flow velocity from the center to outside. This could be overcome by a rectangular design (see e.g. Azong Wara et al., *J. Nanopart. Res.* **11**: 1611-1624, 2009)

Reply: Thanks for the comment. The thermal precipitation can be done either in the parallel-plate, parallel-disk and coaxial tube configurations. The selection of the radial

configuration for TPAE was because it will be a part of the particle sizer under the development and the radial design best fits the limited space requirement. The uniformity of particle deposition is not our concern.

The suggestion is, however, excellent. We will investigate the rectangular configuration of TPAE in the near future.

Why was air-cooling preferred over the more commonly used liquid cooling?

Reply: Thanks for the question. The primary reasons for using air cooling in TPAE (instead of liquid cooling) is to suppress the noise of current signal, and (2) to reduce the liquid handling for the TPAE operation (convenient for field applications). Liquid cooling is used in published thermal precipitators because of the high specific heat of liquids (carrying more heat). However, the current input needle of TPAE is directly connected to the TPAE cold plate. If a liquid is used for colling, the contact of cold plate and liquid (with the liquid conductivity much higher than air) significantly increases the noise of current signal. The other reason for air colling is to remove the liquid handling for the TPAE operation (a welcome for field applications).

With such rather high temperature gradients, did you check if buoyancy may affect the deposition efficiency?

Reply: Thanks for the question. The buoyancy was not concerned because of the tiny spacing between two disks (i.e., only 0.5 mm). With the tiny spacing, the temperature difference between two disks is small to achieve the high temperature gradient.

Section 4.1.2 provides only observations. I would expect to see at least some quantitative analysis, such as the t_{10-90} and t_{90-10} time constants of both the TSI electrometer and the TPAE.

Reply: Thanks for your suggestion. For sample flowrate of 0.3L/min and 0.6L/min, t_{10-90} and t_{90-10} are calculated for TPAE and TSI3068B.

For 0.3L/min, TPAE: $t_{10-90}=36.42s-32.46s=3.96s$; TSI: $t_{10-90}=36.23s-32.4s=3.83s$.

For 0.3L/min, TPAE: $t_{90-10}=134.09s-127.5s=6.59s$; TSI: $t_{90-10}=134.01s-127.38s=6.63s$.

For 0.6L/min, TPAE: $t_{10-90}=20.96s-19.13s=1.83s$; TSI: $t_{10-90}=21.53s-19.45s=2.08s$.

For 0.6L/min, TPAE: $t_{90-10}=77.21s-74.56s=2.65s$; TSI: $t_{90-10}=78.58s-75.06s=3.52s$

For 0.3L/min, the response time of TPAE almost equaled to TSI3068B while for 0.6L/min, TPAE responded faster.

The above data has been added in Line 256~260 of revised manuscript.

While the manuscript in principle well written, it would benefit from a language check by a native speaker.

Reply: Thanks for your suggestion. The revised manuscript has been checked by a native speaker with a PhD degree in engineering.

Minor comments:

Line 51: what is meant with “early generation” of TSI model 3068B? To the best of my knowledge, this model is still commercially available.

Reply: Thanks for the comment. The description has been deleted.

Line 52: it should read Faraday cup, not Faraday cage

Reply: Thanks for the comment. It has been corrected.

Line 55: This paper refers to the mini DiSC, not DiSC

Reply: Thanks for the comment. It has been corrected.

Page 3, first paragraph: The Nanomater Aerosol Sampler (TSI model 3089, Dixkens et al., *Aerosol Sci. Technol.* **30**: 438-453, 1999) is missing in the discussion. This has been developed to electrostatically sample monodisperse particles downstream of a DMA.

Reply: The missing reference has been added in Line 71-72 of the revised manuscript.

Line 77: particles move in counter-direction of a thermal gradient, not in the direction.

Reply: Thanks. It has been corrected.

Line 100: it appears that height and diameter are reversed here; it should thus read ...120 and 0.5 mm in diameter and height, respectively. I would also suggest the term “gap distance” rather than height.

Reply: Thanks for the suggestion. It has been revised.

Line 220-225: When I calculate the temperature gradient based on the provided values, I get 265 K/cm, not 254 K/cm. Either the temperature values or the value for the gradient seems to be incorrect.

Reply: Thanks for pointing it out. Accordingly, it has been corrected throughout the revised manuscript.