General comments:

1. What is the motivation for designing this new electrometer? Compared with the TSI electrometer, why do we need this TPAE?

Reply: Thanks for the question. This development was motivated by the fact that TSI electrometer can only measure the aerosol current. The recovery of collected particles for future offline characterization such as the particle morphology by SEM and chemical analysis is not possible. Although ELPI can be used to collect particles, the collection by the inertial impaction may alter the shape of particle agglomerates, e.g., soot particles. By combining thermal precipitation with aerosol current detection, the device can measure the aerosol current while collecting particles by soft landing. Moreover, the requirement of changing HEPA filters used in existing AEs is not needed (a welcome for instruments to be used the field study).

The above advantages have been stated in Line 29~31 of revised manuscript.

The motivation had been added in Line 71~73 of revised manuscript: "*The collection of charged particles by the filtration, inertial impaction … is not favored for the off-line SEM analysis if required*".

2. Experimental design issues

(1) The singly charged assumption limits the application of this electrometer. Therefore, the author should consider adding a CPC or another SMPS in the setup to measure the charge distribution for the testing particles. Then implement that into the calculation in equations 1-3. Or at least provide the uncertainty caused by the single charge assumption.

Reply: Thanks for your suggestion. Because our test particles were obtained by the DMA classification, it is the reason why we assumed particles are singly charged. For the general application of the device, a particle charger must be used in front of the developed device. The charge status of particles is thus dependent on the charging performance of selected chargers. Accordingly, Eq. 2 has been modified to include "x is the average charge of particles, which shall be obtained for the charger calibration (Line 123). The accuracy of x did affect the accuracy in the conversion from the aerosol current to the particle number concentration. However, the performance of the TPAE would not be affected.

(2) The size effect on the performance of the TPAE covered 25-200 nm. Are those results under the singly charged assumption? If so, will the charge distribution affect the result?

Reply: Thanks for the question. The result shown in the manuscript were under the singly charged particle assumption. In our experiments, the fraction of multiple-charged particles was minimized by selecting test particles from the right-hand side of the peak size in the size distribution of particles generated from aerosol generators. In addition, the ratio of multiple charged particles to singly charged ones is low (lower than 2.1%, $d_p \leq 40\text{nm}$) according to Wiedensohler Formula, as shown

in Figure R1. The combination of the above factors will significantly reduce the fraction of multiple-charged particles in test particles.

Table 1-1
Distribution of Charges on Aerosol Particles According to the Wiedensohler Formula

Percent of Particle Carrying Np Elementary Charge Units													
Dp(μm)	Np=–6	–5	–4	–3	–2	–1	0	+1	+2	+3	+4	+5	+6
0.01						5.14	90.75	4.11					
0.02					0.02	10.96	80.57	8.64	0.01				
0.04					0.54	19.50	64.79	14.86	0.31				
0.06				0.02	1.92	24.32	54.13	18.51	1.09	0.01			
0.08				0.11	3.73	26.81	46.75	20.46	2.10	0.05			
0.10				0.37	5.63	27.31	42.28	20.91	3.30	0.17			
0.20		0.05	0.53	3.40	12.38	25.49	29.66	19.51	7.26	1.53	0.18	0.01	
0.40	0.27	1.14	3.60	8.54	15.24	20.46	20.65	15.66	8.93	3.83	1.24	0.03	0.05
0.60	1.21	3.00	6.19	10.53	14.82	17.25	16.60	13.20	8.69	4.73	2.13	0.79	0.24
0.80	2.42	4.64	7.71	11.12	13.90	15.06	14.15	11.53	8.15	4.99	2.65	1.22	0.49
1.00	3.56	5.84	8.53	11.13	12.96	13.45	12.46	10.30	7.59	5.00	2.93	1.54	0.92

Figure R1. Charge fraction given in TSI3088 (X-ray Neutralizer) manual

(3) This study didn't characterize the real concentration range of the electrometer. At least, it should give the lowest limits for reliable concentration detection for different sizes of particles.

Reply: Thanks for your suggestion. The lowest limits for reliable concentration detection for particles are decided by $C = \frac{\Delta N_p}{q_s t_s} = \frac{\int \Delta I_p \mathrm{d}t}{ex\eta q_s t_s}$, where C is the number concentration of sampled particles, t_s is sample time and q_s is sample flowrate. The concentration range of the device does not depend on the particle size directly.

By the calculation, the number concentration range of TPAE is $2{,}500\#/\mathrm{cm}^3$~$6.25\times10^7\#/\mathrm{cm}^3$ at $q_s = 0.3\mathrm{L/min}$ and $x = 1$. The following is the calculation: TPAE collects 98.9% of particles at the sampling flow rate of 0.3 lpm according to Figure 5(a). Thus, with $q_s = 0.3\mathrm{L/min}$, $\eta = 98.9\%$, $x = 1$, $t_s = 1\mathrm{s}$, we obtained $C \approx (1250\mathrm{cm}^{-3}\cdot\mathrm{fA}^{-1})\Delta I_p$. The lower limit of aerosol concentration is based on the lower limit of current detection, $\pm2\mathrm{fA}$, resulting in the number concentration of $2{,}500\#/\mathrm{cm}^3$. The upper limit of particle concentration was calculated based on the upper limit of current detectable by the used micro-circuit, which is $\Delta I_p = 5\times10^4\mathrm{fA}$.

The upper limit range of TPAE is difficult to be experimentally determined because the concentration of test particles (after a DMA) is very low compared to the required concentration ($6.25\times10^7\#/\mathrm{cm}^3$).

The above information is included in the abstract (Line 22-23) of the revised manuscript.

Specific comments:

Abstract: What are the "other physical parameters"? It might be good to mention them here to emphasize the need for TAPE development.

Reply: Thanks for the question. Instead of “other physical parameters”, we have revised the paragraph as “Note that the charger-AE assembly…….” (Line 51-54).

Line 22: Is this range based on theoretical estimation or experimental confirmation? It seemed that this was only a theoretical range. If so, please provide a practical range. In addition, it is even desirable to convert it to the aerosol concentration range. Please also specify the corresponding size range.

Reply: Thanks for the question. Please see our detailed reply in Q#3. The range is based on theoretical estimation. The calculated number concentration range has been added in Line 22-23 of revised manuscript.

Line 24: The evaluation used one TSI aerosol electrometer to determine another aerosol electrometer – TAPE? Why not use CPC? How does the author determine the charging state for the testing particles?

Reply: Thanks for the question. The reason for not using CPCs for the comparison is because electrometers are used to calibrate the performance of CPCs (ISO 27891:2015). The number concentration of particles (#/cm^3) must be traced to the current (fA) eventually.

Test particles are assumed to be singly charged because of the use of a DMA for particle classification. The DMA classification of particles cannot completely avoid the inclusion of multiple-charged particles in DMA-classified particles. To minimize the fraction of multiple-charged particles, we classified test particles from the right-hand side of the peak size of particles generated from the aerosol generators. The details can be found in our reply of Q#2.

Line 27: "The effect of particle size on the above efficiency was minor for sodium chloride particles." This is not true in general. It should depend on the size range of the testing particles.

Reply: Thanks for the question. The conclusion arrived according to our experimental results, Figure 8, in addition to previous studies on the thermal precipitation of sub-micro-meter particles (such as http://dx.doi.org/10.1016/j.jaerosci.2012.04.004: Performance study of a disk-to-disk thermal precipitator). Please note that “minor” does not mean “no”.

Accordingly, the description has been revised as “The effect of particle size on the above efficiency was minor for sodium chloride particles in the sizes of 23-200 nm” (Line 27-28).

Line 55 and 58, please spell out DiSC and TEOM.

Reply: Thank you for the suggestion. DiSC (Diffusion Size Classifier), TEOM (Tapered Element Oscillating Microbalance). The above has been added to Line 59 and 63.

Line 73: How does the author characterize the aerosol particles as "small" or "large"? Please specify the size range.

Reply: Thank you. “Small” and “large” are adjectives for the particle sizes. There is no scientific

consensus in the aerosol community on how "small" is considered as small and how "large" is considered as "large". By electrical classification and detection, "small" particles are for ones of $d_p < 800\text{nm}$ while "large" is for particles of $d_p \geq 800\text{nm}$. The above is based on the upper size limit for TSI DMA classification.

Accordingly, the above statement has been revised as "the collection by the inertial impaction favors for inertial particles, and the electrical collection favors for diffusive particles' (Line 79)

Introduction: It is not clear what the advantages are of developing a TPAE. Will it extend the lower detection efficiency for small-size particles? Or will it cover a wide range of aerosols?

Reply: Thanks for the question. Please see the detailed reply in Q#1

Fig. 1: Where are the thermistors? And pre-amplifier? Please add them to the figure. Will the location affect the temperature control? If so, please explain the effects. How do you maintain the thermal gradient under different environmental conditions? Are there any feedbacks to control the cooling flow or heating plate? Will the device be used in an outdoor environment?

Reply: Thanks for the question. The thermistors were black arrows in Figure 1 and the captions in Figure 1 had pointed it out. The "amplifier" in Figure 1 has been revised as "pre-amplifier".

The uniformity of plate temperature was examined (using a thermocouple probe) prior to our testing. The variation of plate temperature was within three degrees (°C). In our lab, the room temperature was relatively stable. As the heating and cooling power of the device were fixed, the temperature gradient in the precipitation zone eventually reached stable. Therefore, the temperature feedback control was not conducted. For the outdoor applications, feedback control on the plate temperature will be included.

Fig 7. Does the fitting line indicate that the efficiency is higher than 120% at 0.2? Is it realistic? Why do we have different fitting parameters for different size aerosol particles? Such as 30 for 70 nm and 26.3 for 200 nm? It is crucial to investigate the size effect on the performance of this electrometer.

Reply: Thanks for your correction. The maximal collection efficiency is 100%. Accordling, the figure has been corrected.

The thermophoresis force is calculated by $\vec{F}_{th} = \frac{3\pi\mu^2 d_p H \overrightarrow{\Delta T}}{\rho_g T}$, and drag force is calculated by $\vec{F}_{drag} = -\frac{3\pi\mu d_p \vec{V}}{C_c}$, where $H = \frac{2C_s(\frac{k_g}{k_p}+C_t K_n)}{(1+3C_m K_n)(1+\frac{2k_g}{k_p}+2C_s K_n)}$. $\overrightarrow{\Delta T}$ is temperature gradient, T is the absolute temperature of particle, ρ_g is the density of air and H is the thermophoretic coefficient. Let $\vec{F}_{th} + \vec{F}_{drag} = 0$, we could get $\vec{V}_{th} = \frac{\mu\overrightarrow{\Delta T} H C_c}{\rho_g T}$, according to which we know that $\vec{V}_{th}$ is not directly related to d_p, while H and C_c are related to d_p. Thus, the effect of d_p on the thermal

precipitation does exist but is minor. Consequently, the fitting result should not be the same. The size effect on the thermal precipitation was shown in Figs. 8 and 9.

Fig 8 and Fig 9, why several typos in the manuscript: TDAE? Should it be TPAE?

Reply: Thanks. It is corrected.