Aerosol charge state characterisation using an ELPI

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Abstract. A new technique has been developed to measure the size distribution and charge state of highly charged aerosols using an Electrical Low Pressure Impactor (ELPI). The internal charger was switched alternately on and off and the time between stable charge states found to be ~10 s. The size distribution of aerosols was found when the charger was on, from which the charge distribution can be estimated when the charger is off using the current at each impactor stage. This method was tested in background conditions, when a candle was burning and when a negative air ioniser was used. The ELPI electrometers were not sensitive enough to accurately measure the charge state on background and candle air, but gave a value for air charged by an ioniser. Comparing results from the ELPI with other techniques showed inaccuracies in this method that need to be addressed before further use of this technique.

1. Introduction
The lung deposition probability of inhaled particles is usually described by the ICRP 66 model [1]. In practice, deposition efficiency is below 100%, with a minimum of only 30% at 200 nm, the remaining fraction being exhaled again. The model, however, takes no account of particle charge state and there is evidence that charged particles deposit more efficiently than uncharged or charge-neutralised particles [2]. High voltage power lines are known to emit corona ions [3] which can create a unipolar excess of ions downwind. This can lead to unipolar charging of atmospheric aerosols [4, 5]. It is hypothesised that this increase in charge state could lead to enhanced deposition within the lung [6].

We intend to test whether 100 nm and 200 nm particles with excess unipolar charge have a higher probability of lung deposition than charge-neutralised particles, when inhaled by healthy human volunteers. A key part of the study is characterisation of the charge state and size of inhaled particles.

In this paper we seek to investigate whether the extent of aerosol charging can be determined using an Electrical Low Pressure Impactor (ELPI) (Dekati, Finland). The ELPI is a cascade impactor that uses an internal corona charger to bring particles to a known charge distribution. The current on each impactor stage is measured and the size distribution is obtained in conjunction with the charging probability. Existing research has used the ELPI to measure the total (net) charge present at each stage during a single pharmaceutical inhaler actuation [7]. Here, the charger in the ELPI is alternately switched on and off to allow alternate, on-line measurement of both size and charge distributions (i.e. the probability of a particle of given size carrying a given number of electronic charges).
2. Methods

2.1. Experimental methods

Ambient indoor air was sampled using an ELPI with volumetric flow 9.87 l min\(^{-1}\). When the internal +5 kV charger is on, the particle size distribution can be found from the current values and known particle charging probability at each filter stage [8]. The electrometer measurement range 0 – 100,000 ±15 fA was used. Three different indoor conditions were examined: i) normal background air, ii) with a burning candle, to increase aerosol concentration and iii) with a commercial negative air ioniser, to alter the aerosol charge state. For each run, the ELPI was operated for 30 minutes, with the internal corona charger alternately switched on and off every 30 s. In the candle and ioniser tests, some time was allowed until the readings reached a steady state.

A second test compared aerosol charging results from ionised air using the ELPI to previously used techniques. The bipolar or unipolar electrical environment, when diffusion charging dominates, can be investigated using Aspiration Condenser Ion Mobility Spectrometers (ACIMS) [4] which measure small ion mobility and concentrations, or a ‘Dual-Differential Mobility Analyser’ (D-DMA) technique [5] which uses size- and mobility-distributions to estimate the ambient charge state of sampled aerosol particles. Both can be used to infer a value for the (steady state) ratio of positive to negative ion concentrations \(c_+ / c_-\), multiplied by their associated mobilities \(\mu_+ / \mu_-\), referred to as the charge asymmetry ratio, \(x\) (1). \(x\) can be used to describe the charging probability for aerosols with a Gaussian charge distribution when aerosol concentration is constant, regardless of ion properties [9]. 1 hour of data were collected using the ELPI, ACIMS and D-DMA with the negative room ioniser on.

\[
x = \frac{c_+ \mu_+}{c_- \mu_-}
\]

2.2. Calculation of size distribution, average charge per particle and overall charge distribution

If the size distribution is known, the ELPI can be used to obtain the average charge per particle at each ELPI stage and potentially to estimate the overall charge distribution i.e. the probability that a particle of a given size has \(n\) elementary electronic charges (where 1 electronic charge is the charge of an electron). The relationship shown in (2) is used to calculate the mean concentration of particles at each filter stage, \(C\), from the current, \(I\), which enables a size distribution to be estimated. \(Q\) is the flow rate of air, \(e\) is the charge of an electron and \(P\) is the penetration of particles through the corona charger. The quantity \(Pn_c\) is a constant for each stage [8].

\[
I = CPn_c eQ
\]

When the mean concentration for each ELPI stage has been calculated for the times when the internal charger is on, the net average number of elementary charges per particle for each stage can be estimated from the current measured within the ELPI when the internal charger is switched using (2). When the charger and trap voltage are on, \(P\) and \(n_c\) can be considered theoretically [10] however, the effects when the charger and trap voltage are off are not known. Glover and Chan [7] tested the difference between the total charge measured with the corona charger in place but switched off and with the charger removed from the ELPI, and reported no significant change. Therefore penetration through the charger, \(P\), is assumed to be 1. A simple analytical expression for aerosol charge distributions was put forward by Clement and Harrison [9], which includes an expression (3) for the average number of charges carried by a particle of diameter \(d\), where \(k\) is the Boltzmann constant, \(T\) is the temperature and \(\varepsilon_0\) is the permittivity of free space.

\[
n = \frac{2\pi\varepsilon_0 kT}{e^2} \ln x
\]
3. Results

3.1. Characterisation of the instrument response time

Figure 1 shows the current measured on two stages of the ELPI corresponding to average diameters of 120 and 200 nm. A negative room ioniser was activated at the beginning of the measurement. At 180 s the ELPI internal charger was switched on, then turned off and on alternately every 30 s thereafter.

After switching the internal corona charger on or off, all particles undergo a change of charge state, between ambient and corona charged. The current on each filter stage changes until a new stable current is reached corresponding to the concentration at a particular size. Particles larger than 1 μm in diameter also exhibit a transient lasting a few seconds due to image charge effects. The time taken for the electrometers (using the 100,000 fA setting) on all filter stages to settle was ~10 s.

![Figure 1. Time series of the current measured in an ELPI in ionised air with the internal corona charger switched on (shaded area) and off.](image)

3.2. Size distributions and average charge of aerosols

The data from the ELPI was separated into times when the internal charger was on and when off. The first 10 s after the charger was switched on or off was discarded so that transitional data was not used in the calculations. Figure 2 shows the size distributions found for background conditions, while an air ioniser was used and when a candle was burning. The size distributions for background and ionised air are broadly similar, while there is a large increase in aerosols smaller than 1 μm when a candle is burnt, consistent with previous measurements of candle burning [11]. The current at each filter stage when the internal corona charger was off, after the first 10 s of settling time was discarded, was used in equation (2) to calculate the average charge per particle for each stage, shown in Figure 3.

3.3. Comparison of \( x \) obtained using ELPI, ACIMS and D-DMA

The charge asymmetry ratios \( (x\text{-value}) \) calculated from the ELPI for each size bin were compared to those obtained from ACIMS and D-DMA results. ACIMS measures ion properties only, while D-DMA infers a value of \( x \) based on aerosol charging over a range of diameters (in this case 10-1100 nm), so only a single \( x \) value is given in both cases. For ACIMS, \( x = 0.032 \) while for D-DMA, \( x \sim 0.03 \) during the period when the room ioniser was switched on, consistent with significant negative unipolar charging. This is in contrast to ELPI results, which were of the order \( x \sim 10^{-5} \) to \( 10^{-7} \) for the largest particle sizes (3-10 μm, first and second impactor stages) and increased to a maximum of \( x\sim 0.2 \) for the smallest sizes (7-30 nm, ELPI filter stage). The very low estimated values of \( x \) for the largest sizes (which suggest an unfeasible bias towards negative charging) may result from space charge effects due to the presence of negative ions, enhancing charge deposition on the upper plates. Also, mean particle charge is obtained using currents around the lower resolution limit of the electrometers (several fA).

4. Discussion and conclusions

This study seeks to characterise the charge state of submicron particles for use in a study on the deposition of charged particles in the human lung. For 120 nm and 200 nm particles, the average charge was 4.2 and 8.3 negative charges per particle respectively when the room ioniser was...
The average charge was ~0 for both sizes for background and candle burning, suggesting little deviation from a Boltzmann charge distribution. However in this case the measured currents were of similar order of magnitude to the accuracy limits of the electrometers for each ELPI stage.

**Figure 2.** Size distributions (dN/dln(d)) of aerosols measured by the ELPI.

**Figure 3.** Average particle charge obtained using an ELPI with switched internal charger.

There is good agreement between the overall values of $x$ obtained from ACIMS and D-DMA measurements, and in general poor agreement between these values and those obtained from ELPI. This suggests that, for these charging levels, the present method using the ELPI cannot be used to determine the aerosol charge state. Previous studies have shown that ELPI may not be satisfactory in measuring bipolarly-charged distributions [12] which casts further uncertainty on the results from the background and candle measurements. The assumption given in section 2.2 that $P = 1$ could be found to be incorrect and further work is recommended to assess the effect of space charge and image charge effects of charged particles within the ELPI.

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**References**

[1] ICRP. 1994. *Human Respiratory Tract Model for Radiological Protection* ICRP Publication 66 (Oxford: Pergamon)

[2] Melandri C, Tarroni G, Prodi V, De Zaiacomo T, Formignani M and Lombardi C C 1983 *J. Aerosol Sci.* **14** 657–69

[3] Matthews J C, Ward J P, Keitch P A and Henshaw D L 2010 *Atmos. Environ.* **44**(9) 5093–100

[4] Few A P, Holden N K, Keitch P A and Henshaw D L 2005 *Atmos. Res.* **76** 29–48

[5] Buckley A J, Wright M D and Henshaw D L 2008 *Aerosol Sci. Technol.* **42** 1042–51

[6] Henshaw D L 2002 *Med. Hypotheses* **59** 39–51

[7] Glover W and Chan H K 2004 *J. Aerosol Sci.* **35** 755–64

[8] Marjamaki M, Keskinen J, Chen D R and Pui D Y H 2000 *J.Aerosol Sci.* **31** 249–61

[9] Clement C F and Harrison R G 1992 *J. Aerosol Sci.* **22** 481–504

[10] Harrison R G 1996 *J. Aerosol Sci.* **27** S1 S191-92

[11] Ouf F and Sillon P 2009 *Aerosol Sci. Technol.* **43** 658-698

[12] O'Leary M, Balachandran W and Chambers F 2008 in Conf. Rec. IEEE-IAS Annual Meeting, 1–5

Belaca R, Abbod M, Balachandran W and Miller P R 2010 *IEEE Trans. Ind. Appl.* **46** 1181–87