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Atmospheric ion spectra and the rate of voltage decay of an aspirated cylindrical capacitor

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Abstract. Aspirated cylindrical capacitors are commonly used instruments for atmospheric ion measurements, more of which are needed for climate and pollution studies. However the classical theory of one operating mode of this instrument is based on an approximation, which may introduce errors in ion measurement. A modification to the classical theory of the instrument is consequently proposed. Applying this modification enables ion mobility spectrum information to be extracted from the rate of voltage decay of the aspirated capacitor in air. It can also be used to improve air conductivity measurements.

1 Introduction

The ventilated cylindrical capacitor or *Gerdien condenser* [1] is a long-established instrument for atmospheric ion measurements. It consists of a cylindrical outer electrode containing a coaxial central electrode, with a fan to draw air through the electrodes. With an appropriate bias voltage applied across the electrodes, a current flows in proportion to the ion concentration. Early ion measurements inferred the current from the rate of decay of voltage across the electrodes, using a gold-leaf electrometer [2]. As electronics technology developed, this technique was superseded by direct measurements of the current. Contemporary instruments based on this principle deploy modern electronics and computer control to use both the Current Measurement and Voltage Decay measurement modes for self-calibration [3,4].

Measurements with modern instrumentation showed that although generally comparable, the two modes were not always completely consistent [3]. This motivated reconsideration of the classical principles of the Voltage Decay mode. The modified approach described here improves conductivity determination and enables ion mobility spectra to be retrieved from voltage decay measurements. Improved ion measurements are needed for solar-terrestrial physics, pollution studies, and assessing long-term changes in atmospheric electrical parameters related to climate change.

2 Classical theory of air conductivity measurement

Atmospheric conductivity is proportional to both the atmospheric ion concentration n, and the average mobility μ of the air ion population. Molecular ions with $\mu > 0.5$ cm^{2}V^{-1}s^{-1} are conventionally defined as "small ions" [5]. The unipolar air conductivity σ can be written as

$$\sigma = e \int_{0.5cm^2V^{-1}s^{-1}}^{\infty} n \quad d\mu \tag{1}$$

where e is the charge on the electron. Measurements averaged over some years show the typical shape of the average small ion mobility spectrum [5]. The mean mobility is defined as the mode of the distribution [6], usually 1.3-1.6 $\mathrm{cm^2V^{-1}s^{-1}}$ for positive and 1.3-1.9 $\mathrm{cm^2V^{-1}s^{-1}}$ for negative ions, (the difference arises because of the varying chemical composition of positive and negative ions) [5,6]. Typical values of atmospheric conductivity are 5-100 $\mathrm{fSm^{-1}}$, depending on pollution levels [7].

To derive a simple expression for atmospheric conductivity, it is convenient to assume that all atmospheric ions have the same mobility, and (1) is commonly simplified to

$$\sigma_\pm = en_\pm \mu_\pm \,. \tag{2}$$

For a classical Gerdien condenser in Current Measurement mode, if the ions reaching the central electrode are constantly replenished, air conductivity is proportional to the ion current measured at the central electrode. (The "conductivity measurement régime" requires an adequate ventilation speed and bias voltage, and can be verified by a linear response of measured current to a changing bias voltage). Using (1), and considering the charge arriving at the central electrode per unit time results in the routinely-used equation for calculating bipolar atmospheric conductivity from current measurements with a Gerdien condenser, considered as a classical capacitor with capacitance C [8,9].

$$\sigma_\pm = \frac{\varepsilon_0 i_\pm}{CV_\pm} \,. \tag{3}$$

In the Voltage Decay mode, a voltage across the electrodes will decay due to the current i flowing through the air, which has a large resistance R. If the charge on the capacitor is Q, elementary circuit analysis gives

$$\frac{dQ}{dt}R = -\frac{Q}{C} \tag{4}$$

from which the familiar expression for the exponential decay of charge from a capacitor to a voltage at time t, V_t from an initial voltage V_0 with time constant τ, is

$$V_t = V_0 \exp\left(\frac{-t}{\tau}\right). \tag{5}$$

The current of negative ions from a positively charged capacitor can be derived from Gauss's Law [8], when the permittivity of free space is ε_0, as

$$i = -\frac{\sigma Q}{\varepsilon_0} \,. \tag{6}$$

Since current i is equal to rate of change of charge dQ/dt then

$$Q = Q_0 \exp\left(-\frac{\sigma t}{\varepsilon_0}\right) \tag{7}$$

By analogy with (5), for a Gerdien condenser in Voltage Decay mode σ is given by

$$\sigma = \frac{\varepsilon_0}{\tau} \,. \tag{8}$$

(8) has been the standard expression for calculating air conductivity from voltage decay measurements throughout the history of the Gerdien condenser instrument, using τ determined from a time series of voltage measurements [2,3,8]. (4) assumes that the resistance of air is constant, to give an exponential rate of decay of charge from a Gerdien condenser. However, measurements show that a non-exponential decay rate is commonly observed [3,10], suggesting this approximation may not be universally appropriate.

3 Modification to the classical theory of the Voltage Decay mode

The minimum mobility of ion contributing to the air conductivity measurement, the *critical mobility* μ_c, [*e.g.* 8] is often approximated from the ventilation speed through a Gerdien condenser u, its length L and electrode radii a and b and the bias voltage V:

$$\mu_c = \frac{ku}{V}. \tag{9}$$

where k is a constant related to the size of the Gerdien condenser

$$k = \frac{(a^2 - b^2)\ln\left(a/b\right)}{2L}. \tag{10}$$

The functional dependence of μ_c on the bias voltage has been exploited to compute ion mobility spectra from the changing ion current at the central electrode [11].

During a Voltage Decay measurement the voltage across the electrodes decreases; from (9), μ_c also varies during the decay. The number of ions contributing to the measurement therefore changes as a function of the decay voltage. This is analagous to voltage decay through a voltage-dependent resistor *R(V)* in a parallel RC circuit, and (4) can be rewritten to include this,

$$\frac{dq}{dt}R(V) = -\frac{Q}{C}. \tag{11}$$

Solving (11) gives a modified form of the classical decay, in which V_t is implicitly defined:

$$V_t = V_0 \exp\left(\frac{-1}{C}\int_0^t \frac{dt}{R(V)}\right). \tag{12}$$

R(V) and the shape of the decay curve are related to the shape of the ion mobility spectrum. If all ions have the same mobility (referred to here as a *flat* ion mobility spectrum) *R(V)* is constant during the decay and (12) reduces to (5), the classical exponential decay. For *all other* spectra, (here called *variable* ion spectra), *R(V)* is not constant and there is no theoretical basis on which expect an exponential decay. This is the general case in the atmosphere [5].

4 Inversion of prescribed mobility spectra

The voltage decays expected from prescribed atmospheric ion mobility spectra can be calculated by numerical integration of (12) [7] (Figure 1). The simplest ion spectrum is flat (Figure 1a). As the number of ions in each mobility category is constant, the effective resistance does not change during the decay, and the voltage decay is exponential (Figure 1b). Another simple shape of mobility spectrum is based on existing average ion spectra [5] (Figure 1c). The voltage decay from this spectrum (Figure 1d) is not exponential, with only 73% of its variance explained by an exponential model. A typical voltage decay time series in atmospheric air would only yield an exponential decay to a first approximation.

Figure 2 shows typical voltage decay time series in atmospheric air. The instrumentation was optimised for ultra-low leakage [3,4], and a calibrator was developed specially for the novel electrometer used in the experiment [12,13]. Over the entire data set with 39 voltage decays measured over two weeks, the mean r^2 was 0.887 with a standard deviation of ±0.134 (the range of values was 0.310-0.998). This non-exponential behaviour is consistent with many independent observations of a variable ion spectrum in atmospheric air [4,5,6,7,8,11].

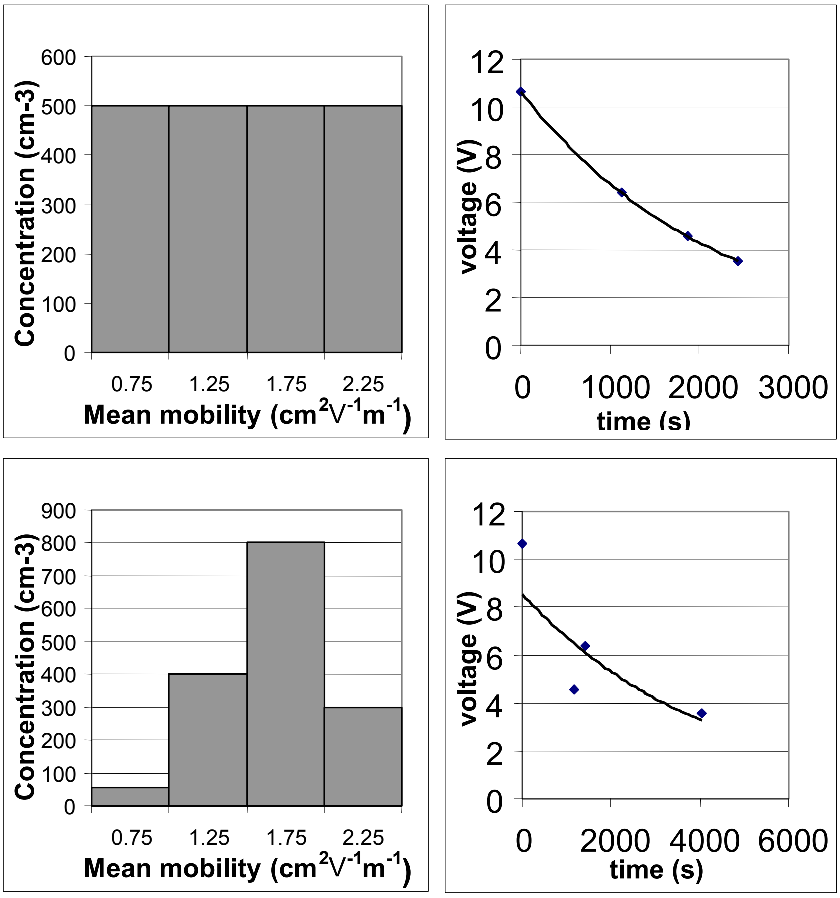

Figure 1 Sample ion mobility spectra with equivalent voltage decays. Ion mobility spectra (left) are simplified and replaced by four equal mobility bands, with the average mobilities indicated. Concentrations and mobilities are typical for surface atmospheric levels. Calculated voltage decays (right) assume a Gerdien condenser with dimensions given in [3]. a) A flat ion spectrum inverts to an exponential decay, c). b) a Gaussian ion spectrum inverts to a non-exponential decay, d).

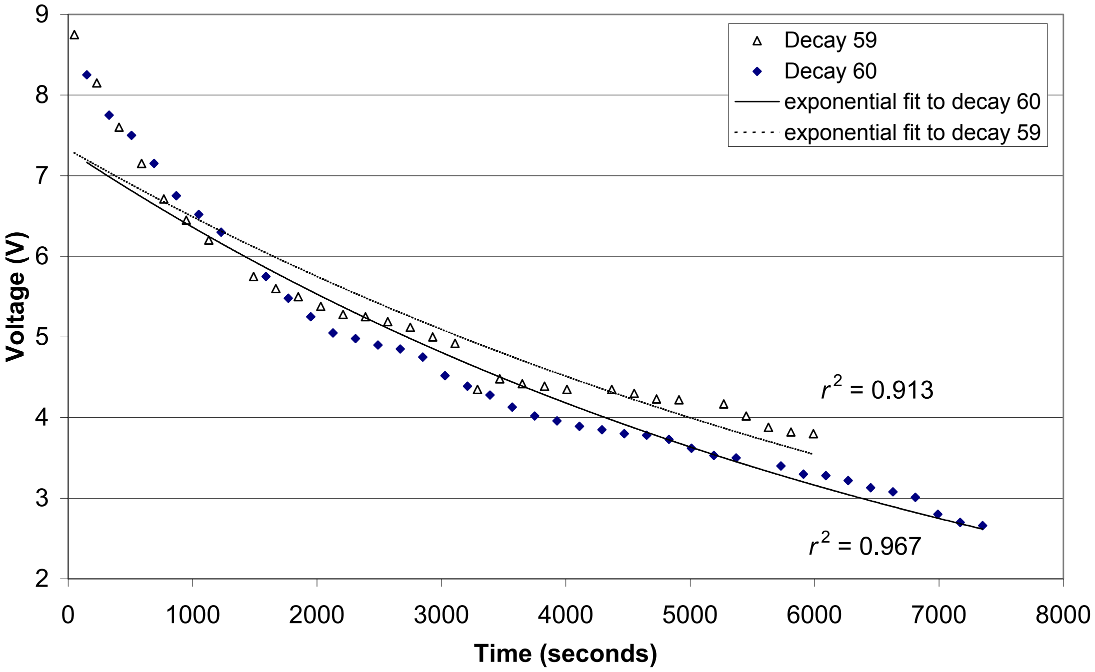

Figure 2 Consecutive voltage decay time series measured in atmospheric air on June 12 1998, showing fits to the classical exponential approximation, and coefficient of determination (r^2) values.

5 Computing ion spectra from voltage decay measurements

As the deviation of voltage decay curves from the exponential is related to the atmospheric ion spectrum, the shape of the voltage time series contains spectral information. Every voltage decay time series could therefore give an ion spectrum within the mobility range defined by the start and end voltage. Although (12) is not analytically soluble in the general case, it can be inverted using a finite difference solution, applicable for small changes, to calculate ion spectra from voltage decay measurements.

5.1 Numerical example

A cubic spline was used to interpolate between the measured values of voltage and time. They were then resampled at constant mobility resolution with the same number of points as the initial data set. (12) was then inverted to give a set of average mobilities and ion concentrations, with the voltage decay measurement, ventilation speed and cylindrical capacitor dimensions as inputs. If the original voltage decay has N points, the inversion yields a mobility spectrum with $N-1$ points. The highest voltage (lowest ion mobility) does not have a corresponding spectral point because it is used as the initial voltage V_0.

A flat ion spectrum with a mean concentration $<n>$ = 2000 ions cm^{-3}, a typical value for clean air, was transformed to obtain an artificial voltage decay using (12). The generated voltage and time values were then used to invert the data back to an ion mobility spectrum. $<n>$ calculated using this method was 1999 cm^{-3}, a mean error of 0.05% compared to the initial spectrum before transformation to a voltage decay; the maximum error in any spectral bin was – 0.3%.

Sets of random errors within conservative time and voltage limits of ± 10s and 0.01V were generated and added to the exponential voltage decay series. These errors were chosen to simulate the uncertainty associated with manually extracting points from a voltage decay time series plotted on a chart recorder. These errors are thought to be conservative as electronic data logging would usually be more reliable, measuring time and voltage up to an order of magnitude more accurately. The mean error in $<n>$ was + 0.32%, and the maximum error was + 2.3%. The inversion procedure can therefore calculate average ion concentrations from voltage decay time series to ± 0.5 %, and within a given mobility band the error is ± 2.5 %.

6 Application of the inversion to calculate conductivity

Ion mobility spectra were calculated from the two example voltage decays shown in Figure 2 using the method described in section 5.1. μ_c for each interpolated point was calculated from (9). Mean mobilities $<\mu>$ for each integrated strip were then determined by averaging consecutive critical mobilities. The conductivity σ was calculated by integration across the measured mobility range as in (1), for both decays, agreeing to 4%. Ion concentrations were normalised to mean ion concentrations $<n>$ using

$$< n >= \frac{\sigma}{e < \mu >} . \qquad (13)$$

The ion mobility range measured during a voltage decay is only related to the start and end voltage. As shown in Figure 2, Decay 60 ran for longer than Decay 59, reaching lower voltages, and therefore $<\mu>$ measured during the decay is higher. Since the conductivities were very close for the two samples, the increased $<\mu>$ for Decay 60 implied a lower $<n>$. Characteristics of the calculated ion spectra are summarised in Table 1.

Decay	***Max*** μ **(cm^{2}V^{-1}s^{-1})**	***Min*** μ **(cm^{2}V^{-1}s^{-1})**	$<\mu>$ **(cm^{2}V^{-1}s^{-1})**	$<n>$ **(cm^{-3})**	σ **(fSm^{-1})**
59	2.53	1.27	1.9	1842	56
60	3.62	1.35	2.49	1458	58

Table 1 Summary of spectral characteristics from consecutive voltage decay time series measurements in atmospheric air from June 12 1998

Typical urban ion concentrations are often lower than the values reported in Table 1. This is likely to be related to errors in absolute values of μ_c, particularly from estimating ventilation speed in the cylindrical capacitor in the presence of turbulent fluctuations in wind speed. Alternative expressions for (9) based on volumetric flow rate exist, but would be subject to similar errors. Further work is required to improve knowledge of the airflow in the cylindrical capacitor ion counter.

7 Conclusions

Modifying the classical theory of air conductivity measurement for voltage decays from an aspirated cylindrical capacitor has two principal advantages. Firstly, ion spectra can be retrieved from simple voltage time series if the instrument characteristics and ventilation speeds are known. Secondly, this spectral information can be used to calculate conductivity directly by integration rather than with simplifying assumptions, such as exponential decay, which introduce error. Previous ion spectrum measurements have usually required bias voltage switching and sensitive current sensing, requiring dedicated electronics and signal processing. The new method described here needs only the simplest logging equipment and is amenable to remote (*e.g.* balloon-borne) applications. Additional information can now be extracted from existing historical data sets.

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