

AEROSOL ELECTRICAL DENSITY: INTERPRETATION AND PRINCIPLES OF MEASUREMENT

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Introduction

The concept of aerosol electrical density was introduced in the paper [1] with an aim to help to use the atmospheric electrical measurements in the monitoring of environmental air pollution. A technique of measurement of aerosol electrical density is theoretically analyzed [2] and successfully tested in practical measurements [3]. However, to date no instrumentation specially designed to measure this quantity is available. All the above publications are in Russian and not too well-known in the scientific community at large.

The Gerdien tube and air pollution

The Gerdien tube is a well-known sensor of air electricity. It operates continually and gives an output signal proportional to the conductivity of the air or the concentration of small air ions for one polarity. The conductivity of the air is proportional to the product of the concentration and the average mobility of small air ions. Variations of the average mobility are small and in this paper we can consider the conductivity of the air and small air ion concentration as proportional quantities.

Aerosol particles adsorb small air ions and diminish their concentration. Some hygiene scientists use the Gerdien tube as an indicator of particulate pollution of the air. This application was introduced soon after Gerdien elaborated his sensor in the beginning of the century. Specifics of the Gerdien tube as an indicator of particulate pollution is its sensitivity to all particles including the smallest particles not counted by optical instruments.

Unfortunately, the Gerdien tube is also sensitive to the radioactive pollution of the environment. After the Chernobyl accident in 1986 the Gerdien tubes in all European atmospheric electricity stations indicated an increase in the air conductivity. The use of the Gerdien tube as the indicator of radioactive pollution is currently under discussion.

Thus, the signal of the Gerdien tube depends on two environmental factors. The particulate pollution of the air decreases, and the radioactive pollution increases its value. The signal of the Gerdien tube as an indicator of air pollution does not have an unambiguous interpretation.

The Allik factor of air pureness

Allik [4] made the first thorough analysis of the electrical conductivity of the air as an indicator of air pollution. He split the conductivity λ into the product of two factors:

$$\lambda = (eq) \cdot (k\tau), \quad (1)$$

where e is the elementary charge, q - the ionization rate, k - the average mobility, and τ the average lifetime of small air ions. The first factor eq characterizes the radioactive pollution of the environment and the second factor $k\tau$ the particulate pollution of the air. Allik denoted the second factor by R and called it the electrical factor of air pureness.

Allik analysed the Schweidler technique [5] of half-saturation voltage of a large ionization chamber to measure the lifetime of air ions. He demonstrated that the half-saturation voltage is inversely proportional to the factor R and that the Schweidler technique can be used to measure air pureness.

The concept of aerosol density

Aerosol density is a quantity making it possible to characterize the amount of particulate matter in the air with one number. There are various definitions of aerosol density useful in various applications. A more detailed characterization of an aerosol is given by the distribution function of number concentration over the particle size $f(d)$. We will not discuss the concept of particle size, instead using the effective diameter determined by a particle volume of $\pi d^3/6$.

If the distribution function $f(d)$ is given, various kinds of aerosol densities can be calculated. For instance, the number concentration or aerosol number density is

$$N = \int f(d) dd, \quad (2)$$

and the volume density is

$$V = \int (\pi d^3/6) f(d) dd. \quad (3)$$

Jaenicke [6] introduced the equation

$$\mu = \int \kappa_L(d) f(d) dd, \quad (4)$$

where $\kappa_L(d)$ is the weight function, to describe various integral effects of aerosol. It should be pointed out that aerosol densities are to be measured as certain integral effects of aerosol and formula (4) can be considered as the definition of the generalized aerosol density.

Specific weight functions $\kappa_L(d)$ should be defined for every concrete kind of aerosol density μ . In the case of the above examples $\kappa_N = 1$ and $\kappa_V = \pi d^3/6$. An additional example: if $\kappa = Q_{\text{ext}}(d)(\pi d^2/4)$, we get the coefficient of light extinction which can be considered as the aerosol optical density.

The balance of small air ions

The simplest mathematical model to describe the balance of small air ions is the equation

$$\begin{aligned} dn_+/dt &= q - \alpha n_+ n_- - g_+ n_+ \\ dn_-/dt &= q - \alpha n_+ n_- - g_- n_-, \end{aligned} \quad (5)$$

where n_+ and n_- are the concentrations of small air ions, t - the time, q - the ionization rate, α - the coefficient of recombination, g_+ and g_- - the coefficients describing the adsorption of small air ions on the aerosol particles.

The coefficient α has a quite stable value about $1.5 \cdot 10^{-12} \text{ m}^3/\text{s}$. The coefficients g are variable, but their ratio g_+/g_- is quite stable and close to the ratio of small air ion mobilities k_+/k_- . The value of this ratio in natural atmospheric air is about 0.87 according the long-term observations [7]. We define

$$g = (g_+ + g_-)/2 \quad (6)$$

and accept the approximation $g_+ = g_- = g$ below. The approximation allows us to describe the steady state ($q = \text{const}$) by the equation

$$q = \alpha n^2 + gn, \quad (7)$$

where $n = n_+ = n_-$. The time constant of the transfer to steady state is about one minute in tropospheric air.

The second term of equation (7) is dominant in tropospheric and indoor air. As an average for the natural near-ground air the first term αn^2 describes 3% of the loss of small air ions.

Definition of the aerosol electrical density

The coefficient g in formula (7) characterizes the adsorption of small air ions in a similar way as the aerosol optical density characterizes the adsorption of light. Therefore, we call this coefficient the aerosol electrical density.

For the interpretation of aerosol electrical density from the point of view of possible applications it is necessary to determine the specific weight function $\kappa_g(d)$.

To calculate the adsorption of small air ions, the term gn in equation (7) is to be expressed through the coefficients of attachment of small air ions by particles of a certain size and preliminary charge.

Let us use $\beta_q(d)$ to denote the partial attachment coefficient of a positive small air ion to a particle with the size d and charge qe , where q is number and e is elementary charge. We hope the double use of the letter q to denote the ionization rate, or to count the elementary charges will not mislead the reader.

On the assumption of charge symmetry, the coefficients of the attachment of a positive small air ion to a particle with the charge qe and of a negative small air ion to a particle with the charge $-qe$ are equal. The error of this approximation is equal to the error of the approximation $g_+ = g_-$.

Let us use $\rho_q(d)$ to denote the probability of the charge qe on a particle with the size d .

The number concentration of particles with the charge qe and size $d \dots d+dd$ is $\rho_q(d)f(d)dd$ and the adsorption of small air ions by these particles is $\beta_q(d)\rho_q(d)f(d)dd$. The total adsorption gn is the sum over all charges and the integral over all sizes:

$$gn = n \int \sum_q \beta_q(d)\rho_q(d)f(d)dd. \quad (8)$$

Comparing this equation with the expression of generalized aerosol density (4) it can be found that

$$\kappa_g(d) = \sum_q \beta_q(d)\rho_q(d). \quad (9)$$

Functions $\beta_q(d)$ and $\rho_q(d)$

In the region of Knudsen small numbers ($d > 200$ nm) the first formula of Fuchs [8] has a good theoretical and experimental foundation

$$\left. \begin{aligned} x &= qe^2/(2\pi d\epsilon_0 kT) \\ \beta_q(d) &= 2\pi dDx/[\exp(x) - 1]. \end{aligned} \right\} \quad (10)$$

k is the Boltzmann constant and T the temperature. The diffusion coefficient of small air ions is expressed by the Einstein formula

$$D = kT/k, \quad (11)$$

where k is the electrical mobility of small air ions. At an average mobility of small atmospheric ions [7] $k = (k_+ + k_-)/2 = 1.45 \cdot 10^{-4}$ m²/(V·s) and $T = 290$ K we have $D = 3.8 \cdot 10^{-8}$ m²/s and $x = q(115 \text{ nm} / d)$.

A large number of papers is dedicated to theoretical and experimental investigation of the function $\beta_q(d)$ in the region of Knudsen large and intermediate numbers. There are no simple formulas and the results are expressed as sophisticated algorithms or numerical tables. We consider the results of Hoppel and Frick [9] to be reliable. It is possible to approximate the tabulated results of Hoppel and Frick with a simple formula

$$\beta_q(d) \approx (1 - \frac{2}{Z + q(q-1) + d/10\text{nm}}) \beta_q^*(d), \quad (12)$$

where $\beta_q^*(d)$ is calculated from formula (10). The mean square relative error of the approximation (12) over the tables of Hoppel and Frick is 5% (very small and zero values are not considered).

The function $\rho_q(d)$ for the steady state can be calculated in the known way using the equations of balance for q -charged fractions of particles:

$$\beta_{-q}(d)N_q(d) = \beta_{q-1}(d)N_{q-1}(d). \quad (13)$$

A recurrent use of this equation beginning from the conventional value $N_0(d) = 1$ gives

$$N_j(d) = \frac{\beta_0(d) \cdot \beta_1(d) \cdot \dots \cdot \beta_{j-1}(d)}{\beta_{-1}(d) \cdot \beta_{-2}(d) \cdot \dots \cdot \beta_{-j}(d)} \quad (14)$$

and

$$\rho_q(d) = N_q(d) / (1 + 2 \sum_{j=1}^{\infty} N_j(d)). \quad (15)$$

Interpretation of aerosol electrical density

The first interpretation of aerosol electrical density is given by its definition as the coefficient of adsorption of small air ions. This interpretation is useful in air electricity.

The second interpretation follows from the character of the weight function $\kappa_g(d)$. The function calculated according to formula (9) is depicted in the Figure. The main conclusion from the analysis of the curve is that the linear diffusion asymptote $\kappa = 2\pi dD \approx d/40$ nm is a perfect approximation for all sizes over 100 nm. An improved approximation

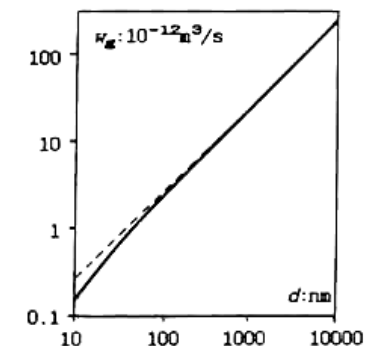


Figure. The weight function of aerosol electrical density and its linear asymptote.

$$\kappa_g(d) = \frac{d - 1 \text{ nm}}{d + 5 \text{ nm}} \cdot \frac{d}{40 \text{ nm}} \cdot 10^{-12} \text{ m}^3/\text{s} \quad (16)$$

deviates from the value calculated according to formula (9) by less than 5% in the whole range of particle sizes $d > 3$ nm. In the case of a standard distribution of atmospheric aerosol [10] the calculation according to the linear approximation $d/40$ nm gives a value of g 8% higher than calculation according to the precise approximation (16). This makes it possible to use the linear approximation with the correction factor 1.08 and write

$$g \approx (2.3 \cdot 10^{-8} \text{ m}^2/\text{s}) \cdot N_d, \quad N_d \approx (4.3 \cdot 10^4 \text{ s/m}^2) \cdot g, \quad (17)$$

where

$$N_d = \int d f(d) dd \quad (18)$$

is the linear size density of aerosol. The standard small air ion mobility $1.45 \cdot 10^{-4}$ m²/(V·s) is assumed in (16) and (17).

In the case of spherical particles, N_d indicates the length of the chain composed of all particles in a volume unit. A typical tropospheric aerosol contains about 1 km of particles in 1 m³ and its electrical aerosol density is about 0.02 s⁻¹.

Two popular measures of aerosol density are the number concentration ($\kappa = d^0 = 1$), and the mass density proportional to the volume concentration ($\kappa = \pi d^3/6$). The number concentration is determined mainly by the smallest particles; the large particles with sizes over 1 μm form a negligible part of its value in atmospheric air. It can be considered as aerosol density only if the effect of a particle in an application is independent of the

size. The large particles are dominant in a volume concentration where the small particles play a minor role. The linear size concentration is between these two popular measures of aerosol and can be used as aerosol density in applications where the effect of a particle is approximately proportional to its linear size.

Measurement with an ionization chamber and a Gerdien tube

Measurement with an ionization chamber directly gives the value of the ionization rate q . If we additionally have a Gerdien tube and measure small air ion concentration n (for one polarity), we can use equation (7) and calculate

$$g = q/n - an. \quad (19)$$

The second term in the formula can be considered as a small correction in normal tropospheric conditions.

A fundamental problem in the use of the above technique is the spatial inhomogeneity of the ionization in the real atmosphere. The ionization chamber gives us the value of q in a small delimited volume. The signal of the Gerdien tube depends on the ionization rate in a large volume with no definite boundaries. The size of the volume is determined by the path of the movement of the free external air during the lifetime of small air ions (about one minute). The values of q in equation (19) and measured using an ionization chamber are not equal. This results in a methodical error the value of which cannot be easily estimated.

The above discussion points to a specific advantage of the Gerdien tube in radioactivity monitoring as an integrator over considerable area and all kinds of radiations. The advantage can be correctly used only if the aerosol electrical density is measured simultaneously.

A practical problem in the use of an ionization chamber is the complicated handling of the equipment. We do not know any realistic way to build a reliable and easily handled device containing an ionization chamber to measure the natural ionization rate.

Measurement with two Gerdien tubes and a standard ion generator

The instrumentation consists of one Gerdien tube installed in a standard mode and another tube equipped with a standard air ion generator and a volume for the ageing of the air between the ion generator and the inlet of the Gerdien tube. The standard air ion generator should provide a stable additional ionization of a known value. It is possible to build such an ion generator using a Sr^{90} preparation with a very low activity of about 1 μ Ci which does not need any safety precautions.

The small air ion concentrations in the standard Gerdien tube n_0 and in the equipped Gerdien tube n_w depend on the environmental parameters q , g , and on the parameters of the air ionizer and the ageing volume $p1$, $p2$, ... :

$$\left. \begin{aligned} n_0 &= f_0(q, g) = (\sqrt{g^2 + 4aq} - g) / (2a) \\ n_w &= f_w(q, g, p1, p2, \dots) \end{aligned} \right\} \quad (20)$$

If n_0 , n_w , $p1$, $p2$, ... are known, the system of equations (20) can be solved and the values of the variables q and g determined.

Theoretically, the simplest way to realize the described principle is to have a uniform additional ionization q_w in the ageing volume and an ageing time sufficient to assume the steady state on the inlet of the Gerdien tube. In this case the second equation in (20) differs from the first by the replacement of q with $q + q_w$ and we get the solution

$$g = [q_w - a(n_w^2 - n_0^2)] / (n_w - n_0). \quad (21)$$

The parameters of the air ion generator should be chosen according to the criteria of minimum measurement error. For instance, if we assume

$$q = 10 \text{ cm}^{-3}\text{s}^{-1}, g = 0.02 \text{ s}^{-1} \text{ and } \Delta n = 1\% + 10 \text{ cm}^{-3} (\Delta n_0/n_0 = 3.1\%),$$

the analysis of formula (20) gives the following dependence :

q_w	20	50	100	200	500	1000	2000 $\text{cm}^{-3}\text{s}^{-1}$
$\Delta g/g$	5.1%	3.2%	2.7%	2.8%	3.4%	4.4%	5.9%

The example demonstrates that the right choice of the parameter q_w guarantees a well-conditioned system of equations (20) without the effect of "error amplification" by data processing.

The simplest way considered above as an example is not expedient in practice. There are more reasonable technical solutions where air ions are generated only near the input aperture of the ageing volume and the time of ageing is about 20 seconds. Some results of theoretical analysis of more sophisticated techniques are published in [2] and results of testing the technique in [3].

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Citation:

Tammet, H. (1991) Aerosol electrical density: interpretation and principles of measurement. Report Series in Aerosol Science, 19, 128-133.