

FAIR-WEATHER ELECTRICITY ON GROUND LEVEL

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ABSTRACT: The paper presents a survey of studies of identification of small air ions, determination of air ion mobilities, electricity of atmospheric aerosols, electrical structure of surface layer, atmospheric electrical observations and electrical methods of indication of air pollution. It is pointed out that the application of atmospheric-electricity methods in monitoring of natural environment might become pivotal in further studies of fair-weather atmospheric electricity.

INTRODUCTION

The Soviet journal of abstracts on geophysics for the year 1987 reviews 59 Soviet and 48 other publications on atmospheric electricity, 13% of these are devoted to fair-weather atmospheric electricity on ground level. Attention to our topic could also be characterized by the following figures:

Conference on atmospheric electricity	Percentage of papers
Garmish-Partenkirchen 1974	26%
Uppsala 1988	14%
Leningrad 1973	24%
Tartu 1986	25%

The high percentage at the Tartu symposium is explained by the large number of reports on the electricity of aerosols. Disregarding those papers the percentage would be 17%.

The start of systematic research of fair-weather atmospheric electricity is dated with 1785 May 28, 6^h32'30" (Coulomb, 1785). Below some contemporary problems of this field will be considered with special attention to the results obtained in USSR.

COMPOSITION OF SMALL AIR IONS

The composition, structure and properties of small air ions are studied in the physics of gas discharges or plasma physics as well as in chemical physics and atmospheric electricity. A survey of the physics of small air ions can be found in a book by Smirnov (Смирнов, 1983). The basic methods of the study of the composition of small air ions are mathematical simulation of the kinetics of ion-molecular reactions and identification of air ions with mass spectrometer.

The principles of mathematical simulation of the kinetics of ion-molecular reactions are thoroughly covered in the literature (cf. e.g., Banks, Kockarts, 1973; Kawamoto, Ogawa, 1986). Computational difficulties ensue from great dimensions and rigidity of the systems of differential equations. Rakitski's method of exponential matrix (Ракицкий, 1979) has mitigated these difficulties. The most important limitation is the lack of reliable data about the concentrations of trace gases and about the rates of reactions with the ions of exotic compositions, vital in the formation of air ion spectrum in real troposphere. Calculation results predict that the most frequent negative air ions on ground level should be $\text{NO}_3^- \cdot \text{HNO}_3 \cdot \text{H}_2\text{O}$ and $\text{O}_2^- \cdot (\text{H}_2\text{O})_n$ dominated by

$n = 4$ and 5 . In addition $\text{NO}_2^-\cdot\text{SO}_2$, $\text{CO}_3^-\cdot(\text{H}_2\text{O})_n$ and some others are possible. Mobilities of these ions agree with observational data (Tammet, Salm, Luts, Iher, 1988). In the case of positive air ions calculations and observations are in conflict. Calculations predict the concurrence of series $\text{H}_3\text{O}^+\cdot(\text{H}_2\text{O})_n$ and $\text{NH}_4^+\cdot(\text{H}_2\text{O})_n$ with relatively low values of n , as well as the presence of $\text{NH}_4^+\cdot\text{NH}_3\cdot\text{H}_2\text{O}$ ions. The mobilities of these air ions, however, are too high to explain the observations in real troposphere. The possible reason for this contradiction could be the fact that the pyridine bases and large symmetrical water clathrates (pointed out already by Siksna (1973)) have not been taken into account in the calculations because of the lack of data.

An extensive survey of experimental studies of small air ions can be found in (Märk, Castleman, 1985). Most of the experimental results have been obtained in laboratories or in the upper atmosphere. Mass-spectrometric studies of natural air ions on ground level are technically difficult to accomplish because of high pressure and contamination of air, and low concentration of ions. Equipment suitable for the study of natural air ions on ground level has been created in recent years (Eisele, 1983, 1986). In Eisele's apparatus a collisional dissociation chamber (CDC) was inserted between the air input and the high-vacuum analysis region. At a pressure of about 0.1 torr the air ions make 10-100 collisions with molecules of neutral gas. The energy of the collisions is controlled with an electric field. At intermediate energies the air ion is released from the molecules of water and the core of the ion gets into the mass-spectrometer. Collisions with high energies make it possible to split air ions into fragments. This substantially enhances the possibility to identify the air ion composition. Usually a great number of possible air ion compositions correspond to one value of the mass. The values of the core mass and the mass of some fragments give important additional information. Already the first observations of natural small air ions (Eisele, 1986) have given significant new results. Observations allow interpretation on the basis of the hypothesis that the cores of a considerable part of natural positive small air ions are pyridine-bases (pyridine, picoline, lutidine). Skilful applications of CDC has given the hypothesis a solid foundation. The described result cannot be theoretically predicted because before Eisele's observations there were no data about the presence of pyridine bases in the atmosphere. These observations give the first estimate of background concentrations of the above substances in the air.

AIR ION MOBILITY

The mobility of small air ions could in principle be calculated according to the kinetic theory of Chapman and Enskog presented, e.g., in a book by Mc Daniel and Mason (1973). To carry out the calculations it is necessary to know the potential of the interaction between the air ion and the molecule of the surrounding gas. In principle the interaction potential could be calculated by the methods of quantum chemistry which have already given practical results at calculations of geometrical structure of small air ions (Контуш et al., 1986). Yet we cannot predict quick success for this way of mobility calculations. An expert opinion about the difficulties of theoretical calculation of interaction potentials can be found in (Wells, Wilson, 1983): "Coulson has compared the calculation of the chemical bonding energies by 'ab initio' with the determination of the weight of a ship's captain by weighting the ship first with and then without captain on board. Kutzelnigg has pointed

out that the calculation of Van der Waals interaction potentials is comparable with the determination of the weight of the captain's cap in an analogous manner".

Classical formulae coinciding with particular expressions of the Chapman-Enskog theory in the case of simplest model potentials or empirical formulae are practically used for mobility calculation. The Langevin formula corresponds to polarization interaction with the potential $\text{const} \cdot r^{-4}$ and the Epstein formula corresponds to the model of rigid spheres. In the case of large ions Millikan's universal empirical formula is used. The formula has been confirmed by calculations (Annis, Malinauskas, Mason, 1972). At $r \rightarrow 0$ Millikan's formula reduces to Epstein's formula and can be used also for the calculation of small ion mobilities if we will use a special interpretation of the particle size and add the fourth term describing the loss of accommodation of molecules on particle surface in the range of small air ions. The universal mobility formula with respective modifications can be written as:

$$k = \frac{e\gamma\lambda\sqrt{1+m/M}}{6\pi\eta\delta} \left[\frac{\delta}{\gamma\lambda} + a + b e^{-\frac{c\delta}{\gamma\lambda}} + \left(\frac{g}{2} - a - b\right) e^{-\left(\frac{\delta}{\delta_0}\right)^\nu} \right],$$

where e is the ion charge, $\gamma\lambda = (\eta/n)\sqrt{(\pi/8mKT)}$, η , n are the viscosity and density of the gas, m , M are the masses of gas molecule and ion, K is the Boltzmann constant, T is the temperature, $\delta = \sqrt{\Omega/\pi}$ is the effective transport size, $a = 2.8$, $b = 0.7$, $c = 0.45$ are empirical constants. In the case of large particles δ is the geometrical radius. For the air in normal conditions $\eta = 1.72 \cdot 10^{-5} \text{ kg/(m}\cdot\text{s)}$ and $n = 2.69 \cdot 10^{25} \text{ m}^{-3}$. The characteristic size of transition from elastic collisions to thermal accommodation δ_0 and the slope of the transition ν are to be determined by the criterion of best fitting of empirical data. If to take as a basis Kilpatrick's (1972) experimental mass-mobility table and the simplest approximation $\delta = \alpha + \beta\sqrt{M:u}$ (u is the atomic mass unit) and to choose α , β , δ_0 , ν by minimizing the error of the above approximation, then $\alpha = 0.41 \text{ nm}$, $\beta = 0.055 \text{ nm}$, $\delta_0 = 0.97 \text{ nm}$ and $\nu = 10$. Whereas the standard error of approximation of δ is 1.2% and the standard error of restoration of mobilities by the mass in Kilpatrick's table is 2.7%. It should be pointed out that the above value of β corresponds to the density of the substance condensed out of air ions 1.2 g/cm^3 and the size δ_0 is close to the physical boundary between clusters and small particles (cf. Петров, 1986).

Among possible methods of air ion spectrum measurement described in the book (Комаров, 1977) the methods of drift tube and aspiration capacitor find practical application. Being more sensitive the aspiration capacitor is preferred in low-concentration natural air ion research. Best results are yielded by differential capacitors of the second order with many parallel measuring electrodes. In the mobility range of small air ions it is possible to resolve up to 10 fractions. An advanced aspiration capacitor was built already by Yunker (1940) but the lack of flexible control means did not allow systematic observations. Sufficient automatic control means were adopted in recent years (e.g., Tammet et al., 1987). The observations show considerable time and space variations. The mobility spectrum is influenced by small impurities in the air, the full inventory of which is yet unknown. A summary of the results of systematic annual observations of small air ion mobility spectrum at a rural site is presented in (Tammet et al., 1988).

AEROSOL ELECTRICITY

Aerosol electricity has the same meaning as physics of large and intermediate air ions. As was the case with small air ions, the problems of large and intermediate ions do not inherently belong to the science of atmospheric electricity. These problems are investigated in the aerosol science also apart from atmospheric electricity. The basis of the studies is general knowledge about aerosol physics most authoritatively stated in Fuchs' book (1964).

The following current problems of aerosol electricity in tropospheric air can be distinguished:

- explanation of the mechanism of nucleation on small air ions and its role in the nature,
- quantitative description of the attachment of small air ions to particles and the formation of charge distribution of particles,
- elucidation of the role of photoeffect in atmospheric electricity,
- development of methods of electrical aerosol measurement.

Nucleation on small air ions has been investigated since the studies of Wilson. A contemporary presentation of the theory of water condensation on small air ions can be found in (Suzuki, 1987). The role of the condensation of water on small air ions in real atmosphere is negligible. Cabane et al. (1978) refer to the connection between condensation on air ions and air pollution. Latest observations (Tammet et al., 1988) show that small impurities in the air may condensate on small air ions. The chemical nature of those impurities was not identified.

The problem of attachment of small air ions to particles has a central role in the electricity of aerosols. Contemporary theoretical foundations were laid in papers (Натансон, 1960; Fuchs, 1963). Fuchs' theory is founded on physical approximation definable with the limiting sphere. Experimental verification (Hussin et al., 1983) confirms the high accuracy of Fuchs' approximation. It is difficult to obtain practical results on the basis of more rigorous theories. The most complete calculations have been carried out by Hoppel and Frick (1986). By the present time there has emerged a relatively complete and accurate picture of attachment processes of small air ions to particles in the lack of external electric field. The presence of external electric field, which is vital in many tasks, complicates the problem. A solution has been pointed out in (Мирзабекян, 1966), more complete results have been obtained by Noppel (1985).

Photoeffect on atmospheric aerosols has been known for a long time but it has not been actively discussed. Only few studies (e.g., Суслов, Позигун, Контуш, 1986) solve some particular physical problems. The quantitative role of aerosol photoeffect in real atmosphere has not been clarified.

Empirical information about fine aerosols in the atmosphere is essential in several branches of atmospheric physics. Counters of condensation nuclei do not satisfy the need for information about the size-distribution of particles. Junge (1963) has noted that most valuable information about fine aerosols in the atmosphere has been obtained by the analysis of observations of large air ions. At the present time an electric analyzer produced by the TSI has taken the lead (Liu, Pui, Kapadia, 1979). This is an air ion mobility spectrometer equipped with a device for preliminary unipolar charging of particles which increases the sensitivity of the apparatus. One of the most advanced apparatus (Mirme et al., 1984) has two differential aspiration capacitors of the second order equipped with various devices for preliminary charging of particles.

ELECTRICAL STRUCTURE OF SURFACE LAYER

The surface layer is the lowest level of the boundary layer of the atmosphere. There turbulent and convective mixing of the air is the basic factor determining electrical processes in fair weather. A survey of atmospheric electricity in the boundary layer is given in (Hopfel, Anderson, Willett, 1986). A specific feature of the surface layer is the electrode effect. Correct theoretical solution of the problem of the electrode effect in the case of linear and power dependence of the coefficient of turbulent exchange on the height has been obtained already in (Тверской, Тимофеев, 1948; Тверской, Милин, Феодоров, 1953). The most advanced theoretical treatment of the electrode effect has been presented in (Морозов, 1986). Experimental results (Norinder, 1921; Suzuki, Tsutsumi, 1985; Петров, Петрова, 1986) agree with the theory. Exact comparison of the theory with experiments is not easy due to the insufficiency of direct data about turbulent mixing. This points to the fact that the measurements of the electrode effect can be looked upon as a source of experimental information about turbulence. Already in 1955 a method for the measurement of the coefficient of turbulent exchange by the profile of the electric field has been proposed and analysed (Милин, 1955). In the same paper Milin discusses some special methods of turbulence measurement involving the use of artificial air ion sources. Milin's proposals deserve attention in the contemporary stage of the development of atmospheric electricity.

New developments in technology have enabled us to measure the parameters of statistical distribution of fluctuation of atmospheric electricity which contain significant information about electrical processes in the surface layer and the boundary layer. Because of the absence of such possibilities the classical studies have considered fluctuations as noise. Ruhnke (1969) has pointed out the possibility to smooth the fluctuations, looked upon as noise, by increasing the size of the measurement antenna. New results (Stening, Ogawa, 1985) have shown that the effect of long antenna is smaller than expected. It is possible that the contradiction between the above results ensues from the differences in stratification of the atmosphere in experiments.

It is necessary to keep in mind that turbulence and convection generate fluctuations on non-uniformities of two kinds. The first is the classical non-uniformity of the electrode effect. Electrical non-uniformity of the second kind emerges as a result of space charge accumulation on clouds of aerosol according to the mechanism described by Hansen (1935).

The results of a quantitative study of signal fluctuation of atmospheric-electricity antennas (Anisimov, 1988) have shown that the frequency spectrum of field strength fluctuations is proportional to $f^{-5/3}$ whereas the frequency spectrum of the Maxwell current conforms to the law $f^{-2/3}$. This result was considered as a consequence of Kolmogorov's $2/3$ -law.

The simplicity of recording the Maxwell current allows us to attempt to use the statistical parameters of the signals of the antennas of various lengths for obtaining information about the structure of the boundary layer. In this case ionospheric variations recorded mainly in high latitudes (Анцев et al., 1988) are considered to be noise.

REGULAR OBSERVATIONS OF FAIR-WEATHER ELECTRICITY

A quantitative picture of meteorological phenomena can be obtained by an observational network operating during a long period. This is equally true in the case of atmospheric electricity. The development of

the network depends on economic resources and as a result the scope of observations depends on public interest in the results. As a matter of fact, the interest in atmospheric-electricity observations is a great deal smaller than in ordinary meteorological observations. In USSR the network for regular atmospheric-electricity observations was set up in 1957. It comprises 8 stations (Шварц, Огуряева, 1987). The measurement results obtained by this network make up about half of the data arriving at the World Data Centre, Leningrad. The World Data Centre gathers and systematizes data about fair-weather electricity and spreads it in regular bulletins.

The development of atmospheric-electricity observations is determined by various factors of which the following should be pointed out:

- formulation of scientific and practical aims of observations in a convincing and comprehensible form,
- development of the measurement technology directed to minimizing the costs,
- progress of the technology for data processing and for its transmission to and from the Centre.

The classical aims of atmospheric-electricity observations determine the funds allotted for the observations. Apparently funding can be increased only if principally new aims emerge. Such aims might appear if the observations of fair-weather electricity are included into the system of environmental monitoring. Certain preconditions for this inclusion already exist. For instance air ionization by radiation from krypton-85 is accepted as an object of environment monitoring (Израэль, 1984).

Basically the measurement technology used in atmospheric-electricity observations has stayed on the level described in Imyanitov's fundamental book (ИМЯНИТОВ, 1957). Nevertheless there are some ideas which hopefully will be elaborated into considerably cheaper and reliable sensors of field strength and air conductivity (e.g., Струминский, Татарин, 1984; Таммет et al., 1986). However, these ideas are not yet technical solutions ready to be industrially manufactured. Insufficient use is made of modern electronics which could facilitate automation of measurements. Attempts at automation of regular observations have been merely experimental (e.g., Шварц et al., 1981). Reorganization of data transmission and processing must be brought about primarily by writing data in a computer-readable form directly at observation stations. If this is not done the amount of labour needed for preliminary handling of data cannot be reduced. Another requirement is data exchange on standardized carriers. The standard chosen for the purpose should be generally available and conservative e.g., floppy disks readable by all IBM PC-compatible computers.

INDICATION OF AIR POLLUTION

The connection between small air ion concentration and air pollution has once been one of the arguments for the hypothesis stating the insufficiency of small air ions is harmful to health. Now and again the hypothesis about direct biological influence of small air ions has aroused considerable interest among both scientists (Минх, 1963; Dolezalek, Reiter, Kröling, 1985) and public (Soyka, 1977). However up to now there are no crucial experimental proofs of biological effect of small air ions.

The value of the analysis of air conductivity observations for the description of aerosol pollution of the air has been convincingly prov-

ed in (Шварц, Огуряева, 1987). Nevertheless conductivity cannot be viewed as an unambiguous pollution indicator as it includes both reverse dependence on aerosol pollution and direct dependence on radioactive pollution. This is demonstrated by observations (Israelsson, Knudsen, 1986; Warzecha, 1987). There are also certain other difficulties discussed by Reiter (1984).

Physically correct setting of the problem of interpretation of air electricity measurements from the point of view of air pollution can first be found in the fundamental work by Allik (Аллик, 1941). His basic idea is simple. If we write the simplified equation of air ion balance $dn/dt = q - \alpha n^2 - gn$ and suppose $\alpha n^2 \ll gn$, then the stationary air ion concentration is presented as the product of two factors $n = q \cdot (1/g)$. Allik calls the second factor the electric factor of air purity. The problem has been repeatedly discussed (Махоткин, 1973; Смирнов, 1983; Таммет, 1979, 1986). Two last papers describe a measurement method with simultaneous use of two aspiration capacitors. One of them has an etalon air ion source and a chamber for ageing ionized air before measurement. The apparatus is described by two equations with two unknowns q and g . The solution of the equations at 1% errors of direct measurements yields the values of q and g with errors of about 3%. An experiment (Семенов, Соколенко, Шварц, 1986) has demonstrated a good agreement between the obtained results and of simultaneous measurements with an ionization chamber. The asset of the new method is in the simplification of apparatus maintenance. Results of measurements can be interpreted as follows.

The ionization rate q is a cumulative measure of ionizing radiation. It takes into account all the sources: radioactive aerosols, radioactive gases, radioactivity of the earth and cosmic radiation. The traditional measurement unit $1 \text{ J} = 1 \text{ e}/(\text{cm}^3 \cdot \text{s})$ is equivalent to $1.73 \text{ } \mu\text{R}/\text{hour}$ at normal density of the air.

The coefficient g can be called aerosol electric density. It gives good approximation for the aerosol diameter concentration $v_d \approx (54000 \text{ s}/\text{m}^2)g$ (Таммет, 1984). $v_d = \int 2rf(r)dr$, where $f(r)dr$ is the numerical concentration of the particles in the interval of radii dr . The value of v_d in natural air is about 1000 m^{-2} and by way of popular interpretation we could say that a chain made of particles contained in 1 m^3 of air has a length of about 1 km. In comparison with the traditional numerical concentration, v_d better characterizes the danger of air pollution both from the viewpoints of ecology and medicine as well as of technology.

There are other atmospheric-electrical quantities which can be interpreted as environmental pollution indicators. For instance, the resistance of the vertical column describes atmospheric aerosol integrated over the height and unlike the respective optical quantity it covers also fine particles. If we could set up a system for monitoring of ionospheric potential then the measurement of the vertical column resistance would become a simple technical matter.

Clarity and comprehensibility of the interpretation of environmental parameters measured with atmospheric-electrical methods is a factor of public interest in the development of fair-weather atmospheric electricity research.

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