Soviet-era science, translated into English

ON DIFFUSIONAL CHARGING OF DROPLETS IN CLOUDS AND FOGS

GEOPHYSICS

1967

SovietRxiv

View the original and related papers at https://sovietrxiv.org/items/ru-196701.98052

Source: Math-Net.Ru and CyberLeninka. Machine translation. Verify with the original.

SovietRxiv

Abstract

Full Text

UDC 537.24+551.594.25

GEOPHYSICS

N. S. SHISHKIN

ON DIFFUSIONAL CHARGING OF DROPLETS IN CLOUDS AND FOGS

(Presented by Academician E. K. Fedorov on 5 I 1967)

The question of the diffusional charging of droplets in clouds and fogs was considered in 1944 by Ya. I. Frenkel (^{3}). He obtained formulas for the influx of ions into a droplet of radius r, having charge Q,

$$I_+ = \frac{4\pi n_+ u_+ EQ}{e^{ED/rkT} - 1}, \qquad I_- = -\frac{4\pi n_- u_- EQ}{1 - e^{-ED/rkT}}, \tag{1}$$

where n_+ and n_- are the concentrations of ions in the space surrounding the droplet; u_+ and u_- are the mobilities of the ions; E is the ion charge (all ions are assumed to be singly charged).

In the equilibrium state the currents must be equal in magnitude, and the charge of the droplet proves to be equal to

$$Q_0 = \frac{rkT}{E} \ln \frac{\lambda_+}{\lambda_-} \tag{2}$$

where $\lambda_+ = n_+ u_+ E$, $\lambda_- = n_- u_- E$ are the conductivities of the air. If $EQ/rkT \ll 1$, then

$$Q_0 = \frac{2rkT}{E} \frac{\lambda_+/\lambda_- - 1}{\lambda_+/\lambda_- + 1}. \tag{3}$$

It follows from formulas (2) and (3) that when $\lambda_+/\lambda_- = 1$ the equilibrium charge of the droplet is zero. Similar formulas were obtained in 1954 by R. Gunn (^{6}).

These results do not agree with the experimental data. Already at the time when Ya. I. Frenkel’ s paper (^{3}) was written it was known that water droplets become charged predominantly negatively. Ya. I. Frenkel supposed that the reason for this was the presence of a double electric layer on the surface of the droplets. He considered that equilibrium charging is attained under the condition that the potential jump $\xi = 0.26$ V (^{5}) of the double electric layer, in which the negative charges are located externally, is compensated by the Coulomb field of the droplet. In this case equilibrium values of the droplet charge were obtained that exceed the experimental values by an order of magnitude. R. Gunn considered that the predominantly negative charging of droplets is connected with the greater mobility of negative atmospheric ions.

In 1958–1959 A. P. Sergieva (1), under the direction of E. K. Fedorov, carried out detailed investigations of the charges of droplets of natural and artificial fogs and established that, in the initial period of their existence, rapid negative charging of 10–30% of the droplets usually occurs. Subsequently the fraction of negatively charged droplets changes little, but gradual positive charging of neutral droplets takes place, so that 30–40 min after fog formation the concentration of charged droplets reaches 40–60% of their total number. Only when the conductivity ratio before fog formation is $\lambda_+/\lambda_- \geq 2$ does predominantly positive charging of droplets occur in the initial period of fog existence.

To explain the experimental data, let us consider in greater detail a possible mechanism of the influence of the double electric layer on the charging of droplets.

Atmospheric ions approaching the surface of a droplet by diffusion transfer their charge predominantly to neutral water molecules of the surface layer (or they are adsorbed by the droplet).

When a negative charge is attached, the particle moves into the liquid owing to the electric field of the double layer, whose thickness, according to T. Olte (4), is $5 \cdot 10^{-7}$ cm. By contrast, in the case of conversion of a neutral water molecule into a positive ion, the double layer hinders the migration of the ion into the liquid. Some of the positive ions may be fixed on the surface owing to solvation, while another part may leave the surface in the process of evaporation, together with neutral water molecules.

Let us suppose that, of the total number N_+ of ions adsorbed per unit area, some fraction χ of ions leaves per unit time. For a small droplet charge Q, one may write, expanding in series the denominators in expressions (2) and retaining terms of first degree in EQ/rkT:

$$I_+ = \frac{4\pi rkT\lambda_+}{E(1+\frac{1}{2}EQ/rkT)} - \chi N_+ E, \qquad I_- = \frac{4\pi rkT\lambda_-}{E(1-\frac{1}{2}EQ/rkT)}. \tag{4}$$

According to experimental data, the equilibrium charge of a droplet must be equal to zero when $\lambda_+/\lambda_- = 2$ (in general, analogous arguments can be carried out for any critical value of λ_+/λ_-). Therefore we obtain

$$\chi N_+ E^2 = 4\pi rkT(\lambda_+ - \lambda_-) = 2\pi rkT\lambda_+. \tag{5}$$

Consequently, the flux of positive ions leaving the droplet must be equal to one half of the flux of positive ions to the droplet. Let us suppose that this relation is valid not only at $Q = 0$, but also for small values of Q. Then, for the resulting fluxes of ions to the droplet, we obtain

$$I_+ = \frac{2\pi rkT\lambda_+}{E(1 + \frac{1}{2}EQ/rkT)}, \qquad I_- = -\frac{4\pi rkT\lambda_-}{E(1 - \frac{1}{2}EQ/rkT)}. \tag{6}$$

The equilibrium charge of the droplet will be

$$Q_0 = \frac{2rkT}{E}\,\frac{\lambda_+/\lambda_- - 2}{\lambda_+/\lambda_- + 2}. \tag{7}$$

For $\lambda_+/\lambda_- < 2$ the equilibrium charge of the droplet is negative, while for $\lambda_+/\lambda_- > 2$ it is positive, in agreement with experimental data.

Calculations by formula (7) can be carried out when the condition $EQ/rkT \ll 1$ is satisfied. If, for example, one puts $T = 280°$ K and expresses r in microns and Q in elementary charges, then the condition takes the form $Q/r \ll 17$.

The values of the equilibrium charge of droplets with radii from 1 to 10μ at $\lambda_+/\lambda_- = 1.1$ agree well with the experimentally measured values (1). The indicated value of the ratio λ_+/λ_- is usual for atmospheric conditions.

It is obvious that, for an air polarization close to unipolar, formula (7) cannot be used. Calculations in this case must be carried out by formula (2). For conductivity ratios tending to zero, the charge of droplets may assume arbitrarily large values.

We now proceed to determine the concentration of atmospheric ions in the equilibrium state. The change in the concentrations of atmospheric ions n_+, n_-, taking into account their production, capture of ions by droplets, and recombination, is described by the equations

$$\begin{aligned} -dn_+/dt &= I_+N_0/E - \nu_+ + \alpha^+ n_+ n_-, \\ -dn_-/dt &= I_-N_0/E - \nu_- + \alpha^+ n_+ n_-, \end{aligned} \tag{8}$$

where N_0 is the concentration of droplets (for simplicity we assume a cloud or fogs monodisperse); $\nu_+ = \nu_-$ are the ionization intensities; α^{+-} is the ion recombination coefficient.

Taking expressions (6) into account, one can introduce the coefficients of combination of ions with droplets

$$\alpha^{+0} = \frac{2\pi rkTu_+}{E(1 + \frac{1}{2}EQ/rkT)}, \qquad \alpha^{-0} = \frac{4\pi rkTu_-}{E(1 + \frac{1}{2}EQ/rkT)}. \tag{9}$$

For the equilibrium state we obtain from (8)

SovietRxiv

$$\frac{n_+}{n_-} = \frac{\alpha^{-0}}{\alpha^{+0}} = 2\,\frac{u_-}{u_+}\,\frac{1 - \frac{1}{2}EQ_0/rkT}{1 + \frac{1}{2}EQ_0/rkT}. \tag{10}$$

Thus, in the equilibrium state the ratio of the numbers of ions depends only on the capture of ions of both signs by the droplets.

From (8) and (10), taking (9) into account, we find for the equilibrium concentrations of ions:

$$n_+ = \frac{-\alpha^{-0}N_0 + \sqrt{(\alpha^{-0}N_0)^2 + 4(\alpha^{-0}/\alpha^{+0})\alpha^{+-}\nu_+}}{2\alpha^{+-}},$$

$$n_- = \frac{-\alpha^{+0}N_0 + \sqrt{(\alpha^{+0}N_0)^2 + 4(\alpha^{+0}/\alpha^{-0})\alpha^{+-}\nu_+}}{2\alpha^{+-}}. \tag{11}$$

If the condition $N_0^2 \ll 4\alpha^{+-}\nu_+/\alpha^{-0}\alpha^{+0}$ is satisfied, then we obtain (in the absence of fog)

$$n_+n_- = \nu_+/\alpha^{+-} = \nu/\alpha^{+-}. \tag{12}$$

In the case of equal ion concentrations

$$n = \sqrt{\nu/\alpha^{+-}}. \tag{13}$$

If the ionization intensity $\nu = 10$ ion pairs $\text{cm}^{-3} \cdot \text{sec}^{-1}$, and the ion recombination coefficient is taken equal to $\alpha^{+-} = 1.6 \cdot 10^{-6}\ \text{cm}^{-3} \cdot \text{sec}^{-1}$, then the equilibrium ion concentration will be $n = 2400\ \text{cm}^{-3}$.

In the case of a large concentration of droplets, i.e., when the condition $N_0^2 \gg 4\alpha^{+-}\nu_+/\alpha^{-0}\alpha^{+0}$ is fulfilled, we obtain for the equilibrium ion concentrations

$$n_+ = \nu_+/\alpha^{+0}N_0, \qquad n_- = \nu_-/\alpha^{-0}N_0. \tag{14}$$

If we put $r = 5\ \mu$, $Q = 50E$, $T = 280°\text{K}$, then the condition under which relations (14) hold gives, for the droplet concentration,

$$N_0 \gg 50\ \text{cm}^{-3}.$$

Let, for example, $N_0 = 400\ \text{cm}^{-3}$, $\nu_- = \nu_+ = 10\ \text{cm}^{-3} \cdot \text{sec}^{-1}$. Then $\alpha^{+0} = 8.0 \cdot 10^{-5}\ \text{cm}^{-3} \cdot \text{sec}^{-1}$, $\alpha^{-0} = 3.3 \cdot 10^{-4}\ \text{cm}^{-3} \cdot \text{sec}^{-1}$. Thus, the coefficient of combination of ions with droplets at the indicated concentration of them is considerably higher than the coefficient of ion recombination.

SovietRxiv

The equilibrium ion concentrations, according to (14), turn out to be $n_+ = 310$ cm^{-3}, $n_- = 76$ cm^{-3}. If the mobilities of ions in a cloud or fog are taken equal to $u_+ = 1.37\ \frac{\text{cm}}{\text{sec}} / \frac{\text{V}}{\text{cm}}$, $u_- = 1.51\ \frac{\text{cm}}{\text{sec}} / \frac{\text{V}}{\text{cm}}$ (^{2}), then the ratio of conductivities under equilibrium conditions will be

$$\lambda_+/\lambda_- = n_+u_+/n_-u_- = 3.8.$$

At the same time the magnitudes of the equilibrium charges of the droplets are the same in absolute value as for $\lambda_+/\lambda_- = 1.1$, and are opposite in sign.

Thus, the assumption made above about the role of the double electric layer leads to values of the equilibrium charge that agree well with the results of measurements.

It is easy to verify that the number of ions present in the atmosphere before the formation of fog is not capable of ensuring equilibrium charging of all droplets. Thus, in a fog with water content $w = 0.25$ g/m^3 and droplet size $r = 5\ \mu$, the required number of charges is $2.1 \cdot 10^4 E$ cm^{-3}. If the total number of heavy and light ions is 2400 cm^{-3}, then it can bring about equilib–

equilibrium charging of 11% of the droplets. This is in satisfactory agreement with the experimental data of A. P. Sergieva cited above. The formation of ions and the change in their equilibrium concentration may cause subsequent charging of droplets.

For the change in the charge of droplets with time it is easy to obtain the formula

$$Q = \frac{2rkT}{E}\,\frac{\lambda_+/\lambda_- - 2}{\lambda_+/\lambda_- + 2}\left(1 - e^{-2\pi\lambda t}\right). \tag{15}$$

As $t \to \infty$, formula (15) passes into (7). With the aid of formula (15) one can calculate the charge of a droplet of radius r at any instant of time, if the values of the conductivities λ_+ and λ_- are known. The quantity $\lambda = \lambda_+/2 + \lambda_-$ represents the effective conductivity.

Main Geophysical Observatory named after A. I. Voeikov

Received 30 XII 1966

REFERENCES CITED

1. A. P. Sergieva, *Izv. AN SSSR, ser. geofiz.*, No. 3, 347 (1958); No. 7, 1018 (1959).

2. P. N. Tverskoi, *Atmospheric Electricity*, L., 1949.

3. Ya. I. Frenkel, *Izv. AN SSSR, ser. geogr. i geofiz.*, 8, No. 5, 325 (1944).
4. T. Alty, *Proc. Roy. Soc. London*, A 106, 316 (1924); 112, 235 (1926).
5. J. Chalmers, F. Pasquill, *Phil. Mag.*, 23, 88 (1937).
6. R. Gunn, *J. Meteorol.*, 11, No. 5, 339 (1954).

Note: Figure translations are in progress. See original paper for figures.

Source: Math-Net.Ru and CyberLeninka. Machine translation. Verify with the original.