THE CLOUD CHAMBER TECHNIQUE IN THE STUDY OF ICE-FORMING CHARACTERISTICS OF ATMOSPHERIC AEROSOL

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We analyze specific features of the cloud chamber technique used for measurements of atmospheric ice nuclei concentration. The chamber "SALYa" at the Central Aerological Observatory and the measurement procedure are described. The results of the study of systematic and random errors of the method are presented. The correspondence of the conditions in the cloud chamber to natural conditions of ice nuclei action is discussed.

Ice-forming ability of atmospheric aerosol is determined by its content of ice-forming particles, the so-called ice nuclei (IN). Although the measurements of concentration and activity characteristics of the atmospheric ice nuclei are being performed more than 40 years throughout the world, the problem of adequacy is still urgent for the methods used for these purposes. The main difficulty of measurements is connected with a great variety of possible mechanisms of ice nuclei action in a cloud medium (vapor formation, condensation-freezing, contact mechanisms) and the impossibility to realize the set in a single method. The accumulated data arrays on ice nuclei concentrations are obtained using two main methods. These are the method of IN deposition and development on filters and the cloud chamber method. The method of filters was most widespread being convenient for field measurements. However, the method uses development of IN not in a suspension in a cloud medium but on a backing, makes the results obtained to be not sufficiently reliable.[1] The cloud chamber technique seems to be more accurate for simulating natural process of ice nuclei activation in clouds. The principle of IN deposition measurements in a cloud chamber is in determination of the number of crystals formed on nuclei due to formation of supercooled water fog in the sample of the atmospheric air studied. During many years, stationary cloud chambers with a long time of the fog's existence (IN activation) were considered as absolute IN counters, and devices of other types were compared with them. However, many problems in ice nuclei measurements with cloud chambers are still poorly understood.

Good experience of using the cloud chamber method has been accumulated in the Central Aerological Observatory (CAO) by making measurements of IN parameters in different conditions during 20 years in which special value was placed on the accuracy of the method.

Some modifications of the cloud chamber "SALYa" (Atmospheric Ice Nuclei Counter)[2,3] designed at CAO are close in construction to known Admirat[4] and Ohtake[5] cloud chambers. " lock-diagram of one of the CAO chambers is presented in Fig. 1.

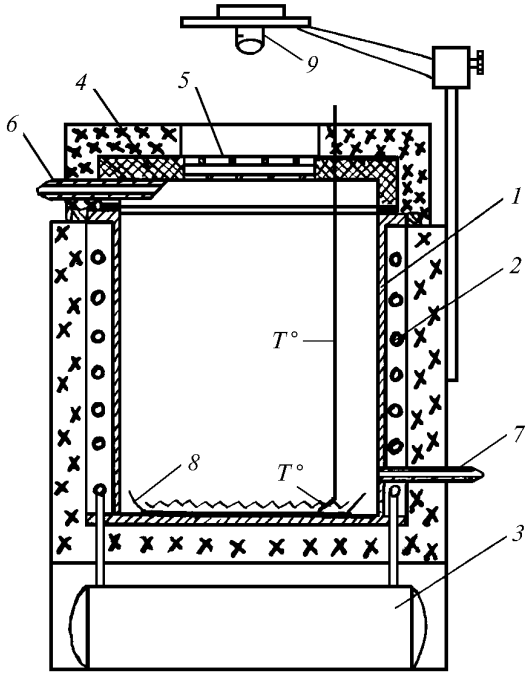

FIG. 1. Block-diagram of the cloud chamber "SALYa–1B.

The main part of the chamber is a brass thick-walled cylinder of 10 liters volume (*1*) cooled from its sides by a coil (*2*) which is the cooler of the refrigerating machine (*3*). The chamber is hermetically closed with an uncooled cap (*4*) having a lateral window (*5*) to observe the fog formation and the growth of ice crystals. To fill the chamber with the air to be studied, there is a connecting pipe (*6*) in the upper uncooled part of the chamber. The output pipe (*7*) is in the lower part of the cylinder. In some modifications, both pipes are placed along the axial line of the chamber. On the bottom of the chamber, a container with an indicator (sugar) solution (*8*) is

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placed. Ice crystals formed on ice nuclei and precipitated onto the bottom grow to visible size (a few millimeters) in sugar solution. A plane heater on the bottom of the container serves to melt crystals after measurements. The chamber is equipped with two thermometers to measure temperature in the central part of the working volume and the indicator solution temperature. To illuminate the working volume, a parallel light beam from a source (9) is used.

The measurement procedure involves several steps: (1) an air sample filling into the working volume of the chamber cooled down to the working temperature; (2) IN activation in the supercooled cloud formed; (3) growth of ice crystals in the indicator solution and visual calculation of their number; (4) heating the solution to melt crystals; (5) recovery of the temperature regime in the chamber and in the solution to working values before a new measurement.

To obtain sufficiently dense supercooled fog in the chamber, the air sample is moistened at the chamber input by passing through a special disk with an internal spiral channel. The bottom of the channel is lined with a moist sponge. The parameters determining the measurement regime are the volume of air, sample intake rate, and moistener temperature. Their optimal values were determined experimentally in a preliminary study.

The necessary volume of air pumped through the chamber was determined as the minimum volume required for complete change of a sample in the chamber. "y measuring the aerosol concentration decrease in the chamber during the pump through of the filtered air, it was established that the complete air change is obtained at a threefold exchange (30 liters). The optimal intake rate was chosen, on the one hand, from the condition of sufficiently quick air change in the chamber and, on the other hand, from the condition of excluding considerable turbulence of the flow. It appeared to be 15 liters/min. The necessary temperature of the moistener was determined from the condition of obtaining a sufficient water content in the fog and, consequently, long existence of the fog at as small temperature increase in the working space of the chamber as possible. This temperature is maintained at the level of 20–25°C.

"efore taking an air sample in, the temperature field in the working volume of the chamber is quite homogeneous due to a relatively slow cooling of the unit (30 min from the indoor temperature to −20°C) and high heat capacity of the cylinder. No vertical gradient is observed in the working space of the cylinder. In the additional uncooled volume under the cap, the temperature is higher by a few degrees as compared with the working space temperature. No radial gradient has also been detected in the main volume. The possible influence of the near wall layer is avoided by using containers whose diameter is less than that of the chamber cylinder by 1–2 cm. During the sampling, the temperature of the working space rises by a few degrees.

After taking the air sample in and shut-off of the studied sample from the outer medium, the temperature reaches its initial value during 2–3 min. Figure 2*a* shows the variations of temperature inside the chamber during the measurement cycle at the temperature of 20°C. The shaded strip denotes the spread in temperature values in different measurements. It is caused by fluctuations of the moistener temperature and unstable work of the refrigerator machine.

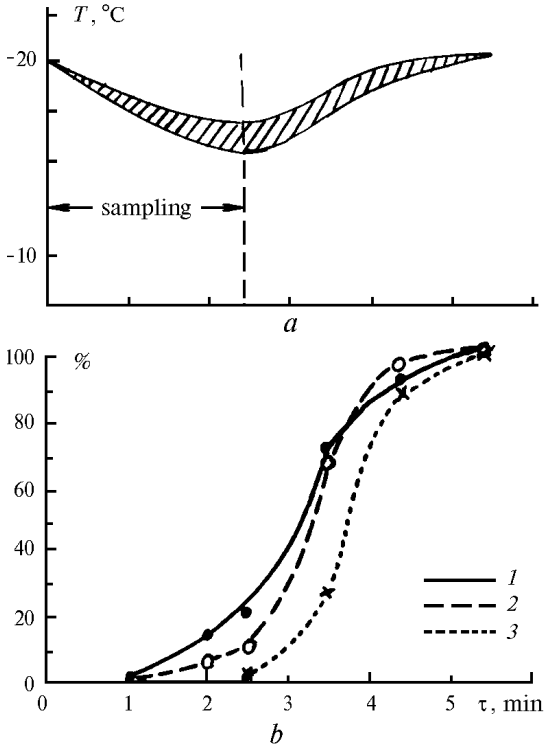

FIG. 2. The behavior of temperature in the chamber (a) and IN development (b) during the measurement cycle at different temperatures of the moistener: 18°C *(1);* 25°C *(2);* 31°C *(3).*

Figure 2*b* presents the relative number of ice nuclei appearing in the chamber and detected in the indicator solution at successive moments in time during the measurements. As is seen from Fig. 2*b*, nuclei activation begins even at the stage of air sampling but the majority of nuclei appears after sampling, at the 3rd or 4th minute of the measurement cycle. The nuclei appear with the change of fog temperature. The measurement temperature is taken to be the minimal temperature $T_{\min}$ achieved in the chamber by the end of measurements. The measured number of ice nuclei is the total number of nuclei activated at $T \geq T_{\min}$.

The influence of the variable field of water vapor supersaturation on the number of nuclei measured is the most complicated problem in estimation of the correspondence between the cloud chamber method and natural conditions of IN activation. Supersaturation occurs in a measurement cycle during the pump of a moistened air sample into the cooled

chamber and cause formation of a supercooled fog in the chamber. Unfortunately this short-lived supersaturation is hard to be detected experimentally. The estimation by the theory of mixing air masses of different temperature[6] demonstrates that supersaturation may reach a few tens percent that considerably exceeds the supersaturation in clouds.

The influence of supersaturation occurring in the chamber on the concentration of ice nuclei measured was studied by varying the moistener temperature T_m. The increase in the moistener temperature increases the moisture content of the air sample in the chamber. This simultaneously leads to an increase in the initial supersaturation in the chamber, increase of water content, and lifetime of the fog formed.

The influence of supersaturation follows from the fact that no dependence on the fog parameters (water content and lifetime) should occur if they reach values sufficient for the development of all the ice nuclei active at temperature of 23–25°C. Supersaturation influence must lead to a permanent growth of the IN number at a supersaturation due to activation of the increased number of nuclei. The dots in Fig. 3 show the numbers of ice nuclei detected at different temperature of the moistener and related to the number of nuclei detected at the temperature of 23°C.

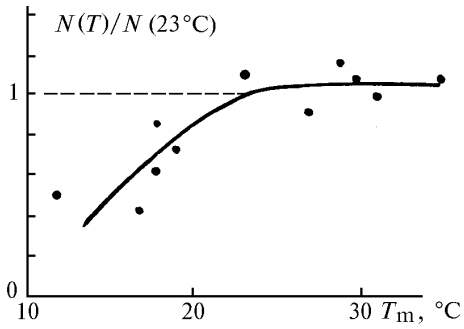

FIG. 3. The IN concentration measured as a function of the moistener temperature.

As seen from the Figure, the number of ice nuclei detected did not depend at $T_m > 23$°C on the moistener temperature, excluding the domain of low temperatures ($T_m < 20$°C) where the number of activated nuclei decreases due to insufficient water content and short lifetime of the fog ($\tau < 3$ min). This makes it possible to conclude that the influence of short initial vapor supersaturation on the number of IN activated in the chamber is insignificant.

The kinetics of IN development at different T_m presented in Fig. 2*b* also shows the absence of supersaturation influence. As T_m increases (curves *2* and *3*), part of the nuclei formed at the stage of sampling, when supersaturation occurs, even does not increase but steadily decreases with the growth of T_m. This result may be explained by a faster increase of temperature in the chamber at pumping of warmer air samples and indicates at least considerably less influence of supersaturation as compared with the influence of the cloud medium temperature.

Additionally, small influence of supersaturation on the activation of atmospheric ice nuclei in a cloud chamber follows from the study of the dependence of measurement results on the total concentration of atmospheric aerosol. The knowledge of this dependence is necessary in order to use cloud chambers at larger heights where the total aerosol content is small.

The measurements of ice nuclei concentration performed at diluting the atmospheric samples with different volumes of air free of aerosol demonstrate that, in a wide range of atmospheric aerosol concentration, from 10^2 to $3 \cdot 10^4$ cm^{-3}, IN concentration decreased proportionally to the total aerosol concentration. The detected IN number begins to increase sharply with the decrease of atmospheric aerosol concentration only for concentrations less than 100 cm^{-3}, when the fog structure changes considerably, the initial supersaturation increases significantly.

These experiments demonstrate that, under normal conditions, when the aerosol concentration in the air is more than 10^3 cm^{-3}, the atmospheric condensation nuclei rather quickly absorb the excess moisture when the air sample falls into the cloud chamber. The ice nuclei have weak condensation properties, so atmospheric nuclei do not permit them to be activated at short-time supersaturation.

All the data presented makes it possible to conclude that short-time supersaturation in a cloud chamber do not create substantial distinctions between the process of IN activation in the chamber and in the natural cloud medium.

The loss of a portion of ice nuclei from the air sample in a moistener introduces a systematic error to the cloud chamber technique. The condensation of atmospheric aerosol in the moistener was studied using the electrical aerosol analyzer TSI–3030 measuring the particles' spectrum in the range from 0.01 to 1 μm, and the device PKZV–906 measuring the concentration of particles of the diameter 0.3–100 μm in seven size intervals. The concentration of particles with the diameter less than 1 μm measured by the analyzer TSI–3030 did not change in the moistener. The results of measurement of atmospheric aerosol parameters obtained using PKZV–906 before and after the moistener are presented in the Table I.

TABLE I. Losses of large aerosol particles in the moistener.

Particle size, μm	0.3–0.4	0.4–0.5	0.5–1	1–2	2–5	5–10	10–100
Decrease of concentration, %	5	5.7	9.5	25.5	40	–	–

As seen from the Table, the losses of atmospheric particles increase with the growth of particles' size in correspondence with the inertial character of their condensation. Although the loss of large particles is rather considerable, it makes insignificant contribution to the total loss of ice nuclei, as their majority has dimensions less than 0.1 μm.[7] The maximal error estimated from data on IN size distribution and as well as from the results presented in the table turned to be less than 7%.

Random errors in measurements of concentration of ice nuclei by the cloud chamber are caused by different factors connected both with the subjective accuracy of measurements and with the characteristic features of the object under study. The estimation based on the instrumental and statistical errors leads to the value of the random error about ±30% when the measured IN concentration is of the order of 10 particles per liter. Direct measurements of the IN concentration by sampling atmospheric aerosol from a big reservoir permitting one to perform several measurements without the change of total aerosol concentration demonstrated that the random error did not exceed ±15% if the demands of the technique were satisfied carefully.

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