DETERMINATION OF THE CONCENTRATION OF CLOUD CONDENSATION NUCLEI

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DETERMINATION OF THE CONCENTRATION OF CLOUD CONDENSATION NUCLEI

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By the present time, natural condensation nuclei that appear in the atmosphere at considerable supersaturations, $S > 10\%$, relative to the pressure of saturated water vapor have been studied fairly well. Theoretical calculations show that, in the formation of clouds, $S \leq 1\%$. Direct measurements of relative humidity in clouds do not contradict these data. The nuclei on which cloud droplets grow at the supersaturations occurring in the atmosphere are called cloud condensation nuclei.

The method of creating supersaturations by adiabatic expansion of moist air is unsuitable for producing small ($< 1\%$) supersaturations. Small supersaturations are produced either through the diffusion of heat and moisture between two moist surfaces of different temperature ([2]), or through the isothermal diffusion of vapors of two different liquids ([3]) (for example, water vapor and hydrogen chloride). The temperature-difference method for producing small supersaturations was used in the Wieland apparatus ([4]). In this method, the concentration of cloud nuclei was determined from the number of droplets that grew to sizes sufficient for them, upon falling to the bottom of the vessel, to leave traces in a layer of lacquer. Smaller droplets were not taken into account. In A. I. Storozhilova' s differential nucleus counter ([5]), based on the same method of producing small supersaturations, droplets that grew on nuclei were counted visually with the aid of an ultramicroscope; the droplet size was not determined. To determine the concentration of cloud condensation nuclei, Twomey ([6]) used the isothermal method of producing small supersaturations. The concentration of cloud nuclei was determined by him from the number of droplet images obtained when the investigated volume was photographed. The smallest size of droplets that the author was able to record by this method is unknown.

Different authors take as the concentration of cloud condensation nuclei the concentration of droplets of substantially different size growing at a given supersaturation. Arbitrary determination of the concentration of cloud nuclei leads to incomparability of the results of their measurements. In addition, the method of determining the number of cloud condensation nuclei from the number of all droplets growing in a chamber at a given supersaturation is incorrect,

Fig. 1. Diagram of the apparatus for determining the concentration of cloud condensation nuclei

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since not all of these droplets growing at constant supersaturation will reach cloud-droplet sizes. It is known that, in the transition from 60 to 99% relative humidity, rapid growth of droplets occurs on salt particles, but the concentration of these growing droplets cannot be taken as the concentration of cloud nuclei active at 99% humidity, since these droplets at such humidity will not reach the sizes of cloud droplets (at a humidity of less than 100%, clouds cannot exist).

From the solutions of the Kelvin, Raoult, and Van' t Hoff equations it is known ([7]) that for each definite supersaturation there exists a critical

size upon reaching which the droplet will continue to grow through condensation of vapor, with a decrease in free energy, and will reach the sizes of cloud droplets provided that the supersaturation remains constant. From these equations it can be found that, at equal supersaturations, the critical size of droplets growing on insoluble nuclei is 1.5 times greater than the critical size of droplets growing on completely soluble nuclei. The critical size of droplets growing on mixed nuclei is greater than the critical size of droplets growing on completely soluble nuclei, but smaller than the critical size of droplets growing on insoluble nuclei at the same supersaturations. The critical size of droplets growing on insoluble nuclei is equal to the critical size of droplets of pure water. Droplets whose size at a given supersaturation is smaller than the critical size of droplets of pure water will not grow to the sizes of cloud droplets, and they should not be taken into account when determining the concentration of cloud condensation nuclei.

Fig. 1. Diagram of the apparatus for determining the concentration of cloud condensation nuclei

On the basis of the foregoing, one can formulate a definition of the concept of cloud condensation nuclei. **Cloud condensation nuclei**, active at a supersaturation S_0, are aerosol particles on which, at a continuously maintained supersaturation, droplets will grow to a size exceeding or equal to the size of droplets of pure water that is critical for the supersaturation S_0. Only measurement of the maximum concentration of droplets that have grown on nuclei, at a continuously maintained supersaturation, to sizes equal to or greater than the critical size of droplets of pure water makes it possible to determine unambiguously the concentration of cloud nuclei active at the given supersaturation, independently of the method by which the supersaturation is produced or of the method by which the droplets are recorded.

On the basis of the definition of the concept of the concentration of cloud condensation nuclei, we have developed a method for determining cloud condensation

Fig. 2. Concentrations of condensation nuclei active at small supersaturations

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nuclei ([8]), distinguished by the fact that the concentration of cloud nuclei is determined from the concentration only of those droplets whose size exceeds the critical size for the given supersaturation. The apparatus on which the investigation of cloud condensation nuclei was carried out (Fig. 1) consists of an isothermal diffusion chamber I, a photoelectric analyzer of the individual sizes of droplets II, and a five-channel electronic recording device III. The air under investigation is drawn into chamber I through tube 1. The growth of droplets in the chamber on the nuclei occurs at a supersaturation determined by the diffusion of water vapor and hydrogen chloride vapor. In the upper part of the chamber there is cotton wool 2, moistened with water, and an aqueous solution of hydrochloric acid 3 is poured onto the bottom of the chamber. The droplets growing in chamber I are drawn into the photoelectric analyzer through tube 4. To prevent spreading of the air jet emerging from tube 4, it is surrounded by a stream of air purified of dust by filter 5.

The photoelectric analyzer II is based on recording the intensity of light scattered by an individual particle within a narrow solid angle in a direction close to the direction of propagation of the light illuminating the particle under investigation. Analyzer II makes it possible to measure droplets of size···

radius greater than 0.1 μ. An incandescent lamp of the Ts-62 type, 6, uniformly illuminates the target 8 through the antireflection lens 7. The objective 9, with a relative aperture of 1 : 1.5, forms an image of the target 8 at the point where the light beam intersects the aerosol jet. Dark-field observation of aerosol particles is effected by covering the central part of objective 9 with screen 10. The size of the screen is chosen so that direct light from the lamp does not fall on lens 11, which collects the light scattered by the particles under study onto photomultiplier 13 of the FEU-35 type.

The use of a photoelectric instrument with a five-channel pulse analyzer made it possible, simultaneously with measuring the concentration of cloud condensation nuclei active at small supersaturations (from 0.1 to 1%), to determine the total concentration of drops of radius greater than 0.2 μ growing in the chamber. The concentration of cloud condensation nuclei was determined from the maximum concentration of drops grown in the chamber that exceeded, for the supersaturation being investigated, the critical size of drops of pure water. For comparison with the data of paper (6), the total concentration of drops grown 30 sec after the chamber was filled with the air under investigation was determined.

Fig. 2. Concentrations of condensation nuclei active at small supersaturations: **A**—according to Twomey' s data (6); **DC**—in continental air during a drought

period, **C**—in continental air; **B**—according to our data processed by Twomey' s method (6): **a**—June 3, 1964; **b**—June 4, 1964; **c**—from the results of the same measurements as in Fig. 1 **B**, but taking into account only those drops whose sizes exceed the critical ones; **g**—according to the data of Wieland (4).

The investigation of the concentration of cloud condensation nuclei active at supersaturations of 0.1, 0.2, 0.3, and 0.5% was carried out in a rural locality 100 km from Moscow in June 1964. Air sampling into the chamber and determination of the nucleus concentration were performed at the sampling site.

Figure 2 presents the results of determining the concentration of cloud condensation nuclei by various authors. Analysis of the curves shows that the concentrations of condensation nuclei determined from our measurements by Twomey' s method (Fig. 2 **B**) and by the method proposed in the present work (Fig. 2 **V**) at a supersaturation of 0.5% practically coincide. This is explained by the fact that the critical size of pure-water drops corresponding to a supersaturation of 0.5% is sufficiently small and equal to 0.25 μ. At supersaturations equal to 0.1%, the concentrations of nuclei determined by Twomey' s method are 10 times greater than the concentrations of the corresponding cloud condensation nuclei. The spectra of condensation nuclei with respect to supersaturations, determined by us by Twomey' s method and presented in paper (6) (Fig. 2 **A**), are in good agreement with one another. On the other hand, the spectra of cloud condensation nuclei calculated from the results of the same measurements (Fig. 2 **V**) are in good agreement with the spectra obtained by Wieland (Fig. 2 **G**). The observed agreement of Wieland' s data with our spectra of cloud condensation nuclei is explained by the fact that Wieland took into account not the total concentration of drops, as in Twomey' s method, but only drops of cloud size. In Twomey' s work all drops growing in the chamber at any supersaturations were determined; therefore the data obtained by him are not concentrations of cloud condensation nuclei.

The data of Fig. 2 clearly show that there are substantial differences in the character of the spectra of condensation nuclei with respect to supersaturations found by Twomey.

in Australia and by Viland in Europe are determined chiefly not by peculiarities in the nature of the nuclei observed, but by differences in the methods used to determine them.

In conclusion I express my gratitude to Academician E. K. Fedorov for posing the question.

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