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OPTICAL INVESTIGATIONS OF ATMOSPHERIC AEROSOL

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CONTENTS

1. INTRODUCTION

THE striking success of the theory of molecular scattering of light, from its very first steps, has for many decades cast a spell not only on physicists, but also on geophysicists, and have determined to a considerable degree the research trends in atmospheric optics. Its creators have presented the main deductions of the theory with convincing clarity and with relative mathematical simplicity—factors of no little importance when it is necessary to cope with the interlinking of different effects and to obtain not only qualitative estimates but also quantitative calculations.

The first touchstone of the theory, as is well known, was the scattering of sunlight by the earth's atmosphere. The good semiquantitative agreement between the spectral and polarization characteristics of the light coming from the sky and the theoretically expected values was justifiably considered by the contemporaries as a triumph of the theory. And although the comparison with the experimental data immediately revealed also quite serious deviations from its predictions (for details see[1]), an erroneous notion has gradually taken hold that these comparisons, so to speak, can be regarded as convincing proof that the main factor responsible for the scattering of light by the earth's atmosphere is its gaseous component.

Actually, however, the atmospheric air is an aerosol, and the scattering of light by its disperse phase, i.e., by the foreign particles suspended in the air, plays a role of no little importance in the optics of the atmosphere. This circumstance was known quite well even before Rayleigh's work and before the experiments of Tyndal, to whom a tradition known to be false relates the discovery of the scattering of light by small particles (see[1]). However, as soon as it became necessary to deal with scattering of light by particles whose dimensions are comparable with or larger than the wavelength of the light, Rayleigh's simple and orderly theory was replaced by the cumbersome theory of diffraction by three-dimensional bodies, the capabilities of which are highly limited[2,3].

The need for operating with poorly convergent series having a structure that is complicated and lends itself with difficulty to a qualitative analysis, has immediately shackled the theoretical thinking and led it on the path of strong idealizations. One must add to this the variety of forms, dimensions, and natures of the particles contained in the air, the limited amount of information concerning their real physical characteristics, and the extreme sensitivity of the laws of light scattering to all the foregoing factors[2,3]. Finally, technical difficulties encountered in the performance of experiments on scattering of light in the real atmosphere and in creating aerosols under laboratory conditions have limited to an extreme degree the nature and the volume of information on the actual optical properties of atmospheric air. In particular, the matrix theory of radiation transfer, developed already in the middle 40's[4-7], could not be applied to an aerosol atmosphere in view of the total lack, until most recently, of any information whatever on the behavior of the components of the matrix of scattering of light by atmospheric air[1,8,9].

All this has created a chasm, not to be bridged until recently, between the theory of the scattering of light by small particles and the theory of propagation (transport) of radiation in a scattering medium, on the one hand, and the experimental research on these phenomena, particularly under conditions of a real atmosphere, on the other. The theoretical conclusions were resorted to primarily for qualitative illustrations, while the quantitative analysis was unavoidably performed from the point of view of the assumption of the molecular character of scattering.

This has gained so much currency, that the very need for taking into account the scattering of light by disperse phases other than clouds and fogs has been regarded by many as doubtful, especially when dealing with clear weather or high altitude. A quarter of a century ago, in particular, no one ever doubted seriously that beyond the limits of the troposphere the disperse component does not exist at all, and that the scattering has a pure molecular character. As research penetrated deeper into the stratosphere, the limit at which the air can be regarded as dust free had to move higher and higher, but the very concept that air becomes cleaner with increasing altitude continued to govern. It persists in many papers published until most recently, especially

when dealing with attempts to construct a systematized optical model of the atmosphere (see, for example,[10-17] and others).

This, in general outline, was the situation at the end of the 50's. It is described in greater detail by the author in his review[1], and in the review chapters of[8,9]. The last decade has introduced radical changes into this picture. The use of latest experimental techniques and modern flying craft, the implementation of complex purposeful programs of complete and exhaustive physical experiments, the widespread use of computers, and the development of the optical sounding of the atmosphere, all have made accessible new sources of information and made it possible to overcome in many respects the existing gap between theory and experiment. Scattering of light has turned into a powerful tool for the investigation of the atmospheric aerosol and the processes of its transformation.

As a result, our opinions concerning the character of the disperse phase of atmospheric air, its optical properties, and the aerosol structure of the atmosphere, and also the character of the condensation phenomena occurring in the atmosphere, have changed noticeably. These changes are indeed the main topic of the present review, which is a direct continuation of the review published eight years ago[1]. Just as in[1], we do not claim to discuss the problem completely, nor do we present an exhaustive bibliography. Attention is focused only on some of the most debatable aspects of the problem, predominantly from the point of view of the results obtained recently under the direction of the author at the Institute of Atmospheric Physics (IFA) of the USSR Academy of Sciences.

Changes took place, however, not only in our notions concerning the atmospheric aerosol. Our views regarding the role of the disperse phase of atmospheric air and the weather-forming processes, and its connection with human activity, have been radically altered. This aspect of the problem, which is important for the understanding of the purpose of the investigations, is beyond the scope of this review, and we shall confine ourselves only to a few remarks.

The transport and the transformation of the disperse phase of the atmospheric aerosol (for example as a result of a condensation process) give rise to extreme variability of the optical state of the atmosphere as a function of the meteorological conditions, and serve as the main cause of this variability. Thus, the scattering coefficient of air in the visible region of the spectrum changes approximately from 10^{-2} km^{-1} for barely noticeable haze to 10^{4} km^{-1} for extremely dense fogs, i.e., by a factor of a million. This gives grounds for speaking of optical weather (i.e., the aggregate of the optical characteristics of atmospheric air at a given state of the weather) and of optical climate (i.e., the aggregate of the statistical laws governing the variability of the optical weather), and makes it necessary to investigate the role of the disperse phase of atmospheric air during the weather-forming processes.

It is observed at the same time that the optical weather determines not only the conditions for light propagation and image transport in the atmosphere, but exerts a decisive influence on the atmospheres radiation regime, and by the same token also on the radiative heat exchange. In order to estimate the extent of this influence, it is sufficient to recall the role of clouds in the regulation of heat exchange between the atmosphere, the underlying surface, and the outer space (see, for example,[18,19]). The unavoidable multiple scattering of the light greatly changes its absorption by both the disperse and the main phase[19-21], in a manner that depends very strongly on the degree of the dispersion of the aerosol[20]. Recently, direct experimental and theoretical indications have appeared that the disperse phase plays an important role in the radiative heat exchange also in the absence of clouds and fog, i.e., in clear weather[22-26] (direct evidence in this favor were recently obtained in our laboratory both with airplane experiments (V. D. Oppengeĭm and G. P. Faraponova) and with satellite experiments (M. S. Malkevich, A. K. Gorodetskiĭ)), and also during the process of cloud formation—at the cloud boundaries[20]. Finally, a very important role is played in the thermodynamics of the atmosphere by condensation processes that are directly connected with the fate of the disperse phase of the atmospheric aerosol.

Thus, the disperse phase comes more and more to the forefront as one of the main regulators of the weather-forming process[235]. However, the state of the atmospheric aerosol not only influences this process, but is determined entirely by its course. In other words, the connection between the optical state of the atmosphere and the thermodynamic processes occurring in it is very close but via an intermediary. This intermediary is the most variable and the least investigated component of the air, namely the foreign inclusions suspended in it. Moreover, this intermediary ensures, as will become clear later, the occurrence of the most important mechanism of feedback between the development of the meteorological processes (including the transformation of the boundary conditions) and radiative heat exchange phenomena that serve as the only source of energy for these processes.

When assessing the significance of this mechanism it is necessary to bear in mind (besides its importance for weather forecasting) two circumstances. First, even relatively negligible changes of the radiation balance of the planet as a whole lead inevitably to catastrophic changes of its climate, i.e., of the conditions of our existence. Thus, the radiative balance of the incandescent atmosphere of Venus differs from the earth's present-day balance by only several per cent; even smaller changes (but in the opposite direction) suffice to cover permanently the earth's sphere with a thick ice cover. On the other hand, the total energy released by all the installations at humanity's disposal already begins to approach the limit of their climatic sensitivity, and further progress in power engineering soon will become impossible without taking effective measures to conserve the climate.

Second, natural variations of the state of atmospheric aerosol are accompanied by very strong disturbances to the radiation balance, which become equalized on the average only over the entire earth's sphere and over sufficiently long periods. During the last decades, human activity has become an important source (on a geophysical scale) of aerosol production, and this is already manifest strongly, for example, in large cities

(or even in entire regions)[27-31], and also at high altitudes, where contamination of the air by combustion products of rocket fuel becomes significant (for bibliography see[32-34]). There are gounds for assuming that this antropogenic aerosol already exerts a noticeable influence on the transformation of the climate. On the one hand, this likewise calls for measures aimed at conserving the climate, and on the other it opens up certain possibilities for the use of artificial aerosol for the purpose of such a conservation (we recall, for example, the protection of orchards against freezing by means of smoke). We mention, finally, an extensive group of very timely problems of social medicine, agriculture, geology, and many other branches of science and national economy, which call for knowledge of the properties of the atmospheric aerosol[27-30].

Therefore a study of the optical properties of atmospheric aerosol and their dependence on its microstructure, and also an investigation of the dependence of the microstructure of the aerosol on the meteorological conditions come to the forefront as most important problems of modern physics of the atmosphere.

An objective measure of the importance of this problem is the consistent growth in the number of researches devoted to it in all the countries of the world. The overall picture of our knowledge in this field at the start of the 60's is represented in splendid monographs[27-30] and also in a surprisingly old-fashioned, not at all reliable, but contentful monograph[31], to which we refer the reader but with an important stipulation. In many cases the authors of these monographs, when speaking of the properties of the aerosol, lean on the results of optical research. As will be shown in what follows, many of these results now must be regarded as unreliable or, at any rate, calling for a critical review, in view of recently revealed singularities of the problem of extracting microphysical information concerning the disperse phase from information about its optical properties.

For the reader's convenience, we preface our review with a brief abstract from the aforementioned monographs[27-31].

2. CERTAIN INFORMATION ON THE ATMOSPHERIC AEROSOL

The substances that enter the atmospheric air in the liquid-drop phase or in the form of solid particles form only a small admixture to the mass of the atmospheric air, as can be seen from Table I. Their role is determined, on the one hand, by the fact that they serve as nuclei for the condensation or sublimation of water vapor, and also as an active adsorbent and catalyst, and on the other hand, by their exclusively high optical activity, which, in particular, makes the scattering of light a very sensitive indicator of the course of the condensation phenomena. Thus, in slight haze, it is possible to discern optically, from the variation of different optical properties of air, changes of 10^{-15}—10^{-16} g/cm^3 (i.e., 10^{-3}—10^{-4} μg/m^3) in the concentration of the finely-dispersed phase, or the appearance of one particle having a radius 5—10 μ in several dozen liters of air.

According to quite firmly established recommendations by H. Junge[27], it is customary to separate three

Table I. Content of slight impurities in the surface layers of atmospheric air.

Impurity	Amount	
	μg/m^3	μg/g
Argon	$1.6 \cdot 10^7$	$9.3 \cdot 10^3$
Neon	$1.6 \cdot 10^4$	1,8
Helium	$9.2 \cdot 10^2$	5,2
Krypton	$4.1 \cdot 10^3$	1,1
Xenon	$5 \cdot 10^2$	$8.6 \cdot 10^{-2}$
Ozone	0—10^2	0—$5 \cdot 10^{-2}$
Hydrogen	36—90	0.4—1,0
Carbon dioxide	(4—8)·10^5	(2—4)·10^2
Carbon monoxide	(1—20)·10^1	(1—20)·10^{-2}
Methane	(8.5—11)·10^2	1,2—1,5
Formaldehyde	0—16	0—10^{-2}
Nitrous oxide	(5—12)·10^2	(2.5—6)·10^{-1}
Nitric oxide	0—6	(0—3)·10^{-3}
Ammonia	0—15	(0—2)·10^{-2}
Sulfur dioxide	0—50	(0—20)·10^{-3}
Hydrogen sulfide	3—30	(2—20)·10^{-3}
Chlorine	1—5	(1—15)·10^{-4}
Iodine	(5—50)·10^{-2}	(4—40)·10^{-6}
Water vapor	$3 \cdot 10^4$—$3 \cdot 10^7$	$4 \cdot 10$—$4 \cdot 10^3$
Water drops:		
a) Outside fogs and clouds	0—10^4	0—10
b) In fogs and clouds	10^4—10^7	10^0—10^3
Solid particles suspended in the air	10^0—10^3	10^{-3}—10^0

ranges of dimensions of the foreign particles suspended in air (Table II).

Inasmuch as the particle dimension changes greatly during the course of condensation, sublimation, or evaporation of the water vapor, it is necessary in the analysis of the aerosol state of the air, first, to separate the solid fraction of the disperse phase into soluble and insoluble (frequently one encounters also insoluble particles having soluble shells) and, second, a clear cut distinction between three states of atmospheric moisture:

a) Saturation state, when the relative humidity is $w \cong 100\%$ and the external conditions determine not the dimensions of the droplets, but the rate of their growth. In this case the condensation nuclei can be either large or even giant particles, solid (soluble and insoluble) and liquid, but the Aitken nuclei cannot serve as condensation nuclei, in view of the fact that the water vapor is insufficiently supersaturated. As seen from Fig. 1, which is taken from[28], under the conditions actually encountered when the supersaturation is low, the droplet dimensions in this state should exceed at least several microns.

b) Equilibrium-solution state, in which the liquid-drop phase results from absorption of atmospheric moisture by particles of soluble salts suspended in the air, and the dimensions of the droplet are determined by the condition that the solution forming them have an equilibrium concentration at the given relative humidity of the air. This state is realized in the relative-humidity interval $w_{cr} \leq w \leq 100\%$, where $w_{cr} \cong 70$—80% is the

Table II.

Range	Particle radius, cm
Highly disperse or "Aitken nuclei,"	10^{-7}—10^{-5}
Medium disperse or "large particles"	10^{-5}—10^{-4}
Coarsely disperse or "giant particles"	10^{-4}

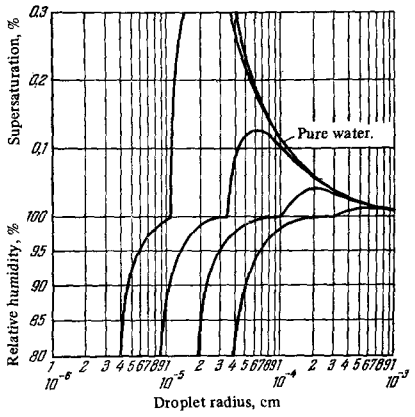

FIG. 1. Equilibrium relative humidity (or supersaturation) as a function of the radius, for drops of the solution produced when particles of potassium chloride of different dimensions are dissolved.

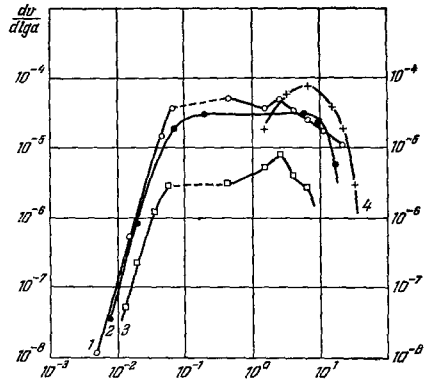

FIG. 2. Spectral distribution of the total volume of the solid fraction of the aerosol with respect to the dimensions of the particles forming it. 1 – Frankfurt-am-Main; 2 – Alps (altitude 3000 m); 3 – particle size distribution model proposed by Junge; 4 – over the surface of the sea.

critical value of w, starting with which the given salt absorbs the atmospheric moisture that dissolves it. The equilibrium size of the drops in this state depends not only on w but also on the mass of the salt nucleus, as shown in Fig. 1. Insoluble solid particles cannot serve as condensation nuclei at these values of humidity.

c) Pre-condensation state, when $w < w_{cr}$ and the particles of the soluble salts are also excluded from the condensation process. In this state, the assimilation of the moisture by the disperse phase is ensured not by condensation in the direct meaning of its word, but by other mechanisms such as absorption, chemosorption, etc.

We shall show below that these three states correspond to entirely different types of aerosol formation. Unfortunately, in most investigations (especially optical) of the disperse phase of atmospheric air (see[27-31]), no distinction is made between these three states, and this leads to a confusion of the picture and to a mystification of the results. This pertains, in particular, to many investigations of the size distributions of the particles.

When analyzing the results of these investigations it must be borne in mind that different ranges must be investigated by different methods[27-31], and that only few of the methods give an idea of the disperse phase in situ. In most cases, on the other hand, the measurement itself changes the state of the disperse phase, leading sometimes to complete dehydration. Therefore, when speaking of the optical properties of atmospheric aerosol, the use of data obtained by other methods must be approached with caution. In particular, data obtained with the aid of impactor traps or an electron microscope pertains certainly only to the solid fraction (including the solid bases of the condensation nuclei) and do not characterize the states of the aerosol.

A sample of average distributions of the total volume occupied by the solid phase with respect to the radii of the particles making up this phase are shown in Fig. 2. The ordinates represent the spectral density of the distribution dv/d log a, where a is the radius of the particle and v is the total volume of the particles contained in 1 cm^3 of air and having radii smaller than a (v assumes the meaning of the filling factor when $a \to \infty$). It is seen that the main contribution to the volume of the solid phase is made by large and giant particles, while not more than 10–20% of the total volume consists of Aitken nuclei. Under marine conditions, the content of the Aitken nuclei and of large particles decreases by approximately two orders of magnitude[27-31].

The upper limit of the distribution of particles by sizes is due to sedimentation processes, which limit the time that the particles remain in the suspended state. This limitation begins to felt quite strongly at a $\gtrsim 10\ \mu$ (the pollen of most plants and particles of less deposits have approximately these dimensions). When a $\gtrsim 20\ \mu$, the sedimentation rate becomes so large that such particles are practically not encountered in the quiet air. Only in clouds, where there are powerful updrafts, is the upper limit of the distribution shifted to approximately 80–100 μ. In rains, the droplet size distribution covers the range from approximately 0.1 mm in the case of weak drizzle to 3 mm in strong showers.

The lower limit of the particle dimension distribution is determined principally by two mechanisms—coagulation, which leads to a growth of the particles at a rate that increases with decreasing particle size, and the mechanism of new-particle production. As to coagulation, outside clouds and fogs the lifetimes of particles smaller than 10^{-6} cm amount to several hours, but particles in the range 10^{-6}–10^{-5} cm can remain in the atmosphere for days and weeks. In clouds and fogs, on the other hand, their lifetime is decreased to several minutes.

The mechanism of formation of the Aitken nuclei has barely been studied, nor its chemical nature. Three possible types of processes are discussed in the literature: condensation and coagulation of volatile substances with high boiling points (for example, in the formation of smoke), reactions between small gas impurities, including those occurring under the influence of irradiation (say, the reaction of formation of sulfuric acid or ammonium chloride), and penetration of the particles into the atmosphere from the earth's surface or from outer space.

At the present time many data have been accumulated offering evidence that the Aitken nuclei have an earth origin (predominantly from the continents). There are serious indications that they are produced directly in the air, principally in the surface layer, and frequently

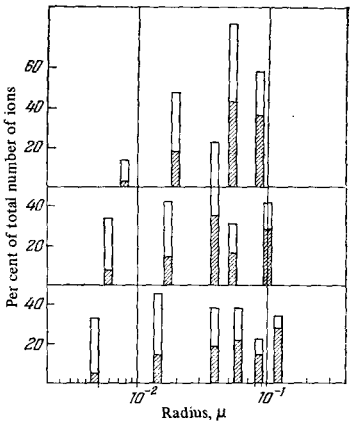

FIG. 3. Examples of the spectra of the dimension distributions of the Aitken nuclei (total height of the columns) and of their ionized fraction (shaded part). [27]

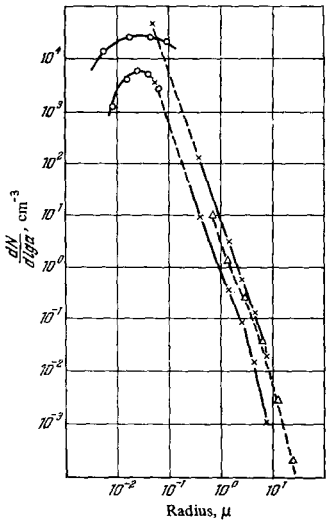

FIG. 4. Examples of dimension distributions of dry particles (the spectrum has been artificially smoothed out in the region of the Aitken nuclei).

as a result of human activity. The hypothesis that the Aitken nuclei are the product of crumbling of the soil or spray from the water surface are doubtful[27-31].

The lower limit of the dimensions of Aitken nuclei lies near $a \cong 4 \times 10^{-7}$ m. Figure 3 shows typical size distribution spectra of Aitken nuclei, showing separately the fraction of the nuclei carrying electric charges[27]. Attention should be called to a peculiarity of the Aitken nuclei, namely that they have a line spectrum. This peculiarity has not yet been explained. Attention is also called to the strong variability of this spectrum and to the absence of correlation between the different fractions of this range[27-31]. The absolute concentrations of the Aitken nuclei are subject to strong oscillations and are characterized by approximately the following numbers (of particles per cm^3):

industrial city	$-(5-40)\cdot 10^4$,
city	$-(5-100)\cdot 10^3$,
farm settlement	$-(1-70)\cdot 10^3$,
island	$-(5-500)\cdot 10^2$,
open ocean	$-(8-50)\cdot 10^2$,
mountains at altitude 500–1000 m	$-(1-40)\cdot 10^3$,
mountains at altitude 1000–2000 m	$-(4-100)\cdot 10^2$,
mountains at altitude > 2000 m	$-(2-50)\cdot 10^2$.

The Aitken nuclei are not washed out to any considerable extent by precipitation or by cloud and fog particles. In the main, the large and giant particles become washed out, and they consequently play the principal role in the formation of the coarsely-dispersed water drops[27-30].

Both large and giant particle of the dry fraction of the atmospheric aerosol constitute predominantly products of crumbling of the soil and salt crystallites (including those of oceanic origin). They include to a lesser degree combustion products and industrial waste (predominantly in industrial centers), and also products of volcanic activity and, finally, spores and plant pollen[27-31].

Numerous measurements, both by H. Junge himself, and by many other authors[27-31], have shown that averaging over many realizations and subsequent smoothing (stylization) of the experimentally observed distributions of particles of the dry fraction of the atmospheric aerosol in the finely-dispersed and coarsely-dispersed bands lead inevitably to the following relation, proposed by H. Junge,

$$\frac{dN}{d\lg a} = Ca^{-\nu}, \tag{1}$$

where ν varies approximately between 2 and 5, depending on the geographical and meteorological conditions, the altitude, the season, etc. Examples of such distributions are shown in Fig. 4. The concentrations of the particles having these dimensions usually amount to several dozen or hundreds of large particles and not more than several giant particles per cm^3.

The products of moisture condensation are also among large and giant particles. Outside the clouds and fogs, the size distribution of the liquid-drop fraction of the aerosol has been practically uninvestigated, principally in view of technical difficulties of performing the experiments (measurements in the range of 0.1–0.5 μ are particularly difficult). Taking into account the different growth rates of particles of different dimensions, and the variability of the nature of the condensation nuclei with changing dimensions, one cannot expect the spectrum of the condensed phase to be the same as the spectrum of the condensation nuclei.

Data are much more plentiful on the spectra of the particle size distributions in clouds, fogs, and precipitation. Averaging over a large number of realizations and subsequent stylization lead to smoothed distributions which are well approximated by the four-parameter distribution proposed by K. S. Shifrin

$$\frac{dN}{d\lg a} = Aa^{\mu}e^{-\beta a^{\gamma}}, \tag{2}$$

where A, μ, β, and γ are adjustment parameters that depend on the type of clouds or precipitation (including their water content[35]; Fig. 5). The total concentration of drops in clouds and fogs is usually from 10 to 500 cm^{-3}.

It must be borne in mind, however, that such averaged stylized distributions reflect the properties of the real distributions only in most general outlines. Concrete realizations are characterized by appreciable deviations from such smooth relationships, as can be

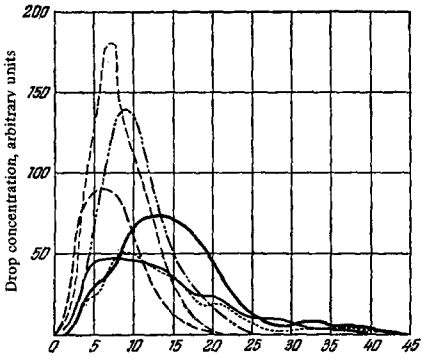

FIG. 5. Smoothed dimension distributions of drops in clouds of different types.

seen in Fig. 6. Distributions similar to those obtained by R. Fenn[36] in desert regions of the USA (Fig. 6a) were observed many times also by others, for example by K. Bullrich and his co-workers in the Hawaii Islands[37] and by J. P. Shedlovsky and I. H. Blifford at 9 kilometer altitude over Colorado[38]. Moreover, even after averaging over many realizations for a given type of formation, as a rule, certain deviations from the smooth distribution curves remain (say bimodal distributions for several types of fogs). The vanishing of these deviations usually is evidence either of mixing of several types of formations, which leads as a rule to a blurring of the main laws, or to artificial stylization of the curves for the sake of a convenient mathematical approximation.

Worthy of particular attention in this connection, in our opinion, is a fact observed by Yu. S. Georgievskiĭ in an analysis of measurements of the size distributions of dry particles of the coarsely-dispersed fraction of aerosol at the Zvenigorod scientific base (ZNB) of IFA. It turned out that the concentrations of the particles of different dimensions correlate only within approximately double the variation of their radius, and do not correlate outside of this interval. It seems that this offers evidence, on the one hand, of independent origins and independent fates of the different fractions of the dry base of the aerosol and, on the other hand, that the laws such as the Junge distribution can be regarded only as very far-fetched approximations on the average (we shall show below that the situation is the same also for the laws governing the altitude variation of the aerosol concentration), but do not reflect the nature of the real processes. We add that analogous results were obtained at ZNB by Yu. S. Lyubovtseva also for the liquid-drop phase of the fog, the only difference being that a distinct anticorrelation is observed here between the number of particles in the ranges 1—5 and 8—12 μ, showing that these ranges have a common origin.

3. THEORY OF OPTICAL PROPERTIES OF AEROSOL

The theory of the optical properties of aerosol is based on the concept that the aerosol is an aggregate of independently scattering particles with different dimensions. Recent investigations[39-44], and also the present author's analysis of the electrodynamic content of the photometric quantities, show that such a concept is valid within the framework of ray optics at time and space

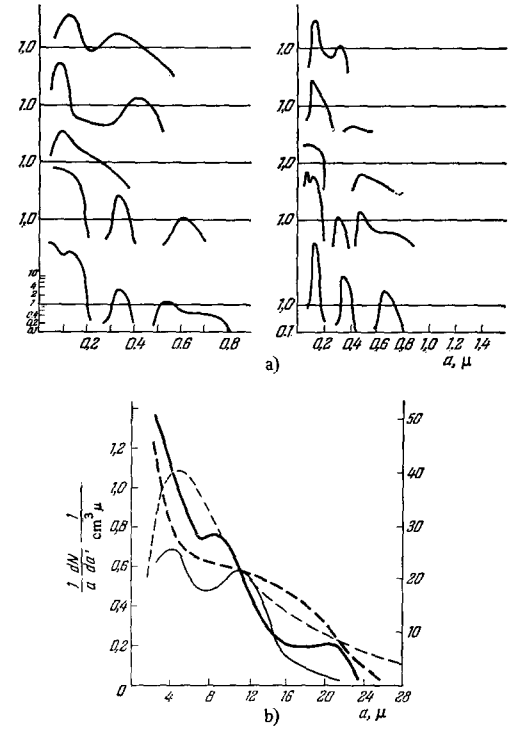

FIG. 6. Examples of concrete realizations of particle size distribution: a) N/cm^3 50 nm (Δa) for clear weather - as measured by R. Fenn; b) for fogs - as measured at ZNB (left scale - for the solid curves, right scale - for the dashed curves).

averaging scales greatly exceeding the time and the area of coherence of the field of the scattered light. At very small scattering angles φ, the coherence time is close in order of magnitude to $2\lambda/u\varphi$ (λ—wavelength of light, u—velocity of Brownian motion of the particles), and can reach several seconds. At not too small φ, the coherence time decreases rapidly to values on the order of $\lambda/u \ll 1$, and the notion that scattering by the different particles is independent is rigorously justified.

If we approximate the scattering particles by spheres or ellipsoids (homogeneous or layered) made up of matter having a known refractive index n and absorption coefficient κ, then the problem of calculating their optical properties reduces to the use of the Love-Mie theory. The initial stage of development of this theory was completed by the well known monograph by K. S. Shifrin[2], published in 1951. The new stage of development of the theory is connected with the use of electronic computers, which have made it possible to tabulate extensively the numerical solutions and subsequently analyze them qualitatively. By now we already have very extensive and continuously growing collections of tables (primarily[45-52]), which make it possible to answer without particular difficulty questions pertaining to the influence of the optical parameters of the particle on its optical properties. A detailed bibliography can be found in[3,53].

A general analysis of the optical properties of the small particles of spherical, elliptical and cylindrical form is contained in[3]. A generalization of the Love-Mie theory to the case of scattering of inhomogeneous plane waves by spherical particles is given in[54], and to the case of scattering by a pair of mutually-illuminat-

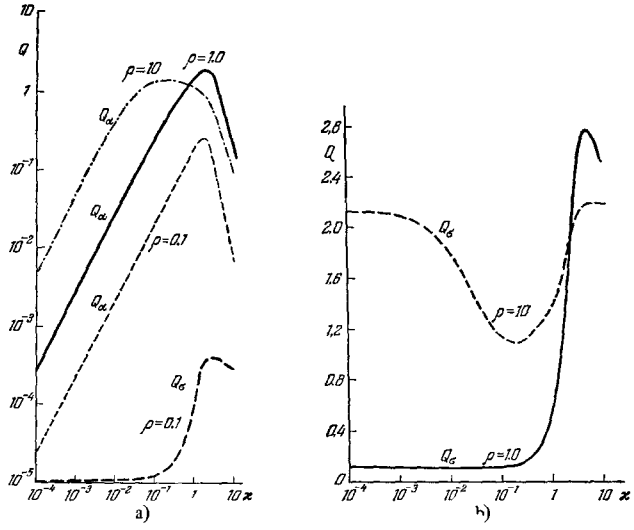

FIG. 7. Plots of Q_α and Q_σ against κ for ρ = 0.1, 1.0, and 10 (n = 1.33).

ing particles is given in[55]. Inasmuch as condensation usually takes place on insoluble particles, it was important to investigate scattering by water drops containing a nucleus of a different substance[56-58]. A number of workers proposed more or less effective approximation formulas on the basis of an analysis of the numerical solutions and the structure of the theory (see[3,59,60]). In particular, the "approximation of soft particles," corresponding to the condition $|n - i\kappa - 1| \ll 1$,[3] has found wide application. A large number of papers have been devoted to an experimental verification of the theory in the microwave band, using polymer models (for a recent bibliography see[32-34]. Referring the reader for details to this extensive group of investigations, which is worthy of a special review, we shall stop to discuss only some aspects which are particularly important for what follows.

The probabilities of scattering and absorption of light by a particle are best characterized by the so-called effectiveness factors Q_i, defined as the ratio of the cross section for the corresponding (i-th) process to the geometrical cross section of the particle. According to the Love-Mie theory, these factors depend only on the dimensionless radius of the particle $\rho' = 2\pi a/\lambda$ and on the refractive index and absorption coefficient of its material. Accordingly, the volume absorption coefficients (α) and scattering coefficients (σ) for the medium in which the particles of given radius are suspended are equal to the effectiveness factor (Q_α or Q_σ) for one particle multiplied by $3\pi v/2\rho\lambda$, where v is the total volume of the particles per unit volume of the medium (the so-called filling factor). Whereas the quantities σ (or Q_σ) characterize essentially the effect of air turbidity due to the disperse phase, the quantities α (or Q_α) determine the direct role of this phase in the radiative heat exchange.

Figure 7 shows plots of Q_α and Q_σ against κ for three values of ρ, taken from[57]. When $\kappa\rho \lesssim 0.1$ (i.e., so long as the attenuation of the beam penetrating through the drop does not exceed 10–20%), the absorption of light by the material of the drop has practically no influence on Q_σ, and Q_α is proportional to κ, the

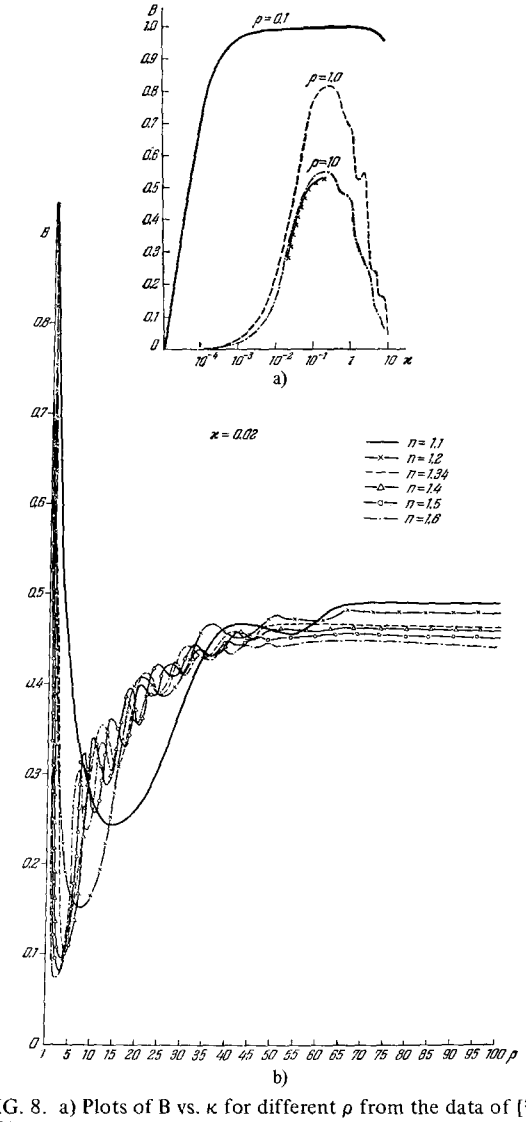

FIG. 8. a) Plots of B vs. κ for different ρ from the data of [52] and [51] (crosses) (n = 1.33). b) Plots of B vs. ρ for κ = 0.02 and different n from the data of [51].

proportionality coefficient being strongly dependent on ρ. The point is that the effectiveness of absorption of light by the drop is determined not only by the value of κ, but also by the effectiveness of penetration of the external electric field inside the drop. The same factors will influence, say, the photoluminescence yield or the Raman-scattering intensity of the disperse phase. This makes it possible, on the one hand, to replace the difficult measurements of Q_α by measurements of other quantities, and also to use these measurements to obtain information on the particle dimension distribution.

When $\kappa\rho \gtrsim 0.1$, an increase of κ already begins to affect strongly the magnitude of the Fresnel coefficients, as a result of which the reflection of light by the particle (i.e., Q_σ) increases and the field penetrating in it (i.e., Q_α) decreases. This can be seen particularly clearly by turning to the behavior of the relative probability of capture of a photon by a particle during the photon scattering, i.e., to the quantity $B = Q_\alpha/(Q_\alpha + Q_\sigma) = \alpha/(\alpha + \sigma)$ (Fig. 8a).

The curves shown in Fig. 8 were calculated by the

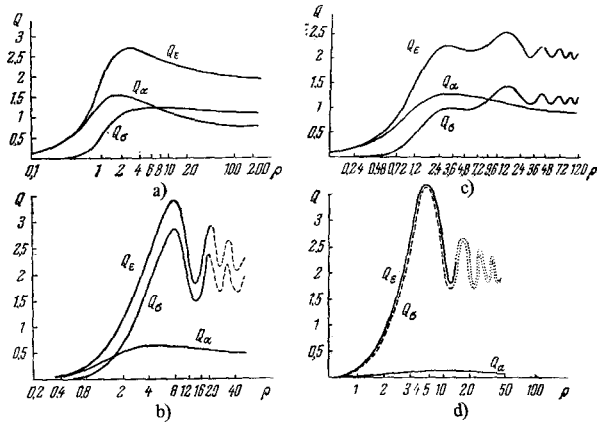

FIG. 9. Plots of Q_α, Q_σ, and $Q_\epsilon = Q_\epsilon + Q_\sigma$ against ρ for water particles having in their center strongly absorbing solid spheres (n = 1.59 and κ = 0.66) with dimensionless radius $\rho' = \rho/c$. a) c = 1; b) c = 1.2; c) c = 2; d) c = 5.

author[61] from the data of[51] and[52]. Attention is called in Fig. 8b to the monotonic decrease of B with increasing ρ in the case of small ρ ($B \sim \kappa\rho$), to the existence of a minimum when ρ is on the order of several units (as a function of n), followed by an approximately linear increase of B with increasing ρ ($B \sim \kappa\rho$)[62], and finally, to the independence of B of ρ ($B \leq 1/2$) when $\kappa\rho \gg 1$. The dependence of B on ρ for different values of κ is qualitatively the same. Thus, a change in the particle dimensions affects strongly the probability of photon capture by the particle, and by the same token its role in the radiative heat exchange. Both sufficiently large ($\kappa\rho \gg 1$) and sufficiently small ($\rho \leq 2-3$) particles turn out to be quite active in this respect, whereas particles belonging to the medium-disperse range take practically no part in the radiative heat exchange. Allowance for this circumstance is important also when determining the rate of radiative heating and evaporation of water drops (see[63]).

Figure 9 illustrates the influence exerted on Q_α, Q_σ, and the extinction effectiveness factor $Q_\epsilon = Q_\alpha + Q_\sigma$ both by the dimension ρ of the water particles and by the presence of a strongly absorbing sphere in the center of the water drop (n = 1.59, κ = 0.66) with dimensionless radius $\rho' = \rho/c$, as calculated by R. Fenn and H. Oser[58]. The presence of such an insoluble nucleus changes greatly almost all the optical characteristics of the drop (for example, the rainbows vanish). However, if we confine ourselves only to the effectiveness factors, then the presence of the nucleus imitates a change in the refractive index and the absorption coefficient of the drop material. In particular, this casts doubts on the methods used by K. Bullrich and coworkers[64,65] to make the constants n and κ of the material of atmospheric aerosol.

Following the experimental observation in 1967, by the present author and his co-workers, of the elliptic polarization of light scattered by the aerosol[66], and the establishment[8] of a strong dependence of the components f_{ik} of the normalized matrix $\hat{f}$ of light scattering by the aerosol on the scattering angle and on the meteorological conditions (concerning the scattering matrix see[1,3,8,9] and in more detail in[67]), it became necessary to calculate theoretically all the components of the matrix as a function of the parameters of the scattering particle.

The general character of the light scattering matrix $\hat{f}(\varphi)$ for spherical particles was established already in the 40's independently by the author and by F. Terrin (see[1,3,62]) namely:

$$\hat{f}(\varphi) = f_{11}(\varphi)\begin{pmatrix} 1 & \tilde{f}_{12}(\varphi) & 0 & 0 \\ \tilde{f}_{12}(\varphi) & 1 & 0 & 0 \\ 0 & 0 & \tilde{f}_{33}(\varphi) & \tilde{f}_{43}(\varphi) \\ 0 & 0 & -\tilde{f}_{43}(\varphi) & \tilde{f}_{33}(\varphi) \end{pmatrix}, \qquad (3)$$

where $\tilde{f}_{ik} = f_{ik}/f_{11}$ is the component of the reduced scattering matrix. For molecular scattering

$$f_{11}(\varphi) = \frac{3}{4+3d}(1+\cos^2\varphi+d), \quad \tilde{f}_{12}(\varphi) = -\frac{\sin^2\varphi}{1+\cos^2\varphi+d},$$
$$\tilde{f}_{33}(\varphi) = \frac{2\cos\varphi}{1+\cos^2\varphi+d}, \qquad \tilde{f}_{43}(\varphi) = 0,$$

but

$$\tilde{f}_{22}(\varphi) = \frac{1+\cos^2\varphi}{1+\cos^2\varphi+d}$$

in place of $\tilde{f}_{22} = 1$ for spherical particles. The quantity d takes into account the depolarization of the light as a result of the asymmetry of the molecules (the Cabannes correction). In general the deviation of $\tilde{f}_{22}$ from unity is evidence of the asymmetry of the randomly oriented scattering particles, while the optical anisotropy of the medium as a whole is manifest by the appearance of new components of the matrix in place of the zeros in formula (3)[3,8,9,66,67].

The first theoretical calculation of the components of the matrix (3) was performed by V. S. Malkova[68], who used approximate methods and $\rho \leq 1$. This investigation revealed incidentally that the method developed by K. S. Shifrin[2] for calculating the scattering matrix not on the basis of the Love-Mie theory, but by solving an integral equation, leads to partially erroneous results, namely, to identical vanishing of f_{43} in any approximation, possibly as a result of an improper approximation of the boundary conditions.

Recently K. S. Shifrin and I. L. Zel'manovich[51] calculated the angular dependences of all the components of the matrix (3) for a broad range of values of ρ, n, and κ. The results of the individual calculations were published also in[64,69,70]. By way of an example, Fig. 10 shows the dependence of the degree of ellipticity of

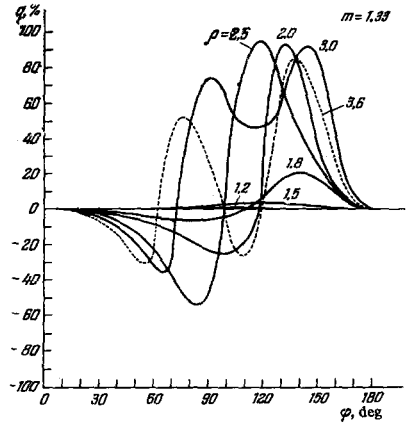

FIG. 10. Angular dependences of the degree of ellipticity of the polarization of the scattered light on the scattering angle φ for drops of water of different dimensions.

polarization q of light scattered by water droplets of different dimensions, on the scattering angle φ (in this case $q = \tilde{f}_{43}$), as calculated by G. I. Gorchakov[70]. However, the extent of the performed calculations is still insufficient for a complete analysis of the behavior of the scattering matrix. This pertains particularly to ellipsoidal particles, let alone particles with irregular shape, for which the theoretical calculations of the scattering matrix are generally nonexistent.

The availability of a sufficiently extensive collection of tables of optical characteristics of spherical particles, and the possibility of their easy supplementation by computer calculations or by approximate formulas, have made it possible to proceed to study the influence of the character of the distribution of the particle dimensions on the optical properties of polydispersed aerosol. This gave rise to a very extensive cycle of investigations by many workers (see, for example,[64,69,71-81]), devoted to calculations of all possible optical characteristics of aerosol at various particle size distributions.

When dealing with spherical particles, the optical characteristics of polydispersed aerosol can be represented in the form of an aggregate of the corresponding volume coefficients

$$G_i(\lambda, \varphi) = \int Q_i(\lambda, \varphi, a)\, u(a)\, da, \qquad (4)$$

where Q_i are effectiveness factors for a single particle and

$$u(a) = \pi a \frac{dN}{d \ln a}$$

is the reduced spectral density of the particle size distribution. Referring the reader to the cited papers for details, we note only that the dependences of Q_i on λ, φ, and a are very erratic, since in final analysis they are determined by the features of the interference phenomena occurring in the drop. Therefore the introduction of the particle size distribution smoothes out appreciably all the obtained curves.

Once $Q_i(\lambda, \varphi, a)$ and u(a) are specified, the determination of $G_i(\lambda, \varphi)$ reduces to direct integration, i.e., it assumes an elementary form. By now the efforts of the already mentioned authors and many others have resulted in the accumulation of many calculations with the aid (4), adding up to a highly developed numerical experiment that gives a fairly clear idea of the influence of different distribution parameters and of the properties of the polydispersed aerosol. By the same token, the difficulties have been transferred to a different region, namely to the question of the extent to which the chosen parameters correspond to reality.

A direct verification of (4) is possible only for fogs and clouds containing only water drops in the coarse dispersed range, for only then can the Love-Mie theory be reliably used to determine Q_i and to measure u(a) directly. A verification of this kind, performed by a number of authors (see, for example,[83]), has led to satisfactory agreement, provided the distribution u(a) is used for a given concrete realization. On the other hand, the use of stylized distributions of the type (2) leads to appreciable divergences.

For haze, when the scattering particles belong in submicron range, a direct measurement of the particle size distribution is so far practically unrealizable. It

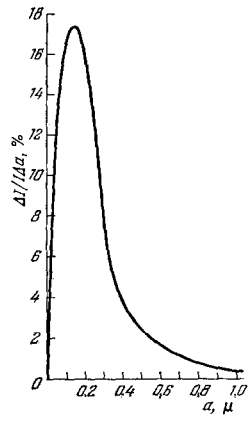

FIG. 11. Efficiency of the scattering of light by different fractions of the atmospheric aerosol, for a Junge distribution, at $\varphi = 45°$ [89].

is therefore customary to use for haze a Junge-type distribution (see (1)), which pertains to the dry core of the disperse phase. The distribution parameters (the quantity ν, the upper and lower limits of the distribution) and the particle constants n and κ remain in this case arbitrarily chosen.

We note in this connection that if the disperse phase is dry, i.e., a distribution of type (1) is applicable to it, the particles have an irregular form and the Love-Mie theory is applicable to them. Moreover, experiments show that the optical properties of an aggregate of randomly oriented particles of irregular form are not modeled in any way by any aggregate of spherical particles, as is sometimes assumed. On the other hand, if the disperse phase consists of liquid drops, then the Love-Mie theory is applicable to it, but the distribution (1) is not. This is confirmed, for example, by the disparity between the scattering indices that follow from (4) and (1) and experiment, as noted by number of authors (say[87,88]) (for details see Sec. 5).

We have already noted that the least reliable are the data on the distribution of particles by dimensions for the 0.05—0.5 μ range and on the nature of the particles for the range of the Aitken-nuclei.

At the same time, the range of large particles is the most active optically, and when account is taken of the increasing particle concentration in haze and the decrease of a—the 0.1—0.3 range is the most active (Fig. 11). Therefore the optical investigation methods, which furthermore do not involve interference with the state of the aerosol, are presently the most promising for the study of the microstructure of the aerosol in the medium-disperse range.

We thus arrive at the problem of finding the form of the particle size distribution, and also of finding the optical properties of the particles themselves, on the basis of a study of the optical properties of a real polydisperse aerosol. This problem is a particular case of the more general problem of applying optical methods to the investigation of the properties of matter, and is worthy of an independent discussion. The point is that, after a certain period of hopefulness and enthusiasm, very serious difficulties have appeared and erased almost completely the results previously obtained by this method.

These difficulties are of fundamental character, and effective means for overcoming them successfully have been suggested only quite recently.

4. OPTICAL METHODS OF INVESTIGATING THE ATMOSPHERE AS PART OF THE GENERAL PROBLEM OF INDIRECT MEASUREMENTS

The starting point for any optical method of sounding of a light-scattering medium, be it a stellar planetary atmosphere, a gas-filled lamp, or a handful of powder on a laboratory table, is a set of various experimental data on the spectral, angular, or spatial structure of the light field in this medium under known conditions of its illumination (see, for example,[20,90]). Knowing, furthermore, the laws of propagation (transport) of the radiation in the medium, we can, in principle, extract from this structure information on the phenomenological parameters G_i of the medium as a whole—its volume absorption coefficient and scattering coefficient, its scattering index, etc.[8,9,20,91-94]. To this end it is necessary to invert the problem of light propagation in a medium, making it necessary in final analysis to solve an integral equation of the type[20]

$$\hat{P}(\lambda, \bar{r}, \bar{s}) = \int_0^\infty \hat{w}(\lambda, \bar{r}, \bar{s}, l)\, p(\lambda, l)\, dl, \qquad (5)$$

where $\hat{P}$ is the experimentally measured transport matrix, characterizing the probability that a photon with wavelength λ will fall on a point $\bar{r}$ from a direction $\bar{s}$; $\hat{w}$ is the probability that the photon reaches this point in a given state of polarization after passing a path l, in the absence of absorption, while $p(\lambda, l)$ is the probability that the photon will traverse the path l without being absorbed.

A distinguishing feature of this equation is, in particular, that $\hat{w}$ and p depend on the properties of the medium only via its phenomenological parameters G_i, and the matrix $\hat{P}$ contains the entire information obtainable by optical methods concerning these parameters. Therefore the role of the transport theory reduces to a determination of the dependence of $\hat{w}$ and p on G_i, and the purpose of the experiment is not only to determine the G_i themselves, but also the relations between G_i and $\hat{w}$ or p corresponding to the given realization of the parameters. This is possible only when we have a system of equations of the type (5), i.e., when the experiment is comprehensive[8,9,20,91-96,98], or when certain model requirements are stipulated ad hoc. Under natural conditions, the described situation becomes aggravated by the variability of the investigated object and the ensuing need for obtaining an entire set of initial data simultaneously, since a real medium will certainly always differ strongly from any a priori model, and the task of the investigation usually becomes precisely the determination of the laws governing the changes of the parameters of the medium.

Further, if we regard (5) as a functional dependence of $\hat{P}$ on G_i, then the theory of radiation transport, which has become one of the most important branches of mathematical physics in the last quarter century, has a bearing only on the direct problem—methods of finding $\hat{P}$ for given G_i. The solution of the inverse problem, i.e., the formulation of an algorithm for finding G_i from the known values of $\hat{P}$, no longer pertains properly to transport theory, and is the subject of the general theory of indirect measurements, which has its own methods of analysis. This constitutes in particular, the principal difference between the work by the author[8,9,20], pertaining to the problem of solving inverse problems, and the fundamental investigations of say, V. A. Ambartsumyan, K. Chandrasekhar, V. V. Sobolev, or E. S. Kuznetsov, which pertain to transport theory proper. We shall show below that the ability to solve direct problems is not necessary and is far from sufficient for the solution of the inverse problem.

The character of the inverse problem, and hence also the degree of its complexity, depends significantly on the choice of the experimental conditions. In particular, when multiple scattering of light does not affect the measured $\hat{P}$, it is possible to establish a direct analytic connection between the matrix $\hat{P}$ and the sought parameters G_i, and by the same token reduce the inversion of the transport theory to an elementary operation (see, for example,[20]). But such a possibility arises quite rarely, and under natural conditions it is necessary as a rule to resort to the general problem.

No matter how the transition from $\hat{P}$ to $G_i(\lambda, \varphi)$ is effected, it does not complete the problem, but leads to Eq. (4), i.e., it confronts us with the necessity of inverting it. If the dependence of the effectiveness coefficients Q_i on a, λ, and φ are known, i.e., if we know a priori the type of particles and the optical constants of the substance of the particles (as is the case, for example, in clouds or in fog), then the solution consists of finding the size distribution of the particles. On the other hand, if the nature of the particles is unknown, as is the case in haze, then it is again necessary to have a set of equations of the type (4) for a simultaneous determination of the Q_i. Replacement of the comprehensive experiment by arbitrarily associating various models with nature, as is still frequently done, usually results in falsification of the solution, just as the inversion of (5).

Finally, whereas inversion of (4) yields the effectiveness coefficients Q_i of the individual particles, to determine the shape of the particles and the optical constants of the material it is necessary to invert the theory of scattering of light by small particles.

Thus, the determination of the microphysical characteristics of the disperse phase from data of optical measurements (i.e., measurements of the matrix $\hat{P}$) is connected with the need for successively inverting three problems: the theory of propagation of light in a medium, the theory of cooperative scattering of light by the disperse phase, and the theory of diffraction by a single particle[20]. Each of these problems is connected with its own difficulties, which require special means to cope with them. The rare attempts made so far to solve such a "three-layer" problem as a whole are usually connected with a misunderstanding of the distinctive features of the theory of indirect measurements, and are doomed to failure from the very beginning, particularly if the initial experimental information is confined only to the dependence of one of the components of $\hat{P}$ on λ or φ.

The most widely used method of solving inverse problems is to construct a certain collection of optical models of the medium, to solve the direct problem for them, and then compare the actually observed properties of the object with the properties of its a priori specified models. This is the procedure used in planetary astrophysics[97], in the study of the optical proper-

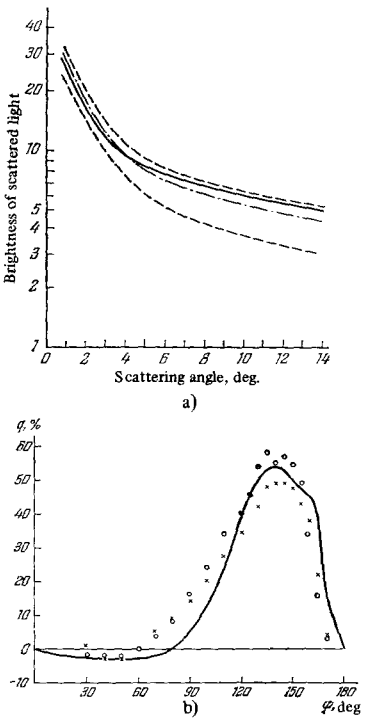

FIG. 12. Examples of the use of a priori models for the solution of the inverse problems of the theory of collective scattering. a) Brightness of the solar aureole. Solid curve - experiment, dashed - theory. Six adjusting parameters do not suffice to reconcile the data with the Junge distribution; agreement is reached when eight parameters are used [65]. b) Ellipticity of polarization of the scattered light. Solid curve - Junge distribution with five adjustment parameters [69]; - experiment of G. I. Gorchakov at ZNB.

ties of the atmosphere, for example by observations from outer space or from the earth's surface[26,65], in the study of the properties of the atmospheric aerosol[64,65,69], and not only in the aforementioned papers but in many others. Examples are shown in Fig. 12.

For natural objects, however, it is impossible to depend on results obtained in this manner. The many parameters involved in optical phenomena lead here to inevitable use of models with many parameters, even if the models are highly stylized. Thus, the Junge model contains as a minimum six parameters (the values of C and ν, the upper and lower limits of the distributions, and the quantities n and κ), which are used, in view of the uncertainty in their values, as adjustment parameters. As a result, the real relations are easily approximated by fictitious ones, with fictitious parameters (which are bashfully called "effective"). The real heuristic role of such model solutions of the direct problem is entirely different. They make it possible to analyze the sensitivity of the problem to variations of different parameters and to various types of noise (see, for example,[64,65,78-82,99,100]), ensuring by the same token the choice of the most effective means of solving the inverse problems.

The nature of the difficulties encountered on the way of solving the inverse problems has a rather general character and is now well known. From the mathematical point of view, we are dealing with the so-called incorrect problems of inversion of an integral operator of the first kind. The inversion of such operators, undertaken without suitable caution, involves the possibility of occurrence of false solutions, and also the "getting out of hand" of the solution, owing to its extremely high sensitivity to small disturbances. Physically, this involves the need for obtaining additional information in order to construct an algorithm that filters out the false solutions, and coarsening the solution as applied to the real volume of the initial information, so as to ensure its stability.

The mathematical aspect of this problem, for the case when the kernel of the equation is known a priori was analyzed by A. N. Tikhonov[101,102] (see also[103]), who developed a general method for the regularization of the solutions of such problems. The method is based on the construction of a regularizing and smoothing functional, the form of which is determined on the basis of the a priori information concerning the general character of the solution (but without its a priori concretization). The latter is found as the limit of a sequence of solutions obtained when the regularization parameter contained in the functional tends to zero. (A similar algorithm as applied to a more particular problem was proposed by Wark and co-workers[104-106].)

As a rule, however, only one general form of the kernel is known, but the values of a number of parameters determining the kernel must be experimentally determined separately for each concrete case. The impossibility of a priori concretization of the properties of the model raises primarily the problem of finding the composition of the experimental information that is necessary and sufficient for a unique determination of the parameters of the kernel as well as of the sought parameter of the atmosphere. Such a problem was clearly formulated and realized by the author, starting with 1946, in a long series of papers on the spectroscopy of light-scattering substances (see[20,91-93]), on projector[8] and twilight[9,95,107] sounding of the atmosphere, and also on the sounding of the atmosphere from outer space[108-114] and the study of the atmosphere of Venus on the basis of earth-based observations[97,98].

As a result, concrete algorithms were developed for the solution of an extensive class of inverse problems of the type of Eq. (5). The gist of such methods reduces to a finding of experimental situations in which the direct problem admits of an approximate analytic solution in general form, without a priori specialization of the properties of the medium, and a subsequent inversion of this analytic solution (numerical solutions do not admit of such an operation). The unknown properties of the medium (i.e., the characteristics of the kernel of the equation) then appear in the form of a few empirically-determined parameters of the solution, and the form of the latter dictates the necessary composition of the auxiliary experimental information for the measurement of these parameters.

The development of such methods is based essentially on introducing into the problem certain physical premises that determine the general structure of the solution and which limit the group of situations to which it is applicable. In particular, its applicability is restricted by the possibility of expansion in terms of a small parameter, which is different for each concrete case, and rapid termination of the series. The latter procedure, which determine the attainable accuracy, is

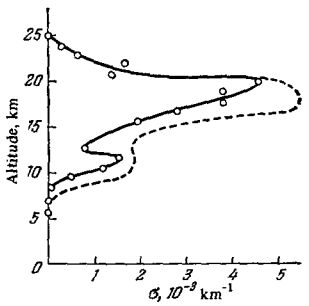

FIG. 13. One of the altitude dependences of the scattering coefficient in the dawn layer [109,113] (solid curve) and its reconstruction upon repetition of the procedure of solving the inverse problem [118] (dashed).

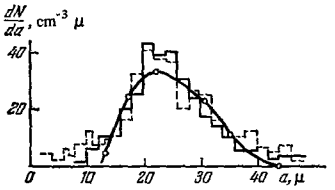

FIG. 14. Example of determination of the spectra of the scattering particles by a model medium (columns) using the aureole part of the scattering indicatrix, by the method of K. S. Shifrin (solid curve).

of fundamental significance, since it is the only feature that ensures regularization of the solution. In essence, we are dealing with an artificial decrease of the resolution of the method, to limits determined by the nature of the matter, when the approximated kernel of the integral equation plays the role of the apparatus function that smooths out the disturbing high frequencies in the noise spectrum[9,115,116].

The effectiveness and reliability of such an approach has been confirmed by extensive and varied material (see, for example,[20,107,117] and the subsequent sections). As one of the many examples, Fig. 13 shows the results of a determination of the altitude variation of the scattering coefficient from observations of dawn from the space ship "Vostok-6." The solid curve represents the results of the reduction of the measurement data[109]; from these data, the brightness of the dawn was obtained again by numerical solution, and the procedure of extracting information by the described method was repeated[118], the results being shown by the dashed curve. The determination of the type of information needed for optical studies of the microstructure of the disperse phase of aerosol, i.e., for the inversion of an equation of the type (4), is the subject of[8,66,70,96,119-127].

If the type of the necessary information is known, the question arises of the optimal organization of the comprehensive experiment, that is to say, the construction of the apparatus ensuring the best sensitivity to the variation of the measured quantity and the greatest independence of other circumstances. This is essentially the question of the interpretational value of the initial information. In particular form, the answer to this question follows from the algorithm for the inversion of the corresponding problem. In general form, for the case when the kernel of the equation is specified a priori, it was discussed by G. I. Marchuk[128], who used a brilliant procedure, that follows directly from the radiation transport theory, employing an equation for the usefulness of the radiation information. On the basis of this equation, it became possible to propose also certain algorithms for the solution of inverse problems, but these algorithms were not tested.

The problem of finding the size distribution of the scattering particles on the basis of optical information, i.e., the problem of inversion of (4) when the kernel Q_i is known, was analyzed in detail by K. S. Shifrin and his co-workers (see, for example,[82,129-137]). The efforts were aimed here at finding experimental situations in which the kernel of the equation can be approximated in a form that admits of exact inversion of the integral operator with the aid of some integral transformation. This turned out to be possible, in particular, for the aureole part of the index of scattering of light by coarsely-disperse particles[129-131], making it possible to propose a concrete method of solving the problem (an example of its realization with laboratory models is shown in Fig. 14[131]).

The method can be extended to larger particles by using either the spectral dependence of the extinction coefficient, or the form of the scattering index at large φ. However, the concrete procedures developed by K. S. Shifrin and co-workers for these cases[133,137] are essentially limited by the requirement of the a priori knowledge of the properties of the scattering particles—their sphericity, the values of n and κ, the admissibility of the so-called "soft" particle approximation (i.e., $|n - i\kappa - 1| \ll 1$). Therefore investigations carried out in this direction have so far predominantly a heuristic value, revealing the most promising experimental conditions and, principally, establishing the minimum amount of information necessary for the uniqueness of the inversion of the problem. One of the important results of this cycle of investigations was the conclusion that most of the information obtained earlier on the microstructure of the atmospheric aerosol by optical methods is scientifically inconsistent (or at best doubtful); this pertains in particular to the series of data reported in[27-31], or, let us say, to the attempt to solve this problem in[64,65,138-140] and many other papers of similar style. Unfortunately, although K. S. Shifrin's methodological investigations have advanced quite far, they have not been used so far to obtain actual data on the natural aerosol or on its transformations.

The possibilities of using any algorithm for the solution of inverse problems, including also the use of rigorous methods of inversion of solutions of the direct problem, are limited by the need for getting rid of the influence of noise of different origin. This includes the noise of the atmosphere itself, the instability and inhomogeneity associated with the atmosphere, noise in the measuring apparatus, "noise" in the measurement procedure (including limitation of the bandwidth) as well as in the procedure and technique of the subsequent calculations, and finally, the "noise" of the theory, which always schematizes both the reality and the experimental conditions. The presence of noise poses a new problem—that of finding the real amount of useful information delivered by the instruments. As applied to apparatus noise, this problem was considered from the general positions of set theory and information theory by V. P. Kozlov, who proposed concrete ways for its solution (predominantly for spectroscopy problems)[141,142]. From another quite particular position, namely the point of view of clarifying the real amount of independent data on the experimental curves for a known signal/noise ratio, the same problem was considered by S. Twomey[105].

If we generalize the already extensive experience in

solving inverse problems of the type considered by us, then we can readily see that any attempt to extract more information from the observations than is contained in them leads inevitably to a disruption of the solution and to false results. Therefore methods of solving inverse problems must be brought in correspondence with the real volume of the observational information. A rigorous inversion of the direct problem even if mathematically possible, far from frequently satisfies this requirement.

Thus, in K. S. Shifrin's methods, special measures turn out to be necessary to limit the information content of the solution and its regularization (see[131-137]). In the methods developed by the author[8,9,20,91-94,107-113], this role of artificially coarsening the solution is automatically performed by a method, justified by the physical analysis of the phenomena, of expanding the solution in a series which is subsequently cut off.

Another method of regularizing the solution of the inverse problem by introducing additional physical information and a smoothing procedure was proposed by M. S. Malkevich (see[143-145,147]). It is based on the use of experimental data on the statistical laws of variability of the very object of the investigation, and also on the noise field of external origin (including apparatus origin).

As shown by A. M. Obukhov[148], the optimal expansion (from the point of view of the speed of convergence) of an empirical function is an expansion in eigenvectors of the correlation matrix. If these vectors are known from direct observations of the variation field of the sought quantity, then the measurement object turns out to comprise a small number of expansion coefficients. The requirement of ensuring stability of the solution, i.e., of filtering out the noise, dictates in this case rapid termination of the series and retention of only the first two or three coefficients. This determines, by the same token, also the natural limits of accuracy of the sought information concerning the concrete realization of the given quantity. The physical content of M. S. Malkevich's method consists in the fact that the first (principal) terms of the expansion in the eigenfunctions of the correlation matrix separate out the influence of the main (most significant) physical parameters of a many-parameter medium, and these parameters determine under the given conditions the main statistical laws governing the variability of the medium, whereas the higher terms of the expansion characterize the influence of secondary factors, causing deviations from these laws. From this point of view, the next step should be a combination of the author's method with the method of M. S. Malkevich, wherein the former serves to reveal the principal law, and the latter is used to reconstruct the statistical deviations from this law. Experience in employing such a complex method, so far applied to a more modest problem of describing the aggregate of the scattering indices of atmospheric haze[120], have led to hopeful results.

The method of M. S. Malkevich was successfully tested in a number of various examples, particularly in[143-147]. Figure 15 shows an example, obtained by Yu. S. Lyubovtseva (ZNB), of the use of this method to find the size distribution of fog particles from measurements of the aureole part of the scattering index. This

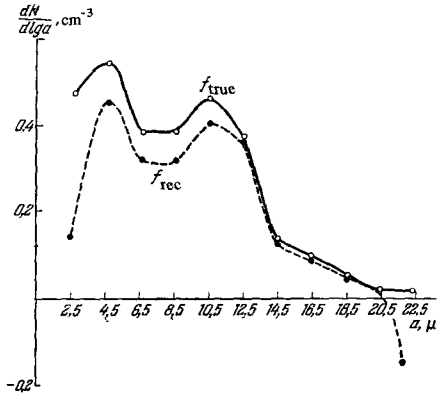

FIG. 15. Example of solution of the same problem for a real fog by the method of M. S. Malkevich. Solid curve - trap data; dashed - optical reconstruction.

method is also based on knowledge of the eigenvectors of the correlation matrix for fog particle spectra obtained from measurements made with flow-through traps. For finely-dispersed systems, such a straight-line procedure is not realizable, owing to the lack of suitable traps. But the method admits of a transformation that eliminates this difficulty. It consists essentially of expanding the optical data pertaining to a concrete realization in terms of the eigenvectors of the correlation matrix; for optical data, in turn, this reduces the problem to inversion, merely a few times, of eigenvectors that are specified once and for all (under these conditions, such an inversion can be performed with respect to an aggregate of many parameters, thus ensuring its reliability).

Thus, the main accomplishment of a decade of development of optical research methods is the clarification of the fundamental methodological problems, making it possible to devise a number of effective methods and to discard at the same time as unsuitable many of the previously employed primitive methods together with the doubtful results obtained with their aid. We present below certain results of investigations of the microphysics of the aerosol and the aerosol structure of the atmosphere, obtained by optical methods in recent years, with allowance for the distinctive features of formulation of inverse problems.

5. OPTICAL PROPERTIES OF AEROSOL AND MICROPHYSICS OF THE DISPERSE PHASE

The number of investigations aimed at studying the optical properties of air is constantly growing at an ever increasing rate. The world's literature already includes data on many thousands or even tens of thousands of realizations, for example[16,17,24,26-35,64-66,69,71,83,119-127] and many others. However, if we examine the entire aggregate of these papers we are struck by the fact that, with a few exceptions, all these papers contain collections of random facts that cannot be systematized, owing to the insufficient completeness of the experimental formulation. Only a few cycles of investigations, in which such completeness was ensured to some degree or another, led to deductions that are of scientific importance. A particular place is occupied here by the cycle of natural investigations performed at the ZNB

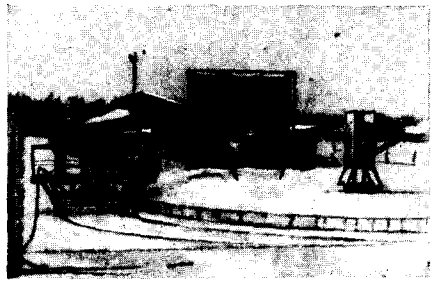

FIG. 16. Automatic-recording goniometric spectropolarimeter, which makes it possible to measure the polarization of light scattered by pure air, with a spectral resolution up to 3 Å. Radius of rail track 7 m.

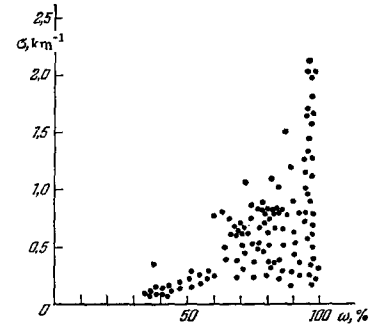

FIG. 17. Influence of relative humidity on the coefficient of scattering of light by air.

IFA[21,119,126,149-154], where a so-far unique assembly of measuring apparatus has been developed for simultaneous measurements of practically all the optical parameters of the atmosphere, under conditions of modern laboratory experimental techniques both in the sense of the technology and the apparatus, and in the sense of the attained degree of reliability and accuracy of the results (one of the installations is shown in Fig. 16). As a result of an analysis of the comprehensive optical data pertaining to several thousand concrete cases, it became clear that certain clearly pronounced regularities can be observed in the apparent chaos of the random realizations; these regularities are masked only by the fact that they occur under exclusively definite conditions, which are frequently confused during the analysis of the experimental data.

O. D. Barteneva[35,156] called attention, as early as in 1959, to the fact that the patterns of the scattering of light by the atmospheric aerosol break up into two or three distinct classes. It was necessary, however, to undertake an extensive program of a comprehensive optical study of atmospheric air in order to reveal the nature of these differences, and to establish that the situation involves qualitatively different states of the disperse phase, manifesting themselves outwardly as different types of optical weather[70,96,119-127]. In particular, it was established that a fundamental role is played by changes in the relative humidity of the air under transformations of its optical state. (The influence of humidity on the coefficient of scattering of light by air is illustrated in Fig. 17.)

These conclusions were subsequently confirmed in an extensive statistical analysis of the variability of the optical characteristics of atmospheric air[35], and also in a number of other independent investigations, which will be discussed later.

Although many details of the picture observed by us still require refinement, its basic features are traced out quite distinctly. It turns out that if one sets aside various types of precipitation (snowfall, rain, etc.), detailed information concerning which can be found in[35], it is possible to distinguish at least five qualitatively different types of optical weather:

1. Haze, due to penetration into the atmosphere of extraneous impurities: dust clouds, smoke from forest fires or from commercial objects, products of volcanic activity, etc.[31]. Haze has not yet been investigated from the optical point of view, and will not be discussed here.

2. Mist, which exists in the pre-condensation state of air and is the result of the growth of Aitken nuclei in the presence of atmospheric humidity. The physico-chemical mechanism of this growth is still not clear, nor is the chemical nature of the Aitken nuclei themselves (see below).

3. Misty fog, produced in the equilibrium-solution state of the atmosphere as a result of condensation of moisture on large and giant particles of soluble salts.

4. Fog and clouds, produced in the condensation state of the atmosphere as a result of a transition of an excess of the vapor-phase moisture into the liquid-drop phase.

5. Mist with drizzle—a heterogeneous formation, representing ordinary mist penetrated by drizzle, which falls from a higher-lying layer (mist does not coexist with rain).

Figures 18—22 illustrate some of the characteristic optical manifestations of each of the aforementioned types of the state of the disperse phase of atmospheric aerosol. (We have to present a relatively large number of these figures, since information of this type is fundamentally new not only in the optics of the atmosphere, but in optics in general.) A simple comparison reveals here the extent of the qualitative differences between these states. At the same time, Figs. 18—22 make it possible to find readily identifiable optical symptoms for each of the types of the optical weather.

The presented classification, based on features of the microphysical processes that regulate the state of

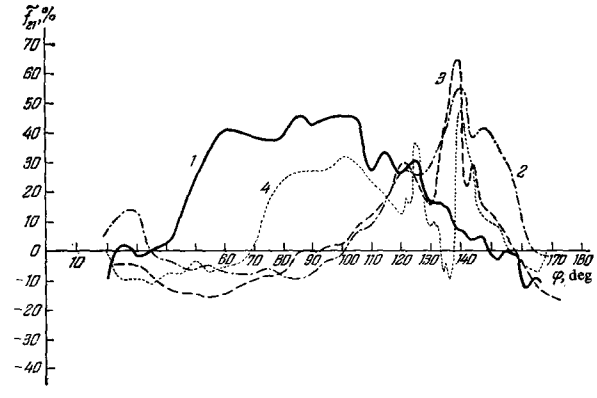

FIG. 18. Polarization indices of light scattering by mist (1), misty fog (2), fog (3), and mist with drizzle (4), obtained with high angular and spectral resolution [123].

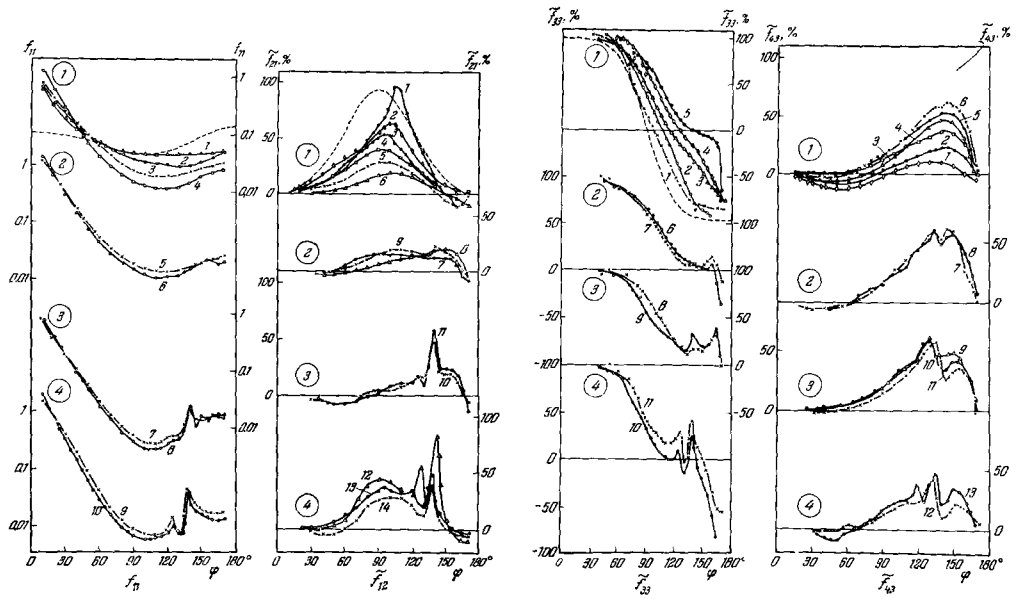

FIG. 19. Plots of the normalized monochromatic scattering indicatrices f_{11} and of the components of the reduced scattering matrix f_{lk} for mist (1), misty fog (2), fog (3), and mist with drizzle (4) vs. the scattering angle φ [121].

the disperse phase[96], follows clearly from the entire aggregate of the optical data[35,70,96,119-127]. However, until direct physico-chemical confirmations are obtained, it must be regarded as a hypothesis, albeit unique, which encounters no serious objections from any point of view, and which explains the entire aggregate of the facts known to us (and furthermore from a unified point of view). All other ideas concerning the variation of the condensation processes in a real atmosphere or concerning the resultant microstructure of the disperse phase, given for example in the monographs[27-30,71,83], must be regarded as inconsistent as soon as they are applied to humidities lower than 100%, since they either contradict the observations or else, at any rate, require a critical analysis.

Such a radical review of the physico-chemical foundation of our concepts concerning the nature of mist and misty fog calls for a radical change in the trend and in the tasks of further research in this field. On this basis, the foregoing classification was recommended by the All-Union Inter-administrative Conference on the Scattering of Light in the Atmosphere (Chrnovtsy, June 1967) for practical introduction. However, in order to implement such a recommendation it is necessary to have certain preliminary specialized investigations, all the more since this classification differs greatly from the presently employed classification in the hydrometeorological service, based on gradations of the visibility distance independently of the nature of the phenomenon (thus, misty fog, fog, and mist with drizzle are all classified as fog).

In this connection we emphasize once more that the distinct statistical regularities can be sought only within types of weather that are physically clearly outlined, and that if they are intermixed (as is done in meteorological practice), these regularities are almost completely erased. This, in particular, is the reason why practically all the earlier information on the optical properties of the real air do not lend themselves to systematization and in final analysis are lost to science.

Proceeding to a description of the main characteristics of each type of optical weather separately, we shall stop to discuss only those which are single-valued and reliable consequences of optical-research data. More complete information can be found in the original publications. We note that these data pertain primarily to the conditions in the vicinity of Moscow, and that in other regions the concrete parameters of the relationships may vary, but the possibility of a change in the general character of the relationships is more than doubtful, all the more since the different details of these

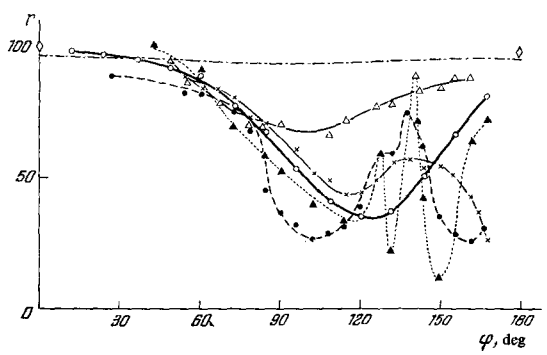

FIG. 20. Degrees of polarization coherence (in %) of the scattered light vs. the scattering angle for mist (○ and ×), misty fog (△), fog (●), and mist with drizzle (▲).

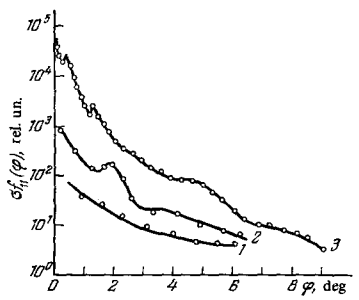

FIG. 21. Aureole parts of the scattering indicatrix for mist (1), misty fog (2), and fog (3) [125].

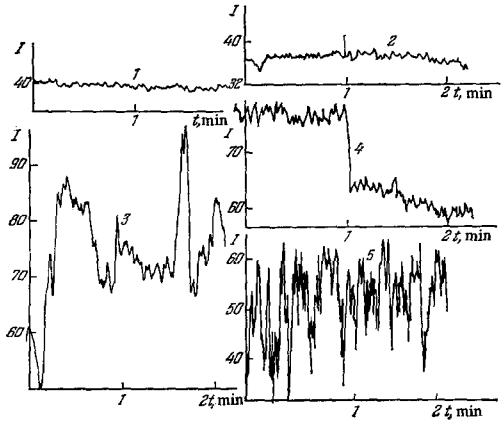

FIG. 22. Fluctuations of the brightness of light scattered at an angle of 20′ by mist (1), misty fog (2), the intermediate state between misty fog and fog (3), stable wreaths of fog (4), and falling snow (5).

relations are confirmed by the entire aggregate of data by many authors, obtained under a great variety of conditions. The only exceptions should be the oceanic regions, where, owing to the fact that the air contains a small number of Aitken nuclei and large particles (see Sec. 2), the mists should be highly rarefied, and misty fogs should be more largely dispersed than over continents. This should lead to an appreciable difference between oceanic air (which has remained practically unexplored) and the continental air also with respect to other optical characteristics. Similar conditions are possible also in the Antarctic. In exactly the same manner, with increasing altitude, the mists should become rarefied as a result of the rapid decrease of the concentration of the Aitken nuclei, in view of which, formations of the misty-fog type should be essentially observed at altitudes exceeding 2–3 km (above the convection layer) in the absence of clouds, as is indeed the case in the tropopause region (10–12 km)[8], in the luminous layer (15–22 km)[27,109,57], and in mother-of-pearl clouds (22–26 km)[8,158-160] according to optical-sounding data (see the next section).

Let us examine briefly the main features of each type of formations separately.

The surface layer of continental air is in the misty state at least 90% of the time. According to data of Yu. S. Georgievskiĭ, under the conditions near Moscow at very low humidities ($w \lesssim 30\%$), the scattering of the light by the disperse phase is due essentially to its dry (dust) fraction, which produces only weak turbidity ($\sigma \lesssim 0.05$ km^{-1}). With increasing humidity, the air becomes more turbid as a result of the appearance of humid mist (this becomes noticeable at $w \cong 30-35\%$), whereas the role of the dry large disperse fraction becomes vanishingly small[96].

A comprehensive analysis of the optical data[70,96,119-127] shows that the disperse phase of humid mist is characterized by not too narrow a size distribution (as is evidenced from Fig. 20) and by a very distinctly outlined upper boundary (this follows from the behavior of $\tilde{f}_{43}$ on Fig. 19). The latter is located near a $\cong 0.07-0.08$ μ at very low humidities, when the randomly oriented particles frequently turn out to be nonspherical (this is evident from Fig. 23), and shifts with

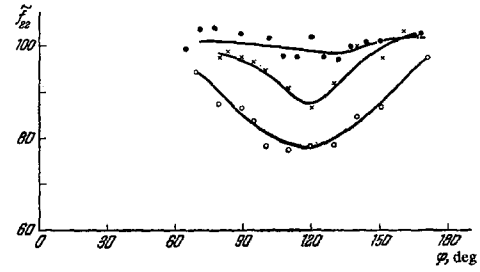

FIG. 23. Angular dependences of $\tilde{f}_{22}$ for mist with low humidity[70]. The difference between $\tilde{f}_{22}$ and unity is due to the nonsphericity of the particles [1,3,67].

increasing humidity gradually to a $\cong 0.20-0.25$ μ, when the particles become already strictly spherical. Consequently, although the details of the distribution of the particles by dimensions are not known to us, we can state that it does not correspond at all to the distribution proposed by Junge (3) (this conclusion is confirmed also by an analysis of the scattering patterns, performed by T. P. Toropova[87,88]), and more likely approaches the types of distributions characteristic of the Aitken nuclei (see Fig. 3) or else those observed by R. Fenn[36] (see Fig. 6) and K. Bullrich[37], and is possibly satisfactorily approximated (in accordance with an idea of L. Foitzik[73,74]) by aggregates of Gaussian distributions. The giant particles, if they do exist in mists, have a concentration not more than several particles in 1 m^3 of air.

Such microphysical characteristics of mist explain fully the entire aggregate of its optical properties, primarily the absence of any traces of rainbows, halos, or crowns, the smoothness of the dependences of each of the optical parameters on all the variables (see Figs. 17–21), and also the unusually high correlation between the values of all the optical parameters of the mist (for certain parameters, the correlation coefficients reach 95–98%) and its high temporal stability (see Fig. 22) and spatial homogeneity (for a climatological investigation of mists see[35]).

By way of an example, Fig. 24 shows the correlation dependence of the maximum value of the degree of ellipticity of the polarization of scattered light with a scattering coefficient σ[120]. Figure 25 shows typical examples of the numerous spectral relations of the scattering coefficient of mist and fog, obtained by Yu. S. Georgiev-

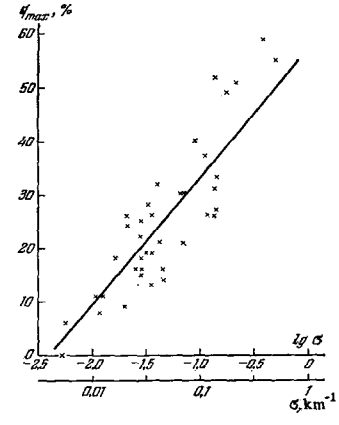

FIG. 24. Influence of the scattering coefficient σ on the value of the degree of ellipticity of the polarization of the scattered light at the maximum.

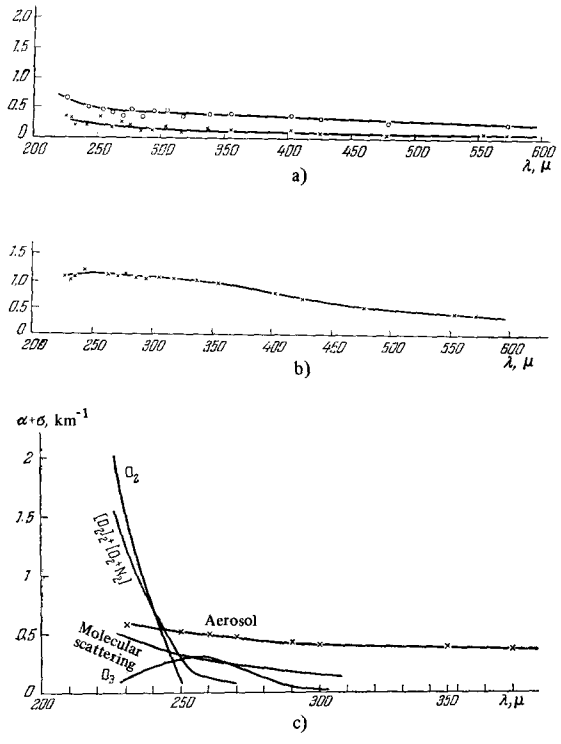

FIG. 25. Spectral dependences of φ for mist (a) and misty fog (b) and comparison (c) of the coefficients of scattering by the disperse phase with the coefficients of molecular scattering[155] and molecular absorption of atmospheric gases: oxygen [166,167], bimolecular complexes $[O_2]_2 + [O_2 + N_2]$ [152] and ozone (in the surface layer) [164]. (The ordinates of Figs. a) and b) are $\sigma_{aer}(\lambda)$, km^{-1}).

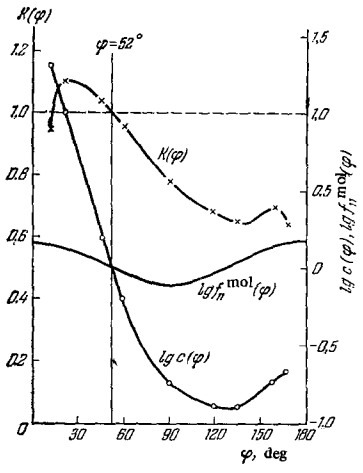

FIG. 26. Empirical functions $C(\varphi)$ and $K(\varphi)$ for $\lambda = 0.55\mu$.

skiĭ (for the first time in such a broad spectral interval and with high spectral resolution)[163]. The first is characterized by a monotonic increase of σ and $d\sigma/d\lambda$ when λ decreases down to $\lambda = 0.24\ \mu$. It turns out that in all the investigated range (including the ultraviolet), the scattering of light by the disperse phase certainly predominates over the molecular scattering, even in weak mists, and only in the region $\lambda \lesssim 0.25\ \mu$ is the principal role in the attenuation of light by the air assumed by molecular absorption[153,154], first by bimolecular complexes $[O_2]_2 + [O_2 + N_2]$, and then by molecular oxygen, and only to a very weak degree (under conditions near the earth's surface) by the ozone (Fig. 25c).

Yu. S. Georgievskiĭ has found that there exists a very close correlation between the variations of σ in different sections of the spectral range (mists are characterized by values of σ from 0.05 to 1.5 km^{-1} at $\lambda = 0.55\ \mu$).

An equally close correlation exists between σ and the form of the scattering pattern $f_{11}(\varphi)$[35,120]. For the scattering of monochromatic light by the disperse phase of mist, the dependence of $f_{11}(\varphi)$ on σ is of the form[120]

$$f_{11}(\varphi) = C(\varphi)\,\sigma^{K(\varphi)-1}, \qquad (6)$$

where the empirical functions $C(\varphi)$ and $K(\varphi)$ for $\lambda = 0.55\ \mu$ are shown in Fig. 26 (for other λ it is necessary to perform additional experiments). In this case $C(\varphi) \cong K(\varphi) \cong 1$ at the angle $\varphi = \varphi_0 \cong 52°$, i.e., the dependence of $f_{11}(\varphi)$ on σ vanishes and all the scattering patterns (including the Rayleigh pattern) intersect one another. For other λ, the angle φ_0 will obviously be different, as follows from observations[9,35,164] and the theoretical analysis[82]. However, as soon as this angle is known, measurement of the brightness of the light scattered at this angle can serve[35,168] for a sufficiently reliable determination of σ, in lieu of the more cumbersome procedure of its determination from the extinction coefficient or the integral of the scattering pattern (see, e.g.,[125]).

Returning to (6), we note that the patterns of scattering of light by mist fluctuate only weakly, for random realizations of the mist, in the vicinity of the average dependence on σ described by (6), and form, as shown by statistical analysis, a family with two essential parameters, one of which is probably the relative humidity and the other in all probability characterises the state of the Aitken nuclei. For a refinement of this question it is necessary to extend greatly the volume of the initial experimental material.

We can trace the following pattern of the transformation of the mist particles during the course of its condensation; this pattern differs in many respects from that adhered to by the authors of[27-31]. The initial dry fraction, consisting of large and giant particles of irregular shape, exerts only a slight dimming action. The Aitken nuclei are also optically indiscernible, owing to the smallness of their size, and also have in all probability an irregular shape. However, even at $w \cong 30$—40%, the Aitken nuclei begin to increase in size, as a result of capture of atmospheric moisture, reaching equilibrium dimensions for a given w. (The mechanism of capture of moisture still remains unclear; it is not excluded that this is a process similar to that of the swelling of polymers, or the dilution of a solution of volatile substances, or else the chemosorption mechanism; the possibility of an adsorption mechanism is completely excluded, since the volume of the particle increases by 8—10 times.) The largest fraction of the Aitken particles migrates over in this case into the most active region, from the optical point of view, of dimensions of approximately 0.1—0.3 μ, leading to a sharp increase of both the dimming of the atmosphere, and the ascertainable concentration of particles populating the medium-dispersion region. With further increase of w, the concentration of the optically active particles remains practically unchanged (obviously, owing to the discrete structure of the Aitken-particle spectrum), and the principal role is assumed by the growth of these particles, which become gradually rounder.

In this connection, the question of the chemical nature and origin of the Aitken nuclei becomes very important. At the present time, apparently, there is only one hypothesis worthy of consideration in this respect, since an attentive analysis of the experimental data makes all other assumptions inconsistent (see[27-31] and Sec. 2 above; in particular, measurements by E. S. Selezneva[155] as well as by a number of other authors[27-31] decisively reject the hypothesis of oceanic origin of the Aitken nuclei and their salt composition). We have in mind here the research by F. W. Went[169-172], which is based, on the one hand, on data of natural measurements with a gas chromatograph, and, on the other hand, on a number of considerations of general nature, which lead him to the conclusion that the Aitken nuclei have an organic origin.

F. W. Went assumes that terpene-like compounds, produced by the biosphere upon decay of organic cells, are subjected under the influence of the visible rays from the sun to a slow photochemical decomposition, forming volatile substances which are sensed by us as the aromas of pastures, forests, the sea, etc. Further, again under the influence of the visible rays from the sun, these substances are further transformed in the presence of ozone, iodine, or nitrogen compounds. Their end products form, as a result of coagulation or condensation (as was observed already by Tindal for photochemical reactions[173] and as is well known for combustion products), small lumps of organic (possibly latex-like) substances which act as the Aitken nuclei.

According to Went's chromatographic measurement data, which prove that at any rate an appreciable fraction of the Aitken nuclei is biogenic or anthropogenic, the average concentration of the decay products of the terpene-like substances in the atmosphere is approximately 10^4 $\mu g/m^3$ (i.e., on the order of 10^6 tons in the entire volume of the atmosphere), and their total production on the earth's sphere is close to 5×10^8 tons annually, in good agreement with the data on the number of Aitken nuclei.

F. Went's notion is in good agreement with the entire aggregate of optical data, and even explains some of its aspects that were previously unclear, for example the distinct change in the condensation mechanism at $w \cong 70-80\%$. However, the subsequent assumption made by F. Went, on the basis of rather superficial optical observations, namely that the mist is produced directly by growth of organic particles, contradicts the observed data on processes of rarefaction of mist, and also on the diurnal and seasonal variation of these parameters. The optical data make it possible to state that the scattering of light by the mist is due not directly to the "condensed odors," as assumed by F. Went, but either to droplets of their aqueous solutions, or else to other products of assimilation of atmospheric moisture.

It thus appears that a study of the nature and of the origin of the Aitken nuclei and the mechanism of their growth in the presence of atmospheric humidity becomes one of the most important problems not only of optics but also of the thermodynamics and chemistry of the atmospheric aerosol, since, as is now clear, the Aitken nuclei are among the most important factors determining the course of the condensational and hence weather-producing processes.

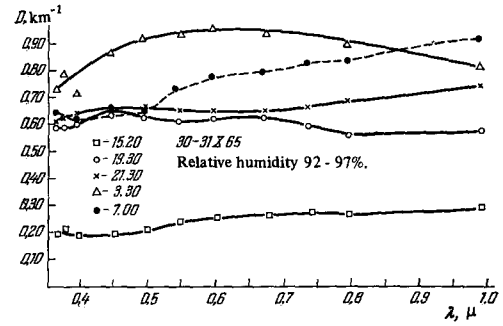

FIG. 27. Typical spectral dependences of the scattering coefficient of misty fog as measured by Yu. S. Georgievskiĭ.

The misty fog, which frequently is produced at the location of the mist when $w \gtrsim 70-80\%$ and condenses noticeably when w increases (with characteristic values $\sigma = 1-3$ km), has been much less thoroughly investigated. From optical data (see Figs. 17–21) it follows clearly that the occurrence of misty fog is connected with the appearance of a broad spectrum of particles in the interval from several tenths of a micron to 12–15 μ, with a distinct quantitative separation, against its background, of a fraction in the 1–5 μ range, which becomes particularly pronounced in the rainbow phenomena, and a relatively small fraction in the range 12–15 μ (causing the existence of halos). Simultaneously, there apparently takes place also a further increase of the mist particles, to dimensions approximately 0.3–1.0 μ, which leads to the formation of a corresponding fraction, also quantitatively separated, which becomes optically manifest in the appearance of a characteristic maximum on the spectral dependence of σ at $a_{max} \cong \lambda_{max}$ (Fig. 25b and 27). The particle dimensions in the 1–5 and 12–15 μ ranges increase regularly or decrease with change of w, as can be quantitatively traced in the behavior of the halos and rainbows.

Misty fog, like mist, is a stable state (see Fig. 22), and the processes of transition of one state to the other have a smooth character and take usually 10–20 min (approximately the same time as necessary to restore the mist after the atmospheric air is washed out by showers).

An entirely different and patently non-equilibrium character is possessed by the transformation of misty fog (less frequently mist) into fog, or the breaking up of a fog (see Fig. 22). The latter is also characterized by a broad spectrum of particle dimensions, encompassing the range from approximately 1 to 25 μ, against the background of which there appear quantitatively two fractions—near 8–12 and 18–25 μ, as is clearly manifest in the behavior of rainbows and the aureole part of the scattering indicatrix (see Figs. 18–22). A clear-cut anticorrelation of the particle concentrations in the ranges 1–5 and 8–12 μ shows that in the processes of occurrence or evaporation of a fog there takes place a repopulation of the particles from one of these ranges into the other. Owing to the coarsely disperse nature of the fog, the scattering coefficient in the visible region is practicalyl independent of λ (see, e.g.,[35,83]). In the infrared region (we are referring to the transparency windows), σ decreases weakly with increasing λ (approximately $\sim\lambda^{-1}$), has one or two weakly pronounced

maxima near $\lambda = 8-12\ \mu$, and then decreases rapidly near $\lambda \cong 20-30\ \mu$ (for misty fog approximately $10-15\ \mu$)[174].

Since the fog particle dimension is sensitive to small fluctuations of the temperature, turbulent fluctuations of the temperature lead to the formation of wreaths of fog (see Fig. 22), which are characterized by an increase of the particle dimensions in the condensation region and a decrease in the rarefaction region, as is clearly observed optically.

The optical properties of mist with drizzle are determined essentially by the finely-dispersed mist, and only in the region of the aureole and rainbows is the presence of the few precipitated mist drops, with characteristic sizes $80-100\ \mu$, become manifest.

It is clear from the foregoing that a comprehensive approach to the study of the optical properties of atmospheric aerosol in conjunction with the use of modern methods for both experiment and analysis has made it possible not only to produce effective methods of investigating atmospheric processes connected with the transformation of the disperse phase, but also obtain essentially new data, which in many respects have modified our concepts concerning the course of the condensation phenomena in the atmosphere. It must be borne in mind, however, that these are only the first steps in the use of the real possibilities of the optical means of investigation, and that the geophysical scales call for more extensive introduction of these methods, all the more since the results obtained in this manner reveal the existence of a perfectly concrete physical foundation for the construction of methods for not only diagnosing but also forecasting of the optical weather.

6. AEROSOL STRUCTURE OF THE ATMOSPHERE AND VERTICAL VARIATION OF THE SCATTERING COEFFICIENT

As a result of the use of new means of obtaining and methods of extracting optical information, major advances were made also in the understanding of the vertical stratification of the atmospheric aerosol and changed in many respects the previously held ideas. It has turned out that indirect methods of measurements not only can compete with direct methods in aerosol investigations, but have in many respects undisputed advantages, especially when dealing with the high layers of the atmosphere.

The most direct although most laborious method of investigating atmospheric aerosol is, of course, to simply gather the disperse phase at different altitudes with the aid of airborne apparatus provided with traps of various types. The data obtained by this method are described in detail in the monographs[27-31,155], and we confine ourselves only to a few examples of altitude dependence of the concentrations of the large particles[27] and of the Aitken nuclei[155], which are shown in Fig. 28.

It is important that the relative content of the Aitken nuclei and of the giant particles decreases rapidly with altitude—this is evidence of their genetic connection with the earth's surface. To the contrary, the altitude distribution of the large particles is characterized by a

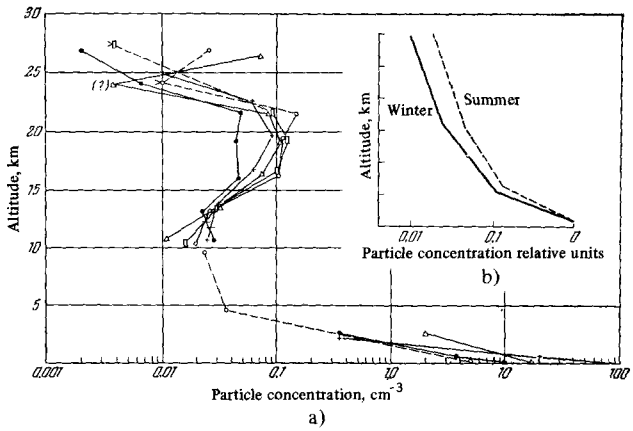

FIG. 28. Examples of altitude variations of the particle concentration, as measured by different authors. a) Large particles [27]; b) all ranges (mostly Aitken nuclei) [155]. In Fig. b), one division on the ordinate axis equals 1 km.

practically constant relative concentration, violated only by the existence of more or less stable maxima—primarily the recently observed maximum at altitudes 15—22 km.

In addition, it has turned out that the disperse phase in the stratosphere consists predominantly of crystallites of ammonium sulfate salts[27]. The latter, most likely, are generated directly at altitudes of approximately 20 km[27], possibly from volcanic gases[157] (this is evidenced by the distinct correlation between the optical manifestations of the stratospheric aerosol and volcanic activity; see[9,157], and also[175-177]), and one might also think that ozone takes part here, too[27,157]. It must be especially emphasized here that the most recent investigations lead more and more persistently to the conclusion that most condensation nuclei are of atmospheric origin, and that they are produced in a great variety of processes from the gas phase and subsequently grow and become transformed.

It is customary to assume that above 25—20 km the atmospheric aerosol is in the main the product of the penetration of meteoritic matter into the upper layer of the atmosphere, and that this matter becomes further transformed (disintegrated, evaporated, recondensed) and gradually settles down to the troposphere, which it reaches after approximately a month. However, analysis of aerosol samples obtained in 1962 in Sweden by rocket sounding of silver clouds (see, e.g.,[178-180]) apparently contradicts such a hypothesis.

The foregoing information pertains only to the dry base of the disperse phase, and not to its state in situ, which, as is now clear, can be quite different.

This is illustrated by Table III, where the data obtained by different methods on the scattering coefficient σ, optical thickness τ, average (or effective) particle radius a, particle concentration N, and specific volume v are compared for the luminous layer of the aerosol at an altitude 15—22 km, mother-of-pearl clouds (21—25 km), and silver clouds (82—85 km).

Both Table III and the data obtained by direct gathering of samples[37,178-180] offer evidence that, at any case below 30 km and in the vicinity of 82 km, the disperse phase of the stratospheric aerosol is practically never

Table III. Comparison of certain data on stratospheric aerosol, obtained by optical methods and by gathering samples

	Dawn layer		Mother-of-pearl clouds		Silver clouds	
	Gathering of samples with an airplane [181]	Optical observations from "Vostok-6" [109]	Projector sounding [8,158-160]	Observation of halos [182]	Gathering of samples with rocket [178-180]	Twilight observations from the earth* [183-185]
σ, km^{-1}	10^{-3}	$(2-5)\cdot10^{-3}$	$(4-6)\cdot10^{-2}$	?	?	10^{-2}
τ	$7\cdot10^{-3}$	$15\cdot10^{-3}$	10^{-1}	?	?	10^{-2}
a, μ	0,15	0,5—0,6	0,7	1,0	0.03—0.04	0.1—0.4
N, cm^{-3}	1	1	4—11	?	?	10^{-2}—10^{-3}
v	10^{-14}	$5\cdot10^{-13}$	10^{-11}	?	—	10^{-15}—10^{-16}

*Polarization measurements show that the particles of the silver clouds are not spherical and that their orientation in space is not isotropic [183].

made up of dry particles, and appears only in the form of droplets of solutions, or else in the form of solid particles clad with water or ice jackets.

This is also the cause of the strong variability of the disperse phase, which is characteristic of the entire altitude interval (at least up to 90 km), posing the problem of investigating the aerosol (and optical) weather in the stratosphere. Thus, according to data of twilight observations[9], the brightness of the sky at large altitudes changes from day to day by several times in the blue region of the spectrum, and several dozen times in the near infrared region. Therefore optical methods turn out to be so far the only ones capable of supplying information on the state and processes of transformation of atmospheric aerosol at large altitudes.

As optical indicators of the state of the atmospheric aerosol and its vertical stratification, one can use the altitude and spectral dependences of the scattering coefficient σ of air, its scattering matrix $f_{ik}(\varphi)$, and its optical thickness

$$\tau(h) = \int_h^\infty \sigma(h)\, dh$$

over the head of an observer situated at an altitude h.

In the lower layers of the troposphere, τ(h) can be easily measured, say by the Bouguer method, from the brightness of the sun (see, e.g., [168]); as shown in[9], the use of a star for this purpose (see[8]) entails noticeable errors due to the scattering of the light of the remaining stars by the air. Similar measurements were made many times (see[1,8,9,168]). From among the later investigations, notice should be taken of[186]. The errors in the determination of σ, with allowance for the natural inhomogeneity and instability of the atmosphere, amount to approximately 30—50% when averaged over a layer approximately 1 km thick. Above approximately 6 km, however, such measurements become difficult, in view of the smallness of the effect, so that other methods must be employed.

A detailed theoretical and experimental analysis[9-13,26,37,64,65,71,168] and others) led to the conclusion that the brightness B of the clear daytime sky at an altitude h, in high sun, is a unique function of τ(h) (in the first approximation it is proportional to τ(h)). Therefore measurements of B at different altitudes can serve as means of determining the altitude dependences of τ and σ, especially if the recommendations proposed by E. V. Pysakovskaya-Fesenkova and her co-workers are adhered to[188].

Earlier work in this direction, described in the reviews[1,8,9], are mostly obsolete, as is also the special review[187] devoted to them. By now such measurements have been extended to considerably greater altitudes, using for this purpose airplanes[188,189] (from 8 to 17.5 km), balloons[190] (up to 30 km), and rockets (up to 80 km[191-193] and from 90 to 450 km[138-140]), and in[188,189] a study was also made of the scattering indices and of the seasonal variability of σ. The error in the determination of the scattering coefficient reaches in this case approximately a factor 1.5—2 (equivalent to an error $\Delta h \cong 3-5$ km), and the error in the averaging over the altitudes from 1 to 5 km depends on the measurement conditions.

The theory of twilight phenomena developed by the authors of[9], which explains their main hitherto-understood features[9,94,107,194,195], led subsequently to the conclusion[196] that land-based measurements of the brightness of the twilight sky can be converted, with sufficient accuracy, to the brightness possessed under the same atmospheric conditions by the daylight sky as seen by an observer located at a certain effective altitude $\bar{h}$, which depends on the observation conditions. This makes it possible to dispense with expensive flying apparatus and create a land-based network of regular observation of the variability of the stratospheric aerosol.

Figure 29 compares the measured altitude variation of the brightness of the daytime sky, obtained by different methods for altitudes from 8 to 450 km. The discrepancies lie within the limits of the normal variability of the atmosphere itself (see[8,9]).

As follows from the specially developed theory[9,107], land-based measurements of the spatial and spectral pictures of the twilight sky at different dips of the sun under the horizon have made it possible to determine σ(h) in the altitude interval from 30—40 to approximately 90—100 km with an error not larger than 150% and uncertainty of approximately 3—4 km in the altitude[107,202]. Similar methods can be extended also to the study of the global variability of the stratospheric aerosol, using observations in the region of the terminator from a spaceship[9,111,112] or from an orbital comprehensive optics station (OCOS) of the "Kosmos-149" type ("cosmic arrow," Fig. 30), which makes it possible to measure the spatial and angular structures of the brightness field of the planet in the ultraviolet, visible, and infrared regions of the spectrum through narrow-band optical filters and specially intended for

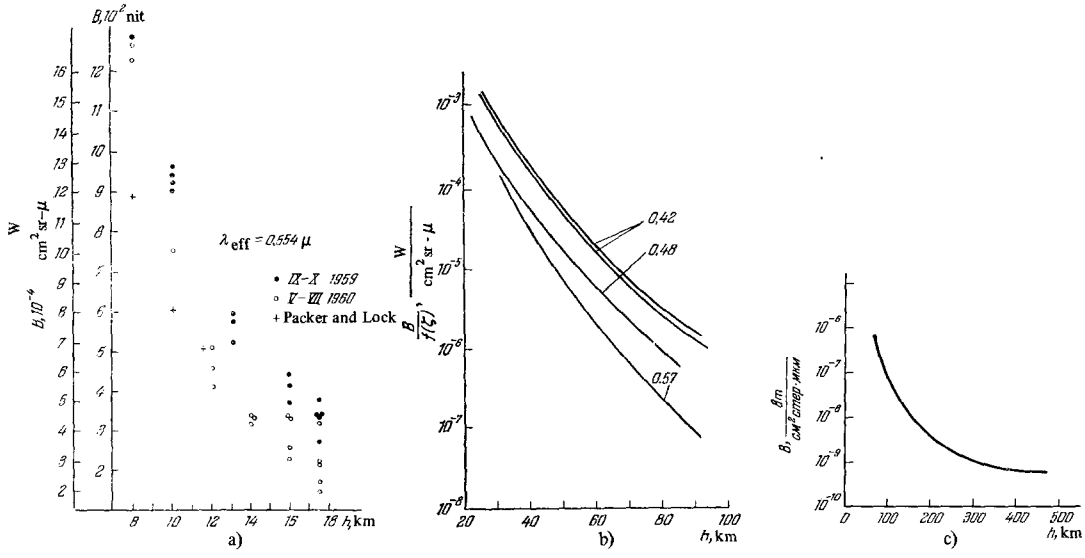

FIG. 29. Altitude dependence of the brightness of the sky as given by different authors. a) Airplane measurements [183]; b) twilight measurements (to obtain the brightness of the sky at the zenith, the presented quantities must be multipled by the scattering indicatrix $f_{11}(\varphi)$ corresponding to the zenith distance of the sun) [191]; c) rocket measurements ($\lambda = 0.53\ \mu$) [140].

the experimental verification of new optical methods of research on the structure and state of the atmosphere[197].

Recently V. G. Fesenkov[204-207] advanced interesting ideas concerning the improvement of the twilight method of sounding the atmosphere, but these unfortunately were not yet used to solve concrete problems by organizing specialized observations. We mention also investigations carried out in this direction by G. Dietze[208] in which the analysis of the stratospheric aerosol is performed by the polarization of the light scattered by the aerosol.

Also included among the twilight methods of investigation are photometric and polarimetric observations of silver clouds[183-185], for which the altitude accuracy is sufficiently high, and the scattering coefficient is determined with an error not larger than 150% (see also[111,112]).

We shall not stop to discuss investigations in which attempts are made to obtain information on the aerosol structure of the atmosphere on the basis of patently insufficient information, or with the aid of untested procedures (a bibliography of the recent investigations can be found, e.g., in[32-34]).

The development of the ideas on which the theory of twilight phenomena are based[9] has also made it possible to determine approximately the main laws governing the altitude, azimuthal, and spectral variations of the brightness of the light aureole as seen surrounding our planet when it is observed from the outside under daylight and twilight conditions, as a function of the structure of the atmosphere and the state of the underlying surface[108-114]. These conclusions were confirmed experimentally by observations from the spaceships "Vostok-6"[109-113] and "Voskhod"[114,198], and also by computer calculations for certain models of the atmosphere[118,199-201]. One of the experimentally observed cases of the brightness profile of the daytime horizon of the earth is shown in Fig. 31 (the point of inflection corresponds to the optical edge of the planet as observed from the outside).

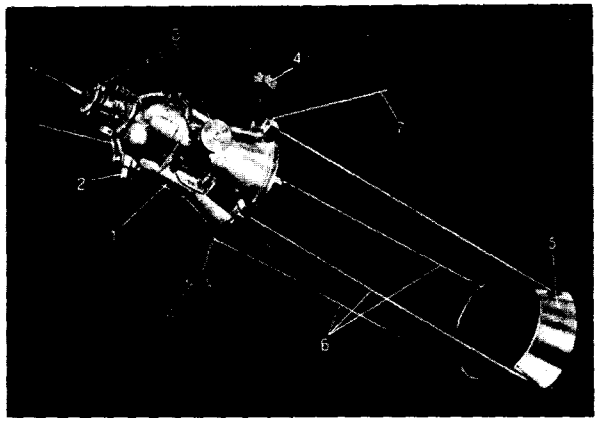

FIG. 30. Orbital comprehensive optical station "Kosmos-149" [197]. 1 – Hermetic housing of the satellite; 2 – illuminator of television apparatus; 3 – scanning telephotometers with interference filters (a narrow-angle narrow-band radiometer is mounted on the invisible part of the satellite); 4 – blocks with pickups for the energy parameters of the radiation of the pilot; 5 – aerodynamic stabilizer; 6 – stabilizer rods; 7 – antennas.

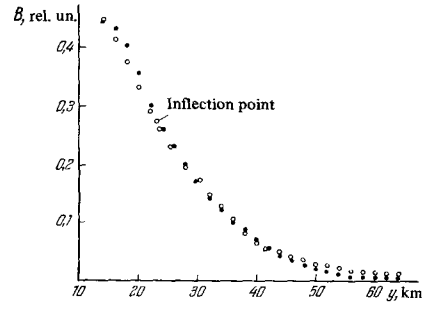

FIG. 31. Brightness profile of the daytime horizon of the earth as observed from outer space [114].

Measurements of the brightness structure of the lower part of the sundown aureole from a spaceship[109,113] makes it possible to determine, on the basis of a specially developed theory[9,108,111,112], the value of σ for the altitude intervals from 8 to 25 km, with a relative error of approximately 25% (cf. Fig. 13), an absolute error of approximately 150%, and an altitude error of approximately 1 km (with averaging along a route of approximately 500–1000 km). Model calculations of the solution of the direct problem in the style of[118,199-201], have, generally speaking, nothing in common with the problem of its inversion, considered in[9,108,111,112], although they are, of course, very important from the point of view of verifying methods of its solution (see[118]). The upper part of the brightness structure of the aureole contains information on the altitude variation of σ up to altitudes on the order of 150–200 km[108,109,112], but the appropriate program has so far not been realized.

For the altitude interval from 12 to 55 km, data on the altitude variation of the scattering coefficient were obtained (also on the basis of a specially developed theory[9,110,114]) by measuring from a spaceship the brightness structure of the light aureole over the daytime edge of our planet[114,198]. The absolute error in the determination of σ was in this case close to the factor 1.5, and the altitude error was approximately ± 3–4 km.

The value of σ can also be determined by measuring the altitude variation of the brightness of the scattered light of a projector (observed from the side). Although the brightness of the projector beam can be measured up to altitudes of approximately 60 km[8], the determination of σ with satisfactory accuracy (approximately 150–200% when the altitude is accurately known) is possible only up to altitudes 45–50 km[8,16,117,158-160,209,210]. This is possible, however, only on the basis of a specially developed theory[8]. In particular, the method of information extraction used by L. Elterman[209] is not correct enough, thus explaining the discrepancies between his latest data[209] and the results of his earlier work[16,17,210].

A direct development of projector sounding is the sounding of the atmosphere with the aid of lidars, i.e., radars with lasers[211-225], which has been realized already up to altitudes of 160 km (to be sure, with reliability and altitude resolution that decrease greatly at large altitudes). The main difference from projector sounding lies in the fact that this method does not make it possible to vary the scattering angle, the measured quantity always being the radar cross section, i.e., the product $\sigma(h)f_{11}(\varphi = 180^\circ, h)$. Owing to the uncertainty and variability of the scattering function $f_{11}(\varphi = 180^\circ)$ as a function of the microstructure of the aerosol and especially as a result of the fact that this quantity can be much smaller for the disperse phase than for molecular scattering, the error in the determination of σ can reach several hundred per cent (this was specially pointed out by D. Deirmendjian[226]). It is possible that this danger is slightly exaggerated, for a relatively high correlation between $f_{11}(\varphi = 180^\circ)$ and σ is observed in the surface layer[227,228].

The main features of the altitude variation of the scattering coefficient in the entire interval of altitudes

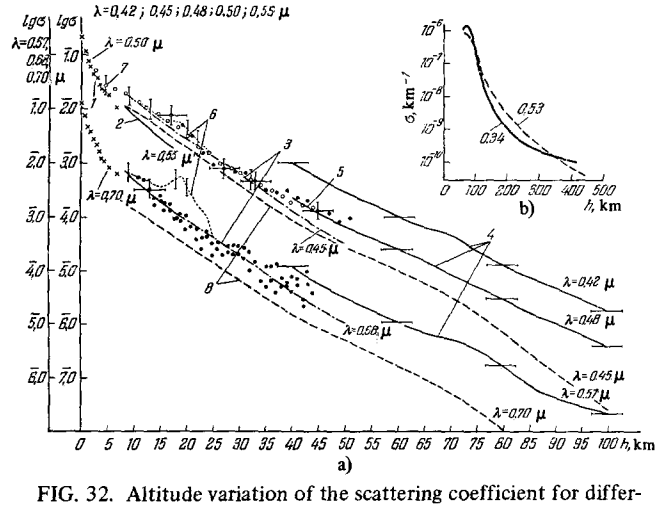

FIG. 32. Altitude variation of the scattering coefficient for different sections of the spectrum: a) up to 100 km, from the following data: airplane measurements of the brightness of the sun [186,203] (1), airplane measurements of the brightness of the sky [188] (2), their extrapolations [117] (3), land measurements of the brightness of the twilight sky [107] (4), measurements of the brightness of the light haze at the daylight edge of the planet from the spaceship "Voskhod" [114] (5), measurements of the dawn from the spaceship "Vostok-6" [109] (6), projector sounding from the earth's surface [8,117] (7), and calculation for the molecular atmosphere (8); b) from 100 to 450 km as obtained by rocket measurements [140].

up to 100 km, as traced out in accordance with the data of optical measurements performed with participation by the author or his co-workers, essentially by means of procedures developed by them, can be seen in Fig. 32a. Fig. 32b shows the data of[140], where the variation of σ(h) is extended to 450 km.

We note first that all the data of Fig. 32a agree with one another, thus demonstrating the effectiveness of all the employed methods, and the reliability of the results. The discrepancies never exceed the expected errors, thus indicating the agreement between the employed methods of coarsening of the solution by the properties of the investigated object (since the resolution of the method is determined by a smoothing algorithm introduced into the solution and playing the role of the apparatus function).

The most significant of the results is the undisputed and already mentioned fact that the entire atmosphere is highly turbid. The scattering coefficient of the disperse phase exceeds that of the molecular phase at any rate by a factor of several times, and this ratio changes relatively little at least up to an altitude of 90–100 km (see also[192]). This is confirmed also by data on the scattering indicatrix, which changes little with altitude[107,189,195].

The only exceptions are two rather narrow altitude intervals where the air is hardly turbid and the ratio σ/σ_M is close to unity (σ_M–the scattering coefficient of light by the gas phase at the same altitude). These are the interval from approximately 5 to 8–9 km (see, e.g.,[1,8,9]), and the 25–30 km interval discovered by us[117], the existence of which has made it possible to improve greatly the reconciliation of the absolute values of σ obtained by projector or laser sounding (see, e.g.,[214]). The reason for the clearing of the air in this interval can be sought in the process of episodic washing

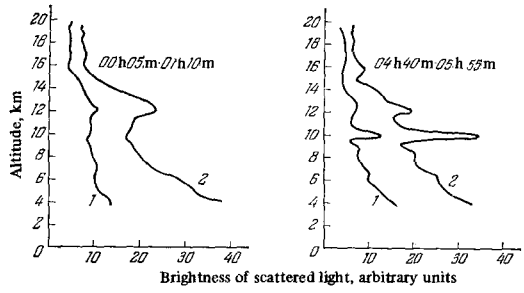

FIG. 33. Altitude variation of the brightness of the scattered light of a projector in the region of the tropopause in two successive soundings in the green (1) and blue (2) regions of the spectrum simultaneously.

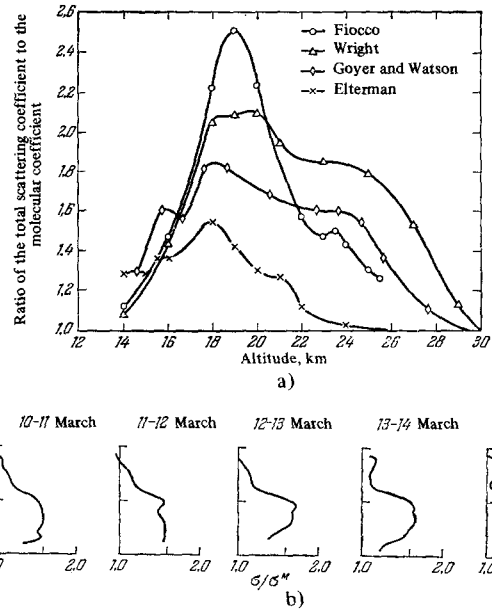

FIG. 34. Altitude variation of the ratio of the scattering coefficient of air to the coefficient of molecular scattering at the same altitude for the dawn layer: a) from data by different authors [221]; b) variation from day to day [214].

out of the disperse phase as a result of formation of mother-of-pearl clouds[157-160].

Another most important feature of the stratification of the atmospheric aerosol is the existence (sometimes for a rather long time) of relatively thin layers, with large horizontal dimensions, in which the concentration of the condensation nuclei is large or else the turbidity of the air is large. These layers have a distinct tendency to occur in the vicinity of temperature inversions. They are therefore usually connected with the phenomena of accumulation of the disperse phase as a result of changes in the dynamic regime of its transport (see, e.g.[8,37,155,179]). Such a process occurs, for example, in the region of the tropopause (9–13 km), where it is clearly observed in the case of projector sounding (Fig. 33).

Equally distinct, from the point of view of optics[1,8,9,186,230] is a surface layer with very high turbidity (approximately up to 500–1000 m), which goes over into a layer of medium turbidity, the upper limit of which coincides usually with the upper limit of the convective layer (3–5 km) (cf. Fig. 28b). This pertains, of course, only to the averaged picture: the individual variations of $\sigma(h)$ are much more complicated and reflect the concrete features of the individual state of the weather (see[8,9]).

A patently unexpected fact, which calls for a review of a number of concepts, was the discovery of a layer of increased turbidity in the altitude region 15–22 km (cf. Figs. 13 and 28). Since this layer is responsible for the coloring of the dawn, we propose to call it the "dawn" layer[157]. Although twilight phenomena frequently indicated its existence[9,157,202], it was first revealed reliably only with the aid of airplane-borne traps by H. Junge and his co-workers, and then optically by the author with V. V. Nikolaeva-Tereshkova[109]. It was soon after subjected to a detailed study with the aid of projector[209] and laser[214,221] sounding. Figure 34 shows examples of the altitude structure of this layer from data of different authors (a) and of the time variation of this structure (b). The latter could not be revealed with the aid of traps, owing to the low effectiveness of this method (the gathering of one sample requires that the airplane travel over a route of approximately 1000 km[37]).

Aerosol layers were observed also in different times at altitudes 30–37 km[8], 43–45 km[8], 50–60 km[192], approximately 70 km[213], approximately 90 km[140,158] (including indirect determination based on the behavior of the glow of atomic sodium[231]), approximately 120 km[215-219], and also at different altitudes following volcanic eruptions and in the vicinity of space-ship launching bases (see[157]).

Although on the whole the optically revealed stratification duplicates the stratification observed by other methods, certain differences are clearly pronounced. First of all, in optical stratification there is a clearly observed altitude variation of the relative humidity[179,220], since it is this variation which regulates the course of the condensation processes. It must be assumed that in many cases the occurrence of layers with increased turbidity (especially in the stratosphere) is connected not with the increase of the particle concentration, but has a patently dynamic character, as was proposed with respect to silver clouds by B. V. Mirtov[230]. Indeed, the particles whose trajectories pass through the region of increased humidity should increase in size in this region, and should again decrease on leaving the region, leading in turn to an increase of the turbidity within the entire region of increased humidity.

It must be assumed that a similar mechanism is responsible for the formation of mother-of-pearl clouds[8,157-160].

The foregoing gives grounds for stating that the optical methods of investigating the atmospheric aerosol may turn out to be a powerful means of studying the dynamics of atmospheric masses, especially at large altitudes. Some attempts in this direction were already made as applied to silver clouds[233]. The promise offered by such searches is evidenced also by the results of determining the spatial spectra of the brightness fields of cloudiness of different types[90,161,162,232], which patently demonstrate the possibility of their use for the identification of various dynamic (turbulent) regimes.

In conclusion we note that the past decade was principally the time of development of new methods and

means of optical investigations of the atmosphere, and also of verifying these methods, for the most part in clearly preliminary fashion. These attempts not only demonstrated the power of the optical methods, but also introduced noticeable changes in our opinions concerning the properties and role of the disperse phase of the atmospheric aerosol, its origin, and its altitude distribution. We are now entering an era in which these methods can turn into a tool for the study of the geophysical processes and for the observation of those physical phenomena on which these geophysical processes are based, primarily, of course, processes of condensation transformation of the aerosol. Consequently the development of specialized apparatus for optical sounding of the atmosphere becomes particularly timely, especially the creation of a wide assortment of highly effective procedures and the development of a network of permanently operating optical-sounding stations.

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