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SUMMARY OF THE CONFERENCE ON THE OPTICAL PROPERTIES OF AEROSOLS

15-17 October 1964
Onchiota Conference Center, Sterling Forest,
Tuxedo, New York

Prepared by

E. A. Chermack
(Editor)

July 1965

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Acknowledgments

The editor would like to thank the survey speakers for their clear comprehensive presentations; the chairmen for their hard work in moderating and channeling the discussions and for summarizing their respective sessions; and the United States Army Electronics Command, Atmospheric Sciences Laboratory, for its sponsorship of this conference.

Introduction

A conference on atmospheric aerosol optics was held in October 1964 at the Onchiota Conference Center, Sterling Forest, Tuxedo, New York. This conference was sponsored jointly by New York University and the U.S. Army Electronics Laboratory. The purpose was to ascertain the state of knowledge and to determine the problem areas in this field of atmospheric physics.

The conference was held over a period of three days, each day being devoted to a problem area as introduced and outlined by a survey speaker. Following the survey talk, morning and evening discussion sessions were convened. Each session was presided over by a chairman who also acted as moderator of the informal discussion. Prior to the conference, the invited participants were asked to submit brief resumes of their current research interests in the problem areas to be discussed and these were forwarded to the chairmen.

The sessions were kept as informal as possible in order to facilitate the free flow of information, opinion, and ideas. The chairmen maintained and directed the thread of the discussion by calling upon participants to present their views as the discussion turned into their spheres of interest.

The number of participants was limited to about fifty meteorologists, atmospheric physicists, chemists, and engineers working actively in the field. This was necessary because the sponsors wished to insure informality, freedom of expression, and of course, to avoid the problem of overlapping on concurrent sessions.

This report is a summary of the conference discussion. Each day's discussion summary is preceded by the survey speaker's remarks. Discussion summaries were prepared by the respective chairmen and represent the salient features of each day's meetings.

AGENDA

The subject of the conference was optical phenomena (affecting the wavelength region uv-50 microns) associated with the presence of naturally occurring aerosol particles of terrestrial origin including those effluents contributed by civilian activities.

First day: Physical and chemical properties of aerosol particles.

Survey Speaker: C. Junge

Chairmen: H. Weickmann (morning)
J. E. McDonald (evening)

Second day: Optical properties of aerosols.

Survey Speaker: H. C. van de Hulst

Chairmen: R. Penndorf (morning)
D. Deirmendjian (evening)

Third day: Influence of atmospheric aerosols on radiation (radiation transfer).

Survey Speaker: F. Möller

Chairmen: Z. Sekera (morning)
J. Dave (afternoon)

FIRST DAY:

PHYSICAL AND CHEMICAL PROPERTIES OF ATMOSPHERIC AEROSOLS

C. E. Junge
Meteorologisch-Geophysikalisches Institut der
Johannes Gutenberg-Universität
Mainz, Germany

I. Introduction

Atmospheric aerosols are not only interesting by themselves but they determine also to some extent the electrical, chemical and optical properties of our atmosphere. This meeting was arranged to discuss the optical properties of atmospheric aerosols and it is therefore necessary to start with a survey about our present knowledge in this field serving as a basis for the subsequent discussions. The following properties of atmospheric aerosol particles will be discussed:

1. Structure
2. Chemical composition
3. Size distribution
4. Large scale distribution.

The optical properties of the atmosphere depend in a complex way on the various parameters of the aerosols. Once we understand this relation optical properties can tell us something about the characteristics of the aerosols. But it is difficult to establish the properties of the aerosol through optical effects. This can only be done by simultaneous and independent investigations of the properties of the aerosols themselves, a fact which was long neglected in the field of atmospheric optics and radiation and which is still today not recognized in all quarters. It is our hope that this meeting will help to eliminate this difficulty.

II. Structure of aerosol particles

A. Basic concept

The most general concept about the structure and composition of atmospheric aerosol particles is that of the mixed particles. A mixed particle consists of a mixture of different substances, soluble and insoluble, inorganic and organic, etc.

It is easy to imagine the large variety of possibilities with the extremes of sea salt particles which consist of an almost pure soluble salt on the one hand, and quartz, i. e., completely insoluble particles on the other hand.

The two parameters which control the optical properties of an aerosol particle, the refractive index and the absorption are strongly dependent on the quality and distribution of the transparent fraction (water soluble as well as insoluble) and the opaque fraction. In case the transparent fraction consists of a solution of inorganic salts in water the optical properties of the particle will also depend on the relative humidity of the air.

B. Evidence for mixed particles in the atmosphere

What evidence is available to support the concept of mixed particles? First, there is rather direct evidence from electromicroscope pictures, which demonstrate the great variability of particle structure. Very common is an opaque central part surrounded by a translucent halo which is composed of water soluble or organic materials.

The growth of particle size with relative humidity is further evidence. The growth for pure hygroscopic material can be calculated and is well known. These growth curves do not differ much for various substances. The observed growth curves for continental aérosols lie between those for pure hygroscopic materials and no growth with humidity for completely unsoluble particles. Measurements from Central Europe seem to indicate that this concept of mixed particles is valid for all particle sizes. Further evidence is also the complex chemical composition of aerosols, which will be discussed later.

C. Formation of mixed particles

The following processes result in the formation of mixed particles:

1. Condensation of heterogeneous smokes in a variety of industrial and other combustion processes (forest fires, e. g.).
2. Coagulation of aerosols of original homogeneous composition in the

atmosphere subsequent to their formation. This coagulation occurs in cloud free air continuously by Brownian motion, but also in clouds, where cloud droplets collect particles but also gather gaseous matter, which remain attached to the particles after evaporation of the cloud droplets. Since condensation and evaporation on condensation nuclei occur many times before their removal by rain, this agglomeration process is very efficient.

3. Finally, reaction products of trace gases in the atmosphere become attached to existing aerosol particles and modify their composition. The omnipresence of sulfate in aerosols is at least to some degree due to the attachment of photooxidized SO_2. Dry mineral dust particles thus obtain a soluble coating and sea salt particles increase their sulfate content.

The majority of processes which result in mixed particles occur over continents.

III. Chemical composition

A. General composition

Our information on this subject is still rather scanty. For a broad survey we may subdivide the substances into three groups:

a) Inorganic, water insoluble
b) Organic, water insoluble, aceton soluble
c) Inorganic, water soluble

For unpolluted atmospheres we have very little information on a) and b), but fairly good information on c). For large cities and populated areas we have some information on a) and b). The total concentrations of atmospheric aerosol material have the following range:

large cities	small cities, densely populated areas	thinly populated continental areas
100-200 $\mu g/m^3$	50-100 $\mu g/m^3$	<50 $\mu g/m^3$

The few data available indicate a composition of polluted and unpolluted aerosols as follows:

component a:	60%
b:	15%
c:	25%

But these figures can give only a general idea and there are certainly large fluctuations. The mass fraction of water can, of course, vary considerably depending on the amount and properties of soluble substance and on the humidity of the air. For Central Europe we can assume the following figures:

Relative humidity	Approximate water content
60%	5%
70%	12%
80%	25%
90%	70%

These are very rough average figures and correspond to approximately 25% soluble material. A value of about 25% soluble material was deduced from growth curves of aerosol particles with humidity in Central Europe.

B. Special compounds

The figures 60% and 15% for a) and b) are primarily based on the analytical data from the Public Health Service network of filter stations, which unfortunately included SO_4 as the only soluble compound. The relative proportion of a) and b) does not seem to vary much with the degree of pollution and it is perhaps permissible to extrapolate to unpolluted atmospheres. Goetz has recently demonstrated that the size range below 0.5μ radius in polluted and clean air contains considerable amounts of material which slowly evaporates; these are most likely organic compounds of low vapor pressure or which are decomposing slowly by reaction. It is possible that this organic material is partly formed by photooxidation of small traces of organic vapors which are produced in large quantities by certain plants and are constantly injected into the atmosphere.

One may ask why rain water analyses do not provide more and reliable information on the chemical composition of aerosols. The answer is that the concentration of rain water is not representative of the aerosol composition:

a) because only certain aerosol sizes, primarily with $r > 0.1\mu$ are removed by rainout in the clouds and with $r > 1\mu$ by washout below the clouds;

b) because the soluble compounds like $SO_4^=$, Cl^-, NO_3^-, NH_4^+ can also be formed in the cloud and raindrops after condensation on aerosols took place due to gas trace reactions. The extent of this influence is not yet certain.

c) because quantitative analysis for insoluble, i.e., also organic matter, is rather difficult due to early separation and sedimentation in the rain water samples and due to the sensitivity of analytical methods for dry material.

Whereas the fraction of insoluble and organic matter and its chemical composition in natural aerosols is still a rather open question we have more detailed information on the soluble compounds, especially on $SO_4^=$, Cl^-, NO_3^-, NO_2^-, N_a^-, NH_4^+, K^+, Ca^{++}.

$SO_4^=$ is a rather regular constituent of aerosols, due to photo-oxidation of SO_2 which is always present in the troposphere. It is also very likely that NH^+, NO_3^- and NO_2^- and perhaps to some degree Cl^- are formed from corresponding gas traces. Most Na^+ and Cl^- comes, of course, from sea salt.

Analyses of aerosols in various geographical locations indicate a general decrease in the concentration of

$$SO_4^= , NH^+ , Ca^{++}$$

and a general increase in

$$Cl^- , Na^+$$

if one proceeds from continental maritime places, indicating the origin of these compounds.

IV. Size distribution

One of the most important properties for the optical behavior of aerosols is their size distribution and we will discuss this a little more in detail.

A. Survey about the observations

It is important to recognize that the distribution of natural aerosols comprises a large size range from about 4×10^{-7} cm radius to about 10^{-2} cm, i. e., of more than four orders of magnitude. Up to the present, very little information is available for the complete size range, but it is nevertheless important to consider the whole size spectrum as a unit. Unfortunately, all methods used to measure aerosols can only be applied to rather narrow size ranges, so that most investigations deal with only limited parts of the complete spectrum.

In the early results the data below 0.1μ were obtained by measuring the mobility spectrum of large ions which are identical to the charged particles. Particles larger than 0.3μ radius were collected by impactors and the size spectrum was obtained with the microscope. If the curves for $r < 0.1\mu$ are not smoothed, they show at least in Central Europe often pronounced "line" structure. Most recent data by Misaki in New Mexico show, however, rather uniform distributions. The main features of these distributions are an apparent lower limit, a maximum in number concentration mostly around 0.03μ and a long uniform slope from 0.1 to 100μ which seem to follow a power law

$$\frac{dn}{d\log r} = c \cdot r^{-\nu^*} \quad \text{with } \nu^* \text{ mostly between 2.5 and 4.0.}$$

For the limited size ranges of $r > 0.3\mu$ more data are now available. These data were obtained in the U. S. by single particle light scattering methods; by and large they agree with a power law.

Another set of data was recently obtained by Goetz and Fenn with the aerosol spectrometer. They found for various parts of the U. S. considerable structure for the range $r > 0.1\mu$ and often no power law.

One of the most striking features is a considerable drop in concentration for the lower end of the size distribution covered by the aerosol spectrometer, i.e., around 0.1μ. It is very difficult to see where in such a distribution the considerable number of Aitken particles would fit. To satisfy these conflicting observations one has to assume a fairly regular minimum around 0.1μ radius, a feature which is difficult to explain. Some of the more pronounced characteristics of the distributions for $r > 0.1\mu$ could also be identified by Fenn in the scattering function, although it must be pointed out that by and large the scattering function and other optical properties are not very sensitive to details of the size distribution.

It appears from these data that the overall distribution often follows power laws with exponents ν^* between 2.5 and 4, but that particularly in individual cases, there are considerable deviations. This is in fair agreement with optical properties, as was shown by Bullrich, Volz, and others primarily in Central Europe. We may recall here that the well known law of Ångström, according to which the total scattering of haze which is equivalent to the extinction has a pronounced wavelength dependence with an exponent of about -1. It can be shown that this fact is a direct result of a power distribution with an exponent ν^* of about 3. The considerable structure in aerosol size distributions found by Fenn and Goetz in the U.S. is of great interest and needs further investigation.

All these data refer to observations over continents. For pure representative maritime air, we have unfortunately data only above 1μ radius; in a few cases down to a few tenths of a micron. If we assume that the particles smaller than 0.1μ radius represent aged aerosols from continents the total size distribution of aerosols in maritime air masses can be estimated.

Whereas the lower size limit seems to be fairly well established between 10^{-7} and 10^{-6} cm, the upper limit over land is still an open question. There is rather good evidence that over the ocean the sea salt particles show an upper limit around 20μ radius, but new data from Central Europe have not produced an upper limit. If methods are used which enable the collection of large air samples so that even the concentration of the very few large particles can be established, the size distribution with the power law of $\nu^* \approx 3$ continues beyond 20μ up to radii of more than 100μ. This extension of the upper end of the size spectrum seems to be a very regular phenomenon. This result poses an interesting problem to the turbulent exchange near the ground and the mode of transport of these particles upward from the ground.

The last feature of the size distribution which we will mention is the location of the maximum. A considerable number of mobility measurements of large ions indicate an average particle size around 3×10^{-6} cm radius. For a size distribution like ours, the average size is approximately identical with the size of the maximum concentration. Because of the large variations of Aitken nuclei, the question arises as to whether there is any relationship between the total concentration and the average size. The approximate average size and thus the size of the maximum concentration can be obtained from two sets of data.

a) From simultaneous counts of the small ions n and the total number N of aerosol particles. If the production rate of small ions is known (it is rather constant) the ratio n/N depends on the average size of the aerosol particles, because the attachment coefficient is a function of the particle size. A large number of data on n and N is available in the literature which was evaluated.

b) From the ratio P of N to the concentration of charged particles. P is a function of particle size under ionization equilibrium conditions. Measurements of P thus give information on the average particle size.

All these data show a decrease of N with increase of maximum or average radius for a large variety of different geographical locations. This indicates a rather general relation and can be interpreted as the growth of aerosol particles as the aerosol ages. Aging of aerosol as a result of agglomeration must result in a decrease of N and a growth of the average particle size.

B. Theoretical considerations

One of the most important processes to modify the size distribution is the coagulation by Brownian motion. In cloud free air this is a very effective process by reducing the number of the smaller particles and increasing the average size. Smolukowski derived an expression for sols which was later also applied quite successfully to aerosols showing good agreement with measurements. Recently it was pointed out by Lassen and others that for particles small compared with the mean free path length of the smaller coagulating particles, this theory is not correct. For most of the size range of interest in atmospheric aerosols this error is not very large and we applied therefore the simpler theory for our model calculations.

In these calculations, several initial size distributions were chosen to investigate the variation with time of these size distributions if subjected to coagulation. The main result can be summarized as follows:

For concentrations encountered in the atmosphere the modification by coagulation is negligible for radii larger than 0.1-0.2μ but is very considerable for decreasing sizes below this value. Any line structure in the distribution for this latter size range is smoothed out by the coagulation process as time proceeds and the number decreases very rapidly with decreasing radius, so that a lower limit for particle concentration must be expected in the atmosphere.

Similar calculations were made for the case when clouds are present in the atmosphere. In clouds a variety of processes occurs which result in a modification of the size distribution. The most important process is the attachment of the small "unactivated" aerosol particles to the cloud droplets, which after evaporation of the cloud, results in a growth of the "activated" aerosol particles. This process becomes rather effective because of the fact that on the average, condensation nuclei go through many cycles of condensation and evaporation before being removed by rain.

The main results of all these processes in cloud filled air on the size distribution are similar to those for Brownian coagulation. These results, therefore, must be considered generally valid for the whole atmosphere.

These calculations force to the conclusion that the features of the size distribution of atmospheric aerosols for radii larger than 0.2μ are not much influenced by processes within the atmosphere but must be due to the processes that produce the aerosols. This applies particularly to the power law or the structures with "holes" found by Fenn. But so far, no explanations for these features have been offered.

IV. Large scale atmospheric distributions

A. Aitken particles

The main features of the large scale vertical distribution of Aitken particles can be demonstrated by the average profile of the U. S. balloon program together with earlier data from aircraft and manned balloons. We can distinguish three main layers:

1. Rapid decrease of concentration in the lowest 3-5 kilometers, due to rainout, washout, coagulation and influx aloft of certain maritime air over continents.
2. More or less constant concentration of around $500/cm^3$ STP in the upper half of the troposphere. We think that these particles represent aged continental aerosols and form a wide-spread background throughout the troposphere (background aerosol). Over the oceans we may find this concentration throughout the whole troposphere, since numerous surface measurements were found to be in the same concentration range.
3. In the stratosphere there is a pronounced decrease of Aitken particles with altitude. Most likely this decrease represents a steady state, for which the upward flux is balanced against decrease in concentration due to coagulation and sedimentation. Since the size distribution is not precisely known it is not possible to obtain reliable estimates on these processes.

B. Particles in the range 0.1 - 1.0μ radius

Within the troposphere the vertical distribution of these particles is essentially similar to that of the Aitken particles. Unfortunately, we have practically no direct data above 5 km, but the indirect information available indicates again a rather constant concentration. As in the case of the Aitken particles, this shows that the processes of removal are slow compared to vertical mixing. In the stratosphere we have a pronounced layer of higher concentration which is now confirmed by various authors and by different methods. The maximum concentration is around 20 km, but there seems to be considerable variation with time and latitude. Although it is at the present time somewhat difficult to compare the various methods because of the lack of accuracy, the peak concentration may differ by a factor of 5 and the altitude by 3 or more kilometers. But since fall 1963 the stratosphere of the northern hemisphere is seriously contaminated by the eruption of the Agung volcano and this increases the difficulty of comparison. By this eruption the northern hemisphere was not contaminated until summer 1963, but from September 1963 on, the volcanic dust also entered the northern stratosphere increasing the concentration by a factor of about 5.

It is now well established that the major constituent of these particles is sulfur, which excludes extraterrestrial origin of most of these particles. So far no definite explanation of the formation of this layer has been advanced.

Thursday morning session - H. K. Weickmann, Chairman

Following Dr. Junge's review, Dr. Fenn was asked to present the salient points of his experimental findings concerning the gaps in the aerosol distributions which were taken in different air masses and locations, and which all indicated a separation into two groups: one group peaks at around 0.2 microns particle diameter, then there is a sharp minimum at 0.4 to 0.5μ, and then there are either one or more groups towards the larger end. Each measurement requires 15 to 30 minutes sampling time; the distributions are consequently averages in time and space. Whether the distribution depends on sampling time is unknown.

The discussion then centered around the sampling technique: level above surface, inside shelter or outside, time of day, etc. Most measurements were made in daytime except for a few which were made at sunset in order to be able to follow up with light scattering methods using a nephelometer in the darkness.

Dr. Volz asked for the reproducibility of two measurements which were made shortly after each other. A few measurements were made one or two hours apart and the reproducibility was very good.

Dr. Hodkinson asked the very pertinent question whether the troughs have also been observed by different methods. Dr. Fenn confirmed this from his parallel measurements of the scattering function. Special minima in the scattering function can be shown as caused by the troughs in the spectrum.

Dr. Goetz confirmed Dr Fenn's findings through his own hundreds of tests.

Errors entering through the analysis process were discussed. The analysis involves differentiating of the raw curve and scatter of the points would cause minima and maxima of the final curve. Dr. Fenn points out that the original plots are very steady curves with little scattering of the individual points. Dr. Arking noticed the steep slope of the distribution curves which is apparently steeper than R $^{-2}$ in the contaminated atmosphere (New Jersey air).

Dr. Twomey then points out that he has found in his investigations of aerosol spectra using diffusion batteries, indications of an active coagulation but also the existence of permanent holes which is a contradiction since holes should disappear by coagulations. Dr. Rooth states that this may be caused by young aerosol coming from a multiple source.

Dr. Fiocco on aerosols in the stratosphere. The author mentions that the new laser techniques offer a way of finding whatever there is in the atmosphere from essentially ground level up to many kilometers. The 20 kilometer dust layer can be studied at a frequency of one cycle; this permits the study of this layer's movements and changes. Dr. Fiocco mentions that in his published data of that layer the ratio of aerosol to molecular scattering was 3 : 1 .

Dr. Bigg then showed U-2 photographs from 20 kilometer height which were taken over Australia before and after the eruption of Mt. Agung, Bali, Indonesia, 8°S 115°E which was on 17 March 1963 in 6°S. Two months after the eruption at latitude 60°S there was not yet a great deal of intervening dust; the sky above is gray while the layer at 20 km shows intensive stratification. These strata stay for distances of several hundred kilometers to 26°S latitude where there is much dust. This was over inland Australia. Here the sky became black and stayed so until latitude 36°. Between 16° and 20°S there was the most intense dust cloud. After a week, photographs were obtained at 40°S which showed extremely "murky" conditions both above and below the aircraft.

Particle sizes of 10 microns diameter were found initially; these lasted up to 100 days, but the maximum size decreased fairly quickly, and one year after eruption the median size of the particles was around one micron to one half in diameter.

Dr. Volz discussed his twilight measurements in connection with Mt. Agung eruption. He showed a graph of the red-green ratios of the twilight measured in Germany. The first high red-green ratios in Germany occurred in July 1963; in his recollection it took almost two months for the dust to show up in the northern hemisphere. Optimum effects occurred around the end of the year and presently (October 1964) a new increase seems to occur. The stratospheric turbidity over the southern hemisphere is much higher than over the northern, and air mass exchange over the equator can still bring new dust over the northern hemisphere. The stratospheric turbidity in the southern hemisphere last summer and winter was comparable to the turbidity in the mornings in downtown New York. Professor McDonald noticed on Dr. Volz's graph of the red-green ratios a recent increase which he had also observed in Arizona due to the twilight coloration. He was considering whether this could have come from the Iceland eruption of November 1963. Dr. Volz's information on the discoloration of the twilight due to the Iceland eruption was that it did not extend into the purple range but he calls attention to other eruptions such as that of Mt. Irazu, Costa Rica (10°N, 84°W), this latter consisting essentially of granular dust which falls nearby. But since the southern hemisphere is still filled up with a large amount of dust, hemispheric air mass exchange may inject periodically dust into the northern hemisphere. Dr. Friend discusses the mechanism through which aerosol particles are carried to altitudes of 20 kilometers. Normally it takes about a megaton nuclear blast to get particles to that altitude. From observations of the eruptions of Kilauea, Hawaii on 14 November 1959, 29 November 1959, and 13 January 1960, the main energy in this case, contrary to Krakatoa, does not appear to come from the eruption, but from the heat (about 2000 degrees in the case of Kilauea); and the transport mechanism into the stratosphere would be convection. This may account for a composition of the material different from that of the Krakatoa eruption. Dr. Friend assumes that the main injection into the stratosphere consisted of carbon dioxide and other gases which subsequently formed aerosol and Aitken nuclei. Bigg, however, states that the particles which he found at latitude 30°S contained large numbers of insoluble angular particles which were certainly not gaseous in origin. (He is referring to Mt. Agung eruption.)

Dr. Martell on Stratospheric Aerosol

Dr. Martell discussed some of the possible mechanisms for the formation of the stratospheric large particle sulfate layer. He pointed out that the very finely divided radioactive aerosol from higher levels became associated with natural aerosol in the lower stratosphere on a number basis. This steady state size distribution for the radioactive aerosols indicates inappreciable formation of sulfate by photochemical oxidation of H_2S or SO_2 within the stratosphere and leads to the conclusion that the stratospheric sulfate aerosol has mixed up from the troposphere.

If this conclusion is correct he agreed that some other explanation must be found to account for the vertical distribution of the large particle sulfate aerosol observed by Junge. He then discussed the vertical flux of large particles on the basis of their observed vertical distribution and their estimated atmospheric residence time versus altitude (measured in days at surface levels, weeks near the tropopause, and about one year at 20 km). On this basis he indicated that the vertical flux of large particles decreases with altitude and that the steady state concentration at 20 km required only an estimated 10^{4} of the total large particle aerosol generated in continental surface air. He suggested the upward transport of tropospheric aerosols in convective systems and storms, or selective upward transport in other latitudes as possible mechanisms. Whatever the actual details of the mechanism he tentatively concluded that tropospheric sulfate aerosol must account for the stratospheric layer.

In an additional discussion Dr. Martell estimated the amount of debris injected into the stratosphere by high yield nuclear explosions on the surface of the ocean and on coral islands as $\sim 1.7 \times 10^{4}$ tons of sea salt and $\sim 10^{5}$ tons of coral, respectively, per megaton of total explosion energy released. The estimate for coral is based on the maximum specific activity of airborne debris following surface coral shots. Sea salt injection was estimated assuming equal energy amounts for vaporization of sea water and contained salts as for coral per megaton of nuclear energy release. He further proposed that the sodium and calcium from sea salt and coral were, in part, carried by the rising thermonuclear fireball and cloud to heights of 30 km and more. Some of the molecular sodium and calcium of such origin should be expected to mix to upper mesosphere levels and may contribute significantly as a source of the sodium and calcium emissions in twilight. Anomalously high levels of lithium have been observed in twilight emissions about two weeks after high altitude thermonuclear explosions. Atmospheric storage times of the order of 10 years in the upper stratosphere and lower mesosphere suggest the accumulation and persistence of trace constituents injected into these levels.

After this, the discussion centered around the validity of the steady state concept as a basis for a theory of the natural stratospheric aerosol layers. Information on the fine structure of the stratospheric dust layers appeared to be particularly suited to investigate this question.

Dr. Friend showed pre-Bali size analyses of stratosphere aerosol with peaks near 0.20μ to 0.23μ radius and 50 percent mean radius of 0.3μ. In the individual analyses both sizes and numbers showed considerable variability in the numbers, varying over two orders of magnitude. He showed twelve samples taken in the northern heimsphere near 30 to 32°N between 13 February 1962 and 13 August 1963. At 60,000 feet (20 kilometers) the highest numbers were 0.26 per cc and the lowest 0.03 to 0.04. In lower altitudes the concentrations were 0.02 per cc. There appeared to exist a post-Bali increase of the concentration, but Dr. Friend attributes this to natural variations as the counts decreased again in August. Dr. Bigg assumes strong stratification of the dust so that it can be missed from one flight to the next. While this possibility exists, no twilight anomalies were observed in Arizona before December 1963, but Dr. Volz recollects that in Germany normal twilight conditions ceased in July 1963. Dr. Friend then showed information on the composition of the aerosol, using electron diffraction methods. In previous work, the composition was found to be ammonium sulfate, but of the twelve samples, eight were ammonium sulfate and four ammonium persulfate at the peak. This is not a normal material in the troposphere at all. One sample showed clearly a mixture of both materials. It was assumed that the particles are liquid when collected and may consist at least partially of sulfuric acid. On the size distribution, Dr. Friend felt very strongly that it is peaked and not continuing. One should be careful in drawing conclusions on an assumed distribution function of stratospheric particles and with the assumption that the distribution is due to a steady state process.

Mr. Elterman discussed the fine structure of the stratospheric dust layer on the basis of his searchlight method. Considerable discussion arose here on the justification of his assumption that the results can be normalized at 35 kilometers to Rayleigh scattering. The outcome of the discussion justified the normalization, since, according to Fenn, at the surface 10^9 to 10^{10} Aitken particles per cc are necessary to match the intensity of molecular scattering while at 35 km about 10^7 nuclei would be required, a number which is certainly never present. According to Junge, the concentration is 1 cm^{-3}. The speaker then showed variations of particle content, based on the analysis from the light-scattering measurements.

Dr. Soberman then discussed data obtained through rocket sampling in the region of noctilucent clouds. Several shots were fired through clouds and on days without clouds. A typical impact pattern appeared in a ring-like structure, which was typical of particles larger than 0.2 micron. In laboratory experiments for the simulation of the impact patterns, the ring-like structures could be obtained only when particles were coated with ice: water coating would not deform the substrate likewise. Chemical analysis of the particles indicated existence of fairly high ratios of nickel to iron, and traces of silica with iron, or nickel iron in combination with silica.

Thursday evening session - J. E. McDonald, Chairman

Speakers: Newkirk, Bigg, Friedlander, Fiocco, Goetz, Kerker, Farone. Closing discussion on gaps in aerosol size-distributions.

1. Newkirk

Newkirk summarized his balloon-borne coronagraph scattering measurements made at altitudes up to 25 km. Sky radiance due to total scattering at angles ranging from 2 to 58 degrees from the sun were directly measured, and the aerosol contributions were then inferred by subtracting the theoretically computed Rayleigh radiances. Newkirk regards the methods as having limited accuracy outside the particle-radius range of 0.2 to 2μ. Evidence was found for a local scattering maximum near 20 km, presumably due to the sulfate layer. Large day-to-day variations in scattering intensities were observed, principally in the 10-15 km region.

Using these estimates, and assuming an eddy diffusivity of the order of 5×10^3 cm^2/sec, Newkirk attempted to deduce the contribution of meteoric particles. An annual meteoric influx of between 10^5 and 10^6 tons/yr (about equal to the total burden from all atomic tests) was indicated. However, only about one particle in 100 near the 20 km level is of meteoric origin in Newkirk's estimate, the rest being of stratospheric or terrestrial origin.

In the discussion, Newkirk explained that he assumed a refractive index of 1.55 for the meteoric particles, but felt that the uncertainties therein were small enough so that one could ignore them as compared with other errors. In reply to a question concerning how much error was introduced by subtracting the Rayleigh portion from the observed total to find the aerosol contribution, Newkirk pointed out that even at 30 km the total is two or three times the Rayleigh component, so the errors of differencing are probably not severe at the lower altitudes with which he was concerned.

2. Bigg

A series of photos taken from U-2 aircraft at about 20 km altitude over northern Australia was presented, with brief commentary, by Bigg. The series constituted a before-and-after view with respect to the Agung volcanic explosion of March 1963. The dark mass of evidently opaque material shown in the slides aroused discussion of the optics involved, but no agreement was reached. Bigg pointed out that, after the Agung eruption, particle contents in the upper troposphere became so large that windscreen erosion on trans-equatorial jet aircraft posed a serious operational problem.

3. Friedlander

A new technique for sampling aerosol particles of diameter below 0.1 micron was outlined in Friedlander's talk. When a disc, say of half-dollar size, is rotated about an axis normal to its face, centrifugal action induces an airflow in which the velocity component normal to the face of the disc is independent of radial distance from the axis. The principle had previously been developed by Russian physical chemists for studies of solution chemistry, and now appears to have promise for certain aerosol sampling problems. The flux dJ contributed by particles in the radius range dr near r has been found theoretically to be given by

$$dJ = 0.62\, D^{2/3}\, \nu^{-1/6}\, \omega^{1/2}\, n(r)dr$$

where D is the particle diffusion coefficient for size r, ν is the kinematic viscosity of air, ω is the constant angular velocity of the disc, and n(r) is the distribution function.

An electron microscope grid is attached to the sampling face of the disc, and the disc rotated at a sufficiently low ω to preclude boundary-layer turbulence. In consequence of this limitation, quite long sampling times may be required (e. g. , 10 hours at 2000 rpm in one run described). After collection, the deposit is shadowcast and the size distribution measured with the aid of an electron microscope, permitting detection of particles down to about 70 Ångströms. The upper limit is set by slowness of diffusion of particles larger than about 0.1μ radius. Results of several runs in laboratory air in Baltimore were shown; but principal emphasis was placed on the technique, itself, as a promising substitute for diffusion batteries and thermal precipitators in the difficult size range below 0.1μ. Some participants were confused as to whether this technique permitted determination of absolute concentrations rather than merely relative concentrations in given size-classes, and Friedlander emphasized that it yields absolute-concentration spectra.

4. Fiocco

Some of the advantages and some of the difficulties of laser techniques of sounding the upper-atmosphere aerosol were outlined by Fiocco. One virtue of the method is that it works well at night, when many other optical techniques fail. Evidence for the sulfate layer appeared on the laser-sounding profiles, and Fiocco stressed the appreciable variability of ratios of concentrations at 19 and 25 km. Problems of shuttering, and of avoiding spurious readings from multiple scattering were discussed. Coulson proposed a simple scheme for experimentally estimating the contribution of multiple scattering by orienting the receiver just off the beam axis.

5. Goetz

Principal emphasis was put on one main point by Goetz: He finds that one of the most frequently encountered properties of natural aerosol particles is their thermal metastability, indicative of the presence of organic constituents. Using the Goetz aerosol spectrometer, one may collect natural aerosols arrayed on the collecting substrate according to their size at instant of collection ("kinetic size"). If the substrate is then held at 50-100°C for 10-20 hours, individual particles are found to lose mass, even when the ambient humidity is held at saturation. Since the temperatures employed are too low to produce evaporation of even such relatively volatile inorganic aerosol components as ammonium sulfate, the mass loss is taken by Goetz to indicate presence of moderately volatile organic components. Goetz suggests that these components are produced by plants, and deposit on other aerosol particles to form mixed nuclei. The Los Angeles smog particles exhibit similar metastability, and there the organic component may be of industrial or vehicular origin; but Goetz reports finding the same type of thermal metastability in samples taken in the northern San Joaquin valley far from urban areas, in Arizona, and even in the air layers contacting the ocean surface in complete calm about 50 miles offshore. His chief suggestion was that much more attention be given to the origin and the influence of organic constituents of the natural aerosol.

6. Kerker

A technique for preparing monodisperse aerosols of spherical particles of sodium chloride and silver chloride was sketched by Kerker (the optical studies thereon being reported in another session of the Conference). His method consists in vaporizing the desired salt, say NaCl, and quenching it in a flow of helium to induce deposition on nuclei. By carrying the aerosol through two further stages of deposition and varying the furnace temperatures and helium-quenching temperatures, uniform sizes can be grown, ranging from perhaps 50 Angstroms up to about 1 μ. Electron micrographs reveal the particles to be perfectly spherical, and X-ray techniques indicate no crystalline microstructure. However, in the case of the NaCl spheres, transition to cubic particles will occur if the initially round particles are merely exposed for a few minutes to laboratory air of average humidity. Evidently, water vapor condenses on the particles to sufficient extent to cause migration in a solution film on the particles, and rearrangement to cubic forms quickly ensues.

A second technique, in which NaCl or AgCl spherical cores are coated with an organic annular shell (linolenic acid) was described by Kerker. These two layer aerosols permit tests of the Mie theory of scattering by non-homogeneous spherical particles.

7. Farone

Current work on airborne dusts at the White Sands Missile Range was briefly mentioned by Farone. Dust particles of one to forty micron diameter lie somewhat outside the usual range of "aerosol particles", but pose many problems in the arid Southwest. Farone invited persons interested in the dust problem to contact his group.

8. Closing discussion on gaps in the aerosol spectrum

During the Thursday session, several speakers brought out the interesting point that they were finding gaps in their observed aerosol size distributions. Since most published distributions are smooth, and since existence of gaps could pose important questions of light scattering, of condensational history during activation of the particles as condensation nuclei, and of origin of the particles, it was decided to close Thursday evening session with a general discussion of what Schotland whimsically proposed calling the "aerosol gap".

It was especially the observations of Fenn that indicated breaks in the aerosol size distribution, so it may be helpful to the reader to note that examples of his distributions may be found in his paper in Beitr. z. Phys. d. Atm., 37, 69 (1964). Distributions measured in localities as widely separated as the Smoky Mountains of Tennessee, the Gila desert of Arizona, and the New Jersey coast exhibited gaps of about 0.2 to 0.3 μ radius. Two New Jersey distributions also showed a secondary gap from about 0.4 to 0.5μ radius. Several of his rural-area distributions showed a marked drop-off below about 0.1μ radius (i.e., a failure to rise towards high counts typical of the Aitken population). Twomey had also described evidence of gaps in distributions measured by diffusional methods, gaps at sizes below 0.1μ. At such smaller sizes there is precedent for gaps, since the mobility-technique of sizing ions has often in the past suggested a "line spectrum" of those smaller particles. The principal topic of discussion thus centered on the more surprising gaps above 0.1μ radius which Fenn had found with a Goetz aerosol spectrometer.

As the discussion proceeded, two principal hypotheses for explaining the gaps were proposed and discussed, mostly inconclusively. First, some felt that the gaps must characterize "young" aerosols, formed recently enough (close enough upwind of the collection site) that coagulation processes had not yet had time to fill in the gaps. Second, others suggested that the gaps were an artifact introduced by aging, through the thermal metastability processes stressed by Goetz.

Several arguments were raised against the "young aerosol" hypothesis. The gaps were found at sizes so large that ordinary Brownian coagulation is not very effective in its smoothing actions. Coagulation is chiefly a matter of collection of the very small particles by the larger; and though this can markedly alter the shape of the distribution at very small sizes (in the Aitken range), it is difficult to see how it could fill in Fenn's gaps in less than many weeks. A further question about the "young aerosol" hypothesis was raised by Volz when he asked whether any known aerosol-production processes (outside the laboratory) were capable of producing sharp peaks such as some of Fenn's distributions showed. (The New Jersey spectra showed peaks of about 0.1μ width, in terms of radius, for example.) No one proposed industrial chimney processes as a plausible source of such narrow peaks, nor was any natural formation-process suggested as an answer to Volz's question. Thus the hypothesis that spectral gaps reflect youthfulness and that adequate aging would lead to smooth distributions of the sort we have mostly seen in the past literature was not acceptable to many discussants.

The second hypothesis, namely the suggestion that the gaps were somehow formed artificially after collections of an actually smoothly distributed aerosol, was not thoroughly examined on the floor. In retrospect, it seems that someone should have pointed out that one of the virtues of the Goetz aerosol spectrometer is that it deposits the particles at locations that correspond to the particle size ("kinetic radius") at instant of collections. Hence, if it had been true that certain cuts of the distribution were populated mainly by particles rich in volatile organics, later evaporation of the organic components would not have confounded the reconstruction of the original size distribution. Since that important point was not considered on the floor, it cannot be settled in this summary.

In the session chairman's opinion, we did not succeed in settling the interesting question of the "gaps". We must hope that these questions will be pursued, both observationally and theoretically, in the literature in the future. Are the nicely smoothed size-distributions which are to be found in our texts and references correct, or are they oversimplified average representations of wiggly individual spectra? If the wiggles are real, how can they originate? Do some spectra have gaps while others are smooth? If the gaps are real, what optical effects do they produce? These questions remained to be answered when the Thursday evening session closed at 2200.

SECOND DAY

OPTICAL PROPERTIES OF AEROSOLS

Survey of Optical Scattering Theory

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1. The subject conveniently divides into: I. Single particles and single volume elements, and II. Slabs and atmospheres. In either subject we deal with computations from a model. The reverse method to obtain particle properties directly from observations on scattering are hardly ever satisfactory.

Part I. Single particles and single volume elements

2. Molecules (Rayleigh scattering) and homogeneous drops (Mie theory) are reviewed. Sample curves show the extinction efficiency against $x = 2\pi a/\lambda$ and scattered intensity in 2 polarizations versus $\cos\theta$. New work by Irvine is available on g = average value of $\cos\theta$ weighted with scattering diagram.

3. Particles of different shape may be important in some applications. The best general method is microwave analogue measurements (Greenberg). Advanced computational methods have been applied with full success to concentric spheres and with some success to ellipsoids.

4. In a volume element with particles of different sizes many details in the scattering pattern are washed out. We can then with some justification represent the scattering diagram by a formula with a few parameters. The most important parameter, besides the albedo a, is the asymmetry factor g defined in 2. We recommend the use of the Henyey-Greenstein function

$$\Phi(\alpha) = \frac{1 - g^2}{(1 + 2g \cos\alpha + q^2)^{3/2}}$$

if only one parameter can be permitted. Good fits to some of Deirmendjian's functions with more parameters can, e. g., be made in the form

$$\Phi(a) = a e^{-ba} + c$$

A simple representation is imperative in most multiple scattering computations.

Part II. Multiple scattering; radiative transfer

5. Multiple scattering (at one wavelength) or radiative transfer (energy transported by radiation in all wavelengths) is an old subject. Much is known but buried in mathematics. The author is writing a book with many tables and graphs and with emphasis on physical interpretation.

6. The "standard problem" refers to a plane homogeneous slab. The independent variables are

μ_0 = cosine of angle of incidence
μ = cosine of angle of emergence
τ = optical depth of an arbitrary point
b = total optical depth
a = albedo
g = asymmetry factor of scattering diagram

7. The most important quantities that can be computed for such a standard slab are

reflection function	$R(a, b, \mu_0, \mu)$
transmission function	$T(a, b, \mu_0, \mu)$
point-direction gain	$G(a, b, \tau, \mu)$

The R and T are normalized by putting $R = 1$ for completely diffuse reflection (Lambert's law). G is normalized by putting it $= 1$ in the absence of the atmosphere. The famous X and Y-functions are the values of G for $\tau = 0$ (top) and $\tau = b$ (bottom). All these functions are now numerically available, at least for isotropic single scattering, for many values of a and b. Also their moments and bi-moments, which are integrals over μ and/or μ_0, are available.

8. Some methods for computing these results are discussed and compared, namely

a. Successive orders (Neumann series)

b. Added thin layer (Ambartsummian's method; invariant embedding)

c. Non-linear integral equations (based on invariance; for homogeneous layers only)

d. Added thick layers (doubling method)

Fast computers have revived the importance of a and have made d a very attractive method because it can be applied with equal ease to anisotropic scattering. Adding the influence of ground reflection is a simple special case of d.

9. The illustrations shown for possible use in the interpretation of observational data on terrestrial cloud layers, or on planetary atmospheres, include the following:

a. Approach of $J_n(\tau)$ to eigenfunction for $n \rightarrow \infty$ in method of successive orders.

b. Point-direction gain in semi-infinite layer; interpretation as escape probability.

c. Behaviour of thick layer; it is often possible to interpolate with certaintiy between $b = 1$ and $b = \infty$.

d. Ratio of total reflection to first-order reflection under various assumptions.

e. Division of incident energy among reflection, absorption and transmission, varying with a, b, and μ_o.

f. Reflection diagrams.

g. Successive orders in multiple Rayleigh scattering showing how most but not all of the polarization is produced in the first order.

10. Much interest is now devoted to the dependence of g and to the behaviour of thick layers. The factor 1-g must simply be added to the effective thickness of a thick layer for radiation diffusing through it. The fact that terrestrial stratus clouds diffusely transmit much light, yet make the solar disk invisible, points at once to a value of g fairly close to 1 (Piotrowski).

Friday morning session - R. Penndorf, Chairman

Single Particle and Small Volume Scattering for Aerosols (Theory)

(1) The scattering functions for *single* particles with real index of refraction n are known for many values of n, and a very large range of size parameters α ($\alpha = 2\pi r/\lambda$). Total scattering coefficient, angular scattering coefficients and degree of polarization are known. Due to the electronic computer technique such computations pose no problems and scattering coefficients can be generated for any desired value of n and size range. For aerosol studies, where integrations over a particle size range and wavelength range are needed, the minor fluctuations, the so-called ripples, are unimportant. Smoothed values without ripples are often completely sufficient. Hence, this problem is well understood; no further basic research and discussion are needed.

(2) The scattering, absorption, and polarization coefficients for *single* particles with *complex* index of refraction $\tilde{n} = n - i\kappa$ are also known for many values of $\tilde{n}$ and α. However, less extensive data have been generated than for real n. The results are well understood, absorption reduces the fluctuations, the ripples vanish first, and the major fluctuations become weaker the larger the absorption, i.e., κ. There exists a steep increase of the scattering coefficient in the Rayleigh range for $\alpha < 1$ and a very simple functional behaviour for $\alpha > 1$. This applies to the total as well as to the angular scattering coefficients. Again the problem is understood, data exist to derive the general behaviour and machine computations pose no difficulties. Hence, no discussion is required.

(3) The scattering functions for single particles of *non-spherical shape* pose some problems. From a mathematical point of view simple forms such as cylinders, oblate spheres, cones, and so on, can be used and the scattering functions have been computed. Forms that are difficult to express analytically create problems. In such cases, experimental studies are helpful. One investigates scaled models at microwave

wavelengths. The models can be made from materials which have refractive indices similar to those for aerosols in the visible and infrared. The models can be given any desired form and measurements can be carried out for any desired angle between incoming and scattered radiation. Such studies have been described by Greenberg (without slides).

A large body of experimental data exists for various materials (conducting surfaces, but also dielectrics, plastic material sheets over conducting surfaces, plastic materials with embedded conducting spheres). The results of such studies are hardly found in scientific journals; they are published in contract reports and some are even classified.

As long as the deviation from a sphere is small, the influence of non-sphericity on scattering coefficients is small, and the solutions for spherical particles seems a reasonable approximation.

The conclusion is reached that the scattering data for simple shapes are available, and more can be computed theoretically, but for odd shapes only experimental investigations will give useful information. Such experimental data exist for other purposes and should be transposed for the aerosol problem. The refractive index, size range, and form have to be specified so that additional microwave measurements for scaled models can be carried out.

(4) The most important point, and this was discussed in detail, is the scattering from a small volume containing an aerosol of uniform refractive index n or $\tilde{n}$. The particles inside the volume are not of uniform size, but possess a size distribution. The size distribution can be described by reasonably simple functions. As such, one may assume an exponential law (as in Junge's distribution function) or a Gaussian distribution function or any other which has been proven to exist.

Kerker described his applications to colloidal systems consisting of various aerosols and hydrosols. Theoretical computations have been carried out for refractive indices ($n = 1.43$, 1.51, and 2.07), a size range $\alpha_M = 1.9\ (0.1)\ 15$ and width parameter $\sigma_o = 0$ [illegible]$0\ (0.05)\ 0.155$[†] Total,

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[†] where α_M and σ_o are the parameters in a logarithmic distribution.

angular scattering coefficients and degrees of polarization for such size distributions have been computed and the data are stored on magnetic tape.

Experimental data are obtained and compared with the theoretical computations. He found that the experimental angular scattering data for the two polarized components in the range $\theta = 30°$ to $130°$ can be fitted to the theoretical data; he obtained the modal size, and the width parameter of the distribution function. The uniqueness of the solution has not posed a problem. The experimental data always fitted one set of theoretical data with the least deviations.

The samples have been checked by the electron microscope technique and the sizes obtained by the optical scattering method and by the electron microscope agree within the expected accuracy. The optical method seems very reliable. Coated particles will also be investigated. However, the problem of uniqueness of the solution will create difficulties. The number of parameters increases when there is a coating. It seems at present that coated particles cannot be investigated optically to determine exact size distribution and thickness of coating in a unique way. The measurement of long cylinders has been carried out by Kerker. Here experiments and theory can be compared. The results give the diameter of these "needles", and good agreement has been found.

Hodkinson presented data for measurements in a size range of $\alpha = 1.4$ to 2.2. Instrumentation has been developed to measure the transmission and determine size ranges.

Hodkinson investigated the problem of what type of instrument should be used to investigate aerosols. This question arises from the fact that instruments are often designed for convenience of handling without due regard for maximizing the effects to be studied. He therefore integrated the scattering functions (Mie) over a range of wavelengths and scattering angles. A weighting function for each type of instrument was added. Some of his conclusions are:

(a) In forward scattering instruments the effects of refractive index (n) and absorption (κ) are small.

(b) At a scattering angle of 40°, the effects of n are small but large for κ.

(c) At 90°, the effects of n are large, but the difference between transparent and absorbing particles can be small for selected values of n and κ (i. e., quartz and carbon).

(d) White light, divergent and convergent beams smooth the scattering functions considerably.

Volz presented data on scattering and polarization of cigarette smoke. Glass filters have been used with effective wavelengths between 0.38 and 0.77μ to obtain some spectral resolution. The laboratory data have been compared with theoretical data for Mie scattering and mean effective radii have been obtained. Polarization data gave a smaller radius (r = 0.12) than scattering (r = 0.21μ).

La Mer pointed out that coagulation studies of cigarette smoke exist and that coagulation may have to be considered in such investigations.

Howell presented computations of the total Mie scattering coefficient and of the intensity function for backscatter for a large range of size parameters. The computations form the basis of a study on the feasibility of using backscattered light to determine the visibility through fog and haze. The computations refer to a single wavelength (λ = 0.7μ) for the laser, and integration (λ = 0.4 to 0.7μ) for white light, and various size distribution models for haze and fog conditions. The single wavelength data contain large variation with size and do not lead to a simple relationship between visibility and backscattered signal, whereas the white light, due to the smoothing in the integration process, leads to a clearer, although still uncertain relationship. The agreement of the computations for white light with actual measurements by other investigators was displayed.

Fenn discussed a similar problem. He assumed models for size distribution and compared the theoretical computation and measurements with the observed visual range. He assumed the total number of aerosols to be constant but he varied the size distribution.

The discussion of these two papers pointed to several difficulties. Backscattering is an inefficient way to use the optical properties of aerosols, because most of the energy is scattered in the forward direction. Furthermore, the angular scattering coefficient for backscattering ($\theta = 180° \pm 10°$) undergoes large fluctuation with α and a single integration over the size distribution introduces not enough smoothing to make the scattering function a simple analytical function, whereas the second integration over the wavelength (white light) introduces the smoothing necessary to obtain a stronger relationship between visibility and backscattered fluxes.

The conclusions for this topic are that several valuable computations for selected models of the size distribution exist. Models for aerosols (turbid air), haze, fog, and clouds are used and the models vary from author to author. Such numerical computations are extremely valuable and a more systematic approach may be desirable. This means a few particular refractive indices should be selected and a number of models computed and (this is an important point) evaluated to see what changes in the assumption lead to significant differences in the results. If a group of assumptions leads to practically identical results (for practical, experimental applications) then the most simple assumption will be sufficient. I am certain that small changes in the distribution function, especially at both ends (large and small r) will contribute very little and are therefore not important.

Experiments should use the forward scattering, because the aerosols scatter most of the incoming radiation in the forward direction, but only a small amount into the backward direction. Due to the large fluctuation of the coefficients for backscattering and the lack of uniqueness, integrated values have to be used in order to obtain significant values.

The choice of wavelength determines the range of radii which can be investigated. Using the range 0.4 to 0.8μ, nothing substantial can be said about particles with $r < 0.1\mu$ and $r > 2\mu$. This had advantages because in theory, one can select the wavelength range in relation to the range size. However, the UV is useless (or at least limited) by ozone absorption and the infrared is limited by absorption bands of CO_2, O_3, and H_2O.

(5) The polarization effects are very important because they give information about large particles. This was discussed by van de Hulst, but not mentioned by Möller. Some of the speakers mentioned it, at least implicitly by referring to the two components i_1 and i_2; the radiation in the plane of polarization and perpendicular to it. Basically the four Stokes parameter can give important additional information about the scattering properties of aerosols. In experimental programs polarization should always be included.

(6) Multiple scattering within small volumes has not been discussed and can be avoided in experimental investigation by proper dilution of the aerosol sample. Hence, it is not a serious problem but very important for investigations in a real atmosphere.

Friday evening session - D. Deirmendjian, Chairman

Optical Properties of Aerosols: Measurements

The chairman introduced the session by posing the question: How can one determine the size, concentration, and nature of atmospheric aerosols from remote optical measurements? After illustrating the difficulties by means of simple examples, he tried to lead an organized discussion by inviting comments from those in the audience who had actually obtained measurements by various techniques. It quickly became apparent that only a limited number of parameters could be determined without elaborate optical measurements *in situ* (angular distribution of intensity and polarization of scattered radiation from a small sample).

Some of the salient comments are reported below *as interpreted by this writer:*

G. Newkirk reported on the success and difficulties of balloon-borne solar aureole measurements, by a method developed by himself and co-workers. In particular, he demonstrated that under certain assumptions, the broad nature of the particle distribution function could be deduced, especially the large particle end of it. Full details have been published in an excellent paper by Newkirk and Eddy (*J. A. S.*, *21*, 35, 1964). There is no doubt that this method is among the best for deducing the total number of particles above the instrument because of the well known forward scattering property of aerosols. Also, it is possible to detect layering when the system is used as a sounding. There is good evidence that thin stratospheric layers of aerosols can and do exist and the aureole measurements are corroborated by other independent techniques.

E. K. Bigg very judiciously disclaimed any possibility of deducing the aerosol particle size distribution from the type of twilight measurements conducted by himself and colleagues. The analysis of this kind of data clearly is fraught with difficulties because of the complex mechanisms involved. Undoubtedly twilight photometry does indicate the presence of stratospheric aerosols, particularly volcanic dusts. Bigg also mentioned evidence of thin layers of the order of 10 to 100 m thick, from twilight measurements, and later showed some remarkable photographs of the Agung volcano dust layer over Australia, taken from an aircraft.

L. Elterman commented on the searchlight method which has been used by himself and others for several years. He claimed that at least the total attenuation coefficient can be thus obtained (always under certain assumptions about the shape of the phase function, as emphasized by R. W. Fenn). Experimentally the simplicity of the searchlight method recommends itself, but again there are inherent theoretical difficulties in the interpretation of the data.

G. Fiocco, who with his collaborators, (cf. Nature, 199, 1963) was the first to announce the detection of very high layers of atmospheric dust by the pulsed laser beam technique, discussed its potentialities. The chairman, in his introductory remarks, had drawn some attention to this method by writing down a sort of laser radar equation and discussing its implications(cf J. Geophys. Res., 70, 743, 1965), mainly because of the novelty of the technique and the current tendency in science to consider research which uses the newest technical discovery as the panacea for all old problems. Although Fiocco mentioned the possibility of designing bistatic systems in order to get scattering at other angles than the backward direction, he emphasized the superiority of monostatic systems mainly because of their simplicity and mobility on a single platform, which could eventually be airborne. At present, it appears that the laser beam technique is best suited for the detection and ranging of scattering layers, particularly very tenuous ones in the mesosphere and near space, which are otherwise unobservable by ground-based techniques. However, future possibilities to make full use of the remarkable properties of laser light of various frequencies should be carefully examined, including the effects of lower atmospheric scattering on the pulses en route to and from the target, as suggested by Z. Sekera.

R. Watson mentioned the high powered laser radar system being developed at NCAR and its potentialities in detecting layering in aerosols. He also made the interesting suggestion that perhaps the invariance of the normalized scattering function at scattering angles 40° to 45° (predicted theoretically by Deirmendjian, 1964[†]) might be used in deducing the concentration by means of future bistatic systems. In the discussion, G. Fiocco concurred that thin layers should be definitely detectable because of the short length of the laser pulses, of the order of 10 m.

R. W. Fenn described his very interesting, though incomplete, measurements of the scattering phase function of ice particles produced in the laboratory, and obtained by means of a nephelometer. His results appear to confirm the existing few measurements in that the intensity function is rather smooth at large scattering angles and the polarization remains constant over a wide range of angles in contrast to the situation with liquid water clouds. In our opinion, nephelometric measurements, especially on natural ice clouds, are extremely important here because of the difficulties of solving exactly the mathematical problem of scattering on finite polyhedral particles.

B. A. Silverman described another interesting application of laser techniques to a determination of particle sizes and distributions by an analysis of their diffraction patterns. The method has the advantage of rapid sampling of a small volume without disturbing the particles. The analysis is based on the diffraction theory of two dimensional particles and hence it seems best suited to the study of opaque particles. For a detailed description, the reader is referred to a paper by B. A. Silverman, B. J. Thompson, and J. H. Ward in J. Appl. Meteor., 3, p. 792 (1964).

In conclusion, as mentioned at the beginning, we believe that the discussion clearly brought out the fact that partial optical measurements cannot by themselves provide all the parameters needed to describe an atmospheric aerosol. Complete optical measurements might, but then the aerosol has to be so accessible that more direct physical and chemical analyses become available. For inaccessible and remote aerosol layers

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[†] See reference in F. Möller's presentation.

(and we are talking only of optically thin matter, so that single scattering theory is applicable) the optical techniques can only be used as an adjunct to check or test other data or hypotheses.

Personally, we felt that having a "theoretical" type preside over a discussion of experimental results is not a bad idea, because the former is mainly concerned with the meaningfulness of the measurements in testing various theoretical models, regardless of the technical difficulties and the fascination of the experimenter with the intricacies of a novel piece of equipment. Conversely, an "experimental" type would be an effective chairman for the theoretical discussion, because he will be able to keep the theoretician's feet on the ground, so to speak, and away from too many non-dimensional quantities and too far out, idealized models, which, while mathematically elegant and tractable, may be meaningless to the man who deals with nature's realities.

THIRD DAY:

THE INFLUENCE OF AEROSOLS ON ATMOSPHERIC RADIATION FLUXES

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I. Introduction

This paper will deal with observations of radiation fluxes and the properties of the atmospheric aerosols which may be derived from them.

There are generally three ways to obtain such information which have been taken by different authors.

A. One chooses a particular model of number density and size distribution of particles, computes the radiation flux affected by them and looks to see whether or not the result is consistent with a general conception of the calculated phenomenon.

B. One repeats such calculations for a very large (10^3 to 10^4) number of different models and selects that model as true which gives the best agreement with observations.

C. One uses an inversion method to infer directly the aerosol characteristics from exact measurements. The advantages and restrictions of this method have been discussed yesterday.

The radiation fluxes that can be observed are

1. direct radiation from solar or articifial sources,
2. scattered radiation from solar or artificial sources including sky radiation and polarization,
3. terrestrial (long wave) radiation emitted by the earth and/or the atmosphere.

II. Direct solar radiation

The earliest attempt to characterize the aerosol content of the atmosphere was made by F. Linke (1922, 1942) who compared the observed depletion of solar radiation with the number T of Rayleigh atmospheres giving the same extinction, in total or only short wave solar radiation. T is the turbidity factor. There exists a comprehensive survey of turbidity factors as observed at many places (Steinhauser, 1935).

From a physical point of view, A. Ångström's (1929) turbidity coefficient B was a better definition. It is based on the assumption of a haze extinction coefficient $a_{D\lambda} = B\lambda^{-\alpha}$ with $\alpha = 1.3$ and it uses only the short wave part of solar radiation, $\lambda < 630\ m\mu$, for the determination of B. Schüepp (1949) left the numerical value of α open and determined it from measurements taken behind different filters (also Ångström, 1964). Knowledge of α permits the determination of the exponent ν^* in Junge's aerosol size distribution law if an assumption of the refractive index m of the particulate matter can be made (Volz, 1954; Junge, 1955). Measurements with spectrographs or interference filters will give more details of both the spectral distribution of radiation and the size distribution of the particles. The refractive index, however, may not be the same for all size ranges of the aerosol or the exponent in the size distribution law may change with the radius r (Bullrich and Volz, 1961) or even different formulae may hold (Deirmendjian, 1964). Therefore measurements in very wide spectral ranges (extending to the long wave infrared) will reveal more and better knowledge of the size distributions.

From observations, the following statements have been inferred. The generalized turbidity coefficient B shows a maximum in the early afternoon hours and an annual variation in middle latitudes with the maximum in summer and minimum in winter. Tremendous annual variations occur at Leopoldville between the dry-dusty and rainy-clean seasons (Schüepp, 1949; Valko, 1962, 1963). The equivalent height of aerosols was determined only by Volz (1955) by comparing the extinction coefficient in a horizontal (visual range) and a slant beam through the whole atmosphere (solar radiation). He found for this equivalent height in winter a value of 1.15 km, in summer forenoon 1.7 km, in summer afternoon 3.6 km, while usually estimations are used ranging from 1.0 to 1.4 km.

For the wavelength exponent α mean values have been determined close to 1.3 or 1.5; Ångström (1964) has given a mean value of 1.9 for Potsdam and an annual variation between 1.3 in January to 2.6 in July. No regular daily variation has been found. A slight decrease is indicated with altitude in the lower part of the troposphere from observations at ground stations (Potsdam 1.9; Davos 1.3) while Newkirk and Eddy (1963) derived an increase from 1.0 at 6 km to 1.5 at above 15 km from their measurements of scattered light in the sun's aureole.

A few spectral investigations are available for the extinction of solar radiation in the infrared spectrum around 10μ, although a clear distinction between absorption by the far wings of absorption lines of H_2O and CO_2 and aerosol extinction can hardly be obtained in this region. In the polluted atmosphere of London the extinction is 3 to 4 times stronger than in the much clearer air of Ascot, England (Roach and Goody) At this place further studies indicated only "small influences of aerosols" (Bignell et al.) A careful inspection, however, of their figures reveals an increase in the extinction coefficient by about 30 to 50% over the value in the cleanest atmosphere with a decrease of the visibility from 15 to 5 km, and an even stronger increase with a visual range of 3 km.

Other studies in this spectral range have been based on measurements of the terrestrial downcoming radiation. Dave et al. found an increase of the absorption coefficient by a factor of 2 to 3 in a day with dense haze and--most remarkably--a change in the shape of its spectral variation. Bolle found in the Negev desert a much higher emission of the atmosphere than in St. Agata, Italy; a disturbance in the general change of the emission at 8-9μ, i.e., in the reststrahlen band of quartz and feldspar, obviously demonstrates a pollution by mineralic dust from the surface.

Summarizing, one has to state that there are not many observations of the spectral extinction by atmospheric haze and a comprehensive survey appears to be needed.

III. Scattered radiation

Measurements of the scattered radiation can give much more information on the scattering particles and their size distribution than extinction measurements due to two parameters involved, i.e., wavelength, λ, and the scattering angle, ϕ. A combination of such measurements with polarization measurements, with simultaneous measurements of high spectral resolution over wide spectral ranges would given an almost ideal program for determining the characteristics of the atmospheric haze. Apparently, such a complete program has never been implemented (with the possible exception of G. V. Rosenberg's observatory close to Moscow).

The dependence of scattering on the angle ϕ, also named phase function, scattering function, or indicatrix, has been measured many times with artificial light sources. The most thorough and complete series has been given by Barteneva. She collected more than 700 measurements of the indicatrix at ground stations between sea level and 3200 m altitude. In the total material she distinguished ten classes of increasing ratio of forward to backward scattering and three types of the shape of the indicatrix: a gradual and a steep type, the latter again subdivided into two groups, one with "peaks" indicating the primary and secondary rainbow and one without "peaks". The ratio between the maximum scattering at $\phi = 0$ and the minimum value varied between 2.2 (almost Rayleigh) and 625 (gradual) or 750 (steep). The rainbow peak first appeared with increasing turbidity at a visibility of 0.6 - 0.8 km but the indicatrix without peak was also still observed in the same class: this means that fog and dry haze may cause the same low visibility. As far as I know these indicatrices have not been evaluated in terms of the aerosol distribution.

The same instrumentation has been flown in aircraft up to 17.5 km. An elongated indicatrix was still observed at all altitudes with an anomalous increase upward of the elongation near the tropopause and further increase above the tropopause up to the top altitude. Obviously, the aircraft entered the upper haze layer first observed by Junge et al. At these altitudes strong variations from flight to flight occurred

(Sandomirsky et al.).

Corresponding measurements in the sky are preferably made in the sun's almucantar from which a much easier evaluation of the indicatrix can be obtained from measurements in the sun's vertical. Volz (1956) has developed to a high degree of precision this kind of evaluation and has shown that different types of angular distribution in the aureole indicate quite different size distributions of particles. A particularly useful measure is the ratio of the angular distribution in two wavelengths. An extension of these measurements up to $\lambda = 1.76\mu$ demonstrated a change of the indicatrix which is not easy to explain; it may perhaps indicate a change of the refractive index with λ or r (Bullrich and Volz).

The principle of studying the forward scattering close to the sun's disc has been used by Newkirk and Eddy also. They sent a balloon-borne coronagraph up to 25 km height and were able to obtain reliable measurements as close as 1.7 degrees from the sun's limb. Their measurements confirmed the existence of the stratospheric aerosol layer. A different method of forward scattering study was used by Volz and Goody in their difficult but careful evaluation of twilight measurements. They were able to detect the aerosol layer between 15 and 30 km in many cases, thus confirming the old hypothesis that the purple light has its origin in a highly elevated haze layer.

Another fine confirmation of the haze layer was obtained by Rössler. He measured the scattered light from the zenith by a rocket-borne instrument up to 30 km. The apparent radiance of the mass unit (in excess of the Rayleigh radiance) was $9 \cdot 10^{-5}$ W kg^{-1} sr^{-1} at 16 km and 6 times stronger at 25 km.

In all calculations of the radiance of the sky or of the illumination from the total sky, the main difficulty is a correct treatment of the multiple scattering in the hazy atmosphere. The problem has been solved only for the clean atmosphere. Sekera (1956) and collaborators, not only gave a complete theoretical computation of the brightness of the sky, but they also found the eventual explanation of the distribution of polarization over the sky and the origin of neutral points (Coulson et al.). Recently Sekera and Kano have shown what influence on the behaviour of the neutral points is affected by a haze layer, either in the high or low atmosphere.

The multiple scattering in a turbid atmosphere has been calculated and published by Feigelson et al. They used some fixed types of indicatrices and it is therefore difficult to take from their tables the flux of scattered radiation in the case of overlapping Rayleigh and Mie scattering where the extinction coefficient as well as the indicatrix change with the wavelength. Comparisons with observations cannot be made easily since the absorption by atmospheric gases was neglected in these calculations.

Robinson (1963) selected from many stations between tropical and arctic latitudes observations of sky and global (sun and sky) radiation on a horizontal surface which show the prevailing influence of scattering or extinction respectively. He compared the observations with calculations of the same fluxes, taking into account absorption by atmospheric gases and Rayleigh scattering only. From these comparisons he demonstrated that there is not only a scattering but also a true absorption by atmospheric haze particles independent of the wavelength. This absorption amounts to about 5-20% of the extraterrestrial irradiation of a horizontal surface; it is larger than the absorption by water vapor in large cities, of the same order of magnitude over island stations and smaller, but still clearly perceptible, in the clean atmospheres of South Africa and Antarctica. Sekihara using the same method confirmed the findings in observations from Japan.

In order to see whether or not these instruments can be confirmed Leupolt derived the scattering coefficient from spectral radiance measurements of the sky and compared it with spectral extinction coefficients obtained from direct radiation measurements. These might contain haze scattering and absorption (in window regions of the spectrum, free of absorption bands) while only the scattering coefficient should be decisive for the radiance. In the polluted atmosphere of Munich, he found an absorption of 35% of the total extinction coefficient of haze.

The effect of haze absorption should be studied thoroughly because it might have strong influence on the interpretation of the extinction pattern by aerosol models and on our conception of the atmospheric radiation budget.

IV. Terrestrial radiation

As early as 1919, A. Defant studied the influence of infrared emission by haze particles upon the daily variation of the temperature in the layers near the ground. This investigation, however, could not yet use our present knowledge of aerosols and their influence on radiation.

Robinson (1950) collected many observations of the downcoming atmospheric radiation at night and found a few days with excessively low amounts of radiation under the same conditions (water vapor content, lapse rate) as many other measurements with much higher intensities. He attributed the surplus of atmospheric radiation to atmospheric haze or dust present in varying quantities. A comparison with absorption coefficients as calculated by Deirmendjian (1960) might be worthwhile.

In recent years a phenomenon has been reported which apparently does not permit any other explanation but by assuming the existence of a haze layer in the high troposphere. Rönicke, Kuhn, and Riehl have reported that in the tropics or at least in tropical air masses the terrestrial upward net radiation flux at night does now show the usual monotonic increase with height. In a layer of about 6 km below the tropopause, occasionally also above it, a decrease of the net flux occurs. This phenomenon cannot be explained by an particular distribution of the absorbing gases which affect the normal upward increase. The only acceptable explanation is given by the hypothesis of a haze layer or a thin cirrus cloud in these altitudes (Zdunkowski). This layer must possess an absorption coefficient, probably gray, within the window region where the other absorbers do not absorb.

Of course, this assumption is unsatisfactory as long as there is no verification of the existence of this layer by other observations. Only one observation which came to my attention quite recently may support it. Ramanathan et al. have shown that a regular increase of the ozone amount from day to night must be derived from Dobson measurements against sun or moon. An increase by the observed amount cannot be understood from physical considerations. If one eliminates the influence of aerosol scattering in the evaluation of the original spectral measurements the increase vanishes. This however means that not the ozone amount but the aerosol scattering increases at night by a factor of about 7, then amounting to 30-50% of the Rayleigh extinction in the total atmospheric column. It is striking to note that the apparent increase of ozone at night as well as the upper tropospheric heating effect by infrared radiation has been reported in tropical air masses only. Thus the two observations, although coming from the UV and the IR end of the spectrum, may support each other.

It has been observed many times that the deepest temperature minimum at night does not occur at the surface of the earth but at a somewhat higher level, about 20 to 50 cm above the ground. An explanation might be sought in some thermal emission of a thin haze layer. However, calculation of this effect has shown that the effect is not strong enough to explain the phenomenon (Möller, 1964).

Bibliography

Ångström, A. (1929): On the atmospheric transmission of sun radiation and on dust in the air I. Geograf. Ann. 11, 156.

Ångström, A. (1964): The parameters of atmospheric turbidity. Tellus 16, 64-75.

Barteneva, O. D. (1960): Scattering functions of light in the atmospheric boundary layer. Bull. Acad. Sci. USSR, Geoph. Ser., 1237-1244 (Engl.).

Bignell, K., F. Saiedy, and P. A Sheppard (1963): On the atmospheric incrared continuum. J. Opt. Soc. Amer. 53, 466-479.

Bolle, H. J (1964): private communication.

Bullrich, K., and F. Volz (1961): Scattering function and polarization of skylight .... J. Met. 18, 306-318.

Coulson, K. L., J. V. Dave, and Z. Sekera (1960): Tables related to radiation emerging from a planetary atmosphere with Rayleigh scattering. U. of Calif. Press, 548 pp.

Dave, J. V., P. A. Sheppard, and C. D. Walshaw (1963): Ozone distribution and the continuum from observations in the region of the 1.043 cm^{-1} band. Quart. J. R. Met. Soc. 89, 307-318.

Defant, A. (1919): Ann.d. Hydrogr. 47, 93. (quoted after Linke (1943))

Deirmendjian, D. (1960): Atmospheric extinction of infra-red radiation. Quart. J. R. Met. Soc., 86, 371-381.

Deirmendjian, D. (1964): Scattering and polarization properties of water clouds and hazes on the visible and infrared. Appl. Opt. 3, 187-196.

Feigelson, E. M. et al (1960): Calculation of the bri .ness of light in the case of anisotropic scattering. Pt. I (1960), Pt. II (1963). Consultants Bureau, Inc., New York (transl. from Russian).

Junge, C. (1955): The size distribution ··· as determined from ··· optical data on the atmosphere. J. Met. 12, 12-25.

Junge, C , C. W. Chagnon, and J. E. Manson (1961): Stratospher aerosols. J. Met. 18, 81-108.

Kuhn, P. M., and V. E. Suomi (1960): Infrared radiometer soundings on a synoptic scale. J. Geoph. Res. 65, 3669-3677.

Leupolt, A. (1964): Determination of aerosol absorption from spectroscopic sky measurements (0.5 to 2.5μ). Paper presented to the International Radiation Symposium, Leningrad, August 1964.

Linke, F. (1922): Transmission Koeffizient u. Trübungsfaktor. Beitr. Phys. Frei. Atm. 10, 91.

Linke, F. (1942): Die Sonnenstrahlung u. ihre Schwächung in d. Atm., in Handb. d. Geophysik Vol. 8, Gebr. Borntraeger, Berlin, Kap. 6, pp. 239-338.

Möller, F. (1964): Einige Überlegungen zur Frage des abgehobenen Temperaturminimums bei Nacht. Met. Rdsch. 17, 86 89.

Newkirk, G., and J. A. Eddy (1964): Light scattering by particles in the upper atmosphere. J. Atm. Sci. 21, 35-60.

Ramanathan, K. R., G. N. Shah and P. D. Angrejii (1964): Comparison of ozone amounts made with sun and moon observations: nighttime increase in large particles scattering. Paper presented to the International Ozone Symposium, Albuquerque, N. M., September 1964.

Riehl, H. (1962): Radiation measurements over the Caribbean during the autumn of 1960. J. Geoph. Res. 67, 3935-3942.

Roach, W. T., and R. M. Goody (1958): Absorption and emission in the atmospheric window from 770 to 1250 cm^{-1}. Quart. R. J. Met. Soc. 84, 319-333.

Robinson, G. D. (1950): Notes on the measurement and estimation of atmospheric radiation. Quart. J. R. Met. Soc. 76, 37-51.

Robinson, G. D. (1963): Absorption of solar radiation by atmospheric aerosol, as revealed by measurements at the ground. Arch. Met. Geoph. Biokl. B 12, 19-40.

Rönicke, G. (1961): Über Messungen d. Verlaufs d. Wärmestrahlungsbilanz i. d. freien Atm. in San Salvador, C. A, Z. Met. 15, 8.

Rössler, F. (1963): Das diffuse Himmelslicht in d. hohen Atmosphäre. Geof. pura appl. 56, 123-134.

Sandormirsky, A. B., N. P. Altovskaya and G. I. Trifonova (1964): Indicatrices of brightness at heights of 8-17.5 km. Isvest. Acad. Nauk. USSR. Geoph. Ser., 958-966 (russ.).

Schüepp, W. (1949): Die Bestimmung d. Komponenten d. atm. Trübung aus Aktinometermessungen. Arch. Met. Geoph. Biokl. B. 1, 257.

Sekera, Z. (1956): Recent developments in the study of the polarization of skylight. Advances in Geoph. 3, 43-104.

Sekera, Z. and M. Kano (1964): Effect of the height of a turbid layer on the polarization of skylight. Paper presented at the Internnational Radiation Symposium, Leningrad, August 1964.

Sekihara, K. (1964): On the absorption properties of atm. dust particles as considered from various kinds of ground level observations. Paper presented to the International Radiation Symposium, Leningrad, August 1964.

Steinhauser, F. (1934): Die mittlere Trübung d. Luft an versch. Orten, Beurteilt nach Linke'schen Trübungsfaktoren. Gerlands Beitr. Geoph. 42, 110-121.

Valko, P. (1962): Untersuchung über d. vertik. Trübungsschichtung d. Atm, Arch. Met. Geoph. Biokl. B. 11, 142-210.

Valko, P. (1963): Über d. Verhalten d. Atm. Dunstes am Alpensüdfuss. Arch. Met. Geoph. Biokl. B 12, 458-474.

Volz, F. (1954): Die Optik u. Meteorologie d. Atm. Trübung. Ber. Deutsch. Wetterd. 2, No. 13.

Volz, F. (1955): Atm. turbidity and its spectral extinction. Geof. pura appl. 31, 119.

Volz, F. (1956): Optik d. Tropfen I, Optik d. Dunstes, in Handbuch d. Geophysik Vol. 8, Gebr. Borntraeger, Berlin. Kap. 14, pp. 842-897.

Volz, F. E., and R. M. Goody (1962): The intensity of the twilight and upper atmospheric dust. J. Atm. Sci. 19, 285-406.

Zdunkowski, W. (1963): Untersuchungen über die langwellige Strahlung der Atmosphäre. Gerlands Beitr. Geoph. 72, 21-47, 103-126.

Saturday morning session - Z. Sekera, Chairman

The chairman opened the session by asking for questions pertaining to the preceding talk by Professor Möller.

Dr. Newkirk found the measurements made on Mt. Everest for clear skies unbelievable as they do not show any aureole, which he had found at 25 km. The discrepancy was attributed to the systematic error in the nephelometer which is not able to measure scattering near the source.

Dr. McDonald asked about the possibility that Robinson's computations might be in error due to the sensitivity of reflection of the solar angle.

Prof. Möller pointed out that the measurements showed the dependence on a solar angle that was taken into consideration; as the amount of virtual absorption is the same in magnitude as the absorption due to the water vapor, it is difficult to explain this absorption by haze by some minor errors in computation.

The Chairman in his closing remarks objected to the way Dr. Robinson separated the scattering by molecules and by aerosol. Such separation is dangerous; the recent computations made by Dr. Kano indicate the importance of the interaction of these two radiative fields. Only by such interaction is it possible to explain observed anomalies in skylight polarization. He therefore recommends that Robinson's computations be revised and this interaction be taken into consideration. The difficulties of computation of multiple scattering by aerosol particles can be eliminated by a new method of separation of forward peak, just developed at UCLA.

The question of whether aerosol particles are absorbing or not can be answered by studies of backward scattering, since Dr. Deirmendjian's computations of the scattering indicate very important differences in this region for absorbing and non-absorbing particles. If in the computation of aerosol scattering their absorptivity is taken into consideration, it may be possible to get different behavior of skylight in the rainbow and anticorona region that can be treated by proper polarization measurements.

The chairman supported by Professor van de Hulst warned against too much confidence in some computations which are lacking in accuracy, having been performed up to now mostly by desk calculators.

Prof. Möller mentioned that he found several errors in published tables. For similar reasons the chairman warned against extensive trust in Waldram's measurements.

Dr. Bullrich mentioned his measurements of ellipticity of light scattered by aerosol particles, which give better agreement with the computation if the absorption of aerosol particles is taken into consideration.

Dr. Volz points to the possible influence of carbon particles in the urban atmosphere on the measurements taken in the Munich area.

The chairman then asked for comments on Professor van de Hulst's paper, presented the day before.

Dr. Irvine gave his report on his computations with asymmetric scattering functions. The results shown in several slides indicated a better agreement with the measurements when the finite Legendre polynomial series are replaced by Greenstein's phase function. As pointed out by the chairman, this agreement can be explained by the fact that Greenstein's phase function represented an infinite series of Legendre polynomials.

Prof. van de Hulst emphasized this fact and warned against the use of a finite number of Legendre polynomials in the representation of large particle scattering.

Dr. Fenn then described some computations made with the use of the Monte Carlo method, following the photon after each scattering. It was used first in the problem of illumination of a plane-parallel layer by an isotropic point source above this layer and the ratio of the flux of the reflected radiation to the flux of the source was computed. Then it was used in a model atmosphere with the aerosol distribution given by the R^{-3} law, and for a visibility of 25 km. The magnitude of the flux of scattered radiation increases with the number of collisions (i.e., with the number of scattering) allowed. This increase levels off by about 15-20 collisions.

Prof. van de Hulst mentioned that with a given optical thickness used (for example, 4.4) the square of the optical thickness (20 in this case) gives the number of collisions needed before some kind of saturation is achieved, which agrees with the presented data.

The chairman pointed out that a similar method was used about ten years ago by Mr. Lake at Imperial College in London. The results of these computations in the total flux were much less accurate than any simple approximation such as that given by Chandrasekhar. Dr. Fenn then replied that the success of this method lay in the capacity of the computer. The newly available computers enable one to take a large number of photons into consideration and thus to get a really statistical distribution.

Answering the questions by Dr. Newkirk about the possible inclusion of polarization due to the scattering, Dr. Fenn replied that this could be done but would increase the computation problems tremendously.

Dr. Irvine added that the flux computations probably could be done but not the angular distribution of emerging intensity. In response to a question from Dr. McDonald, Dr. Fenn replied that the computations had been made on the 7094 and each run took one to two minutes. To Dr. McDonald's questions, Dr. Fenn replied that the random walk was three-dimensional.

Dr. Zdunkowski was then called upon to give his report on the computation of the effect of haze on the divergence of heat fluxes in the long wave region. He considered an atmosphere consisting of water vapor and haze, represented by Deirmendjian's model of continental haze, with exponential decrease in the total number of haze particles with height. In this computation the attenuation by water vapor and haze particles, acting as emitters and primary scatterers, were taken into consideration in addition to the reflectivity of the ground. The results of the computations showed that near the ground the cooling rates are little affected by the haze. If, however, a temperature discontinuity of -3° is assumed at the ground and the reflectivity of the surface is assumed to be four percent, a very large instantaneous cooling rate of about 32°C per hour appeared in the air directly overlying the surface. In the upper atmosphere the effect of haze was also small.

It would require 100 times larger aerosol content to double the cooling of the atmosphere within the lower 2 km layer. However, if the amount of haze is increased 100 times, an increase in the downward flux by about 50 percent will follow, which has never been observed. The speaker then reached the conclusion that 15 times larger haze content than that in Deirmendjian's model could still produce a normal situation.

The lack of time prevented any further discussion, and the session was adjourned at 12:15 p. m.

Saturday afternoon session - J. Dave, Chairman

"Airborne Radiometer Measurements of Effects of Particulates on Terrestrial Flux"

Dr. P. M. Kuhn

Thin layers of particulates or clouds in the high troposphere and lower stratosphere are generally not visible to an observer at the surface, even under the favorable viewing conditions at sunset. They are not easily detected by a television camera in a satellite from above. Yet these layers affect upwelling infrared flux.

A flight program employing balloon borne radiometers and a jet aircraft from which visual observations of particle layers were made was conducted to measure the attenuating effects of these layers on upwelling terrestrial flux. An estimate of the error such attenuation can cause in surface temperatures deduced from the upward flux measurements was then made. The balloon borne radiometers were launched from a desert area of California. During the ascents the presence of cirrus or particle layers could not be visually or photographically detected from the surface. They were, however, detected by the radiometer during ascent and by observers in a jet aircraft.

The observations indicated an attenuation of from to 15 percent in upward infrared flux as a result of an observed particle layer beneath the higher balloon borne measurements. This attenuation could cause a 5°C surface temperature estimate error. Several similar observations and calculations without observers aloft show the same result.

Question from Dr. Albert Arking: Do you take into account the emmisivity of the surface?

Dr. Kuhn: We used the radiometer at Edwards Air Force Base over Rosamond Dry Lake where the humidity is rather low, and a meter above the surface. This took care of any infrared shadowing and gave us an interface temperature.

Dr. McDonald: What is the chance that the attenuation is due to volcanic dust?

Dr. Kuhn: I didn't mean to say that I know what it is. In fact, I'm really here on the edge of the meeting, and not into your work. I don't claim to know what exactly it is. We have to use a gross method and just say quantitatively it's something up there that is attenuating. I personally believe that it's ice, as does Stuckey, but I will not deny that on 20 February 1963 that it could have been Bali. I just don't know what its composition is.

"Measurements of Sky Radiation on Haleakala, Hawaii"

By K. Bullrich

The absolute spectral measurements of the solar radiation and of the intensity and polarization of the sky radiation allow the determination of the size distribution and number density of the atmospheric aerosol. The observations could be corrected for multiple scattering following an approximation given by Dr. de Bary. The size distribution obtained from such a study at Mainz (W. Germany) was found to agree well with the continuous power law distribution given by Prof. Junge for continental aerosol.

Such observations were taken on the island of Maui (Hawaii) at two levels: (a) 3400 m above sea level (above the trade wind inversion) and, (b) sea level during April 1964. Simultaneous direct measurements of aerosol number density at both levels of observation taken with a Schultz counter show a particle number density of 10 per cc. However, a turbidity factor of the order of 2 was measured at $\lambda = 0.5\mu$. There was no significant difference between the observations of the intensity and polarization taken at two levels. The intensity values are relatively higher than the other places and the degree of polarization about 50 percent.

These results can be explained only in the terms of the presence of an upper atmospheric aerosol layer which can be attributed to the March 1963 volcanic eruption of Mt. Agung. The detailed analysis shows that the particles in this layer do not have a continuous power law distribution. The comparison of observations with theory indicates the presence of particles of radius 0.4μ and 2.5μ.

Some results of the computations of intensity were also presented for particles with complex refraction index.

Prof. Sekera: Similar measurements of the polarization taken at the same location by us in 1961 showed that the degree of polarization at that time was about 10 percent higher than that given by Dr. Bullrich. This shows a presence of aerosol due to volcanic eruption.

As mentioned before (p. 51), one of my students, Dr. Kano, computed the degree of polarization of the skylight with two different models. A thin aerosol layer (1) at the bottom, and (2) at the top of a Rayleigh atmosphere. Standard power law of distribution was used for the aerosol size. The variations observed under conditions of strong low level haze agreed very well with case (1). However, the anomalies observed after the volcanic eruption were not brought out by case (2). I therefore feel the size distribution of aerosols from volcanic eruption is Gaussian in nature.

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