Translated from *Uspekhi Khimii* **59** 1654–1675 (1990) U.D.C. 551.510.42

Problems of the distribution of the global aerosol

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ABSTRACT. In view of the short residence time of the aerosol in the atmosphere, knowledge of its distribution in space is essential for the understanding of many processes associated with this distribution, for example, the formation of clouds and hence of the entire cloud cover over the Earth's sphere. This review considers both the distribution of aerosol particles with respect to size (from the standpoint of chemical and physical characteristics) and the distribution of aerosols in space in a broad sense of this term.

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I. Introduction

Apart from the main gases (nitrogen and oxygen), the atmosphere contains small admixtures of other substances. Investigators working in the following fields of meteorology and atmospheric chemistry are concerned with the study of these admixtures: (1) the physics and chemistry of clouds (water); (2) the chemistry of gaseous impurities; (3) the physics and chemistry of aerosols.

Compared with the bulk of the main gases contained in one cubic metre of dry air, which is ~1300 g (this quantity depends on temperature), the overall content of water in all three states of aggregation is less than 10 g, that of gaseous admixtures (excluding water) is up to 0.5 g, and that of aerosol particles is up to 50 mg. Thus the mass concentration of the aerosol does not exceed that of minor gaseous admixtures. For this reason many scientists and unfortunately society in general underestimate the importance of aerosols in view of the low ratio of the mass concentration of the aerosol to the mass concentration of the gaseous admixtures.

Nevertheless, aerosols play a key role in the atmosphere. For example, certain aerosol particles serve as condensation nuclei for water vapour in clouds, so that the turnover of water in the absence of these particles would change significantly and hence the climate of the Earth would also change.

For example, half of the total amount of the aerosol is formed as a result of the gas–particle transformation (including both homogeneous and heterogeneous condensation) and this process is therefore an important route leading to the removal of many gaseous species from the atmosphere.

Aerosol particles consist of a large set of chemical substances (organic and inorganic) minerals, and particles of biogenic origin. The influence of aerosol on man and the environment is due to the presence of these compounds, whose concentrations are sometimes very low (of the order of a nanogram per cubic metre as in the case of certain specific organic compounds or pollen particles).

The question arises why aerosols play such an important role in the atmosphere. Firstly, they influence the field of the Earth's electromagnetic radiation as a whole, since they absorb and scatter a large proportion of the incident solar radiation; secondly, they ensure the presence, directly in the gas phase, of a large surface area for the occurrence of chemical reactions of various gaseous substances; thirdly, they influence the turnover of atmospheric water, since they serve as condensation nuclei in clouds; fourthly, they provide a surface on which natural radioactive nuclides are adsorbed; fifthly, they influence the chemical and radiation (albedo) properties of the surface of the planet as a result of the dry and wet precipitation on its surface; sixthly, they alter the electric field of the atmosphere, since aerosol particles can be charged; finally, they promote the spread of plants and diseases.

It is difficult to estimate the influence of the aerosol on the state of the atmosphere, because in a geochemical sense the aerosol particles have a very short lifetime. Furthermore, there are additional meteorological characteristics (horizontal, vertical, and time distributions of the aerosol) and there exists a distribution of the particles with respect to size (the number, surface, and mass concentrations, the chemical properties, and the refractive index are different for particles of different size) which hinders in its turn the determination of their influence. It is also noteworthy that, among all the components of the atmosphere, the suspended substances are unique as regards the complexity of their chemical composition.[1]

To facilitate the description and study of such a complex system and to investigate its influence on the environment, scientists have introduced various parameters such as the electrical conductivity of air, the mass concentration of the aerosol, the extinction coefficient, etc. The present state of knowledge about the entire wide variety of the parameters of the atmospheric aerosol will be described in this review.

II. Distribution of the aerosol in space and time

The term "distribution" has many meanings even if it is understood in a fairly narrow sense. In the physics and chemistry of aerosols, this term usually means the number or volume distribution of particles with respect to size and more rarely the spatial or geographical distribution.

Henceforth we shall discuss the fact that aerosol particles have a polydisperse distribution with respect to size associated with different mechanisms of their formation. After their appearance, aerosol particles undergo a change in their chemical and disperse compositions.

Meteorological processes mix the aerosol in the vertical and horizontal directions. This process involves all the particles regardless of their size and chemical composition.

Other processes act only on specific particles depending on their size and chemical properties. Gravitational influences on the largest particles regardless of their composition forces them to move towards the surface of the Earth. Brownian motion is responsible for the process called "thermal diffusion", which is also independent of the chemical nature of the particles. As a result of thermal diffusion, small particles, having the maximum mobility, collide with other particles and thus coagulate. Cloud drops are formed on aerosol particles with a specific chemical composition, coagulate into larger drops, and are finally precipitated as rain or evaporate once more.[2]

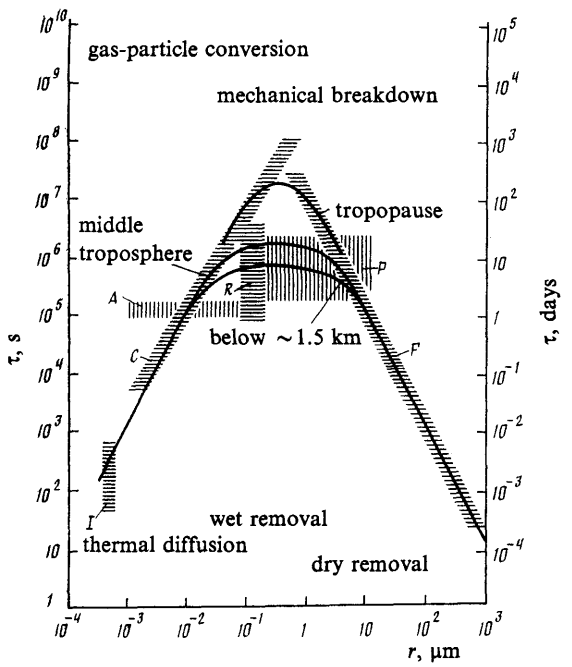


Figure 1. Dependence of the lifetime (τ) of aerosol particles on their radius.[3] The calculation was performed taking into account different independent data: *A*—distribution of Aitken particles in space; *C*—diffusion of the aerosol in dynamic equilibrium; *R*—radioactivity data; *P*—results of studies of the atmospheric water cycle; *F*—sedimentation data.[4]

All these processes limit the residence time of the aerosol particles in the atmosphere, altering thereby their size distribution. The principal result of the influence of these processes is the removal from the atmosphere of the smallest and largest particles and hence the conversion of the polydisperse aerosol into a monodisperse aerosol. This process is called "ageing". Fig. 1 illustrates the dependence of the lifetime of the aerosol particles in the atmosphere on their radius.[3] Small and large particles have shorter lifetimes and only particles with radii in the range 0.1–1 μm exist longer in the suspended state. This time might be longer in the absence of interactions of the aerosol with clouds, a situation which occurs in the upper troposphere and in the stratosphere.

In this connection, it must be emphasised that the term "lifetime" used in this article is not identical with the chemical or geochemical concept of the lifetime of a chemical compound, since the compounds contained in aerosols can continue to be present in the atmosphere after the coagulation of the aerosol particles. There is also another possibility: chemicals can pass from one set of particles to another, differing in size and behaving quite differently in the atmosphere. Yet another remark may be made—the significance of the lifetime in a reactor such as the atmosphere is that of the period during which the concentration of the compound diminishes by a factor *e*. Fig. 1 shows that the lifetime is a maximum for particles with radii in the range 0.1–1 μm and amounts to ~8 days (for the lower troposphere).

It has been suggested[5] that there is an inverse relation between fluctuations in time and space and the lifetime. Substances with a long lifetime are distributed more uniformly. Atmospheric oxygen, for which only rare, episodic measurements of its concentration have been recommended, may serve as a good example.[6] The lifetime of sulphur dioxide is short and SO_2 therefore exhibits pronounced fluctuations in time and space. An equation describing these fluctuations was obtained subsequently[7] using a simple model:

$$V^2 = \frac{1 - \exp(-2T_{max}/T)}{2[1 - \exp(-T_{max}/T)]^2} \frac{T_{max}}{T} - 1,$$

where V is the relative standard deviation (variance), T the lifetime in arbitrary units of time, and T_{max} the "maximum" time (in arbitrary units) during which the admixture exists as an individual compound (T_{max} varies from 10 h to 30 days).

It must be emphasised that there are contradictory attitudes to this statistical approach.[8] Nevertheless, there is qualitative agreement concerning the inverse proportionality between the lifetime and the fluctuation and also about the fact that substances with a lifetime amounting to days should have a standard deviation in their distribution in time and space equal to ~2, while for substances with a shorter lifetime the standard deviation should be much greater.

The dependence of the lifetime of aerosol particles on their size influences the behaviour of the aerosol as a function of time and its distribution in space.

In evaluating the data presented in this review, one must take into account the possible long-time changes in the characteristics of the aerosol under the influence of natural and anthropogenic factors. To some extent, such changes have been summarised in the author's previous communication.[3] Thus any aerosol distribution can reflect the situation only at the time to which it refers.

III. Distribution of aerosol particles with respect to size

Among all the types of distribution of the atmospheric aerosol with respect to size, the most important is the distribution of the number concentration of the particles with respect to diameter,

Table 1. Parameters of the number distribution of the aerosol and its integral characteristics for the size ranges indicated and for the aerosol as a whole.

Type of aerosol	Size ranges	Subrange $10^{-7} < r < 10^{-5}$ cm			Subrange $10^{-7} < r < 10^{-4}$ cm			Subrange $r > 10^{-4}$ cm			Total range $r > 10^{-1}$ cm		
	type of particles according to Junge	Aitken			large			giant			sum		
	fraction according to Whitby	nucleation			accumulation			coarsely disperse					
	aerosol parameters i	n_i	R_i	$\lg\sigma_i$	n_i	R_i	$\lg\sigma_i$	n_i	R_i	$\lg\sigma_i$			
	sum for subranges I'	N'	S'	V'	N'	S'	V'	N'	S'	V'	N	S	V
Polar	$i=1$	$2.17\cdot10^{1}$	$6.89\cdot10^{-6}$	0.245							$2.17\cdot10^{1}$	$2.45\cdot10^{-8}$	$1.24\cdot10^{-13}$
	$i=2$				$1.86\cdot10^{-1}$	$3.75\cdot10^{-5}$	0.300				$1.86\cdot10^{-1}$	$8.54\cdot10^{-9}$	$3.52\cdot10^{-13}$
	$i=3$							$3.04\cdot10^{-4}$	$4.29\cdot10^{-4}$	0.291	$3.04\cdot10^{-4}$	$1.73\cdot10^{-9}$	$7.56\cdot10^{-13}$
	I'	$1.63\cdot10^{1}$	$8.01\cdot10^{-9}$	$1.94\cdot10^{-14}$	$5.83\cdot10^{0}$	$2.13\cdot10^{-8}$	$1.98\cdot10^{-13}$	$1.53\cdot10^{-2}$	$5.92\cdot10^{-9}$	$1.02\cdot10^{-12}$	$2.19\cdot10^{1}$	$3.47\cdot10^{-8}$	$1.23\cdot10^{-12}$
Background tropospheric	$i=1$	$1.29\cdot10^{2}$	$3.60\cdot10^{-7}$	0.645							$1.04\cdot10^{2}$	$1.73\cdot10^{-8}$	$5.12\cdot10^{-13}$
	$i=2$				$5.97\cdot10^{1}$	$1.27\cdot10^{-5}$	0.253				$5.97\cdot10^{1}$	$2.39\cdot10^{-7}$	$2.36\cdot10^{-12}$
	$i=3$				$6.35\cdot10^{-1}$	$2.59\cdot10^{-5}$	0.425				$6.35\cdot10^{-1}$	$3.63\cdot10^{-8}$	$3.44\cdot10^{-12}$
	I'	$1.03\cdot10^{2}$	$1.84\cdot10^{-8}$	$4.55\cdot10^{-14}$	$4.19\cdot10^{1}$	$2.44\cdot10^{-7}$	$2.56\cdot10^{-12}$	$7.71\cdot10^{-2}$	$3.20\cdot10^{-8}$	$3.71\cdot10^{-12}$	$1.65\cdot10^{2}$	$2.92\cdot10^{-7}$	$6.31\cdot10^{-12}$
Marine	$i=1$	$1.33\cdot10^{2}$	$3.90\cdot10^{-7}$	0.657							$1.09\cdot10^{2}$	$2.47\cdot10^{-8}$	$9.66\cdot10^{-13}$
	$i=2$				$6.66\cdot10^{1}$	$1.33\cdot10^{-5}$	0.210				$6.66\cdot10^{1}$	$2.36\cdot10^{-7}$	$1.88\cdot10^{-12}$
	$i=3$				$3.06\cdot10^{0}$	$2.90\cdot10^{-5}$	0.396				$3.06\cdot10^{0}$	$1.74\cdot10^{-7}$	$1.32\cdot10^{-11}$
	I'	$1.25\cdot10^{2}$	$1.90\cdot10^{-8}$	$4.76\cdot10^{-14}$	$5.26\cdot10^{1}$	$2.90\cdot10^{-7}$	$3.12\cdot10^{-12}$	$2.84\cdot10^{-1}$	$1.22\cdot10^{-7}$	$1.29\cdot10^{-11}$	$1.78\cdot10^{2}$	$4.32\cdot10^{-7}$	$1.60\cdot10^{-11}$
Background continental	$i=1$	$9.97\cdot10^{2}$	$1.00\cdot10^{-7}$	0.328							$5.05\cdot10^{2}$	$3.67\cdot10^{-10}$	$5.37\cdot10^{-17}$
	$i=2$	$8.42\cdot10^{2}$	$2.18\cdot10^{-6}$	0.505							$8.39\cdot10^{2}$	$7.51\cdot10^{-7}$	$1.60\cdot10^{-11}$
	$i=3$							$7.10\cdot10^{-4}$	$6.24\cdot10^{-4}$	0.277	$7.10\cdot10^{-4}$	$7.83\cdot10^{-9}$	$4.51\cdot10^{-12}$
	I'	$1.26\cdot10^{3}$	$1.19\cdot10^{-7}$	$2.42\cdot10^{-13}$	$8.11\cdot10^{1}$	$5.13\cdot10^{-7}$	$6.59\cdot10^{-12}$	$4.37\cdot10^{-1}$	$1.35\cdot10^{-7}$	$1.39\cdot10^{-11}$	$1.34\cdot10^{3}$	$7.60\cdot10^{-7}$	$2.05\cdot10^{-11}$
Desert sandstorm	$i=1$	$7.26\cdot10^{2}$	$1.00\cdot10^{-7}$	0.247							$3.69\cdot10^{2}$	$1.53\cdot10^{-10}$	$1.25\cdot10^{-17}$
	$i=2$	$1.14\cdot10^{3}$	$1.88\cdot10^{-6}$	0.770							$1.08\cdot10^{3}$	$2.72\cdot10^{-5}$	$3.51\cdot10^{-8}$
	$i=3$							$1.78\cdot10^{-1}$	$1.08\cdot10^{-3}$	0.438	$1.78\cdot10^{-1}$	$2.00\cdot10^{-5}$	$8.49\cdot10^{-8}$
	I'	$1.26\cdot10^{3}$	$1.28\cdot10^{-7}$	$2.74\cdot10^{-13}$	$1.85\cdot10^{2}$	$2.52\cdot10^{-6}$	$4.68\cdot10^{-11}$	$1.47\cdot10^{1}$	$4.43\cdot10^{-5}$	$1.35\cdot10^{-7}$	$1.45\cdot10^{3}$	$4.72\cdot10^{-5}$	$1.20\cdot10^{-7}$
Agricultural region	$i=1$	$6.65\cdot10^{3}$	$7.39\cdot10^{-7}$	0.225							$6.65\cdot10^{3}$	$7.81\cdot10^{-8}$	$3.76\cdot10^{-14}$
	$i=2$	$1.47\cdot10^{2}$	$2.69\cdot10^{-6}$	0.557							$1.46\cdot10^{2}$	$3.59\cdot10^{-7}$	$1.95\cdot10^{-11}$
	$i=3$	$1.99\cdot10^{3}$	$4.19\cdot10^{-6}$	0.266							$1.99\cdot10^{3}$	$9.30\cdot10^{-7}$	$3.32\cdot10^{-12}$
	I'	$8.61\cdot10^{3}$	$6.44\cdot10^{-6}$	$1.23\cdot10^{-12}$	$1.71\cdot10^{3}$	$5.80\cdot10^{-7}$	$5.14\cdot10^{-12}$	$3.44\cdot10^{-1}$	$1.42\cdot10^{-7}$	$1.65\cdot10^{-11}$	$8.79\cdot10^{3}$	$1.37\cdot10^{-6}$	$2.29\cdot10^{-11}$
Industrial region	$i=1$	$9.93\cdot10^{4}$	$6.51\cdot10^{-7}$	0.245							$9.93\cdot10^{4}$	$9.99\cdot10^{-7}$	$4.81\cdot10^{-13}$
	$i=2$	$1.11\cdot10^{3}$	$7.14\cdot10^{-7}$	0.666							$1.00\cdot10^{3}$	$7.84\cdot10^{-7}$	$6.43\cdot10^{-11}$
	$i=3$	$3.64\cdot10^{4}$	$2.48\cdot10^{-6}$	0.337							$3.64\cdot10^{4}$	$9.38\cdot10^{-6}$	$3.49\cdot10^{-11}$
	I'	$1.35\cdot10^{5}$	$6.72\cdot10^{-6}$	$1.12\cdot10^{-11}$	$1.41\cdot10^{3}$	$4.20\cdot10^{-6}$	$3.00\cdot10^{-11}$	$7.61\cdot10^{-1}$	$3.53\cdot10^{-7}$	$5.81\cdot10^{-11}$	$1.37\cdot10^{5}$	$1.12\cdot10^{-5}$	$9.87\cdot10^{-11}$
Stratospheric (20 km)	$i=1$				$4.49\cdot10^{0}$	$2.17\cdot10^{-5}$	0.248				$4.49\cdot10^{0}$	$5.10\cdot10^{-8}$	$8.34\cdot10^{-13}$
	$i=2$												
	$i=3$												
	I'	$4.07\cdot10^{-1}$	$3.36\cdot10^{-10}$	$9.54\cdot10^{-16}$	$4.10\cdot10^{0}$	$4.76\cdot10^{-8}$	$6.97\cdot10^{-13}$	$1.78\cdot10^{-2}$	$3.32\cdot10^{-9}$	$1.44\cdot10^{-13}$	$4.49\cdot10^{0}$	$5.10\cdot10^{-8}$	$8.34\cdot10^{-13}$

Note. n_i (i = 1, 2, 3) is the number concentration (in cm^{-3}) for the logarithmic-normal distribution of particles in different size ranges. The primed quantities indicate the additive contribution of various modes to the number (N', cm^{-3}), surface (S', cm^{-1}), and volume (V') particle concentrations in each size range indicated.

surface area, and volume. We shall therefore distinguish this type of distribution from the distributions of other parameters which will be discussed below.

The first continuous distribution of the number concentration with respect to size was proposed by Junge[9] in 1952 as a power function. This function was updated[10] in the following form:

$$dN(r)/d\lg r = n^*(r_0)(r/r_0)^{-\nu^*},$$

where $dN(r)/d\lg r$ is the density of the distribution of the number concentration, $N(r)$ the number concentration of particles with a radius greater than r, r the particle radius, r_0 an arbitrarily selected radius, $n^*(r_0)$ the density of the distribution for $r = r_0$, and ν^* the power exponent.

This formulation of the equation has the advantage that the distribution density has the dimensions of concentration despite the fact that it is the concentration density. The form of the distribution of particles with sizes in the range 0.1–10 μm is represented, according to this equation, by a straight line in terms of double logarithmic coordinates.

The advantage of this well known Junge distribution is its simplicity—only three parameters are used: the minimum radius r_0 of the fiducial recovery of the function, the coefficient $n^*(r_0)$ of the expression, and the power exponent $-\nu^*$. This distribution describes satisfactorily certain optical characteristics of the atmosphere.[11] Nevertheless, it does not describe the behaviour of the particles outside the range of their sizes indicated.

Studies carried out during the last two decades have yielded much new information about the distribution of the atmospheric aerosol with respect to size and this required a refinement of the formula describing the distribution.

At the beginning of the 1970's,[12] attempts were made to introduce for the description of the distribution with respect to size the Gaussian probability function related to the aerosol formation and transformation processes. As a result of this approach, certain particle size ranges were differentiated as "modes": nuclear, accumulation, and coarsely disperse.

Although the legitimacy of this approach is still questionable, the use of logarithmic-normal functions and their superposition leads to the following form of the equation for the distribution function:

$$\frac{dN}{d\lg r} = \sum_{i=1}^{3} n_i \frac{1}{\lg \sigma_i \sqrt{2\pi}} \exp\left\{-\frac{[\lg (r/R_i)]^2}{2(\lg \sigma_i)^2}\right\},$$

where $\lg \sigma_i$ is the degree of polydispersity of each logarithmic-normal distribution, R_i the average radius for each mode, and n_i the weight multiplier for the ith distribution.

In this equation, there are nine parameters but it can cover the entire range of sizes of the atmospheric aerosol—from 0.001 μm to several hundreds of microns.

The number distribution can be converted into a distribution with respect to the surface or volume (mass) of the particles, bearing in mind that

$$S = 4\pi r^2 \text{ and } V = {}^4/_3\pi r^3,$$

where S is the surface area of the sphere and V its volume.

Table 1 presents the parameters for all types of aerosol which are nowadays believed to be present in the atmosphere. The data quoted refer to the average relative humidity and a wind strength equal to 4 points on the Beaufort scale. Fig. 2 presents the corresponding types of distributions. The "polar" aerosol denotes particles of the layer near the Earth's surface in the Arctic and Antarctic regions.[13–15] The term "background tropospheric" aerosol is understood as the aerosol at heights above 5 km over dry land and above 3 km over the surface of oceans. This aerosol is fairly homogenous and occupies ~70% of the volume of the troposphere. The "background tropospheric" aerosol has been studied in mountain [observation] stations in air masses which move from above downwards and over the surface of oceans in order to exclude sea salt aerosol.[16–22] Together with sea salt aerosol, the background aerosol constitutes the "marine" aerosol. However, owing to the approximate nature of the averaging procedure, a claim of the type "marine aerosol – background aerosol = salt aerosol" is incorrect.[16,23,24] The term "background continental" aerosol refers to the aerosol in regions not subject to anthropogenic influences.[10,22,25] However, there is a minimal amount of information about this aerosol. The "dust sandstorm" aerosol was differentiated as a special type of natural aerosol in view of the enormous power of the source of these particles, their ability to travel over long distances, and their possible influence on the climate.[24,26,27] The "aerosol in agricultural regions"[10,28–30] near populated agricultural areas as well as the "aerosol in industrial regions",[10,29] formed as a result of anthropogenic influences, have been differentiated as separate types. The "stratospheric aerosol" differs from all the types indicated above by the fact that it is described by a unimodal logarithmic-normal distribution and is fairly monodisperse.[31] It is noteworthy that, after volcanic eruptions, the concentration of particles in the layer of stratospheric aerosol is a maximum but then rapidly diminishes.[32] The data presented on the stratospheric aerosol refer to a height of 7 km above the tropopause and the samples were taken one year after volcanic eruptions during the period between 1970 and 1981. This type of distribution is in fact to be expected bearing in mind the ageing effect and the long residence time of the particles in the stratosphere.

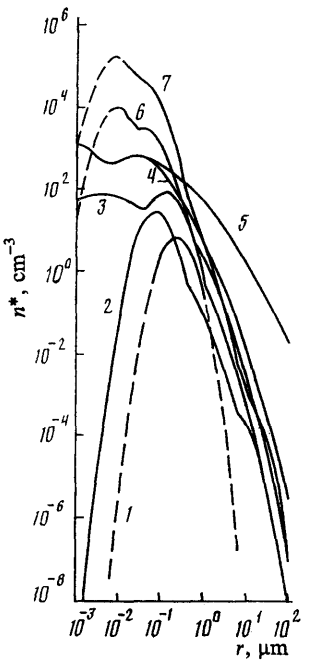


Figure 2. Distribution of the number concentrations of different types of aerosol in the form of the dependence of $dN/d\lg r$ on the particle radius. Type of aerosol: (*1*) stratospheric; (*2*) polar; (*3*) background tropospheric; (*4*) background continental; (*5*) desert sandstorm aerosol; (*6*) aerosol from agricultural regions; (*7*) aerosols from industrial regions.

Fig. 2 illustrates interesting features in the size distribution of the particles of different aerosols. The polar and stratospheric aerosols have fairly narrow, monodisperse distributions typical for "aged" aerosols. Therefore, in this case, on passing from the number distribution to the surface or volume distribution, the form of the distribution does not change while the average radius undergoes a slight alteration. The dust storm aerosol differs from the background continental aerosol only by the higher concentration of particles whose size exceeds 0.1 μm and such particles constitute the main bulk of the aerosol. Although in dust storms the number concentration of the Aitken particles exceeds very slightly that for the particles of the background continental aerosol,[24] their mass concentration is much higher.

The background tropospheric aerosol is of great interest. It has been suggested[33] that this aerosol represents the aged continental background aerosol with a number concentration of ~300 particles cm^{-3}. Indeed, the number concentration is of this order of magnitude but the form of the distribution of the background tropospheric aerosol shows that in this case a continuous gas–particle transformation process takes place, since the fraction of fine particles is high. It is unlikely that such an aerosol is "aged". Similar gas–particle transformation processes have been observed for the background continental aerosol and also for aerosols from agricultural areas and industrial regions. This was in fact to be expected. However, it is essential to note that the distribution of these submicron particles with respect to size has been inadequately studied and further investigations are needed. The concentration of the aerosol with particles larger than 0.1 μm diminishes with increase in the particle radius, as predicted by the Junge distribution. On passing to the distribution of the particles with respect to volume, the distribution function assumes the form of a fairly even plateau and in the determination of the mass of the aerosol this range of sizes is therefore most important, while the Aitken particles make an insignificant contribution.

In conclusion yet another important remark has to be made. For all types of aerosols, the most significant differences in the form of the distribution are observed in the case of the finest and largest particles. For particles of intermediate size, such differences are insignificant. It seems that this finding confirms the earlier hypothesis of a relationship between the lifetime of the aerosol and the fluctuations in its parameters: the maximum variations occur for particles with a minimal lifetime in the atmosphere.

Fig. 2 does not present the confidence intervals for individual distributions. The latter are therefore in no way a result of a large number of averaging steps but are rather a result of certain hypotheses and approximations based on the few published data. The main reason for the deficiency of information is the enormous experimental difficulty of measuring the distribution of particles over the entire range of sizes. Much still remains to be done in this field in the future.

IV. Dependence of the properties of the aerosol on the particle size

In this section we shall not discuss the properties of the aerosol which can be readily calculated from the particle distribution function with respect to size, such as the rate of deposition, mobility, and others, but shall concentrate attention on the measurable properties of the aerosol. The parameters describing the distribution of these properties are important for all types of aerosols and not only for the atmospheric aerosols. The majority of these parameters are calculated on the assumption that the aerosol particles are spherical.

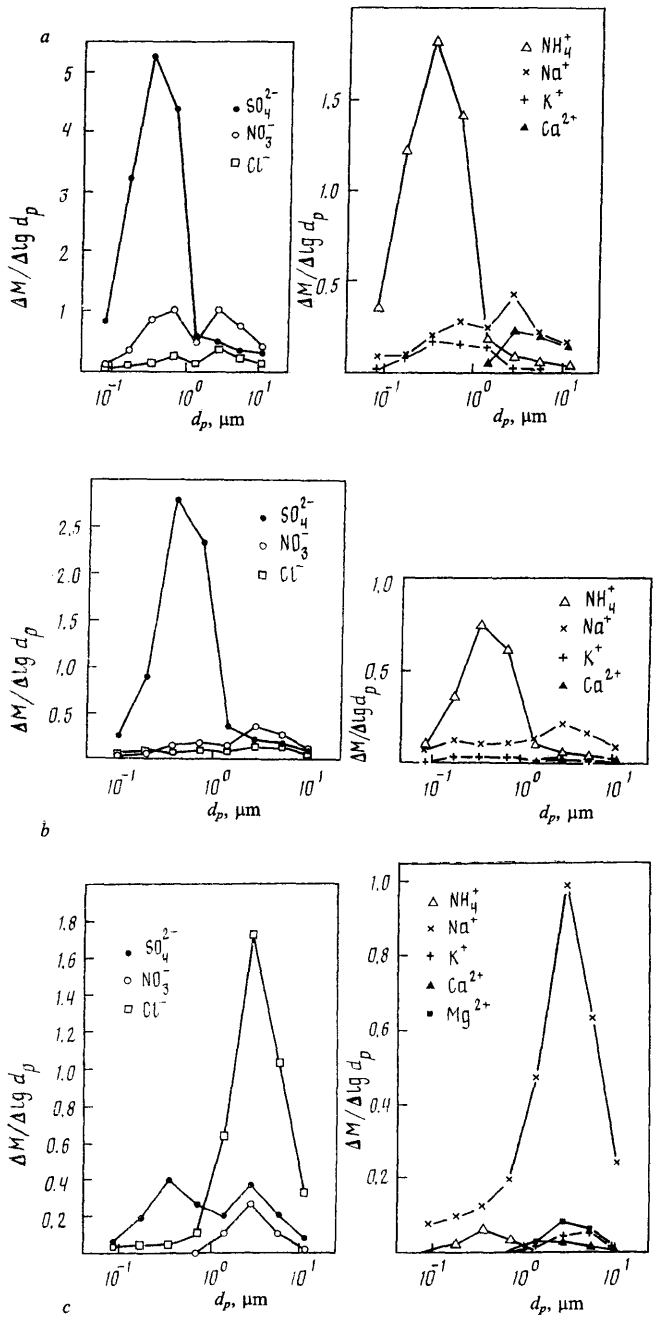


Figure 3. Differential mass distribution of the aerosol $\Delta M/\Delta \lg d_p$ (μg m^{-3}) with respect to size (diameter d_p, μm) of the most widely distributed ions for industrial (*a*), marine (*b*), and background continental (*c*) types of aerosol.[37] Data obtained using impactors.

In order to investigate the distribution of any kind of properties of the aerosol, it is essential to know the size distribution of the particles. In most cases, in order to be able to investigate properties of the aerosol such as the elemental or ionic composition, it is necessary to accumulate a sufficient mass of the substance. Such accumulation usually requires a long time (several days or even weeks). During this period, the aerosol can change or can be contaminated by impurities from local random sources. In order to avoid this and to obtain reliable results, it is necessary to exercise much caution. One of the ways of avoiding such faults, involves a decrease in the sampling time although it is then sometimes necessary to sacrifice information about the size distribution of particles. The distribution function is very frequently not measured and only the overall mass concentration of the aerosol, its overall chemical composition, or the content of only one chemical compound are investigated. Investigators sometimes restrict their study to only specific particles, for example, giant aerosol particles. The main bulk of the substance of the aerosol is very frequently concentrated in such particles.

One should always remember, particularly in the study of the dependence of the elemental or chemical composition of the aerosol on the particle size, that the aerosol has a mixed composition.[34] It is necessary to distinguish external and internal mixing of the chemical composition. By external mixing, one understands the presence of a multiplicity of particles having different chemical compositions, each particle consisting of one pure substance or specific set of compounds, determined by the source of the aerosol, for example, ammonium sulphate of sea salt. Internal mixing describes various stages of the disappearance of this "purity", which leads ultimately to the formation of a mixture of substances in each particle. Evidently particles of the first type are encountered only in the vicinity of specific sources of the aerosol, while those of the second type are a result of a secondary transformation of the aerosol (ageing). In describing the chemical composition of a mixed aerosol, the authors frequently do not indicate how the reported results have been treated. Evidently the content of any substance in the aerosol can be represented by the dependence on the effective size of a particle seemingly consisting of only the given pure substance or by the dependence on the true size of the particle (containing also other substances) whose composition includes the given compound.[35] The concentration of ions and not chemical compounds is more frequently quoted.

The first studies of the dependence of the ionic composition of large and giant aerosol particles on their size were carried out in 1953.[36] Fig. 3 illustrates the dependence of the concentration of sulphate ions on the size of aerosol particles. The data were obtained at a [research] station in an agricultural locality in the Federal Republic of Germany. This dependence has several maxima in the range of sizes 0.1 – 10 μm. Their positions coincide with the positions of the maxima in the distribution of the mass concentration of the NH_4^+ ions, which confirms the claim that the most widely distributed water-soluble compound in aerosols is ammonium sulphate. The maxima in the distribution of the concentrations of other ions investigated (K^+, Na^+, Ca^{2+}, NO_3^-, and Cl^-) are in the range of sizes 1 – 10 μm.

Similar measurements have been carried out in England for the marine aerosol and the aerosol from industrial regions.[38] If the contents of cations and anion are compared with aerosol particles whose sizes are in the range 0.1 – 1 μm, a deficiency of cations is observed. It seems to the author that the main cation which was not taken into account in these investigations was H^+. The above hypothesis has also been confirmed by studies of other investigators.[39] Hence one may conclude that the atmospheric aerosol should possess acid properties.

The most widely distributed ions over the surface of oceans are sodium and chloride ions. The maximum in their mass distribution lies in the range of aerosol sizes 1 – 10 μm. It is most likely that these are water droplets containing sea salt. It would appear that exactly the same distribution should have been observed also for the NH_4^+ and SO_4^{2-} ions if the origin of ammonium sulphate had been similar. However, sulphate ions are mainly supplied by aerosol particles with sizes in the range 0.1 – 1 μm brought from the continents (the so called "excess" sulphates[40]). Only one communication[16] reports the distribution of ammonium sulphate over the entire range of sizes of aerosol particles. The number distribution of the $(NH_4)_2SO_4$ concentration has a distinct maximum in the range of sizes 0.01 – 0.1 μm, after which the concentration of the sulphate diminishes with increase in particle size. If one goes over to the differential mass distribution, then a second maximum appears in the range 0.1 – 1 μm (the results were obtained from an electron microscope analysis of the morphology of the particles). These data are open to some doubt because the ammonium sulphate particles can vaporise *in vacuo* during preparatory operations.

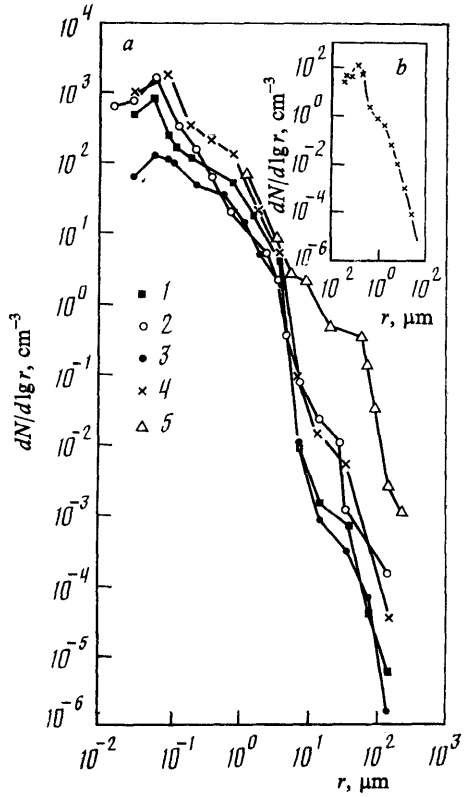


Figure 4. Differential distribution function of the number concentration of the mineral aerosol from the Sahara desert with respect to size (*a*). Data obtained by taking samples on filters in Mali (*1* and *2*), Senegal (*3* and *4*), and Libya (*5*). The inset (*b*) shows the distribution for the aerosol over the Atlantic ocean:[16] (*1*) and (*3*) pure air; (*2*) and (*4*) dust containing air; (*5*) dust storm.

Table 2. Mass concentrations of substances in the aerosol according to X-ray diffraction data.

Step (D_{50}, μm)	QU	CA	DOL	GYP	FELD	NaCl	AmCl	AmS	AmS2N	AmS3N	CaAmS	PbAmS	LET	TSP	%XTAL
A (2)	0.87± 0.53 (6)	1.47± 0.89 (6)	0.98± 0.71 (6)	0.03± 0.07 (1)	0.86± 0.50 (6)	0	0	0	0	0	0	0	0	10.2± 5.8 (6)	43±8 (6)
B (0.65)	0.30± 0.18 (6)	0.48± 0.28 (6)	0.29± 0.16 (6)	0.09± 0.18 (2)	0.21± 0.23 (4)	0	0.01± 0.02 (1)	0.16± 0.25 (2)	0	0	0.18± 0.27 (3)	0	0.03± 0.07 (1)	3.9± 1.8 (6)	44±8 (6)
C (0.37)	0.04± 0.04 (4)	0.12± 0.18 (2)	0.03± 0.04 (2)	0.02± 0.04 (1)	0	0	0	0.96± 1.2 (6)	0.25± 0.22 (4)	0	0.08± 0.10 (3)	0.008± 0.20 (1)	0.14± 0.22 (2)	3.2± 2.3 (6)	57±14 (6)
F (<0.37)	0.17± 0.11 (5)	0.36± 0.24 (5)	0.11± 0.12 (5)	0	0	0	0	2.22± 1.52 (6)	0	0	0.32± 0.49 (2)	0.14± 0.13 (4)	0.20± 0.33 (2)	8.8± 4.4 (6)	47±19 (6)
Sum of specimens	1.37± 0.67 (6)	2.43± 1.42 (6)	1.41± 0.89 (6)	0.13± 0.21 (2)	1.07± 0.71 (6)	0	0.01± 0.02 (1)	3.36± 2.9 (6)	0.25± 0.22 (4)	0	0.58± 0.51 (5)	0.14± 0.13 (4)	0.38± 0.60 (2)	26.2± 14 (6)	46±10 (6)

Overall content of ions

SO_4^{2-}: 3.3±1.9 (1.6–7.0) $n=6$ (34 nequiv. m^{-3})
NO_3^-: 0.21±0.29 (0–0.8) $n=4$ (3 nequiv. m^{-3})
NH_4^+: 1.1±0.70 (0.5–2.5) $n=6$ (61 nequiv. m^{-3})
Cl^-: 0.004±0.01 (0–25) $n=1$ (0.1 nequiv. m^{-3})
CO_3^{2-}: 1.7±1.1 (0.7–4.2) $n=6$ (28 nequiv. m^{-3})

Note. The aerosol was collected in April–June 1986 on the outskirts of Toronto (Canada). The arithmetical-mean mass concentrations and standard deviations (in μg m^{-3}) are presented. The number of measurements on which the averaging is based is indicated in brackets. The steps of the impactor are designated by the letters *A*, *B*, and *C* (*F* is the output filter). Abbreviations: QU = quartz, CA = calcite, DOL = dolomite, GYP = gypsum, FELD = feldspar, NaCl = halite, AmCl = NH_4Cl, AmS = $(NH_4)_2SO_4$, AmS2N = $(NH_4)_2SO_4.2NH_4NO_3$, AmS3N = $(NH_4)_2SO_4.3NH_4NO_3$, CaAmS = koktaite, PbAmS = $PbSO_4.(NH_4)_2SO_4$, LET = letovicite, TSP = total suspended particles, and %XTAL = percentage fraction of substances analysed by X-ray diffraction.

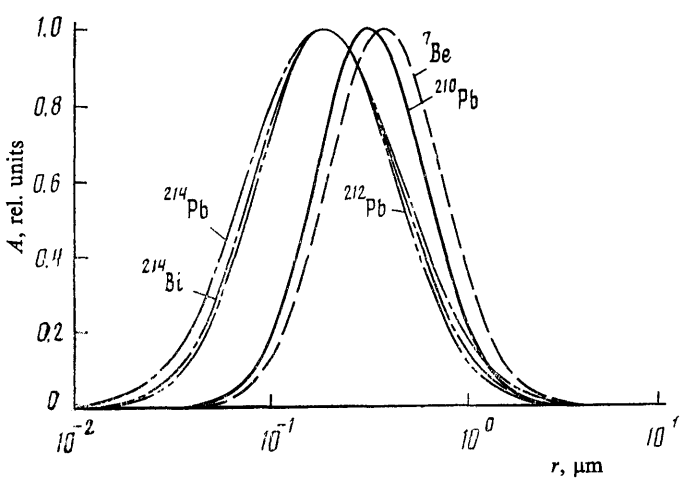


Figure 5. Dependence of the radioactivity *A* (in arbitrary units) on the radius of aerosol particles (for natural radionuclides).

The dependence of the concentrations of ions such as Br^-, Mg^{2+} [39] as well as substances of mineral origin (Al_2O_3, SiO_2, Fe_2O_3, CaO) on the particle size is not described in the literature. The dependence of the composition on particle size is also not available for the organic aerosol.

The distribution of water-insoluble minerals with respect to particle size has been reported.[27] Fig. 4 illustrates the number distribution of the continental aerosol compared with the marine aerosol. In all cases the maximum in the distribution is observed for particles with a radius less than 0.1 μm. The decrease in concentration with increase in particle radius is non-monotonic. On passing to the mass distribution, maxima arise in the range of sizes 1–10 μm, 10–100 μm, and higher.

The first studies on the distribution of minerals with respect to particle size in the continental aerosol have been reported.† Table 2 presents the results of the analysis of specimens selected in the Spring and in the Summer. It was established that minerals constitute ~50% of the mass of the aerosol. These compounds are present over the entire range of particle sizes.

The natural radionuclides entering the atmosphere are rapidly adsorbed on aerosol particles and, since such adsorption is due to diffusion, the distribution of the radioactivity of the aerosol depends on its distribution with respect to the size and surface area of the particles. Although theoretical calculations predict that the maximum radioactivity should be observed for particles whose size is about 0.1 μm, the latest results of the study of the distribution of the

†W T Sturgess, R M Harrison *Atmos. Environ.* 1989, **23**, 1083–1098.

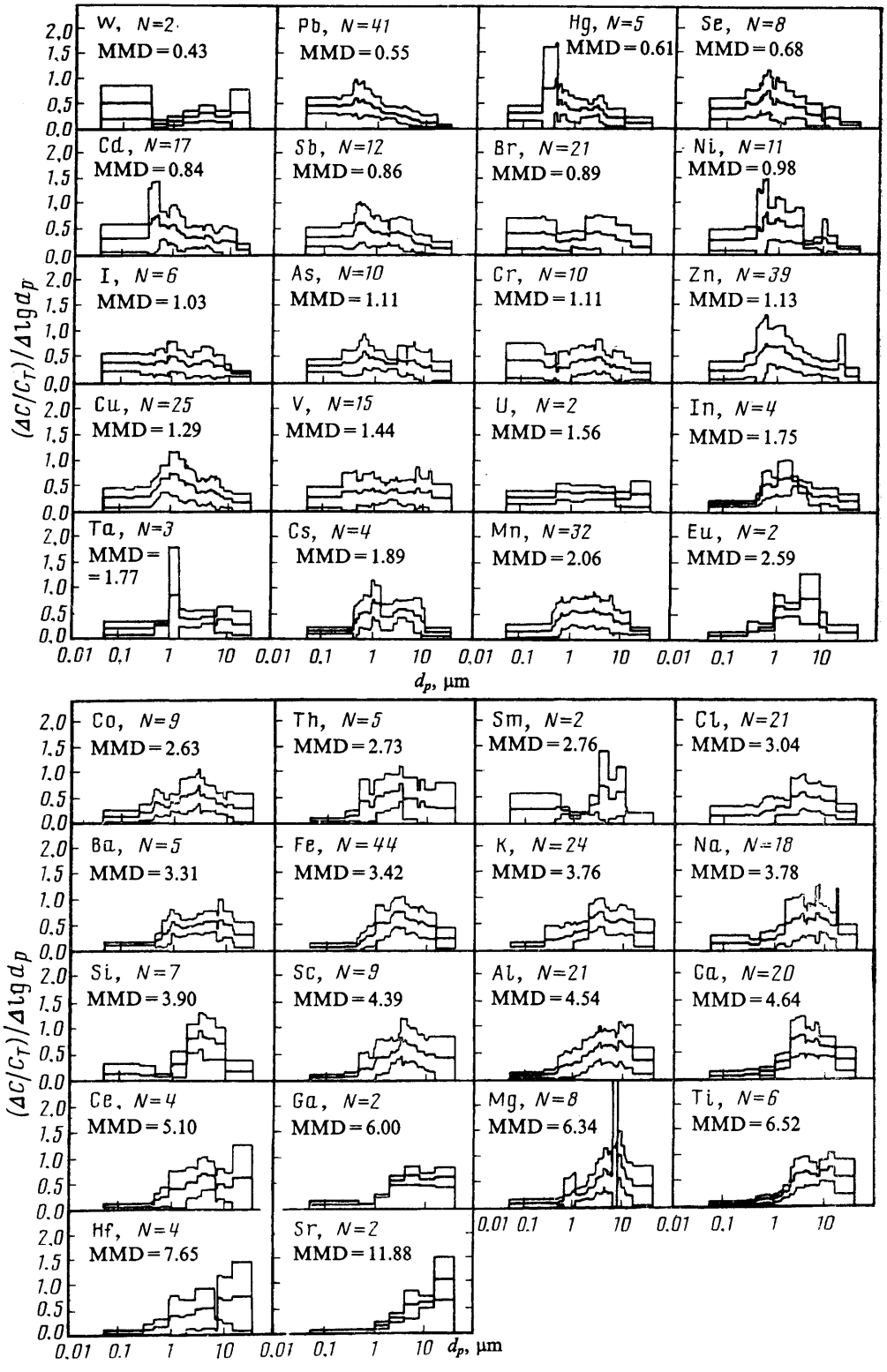


Figure 6. Normalised differential mass distributions for 38 microelements. The data (central line) are presented together with standard deviations (upper and lower lines). *N*—number of averaged distributions; MMD—median mass diameter d_p.[43] The distributions are arranged in order of increasing MMD.

radioactivity of ^{214}Pb and ^{212}Pb in the aerosol from agricultural and industrial regions show[41] that the maximum radioactivity is observed for particles whose radius is about 0.2 μm and in the case of the radioactivity of ^{7}Be and ^{210}Po for particles with a radius of 0.4 μm (Fig. 5). The size distribution for the isotopes formed during tests of nuclear weapons has not been analysed.

Biological aerosols have so far been rarely differentiated as a separate type. Such aerosols contain not only pollen, spores, and bacteria (living organisms) but also ground up pieces of plants and insects and in general decomposing organic material. Pollen, spores, and bacteria are very often considered separately from other types of aerosol. These particles are fairly monodisperse. The distribution of the biological aerosols with respect to the sizes of particles having a radius greater than 2 μm has been investigated.[42] It was found that during Summertime the number concentration of the biogenic aerosol in agricultural localities within the Federal Republic of Germany is between 20 and 40% of the total number concentration of the aerosol.

The aerosol particles can carry an electrical charge, forming the so-called large ions. Ideas concerning the electrical properties and the distribution of large ions have not changed significantly compared with the 1930's. However, the study of these "ionic" spectra has in fact led ultimately to the concept of a continuous distribution of particles with respect to size.[9]

Investigators have devoted much attention to the microelemental composition of the atmospheric aerosol. There are numerous data on this question. A fairly detailed review[43] quotes 38 elements as present in their aerosol particles with diameters ranging from 0.05 to 40 μm (Fig. 6). The curves for the distribution of the elements with respect to particle size for aerosols collected at different times (over a period greater than 20 years) and by different methods (each of which has its limitations) agreed fairly well as regards their shape. A small number of distributions have a fairly monodisperse character (for example, for Mg and Ta). (Naturally, only in the case of distributions of this kind does a parameter such as the median mass diameter have a physical significance.) On the other hand, the majority of distributions are polydisperse without a distinct average radius. A typical example here is provided by vanadium. Elements for which less than two distributions are known from the literature are not considered in this list. Microelements such as sulphur are also not included, since they could not be determined by the methods employed (neutron activation analysis, X-ray fluorescence, and proton-induced X-ray emission).

V. Horizontal or geographical distribution of the aerosol

The distribution of the global atmospheric aerosol is subject to appreciable fluctuations in view of its short lifetime. This is particularly notable if one considers the two hemispheres. Thus the time required for the exchange of the atmospheric aerosol between the Southern and Northern Hemispheres is of the order of two years. This exchange time is significantly greater than the lifetime of the aerosol and it is therefore unlikely that any aerosol particles from the Northern Hemisphere could enter the Southern Hemisphere. In order to achieve a reliable statistical study of the expected fluctuations in the aerosol characteristics, an enormous number of measurements are necessary. Clearly, such a large volume of information can be obtained only for certain parameters of the aerosol and for the majority of the parameters data concerning the distribution of the particles with respect to size are lacking.

An approximate idea about the distribution of the aerosol in space can be obtained by considering the data in Tables 1 and 2 where the characteristics of the polar and background continental aerosols, the dust sandstorm aerosol, as well as the aerosols from agricultural and industrial regions are presented. Fig. 7 illustrates the overall number, surface, and volume concentrations of particles in order of increasing number concentrations. In most cases, these quantities are correlated. The dust sandstorm aerosol is an exception. Whereas the surface area and volume of this aerosol are much higher, the number concentration remains comparable to the number concentration of the particles of the background continental aerosol. The total areas and volumes of the agricultural and background continental aerosols are comparable, but the number concentration of the particles in the agricultural aerosol is significantly higher, which indicates a greater strength of the gas–particle transformation.

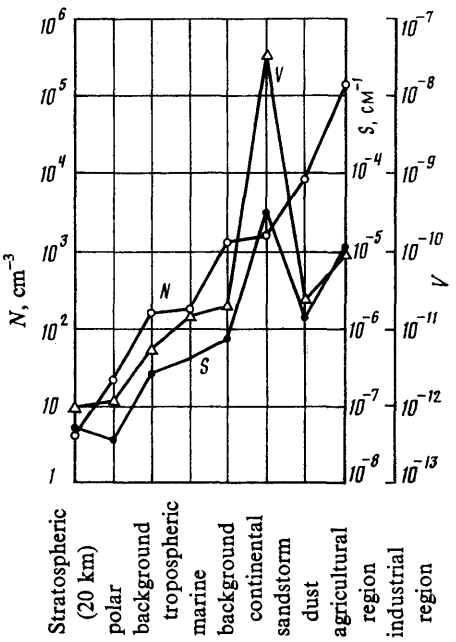


Figure 7. Total number (N), surface (S), and volume concentrations for the types of aerosol presented in Table 1 (in order of increasing number concentration of the particles).[3]

When the detailed distribution in space is described in the literature, the treatment is confined to homogeneous localities. Apparently the authors feel more confident when selecting data on the assumption that a fairly homogeneous source of the aerosol is present or a fairly homogeneous mechanism for the removal of the particles from the atmosphere operates.

Fig. 8 presents a map of the distribution of the number concentration of the condensation nuclei (Aitken particles) over the ocean. Evidently, in the central regions of the oceans the

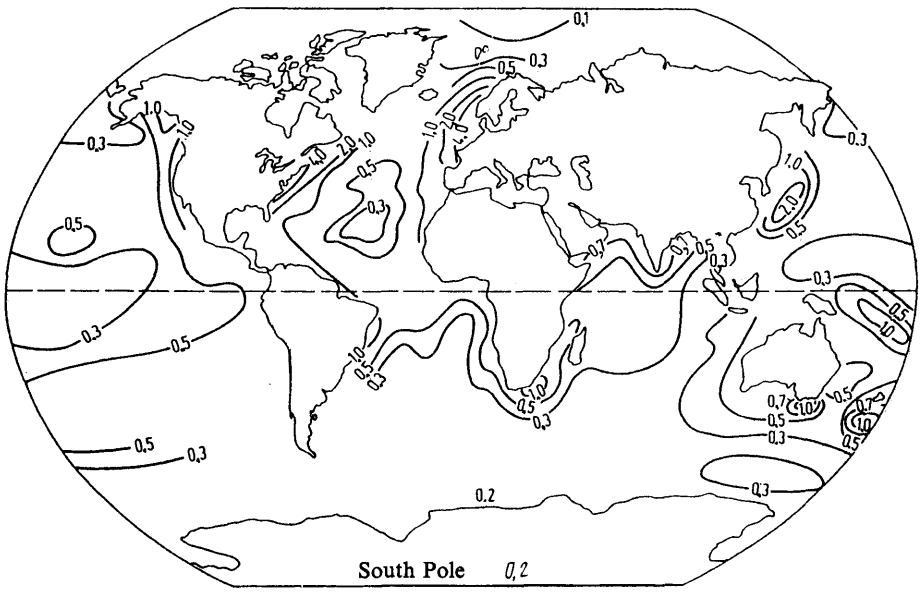


Figure 8. Map of the distribution of the average concentration of Aitken nuclei (10^3 cm^{-3}) over the surface of the oceans. Data taken from Jaenicke[3] and Podzimek.[44]

concentration of Aitken particles diminishes. This does not conflict with the seasonal maps published in 1966 for particles of sea salt greater than 0.7 μm.[45] The data were calculated on the basis of a climatic atlas of the world[46] using the relation between the mass concentration of these particles, transported by air, and the percentage frequency of the repetition of winds with a specified force. One may conclude that the map reflects the storm activity of the oceans. It can be refined by using new data for the relation between the mass concentration of sea salt particles and the wind strength.[3] A similar map can be plotted by using the published[3] relations between the mass concentration of the mineral dust and the wind velocity in the desert. We may note that the mass concentration of this dust is approximately 10 times greater than the mass concentration of the oceanic aerosol.

A minimum concentration of the aerosol (Fig. 8) is observed in polar regions. This was in fact to be expected, since the principal aerosol sources are absent here. The concentration of the particles increases when approaching the continents, which are active sources of aerosol. A more detailed map has been published for the Arctic part of Europe,[13] showing that the average concentration of the aerosol is below 300 particles cm^{-3}. Near populated areas and territories, subject to anthropogenic activity, the concentration of aerosol is higher.

In many industrial countries, the parameters of atmospheric pollution, including certain characteristics of the atmospheric aerosol, are measured at meteorological stations. This is done mainly in order to avoid the formation of smog, acid rain, and the destruction of forests. Thus, using a network of meteorological stations, the extinction coefficients have been calculated for the surface air in the eastern part of the USA.‡ As early as 1948, an increase in the number of sites where the surface air has high extinction coefficients was observed. During the period 1970–1974, a high value of this coefficient was noted for regions along the Great Lakes and the urban conurbation on the Atlantic littoral. As one moves West, the extinction coefficient falls.

‡R B Husar, D E Patterson, J M Holloway 4th Symposium on Atmospheric Turbulence, Diffusion and Air Pollution. Reno, NV, USA, 1979.

Table 3. Concentrations (μg m^{-3}) of organic substances of different classes in certain types of aerosol at the standard pressure and temperature.[3]

Substance	Marine aerosol		Background continental aerosol	Aerosol from agricultural regions	Aerosol from industrial regions
	Northern Hemisphere	Southern Hemisphere			
Total organic carbon	0.60	0.36	1.1	2.5	
Aliphatic neutral substances	0.29		0.26	0.53	6.8
Aromatic neutral substances	0.25	0.25	0.12	0.26	3.1
Polar neutral compounds	0.16		0.20	0.35	4.9
Acids	0.29	0.18	0.20	0.70	10
Bases	0.08	0.09	0.03	0.20	2.0
Total hydrocarbons ($>C_{15}$)	0.014		0.14		0.27
Normal aliphatic acids and ketones	0.09		0.27		0.27
Normal aliphatic alcohols			0.83		1.64
Formaldehyde	0.0054			0.041	

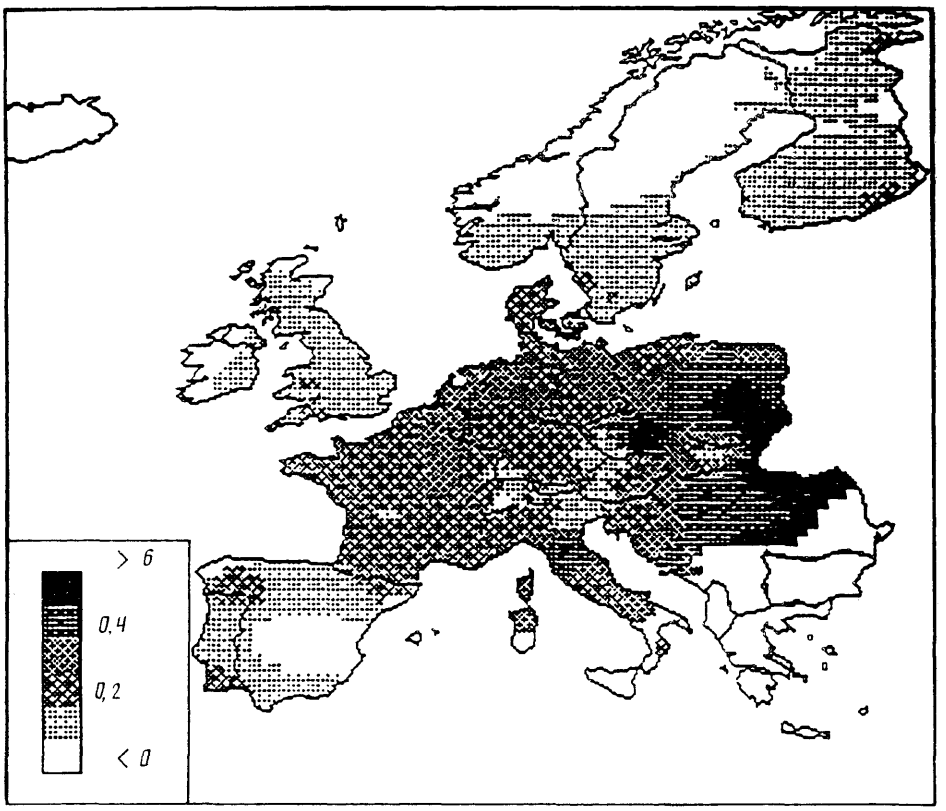


Figure 9. Map of the distribution of SO_4^{2-} in aerosols (in μg S m^{-3}) based on measurements within the framework of the combined program on the monitoring and evaluation of the large scale transport of pollutants in the atmosphere (EMEP).[47]

A map of the distribution of sulphates as aerosol over certain regions of Europe was published recently.[47] Fig. 9 shows that the concentration of sulphate is low over the sea littoral of northern Europe and the Mediterranean and is high in the eastern part of the continent.

The study of organic compounds in the aerosol is extremely difficult because of the great variety of these substances and their low concentration. Concentration data are available for only 95 compounds out of more than 1000 present in the marine and background continental aerosols.[3] Table 3 presents the concentrations of organic compounds of different classes in the marine aerosol of the Southern and Northern Hemispheres, the background continental aerosols and in the aerosols of the agricultural and industrial regions. The contents of organic substances in the marine aerosol and the aerosol from industrial regions differ by ~2 orders of magnitude.

VI. Vertical distribution of the aerosol

The types of distributions presented above show approximately, in relation to the background tropospheric and stratospheric aerosols, how the distribution of the particles with respect to size can vary with height.

As regards the distribution of the aerosol with respect to size as a function of the height over sea level, such studies have only just begun. Measurements performed over the Pacific Ocean in the course of the GAMETAG experiment[48] are most probably subject to instrumental errors. In the continental part of the USA, impactors for the study of the size distribution of the aerosol (in the range of particle sizes 0.2–3 μm) were inserted into the troposphere at heights up to 9 km. Fig. 10 presents data for the size distribution of the particles. At low altitudes, the concentration of the aerosol diminishes with increase in the particle radius. In the free troposphere, this decrease in concentration is modulated. This fact indicates a change in the form of the size distribution of the aerosol from that typical for the background continental aerosol to one characteristic of the background tropospheric aerosol.

The vertical distribution of the sea salt aerosol has been studied up to the height of the cloud cover (~1000 m).[50] The mass concentration of the salt diminishes from 2 mg m^{-3} in the layer where large spray particles are present (at a height of 0.1 m) to 1 μg m^{-3} at an altitude of 2000 m. A similar inversion of the salt layer under the cloud layer is a fairly common phenomenon. Above the cloud layer, the concentration of sea salt is so low that it is not susceptible to measurement.

A cross-section of the spatial distribution of the mass concentration at an altitude of 5.5 km from the South Pole to the North Pole was constructed for the tropospheric background aerosol.[3] At this altitude, the average concentration of the aerosol in the Southern Hemisphere is less than 0.3 μg m^{-3}. The maximum in the mass concentration is observed between 40 °S and 50 °S and amounts to 2.2 μg m^{-3}. In Arctic regions, the concentration of the aerosol is 1 μg m^{-3}.

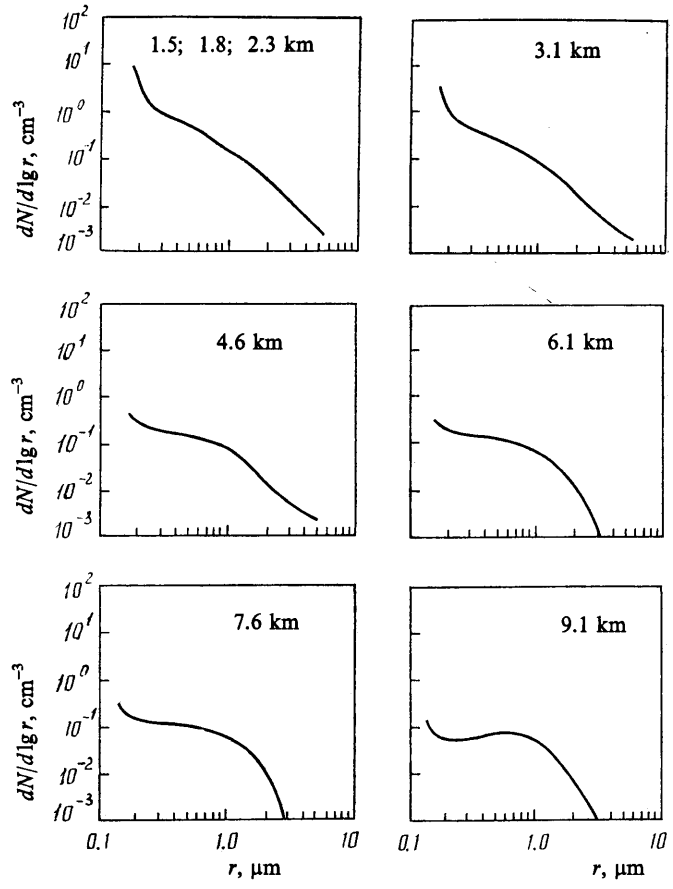


Figure 10. Average distribution functions for the number concentration of the aerosol at different heights in the continental part of the USA.[49]

A vertical section through the distribution of the number concentration of the aerosol was constructed in 1972–1973[51] and is shown in Fig. 11. One can easily note the decrease in the concentration of particles on moving from the surface of the Earth to the tropopause. In the stratosphere there is a layer with the distinctly enhanced concentration of particles. This so called "Junge layer" or the "sulphate layer" is located approximately 7 km over the tropopause. The concentration of the aerosol in this layer is a maximum over the equator. During periods of high volcanic activity, particles enter the Junge layer directly from the volcanic eruptions and this layer then undergoes marked changes. These changes were manifested particularly strikingly in a lidar study of the vertical distribution of the background scattering coefficient before and after the eruption of the El Chichon volcano (Mexico) in 1982[52] (Fig. 12).

The Junge layer continues to exist also during the period of low volcanic activity. During this time, it consists mainly of sulphuric acid particles, formed mainly as a result of the gas–particle transformation from dimethyl sulphide, which is the gaseous tropospheric precursor of H_2SO_4.[53] This layer has been thoroughly investigated from satellites and by terrestrial lidars, because it influences the transparency of the atmosphere, which is an important factor for the Earth's climate. Polar stratospheric clouds are part of this layer, influencing the geochemical ozone cycle and determining the dynamics of the development of the Antarctic ozone hole.

The vertical distribution of the mass concentration of the aerosol over deserts is naturally different. As already reported, mineral particles are distributed uniformly up to a height of ~6 km and this is probably characteristic of all deserts.

The vertical distribution of the number concentration of Aitken particles differs significantly from that for the majority of other particles. In it, there is no distinct layer in the stratosphere. The concentration of these particles diminishes with increased altitude and assumes a value of 1–10 particles cm^{-3} in the middle stratosphere.[54] The number concentration of condensation nuclei

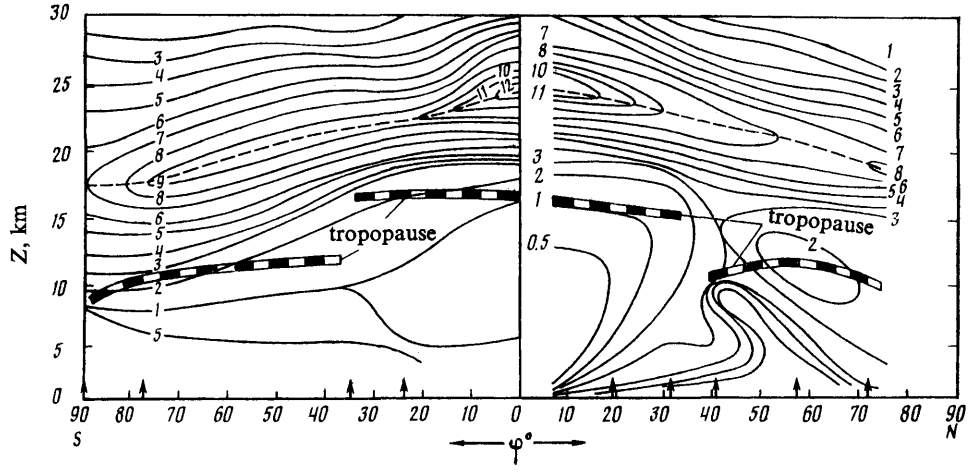


Figure 11. Number concentration contour lines for aerosol particles larger than 0.15 μm in radius as a function of the geographical latitude and height above sea level. The data refer to the period 1972–1973.[51]

increases only for a short time after volcanic eruptions.[55] Fig. 13 illustrates typical examples of the distribution of these particles.

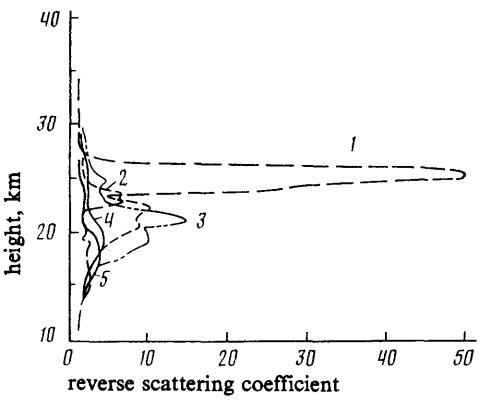


Figure 12. Vertical distribution of the coefficient of backward scattering obtained in terrestrial lidar studies in the region 37 °N, 76 °W. Probing laser wavelength—0.6943 μm:[52] (*1*) data for 1st July 1982; (*2*) data for 27th October 1982; (*3*) data for 3rd January 1983; (*4*) data for 30th June 1983; (*5*) data for 8th November 1983.

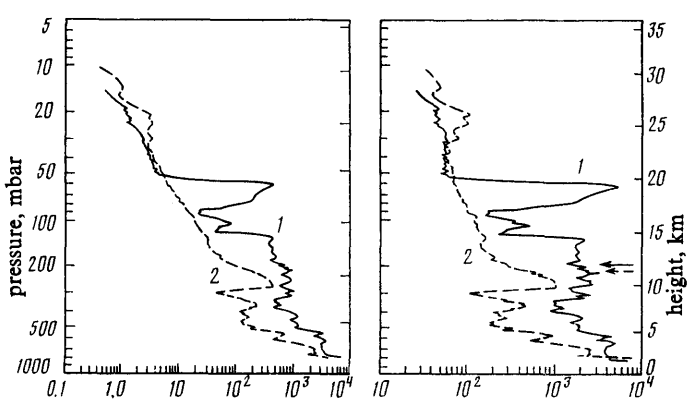


Figure 13. Vertical distribution of the concentration of condensation nuclei before and after the eruption of the volcano on the island of St. Helena.[55] The arrows indicate the position of the tropopause: (*1*) data for 20th June 1980; (*2*) data for 12th November 1979.

VII. Conclusion

In the last decade, our knowledge about the global distribution of the atmospheric aerosol increased immeasurably. Nevertheless, it is seen from the present review that we still know too little about it. A complete global map cannot be drawn for any one individual parameter of the aerosol and we are therefore forced to make do with incomplete information. This situation is unsatisfactory at a time when problems of vital importance for humanity, such as the changes in the climate and in the air require a detailed understanding of the state of the atmosphere, including the aerosol. The global computer meterological and climatic models suffer particularly from the deficiency of information about the distribution of the aerosol. Hitherto the question of the influence of the aerosol on the climate has given rise to controversy. Nevertheless, it may be that the facts presented in this review will inspire investigators to fill the existing gaps.

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