NATURAL AEROSOLS*

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INTRODUCTION

In their pioneering work, Schmauss and Wigand[1] stated that the atmosphere only rarely is a molecular-disperse system. Only after extended rainfalls and snowfalls can it come close to that condition. Usually it is a colloid-disperse system, having gas as the continuous phase, with solid and liquid material in finest distributions dissolved or suspended. This is true not only for aerosols influenced by man, but for the natural aerosol in general. The aerosol load of the atmosphere usually is smaller by one or two orders of magnitude than the load of atmospheric trace gases. However, the effects of the aerosol—such as its contribution to the cloud formation and to the budget of the atmospheric radiation—cannot be overlooked and are crucial for life on earth. To know the atmospheric aerosol, a large number of points must be studied:

1. Sources of the aerosol;
2. Aging while airborne;
3. Residence time;
4. Sinks of the aerosol;
5. Global distribution;
6. Properties and composition of the aerosol;
7. Effects of the aerosol and secular variations.

Our knowledge about the natural aerosol is quite limited. Studies of the aerosol began some 50 years ago but have been done in a systematic way only since 20 years. However, measurements always carry the possibility of being influenced by anthropogenic aerosols, and we will see later to what extent it is possible to study the natural aerosol not influenced by man.

SOURCES

There are two major processes by which aerosol particles and aerosol mass are formed. One is disintegration and dispersion of bulk material, such as the weathering of crustal material, the disintegration and dispersion of biomass, and the production and dispersion of sea-salt droplets. The second process is the modification of airborne substances in such a way that new particle mass is formed. Such processes are gas-to-particle conversion by condensable gaseous species and the evaporation of cloud elements and rain droplets to leave behind aerosol particles. These different types of

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sources lead to a certain chemical composition of the aerosol and determine the size distribution; the disintegration of bulk material mainly forms particles larger than 0.1 μm in radius, while the airborne modification usually results in particles smaller than 1 μm in radius.

Depending on their linear dimensions, the sources can be subdivided into several categories. The surfaces of the oceans and of the continents act as *area sources*. Volcanoes and isolated meteorological events, like thunderstorms and low pressure systems, act as *point sources*. The gas-to-particle conversion and the droplet-to-particle conversion must be considered as *volume sources*.

Still the most complete study about the strength of natural aerosol sources is given in the SMIC Report[2] and results in an estimate of some 2000×10^6 tons per year released by nature, as compared to some 300×10^6 tons per year produced by anthropogenic activities. This estimate, however, is somewhat problematic because it
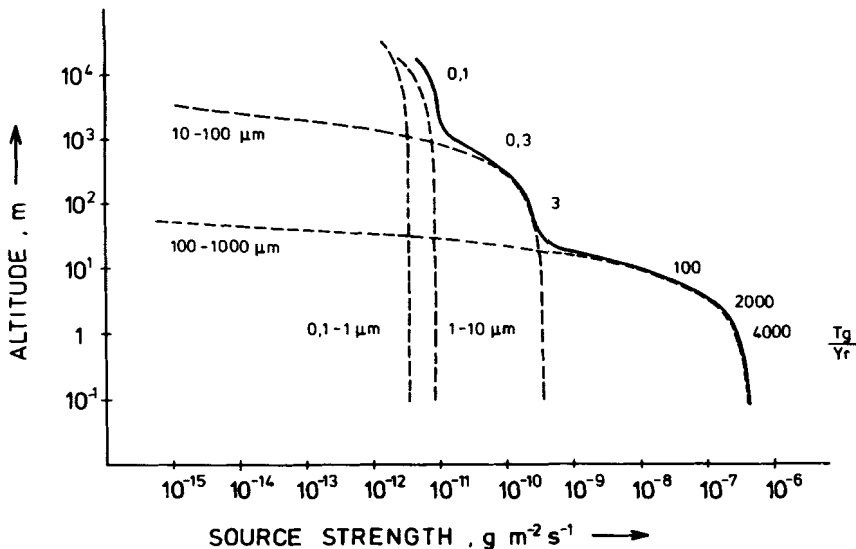

FIGURE 1. The apparent source strength for aerosol particles, as a function of altitude. For various particle size ranges (given in μm) this was calculated on the basis of an average aerosol mass and the limited residence time of particles (FIGURE 2). At various altitudes, the total source strength is given in Tg/yr. The apparent source strength varies rapidly with altitude, and only above some 10 m is the source strength less dependent with altitude.

is limited to particles smaller than 20 μm in radius. This can be demonstrated in the following example. The total particulate load of the atmosphere in natural aerosol is roughly 20 μg m^{-3}.[3] As we have seen above, the particles larger than 0.1 μm mainly come from an area source, the surface of the oceans and the continents. From this source, the particles must be transported horizontally and vertically (a rate of 2 cm s^{-1} is assumed for the purpose of this calculation). As we will see later, the particles have a certain residence time in the atmosphere. All this leads to FIGURE 1, where the apparent strength of the area source is a function of the altitude. It is obvious that the

largest particles cannot be transported to higher altitudes, while the smallest particles under consideration are more or less evenly distributed with altitude. The accuracy of the figures given in FIGURE 1 is of minor importance. However, I would like to stress the fact that the apparent strength of the assumed area source at 10 m altitude is roughly two orders of magnitude smaller than at 1 m altitude. This might explain some variations in the source-strength data given in Reference 2.

Following Reference 2, the volume source from gas-to-particle conversion is of equal importance to the area sources mentioned. However, no data are given about the strength of the droplet-to-particle conversion as a particle source. The formation of cloud droplets around an aerosol particle acts as sink on the one side. On the other side, such a droplet takes up gaseous substances from the atmosphere and will evaporate in 9 out of 10 cases.[4] So it acts as a volume source for particles. This source certainly has a maximum in the cloud layer, but no estimates are available.

AGING

After being released as particles from the source, the aerosol is acted upon by a number of different processes, which are summarized as *aging*. The aerosol is transported in the atmosphere through meteorological events, and mixed with other aerosols or clean air; this is called dilution. The particles might collide with each other due to thermal diffusion (coagulation), become center condensation nuclei of cloud droplets (rain-out), be collected by falling rain droplets (wash-out), or sediment out by their own vertical velocity or by impact on ground obstacles (dry removal). The particles might gain mass through condensation of gaseous species or might lose mass due to evaporation.

Only some of these processes are known to a certain extent. Some of them occur only in the lower layers of the troposphere with an abundance of clouds, rain, and ground obstacles. Coagulation is most rapid in highly concentrated aerosols. The most rapid transport and the most effective dilution certainly take place if an aerosol penetrates into the higher layers of the atmosphere, where the air masses are transported rapidly without the friction from the earth's surface. Major effects of this aging are the formation of the aerosol size distribution with its unique features, the changing of the chemical composition of the aerosol—which might be quite different from that of the source, and the formation of rather uniform aerosol bodies, which are summarized as continental, maritime, and background aerosols.

RESIDENCE TIME

Most atmospheric processes described under "aging" affect the time during which an aerosol particle remains airborne. This time is called *residence time*. As for gaseous species, one single residence time was estimated for the aerosol[4,5] in the past. This, however, describes only that fraction of the aerosol most sensitive to the process used for estimation. In addition, it is a figure that averages over the very short residence times of coarse particles (with high deposition velocities) and the much longer residence times of smaller particles (with practically no deposition velocities). For aerosols with a small particle load, the residence time was calculated as a function of

particle size.[6] This resulted in the analytical expression:

$$\frac{1}{\tau} = \frac{1}{C_F}\left(\frac{r}{R}\right)^2 + \frac{1}{C_D}\left(\frac{r}{R}\right)^{-2} + \frac{1}{\tau_{wet}} \tag{1}$$

which is shown in FIGURE 2 with: τ = residence time, in s; r = particle radius, in μm; $R = 0.3$ μm, standard radius; C_F, C_D = constants, in s; and τ_{wet} = time limit, in s, mainly controlled by wet removal processes.

Equation 1 is the mathematical description of the compilation of experimental findings and estimates about the residence times of atmospheric aerosols. The residence time in this sense means the time a particle is suspended as an entity. After this time, its mass is removed from the atmosphere or incorporated into another particle. Except for τ_{wet}, which is somewhat coupled to the wet removal processes, the constants in Equation 1 cannot be interpreted in terms of physical meaning. The constant C_F, coupled to that branch of the curve where sedimentation is of importance, seems to be rather independent from the particle concentration. So it applies in background aerosols as well as in continental aerosols. It is, however, coupled to the height of the homogeneous dust layer, which was estimated to be 1.5 km. The constant C_D, in the branch mainly controlled by coagulation, depends on the particle concentration. As can be seen from Reference 7, the residence time—defined as the ratio of the particle concentration over the change of concentration with time due to
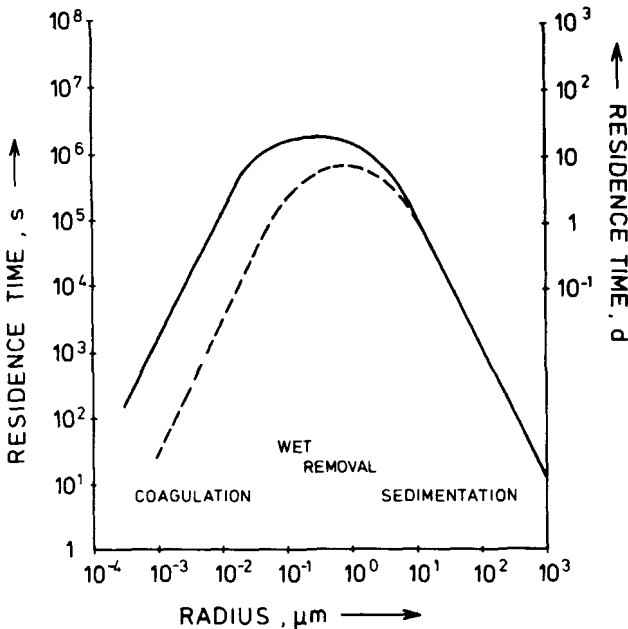

FIGURE 2. The residence time of aerosol particles, based on Equation 1. Two different cases have been calculated: —background aerosol, N = 300 cm^{-3}, τ_{wet} = 21 d; ---continental aerosol, N = 15000 cm^{-3}, τ_{wet} = 8 d. In both cases, the height of the homogeneous aerosol layer was assumed to be 1.5 km. This only has effects on the sedimentation branch of the curve.

TABLE 1

DISPLACEMENT OF AEROSOLS*

Particle Radius (μm)	Horizontal Distance (km)	Vertical Elevation (m)
0.001	8	20
0.01	800	2,000
0.1	8000	20,000
1	8000	20,000
10	800	2,000
100	8	20

*For the residence times given in FIGURE 2, the horizontal and vertical displacement of an aerosol particle is calculated. The actual numbers are of minor importance; only it should be shown that the particles in the range 0.1–1 μm radius can be transported intercontinentally and be distributed in the whole troposphere.

coagulation—is inversely proportional to the particle concentration. Thus, C_D depends on the concentration of the aerosol. FIGURE 2 indicates the ranges of residence times we have to expect in the natural aerosol. The residence time is shortest for the smallest and the largest particles. The particles in the range 0.1–1 μm have the longest residence times. Based on these residence times, FIGURE 1 was calculated.

With these residence times, it is possible to calculate how far particles from a point source—i.e., anthropogenic activities—might be transported. Under the assumption of a horizontal transport velocity of 8 m s^{-1} and a vertical uplift of 2 cm s^{-1}, which seems to be reasonable for atmospheric conditions, TABLE 1 shows (the actual figures are rather unimportant) that particles in the range 0.1–1 μm can be spread over the vertical extension of the troposphere and transported from one continent to the other. This range certainly is most easily influenced by man on a worldwide basis.

A good example of this intercontinental transport is the desert dust. The transport of desert particles from the Sahara can be followed across the Atlantic Ocean to the Lesser Antilles. This transport occurs only for the particles in the size range 0.1–1 μm.[37,24] Smaller and larger particles are transported only in the mesoscale; at selected locations, these particles certainly can be studied free of anthropogenic influences.

It should be mentioned, however, that some gaseous precursors of the smallest particles might have residence times long enough for worldwide (or hemispheric) mixing and distribution. Particles produced from these precursors are, of course, not free of anthropogenic influences if the precursors are released by man. To a certain extent, this is the case for sulfur and might apply to the transport of sulfurous aerosol to Scandinavia from central Europe.[27]

SINKS

The processes responsible for aging and residence times of aerosol particles are sink processes. These sinks—similar to the sources—act as *volume sinks* (so the formation of clouds) and *area sinks* (so the removal by dry deposition). In addition, one could consider the transport of aerosol particles into other volumes of the atmosphere as a sink, too. In principle, such a process could be the transport of aerosol

particles into the stratosphere. FIGURE 1 would favor such a sink; however, FIGURE 1 does not take into account the circulation of the atmosphere. And indeed, it is assumed at present[8] that the stratospheric dust layer is more or less produced *in situ* by gas-to-particle conversion of SO_2, rather than by transport of particles. In this sense, the removal of this stratospheric aerosol with a residence time of roughly 2–3 years must be considered as a source of tropospheric aerosol particles. Because up to now only gas-to-particle conversion has been reported in the atmosphere, and never particle-to-gas conversion (an exception is water vapor), more particulate mass must be removed from the atmosphere than was injected. It has been estimated that roughly 50% is removed in the above-mentioned area sink, while 50% is collected by wet removal.[36]

GLOBAL DISTRIBUTION

The interaction of source function, aging, residence time, and sink function in connection with atmospheric transport processes leads to a global distribution of the atmospheric aerosol. It is now rather well established that four different aerosol bodies exist in the atmosphere. One is the *stratospheric aerosol*. As explained earlier, this aerosol is separated from those in the troposphere by the tropopause. The meteorological conditions at the tropopause permit only a small leakage of particles. The intrusion of gaseous precursors of aerosol particles into the stratosphere was discussed earlier.

Based on measurements of the concentration of condensation nuclei (a rather early, but comprehensive, study was published in 1939[9]), *continental aerosols* and those of the free atmosphere (this was later[10] called *background aerosol*) were distinguished. For the condensation nuclei, it means that the background aerosol can be observed at the surface of the ocean, because sea spray particles are hardly present in this size range.[11] From chemical evidence,[12,13] it became obvious that the *maritime aerosol* should be distinguished from the background aerosol. It was shown convincingly[12,14] that sea salt is practically nonexistent above the cloud layer. This differentiation of atmospheric aerosols then leads to FIGURE 3. For the large particles and the condensation nuclei, the model vertical distribution of the particle concentration is given. The aerosols mentioned above, maritime, continental, background, and stratospheric, can be seen. While only three aerosols can be seen for the condensation nuclei, the large particles show four aerosols.

In studies of the atmosphere, this concept of aerosol bodies has proven very helpful. Of course, the borders are not very sharp, and often *transition aerosols*[15] can be observed. In these four aerosols, the chemical composition and the size distributions are quite different and will be discussed next.

PROPERTIES AND COMPOSITION

The properties and the chemical composition of the natural aerosols are produced by the combined effects of sources, sinks, and aging of the aerosols. At present, it is not known how the effects act together to produce the observed properties.

The size distribution of the aerosol most often has attracted researchers—as

Reference 16 shows—to investigate how it is generated. The "self-preserving" size distribution[19] is one of these cases but is only applicable if sufficient time is given to the aerosol for evolution from its source functions, which rarely is the case in the· atmosphere. The size distribution of the aerosols is not yet fully understood. The most recent compilation of measurements,[6] in the form of model distributions, shows for all aerosols the typical concentration decrease with increasing particle size for radii larger than 0.1 μm. Smaller particles show a more or less constant concentration, with some

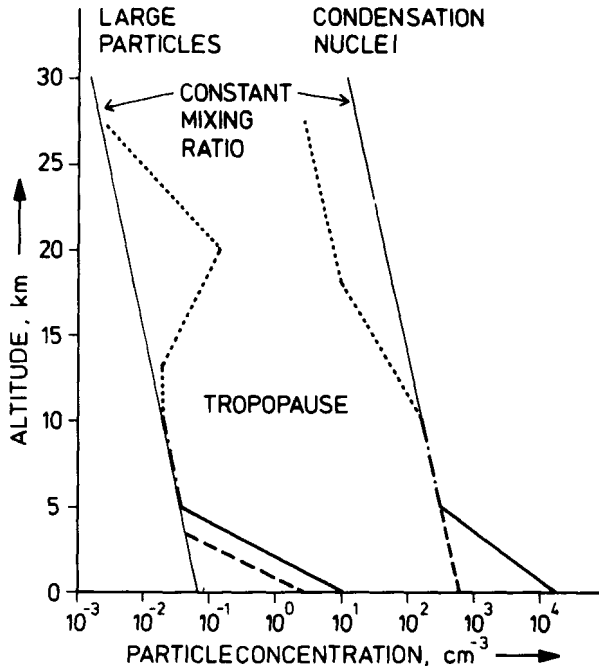FIGURE 3. Model of the vertical distribution of the natural aerosol. The particle concentration is given for condensation nuclei ($r < 0.1$ μm, determining the number concentration) and for large particles (0.1 μm $< r < 1$ μm, proportional to the aerosol mass). Four aerosol bodies can be distinguished: — continental; – – maritime; –·– background; and ···· stratospheric aerosols. For the condensation nuclei, the background and maritime aerosols are equal.

distinct maxima and minima. The smallest particles that are observed ($\sim 10^{-3}$ μm) are constantly present in the aerosol. That only can be understood if a continuous aerosol source is effective.[20]

FIGURE 4 shows model size distributions for three aerosols: background, maritime, and (remote) continental aerosols. In contrast to earlier presentations,[4] these distributions are given as a combination of number and volume size distributions. This is done to emphasize those parts of the distributions that are suppressed if only the number or volume size distribution is presented. In a number size distribution of the maritime aerosol, usually the sea spray is suppressed, because it is limited to particles larger
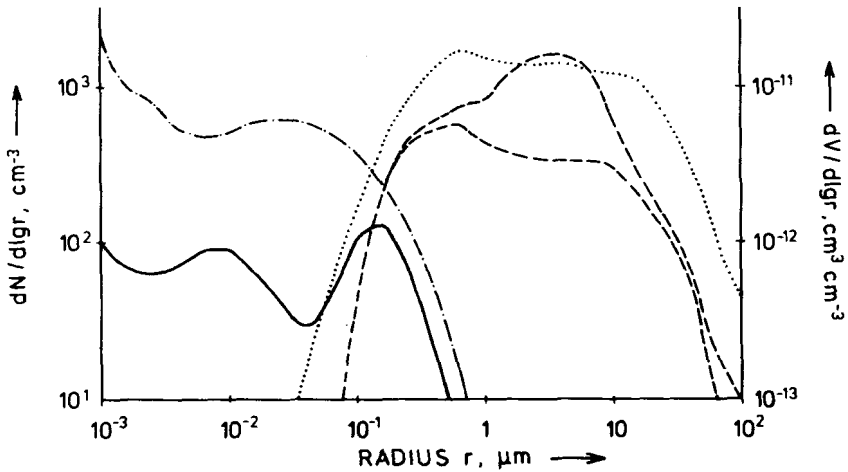
FIGURE 4. Model aerosol size distributions given as number (dN/dlog r) or volume (dV/dlog r) distribution. Number distribution for: background (maritime) —; and remote continental —·—, Volume distribution for: background, lower branch ———; maritime, upper branch ———; and remote continental ········ aerosols.

than 0.1 μm in radius and, thus, to that branch of the distribution with the steep concentration decrease.

The author is convinced that these difficulties in presentation of the size distributions have caused a number of empty controversies. It is certainly because of the work of Whitby[18] that more emphasis is placed, at present, on volume and other distributions.

FIGURE 4 shows clearly that background and maritime aerosols differ only in the volume distribution for particles larger than 0.1 μm. Between 1 and 10 μm, maritime and remote continental aerosols differ only fractionally in volume distribution. The chemical compositions of these particles, however, are different.

FIGURE 4 indicates how problematic it is to describe those distributions with a power function, as it was done in Junge's[17] pioneering work. This kind of mathematical expression was used for a long time and served well for a number of applications. Its limitations, however, were understood rather early.

Currently, the size distributions are often described with normal distributions.[21,22] FIGURE 5 shows how well the model distribution for background aerosol (FIGURE 4) can be described with normal distributions. Shown are number, surface, and volume distribution. The mathematical expression is:

$$n^* = \frac{dN}{d \log r}$$

$$= n_1 \, f \, (r, R_1, S_1) + \frac{1}{4\pi r^2} \, n_2 \, f \, (r, R_2, S_2) + \frac{1}{\frac{4}{3}\pi r^3} \, n_3 \, f \, (r, R_3, S_3) \qquad (2)$$

$$f(r, R_i, S_i) = \frac{1}{S_i \sqrt{2\pi}} \exp \left\{ -\frac{[\log (r/R_i)]^2}{2S_i^2} \right\}$$

This expression was selected because, at present, there is no reason known why the processes responsible for the shape of the aerosol size distribution tend to produce normal distributions in one of the moments: number, surface, or volume only. The above expression produces a normal distribution for each of the moments, with some additional influence from the other moments.

In TABLE 2, the parameters of the above expression are listed for the three aerosols mentioned above: background, maritime, and remote continental. These values have been obtained by minimizing:

$$\sum_i \log \frac{n^{*\prime}(r_i)}{n^*(r_i)} = \min$$

with $n^{*\prime}(r_i)$ given the values shown in FIGURE 4. The purpose of the table is to give a parameterized version of the model distributions, for further calculations. The
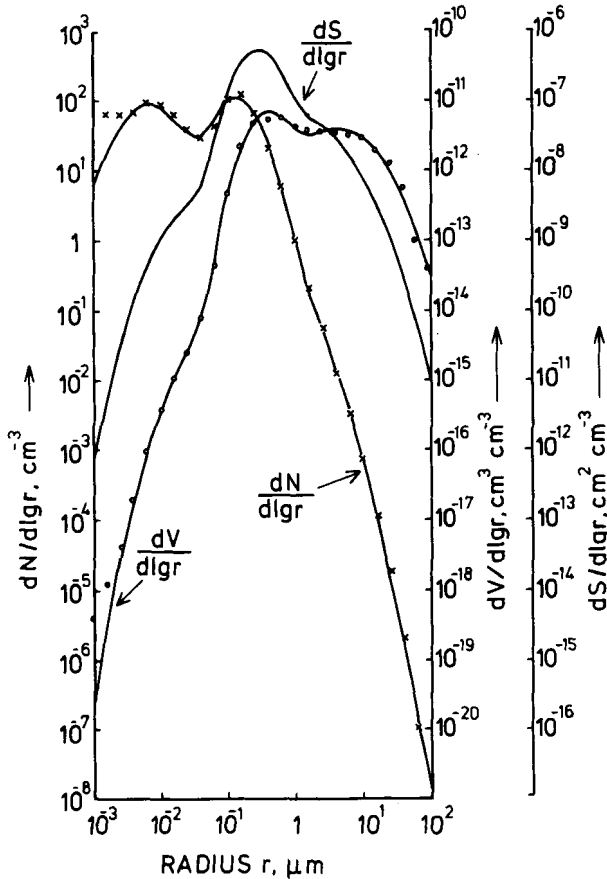

FIGURE 5. For the background aerosol, the model size distribution (FIGURE 4, x – x number, o – o volume) is described by an analytical function (Equation 2). Shown are number (dN/dlog r), surface (dS/dlog r), and volume (dV/dlog r) distribution.

stratospheric aerosol is not included in this table because it is explored only occasionally and undergoes rapid changes due to the nature of its production. The main sources are volcanic eruptions, forming particles mainly by gas-to-particle conversion (see Reference 23). The number size distribution is believed to be normal, with a maximum around 0.3 μm.[16,27]

FIGURES 4 and 5 show clearly that the range 0.1–30 μm is the most important size range for the volume of the aerosol and, consequently, the mass. Any statements about the chemical composition, therefore, must be interpreted in this sense. Continental aerosols consist of roughly 40% water-insoluble substances, mainly minerals[25] and biogenic material.[26] Some 10–20% is ether-soluble organic material.[3] Roughly half of the total organic material is water soluble, and 30% of the total aerosol mass has to be considered as inorganic and water insoluble. The water-soluble fraction mainly consists of inorganic salts with the following ions: $SO_4^=$, NO_3^-, Cl^-, NH_4^+, Ca^{++}, Mg^{++}, Na^+, and K^+. The presence of ammonium sulfate has been postulated, and thus the natural aerosol is coupled to the cycle of natural sulfur.[28] If the background

TABLE 2

THE PARAMETERS FOR THREE AEROSOLS—MARITIME, BACKGROUND, AND REMOTE CONTINENTAL—FOR EQUATION 2

Aerosol	n_1 (cm^{-3})	R_1 (cm)	S_1	n_2 (cm^{-1})	R_2 (cm)	S_2	n_3	R_3 (cm)	S_3
Maritime = Background + Sea Spray	8.20 × 10^1	7.37 × 10^{-7}	3.51 × 10^{-1}	2.74 × 10^{-7}	2.53 × 10^{-5}	2.30 × 10^{-1}	1.57 × 10^{-11}	3.06 × 10^{-4}	4.65 × 10^{-1}
Background	8.70 × 10^1	7.63 × 10^{-7}	3.72 × 10^{-1}	3.71 × 10^{-7}	2.94 × 10^{-5}	2.67 × 10^{-1}	4.51 × 10^{-12}	4.56 × 10^{-4}	4.40 × 10^{-1}
Remote Continental	1.24 × 10^3	1.42 × 10^{-6}	5.70 × 10^{-1}	1.51 × 10^{-1}	1.87 × 10^{-6}	8.36 × 10^{-3}	7.88 × 10^{-12}	1.96 × 10^{-4}	2.77 × 10^{-2}

aerosol is seen as an aged continental aerosol, its chemical composition should be roughly the same.[12] In the maritime aerosol, sea salt is added and, as FIGURE 4 shows, it then makes up the main fraction of the aerosol mass.[29]

Not covered by this survey is the composition of Aitken particles (with radii smaller 0.1 μm). These particles contribute only 5% to the aerosol mass. We now have a good deal of evidence that sulfur and organic compounds are the main constituents of these particles.[30] This is a first indication about the production mechanism for gas-to-particle conversion, which must be effective in the natural aerosols.

An immediate consequence of the chemical composition of the aerosol is its behavior in a humid atmosphere. Water-soluble substances in the aerosol particles adsorb water vapor from the humid atmosphere and form droplets of solution. This process starts at rather low relative humidities[31] and leads to a growth of the aerosol particles. This growth is rather small at low humidities but is important at high humidities. With more water vapor available, these liquid aerosol particles finally form cloud droplets. As an effect of the mixed nature of the aerosol particles with numerous ions, the growth with increasing humidity is rather monotone but smaller than with pure salts.

EFFECTS OF THE NATURAL AEROSOL AND SECULAR VARIATIONS

There are numerous effects of the aerosol, and they have been summarized frequently.[2] It would go beyond the scope of the paper to cover all effects. However, some general remarks seem to be necessary.

Usually the aerosol has no effects through individual particles. Mass and surface of such a particle are much too small to start any macroscopic effect. Any aerosol effect is caused by the particles contained in a certain volume of air. This can be described by:

$$E = E\left(\int_V \int_{r=0}^{r=\infty} e(r)\,w(r)\,n^*(r)\ \mathrm{d}\log r\ \mathrm{d}V\right) \qquad (3)$$

The effect E is a function of the aerosol size distribution $n^*(r)$ weighted with $w(r)$ and $e(r)$ and integrated over all radii and a certain volume V; $e(r) = 1$, if the number of the particles contributes to the effect; or $e(r) = 4\pi r^2$ or $e(r) = (4/3)\pi r^3$, if surface or volume are involved, respectively; $w(r)$ is a weighting function characterizing the effect. Some examples are: mass, $e(r) = \rho(r)$ with $\rho(r) =$ particle bulk density; extinction of radiation, $e(r) = \sigma$ with $\sigma =$ volume extinction coefficient; electrical conductivity of the air, $e(r) = \beta$ with $\beta =$ ion attachment coefficient.

Some effects of the atmospheric aerosol, such as the extinction of sun radiation, are influenced by all particles within a beam of sun radiation. Thus, the effect must be integrated over the whole atmosphere, rather than over a limited volume:

$$E = E\left(\int_{\mathrm{atm}} \int_{r=0}^{r=\infty} e(r)\,w(r)\,n^*(r)\ \mathrm{d}\log r\ \mathrm{d}(\mathrm{atm})\right) \qquad (4)$$

All the functions $e(r)$ and $w(r)$ are weighting functions, and thus only certain size ranges of the aerosol make contributions essential to the effect. As mentioned above, the mass is influenced only by particles of 0.1–30 μm size. This has consequences upon the question of in how much detail an aerosol must be observed or measured in order to understand a certain effect. It is useless to study the Aitken particle concentration if optical effects of the atmospheric aerosol are to be explained.[30]

This discussion clearly shows how carefully any reports about secular trends in the aerosol must be discussed. In 1970, it was reported[32] that the concentration of the atmospheric aerosol has increased since the beginning of the century. This conclusion was made from monitoring of the electrical conductivity of the atmosphere. Electrical conductivity is a function of the number of small ions in the atmosphere, and this is controlled by the presence of aerosol particles. If one calculates the weighting function of this effect of the aerosol, it turns out that only particles with radii 0.02–0.4 μm contribute to the effect. A decrease in conductivity thus only means that, in this range of particle sizes, the concentration was increased. Such an increase in concentration is very likely, because particles in this size range might be transported far over the oceans, as shown earlier. The conclusion, however, that the aerosol in its entirety has changed cannot be supported.

It could be shown[33] that the atmospheric turbidity decreased from the beginning of the century until the mid-1930s. Since turbidity is influenced by particles in the range 0.1–1 μm, a decrease in the concentration of only these particles can be concluded. Indeed, it could be shown[33–35] that the stratospheric aerosol around 0.3 μm declined in concentration due to low volcanic activity.

CONCLUSIONS

The atmospheric natural aerosol is of very complex structure, and it is impossible to describe it with single parameters. If we generalize the aerosol with a small number of aerosol types, some of its properties can be given as first approximations. We still do not fully understand why the natural aerosol is as we observe it.

SUMMARY

The natural aerosol can be described as four different aerosols: stratospheric, background, maritime, and continental aerosol. Two major processes, the dispersion of mechanically disintegrated material and the transformation of airborne precursors, are the sources of the natural aerosol. The freshly formed aerosol is transported, mixed, and aged, and removed from the atmosphere after a certain residence time. This time depends on the particle size and varies between hours and days. This has consequences as to how far a particle can be transported in the atmosphere and makes it possible to estimate the influence of anthropogenic particulate sources upon the natural aerosol.

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