

On the removal time of aerosol particles from the atmosphere by precipitation scavenging

By HENNING RODHE, *Institute of Meteorology, University of Stockholm*¹,
and JAN GRANDELL, *Institute of Mathematical Statistics, University of Stockholm*

ABSTRACT

A statistical investigation is presented on the distribution of dry and precipitation periods at a Swedish station. It is shown that the expected length of time from an arbitrary moment to the onset of the next precipitation period is about 90 and 35 hours for summer and winter conditions respectively. Based on the results of these investigations we have derived expressions for the expected length of life of an aerosol particle in the lower layers of the troposphere. Estimates of effective precipitation scavenging coefficients give expected lengths of life of the order 100–300 hours in summer and 35–80 hours in winter. The connection between the distribution of the length of life of particles in the atmosphere and the distribution of particles on the ground away from a source, is discussed and the importance of studying such age distributions is emphasized.

1. Introduction

Precipitation scavenging is a very important process for removing aerosols, and also some gases, from the troposphere. It is therefore of interest to study how this process affects the age distributions of tropospheric aerosols and thereby also the distances over which they are transported before being deposited on the ground.

The scavenging process is often separated into *rainout*—the incorporation of the contaminant into cloud droplets *within the cloud*— and *washout*—the removal by the falling precipitation *below the cloud*. We shall not discuss in any detail the physical processes that determine the precipitation scavenging, but only make some rather schematic assumptions about their effectiveness. Our aim is rather to combine such assumptions concerning the scavenging processes with precipitation statistics and try to arrive at some conclusions regarding the shape of the frequency distribution of the total length of time spent by individual particles in the troposphere. This distribution function is obviously closely related to the distribution of the emitted substance on the ground away from the source. Information about this distribution is of great interest in

connection with air pollution problems and, more generally, in the study of circulation of chemical substances in the atmosphere.

From such an age distribution function it will also be possible to derive an expected average length of life of tropospheric aerosols. Several estimates have been made of the turnover time of tropospheric aerosols, ranging from a few days to a few weeks. Some of these estimates have been based on a study of the distribution of radioactive particles (e.g. Machta et al., 1970). Junge & Gustafson (1957) compared the precipitation amounts with an average liquid water content in clouds and were able to estimate a turnover time of tropospheric aerosols by making certain assumptions about the rainout and washout efficiencies. Their calculations, reviewed by Junge (1963), gave as a result an average value for the residence time of about 8 days. For aerosols formed at the earth's surface and concentrated in the lower layers of the troposphere a residence time of only 2–4 days was estimated.

A shortcoming of the model used by Junge & Gustafson is the assumption that the average daily rainfall is the result of a large number of independent rainfalls. No information about the real distribution of rainy and dry periods was included.

In the present study a quite different ap-

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proach has been used. A statistical investigation of the distribution of dry and precipitation periods forms the basis for our calculations. From these investigations we can estimate an expected length of time that a particle will spend in the atmosphere before it experiences its first precipitation. In the case of an infinitely rapid washout, this time will be identical with the expected length of life for the particle in the atmosphere. In reality, of course, the length of life will be longer and the modification will depend upon the actual scavenging efficiency.

When interpreting the results of Junge & Gustafson (as well as the results derived in the subsequent sections) it should be borne in mind that they refer only to removal by precipitation. In reality there are also other mechanisms by which aerosols are removed from the atmosphere: dry sedimentation becomes important for particles with a diameter above about $10\ \mu\text{m}$ and impaction on the ground and on vegetation may contribute significantly to the removal (Eriksson, 1955; White & Turner, 1970). Thus the actual turnover times for aerosols in the lower troposphere is shorter than that estimated from precipitation scavenging only.

2. The scavenging process

According to Junge (1963) three separate processes can be separated as responsible for the uptake of aerosol particles by cloud droplets (rainout): (1) the consumption of condensation nuclei, (2) the attachment of aerosol particles, primarily Aitken particles, to the cloud droplets by Brownian motion, (3) the transfer of aerosol particles to the droplets by the water vapor gradient (Facy-effect).

In a detailed evaluation of the rainout efficiency, the effects of electrical forces will furthermore have to be considered. In addition to these processes the precipitation elements (raindrops or snowflakes) pick up aerosol particles as they fall through the subcloud layer (washout).

Theoretical calculations of the efficiency of these processes have been made by several authors (see e.g. Chamberlain, 1953; Greenfield, 1957; Zimin, 1962; Junge 1963 and contributions to the Precipitation Scavenging Symposium held at Richland, Washington, 1970). Junge concluded that the condensation process (1) itself is the most efficient of the rainout

mechanisms for changes of the total mass of natural aerosols, but that the mechanisms (2) and (3) may become important for aerosol particles smaller than $0.1\ \mu\text{m}$. Except in areas where the aerosols are concentrated in the lowest layers of the atmosphere and in circumstances in which a considerable part of the mass is contained in particles larger than about $1\ \mu\text{m}$, the washout process is believed to be less effective than the rainout processes.

For a quantitative estimation of the scavenging efficiency—i.e. the probability of a given particle being removed from the atmosphere in a rain with a certain intensity—it seems appropriate to use the result of Makhon'ko (1967). He studied the integral effect of precipitation scavenging by looking at the time variation of contaminant concentrations (e.g. salts, dust and fission products) during the course of individual rains. Without having to make assumptions about the details of the physical processes, he arrived at estimates of scavenging coefficients which were of the order of 10^{-5} – $10^{-4}\ \text{sec}^{-1}$ for sub-cloud washout and 10^{-4} – $10^{-3}\ \text{sec}^{-1}$ for rainout. These figures apply to rain intensities of about 1 mm per hour. The coefficients are approximately proportional to rain intensity (Greenfield, 1957; Engelman, 1968).

A considerably larger range of variation of the washout coefficient has been given by Zimin (1962) by considering particles of different sizes. When taking into account the distribution of mass between different particle sizes for natural aerosols (c.f. Junge, 1963), Makhon'ko's estimate of the washout coefficient does not seem unreasonable.

In order to estimate an effective scavenging coefficient, including removal by rainout as well as washout, some information about normal vertical distributions of the aerosol particles is needed.

Data on the height distribution of natural aerosols compiled by Junge (1963) show that in most cases more than 50 % of the total aerosol mass is found in the sub-cloud layer ($\approx 1.5\ \text{km}$). Georgii (1970), on the other hand, has presented measurements of sulfate aerosols which indicate a much less pronounced decrease with altitude, at least outside industrial areas.

In our calculations we will not include any information about the size distribution of the aerosol or its height variation, but treat the scavenging in a very schematic way. Thus we

introduce an effective scavenging coefficient λ , defined as the average intensity for the removal of an aerosol particle situated somewhere in a vertical column of air in which precipitation is falling. On the basis of Makhon'ko's result and the above considerations we assume that this coefficient is in the range 5×10^{-4} to $3 \times 10^{-5} \text{ sec}^{-1}$ (or 2 to 0.1 h^{-1}) for a precipitation intensity of 1 mm h^{-1} . This estimate of the scavenging coefficient must of course be considered as somewhat uncertain. If, on the other hand, the scavenging coefficient for a particular contaminant can be shown to be outside this interval, the figures derived in section 4 can easily be modified (c.f. eq. (8) and Figs. 4 and 5).

The precise definition of the effective scavenging coefficient, in terms of the terminology used in section 4, is given by

$$\lambda = \lim_{\Delta \downarrow 0} \frac{P(\Delta)}{\Delta}$$

where $P(\Delta)$ is the probability of removal of an aerosol particle during the time interval Δ .

3. The statistical distribution of dry periods and periods with precipitation

The next step towards an estimate of the scavenging efficiency of precipitation is an investigation into the statistical distribution of rainy and dry periods. Explicitly, what is needed are the frequency distributions of (a) the length of time from an arbitrary moment during a dry period to the onset of the next precipitation period, (b) the length of time from an arbitrary moment during a precipitation period to the beginning of the next dry period.

We will denote the corresponding frequency functions η_d and η_p respectively.

Since we are primarily interested in the fate of those particles emitted into the atmosphere which follow the atmospheric motions, the η -functions should be referred to a Lagrangian reference frame. For practical reasons, however, we have confined ourselves to the study of precipitation statistics in an Eulerian coordinate system; i.e. the calculations have been based on records of precipitation intensity as a function of time at a certain station. The error introduced by this approximation will depend upon how

the air is moving relative to the rain-clouds. If the precipitating cloud were moving with the same speed as the particles, for example, the turnover time for the particles in the atmosphere would be either very short (if they were emitted within or below the cloud) or very long if they were emitted outside the cloud). In this extreme case, therefore, the distribution functions defined above would be very much different in a Lagrangian and an Eulerian reference frame. In the case of orographic precipitation, on the other hand, the clouds have a fixed position relative to the ground and the Eulerian statistics would be similar to the Lagrangian (provided that the precipitation is measured on the windward side of the mountains).

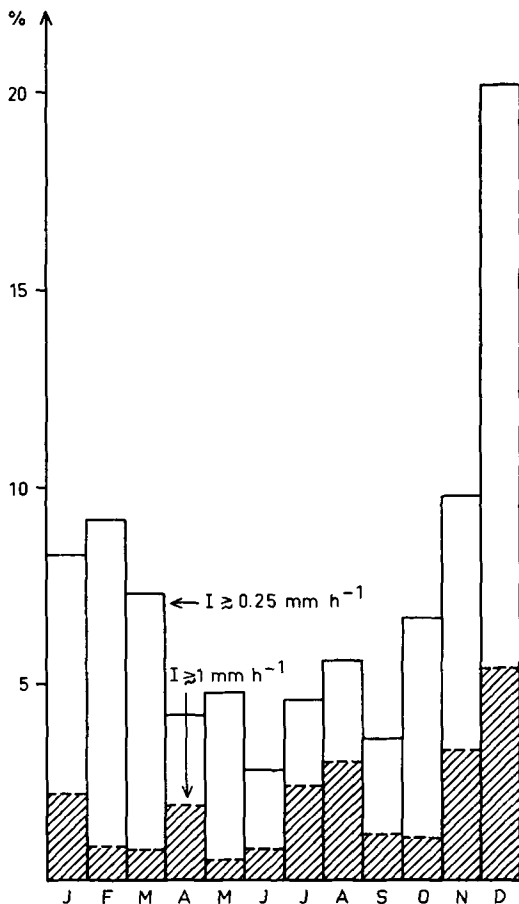


Fig. 1. Percentage of the total time during which precipitation occurred. Two different limits for the precipitation intensity have been used and the data refer to one year only.

Table 1. *Precipitation time as percentage of the whole period*

Intensity of precipitation	Summer (Apr.- Sep.) (%)	Winter (Oct.- Mars) (%)	Yearly average (%)
$I \geq 0.25 \text{ mm h}^{-1}$	4	10	7
$I \geq 1 \text{ mm h}^{-1}$	2 (1.6)	2 (2.2)	2

Since there is, in reality, an appreciable flow of air through a precipitating cloud, we believe that it is a reasonable approximation to study the precipitation statistics at fixed positions.

The data presented below refer to precipitation measured during one year (1966) at the station Västerväradukten near Stockholm. The station has been using an automatic recorder and two hour values of precipitation amounts have been evaluated.

In order firstly to obtain an idea of the frequency of occurrence of precipitation periods, we have computed the percentage of the total time each month during which precipitation occurred (Fig. 1). Average values for summer and winter are given in Table 1.

It is clear from these figures that the low intensity precipitation periods are appreciably longer in winter than in summer.

From the present precipitation data it seems reasonable to use the exponential distribution as a model for the frequency functions η_d and η_p . Furthermore, we assume a Markov dependence for the precipitation model. One formulation of this dependence is that, given the present weather, the past and future weather are independent of each other. We will return to the Markov dependence in section 4.

The first moments of the computed frequency

Table 2. *Time-scales (in hours) describing the distribution of dry and precipitation periods respectively*

For definition see text

	$\tau_d(h)$	$\tau_p(h)$
Summer	92	4
Winter	40	9
Year	68	7

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functions $\tau_j = \int_0^\infty \tau \eta_j(\tau) d\tau$ are given in Table 2. τ_d is thus the expected length of time from an arbitrary moment in a dry period until precipitation begins and τ_p the corresponding time for a precipitation period.

Figs. 2 and 3 show the accumulated distribution functions $\int_0^t \eta_j(\tau) d\tau$ for the two seasons. In the derivation of these distribution functions, all periods with an intensity of precipitation $I \geq 0.05 \text{ mm h}^{-1}$ have been classified as precipitation periods. Naturally, as this requirement for precipitation is limited to larger intensities of precipitation, τ_d increases and τ_p decreases. Thus the yearly average for τ_d increases to 76 h if the limit is set at 0.25 mm h^{-1} , and 170 h for 1 mm h^{-1} .

The figures derived in this section have been obtained from precipitation measurements at one Swedish station and for one year only. It is likely that the use of a longer time period would have given a somewhat different result. In other climates, the figures may certainly be very much different.

4. The removal time of aerosol particles from the atmosphere by precipitation scavenging

In this section we will combine our assumptions about the probability of a particle being removed during a precipitation period with the data on the statistical distribution of dry and precipitation periods given in the preceding section. A detailed treatment of this problem would demand an integration over different intensities of precipitation. The following calculation, however, is based on the simplifying assumption that all precipitation periods have a uniform intensity of precipitation.

In order to make the derivation slightly more general, we introduce a second effective scavenging coefficient allowing for a finite scavenging probability even during a dry period. In the numerical examples this coefficient will be put equal to zero.

We thus define λ_d and λ_p as the scavenging coefficients during a dry period and a precipitation period respectively. Let $\lambda(t)$ be a stochastic process defined by

$$\lambda(t) = \begin{cases} \lambda_d & \text{if dry period at time } t \\ \lambda_p & \text{if precipitation period at time } t \end{cases}$$

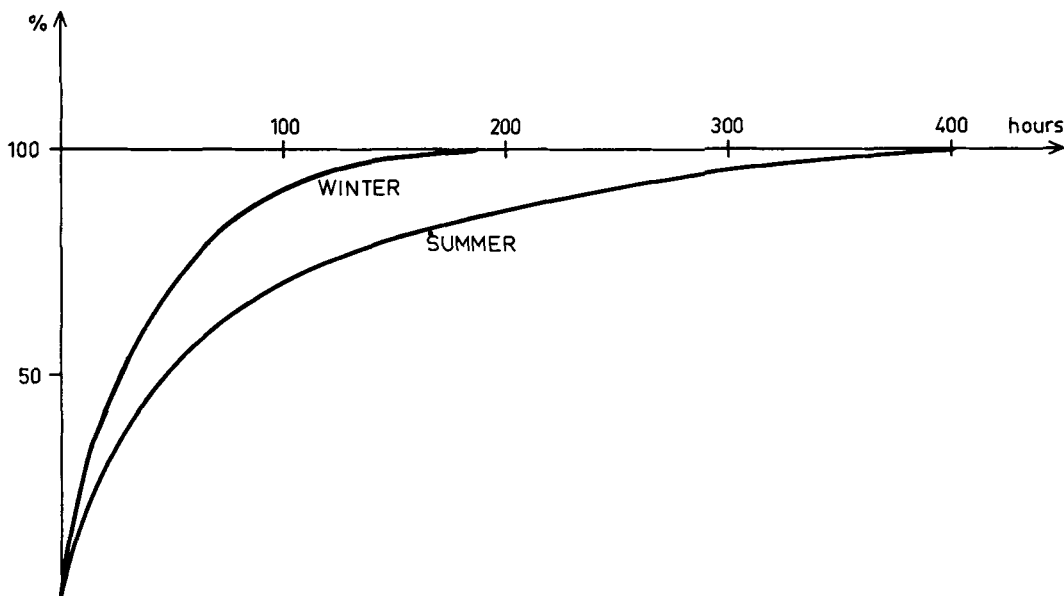


Fig. 2. Distribution function for the time from an arbitrary moment during a dry period until precipitation starts falling.

According to the assumptions made we have

$$\lim_{\Delta \downarrow 0} \frac{1}{\Delta} \Pr\{\lambda(t+\Delta) = \lambda_d \mid \lambda(t) = \lambda_p\} = \frac{1}{\tau_p} \quad (1a)$$

and

$$\lim_{\Delta \downarrow 0} \frac{1}{\Delta} \Pr\{\lambda(t+\Delta) = \lambda_p \mid \lambda(t) = \lambda_d\} = \frac{1}{\tau_d} \quad (1b)$$

As indicated in section 3, $\lambda(t)$ is assumed to be a Markov process. The formulation given in section 3 is equivalent to saying that, knowing $\lambda(t)$, an additional knowledge of $\lambda(\tau)$ for $\tau < t$ does not change the conditional distribution of $\lambda(t+\Delta)$ for $\Delta > 0$.

Thus $\lambda(t)$ is a Markov process with stationary transition probabilities. This is important for the further calculations.

If T is the time for the removal of a particle from the atmosphere since it entered the atmosphere then

$$\Pr\{T > t\} = E \left\{ \exp \left(- \int_0^t \lambda(\tau) d\tau \right) \right\} \quad (2)$$

where E denotes the "expected value of".

The model is connected with a class of doubly stochastic Poisson processes studied by Rudemo (1972).

Let $p_d = \Pr\{\lambda(0) = \lambda_d\}$ and $p_p = 1 - p_d$. p_d is thus the probability of a dry period when the particle enters the atmosphere. Of particular interest are the p_d values of 0, $\tau_d/(\tau_d + \tau_p)$ and 1. $p_d = 0$ or 1 corresponds to known weather at the time the particle enters the atmosphere and $p_d = \tau_d/(\tau_d + \tau_p)$ corresponds to the situation where the particle enters the atmosphere at an arbitrary time, independent of the weather. As distinguished from the symbols τ_d and τ_p in eq. (1), τ_d and τ_p should here refer to an Eulerian frame of reference.

Put

$$G_d(t) = \Pr\{T > t \mid \lambda(0) = \lambda_d\} \quad (3a)$$

$$G_p(t) = \Pr\{T > t \mid \lambda(0) = \lambda_p\} \quad (3b)$$

and

$$G(t) = \Pr\{T > t\}$$

where thus

$$G(t) = p_d G_d(t) + p_p G_p(t) \quad (4)$$

Gaver (1963) calculated the Laplace transform of $G(t)$ for a more general situation than the present. Due to our assumption about an exponential distribution it is possible to derive an expression for $G(t)$ by using simpler methods. Since it seems complicated to invert the Laplace

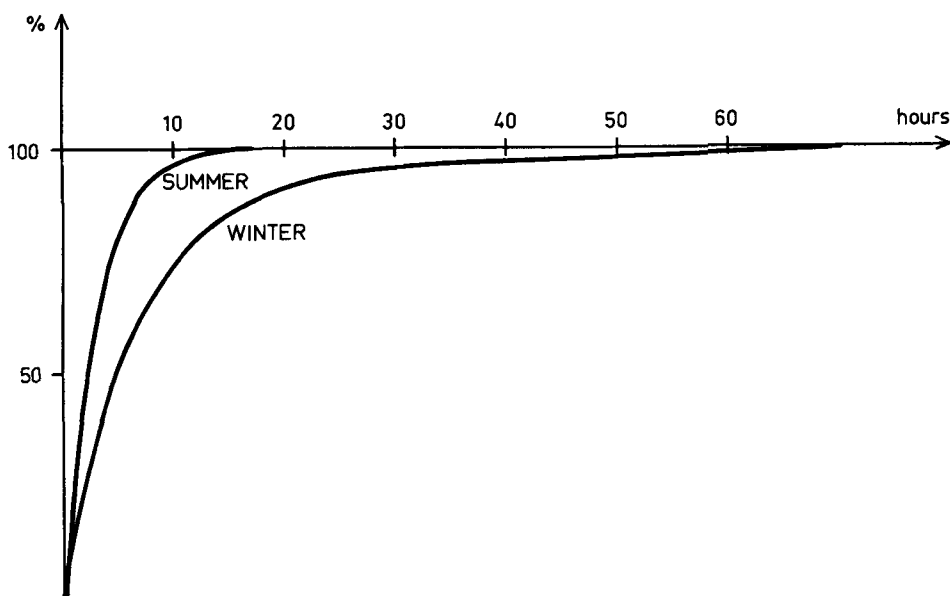


Fig. 3. Distribution function for the time from an arbitrary moment during a precipitation period to the end of the period.

transform given by Gaver (1963), we will derive the expression with these simpler methods.

It follows from the properties of $\lambda(t)$ that

$$E \left\{ \exp \left(- \int_{\Delta}^{\Delta+t} \lambda(\tau) d\tau \right) \middle| \lambda(\Delta) \right\}$$

is independent of Δ .

Considering the possible changes of $\lambda(t)$ for $t \in (0, \Delta)$ we get

$$G_d(t+\Delta) = \left(1 - \frac{\Delta}{\tau_d} \right) e^{-\Delta \lambda_d} G_d(t) + \frac{\Delta}{\tau_d} e^{-\Delta \theta} G_p(t) + o(\Delta)$$

where θ is an average of λ_d and λ_p . Letting $\Delta \downarrow 0$ and using the fact that $e^{-x} = 1 - x + o(x)$ we obtain

$$G'_d(t) = - \left(\frac{1}{\tau_d} + \lambda_d \right) G_d(t) + \frac{1}{\tau_d} G_p(t)$$

and similarly

$$G'_p(t) = \frac{1}{\tau_p} G_d(t) - \left(\frac{1}{\tau_p} + \lambda_p \right) G_p(t)$$

From the general theory of systems of linear

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differential equations (cf. e. g. Karlin, 1966, p. 208) it follows that

$$G_d(t) = \alpha_d e^{tr_1} + \beta_d e^{tr_2} \quad (5a)$$

and

$$G_p(t) = \alpha_p e^{tr_1} + \beta_p e^{tr_2} \quad (5b)$$

where r_1 and r_2 are the eigenvalues of the matrix

$$\begin{pmatrix} - \left(\frac{1}{\tau_d} + \lambda_d \right) & \frac{1}{\tau_d} \\ \frac{1}{\tau_p} & - \left(\frac{1}{\tau_p} + \lambda_p \right) \end{pmatrix}$$

Thus we have

$$r_1 = - \frac{1}{2} \left(\frac{1}{\tau_d} + \frac{1}{\tau_p} + \lambda_d + \lambda_p \right) - \sqrt{\frac{1}{4} \left(\frac{1}{\tau_d} + \frac{1}{\tau_p} + \lambda_d + \lambda_p \right)^2 - \lambda_d \lambda_p - \frac{\lambda_d}{\tau_p} - \frac{\lambda_p}{\tau_d}}$$

and

$$r_2 = - \frac{1}{2} \left(\frac{1}{\tau_d} + \frac{1}{\tau_p} + \lambda_d + \lambda_p \right) + \sqrt{\frac{1}{4} \left(\frac{1}{\tau_d} + \frac{1}{\tau_p} + \lambda_d + \lambda_p \right)^2 - \lambda_d \lambda_p - \frac{\lambda_d}{\tau_p} - \frac{\lambda_p}{\tau_d}}$$

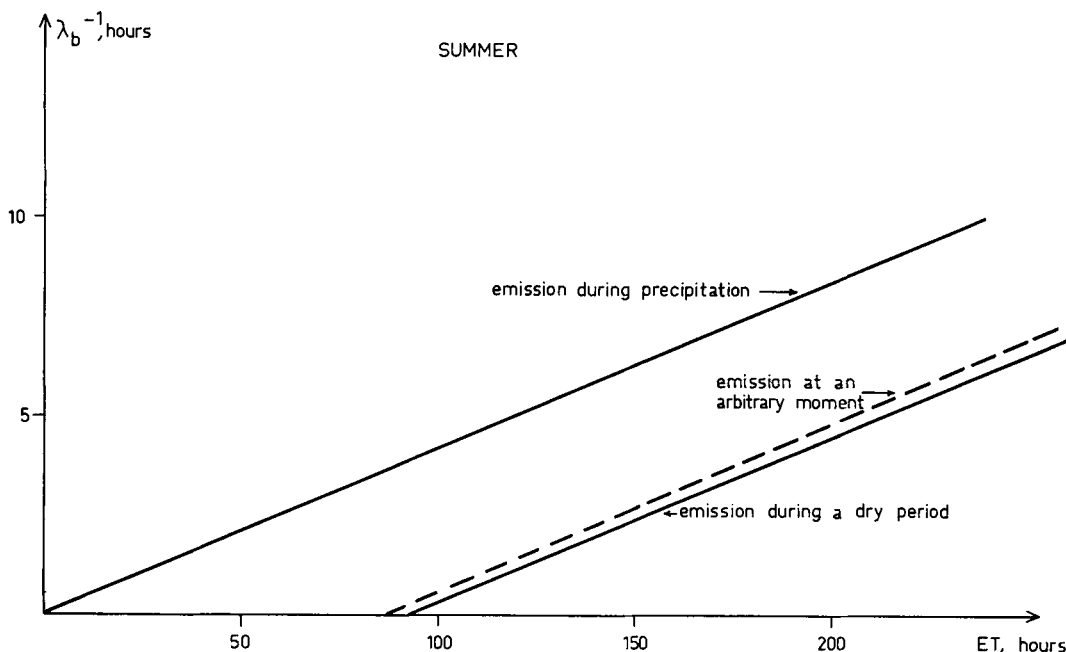


Fig. 4. Expected length of life (turnover time) of a particle as a function of the reciprocal scavenging coefficient. Summer.

From eqs. (4) and (5) it follows that

$$G(t) = \alpha e^{tr_1} + \beta e^{tr_2}$$

where, of course, α and β are dependent upon p_d . Since $G(0)=1$ we have $\alpha + \beta = 1$ and thus

$$G(t) = \alpha e^{tr_1} + (1 - \alpha)e^{tr_2} \quad (6)$$

From the above it is seen that a further condition is required in order to calculate α . Since $G(\infty)=0$ we obtain after integration of the differential eqs.

$$\begin{aligned} -1 &= -\left(\frac{1}{\tau_d} + \lambda_d\right) \int_0^\infty G_d(t) dt + \frac{1}{\tau_d} \int_0^\infty G_p(t) dt \\ -1 &= \frac{1}{\tau_p} \int_0^\infty G_d(t) dt - \left(\frac{1}{\tau_p} + \lambda_p\right) \int_0^\infty G_p(t) dt \end{aligned}$$

and consequently

$$\int_0^\infty G(t) dt = \frac{\tau_d + \tau_p + \tau_d \tau_p (p_d \lambda_p + p_p \lambda_d)}{\tau_d \lambda_d + \tau_p \lambda_p + \tau_d \tau_p \lambda_d \lambda_p} \quad (7)$$

The left hand side of eq. (7) is by definition equal to ET, i.e. the expected time of removal. α is thus determined by

$$ET = \alpha \int_0^\infty e^{tr_1} dt + (1 - \alpha) \int_0^\infty e^{tr_2} dt = \frac{\alpha}{-r_1} + \frac{1 - \alpha}{-r_2}$$

or after some calculations

$$\alpha(r_1 - r_2) = r_1 + \frac{1}{\tau_d} + \frac{1}{\tau_p} + p_d \lambda_p + p_p \lambda_d$$

where r_1 and r_2 are given above.

In the numerical examples presented below, λ_d will be put equal to zero. The expression for ET, eq. (7), is thus simplified to

$$ET = \tau_d p_d + \frac{\tau_d + \tau_p}{\tau_p} \frac{1}{\lambda_p} \quad (8)$$

It can be shown (Eriksson, 1971) that for a reservoir with a constant rate of exchange with its surroundings, the expected length of life for a particle will be equal to the total mass in the reservoir divided by the flux out of it. The latter timescale is commonly referred to as the turnover time, and we will therefore use this term as synonymous with the expected removal time, ET.

Figs. 4 and 5 show ET as a function of the reciprocal scavenging coefficient λ_p for winter

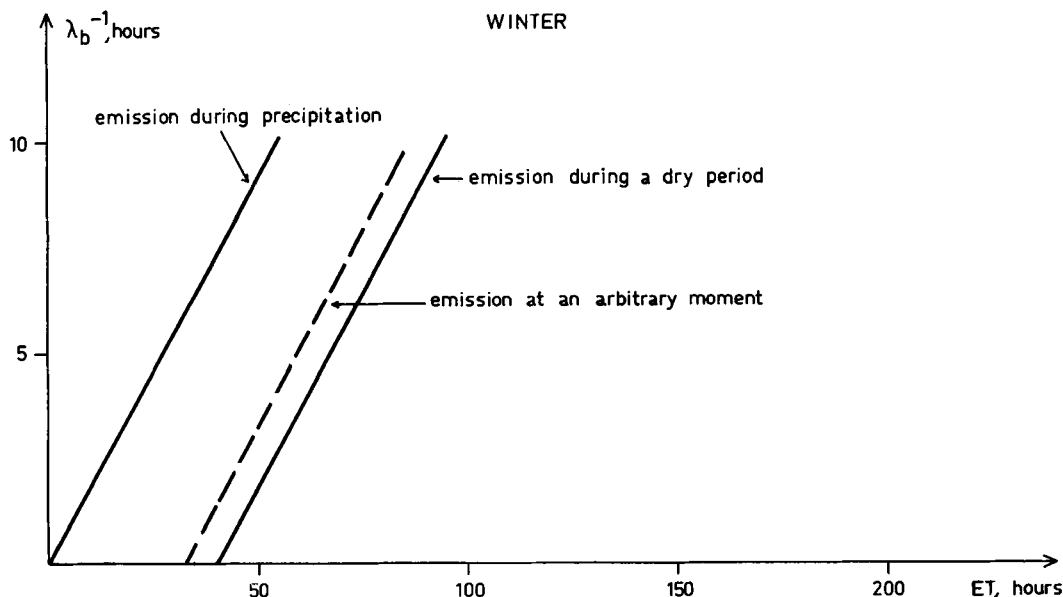


Fig. 5. Same as Fig. 4. Winter.

and summer conditions respectively. The three curves in each figure refer to different values of p_d , i.e. different weather at the time of emission. The intercepts with the abscissae correspond to an infinitely rapid washout as soon as precipitation begins and thus represent lower limits to the expected length of life. For emission during a dry period these intercepts are equal to τ_d (c.f. section 3) and for emission at an arbitrary time equal to

$$\tau_d \frac{\tau_d}{\tau_d + \tau_p}$$

i.e. about 88 and 33 hours for summer and winter respectively. It is seen that the value of ET in summer depends to a high degree on the scavenging coefficient, whereas in winter this dependence is less pronounced. This difference is due to the much longer precipitation periods in winter.

For an arbitrary time of release and with the range of values given in section 2 we obtain the following figures for ET: 35–80 h in winter and 100–300 h in summer. Noting that the average precipitation intensity in summer is higher than in winter, one may choose as typical scavenging coefficients λ_p , 0.4 h^{-1} for summer and 0.25 h^{-1} for winter conditions. These figures correspond to ET values of roughly 150 and 50 h respectively.

For convenience we introduce the distribution function $F(t)$, defined as

$$F(t) = \text{Pr}\{T \leq t\} = 1 - G(t) \quad (9)$$

The shape of the F function is shown in Figs. 6 and 7. λ_p has been set to 0.4 h^{-1} for both summer and winter cases and the corresponding ET values have been indicated. The curves in this figure can be interpreted either in terms of removal probabilities for a single particle—in accordance with the previous derivations—or, in terms of actual distribution of the length of life of a large number of particles emitted independently of each other, but in the same weather conditions.

It is clearly seen from Figs. 6 and 7 that a large part of the particles will have been removed from the atmosphere within the first 3–10 hours in the case where they are emitted during a precipitation period.

5. The frequency distribution function for the length of life of the particles

As pointed out in the introduction the distribution of the length of life of particles in the atmosphere is closely connected with the distribution of particles on the ground away from the

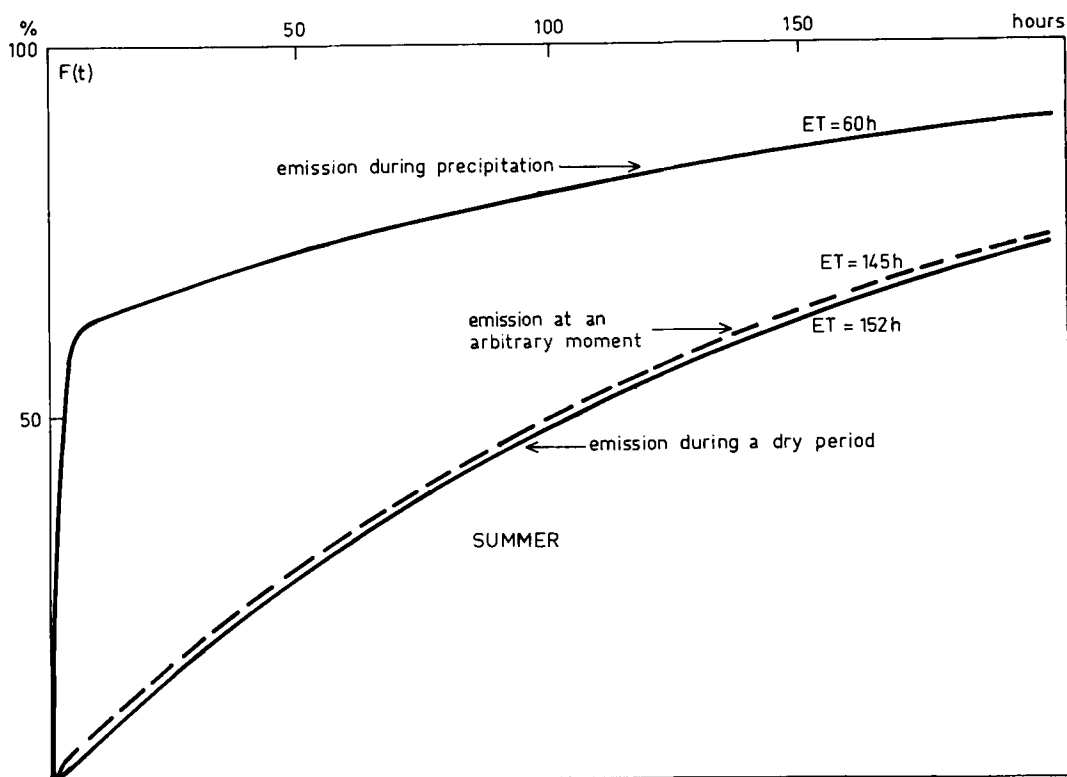


Fig. 6. Distribution function for the length of life of a particle emitted under various conditions. Summer.

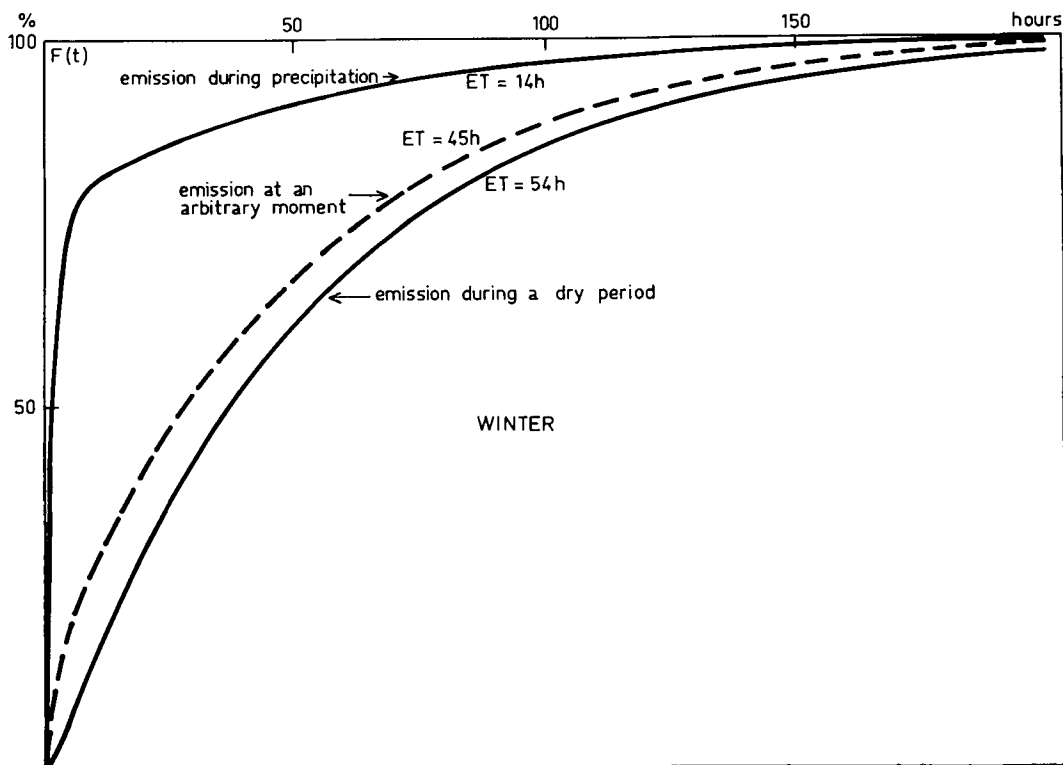


Fig. 7. Same as Fig. 6. Winter.

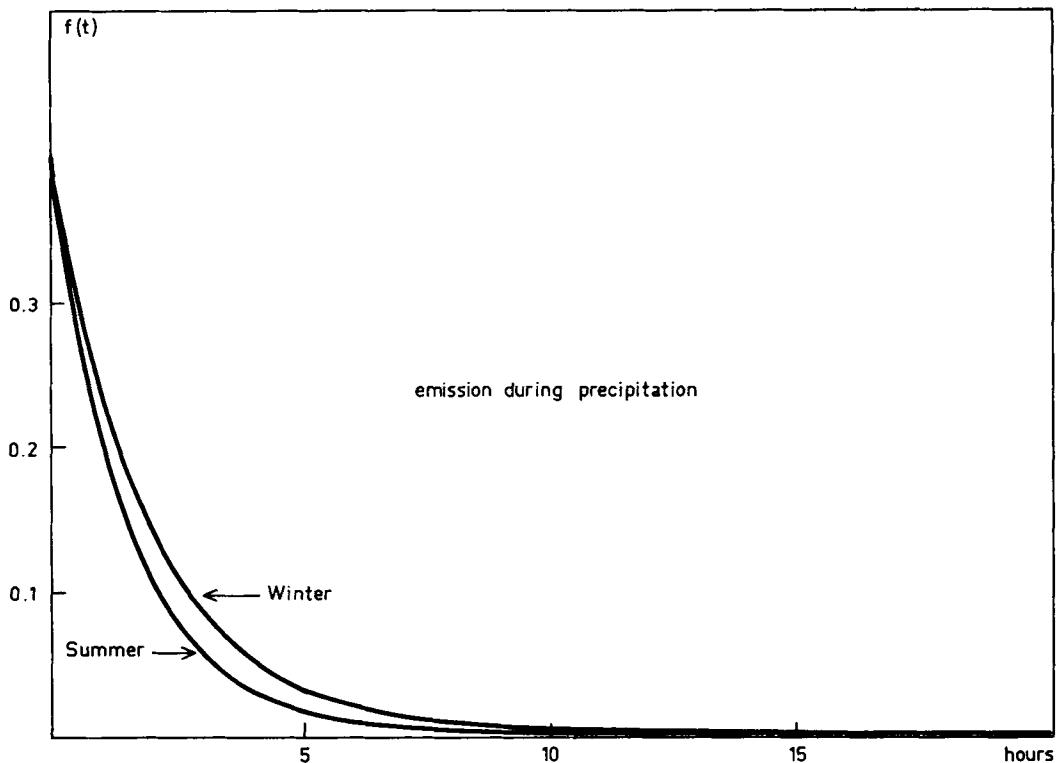


Fig. 8. Frequency function for the length of life of a particle emitted during precipitation.

source. In a discussion of the deposition pattern of particles on the ground it is of interest to know the frequency function for the length of life of the particles in the atmosphere—i.e. the age distribution for the particles leaving the atmosphere. This function $f(t)$ is related to the distribution function $F(t)$ by

$$f(t) = -F'(t)$$

Figs. 8, 9 and 10 show the shape of $f(t)$ for varying weather at the time of emission. (Note the differences in the scales.)

Assuming a constant transport velocity away from the source and no lateral diffusion—as in the case of an infinite line source oriented at right angle to the wind—the deposition $d(x)$ of particles on the ground at a distance x from the source will be proportional to $f(x)$. It is seen from Fig. 10 that with these assumptions the average deposition in winter time will have decreased by a factor two within the first 30–50 km (corresponding to $2\frac{1}{2}$ hours travel-time) and by a factor five within 10 hours. In summer, the

increased deposition close to the source is less pronounced (cf. Fig. 9).

When studying the deposition around a point source it is necessary to take into account also the effect of the horizontal turbulent diffusion. A simple way to do this (cf. Rodhe, 1972) is to assume a constant transport velocity away from the source and a uniform spread out of the trajectories within a certain sector or possibly around the whole circle. In this case the deposition per square meter would be

$$d(r) = \frac{A}{r} f(r)$$

where A is a constant.

At distances from the source that are longer than the length scale of the most important dispersive motions (in the case of a continuously emitting source in mid-latitudes this would correspond to the so-called synoptic scale, $\sim 1\,000$ km) the factor $1/r$ should be modified to give a less rapid decrease.

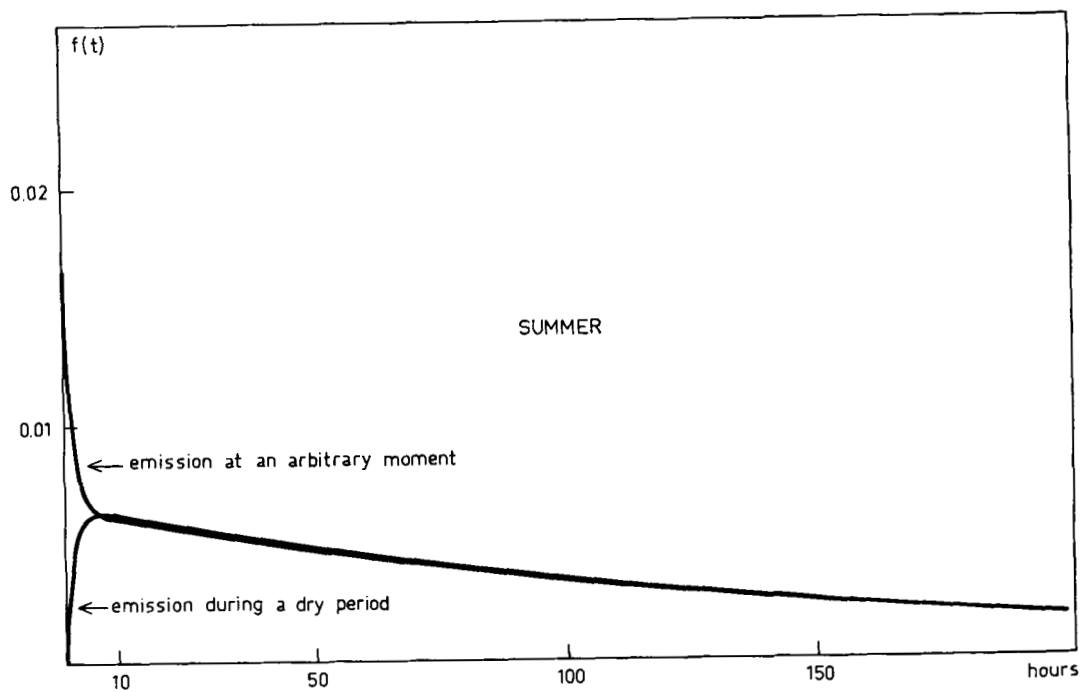


Fig. 9. Frequency function for the length of life of a particle emitted during dry period or at an arbitrary time. Summer.

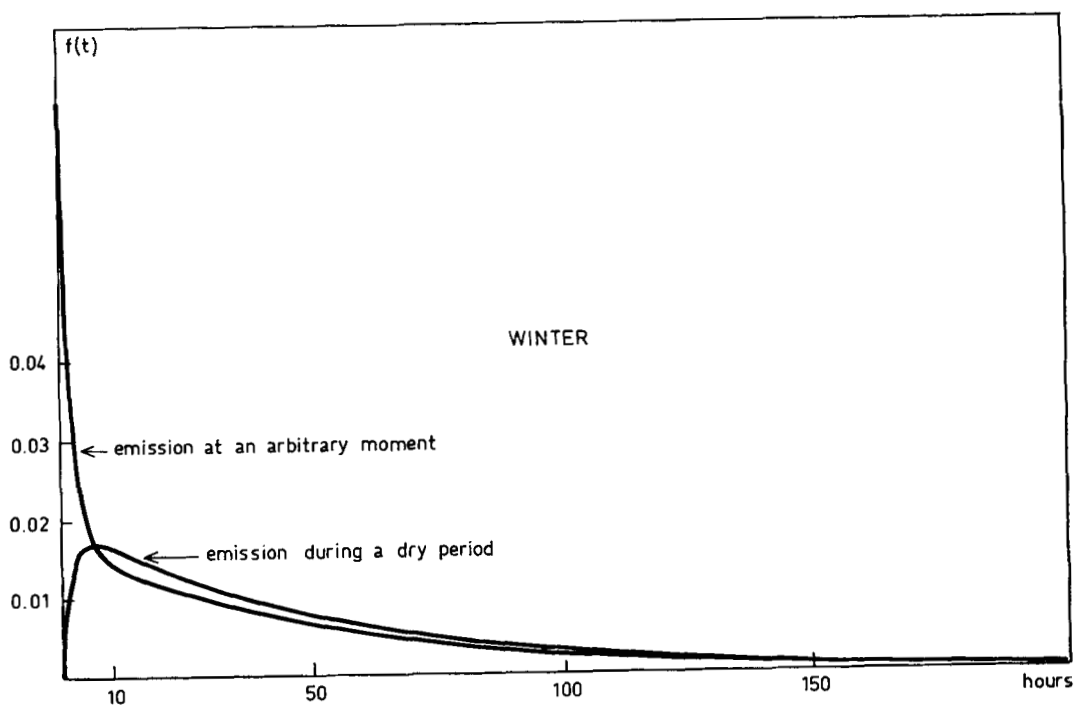


Fig. 10. Same as Fig. 9. Winter.

6. Concluding remarks

Based on an assumption about the precipitation scavenging efficiency and on a statistical investigation of the distribution of dry and precipitation periods, we have estimated the turnover time for tropospheric aerosols to be 35–80 h in winter and 100–300 h in summer. These figures agree reasonably well with those derived by Junge & Gustafson (1957), reviewed by Junge (1963), though it is to be noted that the derivation made in this paper is rather different from that of Junge & Gustafson.

As was pointed out in section 3, an uncertainty in our calculations arises in the assumption that precipitation statistics measured at a certain station are similar to those experienced by a particle moving with the winds. Since it is known (Pasquill, 1962) that the large scale spectrum of motion measured in a Lagrangian frame of reference is shifted to longer wavelengths (as compared to the Eulerian spectrum), one might suspect that the turnover times we have derived are underestimated. It follows from the expression for the turnover time—eq. (8)—how a proportional increase in the time-scales τ_d and τ_p will modify the value of the turnover time. If the emission takes place during a precipitation period ($p_d = 0$) there will be no change in ET. For emission at an arbitrary moment (cf. the dashed lines in Figs. 5 and 6), however, ET will increase.

Of course, one has to be careful when generalizing the results of this study to other climates. In subtropical regions, for example, the turnover time may be very much longer. In such a case the relative importance of the dry deposition becomes consequently larger.

Although it is convenient to use a single parameter—e.g. the turnover time ET—to characterize the distribution of the length of life of particles in a reservoir, the value of such

a parameter tells very little about the shape of the distribution function. We have therefore explicitly computed such distribution functions for some different conditions (Figs. 6 and 7), and discussed the connection between the frequency distribution function of the length of life of particles emitted into the atmosphere and the distribution of particles on the ground away from the source.

The deposition patterns that arise in our model as a result of the precipitation scavenging have been discussed in section 5. It was shown that, particularly during winter conditions, an appreciable increased deposition could be expected within the closest hundred kilometres to the source (corresponding to about 5 hours travel time, cf. Fig. 10). The number of particles deposited within this distance would, however, only amount to some 10 % of the total emission (cf. Fig. 6).

Certain atmospheric contaminants are emitted mainly in gaseous form and aerosols are created only gradually in the air. This is the case for example, with anthropogenic sulfur emissions (Georgii, 1970; Rodhe et al., 1972), where most of the sulfate containing aerosols are not formed until the sulfur dioxide has become oxidized. Since the precipitation scavenging of sulfur dioxide is probably less efficient than that of the sulfate aerosols (Hales et al., 1970), one may expect a less pronounced local precipitation deposition in this case.

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О ВРЕМЕНИ УДАЛЕНИЯ АЭРОЗОЛЬНЫХ ЧАСТИЦ ИЗ АТМОСФЕРЫ ОСАДКАМИ

Описываются результаты статистического исследования продолжительности периодов с осадками и без на одной из шведских станций. Показано, что ожидаемая длительность времени от произвольного момента до начала следующего периода с осадками составляет 90 и 35 часов для лета и зимы, соответственно. На основе этих результатов выведены выражения для ожидаемого времени жизни аэрозольной частицы в нижних слоях тропосферы.

Оценки эффективных коэффициентов вымывания осадками дают ожидаемые времена жизни порядка 100–300 часов летом и 35–80 часов зимой. Обсуждается связь между распределениями времен жизни частиц в атмосфере и распределением частиц на поверхности земли вдали от источника и подчеркивается важность изучения распределений возраста частиц.