Estimates of Characteristic Times for Precipitation Scavenging

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ABSTRACT

We address the problem of using climatological data to estimate residence times in the atmosphere of particles subjected to precipitation scavenging. Basic parameters are the scavenging coefficient \( \lambda \), the rainfall intensity \( R \) and the length of dry and precipitation periods \( T_D \) and \( T_P \). Previous models used to estimate residence times are reviewed and compared with "real" data. It is found that some of the earlier models substantially underestimate the average residence times, particularly for particles with a high value of the scavenging coefficient. An approximate model valid for long-lived particles with a relatively small scavenging coefficient \( [\lambda \leq (5 \text{ h}^{-1}) \text{ at } R = 1 \text{ mm h}^{-1}] \) is formulated and tested. Daily rainfall values are adequate as input in this model but several years of data are required in order to arrive at reasonable estimates of the residence time. The fundamental difficulty associated with all the models discussed is that a proper application requires Lagrangian rainfall data, i.e., information about the weather experienced by the particle as it is carried along with the winds, whereas virtually all data available from meteorological observations are obtained at fixed locations (Eulerian data).

1. Introduction

A good knowledge about the processes that remove pollutants, as well as naturally occurring compounds, from the atmosphere is essential when it comes to making quantitative estimates of transport of such compounds through the atmosphere or their fallout on the surface. Whereas direct uptake at the surface—dry deposition—is a process which is more or less continuous in time, precipitation scavenging occurs only intermittently in connection with the precipitation events. In order to describe and model the process of precipitation scavenging, it is therefore necessary to take into account not only the physical processes responsible for the uptake of pollutants into the hydrometeor and the possible chemical transformation occurring inside it, but also the frequency with which an air parcel finds itself in contact with the hydrometeors. In a previous paper (Rodhe and Grandell, 1972, hereafter referred to as RG) we studied the problem of how the lifetime of a particle subjected to randomly occurring precipitation events could be related to the scavenging coefficients for the dry and wet periods and to time scales characterizing the lengths of the dry and wet periods, respectively. In the present paper we address the same problem with the aim of 1) reviewing some recent work along similar lines; 2) generalizing our previous treatment of the problem; and 3) providing better climatological estimates of the parameters. In a final paragraph we discuss the problem of the relation between removal times estimated in Lagrangian and Eulerian frames of reference.

As in RG, little emphasis will be put on the description of the microphysical and chemical aspects of precipitation scavenging. The reader is here referred to review papers like those of Engelmann (1968), Hales (1972), Slinn (1977) and Slinn et al. (1978).

The question of the relative importance of subcloud and in-cloud scavenging is not made a central issue in our arguments. We believe that in most cases in-cloud scavenging is the dominant mechanism of the two. But as long as the material scavenged inside the cloud can be considered as drawn from the mixed layer below it, for our purposes the distinction between in-cloud and subcloud scavenging is not so important for the qualitative description of the overall process. However, when it comes to estimating the parameters from real data it is likely to be an important problem (cf. Section 6).

Although it is straightforward matter to include a finite rate of removal during non-precipitating periods into the calculations, we have chosen not to do so, mainly for pedagogical reasons. Thus, scavenging will subsequently refer only to precipitation scavenging.
2. Review of earlier models

In this section we describe briefly the basic characteristics of some of the models previously published which address the question of estimating time scales for precipitation scavenging.

Let \( \lambda_p \) (dimensions: \( T^{-1} \)) be a scavenging coefficient describing the rate of removal from the atmosphere of a pollutant subjected to a precipitation event, be it subcloud or in-cloud. In the general case \( \lambda_p \) will be a function of the characteristics of the pollutant, the type of precipitation (rain or snow), the intensity of precipitation, the temperature, etc. For simplicity we consider only one particular type of pollutant at a time and retain only the dependence of the scavenging coefficient on the intensity of precipitation \( R \).

Taking into account the fact that precipitation events generally occur only during a limited fraction of the time, how long a time will a particle on the average remain in the atmosphere before it is brought down by the precipitation? In the following subsections we will review briefly previous attempts to answer this question.

A common assumption in all models considered below, except in the classical model, is that the scavenging coefficient varies with time and that it is described by a stationary stochastic process \( \lambda(t) \). Thus, \( \lambda(t) \) is the scavenging coefficient at time \( t \). The residence time in the atmosphere of a particle is a random variable \( T \). The probability \( G(t) \) for a particle surviving a time \( t \) is then given by

\[
G(t) = P(T > t) = E\left[ \exp\left( -\int_0^t \lambda(s)ds \right) \right]
\]

and the average residence time by

\[
E(T) = \int_0^\infty G(t)dt.
\]

The latter expression follows by partial integration from \( \int_0^\infty t g(t)dt \), where \( g(t) \) is the corresponding density function.

a. The classical model

A simple approach to the problem just formulated is to neglect an explicit consideration of dry and precipitation periods experienced by the particle and to assume that the average removal rate \( \lambda_0 \) is given by \( f \lambda_p \), where \( f \) is the fraction of time—as a climatological average—that a volume of air experiences precipitation (see Appendix for list of symbols), i.e.,

\[
\lambda_0 = f \lambda_p. \tag{2.1}
\]

It should be noted that \( f \) has to be evaluated in a Lagrangian frame of reference. In many studies Eulerian rainfall data have been used to estimate \( f \).

An implicit assumption underlying Eq. (2.1) is that precipitation occurs during many short events separated by dry periods which on the average are short compared to \( (f \lambda_p)^{-1} \). Junge and Gustafson (1957) made this basic assumption and furthermore assumed the scavenging coefficient for both in-cloud and subcloud scavenging to be proportional to the rainfall intensity so that

\[
\lambda_0 = aR_0, \tag{2.2}
\]

where \( R_0 \) is the precipitation rate averaged over both precipitation and dry periods. In the model of Junge and Gustafson (1957) the constant \( a \) is given essentially by \( \epsilon \rho_w/LH \), where \( \epsilon \) is a nondimensional efficiency factor, \( \rho_w \) the density of water, \( L \) the average liquid water content of a precipitating cloud and \( H \) the tropopause height.

Expressions equivalent to (2.1) and (2.2) have been employed by several authors in connection with transport models particularly on regional and global scales (see further Section 7). Let \( T \) denote the time for the removal of a particle from the atmosphere since it entered it. The probability for a particle of surviving a time \( t \) in this model is then given by

\[
G(t) = P\{T > t\} = e^{-\lambda_0 t} \tag{2.3}
\]

and the average time to the removal is given by

\[
E(T) = \lambda_0^{-1}. \tag{2.4}
\]

b. The Markov model

In this model, which was first applied to the scavenging problem by Rodhe and Grandell (1972), explicit consideration is given to the occurrence of dry and precipitation periods.

Let \( \lambda(t) \) be a stochastic process defined by

\[
\lambda(t) = \begin{cases} 
0, & \text{if a dry period at } t \\
\lambda_p, & \text{if a precipitation period at } t.
\end{cases}
\]

The probability for a particle to survive a time \( t \) is then given by

\[
G(t) = E\left[ \exp\left( -\int_0^t \lambda(s)ds \right) \right].
\]

If the assumptions are made that \( \lambda(t) \) is a Markov process with stationary transition probabilities so that the lengths of the dry and the precipitation periods are exponentially distributed, the distribution function and the expected value of the residence time can be readily derived. The assumption of a Markov process may be formulated as follows: knowing \( \lambda(t) \), an additional knowledge of \( \lambda(\tau) \) for \( \tau < t \) does not change the conditional distribution of \( \lambda(t + \Delta) \) for \( \Delta > 0 \) or equivalently: given the present weather, the future and past weather are independent of each other.

The relationship between the length of dry and precipitation periods and the transition intensities of
\( \lambda(t) \) is given by

\[
\lim_{\Delta \to 0} \frac{1}{\Delta} \mathbb{P}\{\lambda(t + \Delta) = 0 | \lambda(t) = \lambda_p\} = \frac{1}{\tau_p},
\]

\[
\lim_{\Delta \to 0} \frac{1}{\Delta} \mathbb{P}\{\lambda(t + \Delta) = \lambda_p | \lambda(t) = 0\} = \frac{1}{\tau_d}.
\]

where \( \tau_p \) and \( \tau_d \) are the average length of precipitation and dry periods, respectively, experienced by the moving particle. [In RG \( \tau_d(\tau_p) \) were defined as the expected length of time from an arbitrary moment in a dry (wet) period until precipitation begins (ends). In view of the assumption of exponential distributions that definition is equivalent to the one given here]. With these assumptions one may show (cf. RG) that

\[
G(t) = \mathbb{P}\{T > t\} = \alpha e^{\tau_d} + (1 - \alpha)e^{\tau_p},
\]

where

\[
\alpha = \frac{1}{\tau_d} + \frac{1}{\tau_p} + \rho_d \lambda_p,
\]

\[
r_{1,2} = \frac{-\frac{1}{2}(\frac{1}{\tau_d} + \frac{1}{\tau_p} + \lambda_p)}{\left[\frac{1}{\tau_d} + \frac{1}{\tau_p} + \lambda_p\right]^2 - \frac{\lambda_p}{\tau_d} \frac{1}{\tau_p}},
\]

\[
p_d = p\{\lambda(0) = 0\},
\]

\[
p_p = p\{\lambda(0) = \lambda_p\}.
\]

\[
E(T) = \tau_d \rho_d + \frac{\tau_d + \tau_p}{\tau_p} \frac{1}{\lambda_p}.
\]

The parameters \( \tau_p, \tau_d, \rho_d, \) \( \rho_p \) have to be estimated from data. If the emission of the pollutant takes place from a point fixed in space (e.g., a chimney), \( \rho_d \) and \( \rho_p \) can be readily estimated from Eulerian rainfall statistics. Actually, \( \rho_p \) may be well approximated by the frequency of occurrence of reported precipitation at climatological stations. A certain bias toward high values of this frequency may be anticipated because of the tendency of meteorological observers to report precipitation even if it is only occurring during part of the finite observation period. \( \tau_d \) and \( \tau_p \) refer to conditions experienced by the particles as they move along a trajectory and are more difficult to estimate. In RG rainfall statistics from a fixed point were used to estimate \( \tau_d \) and \( \tau_p \). This would correspond to the unrealistic situation when the pollutant remains immobile and the precipitating clouds move by. A further discussion about the relations between Eulerian and Lagrangian time scales is given in Section 6.

If all parameters are measured in an Eulerian time scale, the choice

\[
\rho_d = \frac{\tau_d}{\tau_d + \tau_p}
\]

corresponds to the case where the particle enters the atmosphere at an arbitrary time. In this case \( \lambda(t) \) is a stationary stochastic process and we have

\[
E(T) = \frac{\tau_d^2}{\tau_d + \tau_p} + \frac{\tau_d + \tau_p}{\tau_p \lambda_p}.
\]

Baker et al. (1979) used a model similar to that of RG applying it to an industrial haze layer. They derived not only an effective removal time scale—corresponding to relation (2.10)—but also the variance and the skewness of the steady-state concentration distribution.

c. The renewal model

This model is a generalization of the Markov model. Details of this generalization are given in Grandell and Rodhe (1978). Like in the Markov model the length of dry and precipitation periods are independent. We denote these periods by \( \tilde{T}_d \) and \( \tilde{T}_p \), respectively. During a precipitation period the removal rate is denoted by \( \tilde{\lambda} \), which also is a random variable. The removal rate \( \tilde{A} \) is assumed to be independent of all other random variables except the length of the period in which it acts.

We assume that the particle enters the atmosphere at an arbitrary time. Put \( \tau_p = E(\tilde{T}_p) \) and \( \tau_d = E(\tilde{T}_d) \), like in the Markov case, and define further the quantities

\[
\sigma_d^2 = \text{var}(\tilde{T}_d),
\]

\[
A = E(e^{-\tilde{\lambda}\tilde{T}_p}),
\]

\[
B = E\left(1 - e^{-\tilde{\lambda}\tilde{T}_p}\right),
\]

\[
C = E\left(e^{-\tilde{\lambda}\tilde{T}_p} - 1 + \tilde{\lambda}\tilde{T}_p\right).
\]

Then

\[
E(T) = \frac{1}{\tau_d + \tau_p} \left(\frac{\sigma_d^2}{2} + C + \frac{(B + \tau_d)^2}{1 - A}\right).
\]

The parameter \( \tilde{A} \) is the probability for a particle to survive a precipitation period.

The important generalizations in this model as compared to the Markov model are 1) no assumption is made about the shape of the distributions of \( \tilde{T}_d \) and \( \tilde{T}_p \); and 2) the scavenging intensity is allowed to vary from one precipitation period to another.

d. Short rain versions

In this subsection we shall treat the case where the length of the precipitation periods are short compared to the dry periods. Formally, we then put \( \tilde{T}_p = 0 \) and thus \( \tau_p = 0 \). Let \( N(t) \) be the number of precipitation episodes in the time interval \((0, t)\).
Assume that $N(t)$ is a stationary renewal process. This means that the length of the dry periods between the precipitation periods are independent and identically distributed random variables. Then (2.12) is simplified to

$$E(T) = \frac{\sigma^2_{\tau} - \tau_d^2}{2\tau_d} + \frac{\tau_d}{1 - A}, \quad (2.13)$$

since, at least formally, in this case $\lambda = \infty$ and thus $B = C = 0$. In this case $A$ is the probability for a particle to survive a precipitation period.

Now we simplify further and assume $N(t)$ to be a Poisson process. Then $T_d$ is exponentially distributed with $E(T_d) = \tau_d$. Since in this case $\sigma^2_{\tau} = \tau_d$ we get

$$E(T) = \frac{\tau_d}{1 - A} \quad (2.14)$$

which is equivalent to formula (21) in Slinn et al. (1978, p. 2063). The same simplification was used by Baker et al. (1979) when they derived a covariance function for the concentration of particles in the atmosphere.

e. The case with scavenging intensity proportional to precipitation intensity

In this section we assume that the precipitation intensity is described by a stationary stochastic process $R(t)$. Furthermore, the scavenging intensity $\lambda(t)$ is assumed to be directly proportional to $R(t)$, i.e., $\lambda(t) = aR(t)$. This assumption is intuitively reasonable and is also supported by observations (see, e.g., Dana and Hales, 1976; Maul, 1978). On the other hand, there also are investigations which indicate a weaker $R$ dependence (e.g., Scott, 1978).

We let $R(t)$ be the precipitation intensity at time $t$ at a certain fixed place (Eulerian frame of reference). The assumptions of stationarity corresponds to the case when a particle enters the atmosphere at an arbitrary time. We then let $R_0$ denote the average precipitation intensity, i.e.,

$$R_0 = E[R(t)]. \quad (2.15)$$

In the following we let $T_d(T_p)$ be the length of an arbitrary dry (wet) period and $\bar{M}$ be the amount of precipitation during that wet period. In order to emphasize the dependence on $a$ we denote the time for the removal of a particle from the atmosphere since it entered it by $T_a$.

Using these notations we shall reconsider the models treated earlier in this section.

1) The classical model

Assume that $R(t) = R_0$. Then $\lambda_0 = aR_0$ and thus

$$E(T_a) = (aR_0)^{-1}. \quad (2.16)$$

2) The Markov model

Let $R(t)$ be defined by

$$R(t) = \begin{cases} 0, & \text{if a dry period at time } t \\ R_p, & \text{if a precipitation period at time } t. \end{cases}$$

The transitions between 0 and $R_p$ are specified as the transitions of $\lambda(t)$ between 0 and $\lambda_p$. Then

$$R_0 = \frac{\tau_p R_p}{\tau_p + \tau_d}$$

and thus

$$E(T_a) = \frac{\tau_d}{\tau_d + \tau_p} + (aR_0)^{-1}. \quad (2.17)$$

It may be noted that the assumptions in this model imply that $\bar{M}$ is exponentially distributed with $E(\bar{M}) = \tau_p R_p$.

3) The renewal model

If we put $\bar{\lambda} = \bar{M}/T_p$, in the definitions of $\lambda$, $B$ and $C$, Eq. (2.12) remains unchanged.

4) Short rain models

We consider the special case where $N(t)$ is a Poisson process. Let $\bar{M}$ be the total amount of precipitation during an arbitrary precipitation episode. Let $\bar{M}_1, \bar{M}_2, \cdots$ be independent random variables with the same distribution as $\bar{M}$ and also independent of $N(t)$. Then $\int_0^t R(t) dt$ corresponds to $M_1 + M_2 + \cdots + M_{N(t)}$. Further, A, E(\text{e}^{-\lambda t}) and (2.13) remains unchanged. In this case

$$R_0 = E(\bar{M})/\tau_d. \quad (2.18)$$

We can now form a correspondence to the Markov model in this situation by assuming that $\bar{M}$ is exponentially distributed with mean $\tau_d R_0$. Then $A = (1 + aR_0 \tau_d)^{-1}$ and thus

$$E(T_a) = \tau_d + (aR_0)^{-1} \quad (2.18)$$

which agrees with (2.17) provided $\tau_p = 0$.

3. An approximate model for long-lived particles

In this section we shall stick to the assumption about the scavenging intensity being proportional to the precipitation intensity. As in Section 2e we assume $R(t)$ to be a stationary process with mean $R_0 = E[R(t)]$ and covariance function $\gamma(t) = \text{cov}[R(s), R(s + t)]$. We shall also consider the process

$$h(t) = \int_0^t R(t) dt$$

which is the total amount of precipitation in the time interval $[0, t]$. Then we have $E[h(t)] = R_0 t$. 

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Table 1. Estimates of covariances of daily rainfall series.

<table>
<thead>
<tr>
<th>k</th>
<th>Summer</th>
<th>Winter</th>
</tr>
</thead>
<tbody>
<tr>
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<td>12.44</td>
</tr>
<tr>
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<td>1.79</td>
</tr>
<tr>
<td>3</td>
<td>1.34</td>
<td>1.27</td>
</tr>
<tr>
<td>4</td>
<td>-0.18</td>
<td>-0.06</td>
</tr>
<tr>
<td>5</td>
<td>0.06</td>
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<tr>
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<td>0.82</td>
<td>1.46</td>
</tr>
<tr>
<td>10</td>
<td>2.34</td>
<td>0.97</td>
</tr>
</tbody>
</table>

b. Estimation of parameters

We now assume that \( R(t) \) is observed during \( n \) days. Then \( R_0 \) is estimated by

\[
R_0^* = \frac{h(24n)}{24n},
\]

i.e., the total amount of precipitation during the period of observation divided by the length of the period. The parameter \( \Gamma \), however, is somewhat more difficult to estimate. \( \Gamma \) is closely related to the spectral density of \( R(t) \) and thus estimation of \( \Gamma \) is related to spectral analysis.

Let \( h_1, h_2, \ldots, h_n \) be the daily amounts of precipitation. The values \( h_1, h_2, \ldots, h_n \) may be regarded as a part of a stationary time series \( h_k \), \( k = 0, \pm 1, \ldots \). This time series is related to \( R(t) \) and \( h(t) \) by

\[
h_k = \int_{t_{k-1}}^{t_k} R(t) dt = h(24k) - h(24(k - 1))
\]

and thus \( E(h_k) = 24R_0 \). If we define the covariances \( \gamma_k \) by

\[
\gamma_k = \text{cov}(h_j, h_{j+k})
\]

and the natural estimates \( \gamma_k^* \) by

\[
\gamma_k^* = \frac{1}{n-k} \sum_{j=1}^{n-k} (h_j - \bar{h})(h_{j+k} - \bar{h}),
\]

where

\[
\bar{h} = \frac{1}{n} \sum_{j=1}^{n} h_j = 24R_0^*.
\]

From the data used in RG, which will be discussed in the Sections 4b and 4c, we get the estimates given in Table 1. In order to get useful estimates of \( \Gamma \), we assume \( \gamma_k \) to be approximately zero for \( k > m_n \). Both Table 1 and general knowledge about Swedish weather indicates that \( m_n = 5 \) is reasonable. Since

\[
\Gamma = \frac{1}{24} \sum_{k=-m_n}^{m_n} \gamma_k = \frac{1}{24} \sum_{k=-m_n}^{m_n} \gamma_k^*,
\]

a natural estimate \( \hat{\Gamma} \) of \( \Gamma \) is

\[
\hat{\Gamma} = \frac{1}{24} \sum_{k=-m_n}^{m_n} \gamma_k^*.
\]

Since, however,

\[
E(\hat{\Gamma}) = \frac{1}{24} \sum_{k=-m_n}^{m_n} E(\gamma_k^*) = \frac{1}{24} \sum_{k=-m_n}^{m_n} \left( \gamma_k - \frac{\Gamma}{n} \right) = \frac{1}{24} \Gamma \left( 1 - \frac{2m_n + 1}{n} \right),
\]

we are led to

\[
\Gamma^* = \frac{1}{24} \left( \frac{n}{n - 2m_n - 1} \sum_{k=-m_n}^{m_n} \gamma_k^* \right).
\]
A stringent derivation of this modification of $\Gamma$, due to the fact that $E(h_k)$ is estimated, is given by Hannan (1960, pp. 133–137). Applying this estimate to our data, we get the $\Gamma^*$ values shown in Table 2 for different values of $m_n$. From Table 2 it is seen that $\Gamma^* = 1.2$ is a reasonable estimate, for both summer and winter for this particular period.

Our proposed estimate is closely related to the "truncated" estimate in spectral analysis. Under certain regularity assumptions on $R(t)$ it follows from Grenander and Rosenblatt (1956, p. 148) that

$$\sqrt{\text{var}(\Gamma^*)} \approx 2\sqrt{\frac{m_n}{n}}, \quad (3.13)$$

when $n$ and $m_n$ tend to infinity in such a way that $m_n/n$ tends to zero. An uncritical use of this formula yields, with $n = 182$ and $m_n = 5$,

$$\sqrt{\text{var}(\Gamma^*)} = 0.33\Gamma. \quad (3.14)$$

which indicates a considerable statistical uncertainty. In order to get statistically sound estimates of $\Gamma$ we must obtain observations from several years. Then it seems natural to form an estimate of $\Gamma$ for each year and to use the average of those estimates.

c. An example

In order to illustrate the approximation we consider the following simple model. Let the daily amounts of precipitation $h_1, \ldots, h_n$ be independent random variables with

$$h_n = \begin{cases} 0 & \text{with probability } 0.6 \\ 2 & \text{with probability } 0.35 \\ 25 & \text{with probability } 0.05. \end{cases}$$

During a day $R(t)$ is assumed to be constant, i.e., $R(t) = h(0)t/24$ for all $t$.

We have $R_0 = 0.081$ and $\Gamma = 1.20$. This model is not meant to be realistic, but numerically it agrees rather well with the winter data discussed in the previous section. From the renewal model with $\tau_d = \sigma_d^2 = 0$ and $\bar{T}_p = 24$, we get an exact expression for $E(T_n)$. In Fig. 1 we compare $E(T_n)$ thus calculated with the long-life approximation [Eq. (3.4)]. We shall return to this example in Section 4.

4. Comparisons between the models

The main difficulty in a comparison between the different models is that no correct values of $E(T_n)$ are available. In order to overcome this difficulty we shall start this chapter by introducing a model, which is independent of any specific assumption about $R(t)$. This model, here referred to as the empirical model, will then be used as a reference in the comparisons.

a. The empirical model

The only assumption we need to make here is the proportionality between scavenging intensity and precipitation intensity. Thus, the estimates are based on a mixture of empirical data and theoretical assumptions. This mixture has motivated the name empirical model.

We assume that the precipitation intensity $R(t)$ is observed at a station during a time interval $[0,T]$. The idea is to consider the time spent in the atmosphere, by a particle entering at an arbitrary time in $[0,T]$, when the precipitation varies according to the observations. Since the precipitation is observed at a fixed station, all results are strictly applicable only to situations, which can be described by Eulerian data.

We now define $G_s(t)$ as the probability that a particle entering the atmosphere at time $s$, $s \in [0,T]$, has a longer residence time than $t$ and define $E_s$ as the mean of that time. Then we have

$$G_s(t) = \exp\left[-a \int_t^{s+t} R(x) \, dx\right], \quad (4.1)$$
was done in order to make the comparisons with the different models more accurate. The value of $E_T$ was taken from the Markov model.

b. Data base for comparison

For the comparison presented in Section 4c we have used the same precipitation data as in RG, namely, one year (1966) of automatically recorded (Slettenmark instrument) data from the station Västerbroviadukten in Stockholm. The temporal resolution of this data is 2 h. In view of the pronounced seasonal variations we have separated the data into two seasons, winter from October–March and summer from April–September. Admittedly, this is a very limited set of data and numerical estimates based upon it will necessarily be uncertain. The reason for sticking to this set in the comparison is mainly pedagogical.

For the comparison in Section 4d between the classical model, the empirical model and the long-life approximation (Figs. 5–8) daily precipitation values from Stockholm for the period 1970–72 have been used. In Section 5 we present estimates based on more extensive precipitation data from different parts of Sweden.

c. Numerical comparison

We now consider the summer and winter data from 1966. We have modified the endpoints in the interval under observation so that each observation consist of $N$ successive values of $\tilde{T}_d, \tilde{T}_p$ and $M$. More precisely, we have observations $\tilde{T}_{d,1}, \tilde{T}_{p,1}, M_1, \tilde{T}_{d,2}, \tilde{T}_{p,2}, M_2, \ldots, \tilde{T}_{d,N}, \tilde{T}_{p,N}, M_N$. In Table 3 we give some characteristics of the two series of observations.

If the length of the dry periods ($\tilde{T}_d$) were exponentially distributed, then one would have $\sigma_d/\tau_d = 1$. For our observation, however, $\sigma_d/\tau_d$ equals 1.73 for the winter period and 1.93 for the summer period. This indicates that an assumption about exponentially distributed dry periods, as made in the Markov model, need not to be quite accurate. In RG, where the Markov model was originally discussed, unfortunately, we estimated $\tau_d$ and $\tau_p$ in a different way and reached very different values (summer: $\tau_d = 92$ and $\tau_p = 4$; winter, $\tau_d = 40$ and $\tau_p = 9$). The difference is due to the fact that in RG we estimated the time from an arbitrary point in a certain period until the end of that period. For exponentially distributed variables this method of estimation is also possible. The values of $\tau_d$ and $\tau_p$, estimated in that way, gave values of $E(T_d)$, which happened to agree quite well with those estimated with the empirical model. This fact may not have been a coincidence as can be seen from the following consideration.

In the renewal model we assume that $\lambda = \lambda_p$ and
that \( \hat{T}_p \) is exponentially distributed Eq. (2.12) reduces to

\[
E(T) = \frac{\sigma_d^2 + \tau_d^2}{2(\tau_d + \tau_p)} + \frac{\tau_d + \tau_p}{\tau_p\lambda_p}.
\]

(4.6)

A comparison with the corresponding expression for the Markov model (2.11) shows a difference only in the first term. If \( \tau_p \) is small compared to \( \tau_d \) the first terms are approximately \( \tau_d \) and \( (\sigma_d^2 + \tau_d^2)/2\tau_d \), respectively. The latter expression is in fact the mean time from an arbitrary point in a dry period until the period ends, and this is what we estimated in RG. If the "renewal" model is used in this special case [Eq. (4.6)] it, however, is better to estimate \( \tau_d \), \( \tau_p \) and \( \sigma_p^2 \) in a natural way.

We may also note in Table 3 that \( \tau_p \) is much smaller than \( \tau_d \). Therefore, those models which disregard the length of the precipitation periods give results which are similar to those derived from models which take these periods explicitly in account. In order to keep this comparison reasonably short we shall not treat such short rain models here.

Consider first the long-life approximation treated in Section 3. In this case it is unnatural to exclude anything in the endpoints of the interval under observation since the estimates of the parameters are based on daily amounts of precipitation only. From this unmodified data we get \( R_0 = 0.060 \) for the summer and \( R_0 = 0.082 \) for the winter. As seen from Section 3b we estimated \( \lambda \) by 1.2 in both periods.

In Figs. 2 and 3 we present estimates of \( E(T_d) \) obtained from the various models as functions of \( 1/a \). The main impression from these figures is probably that they consist of rather parallel curves at different levels. The fact that the classical model yields small values of \( E(T_d) \) is natural, since any random variation in the scavenging intensity increases the mean residence time (Grandell and Rodhe, 1978, p. 248). The very specific assumptions in the Markov model also may explain its position quite far away from the empirical model. More interesting is the great, and very similar, difference between the empirical model and the long-life approximation. There seems to be three possible explanations of this difference:

1) Some systematic error in the methods of estimation.
2) Poor accuracy in the long-life approximation.
3) Great random variation in the estimates.

In order to judge these explanations further investigations are necessary. In Section 4d we shall perform such investigations based on daily precipitation data.

d. Numerical comparisons based on daily precipitation data

Let \( h_1, h_2, \ldots, h_n \) be the amounts of precipitation during periods of length \( \Delta \). If \( \Delta = 24 \) h, we talk about daily data. As in Section 3c, we treat the precipitation intensity as constant during each period.

The estimates of the parameters in the classical model and in the approximation are based on this kind of discrete data. However, the parameters in the Markov model and in the renewal model seem difficult to estimate, unless \( \Delta \) is very small. This problem is discussed by Alexander (1980) for the Markov model.

Now we shall consider the empirical model. Although the discussion in Section 4a applies to this
kind of data, we shall give an algorithm suited for computer calculations. Let \( E_k(E_k) \) be the mean residence time for a particle entering the atmosphere at the beginning of (arbitrarily during) period \( k \).

Then if \( h_k = 0 \),

\[
E_k = \Delta + E_{k+1}^b,
\]

and if \( h_k > 0 \)

\[
E_k = \frac{\Delta}{ah_k} (1 - e^{-ah_k}) + E_{k+1}^b e^{-ah_k},
\]

The value \( E_{n+1} \) is taken from the classical model, i.e., \( E_{n+1} = (aR_0^n)^{-1} \). The mean residence time is now estimated by \( 1/n \) \( \sum_{k=1}^{n} E_k \). A natural question is now how much the choice of \( \Delta \) influences the result. In Fig. 4 we compare estimates based on the detailed data used in Section 4c with estimates based on daily data.

Fig. 4 indicates that daily data is good enough for our purposes. Only if very large values of \( a \) are important more detailed data would be required.

For the comparison between the models we therefore use daily precipitation data from Stockholm for the years 1970-72. Since the parameters in the Markov model and in the renewal model are difficult to estimate from daily data we restrict the comparison to the “classical” model, the empirical model and the long-life approximation. In Table 4 we give characteristics of the different sets of data. Figs. 5–8 show the results of the comparison. From Fig. 7 it is seen that for the summer 1971 the agreement between the empirical model and the long-life approximation is very bad. The reason is probably that during that period 55% of the total amount of

---

**Table 4. Estimates of parameters used in the comparisons shown in Figs. 5–8.**

<table>
<thead>
<tr>
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</thead>
<tbody>
<tr>
<td>( n )</td>
<td>183</td>
<td>182</td>
<td>183</td>
<td>183</td>
</tr>
<tr>
<td>( R_0^* )</td>
<td>0.0508</td>
<td>0.0531</td>
<td>0.0594</td>
<td>0.0523</td>
</tr>
<tr>
<td>( \gamma_k^*; k = )</td>
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<tr>
<td>0</td>
<td>7.46</td>
<td>5.40</td>
<td>16.00</td>
<td>5.72</td>
</tr>
<tr>
<td>1</td>
<td>1.01</td>
<td>1.03</td>
<td>5.22</td>
<td>0.69</td>
</tr>
<tr>
<td>2</td>
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<td>-0.28</td>
<td>1.24</td>
<td>0.89</td>
</tr>
<tr>
<td>3</td>
<td>0.64</td>
<td>0.38</td>
<td>3.73</td>
<td>0.17</td>
</tr>
<tr>
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<td>5.86</td>
<td>0.46</td>
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<td>-0.10</td>
<td>0.54</td>
<td>-0.30</td>
</tr>
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<td>-0.50</td>
<td>-0.70</td>
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<tr>
<td>7</td>
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<td>-0.16</td>
<td>-0.55</td>
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<td>0.16</td>
<td>-0.03</td>
<td>-0.64</td>
</tr>
<tr>
<td>9</td>
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<td>-0.38</td>
<td>-1.17</td>
<td>-0.07</td>
</tr>
<tr>
<td>10</td>
<td>-0.14</td>
<td>0.02</td>
<td>-0.81</td>
<td>-0.49</td>
</tr>
<tr>
<td>( \Gamma^*; m_n = )</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>0.31</td>
<td>0.23</td>
<td>0.67</td>
<td>0.24</td>
</tr>
<tr>
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</tr>
<tr>
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<td>0.55</td>
<td>0.30</td>
<td>1.24</td>
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</tr>
<tr>
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<tr>
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<tr>
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<td>7</td>
<td>0.77</td>
<td>0.34</td>
<td>2.12</td>
<td>0.41</td>
</tr>
</tbody>
</table>
precipitation fell in two different weeks separated by three weeks of more dry days. As seen from Table 4 no specific value of \( m_n \) was natural to choose.

We concluded Section 4.3 with three possible explanations of the difference between the "empirical" model and the long-life approximation. Several simulations using the model described in Section 3c with \( \Delta = 24 \, \text{h} \) and \( n = 182 \) indicate that systematic errors in the estimation methods are negligible. The Figs. 2, 3 and 5–8 as well as these simulations indicate that the difference between the empirical model and the long-life approximation varies much.

The accuracy in the long-life approximation is difficult to describe, in general. Since the long-life approximation is exact in the classical and the Markov models and since Fig. 1 indicates a fair agreement for \( 1/a > 5 \) (that is a residence time in precipitation larger than 5 hours in precipitation of 1 mm h\(^{-1}\)) we believe that the accuracy is reasonably good for \( 1/a \approx 5 \). Because of the year to year variation we recommend a use of the long-life approximation only if several years of data are used in the estimation.

For small values of \( 1/a \) seems more natural to approximate \( E(T_n) \) with a straight line of the form \( K + (a R_0)^{-1} \), where \( K \) is the value of the residence time calculated from the "empirical" model with \( 1/a = 0 \). In the case where high-resolution data is used this agrees with Eq. (4.6).

5. Climatological estimates of \( R_0 \) and \( \Gamma \)

We have used 11 years of daily precipitation data from 30 meteorological stations in Sweden to estimate the quantities \( R_0 \) and \( \Gamma \) according to the outline given in Section 3b. We recall that \( R_0 \), the average rate of precipitation, and \( \Gamma \), a measure of the variability of the daily rainfall amounts, are parameters needed for an estimate of the average residence time of long-lived particles subjected to removal by precipitation scavenging (cf. Section 3).

For each station, the precipitation data was divided into 22 separate series each covering a 6-month period: 1 April–30 September (summer) and 1 October–31 March (winter). The maximum time lag (\( m_n \)) was set at five days for all stations and for all periods. Finally, arithmetic mean values were calcu-
eral lower than the summer values, indicating a higher steadiness in winter precipitation. For both seasons there is a slight tendency for high values to occur in the southwest parts of the country. This is also where the highest precipitation amounts are found. The resulting data pairs \((R_0, \Gamma)\) are plotted in Fig. 10. Generally, \(\Gamma\) increases with increasing \(R_0\). For the winter seasons the following linear relation gives the best fit (correlation coefficient = 0.94)

\[
\Gamma = 0.04R_0 - 0.7.
\]

Here \(R_0\) is measured in kg m\(^{-2}\) h\(^{-1}\) and \(\Gamma\) in kg m\(^{-2}\) m\(^{-4}\) h\(^{-1}\). For the summer seasons no clear relation is evident.

In Table 5 we show average values and standard deviations of the magnitude of the second term on the right-hand side of Eq. (3.4) \((\Gamma/2R_0^2)\). This term represents part of the average residence time—in the case of long-lived particles—due to the temporal variability of the precipitation.

It is possible that the estimates presented here are roughly representative also for other areas with a similar climate. However, no extrapolations can be made to other climatic regimes.

6. Transformation to a Lagrangian system

The precipitation data, which has been used so far in this study to estimate the scavenging parameters, has been based on measurements taken at fixed points, i.e., Eulerian type data. Since we are considering the fate of individual particles or parcels of air such data are only applicable in a hypothetical situation when the particles remain fixed in space while the precipitating clouds move by. To be correct the parameters \(R, \tau_d, \tau_p, \Gamma\), etc., should all be evaluated in a system moving with the particles, i.e., a Lagrangian system. Direct measurements in a Lagrangian sense of these quantities are virtually impossible to obtain: they would have to entail precipitation gauges mounted on free floating balloons which could be made to follow both horizontal and vertical air movements.

A possible way of estimating the precipitation parameters in a Lagrangian system is to use observations of wind (and/or pressure) to calculate air trajectories and to use areal mappings of precipitation at regular time intervals to associate trajectory positions with precipitation events. However, such an approach has several shortcomings. First, the accuracy with which air trajectories can be calculated is not very high. This is particularly true for air parcels experiencing precipitation since precipitating clouds are associated with complex circulation patterns which are difficult to estimate. Second, the problem of interpolation of precipitation data from observation stations to trajectory positions may introduce a significant uncertainty, particularly

Fig. 9. Geographical distribution of estimates of \(\Gamma\). Based on 11 years of daily precipitation data. Numbers above station names refer to summer season, numbers below to winter.
in situations where the precipitating clouds have a small areal extent (convective precipitation).

In the following subsections we will go through some very much simplified arguments and try to derive qualitative relations between \( \tau_{a}^{h} \) and \( \tau_{a}^{f} \) and their Eulerian counterparts.

a. Frontal precipitation

We make the following two basic assumptions:

(i) The precipitation system remains as an entity during a time period which is long compared to the time scale characterizing the scavenging \( (E(T)) \).

(ii) The precipitation systems move with a constant speed \( V_{r} \) which is related to the constant speed of the particles \( V \) by a constant factor \( \beta \), i.e., \( V_{r} = \beta V \).

Of course, both these assumptions represent considerable simplifications of the real situation. If we let \( D \) denote the distance between two frontal precipitation systems, for simplicity taken to be constant, it follows that

\[
\tau_{a}^{f} = \frac{D}{V_{r}} = \frac{D}{\beta V},
\]

\[
\tau_{a}^{h} = \frac{D}{V - V_{r}} = \frac{D}{V|1 - \beta|}.
\]

so that

\[
\tau_{a}^{h} = \tau_{a}^{f} \frac{\beta}{|1 - \beta|}.
\]

We now look at a few special cases of this relation

1) \( \beta \rightarrow 0 \). This case, with a non-moving precipitation system, corresponds to the situation with orographic precipitation, i.e., rainclouds that are caused by updrafts produced by mountain ridges and therefore not moving. Formally, we have

\[
\tau_{a}^{h} = \frac{D}{V}; \quad \tau_{a}^{f} \rightarrow \infty.
\]

A better approximation for \( \tau_{a}^{h} \) in this case would be

\( \tau_{a}^{h} \sim L/V \), where \( L \) is the distance from the source area to the nearest mountain in the downwind direction.

2) \( \beta \rightarrow 1 \). The clouds are completely passive and move with the horizontal speed of the air, i.e.,

| Table 5. Climatological estimates of the term \( \Gamma/2R_{s} \) for a period of 11 years at 30 Swedish stations. Units = h. |
|-------------|--------|--------|
|             | Average | Standard deviation |
| Summer      | 127    | 23     |
| Winter      | 98     | 13     |

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\[
\tau_d^b \rightarrow \infty; \quad \tau_d^b = \frac{D}{V}.
\] (6.5)

This is an unrealistic situation which implies that only those particles that are emitted during a precipitation period will be scavenged.

3) \( \beta \rightarrow \infty \). The particles do not move relative to the ground but the precipitation systems move by

\[
\tau_d^b = \tau_d^b.
\] (6.6)

If this were true, Eulerian statistics would be adequate.

4) \( 0 < \beta < 1 \). This is the situation, believed to be the most realistic, when the particles move in the same direction but at a greater speed compared to the precipitation systems. The physical interpretation is that the warm air at a warm front is forced to rise above the cold air thereby reducing the speed at which the front (and the precipitation system) is advected.

\( \tau_d^b \) will be \( \cong \tau_d^a \) depending on whether \( \beta \cong 0.5 \). Meteorological data relating to the movements of warm fronts seems to indicate that warm fronts normally move with a speed about \( \frac{5}{2} \) of that of the surface air in the warm sector. Such a \( \beta \) value would imply that \( \tau_d^b \approx 2 \tau_d^a \).

b. Convective precipitation

In a meteorological situation with widespread convective precipitation occurring the length of the average dry period, \( \tau_d^b \) is of the order of a few hours. It seems very likely that the corresponding Lagrangian time scale \( \tau_d^a \) is of comparable magnitude. In the extreme case with very short duration of the convective clouds and with the clouds forming and disappearing at random locations in the area, one would actually expect \( \tau_d^b \) and \( \tau_d^b \) to be equal.

In any case, both \( \tau_d^b \) and \( \tau_d^b \) evaluated in a situation with convective precipitation are short compared to long-term climatological estimates of the same quantities. Therefore the important quantity to consider should be the time it takes to encounter a region with convective precipitation. Let us look at the following two simple situations:

1) Convective clouds producing precipitation occur over a widespread area with a pronounced daily cycle. Normally the precipitation would be concentrated to the afternoon hours. This may be typical for certain continental areas in the tropical belt. Under such conditions one would expect \( \tau_d^b \) to be equal to \( \tau_d^b \) both being of the order of 20 h.

2) The area of convective clouds is associated with the outbreak over a warm surface of cold air behind a midlatitude cyclone. Since the movement of such areas is coupled to the movement of frontal zones and the corresponding precipitation systems, this situation may be similar to the one described in Section 6a.

c. Concluding remarks

In our discussion about the relation between Eulerian and Lagrangian frames of reference we have so far only considered the parameter \( \tau_d^a \). No firm conclusion could be drawn based on simple theoretical arguments about the relative magnitude of \( \tau_d^a \) and \( \tau_d^b \), but there are some weak indications that one might expect \( \tau_d^b \) to be longer than \( \tau_d^b \). This question will probably have to be resolved through further detailed calculations of surface air trajectories and systematic comparisons with precipitation data. Such a study, based on trajectory and precipitation data from Europe, is being undertaken and will be reported separately (Hamrud et al., 1981). The study indicates that the difference between \( \tau_d^a \) and \( \tau_d^b \) is rather small.

The parameters \( \tau_d^b \), \( R_0 \) and \( \Gamma \) are much more difficult to evaluate directly in a Lagrangian frame, mainly because true particle trajectories in or near precipitating system have a vertical component which is very hard to estimate. One may argue that \( R_d^b \) would be greater than \( R_d^b \) since particle trajectories in view of the horizontal convergence associated with precipitation systems should have a tendency to hit such areas more often than "dry areas".

7. Parameterization of precipitation scavenging in transport models

The approach to parameterization of precipitation scavenging in transport models will depend, among other things, on the space and time scales considered. For example, if one is only interested in the fate of an instantaneous emission over a travel time of a few tens-of-minutes one may neglect the probability of a change in weather affecting the emitted pollutant and simply assume that the weather at the time of emission remains unchanged during the period considered. For longer travel times and for longer averaging periods changes in weather become important.

We have chosen to divide this section into three sections according to length of travel time considered:

- \( t < 1 \) h, i.e., short compared to the average length of the precipitation periods \( \tau_d^b \). Chimney-plume models and most urban diffusion models fall into this category.
- \( t \approx \text{a few days}, \) i.e., the time period over which reasonably accurate trajectories may be computed. This category would include mesoscale and regional scale (long-range transport) models.
- \( t > \text{a few days}, \) corresponding to continental, hemispheric and global models.
a. Short-range transport models

For modeling of individual situations the removal probability ($\lambda_p$ or $\lambda_d$) may be taken as a constant determined by the weather at the time of the emission and the problem (in case of precipitation) is reduced to determining a realistic $\lambda$ value.

For long-term average conditions the only meteorological parameter of significance (except those which affect the value of $\lambda$) is the frequency of occurrence of precipitation $f$ possibly distributed between the different classes of wind direction and stability considered in the model. The problem of the long-term distribution on the ground of pollutants emitted at a point source has been treated recently by Rodhe (1980). He showed that for travel times less than $\approx 1$ h the amount deposited by precipitation could be estimated by calculating the emission during the precipitation periods only and applying a decay rate equal to $\lambda_p$. For travel times up to several hours changes in weather during the transport has to be considered and no simple exponential decay can be assumed. In such situations the use of the explicit frequency function for residence times of the Markov model provides a possible approach (Rodhe, 1980).

b. Long-range transport

If the transport model is based on calculation of air trajectories it is, in principle, possible to keep track of the weather along each trajectory and apply an appropriate, deterministically varying, decay rate (see, e.g., Szepesi, 1978). Particularly when dealing with the fate of an instantaneous emission or individual air pollution episodes such an approach would seem to be reasonable. For long-term average conditions this method becomes impractical because of the large amount of meteorological data involved. Here statistical methods represent a possible alternative.

A very simple way to model the scavenging in a statistical sense is to apply an average removal rate which is constant during the transport (see, e.g., Bolin and Persson, 1975). Such a rate must include implicitly information on $\lambda_p$ as well as of the distribution of precipitation and dry periods. A zero-order estimate of this decay rate is $f\lambda_p$ or $aR_0$ which corresponds to the classical model (cf. Sections 2 and 3). Such a formulation has been employed by Scriven and Fisher (1975), for example. This is a good approximation only if the precipitation is actually spread out as a constant light intensity all the time. As we have shown earlier the existence of finite dry and precipitation periods will extend the average residence time of the particles in the atmosphere beyond $(f\lambda_p)^{-1}$. Another consequence is that the distribution of residence times is no longer exponential. Thus one may not uncritically apply an average removal rate given by the inverse of the time scale $E(T_d)$ taken from Eq. (2.17) or (3.4). The examples given by Rodhe (1980) show that this may lead to substantial differences compared to a correct application of the Markov model at least for travel times of only a few hours. For longer travel times this difference becomes less significant. To be honest, it should be pointed out that the uncertainties in the estimate of $a$ or $\lambda_p$ are so large that differences between the various models in many cases are of limited significance.

Fisher (1975, 1978) used a parameterization scheme based directly on the Markov model described in Section 2b in his modeling of the long-term distribution of sulfur pollution over northern Europe. He made explicit use of the distribution function for residence times [Eq. (2.6)] and generalized it to allow for variations in the parameters $\tau_p$ and $\tau_d$ according to climatic differences between different regions. The separation into regions in this case was done in a schematic way and based on rainfall totals only. A much more elaborate mapping of the parameters $\tau_p$ and $\tau_d$ was made for eastern United States by Henmi and Reiter (1978) but no attempt has been made to include such variable fields in a transport model.

In the model used in the OECD project on long range transport (Eliassen, 1978) precipitation scavenging was treated in a semi-empirical manner, which avoids many of the shortcomings of the previously mentioned methods. On the other hand, extrapolations to other regions and to future conditions can not be readily made by such a method. In this model a constant loss rate was employed for sulfate removal (including dry deposition and precipitation scavenging) whereas for SO$_2$ separate decay rates were used for dry and precipitation periods. The forecast concentration of sulfate in precipitation was not explicitly related to these removal rates but rather estimated from an empirical relation between model values of the concentration of total sulfur in the air and observed values of sulfate in precipitation.

c. Global-scale transport

In this case of travel times of weeks and months it is out of the question to keep track of the actual weather affecting an air parcel as it moves along and a statistical approach to the parameterization of scavenging becomes a necessity. At such travel times the pollutants are also no longer confined to the boundary layer and the question of the vertical distribution of the scavenging rate becomes an additional difficulty.

A simple approach in the case of tropospheric transport is to apply average removal rates determined by studies of atmospheric tracers such as radionuclides in particulate form. This seems to have
been the basic philosophy underlying many of the removal rate assumptions employed in one- and two-dimensional dispersion models (see, e.g., Davidson et al., 1966).

Attempts have also been made to relate the removal rate to the frequency of occurrence of clouds and/or precipitation. It is then assumed that the removal rate is given by \( f \lambda_m \) with \( f \) representing the "probability of cloud formation." According to the considerations presented earlier in this paper such an assumption is likely to overestimate the rate of removal.

In the two-dimensional (height/latitude) transport model described by Isaksen and Rodhe\(^2\) subcloud precipitation scavenging was treated with a removal rate given by \( [E(T)]^{-1} \) with \( E(T) \) taken from the Markov model and with the parameters \( \tau_u \) and \( \tau_d \) estimated from climatological data. For inclusion scavenging the rate of removal in this model was made proportional to the rate of formation of precipitation in the different height intervals. The latter rate, in turn, was estimated from energy balance considerations. The absolute values of the removal rates deduced in this manner may be as uncertain as those derived from tracer considerations. One advantage with the former though is that the assumptions about spatial variations of the removal rates are based on physical principles.

8. Conclusions and recommendations

Before stating any conclusions it may be appropriate to reiterate some major limitations of the present study.

1) We have not discussed the problem of estimating the scavenging coefficients \( \lambda_u \) and \( \lambda_d \), i.e., the rates of removal during precipitation and dry periods, respectively. Before any of the models discussed in this paper is applied to a specific pollutant it is necessary to estimate realistic values of these rates.

2) The approximate model for long-lived particles is based on the assumption about a proportionality between the scavenging rate and the precipitation rate. If this assumption is not valid, the results derived with the aid of the long-life approximation are not reliable. In particular, if low precipitation rates in reality do not correspond to proportionally low scavenging rates, the average residence time \( [E(T_m)] \) may be overestimated with the long-life approximation.

3) The derived residence times represent statistical averages over long time periods and may not uncritically be applied to specific periods during which the weather may have deviated significantly from average conditions. In situations when the actual weather is known along a trajectory, it is better to treat the scavenging as a deterministic rather than as a stochastic process.

4) In view of the large year-to-year variability of precipitation statistics, the data used for the comparison between the different models was very limited, indeed.

5) The parameters \( R_0 \) and \( \Gamma \), which enter into the expression for the average residence time in the approximate model, have been estimated from climatological data obtained from some Swedish stations. These estimates may not readily be extrapolated to other climatic regions.

In spite of these limitations we believe that the following tentative conclusions can be drawn from our review of previously published models:

1) The classical model, where no consideration is given to the variability of precipitation, grossly underestimates the average residence time, particularly in situations with high values of the scavenging coefficient.

2) At least in climates like that in northern Europe, the Markov model also seems to underestimate the residence time, although to a lesser degree than the classical method.

3) The renewal model seems to provide a fair estimate of the residence time but it is probably too complicated for most practical applications.

4) In climates where the average length of the precipitation periods is much shorter than the average length of the dry periods, an assumption about infinitely short precipitation periods does not significantly add to the uncertainty in the estimates of the residence time in the Markov and the renewal models.

We have formulated an approximate model which is simple enough for practical applications and which seems to give a reasonable estimate of the average residence time. This model is based on the assumption of a proportionality between scavenging rate and precipitation rate. It is only valid for relatively low values of the proportionality constant. A practical limit of its applicability may be set at \( \lambda_m \leq (5 \text{ h}^{-1}) \) for a precipitation rate of \( 1 \text{ mm h}^{-1} \).

Estimation of the parameters \( R_0 \) and \( \Gamma \) used in this model requires relatively long records of precipitation, of the order of 10 years. On the other hand, the temporal resolution of the precipitation data need not be better than 24 h. This means that standard climatological data may be used for this purpose.

We have used climatological data from several Swedish stations to estimate the parameters $R_0$ and $\Gamma$. The difference in the estimate of the average residence time between the long-life approximation and the classical model turned out to be of the order of 100 h for the winter seasons and 125 h for the summer seasons. For compounds with a very high value of the scavenging coefficient $\lambda_p$ [$\lambda_p > (5 \text{ h})^{-1}$ at a precipitation rate of 1 mm h$^{-1}$] we recommend the use of the special case of the "renewal" model discussed at the end of Section 4d.

The largest uncertainty associated with all these estimates is probably introduced by the fact that the parameters have been estimated using precipitation data from fixed points (Eulerian data) whereas the models in reality require information about the weather experienced by the particles as they are carried along by the winds (Lagrangian data). It follows from the discussion in Section 6 that it is difficult to derive even a qualitative relation between precipitation parameters estimated in an Eulerian and a Lagrangian system. It seems to us as if little further progress can be made in the estimate of statistical precipitation scavenging rates and its parameterization in transport models before the problem of how to obtain Lagrangian precipitation statistics has been resolved.

APPENDIX

List of Symbols

<table>
<thead>
<tr>
<th>Definitions</th>
<th>Dimensions</th>
</tr>
</thead>
<tbody>
<tr>
<td>$R^L(t)$</td>
<td>intensity of precipitation in a Lagrangian system</td>
</tr>
<tr>
<td>$R^E(t)$</td>
<td>intensity of precipitation in an Eulerian system</td>
</tr>
<tr>
<td>$h(t)$</td>
<td>$\int_0^t R(\xi) d\xi$</td>
</tr>
<tr>
<td>$\lambda(t)$</td>
<td>scavenging coefficient at time $t$</td>
</tr>
<tr>
<td>$\lambda_p$</td>
<td>scavenging coefficient during precipitation</td>
</tr>
<tr>
<td>$\lambda_d$</td>
<td>scavenging coefficient during dry period</td>
</tr>
<tr>
<td>$\lambda_o$</td>
<td>scavenging coefficient averaged over precipitation and dry periods</td>
</tr>
<tr>
<td>$a$</td>
<td>ratio of scavenging coefficient and precipitation intensity</td>
</tr>
<tr>
<td>$T_a$</td>
<td>residence time in the atmosphere for an individual particle</td>
</tr>
<tr>
<td>$\tau_d$</td>
<td>length of dry period</td>
</tr>
<tr>
<td>$\tau_p$</td>
<td>length and precipitation intensity of a precipitation period</td>
</tr>
<tr>
<td>$\bar{M}$</td>
<td>total amount of precipitation during a precipitation period</td>
</tr>
<tr>
<td>$G_a(t)$</td>
<td>distribution function for residence times</td>
</tr>
<tr>
<td>$E(T_a)$</td>
<td>mean residence time</td>
</tr>
<tr>
<td>$\tau_d$</td>
<td>mean length of dry periods</td>
</tr>
<tr>
<td>$\sigma_d^2$</td>
<td>variance of length of dry periods</td>
</tr>
<tr>
<td>$\Gamma$</td>
<td>a measure of the variability of rainfall intensity</td>
</tr>
</tbody>
</table>

REFERENCES


Maul, P. R., 1978: Preliminary estimates of the washout coefficient for sulphur dioxide using data from an East Midlands ground level monitoring network. *Atmos. Environ.*, 12, 2515–2517.


