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Simple stochastic models for the sources and sinks of two aerosol types

By M. B. BAKER, *Department of Civil Engineering, Division of Water and Air Resources, University of Washington, Seattle, Washington 98195, U.S.A.*, HALSTEAD HARRISON, *Department of Atmospheric Sciences, University of Washington*, JOHN VINELLI, *Department of Civil Engineering, Division of Water and Air Resources, University of Washington*, and K. B. ERICKSON, *Department of Mathematics, University of Washington*

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ABSTRACT

The statistical properties of two time-dependent and steady-state aerosol concentration distributions are derived from simple stochastic models of the particle sources and sinks. The analysis is applied to statistics of area averaged aerosol concentrations in two examples: (a) anthropogenic sulfate particles, for which the source is assumed constant in time, and removal is by precipitation at random times, and (b) windblown dust, for which we assume the sources are a constant background plus randomly occurring dust storms, and the sinks are time independent advection and dry deposition. The models are solved both analytically and by numerical simulation. For both models the time-lagged autocorrelation function of the pollutant is a simple exponential, and the ratio of the steady-state variance to the squared mean concentration varies inversely with the effective lifetime.

Application of data from the World Meteorological Organization to model (a) shows that the expected lifetimes for sulfates are between 50 and 175 h, and the deposition distances are between 800 and 2000 km.

1. Introduction

One of the central problems in atmospheric research is the interpretation of the fluctuations of measured variables in terms of their governing processes. Measurements of atmospheric aerosol concentrations (or optical parameters which depend on them) exhibit variability in time and space which results from fluctuations in the source, sink, transformation, and transport processes undergone by the particles prior to sampling. This set of interrelated processes, called the aerosol life cycle, is far too complex for detailed analytic description. Numerical models which integrate the dynamical equations for specified initial and boundary conditions are unwieldly, expensive, and often ineffective for obtaining insight into the mechanisms involved. We present here an alternative analytic approach to the study of aerosol life cycles in which the component parts are examined individually and considered to be stochastic. That is, we model or measure the variability or "noise" in the cycle parameters without describing the details of the mechanisms producing the variability. The goal of our work is to present a framework for investigation of the relationships between the statistical properties of individual cycle processes and those of the atmospheric aerosol concentrations.

This study follows the work of Junge (1974) and Gibbs and Slinn (1973), who predicted the variance in trace gas concentrations for general classes of life cycles, and that of Rodhe and Grandell (1972), who computed the lifetimes for individual particles assuming the sink to be rainout, under the assumption that the rain process is Markovian. In the present work we have attempted to construct simple statistical models for the sink and source processes of two identifiable aerosol types, and to compare the results with data.

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We begin by considering the conservation equation for the concentration $c(x, y, z, t)$ of an atmospheric constituent:

$$\frac{\partial c}{\partial t} = -\nabla \cdot (\mathbf{u}c) + q - s \tag{1}$$

where $\mathbf{u}$ (m/s) is the wind velocity vector, $q(x, y, z, t)$ (kg/m^3 s) is the source strength and $s(x, y, z, t)$ (kg/m^3 s) is the sink. $\mathbf{u}$, q, s, and therefore c are stochastic variables. We propose to relate the statistical properties of c, the dependent variable, to those of the independent variables u, q, and s, for two idealized cases.

As a first simplification, we choose to consider systems for which the transport term $\nabla \cdot (\mathbf{u}c)$ can be neglected. That is, we shall investigate solutions to the equation:

$$\frac{dc}{dt} \cong q - s \tag{1'}$$

Equation (1′) is a legitimate representation of the conservation equation in two cases. The first is that in which all variables are measured in a system traveling with an air parcel whose velocity is $\mathbf{u}$. In such a Lagrangian system the functions q and s are to be understood as the source and sink, respectively, of particles in the moving parcel. The effective sink for this system as seen in the stationary frame is $s + (uc/D)$. The second interpretation of eq. (1′) is that it is the conservation equation in a frame at rest relative to the moving parcel, in which all the variables are averaged over linear scales D such that $\nabla \cdot (\mathbf{u}c) \approx uc/D \ll s$. This averaging implies that fluctuations over scales less than D will be ignored. Choice of the parameter D depends on the nature of each problem; that value of D for which $uc/D \approx s$ is the effective spatial dimension of the "cloud" of the substance under study.

We shall use eq. (1′) in the rest of this paper, parameterizing s and q in two different examples which represent idealized forms of the conservation equation for (a) industrial sulfate particles and (b) windblown dust particles. These examples were chosen because they represent aerosol types of major potential importance in radiative climate, because their life cycles can be approximated by simple q and s functions of two extreme types, and because there are many other substances whose sources and sinks are, to first approximation, combinations of the models used here.

In Section 2, we present model (a) for industrial haze, the methods for obtaining the parameters needed from data, the numerical results, and a confirmatory simulation of the model using parameter values derived from the data. Section 3 consists of a similar mathematical analysis of model (b) for windblown dust presented in order to demonstrate certain symmetrical properties of the two models. Data analysis in this section is rudimentary. We present our conclusions and suggestions for further study in Section 4.

2. Industrial haze

2.1. *The model*

The major source areas for industrial haze, which we assume is predominantly composed of sulfates (Bolin and Charlson, 1976) are the eastern half of the U.S. and Western Europe, both of linear dimension $L \approx 2000$ km. We assume that in the source region

$$q(t) = Q, \qquad \text{a constant,} \tag{2a}$$

and that the sinks are dry deposition and rainout, each of which is characterizable in the simplest approximation by a single time constant, i.e.

$$\begin{aligned} s(t) &= c(t)/T_p \qquad \text{when it is raining} \\ s(t) &= c(t)/T_d \qquad \text{when it is not} \end{aligned} \tag{2b}$$

(where "rain" implies snow or rain).

There are two extreme models we might choose for the statistics of the rainy episodes. The first is to assume all rain events in the source area are synchronized, the second that they are totally independent. The true picture lies somewhere between these extremes. For simplicity we have adopted the first, because frontal storms can sweep out areas comparable with L^2 in times somewhat shorter than the intervals between storms. Then, choosing $D \approx L \approx$ the scale of the source region, we examine some consequences of eq. (1′) assuming the advective flux can be ignored, an assumption to be evaluated later.

The analysis of the conservation equations for substances with this source and sink combination depends on the nature of the precipitation events, as defined by the density functions $F_{\{\substack{+\\0}\}}(s)$ where $F_{\{\substack{+\\0}\}}(s)\,ds$ is the probability a rainy/dry event lasts a time between s and $s + ds$. On the basis of available data, we assume

(2) a Markovian rain/dry system in which

$$F_+(s) = 1/T_+\, e^{-s/T_+} \tag{3a}$$

$$F_0(s) = 1/T_0\, e^{-s/T_0} \tag{3b}$$

where T_+ and T_0 are constants. This is the assumption underlying the work of Rodhe and Grandell (1972). In the case $T_+ \ll T_0$ (case (ii)) the mathematics becomes particularly simple. We have in the limit

$$F_+(s) = \delta(s) \tag{3c}$$

$$F_0(s) = 1/T_0\, e^{-s/T_0} \tag{3b}$$

From an examination of rain statistics in Western Europe, presented later in this section, it appears that there are cases in which both (3a) and (3c) provide moderately good descriptions of the data. We therefore derive the results of interest under both assumptions.

2.1.1. *Markovian rain/dry system.* To derive the particular form of area-averaged conservation equation appropriate for this case, we next define the functions $p_{\{\substack{+\\0}\}}(c,t)\,dc$ the probabilities the concentration is between c and $c + dc$ and it is rainy/dry at time t. Also let

$$\int_0^\infty dc\, p_{\{\substack{+\\0}\}}(c, t) = p_{\{\substack{+\\0}\}}(t) \tag{4a}$$

We further define

$$p(c, t)\, dc \equiv [p_+(c, t) + p_0(c, t)]\, dc \tag{4b}$$

the probability the concentration at the time t lies between c and $c + dc$ regardless of weather.

It is shown in Appendix A that the functions $p_{\{\substack{+\\0}\}}(c, t)$ obey the following equations:

$$\frac{\partial p_+(c, t)}{\partial t} = -\frac{p_+(c, t)}{T_+} + \frac{p_0(c, t)}{T_0} + \frac{\partial}{\partial c}(c/T_p - Q)\, p_+(c, t) \tag{5a}$$

$$\frac{\partial p_0(c, t)}{\partial t} = \frac{p_+(c, t)}{T_+} - \frac{p_0(c, t)}{T_0} + \frac{\partial}{\partial c}(c/T_d - Q)\, p_0(c, t) \tag{5b}$$

The expectation value of a quantity $f(x(t))$ is defined in terms of $p(x, t)\, dx \equiv$ the probability $x(t)$ lies in the interval $(x, x + dx)$ at time t:

$$E\{f(x(t))\} \equiv \overline{f(x(t))} = \int_0^\infty f(x)\, p(x, t)\, dx \tag{6}$$

From eqs. (5a), (5b) and (6) we find that the analog of eq. (1′) is the pair of equations

$$\frac{d\bar{c}_+}{dt} = -\bar{c}_+\left(\frac{1}{T_+} + \frac{1}{T_p}\right) + \bar{c}_0/T_0 + Qp_+(t) \tag{7a}$$

$$\frac{d\bar{c}_0}{dt} = -\bar{c}_0\left(\frac{1}{T_0} + \frac{1}{T_d}\right) + \bar{c}_+/T_+ + Qp_0(t) \tag{7b}$$

where $\bar{c}_+$ and $\bar{c}_0$ are the contributions to the mean concentration due to rainy dry periods, respectively. For $t \to \infty$ eq. (7a) and (7b) yield

$$\overline{c_+(t)} \underset{t\to\infty}{\longrightarrow} \frac{QT_+\left[1 + \left(\dfrac{T_+ T_0}{T_0 + T_+}\right)\dfrac{1}{T_d}\right]}{T_+/T_p + T_0/T_d(1 + T_+/T_p)}$$

$$\overline{c_0(t)} \underset{t\to\infty}{\longrightarrow} \frac{QT_0\left[1 + \left(\dfrac{T_+ T_0}{T_0 + T_+}\right)\dfrac{1}{T_p}\right]}{T_+/T_p + T_0/T_d(1 + T_+/T_p)}$$

and thus the average steady-state concentration is given by

$$\bar{c}(t) \equiv \bar{c}_+(t) + \bar{c}_0(t) \underset{t\to\infty}{\longrightarrow} \frac{Q\left[T_0 + T_+ + \left(\dfrac{T_0 T_+}{T_0 + T_+}\right)\left(\dfrac{T_+}{T_d} + \dfrac{T_0}{T_p}\right)\right]}{\dfrac{T_+}{T_p} + \dfrac{T_0}{T_d}\left(1 + \dfrac{T_+}{T_p}\right)} \equiv QT_{\text{eff}} \tag{8}$$

T_{eff} is the effective residence time of the average concentration. Equation (8) gives the same result for T_{eff} as the single-particle treatment of Rodhe and Grandell (1972), calculated under the assumption that the rain/dry system is Markovian. Further

$$\overline{c_+^2(t)} \underset{t\to\infty}{\longrightarrow} \frac{QT_+\left[\bar{c}_0(\infty) + \bar{c}_+(\infty)\left(1 + \dfrac{2T_0}{T_d}\right)\right]}{T_+/T_p + T_0/T_d(1 + 2T_+/T_p)}$$

$$\overline{c_0^2(t)} \xrightarrow[t\to\infty]{} \frac{QT_0\left[\bar{c}_+(\infty) + \bar{c}_0(\infty)\left(1 + \frac{2T_+}{T_p}\right)\right]}{T_+/T_p + T_0/T_d(1 + 2T_+/T_p)}$$

so that

$$\overline{c^2(t)} \xrightarrow[t\to\infty]{} \frac{Q\left[\bar{c}_+(\infty)\left(T_0 + T_+\left\{1 + \frac{2T_0}{T_d}\right\}\right) + \bar{c}_0(\infty)\left(T_+ + T_0\left\{1 + \frac{2T_+}{T_p}\right\}\right)\right]}{T_+/T_p + T_0/T_d(1 + 2T_+/T_p)} \tag{9}$$

Similarly after some algebra we find that

$$\overline{c^3(t)} \xrightarrow[t\to\infty]{} \frac{Q\left[\overline{c_+^2}(\infty)\left(T_0 + T_+\left\{1 + \frac{3T_0}{T_d}\right\}\right) + \overline{c_0^2}(\infty)\left(T_+ + T_0\left\{1 + \frac{3T_+}{T_p}\right\}\right)\right]}{\frac{T_+}{T_p} + \frac{T_0}{T_d}\left(1 + 3\frac{T_+}{T_p}\right)} \tag{10}$$

For T_d, the dry deposition time, much longer than the other times in the problem we have

$$\bar{c}(\infty) = Q\left[T_p\left(\frac{T_0 + T_+}{T_+}\right) + \frac{T_0^2}{T_0 + T_+}\right] \tag{8'}$$

which agrees with the result of Rodhe and Grandell (1972) for this case. We note that the climatological average of the removal time is

$$(T_p)_{\text{ave}} = T_p\left(\frac{T_0 + T_+}{T_+}\right)$$

Thus, the first term in eq. (8′) is the removal time which would be derived by considering the function $s(t)$ in eq. (1′) to be deterministic $s(t) = c(t)/(T_p)_{\text{ave}}$. The second term in T_{eff} represents the average waiting time from an arbitrary moment in a dry period until the onset of the next rainy episode, and it can be thought of as the perturbation in T_{eff} due to the stochastic nature of the removal mechanism.

For $1/T_d \to 0$ we have from eq. (9) the approximate relationship

$$\overline{c^2(\infty)} = Q^2\left[T_p\left(\frac{T_0 + T_+}{T_+}\right)\left\{T_p\left(\frac{T_0 + T_+}{T_+}\right) + \frac{T_0^2}{T_0 + T_+}\right\} + 2T_0^2\left\{\frac{T_p}{T_+} + \frac{T_0}{T_0 + T_+}\right\}\right]$$

We define $\sigma_c^2 \equiv \overline{c^2(\infty)} - \bar{c}^2(\infty)$. Then in this approximation

$$\frac{\sigma_c^2}{\bar{c}^2(\infty)} = \frac{\left[T_p/T_+ + \frac{T_0}{(T_0 + T_+)^2}(T_0 + 2T_+)\right]}{\left[\frac{T_p}{T_+}\left(1 + \frac{T_+}{T_0}\right) + \frac{T_0}{(T_0 + T_+)^2}(T_0 + T_+)\right]}$$

$$\approx \frac{T_0}{T_{\text{eff}}} \qquad \text{if } \frac{T_+}{T_0} \ll 1 \tag{11}$$

This result can be understood by recalling that the average residence time for the average concentration is T_{eff} and therefore the average number of rainy episodes experienced by the concentration is T_{eff}/T_0, if $T_+ \ll T_0$.

Thus eq. (11) states that the relative variance of the concentration distribution decreases as the reciprocal of the number of episodes experienced. Note that σ_c^2 is that contribution to the variance of the steady-state concentration distribution due only to fluctuations in the weather. There will be additional variance in the concentration due to the fact that different particles reside different times in the atmosphere. For large numbers of particles, however, this contribution to the variance becomes small and we shall neglect it.

Finally, we define the skewness S of the steady-state concentration distribution;

$$S \equiv \overline{\left[\frac{c(\infty) - \bar{c}(\infty)}{\sigma_c}\right]^3}$$

Again in the limit $1/T_d \to 0$ we have

$$S = \frac{\left[\begin{array}{l}\dfrac{T_p^3}{T_+}(3T_0 + T_+) + \dfrac{6T_0^2 T_p^2}{T_0 + T_+} \\ + \dfrac{2T_p T_0^3}{T_+}\left(4 - \dfrac{3T_0}{T_0 + T_+}\right) + \dfrac{2T_0^6}{(T_0 + T_+)^3}\end{array}\right]}{\dfrac{T_0^2}{T_0 + T_+}\left(\dfrac{T_0(T_0 + 2T_+)}{T_0 + T_+} + \dfrac{T_p(T_0 + T_+)}{T_+}\right)}$$

$$\approx 2(T_0/T_{\text{eff}})^{1/2} = 2(\sigma_c/\bar{c}(\infty)) \text{ for } T_+/T_0 \ll 1 \quad (12)$$

For a Poisson process $S = 1(\sigma_c/\bar{c})$ and for a log-normal process where $\sigma_c/\bar{c} \ll 1$, $S \approx 3(\sigma_c/\bar{c})$. Thus the skewness given by eq. (12) is between that for these two asymmetrical distributions. The inadequacy of any description of the concentration distribution which does not include higher moments is evident.

The time-dependent characteristics of the concentration distribution can be derived with straightforward though cumbersome algebra from eqs. (4) and (5). It is both realistic and convenient, however, to derive those properties of interest under the point-process assumption, (ii); namely, that the rain episodes are short compared to the dry ones.

2.1.2. *Point-process rain system.* We now let $c\,dJ/dt$ be the rate at which the concentration is diminished by precipitation. Then we have

$$s(t) = c(t)\frac{dJ(t)}{dt} + \frac{1}{T_d} \quad (13)$$

and eq. (1′) becomes

$$\frac{dc(t)}{dt} = Q - c(t)\left(\frac{dJ(t)}{dt} + \frac{1}{T_d}\right) \quad (14)$$

which has the formal solution

$$c(t) = c(o)\exp[-(t/T_d + J(t))] + \exp[-(t/T_d + J(t))]Q\int_0^t \exp[t'/T_d + J(t')]\,dt' \quad (15)$$

If we assume the rainy episodes are brief compared to the dry intervals between them, we can write

$$J(t) = X_1 + X_2 + X_3 + \cdots X_{N(t)} \quad (16)$$

where $N(t)$ is the number of precipitation episodes in the interval $(0, t)$ and the fraction of the concentration removed by the ith episode is X_i. X_i is assumed independent of i, and the probability that any of the X_i lies in the interval x, $x + dx$ is $p(x)\,dx$. We assume further that the times t_i at which the rainy episodes occur are Poisson distributed with parameter $1/T_0$. (That is, T_0 is the mean time between episodes.) Thus, the probability there are $N(t)$ episodes in $(0, t)$ is

$$p[N(t) = n] = \frac{e^{-t/T_0}(t/T_0)^n}{n!} \quad (17)$$

From eq. (15) we find the average value of $c(t)$:

$$\bar{c}(t) = \bar{c}(0)\,e^{-t/T_d}\,\overline{(e^{-J(t)})} + e^{-t/T_d}\,Q\int_0^t \overline{\exp[-J(t) + J(t') + t'/T_d]}\,dt' \quad (18)$$

From eqs. (16) and (17) we have that

$$\overline{e^{-J(t)}} = \sum_{n=0}^{N(t)} \frac{\overline{(e^{-x})}^n (t/T_0)^n}{n!}\,e^{-t/T_0}$$
$$= \exp\{-[t/T_0(1 - \overline{e^{-x}})]\} \equiv e^{-\beta_1 t} \quad (19)$$

where $\overline{e^{-xn}} \equiv \int_0^\infty e^{-xn}p(x)\,dx$ (see eq. 6), and we define

$$\beta_n \equiv \frac{1}{T_0}(1 - \overline{e^{-xn}}) \quad (20)$$

From eqs. (18) and (19) we then have

$$\bar{c}(t) = \bar{c}(0)\exp\{-[\beta_1 + 1/T_d]\,t\} + \frac{Q[1 - \exp\{-[\beta_1 + 1/T_d]\,t\}]}{\beta_1 + 1/T_d} \quad (21)$$

and as $t \to \infty$

$$\bar{c}(t) \xrightarrow[t\to\infty]{} \frac{Q}{\beta_1 + \dfrac{1}{T_d}} \equiv QT_{\text{eff}} \quad (22)$$

The autocorrelation function $\text{cov}(c(t)c(t + h)) \equiv \overline{c(t)c(t + h)} - \overline{c(t)}\cdot\overline{c(t + h)}$ can also be found from eq. (15). If we assume the system is stationary we can neglect the transient term $c(0)e^{-[J(t) + t/T_d]}$ to find that asymptotically

$$\overline{c(t)c(t + h)} \xrightarrow[t\to\infty]{} Q^2\int_0^{t+h} dt' \int_0^t dt'' \cdot$$
$$\cdot \exp\left\{-\left[J(t + h) + J(t) + \frac{2t + h}{T_d}\right] + J(t') + J(t'') + \left(\frac{t' + t''}{T_d}\right)\right\}$$

It is shown in Appendix B that this leads to

$$\operatorname{cov}\,(c(t)\,c(t+h)) \underset{t\to\infty}{\longrightarrow} Q^2 \cdot$$

$$\cdot \left[\frac{(2\beta_1 - \beta_2)\, e^{-(\beta_1 + 1/T_d)h}}{(\beta_1 + 1/T_d)^2\,(\beta_2 + 2/T_d)}\right];$$

$$\frac{\sigma_c^2}{(\bar{c}(\infty))^2} = \frac{2\beta_1 - \beta_2}{\beta_2 + 2/T_d} \tag{23}$$

The skewness of the steady-state concentration distribution is

$$S = \left[\frac{6(\beta_1 + 1/T_d)}{(\beta_2 + 2/T_d)}\left\{\left(\frac{\beta_1 + 1/T_d}{\beta_3 + 3/T_d}\right) - 1\right\} + 2\right] \cdot \left[\frac{2\beta_1 - \beta_2}{\beta_2 + 2/T_d}\right]^{3/2} \tag{24}$$

2.2. *The parameters*

To obtain numerical results from our model equations under assumption (i) we need Q, T_0, T_+, T_d and T_p, and under assumption (ii) we need T_0, T_d, Q and the β_0. A rain episode threshold is determined by setting a minimum value for rainfall intensity under which rainout is presumed negligible. Rainfall is then put equal to 0 if the intensity is below this threshold. T_+ is the mean duration of the episodes of higher-than-threshold rainfall intensity and T_0 the mean duration of the episodes between them. The proper calculation of T_p, the wet deposition time, involves considerations of drop and particle size distributions which are outside the scope of this paper. We simply let T_p be a size averaged removal time, assumed here to be independent of precipitation intensity. To estimate T_p, we assume an average intensity for precipitation of 0.1 mm/hr. If the concentration of sulfates in rainwater is c' (kg/m^3) and the rainfall intensity is j (m/s) then

$$T_p = \frac{cH}{c'j} \tag{25}$$

where H is the average scale height for sulfate. From European rain chemistry data (OECD 1975) $c' \sim 10^{-2}$ kg/m^3. Then if $c \sim 10^{-8}$ kg/m^3 and $H \sim 1000$ m we find $T_p \sim 10$ h. Makhon'ko (1967) concluded from observations on radioactive debris that for $j = 1$ mm/h, 3 h $\le T_p \le$ 10 h. We shall use $T_p = 10$ h in these calculations. This is appreciably longer than the values used by Rodhe and Grandell (1972), although they used a minimum rainfall intensity of $j = 0.05$ mm/h.

To estimate T_d, the dry deposition time, we put

$$T_d = \frac{H}{v_s} \tag{26}$$

where v_s (m/s) is the settling velocity of a "typical" particle. In air $v_s \cong 0.001\ (r(\mu\mathrm{m}))^2$. Thus if $H \sim 1$ km $T_d \sim 280$ h for a 1 μm particle and $T_d \sim 11$ h for a 5 μm particle.

To find T_0 we set a threshold for total rainfall per rainy episode under which it is assumed rainout is significant. T_0 is then the mean time between episodes characterized by rainfalls in excess of that threshold. The parameters β_n are calculated from $p(x)$, which can be determined from rain chemistry measurements. Interpretation of such data and its application to our model we intend for subsequent papers. For the present we use a crude model for the estimation of the parameters in part (2) of our sulfate model, to compare our results with those from part (1) We assume all rain episodes are of equal intensity and that $X_i \approx T_i/T_p$, where T_i is the duration of the ith storm and x_i the fraction removed by that storm. (We are grateful to Dr Rodhe for suggesting this approximation.) It will be shown in the data section which follows that rain episode durations in Western Europe are in general exponentially distributed. That is, $p(x)\,dx = (T_p/T_+)\exp(-xT_p/T_+)\,dx$, where T_+ is the mean duration of the rainy episodes. We then have from eq. (20) that

$$\beta_n = \frac{1}{T_0}\,\frac{n}{n + T_p/T_+} \tag{27}$$

Then from eqs. (21)–(24) we find

$$T_{\mathrm{eff}} = \frac{T_0(1 + T_+/T_p)}{T_+/T_p + T_0/T_d(1 + T_+/T_p)} \tag{28}$$

Also

$$\overline{c^2(\infty)} = \frac{QT_0\,\bar{c}(\infty)\left(1 + \dfrac{2T_+}{T_p}\right)}{T_+/T_p + T_0/T_d(1 + 2T_+/T_p)} \tag{29}$$

and

$$\overline{c^3(\infty)} = \frac{\overline{c^2(\infty)}\,QT_0\left(1 + \dfrac{3T_+}{T_p}\right)}{\dfrac{T_+}{T_p} + \dfrac{T_0}{T_d}\left(1 + 3\dfrac{T_+}{T_p}\right)} \tag{30}$$

Equations (28), (29) and (30) agree with eqs. (8), (9) and (10) in the limit $T_+/T_0 \ll 1$. Again a pattern for calculation of higher moments of c is evident.

Extraction of numerical values of the parameters T_0 and T_+ from data and the corresponding numerical predictions of the model are discussed in the next section.

2.3. *Data analysis*

2.3.1. *Persistence times T_+ and T_0.* A time series of precipitation episodes was generated for each of eight European meteorological stations from the WMO records for 1974. The precipitation series consists of a two-state process (Z_n: $n = 1, 2, 3, \ldots$) such that $Z_n = 1$ if precipitation occurs in the nth period (reported in intervals of 3 h), otherwise $Z_n = 0$. For the purposes of this analysis Z_n was set equal to 1 if two criteria were met: first, during the preceding three-hour period rain or snow was observed at the station, and second, the total precipitation during the prior six-hour period was in excess of 0.3 mm. We define the frequency distribution of Z_n as (f_i: $i = 1, 2, 3, \ldots$) such that f_i = number of episodes which lasted i three-hour periods, divided by the total number of intervals in the record.

The durations of most precipitation episodes are comparable with the three-hour reporting period of the meteorological network. In an attempt to improve the resolution of the duration statistics, we estimated a distribution of persistence times resolved at the reporting period, and extrapolated this distribution into the subscale of unresolved times to obtain the persistence times listed in Table 1.

2.3.2. *Visibility.* The visibility records for the eight European stations of Table 1 were processed through a running average low pass filter to remove the diurnal fluctuations and then averaged for episodes when relative humidities were less than 70%. These visibilities were then converted to equivalent sulfur-aerosol concentrations by the empirical ratio $\bar{c}/b_{scat} = 0.2$ (g/m^2) (Waggoner et al., 1976), where b_{scat} (m^{-1}) is the light scattering coefficient calculated from $b_{scat} = 3.9/L_v$ (m) is the visible range (Middleton, 1968). This procedure yielded a mean concentration for the eight stations of 5.0×10^{-8} kg/m^3, a standard deviation to mean ratio of 0.6, and a skewness of 2.

Table 1. *Persistance times for various places*

	Hours			
Station	T_0 Winter	T_+ Winter	T_0 Summer	T_+ Summer
Oslo	29	8	39	6
Stockholm	37	7	69	5
Manchester	22	6	31	5
London	26	7	72	5
Frankfurt	28	6	46	5
Poznan	36	6	53	5
Leningrad	24	6	64	6
Moscow	25	7	61	4

2.4. *Simulation and comparison with data*

To complement and check the analytical results we have described in the preceding section, we have written a brief simulation program in which the parameters in the model could be approximated by Monte Carlo methods. The appropriate moments and correlations of the resulting time series of simulated concentrations were then averaged over epochs long enough to establish a "climate". Fig. 1 shows simulated concentrations resulting from such an exercise.

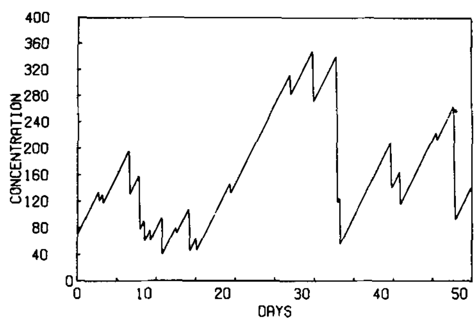

Fig. 1. Time course of simulated sulfate concentration: $T_p = 10$ h, $T_0 = 50$ h, $T_+ = 5$ h, $Q = 1$.

On the abscissa were the last 50 days of a one-year simulation, calculated with 18 min time steps. The parameters of Fig. 1 were chosen to resemble those appropriate to the European sulfate cloud, under summer conditions, where the mean duration of rainy events was taken to be 5 h, the mean duration of dry episodes was taken to be

50 h, dry deposition was neglected, and the characteristic time for pollutant removal during rain was taken to be 10 h. The ordinate scale, in arbitrary units, is proportional to a unit injection rate times T_{eff}.

Fig. 2 shows a concentration histogram for the second half year of the simulations. This reveals considerable skewness and thereby re-emphasizes the potential importance of the higher moments of concentration distributions.

Fig. 3 shows a lagged autocorrelation, again computed for the second half year of the simulation. This illustrates the utility of eq. (23), which predicts simple exponential decay. The concentration power spectrum, which is related to its autocorrelation through the Fourier transformation, is consequently Lorentzian.

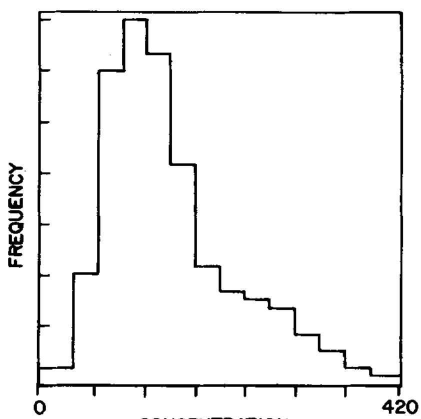

Fig. 2. Histogram of simulated sulfate concentrations for second half year. Parameters those of Fig. 1. Concentration in units of QT_{eff} (see text).

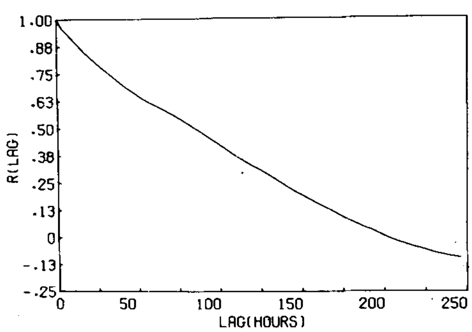

Fig. 3. Lagged autocorrelation function of simulated sulfate concentrations for second half year. Parameters those of Fig. 1.

Table 2 summarizes these simulations by comparison of their derived moments with the analytical expressions of eqs. (8′), (11) and (12) and with those derived from visibility records from the same period for the summer months. The steady-state concentration is predicted from eq. (8′) using the computed values of T_{eff} and $Q = 4 \times 10^{-3}$ kg/m² yr. This value of Q is derived from data presented by Bolin and Persson (1975) for annually averaged SO_2 production rates in Europe. We assume oxidation of SO_2 to sulfate is rapid compared with SO_2 removal, so that the SO_2 production rate and sulfate production rate are approximately equal.

Table 2. *Comparison between simulation, analysis and observation. Summer conditions*

	Simulation	Analysis	Observation
T_{eff} (h)	144 ± 11	150	Not Available
c (kg/m³)	arbitrary	6×10^{-8}	5×10^{-8}
Sigma/mean	0.5 ± 0.12	0.58	0.6
Skewness	1.0 ± 2.0	1.2	2.7

Our results are consistent with the hypothesis that the mean and variance of visibilities in Western Europe are controlled by a balance between nearly constant emission and stochastic rainout. The shape of the visibility histogram appears more skewed than that of our own analysis and simulation; however, the large standard error in our simulation of the skewness, even with half a year's "observations", if of similar magnitude to the real standard errors in measuring the real skewness, suggests that for this parameter also the analysis and observations are "not inconsistent".

3. Windblown dust

In this section we sketch the analysis of a life cycle which is, crudely speaking, the "inverse" of the sulfate cycle, in that we assume a stochastic on/off source and a constant removal rate. The purpose of this section is largely pedagogical: to show the similarities and differences in the statistical properties of the concentration distributions resulting from the two source-sink combinations. Detailed numerical predictions based on this model have not as yet been made, due to insufficiencies in the data available to us; this task remains a goal for future work.

3.1. *The model*

Since there is little rain in the dusty or arid regions of the earth, the main sinks in those regions are dry deposition, which we again parameterize by a single (size-dependent) removal time, T_d, and advection. The sources of wind-blown dust are temporarily assumed to be (a) the dust storms which occur at random intervals, particularly (in the Near Eastern and African deserts) in spring and summer, and (b) a constant background B (kg/m³ s). We let the amount of dust injected into the atmosphere by these storms in the interval $(0, t)$ be $A(t)$. Then eq. (1) becomes

$$\frac{\partial c}{\partial t} = B + \frac{dA}{dt} - \nabla \cdot (\mathbf{u}c) - \frac{c}{T_d} \approx \frac{dA}{dt} - \left(\frac{u}{D} + \frac{1}{T_d}\right) c \tag{31}$$

For each particle size the value of T_d is fixed, which defines a value of D, a linear scale for which $1/T_d = u/D$, within which we ignore advection. We consider here particles of sufficient size that D = the dimension of the heavy cloud characteristic of the most active region of a dust storm. For these particles the formal solution of eq. (31) is

$$c(t) = c(0)^{-t/T_d} + e^{-t/T_d} \int_0^t e^{t'/T_d}\, dA(t') + BT_d(1 - e^{-t/T_d}) \tag{32}$$

Examination of the WMO records for 1974 from the Near Eastern and African deserts shows that to first approximation the storms reported at individual stations are independent of one another, are relatively rare, and that their durations are on the average much shorter than those of the intervals between storms. We therefore assume that for the desert as a whole the total amount of dust, $A(t)$, injected into the atmosphere by the storms during the interval $(0, t)$ is a compound Poisson process (Feller, 1971, page 180, Example (a)). Thus, letting $N(t)$ denote the number of storms during time $(0, t)$ and letting g_k be the increase in dust concentration due to the kth storm, we have

$$\begin{aligned} A(t) &= 0, & &\text{for } N(t) = 0 \\ &= g_1 + g_2 + \cdots + g_{N(t)}, & &\text{for } N(t) \geq 1 \end{aligned} \tag{33}$$

The random variables $g_1, g_2, \cdots$, are stochastically independent and have a common distribution F. The storm-counting process $\{N(t), t \geq 0\}$ is a Poisson process with rate $1/T_0$ = mean number of storms per unit of time. (The distribution of $N(t)$ is given by eq. (17).) The process $\{N(t), t > 0\}$ and the sequence $g_1, g_2, \cdots$, are stochastically independent. For our purposes the exact form of the distribution F is unimportant, but we do assume that it has a finite mean, $\bar{g}$, second moment $\overline{g^2}$ and third moment $\overline{g^3}$. With these assumptions in force, we show in the appendix C that

$$\bar{c}(t) = [\bar{c}(0)]\, e^{-t/T_d} + [\bar{g}T_d/T_0 + BT_d]\,(1 - e^{-t/T_d}) \tag{34}$$

where

$$\overline{g^n} \equiv \int_0^\infty g^n p(g)\, dg$$

Thus

$$\bar{c}(t) \underset{t\to\infty}{\longrightarrow} \bar{g}T_d/T_0 + BT_d \tag{35}$$

Further,

$$\mathrm{cov}(c(t)\, c(t+h)) \underset{t\to\infty}{\longrightarrow} \frac{\overline{g^2}\, T_d}{2T_0} e^{-h/T_d};$$

$$\sigma_c^2/[\bar{c}(\infty)]^2 = \overline{g^2}[2T_d T_0(\bar{g}/T_0 + B)^2]^{-1} \tag{36}$$

and

$$S = \left(\frac{T_0}{T_d}\right)^{1/2} \left[\frac{\overline{g^3}/3}{\overline{g^2}/2^{3/2}}\right] \tag{37}$$

Equation (36) exhibits the same dependence of $\mathrm{cov}[c(t)c(t + h)]$ on the residence time, $T_{\mathrm{eff}} \equiv T_d$, as does its counterpart in eq. (23), which is a consequence of the Markovian nature of both models.

Note that the distribution of the durations of dust storms is *not* a simple exponential, and thus a treatment of the dust system analogous to that in part 2.1 of Section 2 for sulfates is not justified. It can be shown, in fact (Ambroziak, cf. Hinds and Hoidale, 1975; unfortunately, this reference is classified) that an exponential approximation may seriously under-estimate the total dust injection rate.

3.2. *The parameters*

The parameters entering our equations for dust are T_0, T_d (for particles of radius greater than 25 μm) and the moments of g (for the large particles). While in principle all of these should be obtainable from currently available data, in practice attempts at extraction of reliable values reveal major deficiencies in the data which preclude any but the roughest estimates.

No single operational definition of a dust storm has been accepted in the literature. We define a dust storm here as a condition in which the current weather indicator as reported by the observer at the WMO station is "dust storm", and simultaneously the visibility is less than 11 km. We have attempted to determine the spatial extent of the "storms" from 1974 data recorded at stations in the New Eastern and North African deserts. Various synchrony indices for all possible pair-wise combinations of stations were plotted as functions of the distance between stations. No significant correlation between synchrony and distance could be noted. To test the method, synthetic "storms" were added to the data. From these studies we conclude that (a) the WMO network in those regions is unsuitable for the detection of synchronous events of linear dimension $\lesssim$200 km, even if numerous, and (b) the threshold for detection of synchrony in the Saharan region (defined here as the area from 16° W to 30° E and from 10° N to 26° N) by this network is approximately 3 events per year of radius over 200 km. The number of such events in 1974 was subthreshold. That is, either the large storms seen on satellite photographs (Grigoriev et al., 1976) and discussed in the literature (Joseph et al., 1973) are exceptions, or our test year was exceptionally quiet. We therefore assume the events are of linear dimensions less than 200 km. The Saharan region was divided into blocks 200 km × 200 km, the number of events reported in each block averaged and the blocks for which there were no station filled in by interpolation. The average number of dust storms in an area 4×10^4 km^2 in that region found by this method was $\sim$4/yr $= 1/T_0$.

As a comparison of the 1974 data record with previous years the number of dust episodes for Mali, Mauritania, Niger, and the Spanish Sahara were compared with those reported by Hinds and Hoidale (1975). This region, which accounted for the largest number of dust storms and dust episodes for North Africa was found to have an average of 34 days per year with dust or haze. The reported eighteen-year average was 21 days with dust. The difference is probably due to the added criterion of haze in our definition of dust episode.

Grigoriev et al. (1976) estimated T_d from observations of a storm in which the average horizontal wind speed was $\bar{u} \sim 13$ m/s, dust cloud height $H = 2$ km, and linear dimension of the cloud $D = 130$ km. Thus for the particles deposited with the boundaries of this cloud $T_d \leq D/\bar{u} \simeq 2.8$ h. The authors conclude that most of these particles are larger than 25 μm in radius.

For the large particles $\bar{g}$ can be estimated from local measurements of dust concentrations made during storms. We assume the source is "on" at a constant rate R (kg/m^3 h) during a storm and that the mean duration of a storm is T_+, so that $\bar{g} = RT_+$. Then the time-averaged local concentration of heavy particles during a storm, neglecting the contribution due to those "left over" from other storms, is

$$\bar{c} \sim T_d R \left[1 - \frac{T_d}{T_+} (1 - e^{-T_+/T_d}) \right] \tag{38}$$

and

$$\bar{g} \sim \frac{\bar{c}_r \geq 25\ \mu\text{m}}{\dfrac{T_d}{T_+} \left[1 - \dfrac{T_d}{T_+} (1 - e^{-T_+/T_d}) \right]} \tag{39}$$

The total column mass loading during dust storms reported by Grigoriev et al. (1976) is 27 g/m^2, which for a dust cloud height of 2 km corresponds to a mean density of 1.3×10^{-2} g/m^3. The masses in the various size ranges can be estimated from size distribution measurements of Schütz and Jaenicke (1974) made during dust storms in the Sahara. They find $dM/d\log r$ is approximately constant with radius r, where dM is the mass of particles for which $\log r$ lies in the range between $\log r$ and $\log r + d\log r$. This relationship holds for 20 μm $\leq r \leq$ 100 μm. Grigoriev's measurement implies that the mass of particles of radius between 25 and 100 microns

$\sim 10^{-2}$ g/m^3. If we assume $T_d \sim 3$ h and $T_+ \sim 5$ h (Hinds and Hoidale, 1975) we find from eq. (39) that

$\bar{g} \sim 2 \times 10^{-2}$ g/m^3

From eq. (35), neglecting B, $\bar{c} \sim 3 \times 10^{-5}$ g/m^3, and for the desert as a whole, assuming a uniform density of 4 storms per year, the yearly injection rate of large particles due to storms is approximately 160 g/m^2.

From the dust statistics compiled by Hinds and Hoidale (1975) as well as the WMO records we have examined, most of the variance in g appears to be in the durations of the dust storms. Estimation of σ_g^2 on the basis of more complete data remains a goal for the future.

4. Conclusions

Analysis of the simplified stochastic conservation equations for area-averaged concentrations of industrial sulfates and windblown dust leads to the results

$$\operatorname{cov}[c(t)c(t+h)] \xrightarrow[t\to\infty]{} \sigma_c^2 e^{-h/T_{\mathrm{eff}}} \quad \frac{\sigma_c^2}{\bar{c}^2(\infty)} \propto \frac{1}{T_{\mathrm{eff}}}$$

for both classes of aerosols, where T_{eff} is the effective residence time of the particles. The latter result is consistent with that of Gibbs and Slinn (1973), who showed that it holds in general if the characteristic time for source and sink variations is short compared to T_{eff}, which is the case we have considered. This result differs from the finding of Junge (1974); namely, that measurements of several trace gases appear to display $\sigma_c^2/\bar{c}(\infty)^2 \propto 1/T_{\mathrm{eff}}^2$.

Numerical estimates based on our models show satisfactory agreement with data wherever comparison was possible. Standardization and upgrading of data recording in the dusty areas of the world will be necessary for full utilization of these or similar statistical methods of analysis there.

The models presented here represent an approach to the treatment of atmospheric phenomena in which the salient statistical features of the processes considered are approximated by functions of sufficiently simple form that analytic solution of the resulting equations is possible. These models are applicable as they stand to the study of other life cycles in which the sources or sinks are approximately point processes, such as volcanic debris. With straightforward modification they may be applied to other random atmospheric processes; for example, in certain cases, to the study of radiative transfer in cloudy media. They differ fundamentally from standard treatments in the use of data in that the "noise" in atmospheric measurements enters as a parameter in the equations, thus utilizing often ignored information.

5. Appendix A

To derive the time rate of change of $p_{\{{+\atop 0}\}}(c, t)$ we consider a time element Δt in which the concentration is changed from c to $c + Q\Delta t - (c/T)\cdot\Delta t + 0(\Delta t)^2$ where $T = T_p$ during a rainy episode and $T = T_d$ during a dry one. Then if the rainy and dry episodes are each exponentially distributed with mean times T_+ and T_0, respectively, we have

$$p_+(c, t+\Delta t) = \left(1 - \Delta t\left(\frac{1}{T_+} - \frac{1}{T_p}\right)\right)\cdot p_+\left(c - \left\{Q - \frac{c}{T_p}\right\}\Delta t, t\right) + \frac{\Delta t}{T_0} p_0(c + \{Q - c/T_d\}\,\Delta t, t) + 0(\Delta t)^2 \tag{A1}$$

Similarly

$$p_0(c, t+\Delta t) = \left(1 - \Delta t\left(\frac{1}{T_0} - \frac{1}{T_d}\right)\right)\cdot p_0\left(c - \left\{Q - \frac{c}{T_d}\right\}\Delta t, t\right) + \frac{\Delta t}{T_+} p_+(c + \{Q - c/T_p\}\,\Delta t, t) + 0(\Delta t)^2 \tag{A2}$$

From (A1) we have

$$\frac{p_+(c, t+\Delta t) - p_+(t)}{\Delta t} = \frac{-p_+(c, t) + p_+(c - \{Q - c/T_p\}\,\Delta t, t)}{\Delta t} + \frac{p_0(c, t)}{T_0} - \frac{p_+(c, t)}{T_+} + 0(\Delta t) \tag{A3}$$

and (A2) yields

$$\frac{p_0(c, t + \Delta t) - p_0(t)}{\Delta t} = \frac{-p_0(c, t) + p_0(c - \{Q - c/T_d\}\,\Delta t, t)}{\Delta t} + \frac{p_+(c, t)}{T_+} - \frac{p_0(c, t)}{T_0} + 0(\Delta t) \quad \text{(A4)}$$

In the limit $\Delta t \to 0$ (A3) and (A4) yield eqs. (5a) and (5b) of this paper.

6. Appendix B

In order to compute the asymptotic autocorrelation function of the concentration we must take expectation values of the expression

$$Q^2 \int_0^{t+h} dt' \int_0^t dt'' \exp\left\{-\left[J(t+h) + J(t) - J(t') - J(t'') + \frac{2t + h - (t' + t'')}{T_d}\right]\right\}$$

$$\equiv Q^2_{\exp}\left[-\left(\frac{2t+h}{t_d}\right)\right] \int_0^{t+h} dt' \int_0^t dt'' f(t, t', t'') \quad \text{(B1)}$$

It is convenient to write the integral as the sum

$$Q^2_{\exp}\left[-\left(\frac{2t+h}{T_d}\right)\right] [\int_0^5 dt' \int_0^{t'} dt'' f(t, t', t'') + \int_0^t dt' \int_{t'}^t dt'' f(t, t', t'') + \int_t^{t+h} dt' \int_0^t dt'' f(t, t', t'')] \equiv I_1 + I_2 + I_3$$

We note that $f(t, t', t'')$ is symmetric with respect to interchange of t' and t'', so that $I_1 = I_2$. For $t'' < t'$ we write

$$J(t+h) - J(t') + J(t) - J(t'') = [J(t+h) - J(t)] + J(t'') - J(t') - 2[J(t) - J(t'')] \quad \text{(B2)}$$

Thus,

$$\bar{I}_1 = \bar{I}_2 = Q^2_{\exp}\left[-\left(\frac{2t+h}{T_d} + \beta_1 h\right)\right] \int_0^t dt' \int_0^{t'} dt'' \cdot \exp\left\{-\left[\beta_2(t - t') + \beta_1(t'' - t') + \left(\frac{t' + t''}{T_d}\right)\right]\right\} \quad \text{(B3)}$$

As $t \to \infty$

$$\bar{I}_1 = \bar{I}_2 \to \frac{Q^2_{\exp}[-(\beta_1 + 1/T_d)h]}{(\beta_1 + 1/T_d)(\beta_2 + 2/T_d)} \quad \text{(B4)}$$

Also

$$\bar{I}_3 \to \frac{2}{(\beta_1 + 1/T_d)^2}(1 - \exp[-(\beta_1 + 1/T_d)h]) \quad \text{(B5)}$$

Thus setting $\mathrm{cov}[c(t)c(t+h)] = \bar{I}_1 + \bar{I}_2 + \bar{I}_3 - \bar{c}^{-2}(t)$ we find the result in eq. (23). By similar algebra we can compute $\overline{c^3(t)}$ which leads to eq. (24) for

$$S = \lim_{t \to \infty} \frac{\overline{[c(t) - \bar{c}(t)]^3}}{\sigma_c^3}$$

7. Appendix C

From our assumptions about $A(t)$ it follows that

$$\overline{A(t)} = \sum_{n=0}^{\infty} \frac{\bar{g} n\, e^{-t/T_0}(t/T_0)^n}{n!} = \frac{\bar{g}t}{T_0}; \qquad \overline{dA}(t) = \frac{\bar{g}\, dt}{T_0} \quad \text{(C1)}$$

Also

$$\mathrm{cov}(A(t)\,A(t+h)) = \frac{\overline{g^2}\,t}{T_0} = \sigma_A^2, \qquad h \geq 0 \quad \text{(C2)}$$

$$\mathrm{cov}(dA(t)\;dA(t+h)) = \delta(t - t')\,\overline{g^2}\,dt/T_0$$

and

$$\overline{dA(t)\,dA(t')\,dA(t'')} = [\overline{dA}(t)]^3 + \frac{3\delta(t-t')\,\overline{g^2}\,dt\,\overline{dA(t'')}}{T_0} + \overline{A^3(t)}\;\delta(t-t')\;\delta(t-t'')\;dt$$

Taking expectation values of eq. (32) leads to

$$\bar{c}(t) = \bar{c}(0)\,e^{-t/T_d} + \overline{e^{-t/T_d}\int_0^t e^{t'/T_d}\,dA(t')} + BT_d(1 - e^{-t/T_d}) \quad \text{(C4)}$$

and from eqs. (C1) and (C4) we find eq. (34). Similarly,

$$\mathrm{cov}(c(t)\,c(t+h)) = \exp\left[-\left(\frac{2t+h}{T_d}\right)\right] \cdot \mathrm{cov}\left(\int_0^{t+n} dA(t')\,e^{t'/T_d} \int_0^t dA(t'')\,e^{t''/T_d}\right) \quad \text{(C5)}$$

and (C2 and C5) yield eq. (35). Finally,

$$\overline{c^3(t)} = e^{-3t/T_d} \cdot \overline{\int_0^t e^{t'/T_d} dA(t') \int_0^t e^{t''/T_d} dA(t'') \int_0^t e^{t'''/T_d} dA(t''')} \quad \text{(C6)}$$

Equations (C3) and (C6) yield eq. (37).

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ПРОСТЫЕ СТОХАСТИЧЕСКИЕ МОДЕЛИ ДВУХ ТИПОВ ИСТОЧНИКОВ И СТОКОВ АЭРОЗОЛЯ

Из простых стохастических моделей источников и стоков частиц получены статистические свойства двух зависящих от времени и установившихся распределений аэрозоля по концентрации. Проанализирована статистика осредненных по пространству концентраций аэрозоля (а) антропогенных сульфатных частиц, для которых источник предполагался постоянным во времени, а удаление осуществлялось осадками в случайные моменты времени, и (б) пылевых частиц, источниками которых предполагались случайно происходящие пыльные бури, а стоками являлись независимая от времени адвекция и сухое выпадение. Уравнения модели решаются как аналитически, так и численно. Для обеих моделей временнáя автокорреляционная функция концентрации примеси — простая экспонента, а отношение установившейся дисперсии к квадрату средней концентрации изменяется обратно пропорционально эффективному времени жизни.

Применение моделей к данным Всемирной метеорологической организации показывает, что: в случае (а) ожидаемые времена жизни для сульфатов от 50 до 175 часов, а характерный масштаб переноса от 800 до 2000 км.