Multiple state stochastic models for the long-range transport and removal of atmospheric tracers

BY GARY D. EGBERT AND MARCIA B. BAKER

Geophysics Program AK-50, University of Washington, Seattle, WA 98195

(Received 20 March 1985; revised 10 October 1985)

SUMMARY

We discuss a general mathematical framework for modelling long-term average concentration and deposition patterns for atmospheric tracers which are subject to removal and transformation. For a generalized 'particle' which can exist in a finite number of states, the joint statistics of random particle transport and inter-state transformation can be used to calculate long-term mean fields for deposition and concentration. We show that a number of models presented in the recent literature can be considered as simple cases of this formulation. These models generally assume a weak statistical relationship between vertical transport, horizontal transport and removal processes. We consider a general approach to linking these processes, based on calculating approximate transport statistics conditional on particle states. We demonstrate this approach with a rather schematic, but almost analytically solvable, stochastic model which allows for the linkage of transport and removal processes. We use these ideas to construct a two-layer model of the transport and removal of sulphur in a region of convective precipitation, demonstrating how the correlation of vertical transport and precipitation can change the precipitation statistics in the particles' Lagrangian frame and hence affect deposition patterns.

1. INTRODUCTION

Since the mid 1970s, when the possible environmental effects of the long-range transport (LRT) of industrial pollutants became apparent, significant efforts have been expended to understand such phenomena, and numerous LRT models have been proposed (see Fisher (1983) for a recent review). These models can be roughly divided into two distinct groups characterized by different modelling approaches and different uses (Eliasen 1980). The first group includes models which, for specified transport winds, precipitation fields, etc., solve the Eulerian mass conservation equations to give time-dependent concentration and deposition fields. Models of this sort are particularly useful for modelling specific pollution episodes. The second group, with which we will be concerned here, comprises the statistical models which compute long-term mean properties of the concentration fields from long-term meteorological statistics.

The first type of model can also be used to compute long-term averages (if many model runs with varying inputs are computed), but this requires a great deal of input data and subsequent computation. Statistical models provide a much more efficient direct method for computing long-term averages. In addition to their computational advantages, the relative simplicity of statistical models can lead to greater physical insight into the problem.

In contrast to the time-dependent deterministic models which use an Eulerian approach, virtually all long-term statistical LRT models use a Lagrangian approach, at least implicitly. To compute concentration and deposition patterns these models use some form of the relationship between the Lagrangian dispersion statistics of a single particle subject to random transport and the (ensemble) average of the Eulerian concentration field given by the well-known formula (e.g. Lamb 1980):

\[ \langle c(x, t) \rangle = \int_{-\infty}^{t} dt' \int dx' p(x, t; x', t') S(x', t'). \]
Here $p(x, t; x', t')$ is the single-particle transition density defined as the probability density at time $t$ of the position $x$ of a particle which started at $x'$ at time $t'$; $c(x, t)$ is the concentration field and $S(x', t')$ is the source density.

Equation (1) is strictly correct only for inert particles. Since an atmospheric tracer may be either removed or transformed to a different form as it moves along its trajectory (for example, sulphur emitted initially as SO$_2$ may be oxidized to SO$_2^+$ or may be removed), a modification of Eq. (1) is required for the study of long-term concentration and deposition patterns of these substances.

To treat this case it is useful to think of the tracer as a generalized particle which can exist in one of $n$ states, and to denote the state of the particle by $Z(t)$ (where $Z(t)$ takes on the discrete values $1, 2, \ldots, n$). A particle trajectory is thus fully specified by the physical location $X(t)$ and state $Z(t)$, as functions of time.

The case of simple removal of aerosol particles is implicitly included in this formulation. For this case, $n = 2$, the particle has survived to time $t$ or has been removed. Although this simple case can be treated without explicitly considering multiple states (for example, with a 'source depletion assumption'), there are good reasons for treating all transformation and removal models of this sort within a unified framework. With such a treatment it is clear how models including only removal may be simply extended to more complex situations. Furthermore, few models of removal are truly single-state models; a common way to treat the difference between wet and dry removal rates is to assume that particles are either in a 'wet' state or a 'dry' state and to allow the removal rates to depend on the state of the particle. As we shall demonstrate, this idea can be usefully extended: by including more states in the model it is possible to describe more complex meteorological situations.

For the multiple-state case Eq. (1) readily generalizes to

$$\langle c_i(x, t) \rangle = \sum_j \int_{-\infty}^{t} dt' \int dx' p_{ij}(x, t; x', t')S_j(x', t')$$

(2)

where $p_{ij}(x, t; x', t')$ now gives the probability that a particle starting in state $j$ at $x'$ at time $t'$ will be in state $i$ at $x$ at time $t$, and $c$ and $S$ have been generalized to give concentrations and sources for each possible particle state. The determination of long-term mean concentrations is thus reduced to the problem of finding the joint transition density of $X(t)$ and $Z(t)$. Finding models for this pair of stochastic processes which are both physically reasonable and mathematically tractable is thus the central task of LRT modelling.

In the first section of this paper we briefly outline a few of the many LRT models which have been used and show how they can be put in the general framework outlined above. In the process we shall review some general features of finite state Markov processes, which have been used (at least implicitly) in all of these models. In the second section we shall describe a very useful approach to linking transport ($X(t)$) and other ($Z(t)$) processes that has seen relatively little use in the literature. We illustrate this approach with a general analytic model which uses multiple-state Markov processes to describe meteorological situations in which transport and removal are correlated. In the final section we illustrate the utility of this approach with an application to a two-layer model of the transport and removal of sulphur in a region of convective precipitation, demonstrating how the correlation of vertical transport and removal can affect deposition patterns.

Note that symbols used repeatedly in the text are summarized in appendix C.
2. BACKGROUND

(a) Formulation of stochastic LRT models

Numerous models have appeared in the recent literature which use some form of Eq. (2) to estimate long-term mean concentration and deposition fields of sulphur compounds. The models of Bolin and Persson (1975), Fisher (1975, 1978), Smith (1981) and Venkatram et al. (1982) are representative of such efforts. We will review very briefly the main features of these models; for brevity the above papers will be referred to by the abbreviations BP, F, Sm, and V respectively.

The general form of Eq. (2) can be simplified if the LRT processes (including sources) are assumed to be stationary in time. In this case we have

$$\langle c_i(x) \rangle = \sum_j \int_0^\infty dt' \int dx' \, p_{ij}(x, t; x', 0) S_j(x', 0).$$  \hspace{1cm} (3)

Since all models discussed here, and in subsequent sections, are stationary we will use this simpler form, and we will abbreviate $p_{ij}(x, t; x', 0)$ by $p_{ij}(x, t; x')$. In the event that transport and transformation are assumed to be spatially homogeneous so that $p_{ij}(x, t; x') = p_{ij}(x-x', t; 0)$ we will further simplify the expression for the transition density to $p_{ij}(x, t)$.

The transition density $p_{ij}$ gives the (joint) statistics of horizontal and vertical transport along with the statistics of removal or transformation of the particles. In practice, these processes must be separated in some way to make the problem tractable. In general the transition density $p_{ij}$ can be written

$$p_{ij}(x, t; x') = Q_{ij} \{X(0) = x', X(t) = x\} \rho(x, t; x')$$  \hspace{1cm} (4)

where $Q_{ij}$ gives the probability for transition between states $j$ and $i$ conditional on the particle reaching $x$ at time $t$, and $\rho$ is the unconditional transport probability density.

The simplest use of Eq. (4) assumes that transport and removal or transformation are statistically independent so that the transition probability $Q_{ij}$ does not depend upon the trajectory $X(t)$. This approach has been used by BP and V. Both F and Sm have used the more general form of Eq. (4) to model transport and removal of sulphur for a case where removal statistics depend in a simple way on wind direction (and hence on particle transport).

(b) Transport

With the separation of Eq. (4) the first task is to specify the statistical properties of $X(t)$. Just as it is possible to separate transport and transformation statistics using Eq. (4), it is possible to separate vertical and horizontal transport. In general these have been assumed to be independent; i.e., the relationship between vertical transport and removal/transportation and/or horizontal transport is largely ignored. For instance, BP assumed that the transport probability density had the Gaussian form

$$p(x, t; x') = p(z, t; z') \{2\pi \sigma_x(t) \sigma_y(t)\}^{-1} \exp \left[ -\frac{1}{2} \left( \frac{x - \mu_x(t)}{\sigma_x(t)} \right)^2 + \left( \frac{y - \mu_y(t)}{\sigma_y(t)} \right)^2 \right].$$ \hspace{1cm} (5)

and, by calculating a series of forward trajectories from measured 850 mb wind fields, estimated the functional forms of $\mu_x(t)$, $\mu_y(t)$, $\sigma_x(t)$ and $\sigma_y(t)$. Vertical transport was assumed to be a simple diffusion independent of all other processes.

V also used Eq. (5) but assumed simple analytic forms for the parameters. For this model it was assumed that vertical mixing was rapid enough that only horizontal transport need be considered explicitly.
A somewhat different approach to modelling horizontal transport was used by F and Sm. These authors assumed that the velocity on any given particle trajectory was a constant. This velocity was then allowed to be random, with the probability of a given velocity estimated from wind roses. F allowed only a few velocity states and associated a vertical eddy diffusivity with each state. The vertical transport was then described by a diffusion equation which was solved analytically. Because the eddy diffusivity depends on the velocity state there is some interdependence of horizontal and vertical transport in this model. Sm did not treat vertical transport.

(c) Removal/transformation

The treatment of the removal/transformation portion of Eq. (4) requires specifying the statistical properties of the finite state process \(Z(t)\). Before describing specific approaches we develop some general theory. We suppose initially that the rate at which particles in state \(j\) are transformed to state \(i\) is given as a fixed function of \(x\) and \(t\), \(\lambda_j(x, t)\). For an individual particle whose trajectory is \(X(t)\) (assumed fixed for the moment) and whose state is given by \(Z(t)\) this has the probabilistic interpretation

\[
Pr\{Z(t + dt) = i | Z(t) = j\} = \lambda_j(x, t) dt, \quad i \neq j.
\]

(6)

If it is assumed that this infinitesimal transition probability is independent of the previous states of the particle then \(Z(t)\) is an \(n\)-state Markov process with (non-stationary) infinitesimal transition probabilities given by Eq. (6) (see e.g. Karlin and Taylor 1975).

Since we will use finite state Markov models throughout this paper we briefly describe a few elementary aspects of the theory. Suppose \(Z(t)\) is an \(n\)-state Markov process with infinitesimal transition probabilities \(\lambda_j(t)\). Define the vector \(q(t)\) whose \(ith\) component is \(q_i(t) = Pr\{Z(t) = i\}\) and let

\[
R_j(t; t') = Pr\{Z(t) = i | Z(t') = j\}, \quad t > t'.
\]

(7)

Set \(\lambda_j(t) = -\sum_i \lambda_j(t)\) and let \(A(t)\) and \(R(t, t')\) be the matrices composed of the \(\lambda_j\)s and \(R_j\)s respectively. Then using matrix notation

\[
q(t) = R(t; t')q(t'), \quad t > t'.
\]

(8)

It is easy to show that \(R(t, t')\) can be written in terms of the matrix exponential

\[
R(t; t') = \exp\left\{\int_t^{t'} A(s) ds\right\}
\]

(9)

which may be easily evaluated in terms of the eigenvectors and eigenvalues of \(\int_0^t A(s) ds\) (see appendix A; note that the integral of the matrix is evaluated entry by entry).

In general the transformation rates \(\lambda_j(x, t)\) will be assumed to be random functions of space and time. Equation (9) gives the form of the conditional transition probabilities for a fixed trajectory through a fixed realization of \(A(x, t)\). Thus

\[
Q(t; t' | X(t) = x, X(t') = x') = \left\langle \exp\left\{\int_t^{t'} A(X(s), s) ds\right\} \right\rangle
\]

(10)

where the average is taken over all trajectories with the specified end points and all realizations of the random fields \(\lambda_j(x, t)\).

The direct use of Eq. (10) requires first some sort of model for the random transformation rates in an Eulerian frame and their relationship to Lagrangian transport statistics. A very simple approach (that used by BP) is to assume that the transformation
rates are fixed and constant in space and time so that the transition probability matrix of Eq. (10) depends only on $t - t'$:

$$Q(t - t') = \exp \{(t - t')\Lambda\}. \quad (11)$$

Perhaps due to the inherent difficulty of modeling the joint Lagrangian statistics of particle transport together with the Eulerian statistics of the random transformation rate fields, this simple approach is virtually the only direct use of Eq. (10) in the literature. More complex models can be constructed by directly assuming the Lagrangian statistics of the transformation rates seen on a particle trajectory.

Since removal/transformation rates can be much greater in wet conditions than in dry conditions it is important to model the sporadic nature of the precipitation fields encountered on a particle trajectory. This was demonstrated most clearly by Rodhe and Grandell (1972, 1981), who modelled the occurrence of wet and dry periods on a particle trajectory as a two-state stationary Markov process with mean lifetimes $\tau_w$ and $\tau_d$ in wet and dry states respectively. Removal rates were then either $\lambda_w$ or $\lambda_d$ depending on whether the particle was in a wet or dry state.

For this simple model it is useful to imagine that the particle can exist in one of three states—wet, dry or removed. It is not hard to see that the state of the particle $Z(t)$ is a three-state Markov process with transition probabilities

$$\Lambda = \begin{pmatrix}
-\lambda_w - \tau_w^{-1} & \tau_w^{-1} & 0 \\
\tau_w^{-1} & -\lambda_d - \tau_d^{-1} & 0 \\
\lambda_w & \lambda_d & 0
\end{pmatrix} \quad (12)$$

This approach is clearly applicable to the more general case of transformation and removal. Thus, for the case of sulphur, starting with the three-state model (SO$_2$, SO$_4$ and removed), allowing removal and oxidation rates to depend upon wet or dry conditions and assuming Markovian precipitation statistics, leads to a five-state Markov process—wet and dry SO$_2$ molecules and SO$_4$ particles, and a removed state.

This five-state model was used by V to calculate transformation and removal statistics for their LRT model. Note that while the transition rates among the original three states are not constant on a particle trajectory, the transition rates among the five states are. By adding more states the model is reduced to the simplest case so that transition probabilities can be computed from a straightforward extension of Eq. (11).

The Markov precipitation model was also used by F and Sm who treated geographic variations of rainfall statistics in a simple manner. For these models the mean durations of wet and dry periods were assumed to be functions of $x$, $\tau_w(x)$ and $\tau_d(x)$. By assuming a constant velocity on the trajectory from $x'$ to $x$ this spatial variability leads to a simply computed temporal variation of the wet/dry transition rates. For example, the wet–dry transition rate on the particle trajectory at time $t$ is

$$\lambda_{wd} = (\tau_w(U(t-t') + x'))^{-1} \quad (13)$$

where $U$ is the particle velocity along the trajectory. Thus for a fixed trajectory from $x'$ to $x$ the state of the particle is a finite state Markov process with non-constant infinitesimal transition probabilities $\lambda_{pi}(t)$. The transition probability matrix for fixed $x$ and $x'$ can thus be computed using Eq. (10).

All of these transport and removal models suffer from several related weaknesses. First, the linkage between the statistics of vertical transport, horizontal transport, and transformation/removal is generally weak. This is particularly true of vertical transport which in these models is treated independently of the other processes, while in fact vertical
air motions are strongly correlated with precipitation (and hence with transformation/removal). Although the models of F and Sm allow precipitation statistics to depend upon transport, they do so only by using a transport model which is both very simple—particle trajectories are straight lines—and rather awkward.

Second, to use Eq. (10) it is necessary to specify the statistics of transformation/removal on particle trajectories from (at best) knowledge of the statistics in an Eulerian frame (Rodhe and Grandell 1981). The common practice is to ignore the distinction and use the available Eulerian precipitation statistics. While there is some evidence to support this practice (Hamrud et al. 1981; Slinn et al. 1979), the effect of vertical motions on particle trajectories is not always clear (Rodhe and Grandell 1981; Fisher 1983). Both of these problems can be traced to the rather difficult task of specifying the Lagrangian removal or transformation statistics for all possible pairs of end points of particle trajectories as required by Eq. (4).

3. A CONDITIONAL TRANSPORT MODEL

(a) Conditional transport models

An alternative, but seldom used, way of linking transport and removal/transformation is to reverse the conditional and unconditional probabilities of Eq. (4), writing $p_{ij}$ as

$$p_{ij}(x, t; x') = q_{ij}(t; x')p(x, t; x|Z(0)=j, Z(t)=i)$$

(14)

where $q_{ij}$ is now the unconditional probability of transition between states $j$ and $i$ (for particles starting from $x'$) and $p$ now gives the transport statistics conditional on the transition. For many purposes Eq. (14) offers significant advantages over the more typically used Eq. (4). In this section we describe the use of Eq. (14) in LRT modelling. Using this formulation we develop a general class of analytic models which allow us to treat more explicitly many aspects of the correlation between vertical transport, horizontal transport and removal/transformation. Because many of the statistical properties of the model can be found (almost) analytically the approach is very useful for testing model sensitivity to parameter values or structural assumptions.

The ASTRAP model described by Shannon (1981) essentially uses a simple form of Eq. (14). This model uses measured wind and precipitation fields to compute (as functions of time after release) (a) first and second moments of horizontal particle positions (conditional on particle survival) and (b) unconditional survival probabilities. This is similar to the standard trajectory tracing approach used by Bolin and Persson (1975) or Sheih (1977), but now particle survival probabilities are estimated on each trajectory and the computed moments of particle position are weighted by these survival probabilities to give conditional moments. In the ASTRAP model these conditional moments for the positions of surviving particles were then used to approximate the conditional (horizontal) transport density of Eq. (14) by assuming the Gaussian form of Eq. (5).

This general approach can also be used for the case of multiple states. If moments of particle position can be computed conditional on particle beginning and end states, then the conditional density for each pair of states can be approximated by a Gaussian density; together with the unconditional state transition probabilities, Eq. (14) then yields the full joint single-particle statistics needed to compute long-term mean fields. Thus the transformation and removal of multiple-state species such as sulphur can easily be treated.

More importantly the notion of particle state can be generalized (as with the Markov wet/dry model discussed above), to simply model more complex meteorological situations. Our goal is to link vertical transport to horizontal transport and removal. In
particular, vertical transport can be modelled by assuming a finite number of layers and including the particle vertical position in $Z(t)$. Or the particle 'state' may be a general weather state, with characteristic wind and precipitation statistics. In the rest of this section we develop a general analytic model which illustrates the use of Eq. (14) in the multiple-state case.

This model has two essential aspects. First, in order to describe more effectively and simply the effect of complex meteorological situations on the joint statistics of transport and removal we introduce more states in the finite state process $Z(t)$, which we will assume to be Markov. Second, we allow the statistics of horizontal transport to depend upon the current state of the particle $Z(t)$. Using simple properties of finite state Markov processes we are able to compute the unconditional state transition probabilities and the conditional first and second moments of the particle positions. Assuming a Gaussian model we can then, using Eq. (14), compute approximate long-term mean concentration and deposition patterns. To be concrete and to motivate the formulation of the model we will consider explicitly a very simple special case as we proceed. In section 4 we apply the model to a more complex situation.

(b) The model

For motivation, then, we consider a simple extension of the Markovian wet/dry system in which the particle is subject to wet and dry removal at rates of $\lambda_w$ and $\lambda_d$ respectively. Now, however, we assume that the times when precipitation is and is not likely to occur are characterized by very different wind fields. To model this simply we assume two 'general weather states'—a 'fair' weather state with mean wind $u_f$ and a 'storm' weather state with mean wind $u_c$. We assume that the characteristic lifetime of the fair weather state is $T_f$ and that the fraction of time that the weather is fair is $\phi$. Precipitation occurs only within the storm weather state. The mean duration of wet periods is $\tau_w$; the fraction of time that precipitation occurs is $\rho$. This situation very roughly describes the situation in the Puget Sound region as described by Vong (1982). It can be summarized by the four-state model depicted in Fig. 1.

![Figure 1. Diagram of four-state model.](image)

Assuming that this four-state process $Z(t)$ is Markovian, the parameters given above simply determine the elements of the infinitesimal generator. With state numbers as in Fig. 1, setting $\psi = 1 - \phi - \rho$, it can be shown that

$$
\Lambda = \begin{bmatrix}
-T_f^{-1} - \lambda_d & \{T_f \psi\}^{-1} \phi & 0 & 0 \\
T_f^{-1} & -\{T_f \psi\}^{-1} \phi + \{\tau_w \psi\}^{-1} \rho + \lambda_d & \tau_w^{-1} & 0 \\
0 & \{\tau_w \psi\}^{-1} \rho & -\tau_w^{-1} - \lambda_w & 0 \\
\lambda_d & \lambda_d & \lambda_w & 0
\end{bmatrix}.
$$

(15)
This will be the first feature of our general model: the particle will be described first by
the random process $Z(t)$ which will be assumed to be an $n$-state Markov process with
constant generator $A$.

To model the dependence of horizontal transport on the weather state we will allow
the statistics of transport at time $t$ to depend on $Z(t)$. In general we assume that the
particle velocity has the form

$$u(t) = U_{Z(t)} + A_{Z(t)}u'(t)$$

where $U_i$ represents the mean velocity for particles in state $i$ and $u'(t)$ represents a random
velocity fluctuation with Lagrangian time scale $T_L$ satisfying

$$\begin{align*}
\langle u'(t) \rangle &= 0 \\
\langle u'(t)u'(t+s)^T \rangle &= \exp(-s/T_L)I
\end{align*}$$

where the superscript $T$ denotes the matrix transport. Note that with this formulation
the velocity fluctuation $u'(t)$ for a fixed trajectory is not affected by changes of particle
state. The fluctuation $A_{Z(t)}u'(t)$ that is added to the state-mean velocity does, however,
depend on the state and allows, via the matrices $A_i$, for variations of the scale and relative
direction of the velocity fluctuations. The Lagrangian time scale $T_L$ is assumed to
characterize the entire system and is independent of the state or state changes. For the
simple four-state model described above we have

$$U_1 = U_i; \quad U_2 = U_3 = U_v; \quad U_4 = 0;$$

for simplicity we will assume $A_i = \sigma I$ for $i = 1, 2, 3$ and, of course $A_4 = 0$ (since the fourth
state is the removed state). This is the second feature of the general model—the transport
statistics, conditional on the current state of the particle, are given by specifying the
particle velocity via Eq. (16). While this velocity model is somewhat oversimplified, it
gives a reasonably general, yet mathematically tractable approach to specifying a state-
dependent velocity.

Note that the velocity assumed in Eq. (16) specifies only horizontal transport.
Vertical transport can be modelled, as we have suggested above, with the finite state
process $Z(t)$. We will use this approach in section 4; for the simple four-state model
described here we keep things simple and ignore vertical transport.

This model is stationary in time and spatially homogeneous. Thus to compute the
long-term concentration statistics it suffices to consider a particle released at time $t = 0$
from $x = 0$. With the state of such a particle given by $Z(t)$ the position at time $t$ is

$$X(t) = \int_0^t u(t')dt' = \int_0^t (U_{Z(t)} + A_{Z(t)}u'(t'))dt'.$$

For this model it is possible to compute exactly certain joint statistics of $Z(t)$ and $X(t)$
and to use these to approximate the joint transport–transformation density needed for
Eq. (14).

(c) Statistical properties of $Z(t)$ and $X(t)$

Since $Z(t)$ is a finite state Markov process the statistical properties of $Z(t)$ may be
found easily by the methods sketched in section 2 and appendix A. In order to specify
fully the joint statistics of $X(t)$ and $Z(t)$ for the model it is thus only necessary to compute
the conditional density of $X(t)$ given $Z(t)$. While the direct computation of this density
is not easy, the computation of the conditional moments of $X(t)$ is rather straightforward;
in appendix A we derive explicit expressions for the first two moments conditional on the beginning and ending state of the particles:

\[
\mu_{i,j}(t) = \langle X(t)|Z(t)=i, Z(0)=j \rangle \tag{19a}
\]

\[
\Sigma_{i,j}(t) = \langle X(t)X(t)^T|Z(t)=i, z(t)=j \rangle - \mu_{i,j}(t)\mu_{i,j}(t)^T. \tag{19b}
\]

It is possible to extend these explicit formulae to compute higher moment tensors so that (formally at least) the complete conditional distribution of \(X(t)\) could be calculated. We do not pursue this, but instead assume a Gaussian form for this distribution, approximating the conditional transport probability by

\[
p(x,t|Z(t)=i, Z(0)=j) = (2\pi)^{-1/2}\left|\Sigma_{i,j}(t)\right|^{-1}\exp\left[-\frac{1}{2}(x-\mu_{i,j}(t))^T\Sigma_{i,j}(t)(x-\mu_{i,j}(t))\right]. \tag{20}
\]

\((d)\) Calculation of concentrations and deposition

Using the above results with Eq. (3) it is straightforward to calculate long-term concentration and deposition patterns for an arbitrary distribution of sources. For the sake of clarity in our examples we will discuss only the simple case of a point source of unit magnitude at the origin, with initial state probabilities \(q(0)\). In this case the (vertically integrated) concentration of particles in state \(i\) is simply

\[
\langle c_i(x) \rangle = \sum_j \int_0^\infty p_{i,j}(x, t)q_j(0)dt. \tag{21}
\]

The integral in Eq. (21) is calculated numerically on a grid using the approximations for \(p_{i,j}(x, t)\) calculated for a discrete set of times.

For the case of a single point source a slightly simpler approach can also be used. For given initial state probabilities \(q(0)\) we have for the joint location/state density:

\[
p_i(x, t) = \sum_j p_{i,j}(x, t)q_j(0) = q_i(t)p(x, t|Z(t)=i) \tag{22}
\]

where \(p\) now gives the location density conditional only on the end state. The Gaussian puff assumption can also be used to approximate this conditional density for each end state. The moments conditional on only the end state can be easily computed from the full conditional moments computed above:

\[
\mu_i(t) = \sum_j q_j(0)\mu_{i,j}(t) \tag{23a}
\]

\[
\Sigma_i(t) = \sum_j q_j(0)\Sigma_{i,j}(t). \tag{23b}
\]

With this notation Eq. (21) is now written

\[
\langle c_i(x) \rangle = \int_0^\infty p_i(x, t)dt. \tag{24}
\]

We will adopt this approach for the remainder of this paper, with \(p_i(x, t)\) approximated by a Gaussian density with parameters \(\mu_i(t)\) and \(\Sigma_i(t)\).

Because of the general meaning we have given to the notion of particle state some care is needed in the proper physical interpretation of Eq. (24). If the states are actual physical states of the 'particle' which could exist simultaneously in the same region (for example if the states are \(SO_2\) and \(SO_2^+\)) then Eq. (24) gives the long-term concentrations of these two forms of sulphur. For the simple four-state model we are considering the
situation is a bit more complicated. To compute the overall mean concentration for this model we need
\[ p_1(x, t) = p_1(x, t) + p_2(x, t) + p_3(x, t) \]
the overall probability density for the location of particles which have not been removed by time \( t \). Thus
\[ \langle c(x) \rangle = \sum_{i=1}^{3} \int_0^\infty p_i(x, t) dt = \sum_{i=1}^{3} \langle c_i(x) \rangle. \]

It may be of interest, in this case, to calculate the average concentration for, say, the fair weather state. This is not exactly \( \langle c_1(x) \rangle \). To calculate this concentration we would need the probability density for the location of particles conditional on the weather state. It is easy to see that for the fair weather state this is \( p_1(x, t)/q_1(0) \), where \( q_1(0) \) is the probability of occurrence of the fair weather state. Thus the average concentration during fair weather is \( \langle c_1(x) \rangle/q_1(0) \).

The spatial pattern of the total mean deposition rate due to a source of unit magnitude (kg s\(^{-1}\)) at the origin can be computed from the conditional density of the removed state. Assuming the \( n \)th state is the removed state, the mean deposition rate \( \langle D(x) \rangle \) is
\[ \langle D(x) \rangle = \sum_{j \neq n} \lambda_{jn} \langle c_j(x) \rangle = \int_0^\infty \sum_{j \neq n} \lambda_{jn} p_j(x, t) dt = \int_0^\infty dp_n(x, t) = \lim_{t \to \infty} p_n(x, t). \]

That is, the mean deposition rate in the steady state is just the conditional density of the removed state.

The first two central moments of the deposition rate are therefore
\[ \lim_{t \to \infty} \mu_n(t) \quad \text{and} \quad \lim_{t \to \infty} \Sigma_n(t) \]
which can be computed from the formulae given in appendix A. This approximate treatment gives only a rough picture of deposition but is easily computed and gives a succinct summary of the deposition pattern. It is thus useful for comparing results of different models or for assessing model sensitivity to parameter values.

A complete calculation of deposition rate (within the accuracy of the Gaussian puff approximation) is also quite simple. For example, for the four-state model we may calculate the mean wet deposition rate as
\[ \langle D_w(x) \rangle = \int_0^\infty \lambda_w p_3(x, t) dt = \lambda_w \langle c_3(x) \rangle \]
since \( p_3(x, t) \) gives the probability that a particle is in a region of precipitation and is at \( x \) at time \( t \), and \( \lambda_w \) gives the wet removal rate. Similar formulae are easily found to compute dry or total deposition rates.

Examples of the results of such calculations for wet and dry deposition are presented for the four-state model in Fig. 2. The parameters used, summarized in Table 1, have been chosen to be consistent with the results presented by Vong (1982) for precipitation and wind fields in the Puget Sound region. The pattern of both wet and dry deposition is distorted about the source, due to the mean south-south-easterly winds. Note, however, that this distortion is much greater for wet deposition, reflecting the correlation of transport and wet removal in the model.
Figure 2a. Mean wet deposition rate for four-state model \( (\times 10^{-7}\text{kg}/(\text{km}^2\text{s})) \).

Figure 2b. Mean dry deposition rate for four-state model \( (\times 10^{-7}\text{kg}/(\text{km}^2\text{s})) \).

**TABLE 1. Parameters for four-state model**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \hat{\lambda}_d )</td>
<td>( 10^{-2}\text{s}^{-1} )</td>
</tr>
<tr>
<td>( \hat{\lambda}_w )</td>
<td>( 10^{-2}\text{s}^{-1} )</td>
</tr>
<tr>
<td>( r_w )</td>
<td>( 2.0 \times 10^3\text{s} )</td>
</tr>
<tr>
<td>( T_f )</td>
<td>( 2.0 \times 10^2\text{s} )</td>
</tr>
<tr>
<td>( \rho )</td>
<td>0.1</td>
</tr>
<tr>
<td>( \gamma )</td>
<td>0.5</td>
</tr>
<tr>
<td>( V_f )</td>
<td>(0.0, 0.0) m s(^{-1})</td>
</tr>
<tr>
<td>( V )</td>
<td>(3.8, 9.2) m s(^{-1})</td>
</tr>
<tr>
<td>( \sigma^2 )</td>
<td>25 m(^2) s(^{-2})</td>
</tr>
</tbody>
</table>

4. **AN APPLICATION: A MULTI-STATE MODEL FOR A REGION OF CONVECTIVE PRECIPITATION**

(a) *The two-layer model*

To illustrate more fully the utility of the general model formulated in section 3 we now consider a more complex application, one which treats the relationship between vertical transport and wet removal of sulphur in a region of convective precipitation. We compare the results of this model with the more usual models which separate out or
ignore vertical transport and show how including this linkage can affect the Lagrangian precipitation statistics seen on a typical 'particle' trajectory. Finally, we demonstrate how the sensitivity of the model to changes in parameter values can easily be assessed.

The model proposed here is essentially a two-layer model with wet and dry regions. The lower layer represents the boundary layer with the upper layer representing the levels between cloud base and the top of the mixed layer. In the dry region a generalized 'particle' can thus be in one of two states depending on which layer it is in. Vertical transport in dry conditions is modelled by transitions between these two states. The storm region includes three states—in-cloud and below-cloud states and a near-cloud subsiding state. Note that particles in the near-cloud region which neither subside nor enter the cloud are considered to have remained in a dry, non-storm state. Particles from either the upper or lower dry layers can enter the in-cloud or below-cloud regions where wet removal may occur. The near-cloud state is used to model vertical transport from the upper to the lower layer which is directly associated with a storm but does not occur in a region of actual precipitation. Once in the cloud, particles can be rained out, can enter the dry upper layer when the cloud evaporates, or can enter the dry lower layer via evaporation of precipitation in the lower layer. Particles in the below-cloud region can be washed out or can enter the dry lower layer when the storm dissipates. Including the removed state the model is thus a six-state model; these states and the allowed transitions among them are summarized in Fig. 3.

![Diagram of six-state model.](image)

There are fourteen transition rates which must be specified for this model and some are not readily measurable. With a few simplifying assumptions, however, it is possible to reduce these to a smaller number of more easily estimatable parameters. The details of this reparametrisation are discussed in appendix B. In addition to the dry removal rate ($\lambda_d$), the washout and rainout rates ($\lambda_w$ and $\lambda_r$) and the parameters describing the precipitation statistics, $\rho$ (fraction of time during which precipitation occurs) and $\tau_w$ (mean storm duration), we will need five parameters. These are $\tau_L$ (the time scale for transfer of particles from the lower to upper layer in dry conditions), $\xi_L$ (the fractional lower layer thickness), $F$ (the fraction of cloud air that comes from the lower layer), $\kappa$ (the convergence rate in the below-cloud region, averaged over the storm lifetime) and $\chi$, the fraction of the surviving (i.e. not washed out) in-cloud sulphate particles which enter the lower layer via below-cloud evaporation of precipitation.
Estimates of $F$ for convective clouds have been obtained by Jensen et al. (1985) and Austin et al. (1985), and approximate values for $\kappa$ are readily available (Ackerman et al. 1983; Kuo and Anthes 1984). Assuming that the sulphate particles are all bound to cloud water and neglecting interstitial SO$_2$, $\chi$ can be estimated from the results presented by Gamache and Houze (1982).

To complete the model specification we must also give the transport parameters. In this example we model the effect of vertical shear by allowing the transport parameters to depend on the layer. Following Bolin and Perrson (1975), we assume that velocities in the lower layer are reduced by one third and turned 30 degrees to the left over those in the upper layer. In terms of the parameters $A$ and $U$ of Eq. (16) we have then

$$
A_2 = A_4 = A_5 = \frac{3}{2} \sigma \begin{vmatrix} \cos 30^\circ & \sin 30^\circ \\ \sin 30^\circ & \cos 30^\circ \end{vmatrix},
$$

$$
A_1 = A_3 = \begin{vmatrix} \sigma & 0 \\ 0 & \sigma \end{vmatrix},
$$

$$
U_3 = U_1, \quad U_2 = U_4 = U_5 = (1/\sigma) A_2 U_1,
$$

where $U_1$ is the upper layer mean velocity.

This model treats the linkage between vertical transport and particle removal during a precipitation event explicitly. A much simpler and more typically used model which ignores this linkage arises as a special case. When $\kappa = 0$ (no convergence), $F = 0$ (all cloud air comes from the upper layer) and $\chi = 0$ (no below-cloud evaporation) the model is essentially a two-layer version of the simple Markov wet/dry model. If in addition the velocity statistics are the same for the two layers then this model completely decouples the vertical transport, horizontal transport and removal processes.

(b) Model results

In order to assess the effect of this decoupling we have compared the results obtained from the simple model with those obtained with the full model with more reasonable values of the parameters. Table 2 gives numerical values of the parameters used for the comparison. Note that some parameters can be varied only in a narrow range if all other

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\lambda_d$</td>
<td>$10^{-5}$s$^{-1}$</td>
<td>Venkatram et al. (1982)</td>
</tr>
<tr>
<td>$\lambda_w$</td>
<td>$10^{-4}$s$^{-1}$</td>
<td></td>
</tr>
<tr>
<td>$\lambda_t$</td>
<td>$10^{-5}$s$^{-1}$</td>
<td></td>
</tr>
<tr>
<td>$\rho$</td>
<td>0.1</td>
<td>Jensen et al. (1985)</td>
</tr>
<tr>
<td>$\tau_w$</td>
<td>10$^5$s</td>
<td>Austin et al. (1985)</td>
</tr>
<tr>
<td>$\tau_t$</td>
<td>10$^3$s</td>
<td>Ackerman et al. (1983)</td>
</tr>
<tr>
<td>$\tau_{sc}$</td>
<td>0.333</td>
<td>Kuo and Anthes (1984)</td>
</tr>
<tr>
<td>$F$</td>
<td>0.8</td>
<td>Gamache and Houze (1982)</td>
</tr>
<tr>
<td>$\kappa$</td>
<td>1.6 x 10$^{-4}$s$^{-1}$</td>
<td></td>
</tr>
<tr>
<td>$\chi$</td>
<td>0.5</td>
<td></td>
</tr>
<tr>
<td>$U_1$</td>
<td>(5.0, 0.0) m s$^{-1}$</td>
<td>Bolin and Perrson (1975)</td>
</tr>
<tr>
<td>$\sigma^2$</td>
<td>0.3 x 10$^4$m$^2$s$^{-2}$</td>
<td></td>
</tr>
<tr>
<td>$\tau_{sc}$</td>
<td>0.18 x 10$^3$s</td>
<td></td>
</tr>
</tbody>
</table>
parameters are fixed; e.g. if $F$, $r_w$ and $\zeta_L$ are fixed the net vertical transport varies between a fixed maximum and minimum corresponding to the cases that the below-cloud air comes all from the lower or upper layers, respectively. This requirement thus constrains the convergence rate, $\kappa$. The parameter values in Table 2 have been adjusted to give a physically realizable system reasonably consistent with published measurements. References cited were used only as guidance in choosing these parameter values; the exact values chosen are not to be attributed to the cited author. Model sensitivity to the chosen parameters will be discussed subsequently.

All parameters for the simple model are identical except for $\kappa$, $F$, and $\gamma$. In addition, the average of the transport statistics for the two layers of the full model were used for both layers of the simple model so that horizontal transport was independent of particle height.

Patterns of deposition and of vertically integrated concentration for each of the two layers were computed using the Gaussian puff model outlined in section 3. Results are presented in Fig. 4. Although the general patterns of mean deposition and concentration

![Figure 4](image-url)

Figure 4. Concentration and deposition patterns for full (solid contours) and simple (dashed contours) models: (a) wet deposition ($\times 10^{-6} \text{kg/(km}^2\text{s})$); (b) concentration: lower layer ($\times 10^{-5} \text{kg/(km}^2\text{s})$); (c) concentration: upper layer ($\times 10^{-5} \text{kg/(km}^2\text{s})$).
are similar for the two models, there are systematic differences. For the full model, wet deposition levels are substantially higher in a region about 500 km in diameter near the source and slightly (10–20%) lower in the large area far from the source. The plots of concentration in the two layers are consistent with this picture: concentrations are lower everywhere for the full model in the lower layer and everywhere except very near the source in the upper layer. Note that the deposition and concentration patterns for the full model are also turned slightly to the north.

The differences between the results for the two models are readily explained. The convergence in the lower layer and divergence in the upper layer in the vicinity of storms causes substantially different precipitation statistics for particles in the two layers. For the parameters of Table 2 the net transition rate from the lower dry layer into one of the two wet states is

\[ \lambda_{32} + \lambda_{52} = 0.29 \times 10^{-4} \text{s}^{-1} \]

while the analogous quantity for the upper layer is

\[ \lambda_{31} + \lambda_{51} = 0.23 \times 10^{-5} \text{s}^{-1}. \]

The overall average dry–wet transition rate for particles mixed randomly through both layers is still \((1 - \rho) \tau_{w} / \rho_{j}^{-1} = 0.11 \times 10^{-4} \text{s}^{-1}\), but if particles are not randomly distributed between the two layers this no longer holds. When all particles are released in the lower layer (as we have assumed for our model calculations) the precipitation statistics are substantially different for a typical particle until the particles have been mixed through the two layers. Thus in the full model wet deposition is enhanced near the source, and overall particle lifetimes are decreased leading to lower concentrations and lower deposition away from the source.

(c) Model sensitivity

The sensitivity of the model results to variations in parameter values can be easily assessed by calculating the moments of the deposition pattern as described in section 3. We have varied the parameters \(\tau_{L}, \tau_{w}, F, \kappa, \text{and } \chi\), leaving other parameters as in Table 2. For the most part, parameters were varied one at a time, although as discussed above, this was not always possible. The parameters varied, values tried and the resulting deposition centroids are summarized in Table 3; selected results are plotted in Fig. 5.

The detailed results of this model are, not surprisingly, rather sensitive to the dry mixing time scale (see Fig. 5). For very small \(\tau_{L}\) the two models give similar results; with rapid mixing between layers, the distinction between the two models is unimportant. For very large \(\tau_{L}\) the difference between the two models becomes very large and the centroid of deposition moves toward the source.

In addition the model results are rather sensitive to the parameter \(\chi\), which is a measure of the cloud-to-lower-layer flux. If \(\chi = 1.0\) (all in-cloud tracer enters the lower layer) the centroid of deposition moves toward the source and the difference between models increases. If \(\chi = 0.0\) (all in-cloud tracer remains in the upper layer) then particles in the cloud that survive the storm enter a region in which particle lifetimes are long. Thus, in this case the centroid of deposition moves further out and the difference between the models decreases.

Changing the storm time scale \(\tau_{w}\) has a similar (and expected) effect on both models (see Table 3 and Fig. 5). All other parameters varied had little effect and are not plotted in Fig. 5.
Figure 5. Plot of deposition centroids for parameter sensitivity tests.

<table>
<thead>
<tr>
<th>Run No.</th>
<th>Parameter(s) varied</th>
<th>Values</th>
<th>Deposition centroid $(x, y)$ (km)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1a</td>
<td>$\tau_0$</td>
<td>$1.0 \times 10^5$ s</td>
<td>(331.9, 71.6)</td>
</tr>
<tr>
<td>1b</td>
<td></td>
<td></td>
<td>(444.7, 57.5)</td>
</tr>
<tr>
<td>2a</td>
<td></td>
<td>$1.0 \times 10^6$ s</td>
<td>(242.4, 73.0)</td>
</tr>
<tr>
<td>2b</td>
<td></td>
<td></td>
<td>(341.2, 44.1)</td>
</tr>
<tr>
<td>3a</td>
<td></td>
<td>$5.0 \times 10^5$ s</td>
<td>(255.7, 72.8)</td>
</tr>
<tr>
<td>3b</td>
<td></td>
<td></td>
<td>(363.2, 46.9)</td>
</tr>
<tr>
<td>4a</td>
<td></td>
<td>$5.0 \times 10^6$ s</td>
<td>(385.7, 70.8)</td>
</tr>
<tr>
<td>4b</td>
<td></td>
<td></td>
<td>(477.5, 61.7)</td>
</tr>
<tr>
<td>5a</td>
<td>$1.0 \times 10^5$ s</td>
<td></td>
<td>(497.3, 69.1)</td>
</tr>
<tr>
<td>5b</td>
<td></td>
<td></td>
<td>(517.8, 66.9)</td>
</tr>
<tr>
<td>6a</td>
<td>$\tau_o$, $\kappa$</td>
<td>$\tau_o = 2 \times 10^4$ s, $\kappa = 0.8 \times 10^{-3}$ s$^{-1}$</td>
<td>(427.4, 80.7)</td>
</tr>
<tr>
<td>6b</td>
<td></td>
<td></td>
<td>(517.8, 66.9)</td>
</tr>
<tr>
<td>7a</td>
<td></td>
<td>$\tau_o = 5 \times 10^4$ s, $\kappa = 3.2 \times 10^{-3}$ s$^{-1}$</td>
<td>(271.2, 58.0)</td>
</tr>
<tr>
<td>7b</td>
<td></td>
<td></td>
<td>(383.3, 49.5)</td>
</tr>
<tr>
<td>8</td>
<td>$\chi$</td>
<td>1.0</td>
<td>(392.1, 70.6)</td>
</tr>
<tr>
<td>9</td>
<td></td>
<td>0.0</td>
<td>(273.2, 72.5)</td>
</tr>
<tr>
<td>10</td>
<td>$F$</td>
<td>1.0</td>
<td>(328.3, 66.9)</td>
</tr>
<tr>
<td>11</td>
<td></td>
<td>0.9</td>
<td>(324.7, 67.3)</td>
</tr>
<tr>
<td>12</td>
<td></td>
<td>0.7</td>
<td>(320.7, 69.4)</td>
</tr>
<tr>
<td>13</td>
<td>$\kappa$</td>
<td>$1.0 \times 10^{-4}$ s$^{-1}$</td>
<td>(318.9, 76.3)</td>
</tr>
<tr>
<td>14</td>
<td></td>
<td>$0.6 \times 10^{-4}$ s$^{-1}$</td>
<td>(324.1, 80.2)</td>
</tr>
</tbody>
</table>

Runs 1–7 (a) are for full model; runs 1–7 (b) are corresponding results for simple model; other runs are done only for full model. Parameters for runs 1a and 1b are as in Table 2. For other runs, all parameters not varied are as in Table 2.
(d) Discussion of model results

For an incompressible (statistically) homogeneous precipitation statistics (not necessarily independent of the flow) it is not hard to show that the average fraction of time during which precipitation occurs is the same in both Lagrangian and Eulerian frames (the proof of this is identical to the proof that the Eulerian and Lagrangian velocity variances are identical for stationary homogeneous turbulence (see Tennekes and Lumley 1972)). For incompressible flow (as one has with two-dimensional geostrophic winds) the principle difference between precipitation statistics will be proportional changes of equal magnitude in the mean length of wet and dry periods. A simple analysis of this effect, due to the relative motions of clouds, particles and stationary observers has been sketched by Rodhe and Grandell (1981). Limited comparisons of precipitation statistics calculated from horizontal wind fields have found little difference between measured Eulerian and Lagrangian statistics (Hamrud et al. 1981; Slinn et al. 1979).

However, as these example model calculations show there are factors other than relative motion of storms and particles which can cause differences between Eulerian and Lagrangian statistics.

With the two-layer model we do not have incompressibility within each layer. In the lower layer, convergence occurs in the region of precipitation, effectively decreasing the mean dry time on particle trajectories without decreasing the mean wet time. Conversely, for particles in the upper layer divergence in the vicinity of the convective storm keeps upper layer particles from entering the storm directly and increases the mean dry time for particles in this layer without increasing the mean wet time.

We thus see that organized vertical transport may modify Lagrangian statistics of particles in a way that is not readily detected from an analysis of the joint statistics of horizontal wind and precipitation fields.

5. Conclusions

We have developed a general formalism for multiple-state transport models. With this formalism we have developed an analytic model which has been used to demonstrate an important effect due to the linkage between vertical transport and removal of sulphur compounds in a region of convective precipitation.

Although the specific model developed is somewhat schematic, we believe that the approach described here is quite general in its utility. The idea of describing a particle trajectory by the position–state pair \((X(t), Z(t))\) has often been used implicitly; with our model we have demonstrated the value of adopting the approach more explicitly. In particular we have seen that when vertical transport is treated in this manner it can be linked with other processes such as removal.

The 'conditional Gaussian puff' approach we have used to approximate the joint transition–transport density should also be applicable to more general situations. Shannon's (1981) application of this method to a data-intensive model based on trajectory tracing could be extended to the multiple-state case described here, allowing, for example, vertical transport to be linked to horizontal transport and removal in an empirical model.

The schematic model we have presented is perhaps most useful as a tool for investigating the sensitivity of a model to assumptions about model structure or parameter values. Multi-state Markov models can, as our examples have illustrated, be used to capture very roughly a variety of meteorological situations. This provides an inexpensive approach to testing the importance of a variety of physical processes for LRT models.
ACKNOWLEDGMENT

This work was partially supported by NSF grant ATM8303636.

APPENDIX A

In this appendix we review a few basic facts about finite state Markov processes and derive expressions for the conditional moments $\mu_i(t)$ and $\Sigma_i(t)$ of section 2. First, let $q(t), R(t; t')$ and $\Lambda(t)$ be the time-dependent state probability vector, transition probability matrix and infinitesimal transition probability matrix, respectively. $\Lambda(t)$ is called the generator for the process.

Using the definition of conditional probability, together with Eq. (6), it can be seen that $q(t)$ satisfies the system of $n$ linear differential equations

$$\frac{dq(t)}{dt} = \Lambda(t)q(t). \tag{A1}$$

Subject to the initial condition $q(t')$, this equation has the formal solution for $t > t'$

$$q(t) = \exp\left\{\int_{t'}^t \Lambda(s) \, ds\right\} q(t').$$

Thus from the definition of $R(t; t')$

$$R(t; t') = \exp\left\{\int_{t'}^t \Lambda(s) \, ds\right\}. \tag{A2}$$

In practice the exponential matrix in Eq. (A2) can be easily evaluated with a digital computer. Let $\Phi_i$ and $\gamma_i$ be the eigenvectors and associated eigenvalues of the matrix $\int_{t'}^t \Lambda(s) \, ds$. Assuming that there are $n$ distinct eigenvectors, we let $\Phi$ be the matrix whose $i$th column is $\Phi_i$ and we let $\Gamma$ be the diagonal matrix $\Gamma = \text{diag}(\exp(\gamma_i))$. Then

$$\exp\left\{\int_{t'}^t \Lambda(s) \, ds\right\} = \overline{\Phi} \Gamma \overline{\Phi}^{-1}. \tag{A3}$$

(If there are not $n$ distinct eigenvectors Eq. (A3) must be modified slightly. We will not pursue this here; see Karlin and Taylor (1975).)

If the infinitesimal transition rates are independent of $t$ the computation is simpler since in this case

$$\int_{t'}^t \Lambda(s) \, ds = (t - t')\Lambda \tag{A4}$$

and the eigenvectors needed for Eq. (A3) are independent of $t$ and are just the eigenvectors of the (constant) generator $\Lambda$. In this case the matrix $\Gamma$ has the simple time dependence

$$\Gamma(t) = \text{diag}(\exp(\gamma_i t)) \tag{A5}$$

where the $\gamma_i$s are the eigenvalues of the generator.

This representation of the transition probability matrix can be used to compute the conditional moments of particle positions for the stationary model discussed in section 3. For the conditional means we have, using Eq. (15)
\[ \mu_{ij}(t) = \langle X(t) | Z(t) = i = 0 \rangle = \left\{ \int_0^t dt' U_{Z(0)} Z(t) = i \right\} \]

\[ = \sum_{k=1}^n U_k \int_0^t dt' Pr\{Z(t') = k | Z(t) = i, Z(0) = j\} \]  
\[ = \{1/q_i(t)\} \sum_k U_k \int_0^t R_{ik}(t-t')R_{kj}(t')dt'. \]  

(A6)

In the last step we have used the definition of conditional probability. The integral in Eq. (A6) can be evaluated by first writing the transition probabilities in terms of the eigenvalues and eigenvectors of the generator

\[ R_{ik}(t') = \sum_{l} \psi_{il} \psi_{ik}^{-1} \exp[\gamma_i t] \]

(where \( \psi_{ik}^{-1} \) is the lk element of \( \psi^{-1} \)), so that

\[ \int_0^t R_{ik}(t-t')R_{kj}(t')dt' = \sum_{lm} \psi_{il} \psi_{ik}^{-1} \psi_{km} \psi_{mj}^{-1} \int_0^t \exp[\gamma_l t + (\gamma_m - \gamma_i) t'] dt'. \]  

(A7)

Thus writing

\[ T_{lm}(t) = \begin{cases} (\gamma_i - \gamma_m)^{-1}\{\exp(\gamma_i t) - \exp(\gamma_m t)\} & \text{if } \gamma_i \neq \gamma_m \\ t \exp(\gamma_i t) & \text{if } \gamma_i = \gamma_m \end{cases} \]

for the integral in Eq. (A6) we have

\[ \mu_{ij}(t) = \{1/q_i(t)\} \sum_{klm} U_k \psi_{il} \psi_{ik}^{-1} \psi_{km} \psi_{mj}^{-1} T_{lm}(t). \]  

(A8)

The computation of the 2×2 matrix of conditional second moments is similar:

\[ \langle X(t)X(t)^\top | Z(t) = i, Z(0) = j \rangle = \]

\[ \left( \left( \int_0^t U_{Z(\gamma)} dt' \right) \right)^\top \left( \int_0^t U_{Z(\gamma)} dt' \right) | Z(t) = i, Z(0) = j \rangle + \]

\[ + \left( \left( \int_0^t A_{Z(\gamma)} u(t') dt' \right) \right)^\top \left( \int_0^t A_{Z(\gamma)} u(t') dt' \right) | Z(t) = i, Z(0) = j \rangle. \]  

(A9)

Setting

\[ S_{aqn}(t) = \frac{\exp(\gamma_q t)}{(\gamma_q - \gamma_p)(\gamma_q - \gamma_n)} + \frac{\exp(\gamma_p t)}{(\gamma_p - \gamma_q)(\gamma_p - \gamma_n)} + \frac{\exp(\gamma_n t)}{(\gamma_n - \gamma_p)(\gamma_n - \gamma_q)} \]  

(A10)

and proceeding as above, the first term in Eq. (A9) is found to be

\[ \frac{1}{q_i(t)} \sum_{lm} \sum_{npq} \left[ U_l U_m^\top + U_m U_l^\top \right] \left[ \psi_{ln} \psi_{lm}^{-1} \psi_{lp} \psi_{pm}^{-1} \psi_{qm} \psi_{pq}^{-1} S_{aqn}(t) \right]. \]  

(A11)

Defining \( S'_{aqn}(t) \) analogously to \( S_{aqn}(t) \) by replacing \( \gamma_p \) in Eq. (A10) with \( \gamma_p' = \gamma_p - 1/T_L \) where \( T_L \) is the Lagrangian time scale, and again proceeding as for the first moments we find that the second term in Eq. (A9) is

\[ \{1/q_i(t)\} \sum_{lm} \sum_{npq} \left[ A_l A_m^\top + A_m A_l^\top \right] \left[ \psi_{ln} \psi_{lm}^{-1} \psi_{lp} \psi_{pm}^{-1} \psi_{qm} \psi_{pq}^{-1} S'_{aqn}(t) \right]. \]  

(A12)
Expressions (A11) and (A12) together then give an expression for

\[ \langle X(t)X(i)^T Z(t) = i, Z(0) = j \rangle \]

from which the particle position covariance matrix \( \Sigma_0(t) \) can be calculated.

**APPENDIX B**

In this appendix we reparametrize the finite state Markov model of section 5. We will show that in addition to the removal rates \( \lambda_3, \lambda_4, \lambda_5 \) and the precipitation parameters \( \rho \) and \( \tau_w \), one must specify five parameters: the dry vertical transport rate \( \tau_L^{-1} \), the relative lower layer thickness \( \zeta_L \), the composition of cloud air \( F \), the average convergence rate in the lower layer of the storm \( \kappa \), and the fraction of cloud water leaving the cloud by below-cloud evaporation, \( \chi \). With some simplifying assumptions the fourteen transition rates can be expressed in terms of these parameters.

First note that the dry deposition rate and the washout and rainout rates directly give the transition rates \( \lambda_{62}, \lambda_{65} \) and \( \lambda_{63} \), respectively. Also, the inverse of the storm duration gives the rate at which particles leave storm states 3, 4 and 5 so that

\[ \lambda_{24} = \lambda_{25} = \tau_w^{-1} \quad \text{and} \quad \lambda_{13} + \lambda_{23} = \tau_w^{-1} \]  

(B1)

The parameters \( \rho \) and \( \zeta_L \) further constrain, but do not uniquely determine, the dry-to-wet transition rates. In addition, \( \zeta_L \) with \( \tau_L \) determine completely the two dry vertical transition rates. By definition \( \lambda_{12} = \tau_L^{-1} \) and the requirement of mass balance can be used to find \( \lambda_{21} \). Specifically, if we assume that the transport statistics of tracers are the same as those of the embedding air masses in the dry state then the vertical flux rate of air from layer two to layer one is \( \zeta_L \lambda_{12} \); the flux rate in the opposite direction is \( \zeta_U \lambda_{21} \). Mass balance then forces equality of these flux rates so that

\[ \lambda_{21} = \zeta_L \lambda_{12} / \zeta_U. \]  

(B2)

A reasonable parametrization of the remaining transition rates, in terms of estimatable quantities, is a bit more difficult.

In order to do this we initially define some additional parameters. First consider the air movement into and out of the storm. Let \( \alpha \) be the fraction of the storm air that originates from the lower layer. A fraction \( \beta \) goes into the cloud region and a fraction \( (1 - \beta) \) goes into the below-cloud region. Of the air originally from the upper layer, a fraction \( \delta \) goes to the below-cloud region, and a fraction \( \gamma \) subsides into the dry region; the remainder \( 1 - (\gamma + \delta) \) goes into the cloud region. We first show that these three parameters can be expressed in terms of \( F, \kappa \) and \( \zeta_L \) if some simplifying assumptions are made.

We assume, then, that the relative thickness \( \zeta_L \) is the same for all meteorological conditions. In addition, we assume that at the end of the storm the air in the dry upper layer and that all below-cloud air goes into the lower region. Note that sulphate particles from the cloud may still go into the lower layer via below-cloud evaporation. Now the air leaving the storm and entering the upper layer all comes from the cloud. Mass balance requires that the fraction of storm air that goes back into the upper layer must equal the fraction of storm air that came from the upper layer, \( (1 - \alpha) \). Thus the fraction of storm air that is in the cloud is \( (1 - \alpha) \). Since a fraction \( F \) of this came from the lower layer we have, recalling the definition of \( \alpha \) and \( \beta \),

\[ F(1 - \alpha) = \alpha \beta. \]  

(B3)
Since a fraction \((1 - F)\) of the in-cloud air is from the upper layer the total fraction of storm air that came from the upper layer and went into the cloud is \((1 - \alpha)(1 - F)\). But from the definitions of \(\gamma\) and \(\delta\) it immediately follows that

\[
\gamma + \delta = F. \tag{B4}
\]

Using the assumption that the ratio of in-cloud to below-cloud air is given by the dry layer ratio \(\xi_U/\xi_L\) leads to a third equation

\[
(1 - \alpha)/\{(1 - \beta)\alpha + (1 - \alpha)\delta\} = \xi_U/\xi_L. \tag{B5}
\]

The average convergence rate in the storm region can be used to obtain a fourth equation. An average convergence rate of \(\kappa\) in the layer of depth \(\xi_L\) for the duration of the storm gives a total vertical mass transport of \(\kappa \xi_L \tau_w\) per unit cloud area (note that this quantity is dimensionless because we are using a dimensionless layer thickness). This must be compensated by subsidence from the upper layer. The volume per unit cloud area of upper layer air in the storm is \(\xi_U\). A fraction \(\gamma\) of this subsides into the lower layer. Hence

\[
\xi_U \gamma = \kappa \xi_L \tau_w. \tag{B6}
\]

These four equations can be solved in terms of \(F, \kappa\) and \(\xi_L\) to give

\[
\gamma = \xi_L \kappa \tau_w / \xi_U \tag{B7}
\]

\[
\delta = F - \xi_L \kappa \tau_w / \xi_U \tag{B8}
\]

\[
\alpha = \xi_L (1 + \kappa \tau_w) / (1 + \xi_L \kappa \tau_w) \tag{B9}
\]

\[
\beta = F(1 - \alpha)/\alpha. \tag{B10}
\]

The definitions of \(\alpha, \beta, \gamma\) and \(\delta\) respectively imply

\[
\xi_L (1 - \alpha)(\lambda_3 + \lambda_2) = \xi_U \alpha(\lambda_{51} + \lambda_{31} + \lambda_{41}) \tag{B11}
\]

\[
\beta \lambda_2 = (1 - \beta)\lambda_3 \tag{B12}
\]

\[
\lambda_{41} = \gamma(\lambda_{31} + \lambda_{41} + \lambda_{51}) \tag{B13}
\]

\[
\lambda_{51} = \delta(\lambda_{31} + \lambda_{41} + \lambda_{51}). \tag{B14}
\]

Balancing the flux rate of air entering the wet region against the flux rate of air leaving the wet region leads to the equation

\[
(1 - \rho)\xi_U (\lambda_{51} + \lambda_{31}) + \xi_L (\lambda_{32} + \lambda_{52}) = \rho \tau_w^{-1}. \tag{B15}
\]

Letting

\[
\tau_d = \{(1 - \rho)/\rho\}[1 - \gamma(1 - \alpha)]\tau_w
\]

and solving Eqs. (B11)–(B15) for the transition rates gives

\[
\lambda_{52} = \alpha(1 - \beta)/\{\tau_d \xi_L\} \tag{B16}
\]

\[
\lambda_{32} = \alpha \beta/\{\tau_d \xi_L\} \tag{B17}
\]

\[
\lambda_{31} = (1 - F)(1 - \alpha)(\tau_d \xi_U) \tag{B18}
\]

\[
\lambda_{41} = \gamma(1 - \alpha)/\{\tau_d \xi_U\} \tag{B19}
\]

\[
\lambda_{51} = \delta(1 - \alpha)/\{\tau_d \xi_U\} \tag{B20}
\]
Finally, assuming that the sulphate particles are all bound to cloud water and neglecting interstitial SO₂, \( \lambda_{23} \) and \( \lambda_{13} \) are easily found in terms of \( \chi \):

\[
\lambda_{23} = \frac{\chi}{\tau_w} \tag{B21}
\]

\[
\lambda_{13} = \frac{(1 - \chi)}{\tau_w}. \tag{B22}
\]

Equations (B16)–(B22) together with (B7)–(B10) give expressions for the transition rates in terms of the estimatable parameters.

**Appendix C: Summary of Symbols**

This list includes only symbols used repeatedly in the text. Symbols used only in the section where they are defined are not included.

**Section 1**

X(t)  
Lagrangian particle trajectory

Z(t)  
Particle state

\( p_{ij}(x, t; x', t') \)  
Probability that a particle starting in state \( j \) at \( x \) at time \( t' \) will be in state \( i \) at \( x \) at time \( t \); \( t'(x') \) is omitted and implicitly equal to zero if the transitions are stationary (spatially homogeneous)

\( c_i(x, t) \)  
Concentration of particles in state \( i \)

\( S_j(x, t) \)  
Source density for particles in state \( j \)

**Section 2**

\( Q_{ij} (Q) \)  
Conditional transition probability between states \( j \) and \( i \) (matrix of \( Q_{ij} \))

\( \lambda_{ij}(x, t) (A(x, t)) \)  
Infinitesimal transition rate from state \( j \) to \( i \) (matrix of \( \lambda_{ij} \))

\( q_i(t) (q_i(t)) \)  
Probability that particle is in state \( i \) (vector of \( q_i \))

\( \tau_w (\tau_d) \)  
Duration of wet (dry) period

\( \lambda_w (\lambda_d) \)  
Wet (dry) removal rate

**Section 3**

u  
Particle velocity

\( U_i \)  
Mean velocity in state \( i \)

\( u' \)  
Velocity fluctuation of particle

\( A_i \)  
Matrix giving relative scale and direction of velocity fluctuations in state \( i \)

\( u_i(t), \Sigma_i(t) \)  
Mean covariance of position for particles in state \( i \)

\( T_L \)  
Lagrangian time scale of velocity fluctuations

**References**

Ackerman, B., Scott, R. W. and Westcott, N. E. 1983  

Austin, P. H., Baker, M. B., Blyth, A. and Jensen, J. B. 1975  
Small-scale variability in warm continental cumulus clouds. J. Atmos. Sci., 42, 1123–1138

Bolin, B. and Persson, C. 1975  
Regional dispersion and deposition of atmospheric pollutants with particular application to sulfur pollution over western Europe. Tellus, 27, 281–310

Eliassen, A. 1980  


