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AEROSOL TRANSPORT AND DIFFUSION DATA

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Various mathematical modeling approaches have been developed over the years to simulate the transport and diffusion of small particle aerosols in the atmosphere. The roles played by some important factors/mechanisms are briefly discussed, including the mean and turbulent components of the wind field, boundary conditions, particle size distribution, mode of dissemination, evaporation, and deposition. Expected-value models generally predict integrated dosage, deposition density, and aerosol "cloud" dimensions as functions of downwind location. Instrumented field trials of aerosol releases under various meteorological and environmental conditions can be valuable in helping to assess model validity. A literataure survey identified 65 documented small particle aerosol field tests, mostly under Department of Defense auspices. Review of these tests enabled initial development of an input-output data base for selected aerosol trials. The scope and some limitations of the test data documents reviewed to date are noted. Recommendations are made, including the need for further development of stochastic aerosol modeling, to capture the highly variable nature of experimental observations.

INTRODUCTION

The objectives of this paper are to report on a survey of historical records of field test data on aerosol releases into the atmosphere and to discuss the applicability of the available data to the validation of aerosol transport and diffusion simulation models.

Aerosols may consist of solid or liquid particulates. Our primary focus is on particle diameters less than 10μm (having very long settling times) and transport distances as great as 100 km. Gaseous release into the atmosphere is included and may be viewed as an "aerosol" of molecular dimensions. Disciplinary areas or applications for which a quantitative understanding of aerosol transport can be useful include: continuous (smokestack) air pollution, accidential releases of hazardous materials, pesticide spraying, defense against chemical or biological warfare, battlefield obscuration, micrometeorology, and aerobiology.

CHARACTERIZING AEROSOL RELEASES

Release conditions may be categorized by a number of idealized forms. With respect to spatial extent, we consider point sources, line sources, and area sources which may occur at ground level or at a fixed height above the ground. In the time dimension, sources may be continuous (with uniform release rate) or instantaneous. Actual releases, while invariably more complex, can often be reasonably approximated by one of these idealized forms. For example, a smokestack may be represented by an elevated, continuous-point source whereas airborne spraying takes the form of an elevated, instantaneous-line source. Some combinations, like elevated, continuous-line or area sources, are not likely to be encountered in practice.

Source strength can be expressed in either mass or particle number units. Thus, instantaneous sources might typically be quantified by gm, gm/m, or gm/m^2 units, or alternatively by no., no./m, or $no./m^2$. Continuous source strengths have comparable units augmented by $(unit\text{-}time)^{-1}$. Mass and number strengths are proportionally related, with the constant of proportionately determined by aerosol particle density, shape, and size distribution. In the simplest case of a monodisperse (uniform size) aerosol of sperical particles with diameter D and density ρ, the constant of proportionality is $\pi D^3/6$. For polydisperse aerosols, common parameters which characterize the size distribution are number median diameter, NMD (that diameter exceeded by 50% of all the particles), mass median diameter, MMD (that diameter exceeded by 50% of the total mass of particles), and the geometric standard deviation, GSD (a number >1 which measures the broadness of the size distribution). MMD always exceeds NMD except for monodisperse aerosols where they are equal. Aerosol size distribution is quite often well-approximated by the log-normal form, under which assumption

$$\ln \text{MMD} = \ln \text{NMD} + 3\,(\ln \text{GSD})^2 .$$

Another factor often introduced to characterize aerosol releases is efficiency of dissemination. If Q gm of a material is sprayed along a line L m in length to produce a small-particle aerosol with efficiency η, then the line source strength will be

$$q = \eta Q/L \text{ gm/m}.$$

Presumably the remaining $(1-\eta)Q$ of material was never actually released or else formed large, rapidly settling particles. Note that dissemination efficiency may be strongly dependent on the defined range of particle sizes of interest in the released aerosol.

UNDERLYING PHYSICAL MECHANISMS

The fundamental mechanism of transport in the atmosphere is the wind field, characterized by a mean wind vector $\bar{v}$ and a zero-mean wind turbulence. The latter is generally a nonhomogeneous, nonstationary vector stochastic process, though often approximated as stationary and quasi-homogeneous (height-dependent only) with specified temporal and spatial spectra. Aerosol particles less than 10 μm effectively follow the local wind field. Gravitational settling is a mere 2 mm/sec or so at 10 μm, and varies with r^2 in that size regime. It is, thus, a generally negligible effect. The wind turbulence is the fundamental mechanism causing diffusive growth of the aerosol cloud, while the mean wind accounts for the overall transport of the center of mass of the cloud. We may represent the aerosol cloud by a concentration field $c(x,y,z,t)$,

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expressed either in mass or number units per unit volume. As a highly idealized example, if (1) there were an instantaneous point source release of strength Q at space coordinates (0, 0, H) at time 0, (2) the diffusion followed Fick's law, and (3) the atmosphere were considered infinite in extent in all directions with mean wind speed u along the positive x axis, then the ensuing aerosol cloud concentration would have an expanding Gaussian distribution as follows:

$$c(x, y, z, t) = \frac{Q}{(2\pi)^{3/2}(K_x K_y K_z x^3/u^3)^{1/2}} \times$$

$$\exp\left\{-\frac{(x-ut)^2}{2K_x x/u} - \frac{y^2}{2K_y x/u} - \frac{(z-H)^2}{2K_z x/u}\right\} \qquad (1)$$

for $t > 0$ and $x > 0$, where K_x, K_y, K_z are constant diffusivities in the three coordinate directions. The magnitudes of the diffusivities which relate to the rapidity of diffusion, are, of course, tied to the intensity of turbulence in the atmosphere. Note that the cloud center travels the world line (ut, 0, H, t) while the cloud dimensions (standard deviations) grow with the square root of downwind distance. This is only one particularly simple solution out of a large diverse set of solutions based on various models.

We consider now some of the complications that derive from numerous physical complexities and constraints. The atmosphere is clearly decidedly nonhomogeneous in the vertical direction. The ground, heat and momentum transfer processes, nonuniform atmospheric density, and the like act to create very significant height dependent phenomena. For one thing, the ground presents a total physical barrier to further downwind diffusion. A comparable phenomenon often occurs aloft where a stable boundary forms through which turbulent mixing and diffusion are greatly attenuated. This produces a so-called mixing layer of finite thickness, typically of the order of hundreds of meters, within which the aerosol cloud would be mostly confined (if initially released within the layer).

Processes at the ground and upper mixing layer boundaries can be quite complex, involving reflection, transmission, deposition, and/or resuspension of aerosol particles. The complexity may be accentuated by the boundary irregularities of terrain, vegetation, and culture. The aerosol particles themselves are not necessarily immutable. Evaporation of water or other volatile components of liquid droplets, the converse process of water accretion in hygroscopic particles, agglomeration or condensation into larger particles, chemical changes, and viability loss in microorganisms are all possible phenomena. These may be mediated by particular environmental conditions of temperature, relative humidity, and solar (UV) radiation. Aerosols are also subject to scavenging by precipitation.

A particularly intriguing special environment is that beneath a forest canopy. Transport and diffusion are influenced by the large surface area and spatial distribution of leaves (or needles) which substantially modify the local wind field and serve as deposition sinks. Depending on source location, infiltration will occur either upwards or downwards through the canopy.

CLOUD TRANSPORT AND DIFFUSION SIMULATION MODELING

Under fairly general conditions, the differential equation for transport and diffusion of an aerosol field can be written as

$$\frac{\partial c}{\partial t} + u\frac{\partial c}{\partial x} = \frac{\partial}{\partial x}\left(K_x \frac{\partial c}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_y \frac{\partial c}{\partial y}\right) + \frac{\partial}{\partial y}\left(K_z \frac{\partial c}{\partial z}\right) + S$$

where S is the aerosol source function and the other quantities are as previously defined except that the diffusivities (K's) are not constrained to be constant. An example of S for an instantaneous point source at (0, 0, H, 0) is

$$S = Q\delta(x)\delta(y)\delta(z - H)\delta(t).$$

where Q is source strength and δ is the Dirac delta function. For a continuous point source at (0, 0, H)

$$S = Q\delta(x)\delta(y)\delta(z - H).$$

In the latter example, at steady state the $\partial c/\partial t$ term vanishes.

A customary simplification is to assume an idealized flat ground with mixed boundary conditions, and only height (z) dependence for wind speed and diffusivities. Various functional forms for these dependencies have been tried. Under most assumptions an analytical solution is not achieved and the differential equation must be numerically integrated, requiring considerable computational resources. There has recently been renewed interest in numerical integration of partially simplified diffusion equations as a means of predicting aerosol cloud transport and diffusion behavior.

An alternate tack followed by many early investigations was to introduce further simplifications or to postulate specific solution forms consistent with special case solutions. A very popular approach was to assume a Gaussian plume distribution. For conditions of an instantaneous point source release at altitude this gives rise to the form presented in Equation (1) except that more generalized expressions for cloud variances as functions of downwind distance

$$\sigma_x^2(x),\ \sigma_y^2(x),\ \sigma_z^2(x)$$

replace the

$$K_x x/u,\ K_y x/u,\ K_z x/u$$

terms, respectively. Some typical forms proposed for these variance functions are $2p^2x^2$ by Bosanquet and Pearson [1], c^2x^{2-n} by Sutton [2], $2kx/u$ (the Fickian solution) by Roberts [3] and Vaughan [4], $(\sigma_w x^\beta)^2$ by Cramer [5], and $(3i^2x)^2$ by Smith and Hay [6], where the various coefficients and exponents, c, k, σ_w, i, n, and β are empirical functions of the intensity of turbulence. In particular, σ_w in Cramer's formulation is interpreted as the standard deviation of fluctuating wind direction (in radian measure) while β is an empirical constant ranging from 0.3 under very stable atmospheric conditions to around 1 for unstable conditions.

In order to account for complications usually encountered, such as multiple sources, complex terrain, forest environment, droplet evaporation, and aerosol decay, various ad hoc appendages theoretically or empirically justified have been proposed. Multiple or complex sources are considered well accounted for by the principle of superposition of results for elemental sources. A recent evaporation model by Wu [7] expresses the rate of decrease of the square of drop radius as a function of ambient temperature, droplet constituent partial pressure (and other constituent physical properties), and mean wind speed (which becomes significant for drop radii greater than 20 μm). The most common approach for handling aerosol decay due to chemical or biological changes is to assume an exponential decay (i.e., decay rate proportional to concentration) with the coefficient of decay empirically dependent on such ambient factors as temperature, humidity, and solar radiation intensity.

FORMS OF MODEL PREDICTIONS

Instantaneous aerosol concentration, which we have focused on above, is not a directly measurable quantity. Any instrumentation must sample over a finite time period. Another consideration is that many effects of interest depend on accumulated action of an aerosol field over time. Accordingly, we define the partial dosage at point (x, y, z) from time t_1 to t_2 as

$$d(x, y, z; t_1, t_2) = \int_{t_1}^{t_2} c(x, y, z, t)dt$$

Typical units are number min/m^3 or gm min/m^3. A special case is total dosage

$$D(x, y, z) = d(x, y, z, -\infty, \infty)$$

This represents the accumulated effect of a total aerosol cloud as it impinged on, enveloped, and finally passed by the point (x, y, z). An even more special quantity is the total dosage at ground level, $D(x, y, 0)$, where most things (people, instrumentation, etc.) are usually located. As an example, consider the total ground level dosage from an instantaneous point source release of strength Q at $(0,0,H)$ at $t = 0$ following Fickian diffusion as represented in Equation (1) for $x >> K_x/u$, i.e., at sufficiently far downwind locations,* yields:

$$D(x, y, 0) = \frac{Q}{2\pi(K_y K_z)^{1/2} x} \exp\left\{-\frac{y^2}{2K_y x/u} - \frac{H^2}{2K_z x/u}\right\}$$

Figure 1 schematically portrays the general form of ground level dosage for this particular example

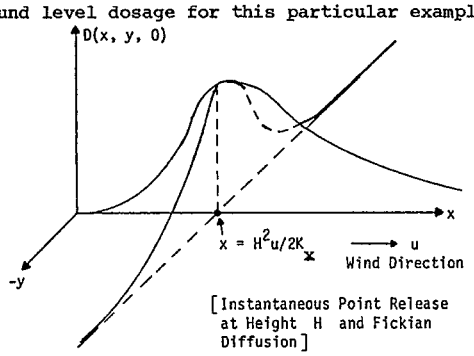

Figure 1. Ground-Level Dosage

As another example, consider an instantaneous infinite line source of strength q along the line

$$x = z - H = 0$$

where the ground level concentration field follows a Cramer-like model form, viz.,

$$c(x, y, z, t) = \frac{q}{2\pi \sigma_w^2 x^{2\beta}} X$$

$$\exp\left\{-\frac{(x - ut)^2}{2(\sigma_w x^\beta)^2} - \frac{H^2}{2(\sigma_w x^\beta)^2}\right\}$$

Total dosage at ground level then becomes

$$D(x, y, 0) = \frac{q}{\sqrt{2\pi}\,\sigma_w x^\beta u} \exp\left\{-\frac{H^2}{2(\sigma_w x^\beta)^2}\right\}$$

which is, of course, independent of y and has its maximum dosage occur along the line

$$x = (H/\sigma_w)^{1/\beta} .$$

Note that for longer ranges downwind, D is predicted to fall off as $x^{-\beta}$ (where typical values for β range from 0.3 to 1) in contrast to the previous example where the fall off was predicted to be as $x^{-1/2}$ independent of atmospheric conditions. It must be emphasized again that these are example results, only, for some assumed diffusion models.

From the abstract quantity, dosage, one can obtain two derived measures having more concrete significance. One is the inhaled dose $\mathcal{D}$ (in number or mass units) accumulated by a person (or animal) exposed to the total cloud passage. The other is deposition density P (in units per area) of total aerosol material deposited on the ground following passage of the cloud. Inhaled dose is given by:

$$\mathcal{D} = Dr$$

where r is the respiration rate in volume rate. For human beings r is of the order of 10 l/min or 0.01 m^3/min. Thus, for example, a total cloud dosage of 10 mg-min/m^3 would lead to an inhaled dose of approximately 0.1 mg. Dose also applies to instruments with given volume sampling rates. It should be noted that the actual dose accumulated by respiring organisms depends also on the fraction retained during expiration and is a complex function of particle size.

Deposition density is given by:

$$P = Dv_d$$

where v_d is deposition velocity. Deposition velocity is not the same as gravitational settling velocity. The latter for aerosol particles smaller than 10 μm is quite small, less than 0.2 cm/sec. On the other hand, v_d which can be in the range of several m/sec, is influenced by intensity of turbulence which leads to inertial impaction of the particles on the ground surface [8]. The nature of the particle and of the surface, and possibly also

* K_x/u will generally be less than 10 m for most atmospheric stability conditions of interest.

local electrostatic fields, determines the extent of particle resuspension. Note that the spatial distributions of dosage, inhaled dose, and deposition density may all be assumed to be the same inasmuch as these quantities are linearly related by coefficients which can be taken as constant (for simple ground models).

Two other predictions of interest that can be made from concentration fields are cloud duration from an instantaneous source at a fixed point downwind and cloud horizontal width normal to wind direction (the latter generally from a point source). If we define durations and widths by intervals over which the concentration is at least 0.1 of maximum concentration, and if the cloud shape is assumed Gaussian (as has generally been the case in past modeling) then cloud duration and width are approximately:

$$T = 4.3\sigma_x/u$$

$$W = 4.3\sigma_y$$

where σ_x and σ_y are the Gaussian standard deviations in x and y, and u is mean wind speed. Thus, for the solution in Equation (1):

$$T = 4.3\sqrt{K_x x/u^3}$$

$$W = 4.3\sqrt{K_y x/u}$$

which implies growth in these quantities proportional to the square root of downwind distance.

DATA SURVEY FOR MODEL VALIDATION

In view of the complexity and variability of atmospheric phenomena and boundary conditions, one is naturally concerned about how well the various proposed simplified model formulations predict actual transport and diffusion behavior. We were, in particular, interested in a specific Gaussian model formulation [7, 9] geared to instantaneous point or line releases which allow for a power law growth of cloud dimensions with downwind distance, i.e.,

$$\sigma_{x_i}(x) = (a_i + b_i x)^{\gamma_i}$$

where

$$x_1 = x,\ x_2 = y,\ x_3 = z$$

and b_i and γ_i are constants related to the intensity of atmospheric turbulence as may be predicted from measured meteorological parameters. The a_i terms allow for finite aerosol source size at time of release. The model has some additional interesting features, for example, a selectable mixing layer height limiting upward cloud growth, flexible boundary conditions at ground level for resuspension of impacting particles (along with a selectable or theoretically computed deposition velocity), aerosol evaporation, and an optional model for transport within a forest canopy. Since deposition, evaporation, and forest transport are strongly dependent on particle diameter, the model also provides for aerosol diameter distribution and the tracking of partitioned individual size groups.

Our major focus was on predicted cloud growth and ground dosage, and deposition as functions of downwind location under various conditions of release and environment. Our objective was therefore to identify actual aerosol field tests for which adequate data exist to enable validation studies on the transport and diffusion model of interest, as well as on other extant or proposed models. As part of such a field test data survey, it is important to assess the completeness, accuracy, and relevance of the reported data.

Some of the constraints or conditions of special interest imposed on the survey were:

- Small particle sizes -- $\leq 10\,\mu m$;
- Instantaneous sources (with emphasis on line sources);
- Stable and neutrally stable atmospheric conditions;
- Flat open as well as wooded and complex terrain;
- Downwind distances from 1 to ~100 km.

A total of 65 small-particle aerosol field test programs with likely potential for useful downwind dosage, deposition, and/or cloud size data were identified. A program generally consisted of a series of individual releases or trials under varying conditions. Test reports for 21 of these programs were obtained and reviewed in detail. The bulk of the identified programs were conducted by or under sponsorship of the U.S. Department of Defense. The variety of conditions covered in the surveyed tests is described for several test attributes in Table 1.

• RELEASE:	Elevated/Surface, Line/Point
• MATERIAL:	Solid/Liquid/Gas Persistent/Decaying
• METEOROLOGICAL:	Stable/Neutral/Unstable Mixing Layer Heights
• TERRAIN:	Flat Open/Wooded/Complex/ Marine
• SAMPLER GRID:	Linear/Array (Ground Level) Vertical (Some) Redundant Samplers (Some)
• SAMPLER RANGES:	30 m to 180 km
• OUTPUTS:	Dosage Partial Dosage (Some) Horizontal Deposition (Some) Cloud Size (Some)

Table 1. Conditions Covered in Surveyed Tests

As an example of the kind of data found in these reports, some dosage and cloud duration results from one trial of the Victoria Diffusion Program [10], conducted near Victoria, Texas in 1965, are shown in Figures 2 through 4. An attempt was also made to measure ground deposition, but this was unsuccessful due to poor techniques. Table 2 gives selected meteorological measurements for this trial. The aerosol material was a zinc-cadmium sulfide particle which fluoresces yellow under ultraviolet light (hence, the nomenclature Yellow FP). Its bulk size distribution was approximately log normal with parameters: NMD = 1.57 μm, MMD = 3.05 μm, and GSD = 1.6. Evaporation was clearly not an issue for this particle. The terrain in the test area was quite flat with very low vegetation (mostly farmland); hence, forest diffusion was also not an issue.

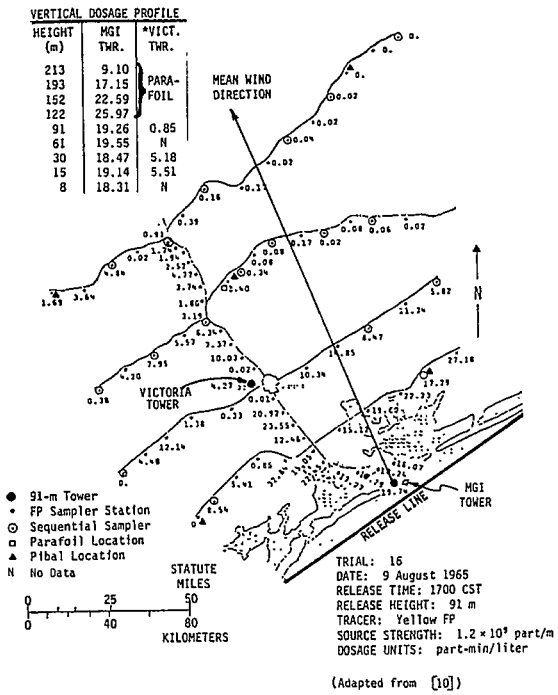

Figure 2. Victoria Diffusion Trial Total Dosage Map

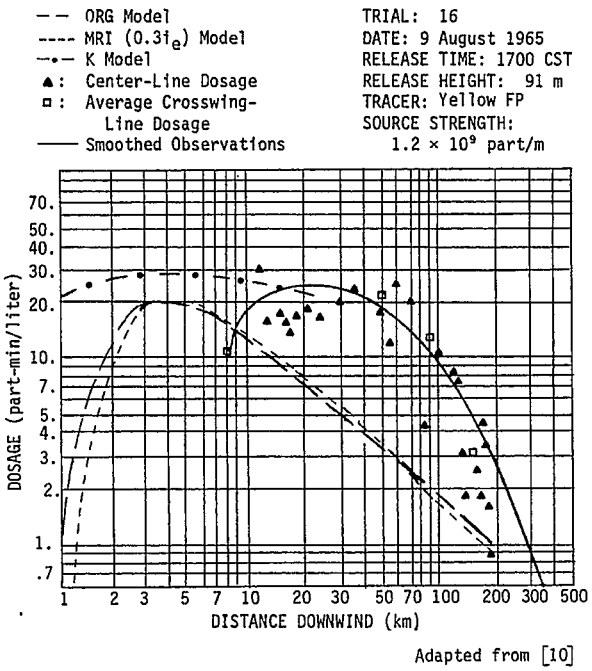

Figure 4. Downwind Dosages in Victoria Diffusion Trial

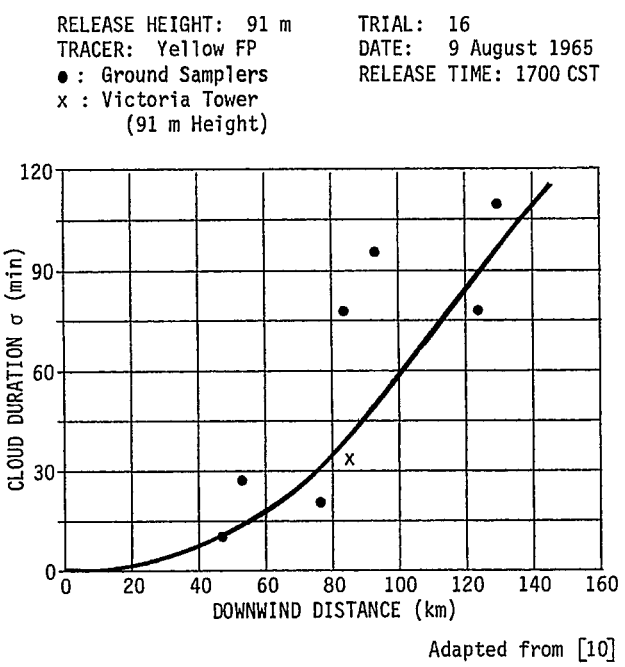

Figure 3. Cloud Duration

HEIGHT (m)	WIND SPEED (m/sec)	TEMPERATURE T_{Z-2} (°C)	TURBULENCE σ_{30} (°)
At Line Source (15 min average)			
2	3.2	0	
4		0.3	
8	2.4	0.1	
15		0	
30	6.2	-0.3	
61	6.6	-1.3	3.7
91	6.3	-1.7	2.4
80 km Downwind (15 min average at arrival time)			
2	1.3	0	
4		0.1	
8	2.6	0.5	
15		0.8	
30	5.1	1.7	
61	7.0	2.7	
91	7.2	2.7	0.6
RELEASE TIME: 1700 CST SKY COVER: Scattered to High Thin Overcast RELATIVE HUMIDITY: 55-70%			

Table 2. Victoria Diffusion Trial 16 -- Selected Meteorological Data

Now, if appropriate inputs to the aerosol transport diffusion model of interest, or for that matter to any other prediction model, were developed based on the conditions of the trial, then a comparison of model outputs with observed data would contribute to an assessment of model adequacy. Clearly, it is desirable to conduct such comparisons over a wide range of tests, and this is the motivation for developing a comprehensive aerosol field test data base. The overall process of model validation has not as yet been carried out under our survey project, but input data sets to run the model so as to correspond to field test conditions were developed for a number of selected trials. This included implementing procedures for estimating diffusion parameters (b_i, γ_i) on the basis of different kinds of available meteorological measurements. Unfortunately, meteorological data varied considerably in form and detail over the documented field trials depending on the specifics of instrumentation setup.

Even without going into the details required to develop model inputs, or applying these to arrive at model predictions, a few interesting observations can be made from the one sample of illustrative data here provided. First, the substantial variability of measured dosages at individual sample points is noted. (Each sample point, incidentially, is the average of two replicate colocated samplers.) This observation should remind us of an implicit assumption in all of the transport and diffusion models described in this paper, namely, that it is the expected value of concentration, dosage, or whatever that is being estimated. The thing, itself, is in reality a random variable consequence of the stochastic process in space and time called atmospheric turbulence. Thus, an important process that ought to be applied to the measured data is smoothing in space (and time, if possible). We shall come back to this issue a bit later. At any rate, one significant impact of variability is the uncertainty it introduces in any attempt to ascertain whether or not a given expected value model is valid.

A second point, which may not be quite apparent from the data shown, is that relative, rather than absolute, dosages merit the greatest attention because of the difficulties involved in measuring actual effective source strengths under field conditions. While some questions would not be fully resolved under such an approach, a great deal could still be said about model fit because just about all theoretical models predict concentration/dosage outputs that are directly proportional to source strength.

APPARENT SHORTCOMINGS IN THE FIELD TEST DATA

On the basis of the test data which have been examined, some preliminary observations are offered regarding apparent deficiencies. These problem areas could conceivably also be characteristic of the remaining documented tests, in which case model validation efforts would be limited without additional properly planned test programs.

Very few tests collected detailed partial dosage data from which time-varying instantaneous concentrations could be inferred. Relatively few tests attempted to measure horizontal deposition, and none attempted vertical (or other surface orientation) deposition measurements. Because of substantial variabilities found between replicate samplers, even those next to each other, questions were raised regarding possible inadequacies of sampling/measurement techniques; these should be resolved. Available sampling devices e.g., Anderson sampler, which can establish particle size distributions at the sampled points were usually not employed. Thus, important questions pertaining to droplet evaporation, condensation, and water accretion phenomena cannot be explored or, if incorporated in a model, validated. As previously noted, absolute source strengths were often not measured nor estimated from auxiliary experiments with the disseminating device. In a number of tests conducted to observe transport within forest canopies, the forest environment was not adequately described; for example, in terms of canopy height and leaf areal density.

Another general problem area is in meteorological and/or direct turbulence measurements which pose an extremely difficult experimental requirement. Because different models use different meteorological data to infer turbulence characteristics or depend on direct turbulence instrumentation, there is no standard for what should be measured. This problem is aggravated by the spatial and temporal inhomogeneity of meteorological conditions which can only be overcome by suitable averaging, and which therefore, imply the need for extensive instrumentation: at the source, at the sampling locations, near the ground, and aloft.

SIMULATION OF CLOUD TRANSPORT AND DIFFUSION VARIABILITY

One conclusion that derives from the preliminary work on field test data collection done so far is the clear need for stochastic modeling of aerosol cloud transport. Stochastic modeling may possibly be approached through more explicit simulation of fundamental turbulent and other random processes. Achievement of such a model would enable one to predict dosage/deposition/cloud size variances in addition to expected values. Some of the input sources of variance that could be considered are: most importantly, the spectral structure of the stochastic wind field (spatial and temporal); incomplete turbulent mixing, i.e., stable laminar flow phenomena, randomness in terrain or vegetation characteristics; and randomness in source strengths and dissemination efficiencies (particularly manifested as variations along a release line). The goal of such modeling is the explicit estimation of spatial and temporal covariances of measurement events. This covariance could also be extended to replicated aerosol releases. The development of a validated stochastic model (validated by means of a comparison of its predicted variability with observed data) would then enable it to be used to provide even more realistic predictions of aerosol cloud transport phenomena than is now possible with existing expected value models.

Some efforts at stochastic aerosol diffusion modeling have been attempted in the past [11]. A recent analysis of instantaneous plume structure by Jones [12] provides some relevant new insights.

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