A Review of Transport and Diffusion Models

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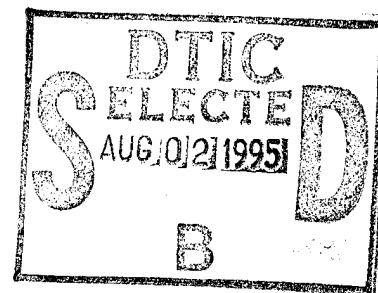
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The modeling techniques for the transport and diffusion of material in the atmosphere is reviewed with emphasis on the impact of terrain and meteorology on the process. Weaknesses and strengths of the various modeling approaches are discussed and a number of models in use by or under development for the Defense community are categorized by type and suggested applicability. The conclusion of this report is that any substantial increase in current modeling capability will require more attention to the high-resolution effects of terrain and meteorology. It is further concluded that analytic Gaussian techniques in use are not capable of efficiently providing the necessary coupling to these effects and of the subsequent increase in desired accuracy.

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Contents

1. Introduction

This report addresses issues associated with the modeling and simulation of atmospheric transport* and diffusion processes. Specifically, currently used methodologies are compared and examined for range of applicability and for appropriateness. The topic should be of interest to Department of Defense (DoD) users and program sponsors that rely on such models for decision. The topic also fulfills a requirement of the Ballistic Missile Defense Organization. To be certain that all readers have the same basic understanding, a substantial portion of this report is concerned with providing a necessary but not exhaustive overview and history of the concepts that provide the foundation for existing models. While some readers may be uncomfortable with the mathematics and others concerned with the lack of detail, every effort has been made to assure sufficient coverage that technically advanced readers are able to draw their own conclusions, while others achieve some understanding without being overwhelmed with mathematical intricacies.

To put things in the proper perspective, it is well to remember: before computers, solutions to complex physical problems could only reasonably be treated in an idealistic parametric manner and were fraught with many simplifying assumptions. Still, beyond the simplest of cases (ones for which exact mathematical results exist) the accuracy of the result is obviously dependent on how well the parameterization replicates the physical process. The prediction of hazards caused by the dissemination of any material into the atmosphere is an extremely complex problem driven primarily by meteorology and affected by source structure, chemical reactions, decay factors, and human response. While each factor is in itself a complex problem, and the final solution is only as accurate as the least accurate treatment in the chain, this paper addresses only the issues associated with atmospheric phenomena that control the dispersion of materials, commonly referred to as transport and diffusion (T&D).

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*Terms in this report are referred to in the index beginning on page 59.

Local meteorological conditions are the primary determinant of the outcome of the T&D processes, and a complete understanding of atmospheric behavior is essential to the description of its influence. To ascertain the effect of atmosphere on the mass dissemination, we must first consider that forces of much different temporal and spatial scales drive these two separate yet coupled physical processes. The scale and coupling are, in turn, dependent on space and time considerations. [1] Using the standard notion of transport (the motion of the center of mass of some defined entity) does not relieve the necessity to examine the temporal and spatial variability that leads to a three dimensional smearing about that center. Traditionally, we treat all motions not associated with the motion of the centroid as diffusion, and we further differentiate molecular or microdiffusion from macroscopic turbulent diffusion. This separation is natural because molecular diffusion is a property of the transporting media while turbulence is a property of the flow. The effects of turbulent diffusion are generally orders of magnitude larger than the effects of molecular diffusion, a fact many researchers use to justify ignoring the later. As will be seen, the common approach is to mathematically treat turbulent diffusion in a manner similar to molecular diffusion. This has led to many physical constructs that attempt to put order into a process that is inherently unordered. A pervasive notion is that turbulence is a random process; therefore, it eludes a rigorous mathematical description necessitating an empirical or statistical treatment. To gain an appreciation for the various methods for the treatment of this phenomenon, it may be best to review the mechanisms that introduce turbulence into the atmosphere and the various methods used to characterized it and its effect.

2. Atmospheric Motions

In the absence of differential energy effects (surface heating and density variations), a very simple picture of a fluid surrounding a spinning globe can be described analogous to well-studied viscous flow over a flat plate. For a smooth, uniform surface and relatively slow, undisturbed flow, two regions can be defined. The lower level is the region directly influenced by the frictional drag (no-slip condition) at the fluid-solid interface and is characterized by an increasing velocity for some distance from the surface. The flow of the upper region is independent of frictional effects resulting in a constant velocity with distance from the surface. In both regions, the flow is typified as laminar, which can be visualized by the introduction of a steady source of dye. In laminar flow, the dye traces a thin ribbon or streamline in the direction of the flow, horizontal and vertical spreading of the dye is caused by molecular diffusion and the transfer of momentum in the region influenced by friction.

Irregular surfaces (hills, grasslands, and wooded areas) disturb the purely laminar aspects of the flow in two ways. Experiments by Osborne Reynolds in the late nineteenth century demonstrated that flow over a rough surface could be characterized by a nondimensional number used to indicate various transitions in flow characteristics. The Reynolds number (Re) results from a ratio between the elements that contribute to and suppress a disturbance in the flow. Roughness and speed lead to an unsteadiness while viscosity tends to dampen it, leading to Re = lU/v, where l characterizes the length (height) of the disturbing element(s), U is the mean flow speed, and v the kinematic viscosity of the fluid. Reynolds experiments showed that for very small Re, the flow remained laminar in the sense that streamlines remained parallel. Returning to the flow over a plate, in this case with surface protuberances, the maximum height at which flow velocity continues to change is simply higher. Two distinct flow regimes are still evident.

The experiments further showed that as Re increased, circulating eddys formed on the downstream side of an obstruction, and at even larger Re, the eddys shed into the laminar flow disturbing its uniform characteristics downstream of the obstacle. Eddy size and the periodicity of shedding are directly correlated

to Re. Extending these results to atmospheric flow in proximity to the very nonuniform surface of the earth and in the absence of any thermal effects, the production of a very large range of eddy sizes is possible. This action is one of the primary sources of mechanical turbulence in the atmosphere, providing the rationale for characterizing turbulence as eddy turbulence and resulting dispersion as eddy diffusivity. As is discussed, turbulence is not as well organized as this view may suggest; it does, however, provide a basis for the definition of turbulence regimes in the atmosphere. The complete picture requires that we also consider thermal eddy sources, and the effects by large air masses distributed throughout our atmosphere.

The simplified notion described is complicated first by temperature-density variations and by differential heating of the surface of the earth. Temperature-density variations are manifested in a tendency for the atmosphere to organize itself into large, identifiable air masses. The boundaries between air masses or weather systems can result in velocity shears and strong gradients providing additional sources of turbulent energy. Each air mass type, exclusive of its boundaries, has its own turbulence structure. Superimposed on the large scale features are the effects of the local surface topography and heating. Surface heating creates small convective or thermal eddys that provide a mechanism for the vertical transport of turbulence. In the absence of the thermal effects, turbulent eddys caused by surface irregularities would remain confined to a very thin layer close to the surface. In the real atmosphere, vertical convection caused by surface heating transports the eddys to higher levels.

Based on these considerations a reasonable separation of the atmosphere into regimes having very different turbulence structures is possible. Meteorologists identify a surface boundary layer (SBL) in which surface features, friction, viscosity, and temperature gradients[*] are the dominant influence on the motion. Above this layer is a transition zone or planetary boundary layer (PBL) in which density gradients, residual friction (caused by convective transport) are

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[*]Strongly heated surfaces create temperature gradients near the surface much larger than those found elsewhere in the atmosphere. In some cases, the gradients lead to isolated rivers of vertical flow that cause mirages.

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the dominant flow determinants. Finally, far removed from the surface we have a region that can be treated as inviscid because the governing forces are primarily pressure gradients and the coriolis effect (caused by rotation of the earth). The highest layer is generally termed the free atmosphere, free in the sense that motions are all but independent of surface characteristics and influences.

The thickness of each layer is highly dependent on prevailing conditions (terrain roughness, wind speed, temperature, time of day/year, cloud cover, etc.), but in general, the SBL extends 50 to 100 m above the surface and the planetary layer extends 500 to 1000 m. Using standard fluid dynamic analyses, [2] we can express these definitions in a more rigorous manner. Accelerations at any point in an incompressible* fluid are described, in Cartesian coordinates, by Newton's second law,

$$\rho\left(\frac{\partial u}{\partial t} + u\frac{\partial u}{\partial x} + v\frac{\partial u}{\partial y} + w\frac{\partial u}{\partial z}\right) = \mu\left(\frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} + \frac{\partial^2 u}{\partial z^2}\right) - \frac{\partial P}{\partial x} + \rho\xi \qquad (1)$$

where

u, v, and w	= the flow velocities in the respective x, y, and z Cartesian coordinates
ρ	= the fluid density
μ	= the dynamic viscosity
P	= the pressure
ξ	= other external forces per unit mass.

Similar equations for the y and z components yield three equations that form the Navier-Stokes equations of motion. The term on the left-hand side is the inertial or acceleration term (the ma of F = ma). On the right are, the viscous, pressure, and additional (coriolis force and gravity) force terms, respectively. It is the viscous force, together with the physical reality that at a surface, the relative velocity must vanish (no-slip condition), which accounts for resulting

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*For Reynolds numbers associated with atmospheric motions, the simplifying approximation of an incompressible atmosphere has been shown to be valid.

frictional and rotational forces that lead to eddys and turbulence. Mathematically, the viscous term can be separated into two terms reflecting frictional and rotational forces. Therefore, the amount of energy existing in the turbulent eddys, created by the rotational forces, can be tracked. The inertial term is nonlinear, which makes universal solutions extremely difficult except in the simplest cases. [3] Separating the atmosphere into regimes based on importance of the various force terms permits convenient generalizations for a layer. Unfortunately, success in applying generalizations in one domain does not guarantee the similar applicability in other regimes.

In the SBL, the temperature gradient and viscous terms in equation (1) can be shown to be much larger than either the pressure or coriolis* terms. The manner in which the temperature gradient is included will be shown in the section on diffusion. As distance from the surface increases, the viscous and temperature gradient terms decrease to a point at which the order of all terms is the same. The process can be visualized as follows. Small volumes of air immediately adjacent to the surface are heated by conduction, and rise similarly to bubbles in boiling liquid. Remaining intact and rising through ever decreasing external pressure forces, parcels cool through expansion; and moving slower than the surrounding air act as obstacles in the flow, effectively extending the influence of the surface through a larger than otherwise layer. The rising volumes constitute an eddy and can encompass eddys formed by the horizontal flow near the surface. The height M to which these thermally driven eddys rise (transporting horizontal momentum, heat, and turbulence) depends on the initial impulse and the diminishing buoyancy force resulting from density differences. M, the effective top of the PBL, referred to as height of the mixed layer, contains the planetary and surface layers. The ability of the atmosphere to produce such a mixed layer can be described through the ratio between competing convective and advective forces. The Richardson's number R_i,

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*The coriolis force is of the order ωv, where ω is the angular velocity of the earth's rotation, and v is the flow velocity.

$$R_i = \frac{\frac{g}{T}\left(\frac{\partial T}{\partial z} + \Gamma\right)}{\left(\frac{\partial \overline{U}}{\partial z}\right)^2} \qquad (2)$$

where

    g = gravitational acceleration

    Γ = the appropriate adiabatic temperature lapse rate (see footnote on page 20).

Comparing actual temperature lapse rates to the adiabatic rate is a standard meteorological means of determining the vertical stability of an atmospheric layer. Examination of equation (2) indicates that single point measurements of T and u are inadequate descriptors of a mixed layer. To correctly characterize the mixing potential, realistic gradients of these quantities are required. For $R_i > 1$, turbulence is suppressed and a tendency to laminar flow is achieved; for $R_i < 1$, the flow tends to remain turbulent and the mixed layer tends to grow in thickness.

3. Atmospheric Turbulence

Section 2 suggests that mechanisms introduce turbulence into the atmosphere, and indicates that the random nature of turbulence lends it intractable. If the processes are completely random, a less rigorous but pliant description can be applied using statistical methods. Before attempting this, it is good to isolate the tractable. Following Reynolds, this can be accomplished through the definition of any variable term through the superposition of its mean (a) and fluctuating (a') components, where:

$$a' = a - \bar{a}; \quad \bar{a} = \frac{\displaystyle\int_{t - .5T}^{t + .5T} a(t)dt}{\displaystyle\int_{t - .5T}^{t + .5T} dt}. \tag{3}$$

Applying this notion to equation (1), it is possible to separate the flow into its mean and fluctuating components. Clearly, the viability of this approach is dependent on choosing a period T such that a represents the mean characteristics of the general flow. T must be sufficiently long to include an adequate number of low-frequency fluctuations, while an overly long period may effectively conceal important features of the higher frequency variability.

Imposing a requirement of steady mean flow, defined by

$$\overline{u'} = \frac{1}{T} \int_{t - .5T}^{t + .5T} (u - \bar{u})dt = \bar{u} - \bar{u} = 0 \tag{4}$$

leads to $\bar{u}' = \bar{v}' = \bar{w}' = 0$. However, fluctuation correlations (eddy velocities represented by $\bar{u}'^2$, $\bar{u}'\bar{w}'$, etc.) do not necessarily vanish and lead to a problem in the solution of the Navier-Stokes equation known as closure. The closure problem is a mathematical result of having insufficient information (equations) to match the number of unknown quantities. Thus, even under ideal conditions of steady flow, energy can exist in the turbulent structure and contribute to diffusion. If the mean flow is not steady, other restrictive conditions must be

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applied. In all cases, equation (3) must be satisfied exactly or to a high degree of approximation before this construct is useful. As appealing as this approach may first appear (separation into a well behaved and solvable equation and a coupled but unsolvable transient equation), it still does not permit a rigorous definition of the effects of turbulent diffusion for one must have *a priori* knowledge of or a valid method for estimating the transient eddy velocities or at least the amount of energy contained in the turbulence. Given the complexities of atmospheric motions compounded by weather, terrain, and diurnal and seasonal variability, the former is certainly formidable. A number of methods, including statistical approximations, for estimating eddy velocities or their effects have been suggested. These will be reviewed; but first, it must be noted that the analyses of atmospheric turbulence are complicated by the range of temporal and spatial scales over which the phenomenon occurs in the atmosphere. Further it must be realized that the regimes, as defined, have completely different spectral forms. As noted by Pasquill, [4] the statistical properties depend considerably on the sampling duration, and often, autocorrelations (e.g., $\bar{u}'^2$) and variances do not satisfy boundary conditions. This is caused by variability in the sources of turbulence such that the atmosphere, particularly in the boundary layer, is not steady or stationary. Both conditions are required for strict application of statistical theory, and more strictly for the generalization of analytic descriptions. A further restriction, homogeneity, is in force when the theory is applied to the diffusion of volume sources. Despite these cautions, statistical approaches are appealing in theoretical applications when energy conservation can be applied or its principals are used to obtain statistical quantities (e.g., $\bar{u}'^2$, $\bar{u}'\bar{w}'$). One technique has been successfully applied in recent diffusion modeling using the Navier-Stokes equations.

The atmosphere can be considered an enormous energy conversion machine, and it was G. I. Taylor [4] who first suggested that eddys in the atmosphere provide the mechanism for the transfer of the mechanical energy, contained in the large scale flow, to thermal energy. He postulated that the process consisted of a cascade of energy from the large eddys to sufficiently small motions that are indistinguishable from molecular motion induced by thermal properties. Kolmogorov's [6] theoretical extension of Taylor's [5] work provided a statistically grounded mathematical description of Taylor's [5] evolutionary

process. The success of this idea and subsequent work is realized in its ability to provide verifiable correlations between the mean flow and the turbulence structure. A physical realization of this process can be gained by the observation that large atmospheric eddys produced by intermittent sources are nonpersistent, and are illustrated by considering a thermally isolated system containing a fluid. Any instantaneous agitation of the fluid that results in a large-scale motion, such as an abrupt movement of a wall or perhaps a stirring action that creates a whirlpool, will eventually be followed by the (slightly warmer) fluid coming, once again, to rest.

4. Diffusion

Given the random nature of turbulent diffusion, the question is: "Is there a method available that can mathematically replicate the physical process?" The hypothesis by Taylor [5] provides one link, albeit microscale and statistical in nature, between the thermal and mechanical properties of the atmosphere. The atmosphere is a gas; therefore, the pressure term in the equation of motion equation (1), coupled with the gas law ($P = \rho RT$) affords the macroscopic relation between the two manifestations of energy. Rendering an effective and useful characterization of the atmosphere at either scale lies in our ability to provide a description that bridges the gap between them. Much of the early work in modeling diffusion processes ignored this important factor. The conservative nature of this physical process, specifically energy conservation, together with the many observations of the correlation between the state of the mean flow and the state of the turbulence [1,4] yields an appealing method for this connection. A satisfactory employment of this methodology requires an adequate scheme to maintain a close inventory on energy conversions in the domain of interest, particularly at the boundaries, and in the presence of cloud formation and water vapor transport. In the absence of water vapor considerations, thermal energy changes can be expressed in terms of variations in temperature T, a measure of heat (energy), by summing the source and sinks of heat energy in a volume:

$$\rho C_v \frac{\partial T}{\partial t} = \rho C_v [u\frac{\partial T}{\partial x} + v\frac{\partial T}{\partial y} + w\frac{\partial T}{\partial z}] + N + k[\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2}] \qquad (5)$$

where

ρ = the media density
C_v = a constant specific heat
k = thermal conductivity.

The first term on the right is the transport (advection) term; N represents terms for energy sources/sinks (viscous dissipation); the last term is the conduction term. The conduction term requires special attention, and to do this, it is instructive to derive equation (5).

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The flux of heat per unit time across a unit area $Q = U\rho C_v T$ (U is the mean velocity in the direction of the flux), can be expressed by

$$Q_i = -k_i \frac{\partial T}{\partial x_i} = -(\overline{k} \cdot \nabla T)_i. \qquad (6)$$

Equation (6), where the subscript indicates summation in the three coordinate directions, implies that the flux of heat is directly proportional to the gradient of temperature, with k representing the proportionality factor. If the conducting material is isotropic (conductivity independent of direction), the equation can be simplified by the dropping of the subscripts. A similar equation can be derived for the flux of a nonuniformly distributed material. The flux of material per unit area per unit time J is

$$J_i = -D_i \frac{\partial X}{\partial x_i} = -(\overline{D} \cdot \nabla X)_i \qquad (7)$$

where

X = the concentration of the material.

Analogous to the conduction of heat in a solid, D represents a proportionality between the flux of a material by self-diffusion and the spatial gradient of the material. In both cases the flux of the physical quantity is an attempt by the system to achieve equilibrium and is assumed to be driven by a force proportional to the gradient of the quantity not in equilibrium. Defining the thermal diffusivity $\kappa = k/(\rho C_v)$ and applying the total derivative $[D/Dt = \delta/\delta t + \nabla \circ]$ (for a more detailed exposition see Sutton [1]) equation (6) becomes

$$\frac{\partial T}{\partial t} + \nabla \cdot (\vec{U}T) = -\nabla \cdot (\kappa \nabla T). \qquad (8)$$

This is equivalent to equation (5) with one major exception; the thermal conductivity in equation (5) is outside of the spatial derivative operator, implying an isotropic assumption. Equation (7) is the more general form of the equation for the transport and diffusion of heat. Likewise, the general equation for selfdiffusion (molecular) of material in a gas is

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$$\frac{\partial X}{\partial t} + \nabla \cdot (\vec{U}X) = -\nabla \cdot (\vec{D}\nabla X), \tag{9}$$

and moving the diffusivity term outside of the gradient operator is only valid for conditions of isotropy (Fickian diffusion). This is a simplification that limits many of the transport and diffusion models in current use. The critical error is not in the application to molecular diffusion, which would be serious enough if used indiscriminately, but in the extension of this concept to the treatment of turbulent diffusion.

The discussion of diffusion, so far, is strictly applicable only to molecular diffusion. In this context, the concept of proportionality between the force and its effect is physically reasonable and well founded. Broadening the definition of D to allow it to represent turbulent diffusion, which, as will be shown, is the basis of many models, jeopardizes the general validity of equation (9). The scientists who first suggested the broader interpretation were cautious in its usage. When D represents molecular diffusion, a linear relationship between $\chi(x,y,z,t)$ and $\nabla\chi$ is reasonable and has been shown to be valid. However, a simple proportionality, when D represents turbulent diffusion, is questionable; the linkage between the rate of change and the gradient has vanished. It is no longer the gradient force that is responsible for the change, rather it is turbulent diffusion; a separate and independent physical process. Although this is not as serious as might first be expected[*], it is probably the singular fact that accounts for the inability of early attempts, using a molecular diffusion analogy, to model the effects of turbulent diffusion. These failures led to searches for the universal factor. The constructs that resulted from these attempts are so ubiquitous that they are often treated with the reverence that should be reserved for real physical quantities.

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[*]It can be shown that equation (9) may be considered valid. However D is no longer a simple proportionality constant but a modifying (and complicated) tensor-based expression (K_{ij}) of a physical force.

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5. Modeling Diffusion

While the earliest approaches to modeling turbulent diffusion were less than successful, a number of useful ideas emerged from the work of a collection of well-known scientists. The constructs developed introduced ideas and terminology (mixing length and friction velocity) that have outlived the theories, necessitating a review to put them in the proper perspective. An early approximation of turbulent diffusion borrowed an approach used in analyses of turbulent vertical transport of horizontal momentum. Defining an eddy diffusivity K analogous to the definition of molecular diffusivity, and replacing T in equation (6) by η, where η is the concentration of any physical species (momentum, or suspended material); Q represents the flux of that quantity. This provided a basis for analyses that proceeded on strictly heuristic arguments. The flux $Q' = K\nabla\eta'$, caused by the fluctuating components, was defined as $Q' = (\bar{\eta}'\bar{u}_i')\bar{\eta}'$ in which the prime has its usual meaning and u_i indicates the velocity in the x_i direction. Further, (with k in equation (6) replaced by an eddy diffusivity K) it was argued that η' was proportional to (and in the direction of) the material gradient leading to $K \equiv \bar{u}'\lambda$. Later Prandtl, [7] using dimensional arguments[*], identified λ as a mixing length analogous to the mean free path in molecular diffusion theory. [8] While this seemingly added physical validity to the approach, now known as K-theory, no tangible, measurable entity corresponding to λ exists, and K continued to be defined on empirical grounds. The introduction of friction velocity u_* was a further attempt at defining a physically reasonable and measurable quantity that could be used to characterize turbulence.

It was demonstrated that u_*, defined as the square root of the ratio between horizontal shear stress and material density, was relatively constant with height very near the ground, and could be related to the velocity correlation $\bar{u}'\bar{w}'$. Careful studies of flows over flat plates showed that an essentially logarithmic relationship characterized the ratio $\bar{u}/u_*$. The promise offered by these ideas, unfortunately, is limited to flat terrain and a narrow set of atmospheric

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[*]Molecular diffusivity has the dimensions of l^2/t, if K is to be treated analogous to diffusivity, it should have similar dimensions.

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conditions. This is evidenced by the numerous attempts and equations available to obtain u_*. However, in the theory of momentum transport, studies along this line continued, and it was shown for adiabatic[*] conditions that the eddy diffusion coefficient could be expressed as $K_M = u_*^{1/2}kz$. It was then argued that such a formulation was equally applicable for any physical entity. The persistent occurrences of nonadiabatic boundary-layer conditions confounds the application of K-theory in the real atmosphere, which was derived from experimental observations under ideal neutrally buoyant conditions. For nonstable conditions a number of functions $K_M(u_*, R_i)$ have been proposed. An alternative approach to the definition of λ (mixing length) was introduced by von Kármán [9] (also see Tatarski [10]). This led to an expression for u_* in terms of another parameter, roughness length (z_0).

Numerous attempts at resolving the inadequacies of K-theory have been attempted, each introducing a new parameter to correct the deficiencies introduced by attempts to generalize. The fault in all the attempts was not the ineptitude of the scientists but the difficulty in describing the behavior of a chaotic atmosphere in terms of well-behaved relationships designed for simple cases and that worked well for relatively quiescent periods (night, flat terrain, neutral buoyancy). The nature of turbulence and the inexact method required to assign values to z_0 and u_* imposes considerable problems in applications of this theory. The inexactness results from the fact that these parameters were constructed to represent artificial concepts rather than measurable physical quantities. The inclusion of the Richardson number, which acknowledges the importance of the vertical temperature gradient in determining the overall diffusion effect, must be noted; for in a highly unstable atmosphere, vertical diffusion is likely to be the dominant factor in the dilution of the material. Conversely, an overly stable condition will dampen any tendency for the material to diffuse vertically. However, a simple appeal to, or over emphasis on, atmospheric stability conditions as anything more than a rule of thumb masks the role of turbulence as the primary influence on material dispersion.

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[*]Adiabatic conditions, also referred to as neutral buoyancy conditions, are achieved when the vertical temperature lapse rate in any layer is equal to the rate of change of temperature, Γ_d ($= \delta T/\delta z$), that a dry parcel of air would experience if it were to rise without exchanging heat with its surroundings. $\Gamma_d \approx 9.76\ °C/km$.

It is the random nature of turbulence, coupled with a questionable correlation to molecular diffusion, that threatened the further generalizations of the approaches so far discussed, and led researchers to return to the consideration of a statistical approach.

Atmospheric processes are naturally Lagrangian* and only weakly correlated to past interactions. In contrast, analytic solutions to the physical equations and standard measurements of physical quantities are Eulerian[†] (time histories of events at a fixed location). Thus, Eulerian statistics are time correlations, while Lagrangian correlations are spatial in nature. Practically, interpretive results are generally portrayed in Eulerian space, while formal expressions of turbulence are Lagrangian. Therefore, the efficacy of necessary time-space transformations must be carefully analyzed. Pasquill [4] argues that errors introduced by this transformation are small, but his arguments are based on carefully controlled experiments in homogeneous-isotropic-stationary (h-i-s) turbulence and limited observations. He also points to the difficulty in making measurements of the Lagrangian time-scale and the wide variability in the limited data available (which may indicate that such time-space correlations are not well in hand). Taylor's hypothesis, [11] a utilitarian approach that attempts to avoid the time-space problem, provides the basis for many turbulent diffusion models. The tenet of the hypothesis, an assumption that turbulence at any point is unaffected by a translation in space by the mean wind, can be shown to be

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*The Lagrangian correlation relating variations with respect to space implies that the effect on a particle is determined by its physical location at a given time and can be expressed by

$$R(\xi) = \frac{\overline{u'(x,t_1) - u'(x + \delta x, t_1)}}{\overline{u^2}}$$

where $\delta x = u'(x, t_0) \, \delta t$.

[†]Eulerian correlations are based on measurements at the same location at different times

$$\mathbb{R}(x) = \frac{\overline{u'(x_1,t_1)u'(x_1,t_2)}}{\overline{u'^2}}.$$

tenuous in even the most benign conditions for which the hypothesis is supposed to be particularly relevant[*].

The Eulerian approach seeks solutions of equation (9) with possible terms for sources and sinks included. The strength of this approach is its capability to address nonlinear reactions (sources and sinks) and wide applicability when a suitable function for K_M is obtained. The Lagrangian approach relies on the ability to define a probability density function (PDF) $\psi(X_i, t)$ where ψ is the probability that a particle is in a given volume, or that it is at a location, $x_i < X_i < x_i + dx_i$. Then, if the concentration $c(x,t)$ is composed of m-number of particles,

$$<c(x_1, x_2, x_3, t)> = \sum_{k=1}^{m} \int\int\int_{-\infty-\infty-\infty}^{\infty\infty\infty} Q(x_i, t|x_i', t') \psi_k(x_i', t') dx_i' \qquad (10)$$

where

$\quad$ Q = the transition PDF that a particle at x′ at t′ will be at x at time t.

$\diamond$ indicates an ensemble average (a statistically averaged result). The form of and ability to evaluate the transition PDF is crucial to obtaining Lagrangian solutions, and is the foremost limitation of this technique. Monin and Yaglom [12] argued that empirical data indicate that Q is a multidimensional normal distribution.

Utilizing this assumption of normally distributed[†] turbulent energy and further restricting turbulence to be homogeneous, stationary, and isotropic provide sufficient simplification that analytic solutions to equation (10) are obtainable for specific cases. Further details of this approach are in the section on model

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[*]This topic is polemic between atmospheric physicists. However, the controversy can be lessened when viewed in terms of comparison of Eulerian and Lagrangian time scales.

[†]Normal distributions are sometimes referred to as Gaussian distributions. The requirement that turbulent energy be normally distributed leads to Gaussian puff/plume solutions.

descriptions. The obvious questions are: How representative are results based on these restrictive assumptions? Can a PDF, $Q(X,t|X't')$, be determined for the more realistic atmospheric boundary-layer case of nonstationary, nonhomogeneous, and nonisotropic turbulence? Extensive experimental measurements could provide these PDFs, but the validity (adjustment to the shape factors for the Gaussian distributions) are limited to those specific situations. Such applications are, at best, diagnostic, and attempts to reproduce anything other than trends are irresolute. Alternatively, numerical simulations of the turbulence structure provide PDFs representative of current conditions that provide a means to circumvent the restrictiveness of the Gaussian assumption and the diagnostic character of current techniques.

A third alternative is to cast the Lagrangian form (see equation (10)) into a differential equation that can be solved as a boundary-value problem. Assuming diffusion to be a Markov process (i.e., that the current physical state of any particle is dependent only on its immediately prior state), [13] then

$$
\frac{\partial <c>}{\partial t} + \frac{\partial}{\partial x_i}(\overline{u_i}(x,t) < c(x,t)>) = \frac{1}{2}\frac{\partial^2}{\partial x_i \partial x_j}[K_{ij} < c(x,t)>]
$$
$$
+ S(x,t) - r(t) < c(x,t)>
$$

(11)

where

S = a source term
r = a sink terms
u_i = wind velocity components
c = three-dimensional concentration
K_{ij} = represents a diffusion coefficient.

We note immediately that equation (11) resembles the advective form of equation (9) with the diffusion coefficient replaced by a tensor and subject to the double spatial derivative. Rigorously, the diffusion coefficient is a tensor. The true difference between K_{ij} and D is that K_{ij} is dependent on Lagrangian statistics; whereas, D relies on Eulerian statistics. The ability to cast the Lagrangian equation into a boundary-value problem does not solve the time-space problem. The properties on which K_{ij} are dependent are difficult to measure and are usually treated as parameters, the value of which are adjusted

to fit experimental concentration measurements. (The same approach that is used in forcing the results from solutions to equation (10) to match experimental data.) Therefore, neither the equation nor the problem has been adequately solved, and the accuracy of the results are dependent on how well the physical conditions of concern correspond to conditions to which the data were fitted.

An additional problem concerns solutions to equation (11). The diffusion equation is a parabolic differential equation. Equations of this type are characterized by a physically unrealistic property that implies that diffusion proceeds at infinite velocities, leading to problematic results that must be interpreted. An extension of the Markov process can be used to yield a hyperbolic (telegrapher) equation that describes diffusion occurring at finite velocities. [14] For all practical purposes, the results obtained by the telegrapher equation are minimally different. Although it solves one concern, the hyperbolic solution does not remove the problem of defining K_{ij}. It was suggested earlier that numerical simulations provide a means for obtaining applicable probability density functions for the turbulence (discussion following equation (9)). Deardorff and Peskin [15] and Deardorff [16] demonstrated a modeling technique now called large-eddy simulation (LES) that has the capability to predict accurate turbulence statistics. The model is a solution technique applied to the full set of the Navier-Stokes equations, permitting the full inclusion of meteorology, terrain, and other factors.

As pointed out by Stull, [17] the predictability achieved by any model of turbulence structure must be viewed in two ways: (1) actual pattern predictability and (2) statistical functions from pattern forecasts. While the simulated patterns of the LES do not necessarily correspond to true structures, it has been shown that for well designed simulations, the accuracy of the turbulence statistics can be maintained for forecasts extending several days. With the ability to numerically produce a PDF for a given terrain/meteorological situation, a specific solution to either form (equations (10) and (11)) of the Lagrangian formulation is possible without the attendant ambiguities introduced by parameterization. The superiority of the direct solution to the diffusion equation over the Gaussian plume/puff technique was demonstrated by Lamb et al. [18] This report also provides a short but excellent theoretical review and discusses the unrealistic Gaussian assumption.

6. Modeling Transport and Diffusion

There is a very large stock of hazard prediction models available. To differentiate among them, we can separate the models into three families: (1) solution technique, (2) level-of-detail within the solution technique, and (3) inventory of materials treated. Solution techniques can be broadly categorized as: (1a) statistical, including Monte Carlo and particle-in-cell, (1b) parametric, including most Gaussian puff/plume models, or (1c) numerical. Level-of-detail considerations include such factors as treatment of multiple sources, phase change, directionality, meteorology, and terrain[*]. The types (gas-liquid-solid) of material handled by a model impact not only the hazard prediction, because of the range of toxicities and effects available, but also T&D (particle mass/density and phase changes affect concentrations). Further, because the parametric family of T&D solutions has dominated modeling for military applications, discussions will focus on this approach. However, it will be immediately clear that the main conclusion is equally applicable to all classes.

All hazard prediction models in use are attempts at obtaining solutions to the advective form of the basic diffusion equation:

$$\frac{\partial \chi}{\partial t} + \frac{\partial (U_i \chi)}{\partial x_i} = \frac{\partial}{\partial x_i}[K_{ij}\frac{\partial \chi}{\partial x_j}] + S(x_i, x_j, x_k, t) + r(x_i, x_j, x_k, t) \qquad (12)$$

The salient difference in solution techniques is the manner in which the stochastic nature of the atmospheric processes, specifically the eddy diffusivity, K_{ij}, is treated. It was first recognized by Lord Rayleigh that the Monte Carlo (alternatively known as the method of statistical trials, or the random walk method) was a suitable method for solving boundary-value differential equations of random processes. It was not, however, until the advent of high speed computers that such methods were realizable. All models in the statistical family attack the problem by examining the behavior of a representative number of entities, eventually assigning the resulting average tendencies to the larger

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[*]See appendix B for a discussion of the interconnection of terrain and meteorology as it pertains to dispersion modeling.

ensemble of particles. The distinction between the models is in the achievement of the multiple trial concept. Pure Monte Carlo codes create the statistical ensemble through multiple runs on a few particles. Particle-in-cell methods employ a larger number of particles that are initially assigned to cells; averaging is accomplished on the collection of cells. In each case, the velocity (direction and speed) of the individual particles during each time step is determined by random functions (random number generators). To be valid, the applications of the random number generator must result in movements from one location to another that represent physical reality. To accomplish this, the random number generator must mimic some PDF. The shape of the probability distribution must be empirically determined so as to have the appropriate atmospheric turbulence characteristics. [19] In contrast to the random walk techniques, many parametric methods utilize particular solutions to equation (10). To arrive at a particular solution a number of simplifying assumption are necessary.

A number of simplifying assumptions are necessary to determine a transition probability for the case of atmospheric turbulent diffusion. First, it must be assumed that the turbulence is homogeneous and stationary. In this case, $Q(x,t|x',t')$ of a particle depends only on the displacements in time and space, not on when or where the particle was introduced into the flow. Secondly, the incorrect assumption that the turbulent energy is normally distributed is applied. Following Monin and Yaglom [12] a normal/Gaussian distribution of turbulent energy results in

$$Q(x - x', t - t') = \frac{1}{(2\pi)^{\frac{3}{2}}|P|^{\frac{1}{2}}} e^{-\zeta^T P^{-1}\zeta/2} \tag{13}$$

where

ζ^T = the transpose of a column vector of space correlations with the components $\zeta_i = x_i - x_1' - \langle x_i - x_i' \rangle$ (i = 1,2,3)

P^{-1} and $|P|$ = the inverse and determinant of the matrix of which the elements are $P_{ij} = \langle \zeta_i \zeta_j \rangle$.

An additional assumption of isotropic turbulence results in $P_{ij} = 0$ for $i \neq j$. For $\tau = t - t'$

$$Q(x - x', \tau) = \frac{1}{(2\pi)^{\frac{3}{2}}[P_{11}(\tau)P_{22}(\tau)P_{33}(\tau)]^{\frac{1}{2}}} e^{-\frac{1}{2}\sum_{1}^{3}\frac{\gamma_i^2}{P_{ii}(\tau)}} \tag{14}$$

Replacing Q in equation (10) with equation (14), defining the source distribution (a point source is given by the probability distribution, $\psi = S\delta(x-x_0)\delta(t') \ dx'$), and assuming the mean wind velocity is along a coordinate axis, we gain a trivariant joint normal time-dependent solution of the form typically found in models of the parametric-analytic class. A specific form (sample solutions are shown in appendix A) is dependent on the imposed boundary conditions as well as other assumptions. The results for any time t are obtained through integration of the appropriate particular solution with respect to time, producing well-behaved Gaussian plumes that are seldom realized in actual conditions. The solutions are characterized by the standard deviation of the PDF σ known also as the Gaussian spread factor. Inspection of the simple solutions (appendix A) immediately reveals the causes for the nonrealistic results:

a. Turbulent diffusion (the entire point of the model) is confined to three parameters or spread factors σ_i. Additionally, σ is a function of time.[*]

b. The transport term U is a constant.

c. The source S is singular and constant.

d. No provision for loss of material is included.

e. Concentrations are relative to a fixed axis, rather than relative to the center of mass of the puff.

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[*]The time dependence of σ, in reality a spatial and temporal dependence, is seldom treated.

Many different adjustments to the basic solutions have been applied, and while the purist may argue that the solution is no longer valid; pragmatically, such results can be forced to resemble experimental measurements. To repair the apparent violation of the Lagrangian spatial correlation implied by e, investigators have modified the equations to include a Lagrangian time scale factor. A large variety of methods have been applied to include the effect of material sinks (d), primarily through evaporation and deposition as well as casting S as $S(x,t)$; thereby, permitting time-dependent additional sources (c). Such adjustments are valid because the independence of x was a requirement for and time was not a factor in the derivation of equation (14). Likewise, allowing $U = \bar{u}_i(t)$ addresses (b) but does not invalidate the results. The most severe deficiency is contained in $\sigma = f(K_{ij},x,t)$ and the underlying assumption of a normally distributed turbulence field. Recall, to achieve this solution, it was necessary to assume that the turbulence was h-i-s, which for the boundary layer is rarely a valid assumption. Ignoring this problem, one is still faced with achieving a valid representation of turbulence commonly through the standard deviation, σ_i, of the normal (Gaussian) distribution[*]. The method most used is attributed to Pasquill, [4] and its further development by Gifford and others. [20,21] These methods that appeal to the physics embodied in the Richardson's number and the friction velocity with empirical modifications for the effects of cloud cover are referred to as the Pasquill-Gifford (P-G) stability category methods. (When the downwind dependence of σ_i is included, the method is sometimes attributed to McElroy-Poole.) [22] As the title implies, these methodologies provide a means for selecting approximate spreading factors σ_i based on estimates of the boundary layer stability condition. The inability of the technique to produce reliable results has led to additional, and often inappropriate, parameterizations. Given a sufficient number of adjustable parameters, a model certainly should be capable of reproducing a single given result, providing grounds for claiming success. The prototype model suggested by Taylor and Pasquill [4,5,11] was designed for use in the boundary layer as

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[*]Atmospheric turbulence is predominantly non-Gaussian. For dispersion, the nonuniform behavior is enhanced by the superposition of large and small eddys. Small eddys do not significantly contribute to the diffusion process and do not move independently as suggested by the Gaussian assumption. Rather, small eddys move within the larger structures producing a skewed distribution of turbulent energy.

were parameters from mixing-length theory that are routinely used in attempts to force a simple model to replicate a complex process. Therefore, if such a model is useful, its functionality must be assumed to be only appropriate to problems within the boundary layer. However, the boundary layer in general and, specifically, near surface releases are the situations least likely to encounter conditions adhering to the Gaussian assumption. [23] The use of a numerical methodology avoids the faults associated with the parametric approach, and offers the possibility of a model of greater general applicability.

For short periods of interest, some parametric approaches may provide an adequate solution for uncomplicated cases if the turbulence can be properly characterized. However, the atmosphere is a complex system that eludes simple parameterization, particularly for long time periods. It was previously noted that the prime deficiency, in the statistical and parametric approaches, is the proper characterization of the turbulence. With valid representations of turbulence as a function of time and space, either equation (10) or (11) can adequately describe concentration of material as a function of time. It was also noted that properly executed LES models have the proven capability of reliably providing statistics for atmospheric turbulence. The LES is the foundation[*] of a numerical approach to solving T&D problems and, ultimately, the prediction of hazard areas caused by the release of toxic materials at any location, height, or time. In addition to correct statistics and removal of the ambiguities that are inherent in parameterizations, advantages gained through use of a LES include the following:

a. complete four dimensional mean wind and temperature fields for a more complete treatment of transport and chemical reactions

b. complete four dimensional treatment of terrain and diurnal effects

c. prognostic capability on time and spatial scales pertinent to T&D

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[*]The LES is particularly suited for PBL applications, but proper structuring the code can extend its applicability. This is also true of other methods for solving the set of meteorological equations.

d. equal applicability to the statistical or parametric approach

In a sense, the numerical class is not a separate family, but a new entry point into the first two families.

7. Hazard Prediction Modeling, A Brief Survey

The point of this report is chemical and biological T&D, the results of which are spatial concentrations and distributions of a given material. T&D is at the core of hazard predictions, which often leads to consideration of such models as atmospheric dispersion codes. Hazard prediction generally requires a time integration of the concentrations produced by T&D calculations yielding dosages that can be assessed in terms of medical impacts. In this sense, T&D is a module within the hazards prediction scheme. In assessing the applicability of a given model to a problem, this modular aspect should be considered.

The modular notion provides further considerations. Added modules will likely increase computational time and memory requirements, but more critical is the proper and judicious combination of modules. To illustrate this point, consider recent uninformed attempt at increasing the reliability of hazard predictions by combining a Gaussian plume/puff model with a mesoscale meteorological model. The Gaussian model is an analytic solution designed to provide diagnostic results for short times and distances and generally has a implicit grid system fixed to the release point with one axis oriented in the direction of the mean wind. The mesoscale model is a numerical, predictive model, with an explicit fixed, independent grid. The distances over which a Gaussian solution might be considered valid are small compared to the typical mesoscale model spacing of 5 km. The entire domain of the Gaussian solution is likely to be subgrid scale and unresolvable, relative to the mesoscale meteorological model. More important, and increasingly so when higher resolution meteorological models are used, is the incompatibility of the methodology used to describe turbulence, the primary influence on changing concentrations. The incompatibility results in neglect of the available turbulence information or the need for substantial additional computations, which defeats the simplicity of the analytic model. Failure to use the turbulence information negates any advantage anticipated from the additional overhead associated with the improved meteorology. The complexity involved in using the data suggests replacing the Gaussian technique, which is not always applicable, with a compatible numerical solution of the basic diffusion equation. The value of improved meteorology can then be scenario driven.

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A point of particular relevance for military application is the persistency of agents. As toxic agents become more persistent, and delivery/intercept mechanisms become ubiquitous, the need for models capable of adequate treatment of transport and diffusion for long time periods and at all levels in the atmosphere become more important. For releases of persistent agents at mid-to-high altitude, prognostic or predictive models are increasingly desirable. The following review includes models that have been used by various military agencies, but is not an exhaustive compilation. However, after the family to which a model belongs can be identified, a cognizant user can immediately determine the level of accuracy to be expected for a specific application. For additional details on a specific model, the reader is directed to cited references.

7.1 Statistical Models

Models in the statistical (Monte Carlo) family (MESO, [24] RAPTAD, [25] and, ADPIC [26]) have general applicability. As pointed out, the accuracy of statistical T&D results is dependent primarily on the accurate portrayal of the turbulence data. Capable of diagnostic results, these models are well suited to running in a predictive mode if coupled to a prognostic meteorological model. RAPTAD is the only statistically based T&D model linked with such a model. However, in general terms the associate model HOTMAC is capable of a minimum grid resolution of 2 km*, which is questionably too large for appropriate turbulence statistics. Additionally, RAPTAD does not address issues associated with change of phase; therefore, it is restricted to use for the dispersion of gaseous material. The Monte Carlo approach is a viable and attractive method for the solution of the hazard prediction problem. Its primary drawback is the fact that its accuracy goes as $N^{-\frac{1}{2}}$. This suggests that a very large number of statistical trials N must be computed. Each of these models are dispersion rather than hazard prediction codes.

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*General applicability demands that a model treat the majority of expected conditions. Meteorological models developed with a "hydrostatic" assumption (e.g., HOTMAC) suffer from the inability to treat convectively unstable atmospheric conditions. For such conditions, it is not advisable to use a computational grid with spacing smaller than approximately 2 km.

7.2 Gaussian Models

The comparison of models within the parametric family, including OB/DG (Ocean Breeze/Dry Gulch), CHARM (Chemical Agent Release Model), AFTOX (Air Force Toxic Corridor), NUSSE (Non-Uniform Simple Surface Evaporation), GAPCAP (Generation of Assessment Patterns for Clouds of Airborne Particles), VLSTRACK (Vapor Liquid Solid Tracking), MACH-LT (Model of Atmospheric Chemical Hazards - Laptop) , requires a comparison of the level of detail included in each of the models. (See appendix B for a discussion of terrain interaction in T&D modeling.) All the mentioned models, except OB/DG, rely on the P-G stability parameterization to determine the effective atmospheric diffusion. The utilization P-G categories limits the applicability of a model to descriptions of releases on or near the surface of the earth.

1. OB/DG - [27] Look-up tables determine atmospheric effects; tables available for 36 gaseous materials; gases considered are neutrally buoyant. Continuous point sources only. No mass loss or gain considered. Single point and time wind and temperature input. Flat terrain. Diffusion parameterized to single vertical temperature variation with height. Dispersion code.

2. CHARM - [28] Standard Gaussian Puff routine with parameterized increase in dispersion in wake of obstructions (buildings). Gases only - data available for 136 gases with wide variety of gas densities. Continuous point sources only. Mass gain from evaporation allowed. Single-point wind and temperature at multiple times. Flat terrain. Diffusion based on criteria similar to P-G methodology. Dispersion/hazard code.

3. AFTOX - [29] Standard Gaussian Puff routine. Handles all chemicals with known molecular weight. Gases only, but treats mass gain from evaporation. Single point and time wind and temperature. Flat terrain. Diffusion based on P-G methodology, boundary-layer parameterization used to calculate stability. A dispersion code.

4. NUSSE4 - [30] Standard Gaussian Puff routine. Continuous-point sources only. Tracks liquid and gaseous clouds. Losses caused by deposition and

(vapor) mass gain from surface evaporation of deposited liquids treated. Extensive use of agent look-up tables. Single-point wind and temperature. Flat terrain. Diffusion based on P-G parameterization. Dispersion/hazards code.

5. GAPCAP - [31] This model is a variation of the NUSSE family of models, modified to treat solid biological agents, accounting for the associated decay of such agents. Uses look-up tables for agent parameters. Flat terrain. Single-point wind and temperature. Diffusion based on P-G parameterization. Dispersion/hazards code.

6. VANDECAP/PACCE - [32] Modified version of GAPCAP. Flat terrain. Attempts to use a single profile of wind and temperature to permit application to high altitude releases. Dispersion/hazards code.

7. VLSTRACK - [33,34] Standard Gaussian Plume/Puff routine. Continuous-point and line sources. Tracks clouds consisting of liquid, gaseous, and solid phases. Losses caused by deposition and mass gain from evaporation surface evaporation of deposited liquids treated in some versions. Agents in database. Various input schemes for wind and temperature (discussed below). Flat terrain. Dispersion based on P-G parameterization. Dispersion/hazards code.

8. MACH-LT - [35] Modified Gaussian Plume/Puff routine. Continuous-point and line sources. Tracks liquid and gaseous and solid clouds. Losses caused by deposition and mass gain from evaporation surface evaporation of deposited liquids treated. Agents in database. Single-point wind and temperature. Flat terrain. Diffusion based on statistics derived from LESs over similar terrain. Diffusion/hazards code.

7.3 Numerical Models

Despite the astounding advances in computation power experienced over the past decade, a complete numerical description of all the physical processes in a complete first principles approach is not yet possible for the scales that would be required for very accurate or reasonable hazard prediction modeling. However, it is possible to use numerical techniques that provide a substantial improvement to either the representation of the problem or the application of the statistical techniques.

1. SCIPUFF - [36] Second-order Closure Integrated PUFF. Continuous-point and line sources. Tracks clouds consisting of liquid, gaseous, and solid phases. Treats mass losses and gains. A direct numerical solution of the diffusion equation (12) with turbulence treated in a manner analogous to standard perturbation theory. Capable of calculations for complex terrain. Diffusion is based on statistics for turbulence spectra inferred from climatological data and more recently derived from a turbulence closure model. The primary limitation of SCIPUFF is in the treatment of K the diffusion coefficient as a scaler quantity. Dispersion code.

2. ABCSIM - [35] Atmospheric Biological Chemical Simulations. Continuous-point and line sources. Tracks clouds consisting of liquid, gaseous, and solid phases. Treats mass losses and gains. A family of modules that include an LES complex terrain meteorological model and a direct numerical solution of the diffusion equation (12). Turbulence statistics are derived from the LES, and the diffusion coefficient is treated in its tensor form. Diffusion/hazards code.

3. ABCSIM-LT - [35] Atmospheric Biological Chemical Simulations - Laptop. A scaled-down version of ABCSIM for use on small computers. Reduced capability relates to the computation of the vertical structure of wind and turbulence. Speed and memory requirements restrict LES computations to approximately the lowest 1 km of the atmosphere. Transport and diffusion calculations are unchanged. Diffusion/hazards code.

8. Conclusions and Recommendations

The inference of this report is that adequate predictions of hazards associated with the dispersion of agents into the atmosphere requires that substantial attention be placed on the efficacy of the method in which T&D are treated. While VLSTRACK is not a T&D model, it does contain and is under consideration to be the model of choice by many DoD agencies. Therefore, a number of specific comments are addressed to that model.

A description of the evolution of a cloud of material suspended in the atmosphere can be separated into transport (the movement of the center of mass) and diffusion or the changing distribution of the material about the centroid. Transport is derived from knowledge of the wind at the location of the centroid at given times, or estimated by assuming a mean wind over the entire travel distance. Either case is the result of a relatively simple calculation. Superimposed on this are other physical processes such as phase change and the more elusive process of turbulence-induced dispersion.

In general, diffusion models based on a parametric paradigm are useful as estimators or indicators of trends, provided atmospheric conditions, including terrain effects, are relatively benign and constant and dispersion times are relatively short. Persistent agents and complex terrain (appendix B) can only be addressed at increasingly lower accuracy levels and at considerable computational expense. The Gaussian model, while physically reasonable for pure molecular diffusion, is not generally representative of boundary-layer turbulent diffusion except for large averaging times. [23] Therefore, Gaussian-based models have a severe inconsistency between the underlying physical realities and modeling assumptions. The basis of the model, the diffusion equation (12), is derived from analyses of molecular motion and, as such, is dependent solely on physical properties of the fluid. Turbulent diffusion is not intrinsic to the fluid or the dispersing species, rather, it is driven by external forces. Without regard to the questionable assumptions required to obtain the Gaussian solution, this suggests caution should be exercised in the application of the equation for the description of events dominated by the mechanism termed turbulence. The limited success of early investigators who

attempted to extended the notions of molecular diffusion to include turbulence effects masks the fragility of a generalization of the approach.

In addition, diffusion resulting from turbulence is a local phenomenon, and unless the cause of the turbulence is uniform over a large area (or minimally the path on which the calculation is occurring), utilizing a parameterization based on measurements at a single location is subject to large errors. Measurements used to verify models, including early models based on the P-G stability category, were conducted in regimes that adhered to this limitation. It is not surprising that there is some agreement between model results and measurements. These comparisons also verify the decrease in accuracy with time. This is an expected result, a single simple solution can not be expected to adequately track the effects of a dynamic atmosphere and Gaussian plume/puff models are based on one simplified solution of the molecular diffusion equation. Coupling such solution methods to a four-dimensional dynamic meteorological model has questionable merit.

Meteorological models useful for application to T&D problems add substantial computational overhead. If there is a benefit to be gained from the additional computations, it should be in a better representation of the mechanism that contributes to the dilution of the species being tracked. If the meteorological model is used solely for wind information, the more important information is being discarded. Using the available turbulence information to improve a Gaussian model would require considerable restructuring, and the benefit is questionable. Certainly, a better estimate of the standard deviation σ should improve model specificity and results, but the model remains Gaussian and not representative. The additional computations and necessary factor fiddling contradicts the purpose of an analytic solution. The contradictions are intensified when a meteorological model of sufficient resolution is used.

Consideration of resolution is an important aspect because it controls computational resource requirements as well as accuracy of results. For example, consider a case used to validate VLSTRACK. The largest total area coverage for any trial was about 41,000 m^2, equivalent to a 114-m-radius circle. Using the HOTMAC mesoscale model with an approximate 2000-m minimum resolution (refer to footnote on a page 28), a single HOTMAC cell is then

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97 times larger than the largest area used in the validation of the VLSTRACK model. Therefore, at best, information from two HOTMAC grid points would be useful to the entire T&D calculation. Because VLSTRACK is not configured to use turbulence information from a meteorological model, only wind information, approximately 20 percent of the relevant information, would be used. From a diagnostic point of view, the single data point input method of the NUSSE model would have been sufficient. HOTMAC (or any mesoscale model) used in conjunction with VLSTRACK can provide prognostic winds but at a resolution insufficient to substantially influence the results. Models for higher resolution winds are available, but the computational overhead required to use these products with VLSTRACK is inefficient.

Additionally, users of the model are cautioned against reliance on the validation and documentation of VLSTRACK. [33,34] These works were incorrectly titled validation. The work reported in these nonreferred records documented a parameter-fitting methodology. The VLSTRACK validation reported by Sterle et al. [37] documents the inability of the analytic Gaussian model, specifically VLSTRACK, to replicate field-trial data collected during less than stressful terrain and meteorological conditions. Sterle et al. attributes the less than acceptable performance of the model to "inherently large variability and unpredictability of the meteorological conditions, which play a critical role in determining the distribution pattern of agents and stimulants following dissemination into the atmosphere. Users of VLSTRACK or any other chemical/biological dispersion model should be cognizant of this limitation and thereby avoid placing too much weight on the model outputs most affecting borderline decision making." [37]

It must be pointed out that such cautions should be applied in any case, but meteorology is not completely unpredictable for the time scales of interest, and the cited limitations are applicable only to highly parametric Gaussian models in which all the variability and unpredictability of the meteorological conditions is contained in a single parameter. Likewise, the abandonment of models such as VLSTRACK would focus funding and labor on more reasoned approaches to the proper inclusion of variable meteorology and turbulence. Regardless of the paradigm used, an adequate description of the diffusion process can only be obtained if the distribution of turbulent energy is known or correctly estimated.

Models based on the Gaussian assumption are confined to a normal distribution and to isotropicity, homogeneity, and stationarity, which do not have general applicability in the boundary layer of the atmosphere. In the absence of proper turbulence statistics, statistical and numerical models can use a Gaussian assumption but are normally free to apply any distribution to the turbulent energy. The accuracy of these simulations is dependent on the number of trials and/or the shape of the energy distribution. In this aspect, parametric models are the least flexible and subject to the largest errors.

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Acronyms and Abbreviations

ABCSIM	Atmospheric Biological Chemical Simulation
ABCSIM-LT	Atmospheric Biological Chemical Simulation - Laptop
AFTOX	Air Force Toxic Corridor
CHARM	Chemical Agent Release Model
DoD	Department of Defense
GAPCAP	Generation of Assessment Patterns for Clouds of Airborne Particles
h-i-s	homogeneous-isotropic-stationary
LES	large-eddy simulation
MACH-LT	Model of Atmospheric Chemical Hazards - Laptop
NUSSE	Non-Uniform Simple Surface Evaporation
OB/DG	Ocean Breeze/Dry Gulch
PBL	planetary boundary layer
PDF	probability density function
P-G	Pasquill-Gifford
Re	Reynolds number
SBL	surface boundary layer
SCIPUFF	Second-order Closure Integrated PUFF
T&D	transport and diffusion
VLSTRACK	Vapor Liquid Solid Tracking

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Appendix A

Gaussian Solutions for Lagrangian Dispersion

The following are examples of solutions to the diffusion equation (12) using Gaussian assumptions of equation (14) for mean concentrations:

1. Instantaneous point source at x_0. Mean wind $\bar{u}_i = U$; source strength, S (g), no boundary conditions.

$$<c(x,t)> = \frac{S}{(2\pi)^{\frac{2}{3}}\sigma_1(t)\sigma_2(t)\sigma_3(t)}e^{-\frac{(x - x_{1_0} - Ut)^2}{2\sigma_1^2(t)} - \frac{(x_2 - x_{2_0})^2}{2\sigma_2^2(t)} - \frac{(x_3 - x_{3_0})^2}{2\sigma_3^2(t)}} \qquad (A\text{-}1)$$

2. Continuous-point source at x_0. Mean wind $\bar{u}_i = U$; source strength, S (g/s), no boundary conditions.

$$<c(x,t)> = \frac{S}{(2\pi)\sigma_2(x - x_{1_0}))\sigma_3(x_1 - x_{1_0})U}e^{-\frac{(x_2 - x_{2_0})^2}{2\sigma_2^2(x - x_{1_0})} - \frac{(x_3 - x_{3_0})^2}{2\sigma_3^2(x_1 - x_{1_0})}} \qquad (A\text{-}2)$$

3. Instantaneous point source at x_0. Mean wind $\bar{u}_i = U$; source strength, S (g), reflecting barrier (ground) at $x_3 = 0$.

$$<c(x,t)> = \frac{S}{(2\pi)^{\frac{2}{3}}\sigma_1(t)\sigma_2(t)\sigma_3(t)}[e^{-\frac{(x - x_{1_0} - Ut)^2}{2\sigma_1^2(t)} - \frac{(x_2 - x_{2_0})^2}{2\sigma_2^2(t)}}][e^{-\frac{(x_3 - x_{3_0})^2}{2\sigma_3^2(t)}} + e^{-\frac{(x_3 + x_{3_0})^2}{2\sigma_3^2(x_1 - x_{1_0})}}] \qquad (A\text{-}3)$$

4. Continuous-point source at x_0. Mean wind $\bar{u}_i = U$; source strength, S (g/s), reflecting barrier (ground) at $x_3 = 0$.

$$<c(x,t)> = \frac{S}{(2\pi)\sigma_2(x - x_{1_0}))\sigma_3(x_1 - x_{1_0})U}e^{-\frac{(x_2 - x_{2_0})^2}{2\sigma_2^2(x - x_{1_0})}}[e^{-\frac{(x_3 - x_{3_0})^2}{2\sigma_3^2(x_1 - x_{1_0})}} + e^{-\frac{(x_3 - x_{3_0})^2}{2\sigma_3^2(x_1 - x_{1_0})}}] \qquad (A\text{-}4)$$

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Appendix B

Aspects of Terrain Influence on Dispersion Modeling

The dispersion of material release near the ground is subject to a number of factors of which all but one are correlated to meteorological considerations. The exceptional influence is the fact that the surface provides a no flux boundary condition. In numerical and statistical models, this condition is easily treated in the natural grid system. In the case of the parametric models, equations (A-3) and (A-4) of appendix A illustrate the methodology. Except in flat terrain, this procedure adds an additional complexity that is not easily treated.

Terrain influences on meteorology that are of major significance to dispersion are (a) steering effect, (b) generation of mechanical turbulence, (c) thermal effects on the general flow, the turbulence, and possibly the chemistry. The steering influence of the terrain on the mean air flow is the easiest to imagine; and, as the discussion in the main text suggests, the simplest to include in any type of dispersion model. The parametric class simplicity demands an assumption of constant wind direction. However, the nonconstancy of wind direction is a major reason for the inclusion of terrain effects. Without this assumption, the same invalidation or stricture associated with the inclusion of surface roughness effects, discussed next, are applicable.

The generation of mechanical turbulence is a function of the surface roughness. Thermally generated turbulence is dependent on surface composition. Both are, in general, localized effects and a most cogent reason for including terrain effects in a dispersion model. The simplicity of a parametric model is destroyed when attempts are made to include the general variability associated with terrain roughness and directional variability. In addition to the requirement to use an alien grid system which substantially increases the necessary number of computations, either the validity of the equations (the form in general use assumes that the wind direction is constant and along one of the solution axis) is destroyed or the need to use a more general and complicated solution to equation (12) is dictated. Current parametric models are not designed to incorporate the variability of turbulence effects. All models of this class negate the advantages of including terrain through reliance on the generalization represented in the Pasquill-Gifford stability categories.

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Thermal effects associated with diurnal features such as mountain/valley winds and land/sea breezes are additionally compelling and important reasons for the inclusion of terrain effects.

The foregoing demonstrates that the terrain only indirectly, through the meteorology, influences transport and diffusion. Therefore, with the single exception of providing a reflecting barrier in the calculation of vertical transport and diffusion, terrain is the providence of the meteorological model.

Appendix C

Comments on the Model VLSTRACK

The Vapor Liquid Solid Tracking (VLSTRACK) documentation has numerous errors and inaccuracies, that would and could not escape the attention of a serious scientific reviewer. The document is useful as a user's manual, but the scientific content reveals a lack of complete understanding of meteorology and of T&D theory. The procedures for model validation as exhibited in the validation document does not meet the requirements of AR-5-11, and further demonstrates a lack of familiarity with the difference between model validation and model testing. It is unlikely that the model is capable of results sufficiently better than non-uniform simple surface evaporation (NUSSE4). The published differences in results [33,34] are most likely due to the manipulation of VLSTRACK modeling factors, during "validation", leading to a minimization of the difference between field data and model results. No such manipulations were performed during the NUSSE4 model evaluations[*]. In this sense as well as in a more specific sense, versions of VLSTRACK later than 1.2 have not been validated. In particular, validation of versions that contain significant changes to accommodate the higher resolution meteorology has not been accomplished. The types of improvements necessary to gain an advantage from "better/representative" meteorology can be more efficiently applied to either the statistical or numerical models. Further, if as noted in Sterle et al., [37] no significant differences exist in the results of version 1.4 and 2.0, one must question the need for the "improvements" as well as the significance of coupling VLSTRACK and a meteorological model, and the increased complexity such a combination introduces with no change in results.

It is recommended that if VLSTRACK is used, that it is used in a stand alone configuration (VLSTRACK 1.2), and that no further development be applied. The model is state-of-the-art for its configuration and "improvements" can only provide minimal changes.

In assessing the value of a model, one must be acutely aware of its limitations and strengths. Hazard prediction simulations are, by nature, multi-disciplined, requiring elements of physical chemistry, meteorology, atmospheric physics,

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[*]In comparison to NUSSE, VLSTRACK does provide a easier-to-use user interface for model initialization and it's graphical output substantially eases the task of output analyses.

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medicine and some esoteric science that is capable of describing the initial conditions. To expect a single individual to possess the all the required knowledge to produce an adequate model for the protection of personnel is courting disaster.

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