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ATMOSPHERIC TRANSPORT OF RADIONUCLIDES

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ATMOSPHERIC TRANSPORT OF RADIONUCLIDES*

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ABSTRACT

The chairman and contributors are members of the Working Group on Atmospheric Dispersion, Deposition, and Resuspension. This group examined the mathematical approaches for determining the direct and indirect pathways to man of releases of pollutants to the atmosphere. The dose-to-man limitations promulgated by the Nuclear Regulatory Commission, the Environmental Protection Agency, and the Energy Research and Development Administration** were presented. The present status of research was discussed, and recommendations for future work were made. Particular emphasis was placed on the need for additional experimental work to develop confidence limits leading to acceptable probability statements of critical pathways for determining the dose-to-man.

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INTRODUCTION

The purpose of atmospheric transport and diffusion calculations is to provide estimates of concentration and surface deposition from routine and accidental releases of pollutants to the atmosphere. These calculations provide the link between emissions to the atmosphere and direct or indirect pathways to man important for dose calculations. To focus the discussions of this working group, Nuclear Regulatory Commission (NRC) Regulatory Guides 1.109¹ and 1.111² were distributed to participants ahead of the meeting. In addition, the meteorological aspects of these Guides were presented as the first talk during the meeting of the working group. Key assumptions and methods were identified, tentative accuracy statements appropriate to these methods were suggested, and recommendations to improve NRC methods (keeping in mind the objectives of the Regulatory Guides) and for further research were developed. In addition to the presentation of the NRC Regulatory Guide methods, there were also presentations on the methods used by Environmental Protection Agency (EPA) personnel and used in WASH-1400.³

The most commonly used atmospheric concentration calculational method is the Gaussian plume equation. This is an empirical formula which is based on an analytical solution to the diffusion equation under the assumptions of constant wind speed, no wind shear, flat topography, and Fickian diffusion. It has shown considerable success under ideal field test conditions. Such a distribution can also be assumed from a statistical consideration. The equation for a continuous point source is

$$\chi = \frac{Q}{\pi u \sigma_y \sigma_z} \exp \left[-\frac{1}{2} \left(\frac{h}{\sigma_z} \right)^2 - \frac{1}{2} \left(\frac{y}{\sigma_y} \right)^2 \right] \quad (1)$$

where:

χ = atmospheric concentration at ground level for a release point h meters above the ground, units/m³

Q = source term, units/second

σ_y = standard deviation of a Gaussian distribution in the cross wind direction, m

σ_z = standard deviation of a Gaussian distribution in the
vertical direction, m

u = wind speed, meters/second

y - cross wind distance, m

The σ_y and σ_z are not defined explicitly by the mathematical assumptions; they must be determined empirically. Thus, a number of different field experiments have been conducted to determine σ_y and σ_z as functions of atmospheric stability conditions and of distances downwind. Our discussions drew heavily upon recent papers by Gifford⁴ and Vogt.⁵ References were also made to a draft position paper on the accuracy of diffusion models [being prepared by the American Meteorological Society (AMS) Committee on Atmospheric Turbulence and Diffusion] and to a summary of recommendations made by the American Meteorological Society Workshop on Stability Classification Schemes and Sigma Curves (being compiled by S. R. Hanna) following the June 27-29, 1977, workshop held at AMS Headquarters in Boston, Massachusetts. Neither of these latter two documents has been finalized or published yet.

The rest of this report from the Atmospheric Working Group consists of a series of short topical summaries important to the atmospheric calculations. The working group as a whole discussed each one of these topics under the guidance of a discussion leader.

STABILITY CATEGORIES

Pasquill Curves

The most widely known method for determining atmospheric stability categories is the one originally proposed by F. A. Pasquill⁶ in association with his first tentative σ_z and σ_y curves. This general classification scheme, based on insolation, cloud cover, and wind speed, was adapted by D. B. Turner⁷ for use with standard National Weather Service observations at airports. This latter computational scheme evolved into what is known as the STAR computer program which is run by the National Climatic Center at Asheville, North Carolina. D. H. Slade⁸ found that σ_θ (the standard deviation of the horizontal wind direction)

stratified the varied data from different diffusion experiments. This method was also simplified by taking the range of wind directions from an analog trace over a half-hour period and dividing by six to get an approximate value for the σ_θ . NRC Regulatory Guide 1.23⁹ also recommends the use of a temperature gradient, with one temperature being at approximately 10 m above the ground, and the other temperature being measured at approximately 40 m (or the top of the stack) to determine stability classifications. Golder¹⁰ developed relations between the Pasquill stability classifications and the Richardson number. Smith, et al.¹¹ used the measurements of σ_θ directly in the equations for σ_y and σ_z as a function of distance.

These various classification schemes and the relationships among them are summarized in Table 1.

Intuitively, one would believe that the direct measurement of turbulence σ_θ and σ_ϕ (the standard deviation of horizontal and vertical elevation angles, respectively) should be the most appropriate method. However, data sets are not available to test adequately all of these classification schemes against measured concentrations. Sensitivity studies have been performed (e.g., Pendergast¹²) where the same σ_y and σ_z curves were used in all cases, but different methods were used to classify atmospheric stability. These tests indicate that the selection of a stability category alone can make a difference in the annual average concentration calculations of a factor of ± 2 .*

Recommendations

Where possible, the working group recommends direct measurements of σ_θ and σ_ϕ should be used in equations for σ_y and σ_z , respectively, as a function of distance, x , such as suggested by Draxler.¹³ The second method of choice would be to use measurements of σ_θ and σ_ϕ as a method of selecting σ_y and σ_z curves, respectively. It is recognized, however, that measurements of σ_θ and σ_ϕ are difficult to obtain routinely over

* In this report, differences in a variable, x , of a factor of $\pm n$ means that the value of the variable ranges from x/n to nx .

Table 1

a. Pasquill Stability Categories

A: Extremely unstable conditions	D: Neutral conditions*	
B: Moderately unstable conditions	E: Slightly stable conditions	
C: Slightly unstable conditions	F: Moderately stable conditions	

Surface Wind Speed (at 10 m), m/sec	Daytime Insolation			Nighttime Conditions	
	Strong	Moderate	Slight	Thin Overcast or >3/8 Cloudiness [†]	≤3/8 Cloudiness
<2	A	A-B	B		
2-3	A-B	B	C	E	F
3-5	B	B-C	C	D	E
5-6	C	C-D	D	D	D
>6	C	D	D	D	D

* Applicable to heavy overcast day or night.

[†] The degree of cloudiness is defined as that fraction of the sky above the local apparent horizon that is covered by clouds.

b. Different Methods of Defining Categories

Pasquill	Turner (STAR Code) ⁷	σ_{θ} (deg) ⁸	$\Delta T/\Delta z$ (°C/100 m) ⁹	Golder (Ri) ¹⁰
A	1	25	<-1.9	Ri <-3.5
B	2	20	-1.9< to ≤-1.7	-3.5 ≤Ri <-0.75
C	3	15	-1.7< to ≤-1.5	-0.75≤Ri <-0.1
D	4	10	-1.5< to ≤-0.5	-0.1 ≤Ri <-0.15
E	5	5	-0.5< to ≤ 1.5	0.15≤Ri < 0.75
F	6	2.5	1.5< to ≤ 4.0	0.75≤Ri < 3.5
G ⁺⁺	7	1.7	>4.0	Ri > 3.5

⁺⁺ The original Pasquill F was split into two categories to allow for extremely stable conditions.

long periods of time (e.g., a year). Also, these measurements are only available from instrumented towers. The working group further recommends that the wind speed (u) at the height of emission be measured directly or a measured wind speed at a lower level after correction to the height of emission by a stability dependent function be used.

These wind speeds should be combined with estimates of the turbulence intensity of the atmosphere based on the Richardson number (Ri), the Monin-Obukhov length (L), the temperature gradient, or a measure of radiant flux (solar radiation, or sky condition and sun angle) to obtain stability categories.

These different methods should be directly tested against concentration measurements, not just against σ_y and σ_z estimates separately. Field tests should include a complete set of data so that all of the different methods of categorization mentioned can be evaluated simultaneously.

Investigations should also be conducted on the use of remote sounding techniques, like acoustic sounders, to characterize atmospheric turbulence.

GAUSSIAN PLUME MODELS

Limitations

It was recognized very early in the development of the atmospheric diffusion equations that the Fickian assumption necessary to derive the analytical form of the Gaussian plume equation does not hold well in the atmosphere. Thus, it has been necessary to conduct field experiments to determine, empirically, σ_y and σ_z as a function of distance and of stability.

Pasquill⁶ presented one set of σ_y and σ_z curves which has become widely accepted. These estimates were adapted by Gifford¹⁴ and Turner.⁷ The data on which these curves were based were obtained at ground level at distances of less than 1 km. The original tentative nature of these curves has been forgotten by many users; for example, these curves have been extrapolated to releases at great heights and to 100 km from the

source. The Pasquill curves were developed from near-surface release experiments conducted on smooth (a roughness coefficient of approximately 1 cm), flat areas with a sampling time of 3 to 10 minutes.

There are additional sets of σ_y and σ_z curves. McElroy and Pooler¹⁵ developed curves from low-level releases over the city of St. Louis, Missouri. These curves include the effects of a roughness coefficient of approximately 1 m and a one-hour sampling time. Smith¹¹ has also developed a set of σ_y and σ_z curves from tower releases (108 m) at the Brookhaven National Laboratory (BNL) for estimating diffusion over the surrounding forest areas. The sampling time was one hour and the roughness coefficient was approximately 1 m. Vogt⁵ has recently conducted a series of experiments near Jülich for releases from elevations of 50 m and 100 m. His data are also from a one-hour sampling time and a roughness coefficient of approximately 1 m. There are differences among these sets of curves, particularly between the Pasquill-Gifford curves and the rest. Vogt⁵ has published a very useful set of comparisons.

The working group suggested some tentative accuracy statements on the estimation of airborne concentrations. These statements are largely based on scientific judgment; there are not enough data upon which to base a reliable statistical estimate. For the ideal situation of a highly instrumented flat-field site from which previous data on meteorology and airborne concentrations were available, it should be possible to estimate to within ± 20 percent the ground-level centerline concentrations from a continuous point source at downwind distances of less than 10 km.

For a specific hour and downwind receptor point, the accuracy is very dependent on the calculation of the exact plume trajectory during a short period. For flat terrain and relatively steady meteorological conditions and distances of 10 km or less, the airborne concentrations for an individual case should be estimated to within about a factor of ± 10 . The ensemble of a large number of hours of air concentration calculations for a specific receptor point compares more favorably with the ensemble of air concentration measurements at that point (e.g., Pendergast¹⁶). Accuracy for the above situation and for the usual annual average concentration estimate is about a factor of ± 2 .

For distances of 10 to 100 km in relatively flat terrain, unpublished data on ^{85}Kr concentrations around the Savannah River Plant presented by Crawford to the working group indicate accuracies of about a factor of ± 4 for monthly and seasonal averages. It is expected that the scatter in these data will be somewhat reduced when these particular calculations are corrected for the time-dependent release of ^{85}Kr instead of using a uniform release rate for the month.

For complex terrain or meteorological situations (e.g., sea breeze regimes) a few experiments have indicated departures from estimates from the Pasquill-Gifford curves of more than a factor of 10. However, there are insufficient data upon which to base even a "scientific judgment" estimate of accuracy.

Recommendations

The use of at least two sets of σ_y and σ_z curves was recommended. One set should be used for surface releases. In this case, the most appropriate curves are those of Pasquill-Gifford with an adjustment for averaging time [$(\sigma_y)_{t_2} = (\sigma_y)_{t_1} (t_2/t_1)^{0.2}$] (see Pasquill¹⁷) and with a roughness coefficient adjustment ($\sigma_z = \nu z^{0.2}$) (suggested by G. A. Briggs during the AMS Workshop on Stability Classification Schemes and Sigma Curves). For elevated releases, it is recommended that either the Smith¹¹ or Vogt⁵ curves be used.

It was pointed out in this particular discussion, as well as in several of the others, that an accurate measurement of the source term, Q , is necessary. Environmental monitoring data have not been usable at many of the U.S. nuclear facilities to test various atmospheric diffusion models because Q is not known as a function of time (hourly values are desirable as that is typical for meteorological data). The common assumption of a uniform release rate is not often realized at nuclear facilities.

The Gaussian plume models must be validated against atmospheric concentrations to distances greater than 10 km, for greater ranges of stability, and in complex terrain. It is also desirable to measure

atmospheric concentration as a function of height in order to determine σ_z as an explicit function of downwind distance, and as a basis for examining the appropriateness of the reflection boundary assumption.

BUILDING WAKES

Status

The NRC guidelines include a building wake correction to the Gaussian plume equation. This correction for a Gaussian plume is indicated as:

$$\chi = \frac{Q}{u(\pi\sigma_z\sigma_y + 0.5 A)} \exp \left[-\frac{1}{2} \left(\frac{y}{\sigma_y} \right)^2 \right] \quad (2)$$

where A is the minimum cross sectional area of the building (m^2). The term $(\pi\sigma_z\sigma_y + 0.5 A)$ is only permitted to have a maximum value of $3 \sigma_z\sigma_y$. For sector-averaged concentrations, the σ_z term is replaced by $(\sigma_z^2 + 0.5 D^2/\pi)^{\frac{1}{2}}$ as long as this is equal to or less than $\sqrt{3} \sigma_z$. The symbol D stands for the height (in meters) of the building.

This relationship has been determined from limited field tests of diffusion around buildings, and from a larger series of wind tunnel tests. An isolated building has been assumed.

The accuracy of a building wake correction is difficult to estimate. The rough estimate might be a factor of ± 2 for routine releases at distances close enough to the building to be influenced by the wake. The use of a building wake correction is probably satisfactory for accidental releases, provided that the release is longer than a few minutes in duration and the wind speeds are greater than 1 m/sec.

Recommendations

It is recommended that the NRC Guides be continually reexamined as new data become available. More field experiments, particularly in stable and unstable light wind conditions, need to be performed. Currently, a set of experiments is being conducted by the NOAA staff at Idaho Falls, Idaho. In addition, some of the wind tunnel data should be reexamined for appropriate scaling. Recent information indicates that the mixed depth should be one of the scaling parameters in such studies, which has not been reported in published wind tunnel work on building wakes.

BUOYANT PLUME RISE

The NRC guidelines make use of the work of Briggs¹⁸ as adapted by Sagendorf.¹⁹ Basically, a few minor corrections were suggested for Sagendorf's¹⁹ adaptation, but nothing of substance. The working group essentially accepted the methods being used by NRC from the above two references.

As the focus in these discussions was on the NRC Guides^{1,2} and nuclear power plants, the actual use of the plume rise formula is not too critical for calculations related to routine operations at nuclear facilities. These facilities seldom have either buoyant or large momentum fluxes associated with their routine plumes; and, thus, the resulting concentration calculations are not very sensitive to methods for plume rise calculations.

MIXED-LAYER DEPTH

The depth of the mixed layer becomes important as a lid for vertical diffusion calculations. The most widely used summaries of mixed-layer depth are those prepared by Holzworth.²⁰ For a morning mixed-layer depth, Holzworth²⁰ has taken the 1200 CUT RAOB (an instrumented balloon temperature sounding), added 5°C to the morning minimum surface temperature, and extrapolated dry adiabatically upward to the intersection of the RAOB temperature-versus-height profile. This intersection is defined as the top of the mixed layer. For the afternoon depth of the mixed layer, Holzworth has used the same 1200 CUT RAOB and the afternoon maximum observed surface temperature extrapolated dry adiabatically to its intersection with the RAOB temperature profile. A large body of National Weather Service data has been used in these calculations, and Holzworth has published contour maps for the United States of monthly averaged morning mixed depths and afternoon mean maximum mixed depths.

Several shortcomings for the above method of obtaining mixed depths are obvious: the mixed depths represent only one time of day and are not necessarily the average for the whole afternoon or the whole morning.

Also, the intersection of the dry adiabatic line and the RAOB may be very oblique; the top of the mixed depth defined in this way may not be a "lid" to the vertical mixing, but a height above which the diffusion rate may just slow down. Furthermore, if radiation or advection causes the temperature profile to vary by as little as 1 to 2°C from the 1200 CUT RAOB, the calculated afternoon mixed depth may be in error by 50 percent or more, depending on the lapse rate (Pendergast²¹). The choice of a 5°C addition to the surface temperature to obtain the morning mixed depth is also somewhat arbitrary; yet a difference of ±1°C can cause differences in the mixed depth of as much as 200 m.

The calculated atmospheric concentrations at distances greater than about 50 km (depending on stability category and height of release) are inversely proportional to the mixed depth used in the calculation. Under these conditions, an error in mixing depth would cause a corresponding error in air concentration.

Recommendations

It is recommended that these mixed depth heights be measured with either temperature or acoustic sounders, and that research be performed to develop diurnal mixed-depth data (from measurements) for different locations and for different seasons. A criterion for defining a "lid" versus a slower rate of diffusion should also be developed.

REMOVAL AND SUSPENSION

Status

During the past decade, a number of advances have been made to understand and quantify removal and resuspension processes. Generally, the NRC Guides^{1,2} have used older methods to describe dry deposition (Markee²²) and wet removal (Engelmann²³). In the Guides, resuspension is not treated explicitly, nor do any of the methods discussed in the Guides (or in this section) include gravitational settling of large particles (say $\geq 10 \mu\text{m}$), as release of large particles from reactors is not probable.

To parameterize dry deposition and resuspension, regardless of the method used to describe the contaminant's air concentration, two boundary conditions must be specified at the interface between the atmosphere and the surface medium. One inviolable condition is equality of the normal components of the fluxes in the two media at the interface. For the special case that the net flux past the interface is zero, then only the single (reflection) boundary condition is needed. For the general case, there are a number of alternative parameterizations for the second boundary condition. Some of these are listed in Table 2.

Dry Deposition

To describe dry deposition, the NRC Guides^{1,2} use Calder's parameterization (cf. Table 2). This method is a replacement of the flux gradient relationship with a deposition velocity times surface concentration.

$$F = K_z \frac{\partial \chi}{\partial z} = v_d^c \chi_i \quad (3)$$

With this formulation, no account is taken of processes such as resuspension or molecular diffusion of gases. Yet, field measurements of F and χ (usually χ_b instead of χ_i) include all of these processes. Thus, v_d^c becomes a "lumped" parameter which is usually only applicable to the particular situation of measurement.

For a gas like molecular iodine, which reacts rapidly and irreversibly with the surface, field measurements of deposition in grass (converted to a unit area of soil surface) typically give deposition velocities (v_d) of a few cm/sec. This indicates that flux is limited by transfer through the atmosphere.

Through the constant flux layer, the transfer velocity is about u_*^2/\bar{u} where u_* is the friction velocity. Typically, u_*^2/\bar{u} is a few percent of the mean wind speed. This explains the frequent use of a value for v_d of a few cm/sec for annual average estimates. This value is appropriate for material whose deposition is limited by transfer through the constant flux layer. Sensitivity studies have been

Table 2. Some Parameterizations for the Second Interfacial Boundary Condition (Adapted from Slinn)^{3e}

Options	Interrelations and Comments
1. Specification of an equilibrium jump discontinuity:	
(a) Gases (liquid adsorber) $\chi_* = H c_*$	χ_* is the pollutant's concentration in the atmosphere, at the interface, that would be in equilibrium with the equilibrium concentration c_* at the interface in the bounding medium; H is Henry's constant for gases which form simple solutions.
(b) Particles $\chi_* = J_* c_*$	J_* is a parameterization for the equilibrium jump discontinuity.
$\chi_* = K_* G$	$G \approx \delta c_i$ is an approximate "surface concentration" where δ is an approximate depth to which the pollutant has penetrated into the surface medium; therefore $K_* \approx J_*/\delta$.
$\chi_b = KG$	K is known as the resuspension factor; note that use of the bulk concentration in the atmosphere, χ_b , rather than the equilibrium concentration at the interface, χ_* , can severely restrict the value of the resuspension factor.
2. Specification of the magnitude of the common flux, F:	
(a) Gases $F = k_a (\chi_b - \chi_i)$	k_a is the transfer velocity in the atmosphere from where the bulk concentration χ_b is measured, to the interface, subscript i; for the measurement at height h and a mean eddy diffusivity K in this interval, then $k_a \approx K/h$; it is usually assumed that $\chi_i = \chi_*$.
$F = k_s (c_i - c_b)$	Similarly, k_s is a net, downward, transfer velocity in the surface medium to where the bulk concentration, c_b , is measured.
$F = k_o (\chi_b - Hc_b)$	k_o is an overall transfer velocity; from previous expressions, $k_o^{-1} = k_a^{-1} + H k_s^{-1}$ which shows the overall transfer resistance, k_o^{-1} , to be like the sum of electrical resistances in series.
(b) Particles	
$F = v_d^c \chi_i$	v_d^c is Calder's deposition velocity; v_d^c can have any value less than infinity.
$F = v_d^{ch} (\chi_h - \chi_i)$	v_d^{ch} is Chamberlain's deposition velocity; h is a convenient reference height; a resistance analogy can be developed.
$F = v_d \chi_i - v_r c_i$	v_r is a resuspension velocity. ^{3e} At equilibrium, $F = 0$ and therefore $\chi_* = (v_r/v_d) c_*$ or $J_* = (v_r/v_d)$.
$F = v_d \chi_i - \Lambda G$	Λ is a resuspension rate; $\Lambda \approx v_r/\delta$.
$F = \Omega A - \Lambda G$	Ω is a similar deposition rate, where A is the average atmospheric concentration of the pollution. For example, if h_d is the average height to which the pollution has mixed, then $A \approx h_d \chi_i$ and $\Omega = v_d/h_d$.

performed (Markee, et al.²⁴) which illustrate that changes in v_d of factors of ten, per se, only change deposition by a factor of 2 to 3 with the model used by Markee. This reduction is due to the compensatory effects on the airborne concentrations of large v_d and then K_z by limiting the downward flux. For a specific release and specific meteorological conditions, transfer may be rate-limited elsewhere in the atmosphere, rather than in the constant flux layer. Possible corrections should also be considered for the attachment of molecular iodine to aerosol particles, which causes subsequent reduction in the effective deposition velocity of the iodine.

More recent results, both experimental and theoretical, can be used to describe the transfer of gases and particles through the atmosphere and past the atmosphere/surface interface. Data for a variety of gases depositing on vegetation can be interpreted in terms of gas solubilities and diffusivities in vegetation.^{25,26} Alternatively, the resistance model can be used to describe the flux of gases to vegetation²⁷ or to water bodies.^{28,29} For dry deposition of particles substantial amounts of new data are available,^{30,31,32} and considerable progress has been made in interpreting the data.^{29,33,34,35} However, more research is definitely needed because uncertainties of about a factor of 10 remain for the dry deposition of submicron particles. This type of deposition depends on particle size and canopy characteristics. Similarly, although there have been both experimental^{36,37} and theoretical^{38,39,40} progress in the study of particle resuspension, there remain uncertainties of many orders of magnitude.

Wet Removal

To describe wet removal processes, the NRC Guides^{1,2} recommend methods described by Engelmann.²³ However, more recent results have shown clearly that there is no major difference between below-cloud and in-cloud scavenging; in particular, the data of Radke, et al.⁴¹ show the necessity for accounting for particle growth by water vapor condensation beneath clouds. Simpler formulae to describe wet removal of particles have been developed⁴² and inadequacies of older methods

for describing wet removal of gases have been shown both experimentally⁴³ and theoretically.^{44,45} Further, a wealth of new data for washout ratios has been obtained^{46,47} and can be used to define a wet deposition velocity for both particles and gases.⁴⁸ This wet deposition velocity, v_w , was introduced by Slinn as follows:

$$v_w = \frac{\omega}{\chi_o} = \frac{\kappa_o p}{\chi_o} = w_r p \quad (4)$$

where ω = wet flux (units/m²-sec)

χ_o = (near) surface level air concentration (units/m³)

κ_o = surface level concentration of the pollutant in the precipitation (units/m³)

$w_r = (\kappa/\chi)_o$ = washout ratio, dimensionless

For example, if $w_r = 0.3 \times 10^6$ and $p = 100$ cm/yr then $v_w \approx 1$ cm/sec. This typical, annual average value for v_w for aerosol particles supports the result of NRC's suggestion to ignore wet removal of particles and to use a dry deposition velocity of about 1 cm/sec. However, it is only the *result* of their suggestion that is supported, not the rationale leading to the result. It would be more appropriate to use best available data for v_d for particles,^{30,32} and then $v_w = w_r p$ with best available data^{46,47} for the washout ratios. Notice that for considerations of the wet deposition from a single event, it is necessary to use the removal rate approach if a vertical distribution of the pollutants is involved.

Accuracy Estimates

In a recent survey paper,⁴⁸ accuracy estimates for state-of-the-science parameterizations for resuspension and for wet and dry removal were presented. For annual-average predictions, the accuracies were estimated to be as follows: (i) a factor of 2 to 5 for v_w given p ; (ii) a similar factor for v_d for reactive or highly soluble gases (e.g., HTO₂, SO₂, and I₂), worse than a factor of ten for slightly soluble gases and submicron particles; and (iii) many orders-of-magnitude uncertainty for the resuspension velocity. For the accident case, parameterizations for wet and dry removal would have accuracies

similar to the long-term average estimates if the trajectory of the pollutant could be well specified. However, as this is not presently feasible, order-of-magnitude poorer descriptions of the removal processes can be expected for the accident case than for routine releases. These estimates of the accuracies reflect current capabilities; the reliabilities of the older methods used in the NRC's Guides^{1,2} are not as good. The description of the dry removal of reactive gases (such as I₂) has improved only slightly, but improvements in the descriptions of the wet and dry removal of particles and all other gases have improved substantially. These improvements should be incorporated into the NRC Guides if they are to be used for materials other than molecular iodine.

The deposition and depletion curves in the NRC Guides are from one particular diffusion calculational model. NRC permits several different diffusion models for air concentration calculations. For consistency, it is desirable that the same model be used for all types of calculations.

Recommendations

Research is needed to improve these parameterizations and to obtain more reliable estimates of accuracies. There are order-of-magnitude unexplained differences among different experimental results for the rain scavenging of submicron particles. There are almost no data available for snow scavenging of particles, uncontaminated by simultaneous dry deposition. Particle modifications and gas reactions and attachment to particles in the atmosphere need substantial further study. Realistic descriptions of the properties of particles and cases at release are also needed. Dry deposition of submicron particles and slightly reactive

gases to vegetative canopies and water bodies needs much further study. Resuspension velocities or rates as functions of time since deposition, and of soil and meteorological conditions are needed to reduce uncertainties in parameterizations. Thus, in summary, and repeating recommendations stated in other recent workshops reports,^{29,49,50,51} more data are needed to test existing models and, almost certainly, improved models must be developed to couple removal and resuspension to diffusion. The practical needs of the user and the availability of local input data to the models must be kept in mind during the research program.

GAMMA DOSE

The application of atmospheric transport calculations to the evaluation of external gamma-ray dose near nuclear facilities was discussed briefly. Because of the relatively long mean-free-path of gamma rays in air, it is possible under some dispersion conditions to have low surface air concentrations under an elevated plume, and yet still have a significant gamma-ray dose. Thus, it is important to consider the entire plume geometry [i.e., effective plume height (h), σ_y , and σ_z] as well as the surface airborne concentration even when sector-averaged concentrations are used at distances less than 5 km downwind. Furthermore, the dose contribution to sectors adjacent to the downwind sector should be included, especially near the point of release. Current NRC practices do include consideration of the geometry of finite plumes, but do not include the contributions of adjacent sectors in dose calculations employing sector-averaged plume concentrations.

Studies, in which measured long-term average external gamma-ray exposures within 10 km of nuclear facilities have been compared to calculations using Gaussian plume models, have usually indicated agreement to within about a factor of two.^{52,53,54,55,56,57}

REGIONAL AND CONTINENTAL MODELS

The NRC guidelines (Reg. Guide 1.111)² for routine releases permit the use of both conventional Gaussian plume models and several alternative kinds of atmospheric transport and diffusion models, particularly for

situations such as multi-day long-range transport for which straight-line, steady-state, Gaussian-plume models should not be expected to apply. Any of several categories of such models are acceptable, and indeed may be preferable to the Gaussian model forms, subject to the NRC's regulatory position that "the preferred model is that which best simulates atmospheric transport and diffusion in the region of interest ... considering the meteorological characteristics of the region, the topography, the characteristics of the ... source and ... receptor, the availability and representativeness of input data," etc.

Particle-in-Cell

Particle-in-cell (PIC) models simulate the atmospheric dispersion by the calculation of trajectories of many particles emitted as a function of time from a particular point source (see Sklarew⁵⁸). The concentration is determined by counting the number of particles per unit volume. The advantage of such a particle and cell method is that complex terrain can be treated explicitly; there is no numerical pseudo-diffusion associated with the calculations, although statistical diffusion may be appreciable. PIC models have the disadvantages of requiring a large specialized computer (not available to most potential users) and large amounts of computer time (making these models impractical for industrial use). Quite complex situations can be treated, particularly with the ADPIC model developed by the Lawrence Livermore Laboratory (Lang⁵⁹).

Trajectory

Another class of models is the trajectory models, where time- and space-dependent wind fields are used to calculate trajectories for either puffs or plume segments. These codes are exemplified by the work of Heffter⁶⁰ and Wendell, et al.⁶¹ For short-range, short-term calculations, these models may use on-site or near-site hourly meteorological data (e.g., the MESODIF model⁶²); for long-range and for long-term computations, they may use routinely available NWS 12-hourly upper-air data. Some models assume uniform concentration through the depth of the mixed layer; other models explicitly represent the vertical concentration profile as a solution to the vertical diffusion equation. Trajectory models

are most practical for small numbers of point sources. Their storage and execution times grow linearly with the number of sources and can easily become unmanageable; therefore, these models may be impractical for area-type sources.

Grid (Eulerian)

A third class is the grid or Eulerian models, where numerical solutions are obtained to the advection-diffusion equation on a grid network. These models have the advantage of being able to handle many sources simultaneously, with little or no cost penalty for multiple sources, but have the disadvantage of inadequate near-field representation of point sources. Computer storage, execution time, and numerical diffusion problems are also typical disadvantages of some of these models, although special approaches are available to minimize numerical diffusion. Grid models allow the treatment of time- and space-dependent meteorology, transport over distances greater than those for which the Gaussian plume model is applicable, and permit the consideration of a variety of complex meteorological or topographical situations, removal processes, etc., for which the Gaussian plume equation would not be expected to apply.

Accuracy Estimates

In general, there is as yet insufficient validation for these three classes of models to permit useful general conclusions about their accuracy ranges for different averaging times, spatial scales, characteristic meteorological regions, etc. At one extreme, preliminary assessment of unpublished radionuclide transport data around the Savannah River Plant suggests, for example, that the annual average accuracy of some of these models is of the order of a factor of ± 4 in relatively flat terrain at distances of the order of 100 km.

These preliminary comparisons indicate about the same accuracy as obtained with the simple wind-rose models. More complex models may reduce the scatter and out-perform the wind-rose models when final comparisons are done using time-dependent Q (on an hourly basis) instead of assuming a uniform rate for the month. Some recent comparisons of regional-scale trajectory and grid models with observational data for

sulfur dioxide transport on regional scales are encouraging. Although the data bases are very sparse, they do suggest that general agreement within a factor of two is possible at much longer ranges (500 to 1000 km) and for much shorter averaging times, as little as 24 hours (see, for example, Rao, et al.,⁶³ and Hildy, et al.⁶⁴). Certainly, considerably more research and validation work with these models is needed to clarify their achievable accuracy ranges and the limitations upon their use for regional scale problems before they can be considered as "routinely operational."

Recommendations

The NRC should continue to leave open the option to use these alternatives to the conventional straight-line Gaussian plume model, if justified by further validation experience and by the complexity of the application situation. Wide industrial use of the more complex of these codes is not presently feasible due to the non-user-oriented research nature, elaborate data needs, expert "babysitting" required, and the very large or unusual computational resource requirements of these codes. However, some of the simpler trajectory-type codes (e.g., MESODIF) are quite practical; indeed they are presently being used both by the NRC and by nuclear electric utilities for routine environmental impact assessment purposes. For such assessment, NRC may often require the use of hourly, on-site, multilevel-tower, high-capture-rate meteorological data which are not available from the NWS. Therefore, the routine operation of such a model at a central "service bureau" is probably impractical at present. Furthermore, for assessment purposes, even these simpler models may require a certain amount of modification for local or site-specific features. They certainly will require skilled interpretation in light of the characteristic local meteorology, which argues against "clearing house" routine operation.

To bring some of these codes to more routine operational status, additional development is needed. All of these codes have to make some assumptions either on the growth of the puff or the segmented plume as a function of time, or on the specification of the horizontal and vertical turbulent exchange coefficients. The basis for these specifications

needs to be improved, particularly for long travel distances. Consistent methods must be developed for obtaining input wind field and diffusivity data. This is very important in the multi-level Eulerian codes and the Particle-in-Cell codes. The laws of mass conservation should not be violated in the input wind fields which are used in these codes. Some Eulerian codes need further improvement to minimize the numerical errors associated with solutions to the advection-diffusion equation and to be able to represent point sources adequately. Most of these codes have large computational requirements. Techniques to reduce computer requirements for production versions of the codes need to be examined. Lastly, but not least, these codes need to be validated against a statistically significant amount of measured concentration data.

GENERAL CONCLUSIONS

The NRC Guides were written specifically as guidelines for safety and environmental assessments associated with power reactors. Even within these constraints, specific methods must be selected to fit specific situations. All applications of the models should be physically consistent.

In all cases, additional validation against field data is needed. Our working group suspects that a large amount of currently available data have not been analyzed adequately. But, even with thorough analysis of current data, some additional carefully selected experiments should be done. It is recommended that the NRC Guides should be kept flexible and adaptable; however, these Guides should be subject to change as new methods or additional data justify. In all cases, atmospheric scientists need to develop confidence limits on the calculations. These confidence limits should be based on adequate data bases, not just on scientific judgment as was largely the case in this summary. To use these confidence limits, regulatory agencies should be willing to accept probability statements on the calculations. With a probability statement associated with each environmental pathway calculation, probability statements associated with the final dose-to-man calculation can be

developed and critical pathways identified to show where new information is needed for significantly improving the confidence in the dose-to-man calculation. This latter comment is applicable to all of the pathways, not just to the atmospheric pathway.

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