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Improved Formulations for Air-Surface Exchanges Related to National Security Needs: Dry Deposition Models

J. G. Droppo

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Summary

The Department of Homeland Security and others rely on results from atmospheric dispersion models for threat evaluation, event management, and post-event analyses. The ability to simulate dry deposition rates is a crucial part of our emergency preparedness capabilities. Deposited materials pose potential hazards from radioactive shine, inhalation, and ingestion pathways. A reliable characterization of these potential exposures is critical for management and mitigation of these hazards.

A review was conducted of the current status of dry deposition formulations used in these atmospheric dispersion models. The formulations for dry deposition of particulate materials considered an event such as a radiological attack involving a Radiological Detonation Device (RDD). The results of this effort are applicable to current emergency preparedness capabilities, such as are deployed in the Interagency Modeling and Atmospheric Assessment Center (IMAAC), other similar national/regional emergency response systems, and stand-alone emergency response models.

The review concludes that dry deposition formulations need to consider the full range of particle sizes, including: 1) the accumulation mode range (0.1 to 1 micron in diameter) and its minimum deposition velocity, 2) smaller particles (less than 0.01 micron diameter) deposited mainly by molecular diffusion, 3) 10 to 50 micron diameter particles deposited mainly by impaction and gravitational settling, and 4) larger particles (greater than 100 micron diameter) deposited mainly by gravitational settling. The effects of the local turbulence intensity, particle characteristics, and surface element properties must also be addressed in the formulations.

Specific improvements recommended for dry deposition formulations are 1) the capability of simulating near-field dry deposition patterns, 2) the capability of addressing the full range of potential particle properties, 3) the incorporation of particle surface retention/rebound processes, and 4) the development of dry deposition formulations applicable to urban areas. Also, to improve dry deposition modeling capabilities, atmospheric dispersion models in which the dry deposition formulations are imbedded need better source-term plume initialization and improved in-plume treatment of particle growth processes.

Dry deposition formulations used in current models are largely inapplicable to the complex urban environment. An improved capability is urgently needed to provide surface-specific information to assess local-exposure hazard levels in both urban and non-urban areas on roads, buildings, crops, rivers, etc.

A model improvement plan is developed with a near-term and far-term component. Despite some conceptual limitations, the current formulations for particle deposition based on a resistance approach have proven to be reasonable dry deposition simulations. For many models with inadequate dry deposition formulations, adding or improving a resistance approach will be the desirable near-term update. Resistance models, however, are inapplicable to aerodynamically very rough surfaces, such as are found in urban areas. In the longer term, an improved parameterization of dry deposition needs to be developed that will be applicable to all surfaces, but particularly to surfaces found in urban environments.

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Acronyms

ADAPT	Atmospheric Data Assimilation and Parameterization Tool
AGDISP	AGricultural DISPersal
CFD	computational fluid dynamics
DERMA	Danish Emergency Response Model of the Atmosphere
DHS	U.S. Department of Homeland Security
EPA	U.S. Environmental Protection Agency
GIS	geographical information system
ISC	Industrial Source Complex Model
JEM	Joint Effects Model
LLNL	Lawrence Livermore National Laboratory
LODI	Lagrangian Operational Dispersion Integrator
MOBLAM	Molecular Oceanic Boundary LAyer Model
NARAC	National Atmospheric Release Advisory Center
PM	particulate matter
RASCAL	Radiological Assessment System for Consequence Analysis
RDD	Radiological Detonation Device

Definitions

Parameter	Definition	Typical Units
B	Sublayer Stanton number	dimensionless
C	Air concentration at some designated location	g m^{-1}
C_0	Air concentration at the surface	g m^{-3}
C_1	Air concentration at the top of the surface quasi-laminar surface layer	g m^{-3}
C_2	Air concentration at the top of the turbulent surface layer	g m^{-3}
c_d	Dimensionless drag coefficient for the canopy	dimensionless
C_i	Efficiency of impaction	dimensionless
d	Aerodynamic (equivalent) diameter - diameter of a unit-density sphere having the same gravitational settling velocity as the particle in question. Aerodynamic diameter takes into account the shape, roughness, and aerodynamic drag of the particle.	cm, µm
D	Brownian or molecular diffusivity	cm^2 s^{-1} or m^2 s^{-1}
E_B	Efficiency of Brownian motion	dimensionless
E_{IM}	Efficiency of impaction	dimensionless
E_{IN}	Efficiency of interception	dimensionless
F	Total contaminant flux	g m^{-2} s^{-1}
F_g	Gravitational dry deposition flux	g m^{-2} s^{-1}
g	Acceleration due to gravity	cm s^{-2} or m s^{-2}
$h(d, x_o)$	Initial plume height	m
h(t)	Plume height as a function of time	m
k	von Karman constant (0.4)	dimensionless
K	Turbulent diffusivity	m^2 h^{-2}
Ko	Average turbulent diffusivity in the canopy	m^2 h^{-2}
L	Monin-Obukhov length	m
L_c	Characteristic length	m
M_H	Density of herbage	kg dry wt per m^2
P	Atmospheric pressure	cm Hg
p	Fraction of radioactive activity retained in foliage	dimensionless
r	Particle radius	cm, µm
r_a	Atmospheric resistance	s m^{-1}
r_b	Resistance across quasi-laminar sublayer	s m^{-1}
r_c	Surface retention resistance	s m^{-1}
Re	Reynolds number	dimensionless
Re*	Roughness Reynolds number	dimensionless
R_R	Rebound fraction (i.e., stickiness factor)	dimensionless
r_s	Total surface resistance through a deposition layer, including flux through the quasi-laminar sublayer and surface retention processes	s m^{-1}
r_t	Total resistance to dry deposition	s m^{-1}
Sc	Schmidt number	dimensionless
S_f	Cunningham correction factor or slip correction factor	dimensionless
S_p	Stopping distance - product of relaxation time and the initial particle velocity; an indicator of a particle's ability to adjust to directional changes in aerosol flow	m
St	Stokes' number - ratio of a particle's stopping distance to a characteristic dimension; generally used as an indicator of similitude in particle behavior in a given aerosol flow configuration	dimensionless
t	Plume travel time	s

Parameter	**Definition**	**Typical Units**
Ta	Air temperature	degrees Kelvin
u	Wind speed	$m\ s^{-1}$
u_*	Friction velocity	$m\ s^{-1}$
u, v, w	Air velocity components	$m\ s^{-1}$
u_1	Free air stream velocity	$m\ s^{-1}$
u_z	Wind speed at height z	$m\ s^{-1}$
u_o	Characteristic wind speed in the canopy	$m\ s^{-1}$
u_p, v_p, w_p	Particle velocity components	$m\ s^{-1}$
V	Relative velocity of particles	$m\ s^{-1}$
v_d	Deposition velocity	$cm\ s^{-1}$ or $m\ s^{-1}$
$(v_d)_b$	Local deposition velocity to a specific surface	$cm\ s^{-1}$ or $m\ s^{-1}$
v_i	Inertial velocity	$m\ s^{-1}$
v_{impact}	Particle velocity at impact	$m\ s^{-1}$
$v_s\ ,v_g$	Settling velocity for particles	$cm\ s^{-1}$ or $m\ s^{-1}$
V_{sm}	Transfer velocity for submicron particles	$m\ s^{-1}$
z	Height over surface	m
z_d	Deposition reference height	m
z_{hl}	Local reference height defined as normal to the receptor surface	m
z_o	Surface roughness length	m
z_{ol}	Roughness length of the local surface element	m
z_v	Roughness length for mass transfer	m
α	Collection area per unit volume of the canopy	m^{-1}
β	Buoyancy effect parameter	dimensionless
γ_v	Particle impaction parameter	dimensionless
λ	Mean free path of air molecules (= 5.53 10^{-5} m)	m
μ	Dynamic viscosity of air	$kg\ m^{-1}\ s^{-1}$ $g\ cm^{-1}\ s^{-1}$
ξ_c	Dimensionless particle collection efficiency for cylinders	dimensionless
ρ	Particle density	$g\ cm^{-3}$
ρ_a	Air density	$g\ cm^{-3}$
ρ_p	Particle density	$g\ cm^{-3}$
υ	Kinematic viscosity of air	$cm^2\ s^{-1}$ or $m^2\ s^{-1}$
Γ	Interception coefficient	$kg\ m^{-2}$

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1.0 Introduction

The Department of Homeland Security (DHS) and others rely on results from atmospheric dispersion models for threat evaluation, event management, and post-event analyses. As such, these models are a crucial part of our emergency preparedness capabilities. Efforts are underway to improve the formulations in these models; the focus of this report is to review and recommend improvements in dry deposition formulations, with specific emphasis on urban environments.

A review of the current status of atmospheric dispersion model capabilities for homeland security applications by the National Research Council (2003) considered the status of modeling capabilities relative to potential chemical/biological/nuclear applications. The report emphasizes the challenges facing effective use of such models and, in particular, the meteorological observations to support such models. For homeland security applications, the National Research Council publication is an important resource to understand current atmospheric dispersion modeling capabilities and needs.

Formulations for deposition processes are considered in this report. It is critical to be able to model the progression of deposition associated with an event because hazards are sequentially created as the transported material deposits to local surfaces. The deposited materials pose potential hazards from direct exposure to radioactive shine, inhalation and redistribution from resuspension, ingestion from dermal contact or contaminated food materials, and other indirect exposure routes. A reliable characterization of these potential exposures is critical for management and mitigation of these hazards.

The objective of this review is to identify areas for improvement of the DHS's capabilities in the area of atmospheric–surface deposition and resuspension formulations for radioactively contaminated particulate matter. Specifically, the review considers emissions from an "event" based on a radiological attack involving a Radiological Detonation Device (RDD). The results of this effort will be used to update current emergency preparedness capabilities such as are deployed in the Interagency Modeling and Atmospheric Assessment Center, other similar national/regional emergency response systems, and standalone emergency response models. While focusing on improving dry deposition formulations, the results of this effort have wide applicably for improving our homeland security capabilities.

The capability to model processes in an urban environment is of special interest for homeland security needs. For urban areas, it is important that the wide variation expected for deposition rates be accounted for when considering potential exposures. Such variations were observed in exposure levels in different areas of the city of Kiev after the passage of the Chernobyl plume.[1] Models should be able to simulate these variations and indicate relative threats posed by operations in the urban environment. Of concern are resuspension exposures from remediation activities as well as day-to-day exposures from outdoor recreational activities in areas such as parks and other walking/cycling pathways and vehicle transportation routes.

The capability of reliably simulating the atmospheric source, dispersion, and deposition processes is critical for threat evaluation, event management, and post-event analyses. A major capability gap in

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[1] Personal Communication, Vitaly Eremenko, November 3, 2005. The observations of spatial variability are based on total activity measurements made with a personal Geiger counter in different areas of Kiev after the passage of the plume from Chernobyl (Eremenko and Droppo 2006).

current dry deposition formulations is the ability to account for the combined effects of local meteorological conditions, surface properties, and aerosol properties. Dry deposition formulations used in current models are based on assumptions that make them largely inapplicable to processes that occur in the complex urban environment. The improved capability is urgently needed to address urban environment applications. This improved capability will address the inability of most current models to provide surface-specific information to assess local exposure hazard levels in both urban and non-urban areas on roads, buildings, crops, rivers, etc.

1.1 Event Progression

Atmospheric exposures occur both as the result of airborne concentrations and deposited materials. Major atmospheric exposure routes include inhalation and shine from the primary airborne contamination, as well as subsequent ingestion, inhalation, and shine from deposited materials.

Figure 1.1 illustrates the sequences of major processes that determine the environmental fate of particulate contaminants in an airborne plume. The ability of reliably simulating the processes shown in Figure 1.1 is thus critical for threat evaluation, event management, and post-event analyses. To evaluate potential consequences of an event, models need to address the release, dispersion, and deposition processes. Although an urban setting is shown as an application of special interest, the same processes apply to all types of surface cover.

The progression of processes starts with a primary *source* of contaminated airborne material from an *RDD event* involving an explosion or secondary dispersal mechanism such as a fire source. To initialize the atmospheric dispersion computation, inputs that define the initial characteristics of the plume source are required. Generally, atmospheric dispersion models do not explicitly treat the circulations and processes within the initial volume of the explosion, but rather depend on a combination of input data and source parameterizations.

The deposition rate of a material is highly dependent on its form, concentration, and size distribution in the air over the receptor surface. The *atmospheric dispersion* processes in Figure 1.1 move, dilute, and change the airborne plume. In-plume processes such as radioactive decay and chemical reaction act as a source for some contaminants and as a sink for others. To effectively model dry deposition rates, in-plume processes such as gas-particle partitioning, coagulation, and evaporation/condensation also need to be considered. Jacobson (1997) addresses modeling constraints for developing an aerosol simulation formulation that can handle these processes. The progressive process of in-plume particle coagulation will tend to increase deposition rates of contamination.

As the prevailing winds take a plume over surfaces, those surfaces act as *sinks* for the airborne material. The materials deposited to soils, roads, cars, vegetation, buildings, etc., reduce the airborne concentrations. However, the resulting surface contamination creates residual threats from radiation shine, direct contact, and resuspension (shown as secondary airborne sources in Figure 1.1) after the primary airborne plume has moved downwind. These potential hazards need to be addressed in terms of both immediate and long-term hazards.

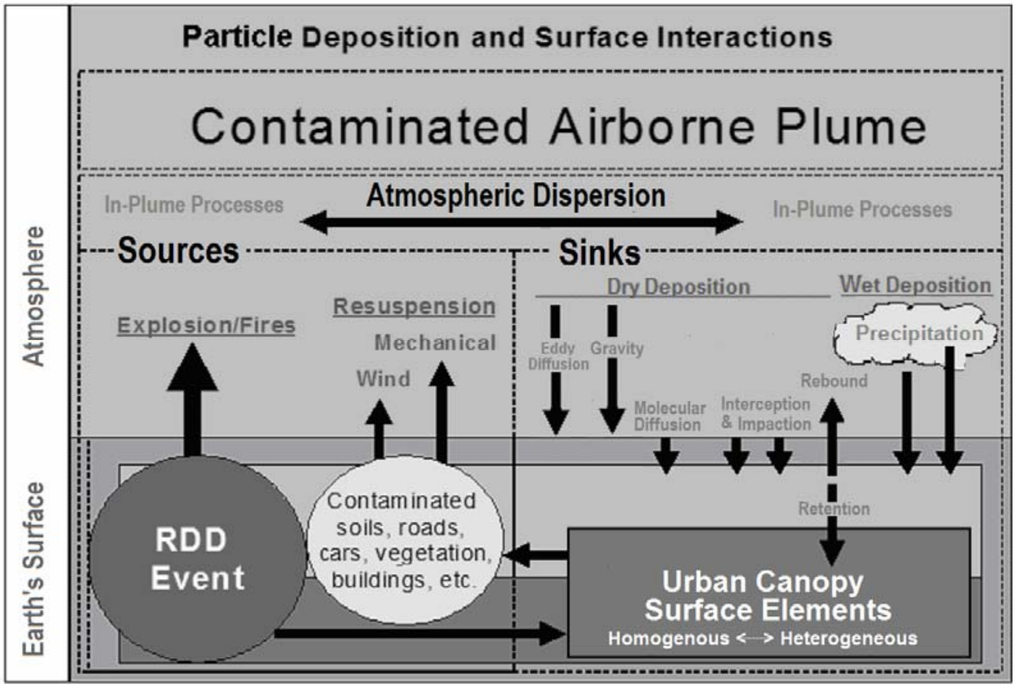

Figure 1.1. Events and Processes for Evaluating Potential Hazards Involving an Airborne Release from an RDD Type of Event

1.2 Surface Contamination

The contamination of earth's surface by airborne material occurs by processes broadly grouped as *dry deposition* or *wet deposition*. Dry deposition, or the flux of an airborne contaminant directly from the air to the surface, occurs by concurrent diffusion, gravitational settling, interception and impaction, and retention/rebound processes. Eddy diffusion and gravity move the particles near to surfaces; molecular diffusion, interception, and impaction move the particles to the surface; and retention/rebound determines if they stay on the surface. The formulations for dry deposition processes are typically coupled to the *atmospheric dispersion* models. The discussion below describes these dry deposition processes in more detail.

Wet deposition (the delivery of contaminants by water droplets or ice particles to local surfaces) occurs as the result of in-cloud or below-cloud scavenging. In contrast to dry deposition, which occurs when particles are impacted directly on local surfaces, wet deposition occurs when water droplets or ice particles carry scavenged particles to local surfaces. When wet deposition of particles occurs, the transfer of material to the surface can be much higher than for dry deposition, depending on parameters such as rain rate and particle size.

This report will show that a major deficiency in current atmospheric dispersion models is their inability to account for the combined effects of local meteorological conditions, surface characteristics, and aerosol properties in complicated environments. Dry deposition formulations used in current models assume idealized conditions, and the application of such formulations to simulate the complex urban environment is questionable. As a result, an improved capability is urgently needed for effective operational response to RDD events or other releases of hazardous materials in urban areas.

2.0 Atmospheric Modeling

The atmospheric pathway simulation of an event (e.g., radiological attack with an RDD) requires models that start with the initial characteristics of the resultant airborne plume and simulate the subsequent downwind dispersion and removal processes for a wide range of possible environments. Current deposition models have been developed largely for non-urban environments, making urban environments an application that needs improvement.

Modeling dry deposition in an urban environment has special challenges. To address hazards from dry deposition in such a complex environment, the model needs to be able to account for deposition to heterogeneous mixes of surface elements, including buildings, roadways, vehicles, sidewalks, parks, grass, trees, lakes, rivers, and agricultural surfaces. Because dry deposition rates will vary with the surface properties, each surface element potentially can have a different deposition rate.

Another key requirement for dry deposition models is their ability to handle the potential range of particle sizes as well as the amount of contamination level on the particles. Although the literature suggests possible distributions, there is a wide range of possible characteristics for an RDD device that will result in widely different size distributions. This wide range of possible distributions requires that the effects of the full range of possible particle sizes be considered in order to be certain that the size distribution generated in an event can be effectively modeled.

Atmospheric dispersion models simulate the dispersion of pollutants in a frame of reference fixed either to the earth (Eulerian) or in a reference system that moves with the parcel's trajectory (Lagrangian). The distribution of airborne material is treated in a number of different ways: Gaussian plume models assume that plumes are continuous with some defined duration, puff models use a series of instantaneous puffs distributed over the release period, and stochastic models track the movement of particles.[2] Models typically include formulations for defining the tendency for the plume to rise or fall (plume rise), for defining flow streamlines (incorporating local wind fields, topography, and other influences), and for accounting for vertical structure (typically winds and temperature) and for dispersion rates (normally schemes for defining vertical, lateral, and transverse rates). Despite major differences in their other formulations, air dispersion models tend to use very similar parameterizations for dry deposition.

A number of atmospheric models are deployed for emergency response applications. For national responses in the United States, the National Atmospheric Release Advisory Center (NARAC)[3], the interim Interagency Modeling and Atmospheric Assessment Center, is located at Lawrence Livermore National Laboratory (LLNL). NARAC uses the Atmospheric Data Assimilation and Parameterization Tool/Lagrangian Operational Dispersion Integrator (ADAPT/LODI) codes (LLNL 2005). Emergency response models have been developed for specific civilian and military applications. In addition, many major facilities have custom implementations of these or similar models, as part of their local emergency

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[2] Subsequent discussions use the term "plume" to generically refer to the airborne material, whether it is continuous plume, discrete puff, particle cluster, etc.

[3] Based on the former Lawrence Livermore's Atmospheric Release Advisory Capability, the MATTHEW-ADPIC models were originally designed for nuclear facilities.

response capabilities.[4] Models such as the Danish Emergency Response Model of the Atmosphere (DERMA) (Baklanov et al. 2006) with similar capabilities have been developed by other countries. The Radiological Assessment System for Consequence Analysis (RASCAL) model was developed by the U.S. Nuclear Regulatory Commission for assessing potential accidents from commercial nuclear power plants and associated facilities in the United States (Sjoreen et al. 2001).

Atmospheric dispersion models for chemical, biological, and nuclear applications include HYSPLIT (Draxler 2004) and CAMEO/ALOHA from National Oceanic and Atmospheric Administration, NARAC from DOE/LLNL, HPAC from the Defense Threat Reduction Agency, VLSTRACK from the Navy, MIDAS-AT from the Marines, the Joint Effects Model (JEM), and the CATS-JACE model being developed by many agencies (National Research Council 2003). Atmospheric dispersion models also of interest in this review of dry deposition formulations include a series of models promulgated by the U.S. Environmental Protection Agency (EPA) for air-quality assessments and long-range transport models developed for acid-rain applications (USEPA 2005b).

High-resolution computational fluid dynamics (CFD) models based on numerical flow simulation methods are used for studying air circulation in urban environments (Lien, Yee, and Cheng 2004; Hamlyn and Britter 2005; Kim and Baik 2004). Their fine-scale simulation resolutions (on the order of 1 m) combined with their ability to simulate large and medium-size eddies make them ideal for simulating flow around urban structures. This class of models is showing promise for urban dispersion applications. Although the setup and input requirements for these computationally intense models may preclude using them as operational tools, their ability to provide simulation, visualization, and analysis of fluid flow in urban environments has a strong potential for use in developing improved parameterizations for operational models.

2.1 Dry Deposition Models

The earth's surface is the interface between the atmosphere and earth – and as such it is the surface through which all energy and mass exchanges occur. The development of models for the deposition of material from the air to the earth's surface has relied heavily on historical studies of momentum, heat, moisture, trace metal, and carbon dioxide surface fluxes. Most current parameterizations for particle deposition include an assumption of analogous particle and mass or momentum transport. At the atmospheric surface-air interface, the formulations and methodologies directly correspond to those considered in filtration theory and practice, such as those detailed by Willeke and Baron (2005).

The current review builds on several previous reviews of dry deposition models. In the early 1990s, an extensive review of dry deposition models was conducted by the EPA (USEPA 1994). That review evaluated model performance based on field measurements for ideal conditions. They provided a basis for formulating a new dry deposition model that is documented in the second volume of the user's guide for the Industrial Source Complex Model (ISC) (USEPA 1995). A review of dry (and wet) deposition computational methods was conducted for radioactively contaminated particles (in the range 0.1 to 10 micron) by the Atmospheric Dispersion Modeling Liaison Committee (NRPB 2001). They stress the importance of combining sound data with an understanding of the underlying processes. They recom-

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[4] Examples of models developed and/or used for local emergency preparedness are APGEMS for Hanford, Washington, operations and Atmospheric Release Advisory Capability, HOTSPOT, CHARM, and EPICODE at Y-12 for Oak Ridge, Tennessee, operations.

mend values and methods for estimating deposition rates and recommend special parameter limits for extrapolation of the dry deposition model to an urban environment. However, neither of these reviews addressed the issues of the applicability of the dry deposition models to non-ideal conditions such as the aerodynamically very rough surfaces encountered in an urban environment.

The aftermath of the Chernobyl accident has provided information on where airborne material deposits and what exposures occur. Anderson and Roed (2006) prepared estimates of doses received from dry-deposited radionuclides from streets, roofs, exterior walls, and landscapes in a residential area in Bryansk, Russia. Ramzaev et al. (2006) considered the contamination of a broader range of surfaces (structures, pastures, gardens, forests, roads, grass, etc.).

The challenges facing the modeling of dry deposition are highly dependent on the spatial scale being modeled. Three sets of application-specific requirements for dry deposition models are considered: near-field, local-region, and long-range. Near-field refers to the locations in the immediate vicinity of the release where peak air and surface exposures normally occur. Local-region refers to affected areas in approximately the first 50-100 miles downwind. The near-field and local-region correspond to the scale normally considered for industrial stack emission impacts. Long-range refers to affected areas beyond the local-region and corresponds to the scale for acid-deposition modeling.

Near-field models need to have relatively fine spatial and temporal scales, incorporate local plume characteristics and influences, and be able to treat a wide range of particle sizes. Detailed near-field predictions of detailed deposition patterns are very difficult to model, given the normally large uncertainties in plume initialization combined with the highly stochastic nature of the initial plume dispersion. The highest activity levels will be in the immediate vicinity for a near-surface plume and at some distance downwind from the source for an elevated plume.

The patterns of deposition as a function of distance will be highly dependent on the particle size distribution. Near-field deposition from an event is normally dominated by larger particles. Models for near-field deposition thus mainly consider gravitational and wind influences. The near-field formulations often do not account for deposition of smaller particles because most of the deposited mass in this range will be associated with larger particles. The larger particles with settling velocities greater than 100 cm s^{-1} will not be effectively transported by atmospheric turbulence or the mean wind speed. Deposition rates are typically based on a ballistics model using wind speed and the gravitational settling velocity to determine at what distances the particles will fall to the surface (Hanna, Briggs, and Hosker 1982).

An understanding of near-field deposition processes is very important in agricultural spraying. The aerial spraying of pesticides depends on deposition processes to insure high rates of application to crops. The material that is not deposited and is carried away from the crops by the prevailing winds is referred to as drift and is of concern in assessment of potential exposure risks (Hewitt et al. 2002). Hewitt (2001) reviewed the literature on drift filtration by natural and artificial collectors and concluded that a drift reduction of 45 to 90% can be achieved using deposition processes on appropriate barriers. The AGricultural DISPersal (AGDISP) near-field spray model addresses the details of release and source processes to simulate potential downwind drift concentrations and deposition rates.

Local-region plume models consider the dry removal of material being transported in a wind-driven trajectory over the terrain in the local region. In this portion of the plume, the peak-to-mean concentration ratios will be much smaller than closer to the source, which will result in more uniform patterns of deposition. In the case of an elevated plume intersecting an elevated local terrain feature, relatively high levels of deposition can occur at some distance from the release. Operational emergency response models typically address this scale of potential impacts.

Most currently deployed atmospheric dispersion models provide near-field concentration and deposition results but only at distances greater than 100 m downwind. The practical limitation is related to the model's ability to adequately define the effects of the source configuration on the initial plume dilution at very close distances. An example of a model that does provide concentration and deposition simulations at closer scales is the air-quality assessment model EPA ISC-PRIME (USEPA 2000), which has routines for simulating building wake effects.

The trajectories of plumes starting with a large fraction of larger particles are modeled as a "tilted plume" model, where particles in designated size ranges fall at representative settling velocities (Hanna, Briggs, and Hosker 1982). As a plume moves downwind, the larger particles will fall to lower portions of the plume. The relatively rapid rates of deposition for larger particles at closer distances will result in a shift to a distribution with higher fractions of smaller particles at extended distances. The tilted plume approach is correct only for larger particles, for which the effects of atmospheric turbulence are relatively small. Intermediate-sized particles in a plume will also settle, but they are carried upward more readily by turbulence. As a result, their rate of transfer to the surface is reduced. Settling is an insignificant effect for small particles. Their rate of deposition is determined by a combination of the effects of turbulent dispersion and the near-surface loss mechanisms.

Long-range transport models extend the dispersion and deposition simulation from the immediate region out to surrounding regions. At regional and long-range distances, one would expect most of the larger particles to have settled out or to have been lost by impaction. For these distances, the formulations for the smaller particle sizes become central to modeling deposition rates. There is some evidence, however, that large particles may contribute to deposition even at long ranges from the plume origin. In a study that noted the failure of eddy correlation methods to fully account for deposition fluxes, Wesely and Hicks (2000) suggested that particle coagulation may be creating new large particles that become important for deposition during long-range transport.

3.0 Dry Deposition

The processes that control the transfer, or flux, of airborne material to local surfaces are of interest in a number of disciplines. Model formulations developed for the deposition of radionuclides, air quality, industrial emissions, fugitive dust, chemicals, trace metals, nutrient fluxes, pesticide applications, acid rain, and climate change all address the same underlying processes. Improvement of particulate dry deposition formulations requires an understanding of the major processes that control dry deposition rates. In addition, it is important to understand how those processes can affect the characterization of potential threats.

3.1 Processes

In atmospheric models, the surface layer is the air layer over the surface whose properties are largely controlled by the local surface fluxes. The strict definition of the surface layer is a fully turbulent layer over homogenous surfaces under steady-state conditions. With this surface layer, a second layer is designated that refers to the laminar, or near-laminar, flow that occurs immediately over the surfaces. This layer, which is referred to here as the “quasi-laminar layer,” may exist only intermittently in nature as the flow changes over the surfaces. In the literature, this layer is also referred to as the “laminar sublayer,” “sublayer,” or “deposition layer.” The processes that control dry deposition to various surfaces involve three sequential sets of processes:

1. Through the turbulent surface layer, an airborne contaminant moves by the combined effects of eddy diffusion (i.e., carried by turbulent movements of air) and gravity.

2. Moving through the quasi-laminar surface layer, an airborne contaminant can reach the surface by molecular diffusion, interception, or impaction.

3. Retention or rebound of an airborne contaminant at the surface depends on a combination of surface and impact properties.

3.1.1 Eddy Diffusion

Eddy diffusion refers to the transport resulting from turbulent movements in the air near the receptor surface. This mechanism is important for the range of particles that tends to follow turbulent air currents. For those particles, eddy diffusion provides an upper limit to the rate at which they can be moved to the deposition surfaces in the absence of gravitational settling. When surface processes (described below) limit the deposition rates, the eddy diffusion is of secondary importance.

3.1.2 Molecular Diffusion

Molecular diffusion by Brownian motion is usually assumed to dominate the diffusion processes in the quasi-laminar surface layer. However, there is the possibility that phoretic forces also can locally influence dry deposition fluxes.

Brownian Motion

Particles in the range of 0.001 to 0.1 micron move like gaseous molecules in flowing air (i.e., they exhibit rapid random Brownian motion). Their motion causes them to collide with any nearby surfaces. These smaller particles tend to adhere to these surfaces as the result of intermolecular forces. Contaminants are subject to diffusion coalescence under both turbulent and non-turbulent flows. This mechanism tends to be an effective deposition process with very small particles depositing at rapid rates on the nearest available surfaces. Under some circumstances, this diffusion mechanism can continue to be the dominant deposition process for particles >0.1 micron.

Phoretic Processes

Electrophoresis (electrostatic attraction), diffusiophoresis, and thermophoresis are processes that can influence the deposition of particles small enough to be influenced by molecular collisions or to have high ion mobility. Phoretic processes are neglected in many dry deposition formulations.

Electrophoresis or electrostatic attraction causes the movement of charged particles in the presence of an electric field. The direction of movement depends on the direction of the field and the sign of the charge on the particle. Attractive electrical forces have the potential to assist the transport of small particles through the quasi-laminar deposition layer and, thus, could increase the deposition velocity in situations with high local field strengths. However, Hicks et al. (1982) suggest that this effect is likely to be small in most natural circumstances.

Diffusiophoresis can change the rate of dry deposition of particles imbedded in a surface gradient of a gas created by a condensation or evaporation of the gas to/from the surface. There is a difference in the kinetic energies imparted by collisions with up-gradient and down-gradient gas molecules. This process imparts momentum to the particles, which tends to move them down-gradient for denser-than-air gases and up-gradient for lighter-than-air gases.[5] In addition, the introduction of new water vapor molecules at an evaporating surface displaces a certain volume of air.[6] This effect, called Stefan flow, tends to reduce deposition fluxes from an evaporating surface.

Thermophoresis results in a net directional particle transport in the presence of a thermal gradient. For a particle in a thermal gradient, the air molecules striking one side of the particle will be more energetic than those on the other side. This effect will tend to move small particles away from a heated surface and towards a cooled surface.

3.1.3 Gravitational Settling

Gravitational settling is the downward motion of particles that results from the gravitational pull of the earth. Gravity is important for the dry deposition of the larger (>10 micron) particles. For smaller

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[5] For example, over an evaporating water surface, a particle is more likely to be impacted by water molecules on the side of the particle facing the surface. Because the water molecules have a lower molecular weight than the average air molecule, there is a net force toward the surface, which results in a small enhancement of the deposition velocity of the particle (USEPA 1994).

[6] For example, 18 g of water vapor evaporating from 1 m^2 will displace 22.4 liters of air at standard temperature and pressure conditions (Hicks 1982).

particles, the effects of molecular forces become larger than the gravitational force. Particulate sizes, densities, and shapes largely define gravitational settling rates.

3.1.4 Interception

The predominant deposition mechanism for particles in the range of 0.2 to 2 microns diameter is often assumed to be interception. These particles tend to move with the airflow streamlines, and interception occurs only on the limited surface areas when streamlines intersect a surface element. Interception occurs most effectively when the surface element structures that the air is flowing through are smaller than the aerosol or solid particle diameter – a condition that does not tend to occur in the natural air-surface interface and limits the effectiveness of this process in dry deposition.

3.1.5 Impaction

Particles with diameters 2 microns and larger are effectively deposited by direct impact. These particles have sufficient momentum such that they only partly, or do not, follow the airflow streamline. As air flows over the surfaces, these particles collide with the surface elements, a deposition process termed inertial or direct impaction.

3.1.6 Particle Surface Properties

In addition to the size, density, and shape characteristics that are critical deposition properties, the types of particulate component materials and the resultant surface properties are important in determining deposition rates. The “stickiness” of the particle surfaces changes the fraction of particles that will actually be deposited on the surface. For example, experimental data show that Lycopodium spores tend to be stickier than fly ash particles (Chamberlain 1991).

3.1.7 Concurrent Processes

These dry deposition processes act concurrently. Starting with the airborne concentrations at the upper levels of the local atmospheric surface layer, eddy dispersion and gravity move particulate matter to the quasi-laminar surface layer. Then, the interception, inertia, and molecular diffusion processes result in the airborne material reaching the surface. The effectiveness of these delivery processes varies strongly with size and density of the particles, surface characteristics, and local meteorological conditions. Finally, retention depends on the properties of the particle surfaces and receptor surfaces along with the kinetic energy of impact.

3.2 Modeling Concepts

The modeling concepts for dry deposition have evolved over the past several decades as products of several related fields of interest. Friedlander (2000) and Seinfeld and Pandis (1998) described the underlying concepts and provide a foundation for understanding dry deposition from the standpoint of aerosol properties and processes. Chamberlain (1991) provided a detailed review of the processes controlling the fate of radioactive aerosols in the atmosphere. A literature review of reported rates of dry deposition and suspension conducted for a wide range of applications was provided by Sehmel (1980). Experimental acid-rain studies that have addressed the issues of modeling dry (and wet) deposition were provided by Hicks (1984) and Cantor (1986).

Resistance-based approaches are widely used as a basis for dry deposition formulations. This approach, explained in more detail below, has the advantage of providing a means of combining a number of the processes controlling dry deposition into a single formulation. In one of the early implementations, Sehmel and Hodgson (1978) proposed an empirical model based on curve fits to wind tunnel deposition results for a range of soil surface covers. Their model combined empirical data with the theory for molecular diffusion of very small particles and gravitational settling rates for larger particles. Sehmel and Hodgson also demonstrated the importance of considering the density of the particles in the dry deposition computation. Subsequent applications have included air quality (e.g., chemicals and trace metals), health physics (radionuclides), and acid rain models.

3.2.1 Settling Velocity

Atmospheric transport models developed in the 1950s and 1960s used relatively simple formulations for computing dry deposition rates. A major concern during that period was simulating the regional to global deposition from atmospheric nuclear explosions. For these particles, the settling velocity was based on a relationship appropriate for the particle size and density. For elevated releases, such as those from atmospheric nuclear weapons testing, the initial height, settling velocity, and mean atmospheric winds determine the downwind locations where the larger particles will tend to fall to the ground. This type of deposition is aptly referred to as “fallout.”

Adapting this concept to a plume near the earth’s surface, the flux to the surface from gravity, F_g, can be approximated using the air concentration C in g m^{-3} measured near the ground:

$$F_g = - v_s C \tag{1}$$

where F_g is the gravitational dry deposition flux, g m^{-2} s^{-1}
v_s is settling velocity, m s^{-1} (often expressed as cm s^{-1})
C is air concentration, g m^{-3}

This approach of using a settling velocity, the fall velocity of a particle in the atmosphere when not significantly affected by forces other than friction, works well for larger particles but greatly underpredicts the dry deposition rates for smaller particles (smaller than a few microns in diameter). These smaller particles are of particular concern for inhalation-pathway hazards (Chamberlain 1991). For the particles in the intermediate range, a combination of inertial and gravitational effects acts on the particles and results in higher deposition rates.

3.2.2 Deposition Velocity

The challenge in the 1960s and 1970s was to develop models that could simulate the deposition rates for smaller particles. Most widely implemented were dry deposition formulations based on an empirical parameter, the deposition velocity, defined in analogy with the settling velocity described in Equation (1) above:

$$v_d = - F/C \tag{2}$$

where v_d is the deposition velocity, m s^{-1} (also often expressed as cm s^{-1}), and F is the dry deposition flux, g m^{-2}. Although functionally equivalent to Equation (1), the important difference is that the velocity term, v_d, includes additional deposition processes not accounted for in the settling velocity. Other methods of formulating the dry deposition flux were proposed (see, for example, USEPA 1994) but were ultimately abandoned in favor of the representation expressed in Equation (2).

The computation of deposition for smaller particles is thus accomplished using the relationship analogous to that used for larger particles, with v_d approaching v_s as particle size increases. In practice, a deposition velocity is specified and the dry deposition flux is computed using

$$F = -v_d C \tag{3}$$

The initial implementations of this approach involved using a “representative value” for the dry deposition velocity for all particles and locations. Using a single deposition velocity (either as a fixed value or as an input parameter) to compute dry deposition is still widely used in the older dose and risk assessment models, including recent updates to several of those models. The single value concept is a convenient way to make fast estimates of the dry deposition flux but it does not take advantage of our current knowledge of the variety of processes, discussed in this report, that control the flux. The factors that determine the values of v_d have been found to be complex and highly dependent on the particle properties, ambient turbulence, and surface properties; on concurrent fluxes of momentum, heat, moisture, and other materials; and on electrical forces. These processes interact to give a wide variation in the reported values for deposition velocity (Sehmel 1980; NRPB 2001).

3.2.3 Resistance Approach

The resistance model was developed to provide a formulation that addressed the interactions of the controlling processes. The dry deposition resistance formulation started as an adaptation of the resistance formulations developed to study micrometeorological and CO_2 fluxes over agricultural surfaces (Droppo 1974; 1980). Businger (1986) provided a critical review of the use of these micrometeorological techniques to measure pollutant flux rates.

The subsequent development of this approach, in particular, the estimation of the atmospheric resistance, was aided by the development of non-dimensional parameterizations of the fluxes and profiles in the atmospheric layer near the earth’s surface (surface layer).

Resistance approaches are the basis of the dry deposition formulations in most currently deployed models that estimate a situation-specific deposition velocity. The resistance approach uses an analogy with electrical circuits to create a model that incorporates the various processes resulting in the dry deposition of a contaminant. The contaminant concentration gradient over the surface corresponds to the potential for deposition (i.e., voltage) and the eddy/molecular dispersion processes correspond to resistances. The resistance approach assumes that eddy and molecular diffusion act as a series circuit. For particles, the approach incorporates gravitational settling as a parallel resistance term. The resistance approach is largely an application of the Monin-Obukhov Similarity Theory of the atmospheric surface layer. Sorbjan (1989) provides a summary of surface layer research based on this theory.

Surface-Layer Concepts

The Monin-Obukhov Similarity Theory (often referred to as surface-layer similarity theory, or just surface-layer similarity) provides non-dimensional parameterizations of momentum, heat, and moisture fluxes and profiles over homogeneous surfaces under fully turbulent steady-state conditions. Under this specific set of conditions, the constants for the parameterizations for temperature, moisture, wind profiles, and fluxes have been defined by a series of field studies summarized by Sorbjan (1989). These parameterizations have been extended to airborne contaminants as a means of both studying dry deposition rates and developing formulations to define dry deposition rates.

Figure 3.1 illustrates the air structure near the earth in terms of the special conditions where surface-layer similarity theory applies. The term "surface layer" refers to the air layer over the earth's surface cover whose properties are largely controlled by the local surface fluxes. Within statistical uncertainties, the fluxes do not vary with height in this layer.

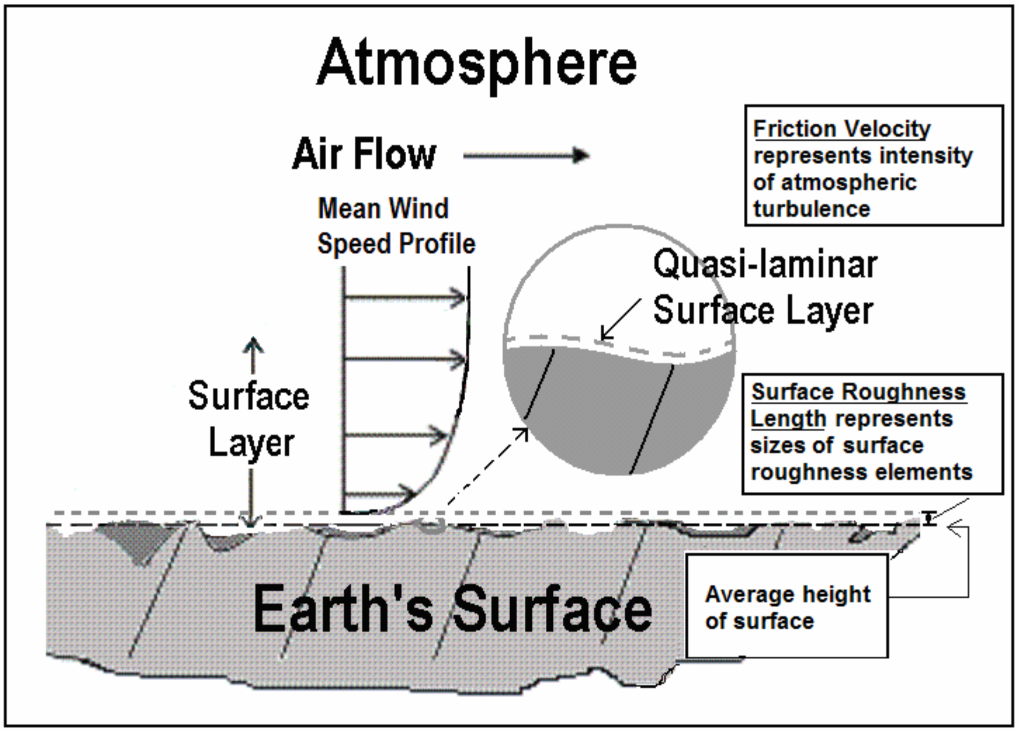

Figure 3.1. Air Structure near the Earth's Surface

The profile of wind speed has an approximately logarithmic shape that under neutral atmospheric stability can be parameterized with a friction velocity and surface roughness length. The friction velocity parameter provides a measure of the intensity of atmospheric turbulence. For an aerodynamically rough but relatively flat surface, an extrapolation of the mean wind speed profile downward shows that it reaches zero at some distance above the physical surface. The height at which this occurs is called the roughness height (or roughness length), z_o. The roughness height is positively correlated with the physical roughness of the surface although a strict functional relationship between measures of physical roughness and z_o do not exist. Some studies have found that the surface roughness length tends to be about one-tenth of the dimensions of the surface elements. In environments with substantial vegetation, such as over agricultural crops and forest canopies, the practice is to displace the entire velocity profile upward such that the height at which velocity profiles reach zero is the sum of displacement height d and a canopy roughness length as functional representations of mean values for characterizing the over-canopy turbulent flow.

Very close to the surface ($z \leq z_o$), a layer with nearly laminar flow will exist. Because turbulence invariably modifies this layer to some extent, it is referred to as the "quasi-laminar surface layer." This layer, for which the structure is largely expected to be controlled by molecular processes, is illustrated in Figure 3.1.

Resistance Analogy Formulations

The dry deposition resistance approach evaluates a total resistance to deposition of a contaminant – a term that is the inverse of the deposition velocity. The deposition rate is computed also using the concentration over the surface:

$$F = -(C-C_o)/r_t \tag{4}$$

where r_t is the total resistance to dry deposition, s m^{-1}; C is the air concentration, g m^{-3}, at a reference height; and C_o is the concentration, g m^{-3}, at the surface. Assuming that $C_o << C$, the formulation is similar to the definition of deposition velocity:

$$F = C/r_t \tag{5}$$

Inspection of Equations (2) and (5) shows that $r_t = 1/v_d$. The condition for C_o to be close to zero occurs when all the material reaching the surface remains on the surface and results in a depletion of concentration immediately over the surface.

Resistance approach implementations define and incorporate application-specific resistance terms. Special terms or layers are included to capture the influence of the processes controlling the deposition. A typical resistance formulation for the dispersion/diffusion-controlled flux of a contaminant is

$$V_d = 1/(r_a + r_b + r_c) \tag{6}$$

where r_a = atmospheric resistance (also referred to as aerodynamic resistance), s m^{-1}
r_b = resistance across quasi-laminar sublayer, s m^{-1}
r_c = surface resistance, s m^{-1}

Formulations for the resistance in the fully turbulent atmosphere are based on surface-layer similarity theory. Process-specific formulations for the sublayer and surface resistances are needed.

During non-neutral conditions, the atmospheric resistances for momentum and mass (moisture and sensible heat) fluxes in the surface layer can have different values. Droppo (1985) showed that pollutant fluxes (ozone) occur at a rate consistent with mass fluxes. Some dry deposition models assume that particles are transported vertically at the same rate as momentum (i.e., an aerodynamic resistance), and other models assume that small particles are passively carried by air movements and carried to the surface at the same rate as mass. Because the atmospheric resistance does not limit particle deposition under most conditions, this transport assumption rate is not normally a critical assumption but is a source for small differences in simulated deposition rates between models.

For particles, the process of gravitational settling acts in parallel with the diffusion-controlled flux. The gravitational settling flux does not depend on a concentration gradient but rather tends to carry airborne materials to the surface at some settling velocity, v_s. The approach assumes that the dispersion/diffusion and gravitational deposition processes act like resistances in parallel:

$$v_d = 1/r_t = 1/(r_a + r_b + r_c) + v_s \tag{7}$$

The fact that diffusion flux will result in the mean concentration varying with height implies that the computed gravitational flux based on settling velocity and concentration will vary with height. However, this is inconsistent with the formulation of Equation (7), which implicitly assumes a height-independent gravitational flux term.

Seinfeld and Pandis (1998) derived a dry deposition flux relationship based on the assumption that $r_c = 0$, by equating the vertical fluxes in two layers over a surface (defined by three vertically spaced horizontal planes, 2 at the top, 1 at the middle, and 0 at the surface) to the total resistance:

$$F = \left(C_2 - C_1\right)/r_a + v_s C_1 = \left(C_1 - C_0\right)/r_a + v_s C_0 = \left(C_2 - C_0\right)/r_t \tag{8}$$

where C_1, C_2, and C_3 are concentrations at the layer boundaries such that C_2 and C_1 are concentrations across the turbulent surface layer and C_1 and C_0 are across the quasi-laminar surface layer. Then, the following relationship for deposition velocity (based on the inverse of total resistance) was derived algebraically from Equation (1), assuming that $r_s = 0$ (and, thus, that $C_0 = 0$):

$$v_d = 1/r_t = 1/(r_a + r_b + r_a r_b v_s) + v_s \tag{9}$$

Equation (9), as proposed by Pleim, Vernkatram, and Yamartino (1984), was documented and evaluated in the dry deposition model review (USEPA 1994). Although the formulations in Equations (7) and (9) fit experimental data as reported in USEPA (1994), these formulations are inconsistent with the mass conservation equation. Sehmel and Hodgson (1978) and Venkatram and Pleim (1999) propose a mass-consistent relationship for deposition velocity:

$$v_d = \frac{v_s}{\left(1 - e^{-r_t v_s}\right)} \tag{10}$$

where r_t is the total resistance is the sum of the aerodynamic layer, quasi-laminar sublayer layer, and surface resistances:

$$r_t = r_a + r_b + r_c \tag{11}$$

Equation (10) yields the equivalent results as the widely used other relationships, Equations (7) and (9), without a mass conservation issue.

3.3 Models

The dry deposition models that have been widely accepted and incorporated in air dispersion models are reviewed in this section. The list of models discussed is not meant to be comprehensive but rather representative of the evolution of dry deposition formulations.

3.3.1 Sehmel and Hodgson Model

Sehmel and Hodgson (1978) proposed an empirical model based on curves fit to wind tunnel deposition results for a range of soil surface covers. They were among the first to combine micrometeorological relationships with experimental deposition data to provide a model for dry deposition that was applicable over a range of surface roughnesses, friction velocities, and particle sizes. Their model combines experimental results in the mid-range with molecular diffusion and gravitation models for the smaller and larger particles, respectively. This model has the advantages of having wide applicability and being based on empirical fits[7] to experimental data.

As noted above, the Sehmel and Hodgson model has the form:

$$v_d = v_g/(1 - e^{(-v_s/u_* e^A)}) \tag{12}$$

where the value of A is computed based on an empirical function,

$$A = f(Sc, r, u^*, z_o, D) \tag{13}$$

where Sc = Schmidt number
r = particle radius
u* = friction velocity
z_o = roughness length
υ = kinematic viscosity of air
D = Brownian diffusivity

Figure 3.2 provides an example of the curves of deposition velocity generated by the Sehmel and Hodgson model with added information on surface roughness and particulate-matter size range. The curves in Figure 3.2 are for stable atmospheric conditions, i.e., relatively low levels of turbulence. This figure clearly shows the importance of accounting for the particle size distribution. An intermediate particle size range (0.1 to 1 micron diameter) has a minimum deposition velocity as well as changes in deposition velocity of several orders of magnitude as a function of particle size. An order-of-magnitude change in deposition velocity also occurs between crops and very smooth surfaces. The model, however, does not address deposition velocities for roughness values associated with urban areas. Also indicated are the EPA's PM 2.5 and PM 10 ("PM n" = "particulate-matter with less than n μm diameter"), which are of particular interest for defining potential health impacts.

Several important dependencies of deposition velocity on ambient conditions are not illustrated in Figure 3.2. Increasing the friction velocity will tend to increase deposition velocity. The Sehmel and Hodgson model predicts minimum deposition velocities for a grass surface of 0.026, 0.031, and 0.2 cm s^{-1} for friction velocities of 30, 50 (as shown in Figure 3.2), and 200 cm s^{-1}, respectively. For more turbulent atmospheric conditions (e.g., neutral and unstable conditions), the rate of dry deposition will tend to increase, bounded by the extent that eddy dispersion rather than surface processes is limiting dry deposition.

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[7] Users have noted that although some small anomalies occur in the behavior of the empirical curves that are counter-intuitive in terms of the physical processes, the magnitudes of the anomalies represent insignificant differences in the magnitude.

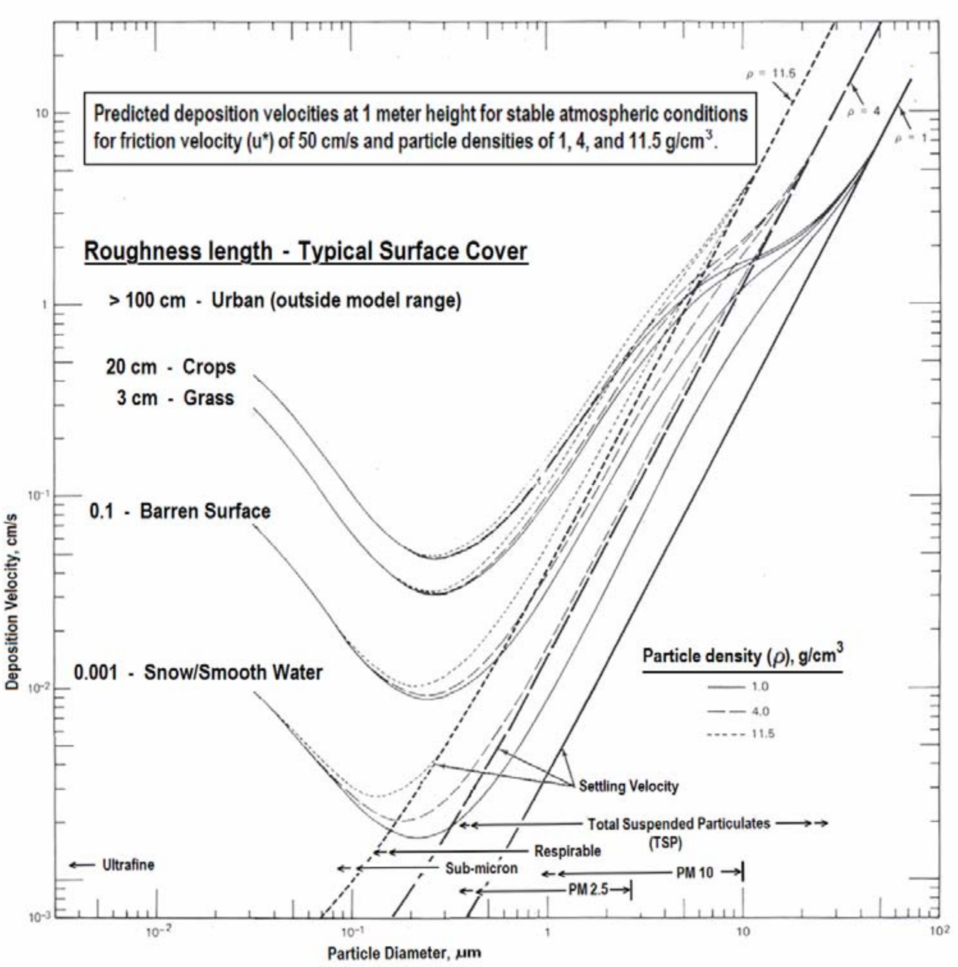

Figure 3.2. Modeled Particle Deposition Velocities (from Sehmel [1984])

3.3.2 Deposition to Vegetation Canopies

Although the Sehmel and Hodgson model and similar models provided a basis for developing dry deposition formulations for relatively low surface covers, these models did not address the more complex processes in higher vegetation canopies and urban areas. Over the past several decades, field studies and model development have addressed the problem of modeling to aerodynamically rougher surfaces.

Most of these efforts have been directed at forest canopies, largely because of the need to understand acid-rain phenomena. The results of studies of dry deposition to forest canopies provide valuable insight into the change in dominant processes with changes in surface characteristics. These forest canopy results provide a basis for developing dry deposition models for urban areas, which are considered below.

An area of controversy in acid rain research has been the magnitude of dry deposition of particles to forest canopies. Wesely, Cook, and Hart (1983) studied the fluxes of gases and fine particles above a deciduous forest in wintertime. In a similar study with the same instrumentation, Hicks et al. (1982) studied sulfur deposition to a pine forest. These eddy-flux[8] studies of fine particles showed the defoliated forest to have lower deposition velocities than the pine forests. Makarov et al. (1996) had similar results in studies of the dry deposition of pesticide aerosols on pine needles and birch leaves.

Several studies of the deposition rates of larger particles have found greater rates than those predicted by the Sehmel and Hodgson deposition model (Kim et al. 1997; Lin, Noll, and Holsen 1994). Lin, Noll, and Holsen (1994) found higher than predicted rates to a smooth greased surface in the 5 to 80 micron range of particle diameters. Kim, Kalman, and Larson (2000) also conducted field studies of dry deposition rates for large particles and obtained similar results. They used three different types of artificially generated particles (perlite, diatomaceous earth, and glass beads) in the size range of 10 to 100 microns diameter to study deposition to sampling plates at two sites – on a building top and in a field. Best agreement or predicted values of particle deposition velocity was found by using Slinn and Slinn's (1980) values of inertial deposition collection efficiencies in the Pleim, Venkatram, and Yamartino (1984) dry deposition model, which is discussed below.

Large-particle deposition results are shown in Figures 3.3 and 3.4 (Kim, Kalman, and Larson 2000). Figure 3.3 compares particle dry deposition velocities of the quasi-laminar layer as a function of particle aerodynamic diameter for wind speed (u) of 2 m s^{-1}, friction velocity (u*) of 30 cm s^{-1}, and surface roughness height (z_o) of 0.1 cm. Figure 3.4 compares friction velocity as a function of particle diameters of 15, 30, and 100 microns and surface roughness height of 0.01 cm.

Figures 3.3 and 3.4 show that particulate material can be locally removed by the combination of gravity and inertia faster than the average rate that the particulate material can be delivered by gravitation alone through the atmosphere. Kim, Kalman, and Larson (2000) suggest that widely used dry deposition models are not well formulated for particles in the 10 to 100 micron range. They show that surface processes can support a faster local rate of deposition on a collection plate than are predicted by the Schmel and Hodgson model.

Improved formulations by Pleim, Venkatram, and Yamartino (1984), described below, account for inertial surface effects as well as gravitation effects for this size range. This change is particularly important for applications such as an RDD event that may have significant mass in the larger particle sizes. Recently developed/upgraded models such as ISC, CALPUFF, and ADOM use formulations that incorporate the improvements.

Efforts to model deposition to vegetation canopies using the resistance approach have met with some success when surface-specific deposition processes are included. For grasses, Davidson, Miller, and Pleskow (1982) proposed a deposition model that accounted for the fine nature of grass structure. For forest canopies, Slinn (1982) proposed a surface-oriented resistance-based model for dry deposition of particles by treating the canopy as a set of cylinders with specified collection efficiencies. Of particular note were the formulations that Slinn proposed for estimating collection efficiencies.

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[8] Eddy-flux refers to a research method of computing the net vertical contaminant flux from a rapid response time series of vertical velocities and concentrations measured at some height over a surface. The method applies to the flux of fine particles that are passively carried by air movements.

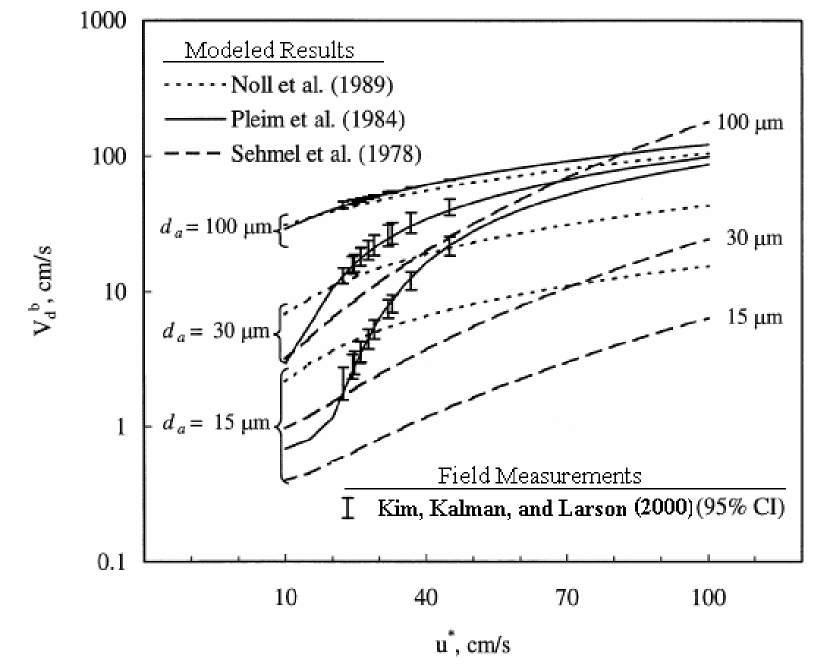

Figure 3.3. Large Particle Deposition Velocity, V_d^b, as Function of Friction Velocity u* for a Range of Particle Diameters, d_a[9]

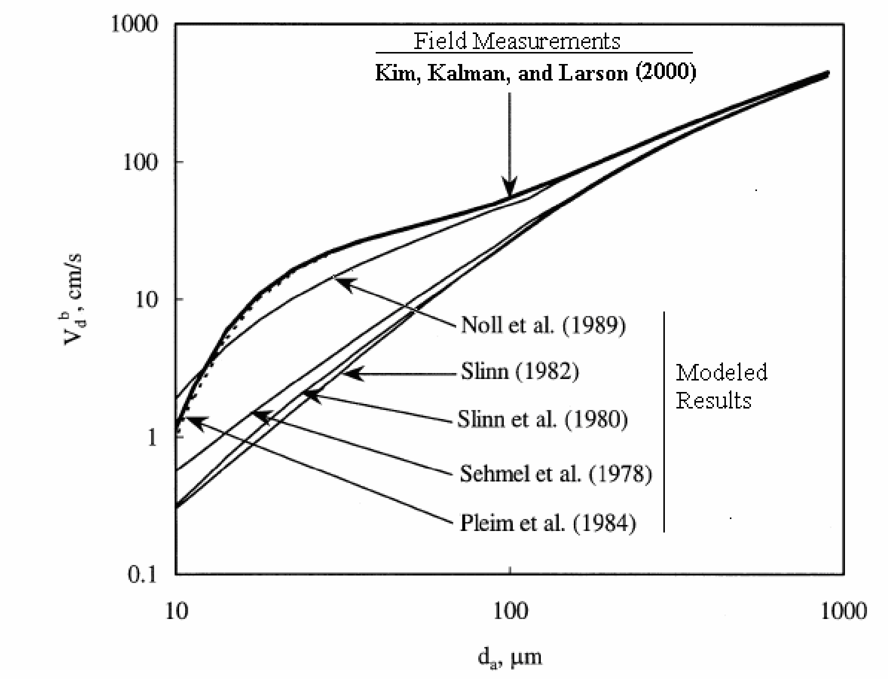

Figure 3.4. Large Particle Deposition Velocity, V_d^b, as Function of Particle Diameter, d_a[12]

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[9] Reprinted from Kim, Kalman, and Larson (2000), by permission from Elsevier.

Slinn's model has been widely used in developing dry deposition formulations for vegetation canopies. The Slinn model uses two resistance terms (aerodynamic and surface) and a gravitational settling term. For Slinn's model, the relationship for deposition velocity is:

$$v_d = 1/r_t = 1/(r_a + r_s) + v_s \tag{14}$$

with r_s including both r_b and r_c. The surface resistance is computed using:

$$r_s = \frac{u_z}{{u_*}_v^{\,2}} f\left(\xi_c, \gamma_v\right) \tag{15}$$

where u_z = wind speed at height z

$\xi_{c\,=}$ collection efficiency for cylinders

γ_v = deposition factors parameter

Slinn combines all the surface deposition processes in the single dimensionless collection efficiency ξ_c for the cylinders. The collection efficiency of the canopy is estimated as:

$$\xi_c = \overline{\mathrm{E}}\mathrm{R}_\mathrm{R} \tag{16}$$

and

$$\overline{\mathrm{E}} = \mathrm{E}_\mathrm{IN} + \mathrm{E}_\mathrm{IM} + \mathrm{E}_\mathrm{B} \tag{17}$$

where R_R = a rebound fraction (i.e., stickiness factor)

E_{IN} = the efficiency of interception

E_{IM} = the efficiency of impaction

E_B = the efficiency of Brownian motion

The dimensionless deposition factors parameter γ_v is defined as:

$$\gamma_v = h_c \left[\frac{c_d \alpha \cdot u_o}{K_o}\right]^{0.5} \tag{18}$$

where h_c = canopy height, m

c_d = dimensionless drag coefficient for the canopy

α = collection area per unit volume of the canopy, m^{-1}

u_o = characteristic wind speed in the canopy, $m\ s^{-1}$

K_o = average turbulent diffusivity in the canopy, $m^2\ s^{-1}$

Wesely Canopy Model

The resistance concept for characterizing local fluxes over a specific surface was extended to a regional scale for long-range modeling for acid-rain dry deposition computations by Wesely (1989). The challenge was to be able to estimate a representative deposition velocity for the relatively large grid cells used in these models.

Wesely addressed dry deposition to high vegetative canopies, mainly forests, which are the receptors of principal interest for acid-rain applications. Figure 3.5 illustrates schematically the multi-level layered resistance model that Wesely developed for deposition of gasses and particles. The proper characterization of grid-scale deposition for long-range acid rain modeling applications has continued to be a topic of interest in the literature, with most contributions using different implementations of resistance approaches.

AIRMON Inferential Dry Deposition Monitoring

Dry deposition has proven to be very difficult to measure directly in monitoring programs. As part of a national acid rain monitoring program called AIRMON, an inferential dry deposition monitoring method for selected atmospheric contaminants was developed by National Oceanic and Atmospheric Administration (Hicks et al. 1987; Hicks et al. 1991). The method is based on a combination of measurements of air concentrations and characterizations of the concurrent surface momentum and heat transfer. Contaminant flux parameterizations from field studies are used to estimate dry deposition rates. The correlation between the deposition flux and a parameter representing the horizontal dispersion rate defined as the product of the wind speed and standard deviation of wind direction was reported by Wesely, Cook, and Hart (1983). This methodology provides a means of estimating long-term dry deposition fluxes from parameters that are available or can be estimated/measured much more easily than making direct flux measurements. The same capability is needed in air models used for emergency preparedness with applicability for much shorter time-scales.

Because AIRMON attempts to use a dry deposition model to estimate routine deposition rates, its performance can provide some insight into how well dry deposition models can simulate actual deposition rates. Studies of the dry deposition inference method conducted over a forest in Tennessee found a consistent overestimate of the deposition velocity for sulfur dioxide that was felt to be related to the failure for the model to account for the concurrent moisture fluxes (Matt et al. 1987). Although the applicability of this result to particle fluxes is uncertain, because of the differences in the relative importance of the dominant processes, the result does indicate a limitation in current modeling formulations.

Chamberlain Foliage Interception Model

To model potential impacts from ingestion of foliage as well as other pathways, the net interception of material by foliage needs to be understood. Chamberlain (1991) proposed a relationship to quantify the fraction of radioactivity retained in foliage:

$$p = \exp(-\Gamma / M_H) \tag{19}$$

where Γ is an interception coefficient, kg m^{-2}, and M_H is the density of herbage, kg dry wt m^{-2}.

A value for Γ of about 3 kg m^{-2} has been observed for a number of studies of mixed grasses and other narrow-leafed foliage from wet and dry deposition of vapors and particulates from sprays, particle spreaders, vapors, spores, and stack fallout. An interesting aspect of this approach is that the interception coefficient seems to be relatively independent of the chemical and physical characteristics, particle size, ambient meteorological conditions, and types of vegetation. Chamberlain does, however, report that the literature results for broad-leaved plants and shrubs are more scattered. An implication for modeling is that a given type of vegetation appears to have consistent fractions of deposited material on the vegetation.

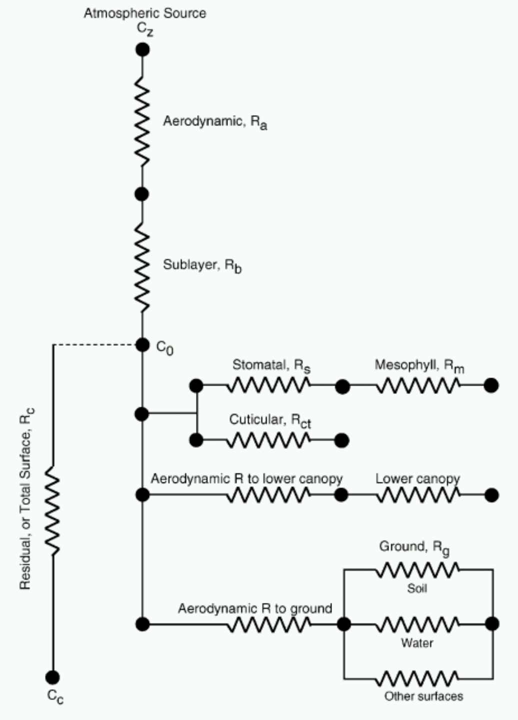

Figure 3.5. Wesely's Approach to Dry Deposition Modeling Using Multi-Layer Resistances[10]

This approach to dry deposition on vegetation canopies is unique in that it stresses the filtration potential of the surfaces as being relatively independent of ambient meteorological processes and particle sizes. It is expected that some dependence on these parameters will be needed in a formulation applicable to a wider range of surface types. In addition, assuming that the deposition will scale better with plant surface area than with the mass of plant material should provide a more general formulation.

Zhang Canopy Model

Zhang, Brook, and Vet (2003) added a term for "non-stomatal" surfaces in their vegetative canopy dry deposition model for gasses. Previous models had assumed that the rate of gas uptake was only a function of the status of the stomata. They found that this improved formulation did a much better simulation of observed deposition rates.

The recognition that deposition processes to a complex canopy must be formulated as multiple flux processes considering the properties of the various surface elements represents a modeling advance that has application to improving deposition models for urban canopies.

Muller and Prohl Model

Muller and Prohl (1993) proposed a model for assessing the consequences of airborne material from a nuclear accident. Their dry deposition model used the leaf area index to estimate the surface areas

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[10] Reprinted from Wesely and Hicks (2000), with permission from Elsevier.

available for deposition. They computed a wide range of deposition velocities for aerosol-bound radionuclides for different surfaces. The maximum deposition velocities at the time of fully developed foliage was 0.05 cm s^{-1} for soil, 0.5 cm s^{-1} for trees, and 1.05 cm s^{-1} for grass. These tree and grass results are counter to other more complex formulations that suggest that dry deposition rates should increase with increasing surface roughness, but they do agree with observational data reported by other authors that found relatively high deposition rates to grasslands.

Ruijgrok et al. Canopy Model

There has been considerable interest in the area of acid rain research over the magnitude of dry deposition of particles to forest canopies. Carefully designed and conducted mass-budget studies of particle deposition at the Speulder forest (The Netherlands) provided important results (Erisman et al. 1997; Ruijgrok, Tieben, and Eisinga 1997). The canopy deposition model proposed by Slinn (1982) was used to evaluate estimates of dry deposition of acidifying aerosols and particles with base cations. Their results indicate that on a given canopy, two forces dominate dry deposition: particle size and friction velocity. They found significantly greater deposition rates (2-3 times greater) than had been determined in earlier studies for similar canopies using measurement based on eddy-flux deposition. The reason for the difference, although not certain, appears to be that gravitational settling of larger particles were missed with the eddy-flux approach.

Gaseous Flux Canopy Models

The Wesley model, which also included particle deposition, and more recently the Meyers Multilayer Model (Finkelstein et al. 2000; Meyers et al. 1998; Wu et al. 2003a; Wu et al. 2003b) address the flux of gases to vegetation using a multilayer approach. Although the details of surface uptake and retention processes are quite different for particles and gases, the overall approach, including the characterization of atmospheric processes, should be quite applicable to passively dispersed particles.

Mechanistic Models

Petroff (2005) developed a canopy dry deposition model using a mechanistic approach to aerosol dry deposition onto agricultural and forest areas. The proposed approach has two steps. First, interaction between aerosols and foliar surface is formulated on the scale of each foliar element. Second, the total deposition is computed, based on the winds within the canopy, aerosol deposition mechanisms, and canopy characteristics. Deposition mechanisms are inertial impaction, gravitational settling, and Brownian diffusion. The canopy characteristics are spatial distribution, orientation, and detailed structure of foliar surfaces.

Simulations showed improved agreement with field measurements compared to the highly empirical models typically used in radiological risk or air quality assessments.

3.3.3 Models for Dry Deposition to Water

Large urban areas are often located near oceans, lakes, and/or major rivers. The capability to model dry deposition to water surfaces is important because of potential contamination of water supplies as well as local aquatic ecosystems. The dry deposition rates tend to be much smaller over water surfaces than over land surfaces. The ability to simulate the deposition to water surfaces is particularly important in urban areas from the viewpoint of defining levels of contamination in water.

Water surface processes, such as wave action, bubble formation, and strong moisture fluxes, affect the deposition rate. Dry deposition is further complicated by variable states of the air-water interface. The air-sea interaction will change both the surface resistance and the rate of eddy transport in the air. Under broken water conditions, factors such as the increase in exposed surface area, breaking bubbles, etc., will decrease the surface resistance for deposition.

It is important to understand the relative importance of the air-water interface processes (Cantor 1986). Although the same surface-layer properties as over land likely dominate the fluxes, the interactions can be much more complex over water. The surface micro-layers affecting the surface roughness are not seen as being a major factor in deposition rates except at a local level. Bubble ejection is seen as a potentially significant counter-flux process. A symposium proceedings addressed the deposition of chemical contaminants to the Great Lakes and coastal waters of the United States (Baker 1997).

A number of authors have proposed assessment models for estimated deposition rates to water. Slinn and Slinn (1980) proposed a detailed model for deposition to water surfaces. A model accounting for different surface mass-transfer rates for smooth and broken water surfaces (as well as condensation effects) was proposed by Williams (1982). Changes in eddy transport with changing conditions in the air-sea interface are implicitly included but not directly addressed. Droppo, Ecker, and Redford (1987) proposed a model for over-ocean plumes from incinerator ships that accounts for influences on the change of water roughness and eddy transport in the air with changes in wind speed. In fact, the exchanges and processes involved in deposition to natural water surfaces are much more complex and dynamic than these models assume.

The deposition rates to surface water have been studied relative to potential acidification of surface waters, including improvements to the Slinn and Slinn (1980) model by accounting for the effects of the rough surface and bubbles (Baker 1997). Zufall, Dai, and Davidson (1999) studied the effect of the wave surfaces on deposition of 4.0 and 6.7 micron uranine particles. They found that deposition rates on waves were increased over flat surfaces. The cases they considered gave an average increase of about 80%.

The fluxes to and from the ocean surface are complicated by the formation of a microlayer on the ocean. The Molecular Oceanic Boundary LAyer Model (MOBLAM) (Schlüssel et al. 1997; Soloviev and Schlussel 1994; Soloviev and Schlussel 1996) was developed to simulate the air-ocean interactions and fluxes that create, maintain, and destroy this layer. These same processes will control contaminant deposition rates.

3.3.4 Pleim, Vernkatram, and Yamartino (1984) Dry Deposition Model

The ADOM/TADAP model was designed for long-range transport and deposition of acidifying pollutants and photochemical oxidants. Pleim, Vernkatram, and Yamartino (1984) documented the dry deposition formulations used in this model.

In the review and testing of dry deposition models (USEPA 1994), the Pleim, Vernkatram, and Yamartino (1984) ADOM/TADAP dry deposition model, and the Sehmel and Hodgson model (1978) showed agreement with experimental dry deposition data for smaller particles (0.1 to 1 micron) as well as for larger particles (greater than 10 microns).

The EPA developed a series of ISC models for evaluation of hourly to annual impacts from industrial emissions. Although primarily developed for nonradioactive pollutants, these models are also used for radionuclide emissions with complex source terms.

A modified form of the Pleim, Vernkatram, and Yamartino (1984) model was developed and used in the ISC models as the result of a review of dry deposition models conducted by the EPA (USEPA 1994) to identify improved formulations for use in their local-region ISC atmospheric dispersion models. In the EPA review, an evaluation was made of the available field and wind tunnel deposition studies. Data sets were selected for testing dry deposition models. Simulations were made for the experimental conditions. The resulting comparisons of modeled and measured dry deposition measurements provide valuable insight into the relative performance of the models.

3.3.5 Noll and Fang Model

Fang et al. (1999) apply a "Noll and Fang" dry deposition model based on measurements of the deposition of atmospheric particles to surrogate surfaces. The empirical relationship for dry deposition velocity is[11]:

$$v_d = v_s + 1.12u^* e^{-30.36/D_p} \tag{20}$$

where v_d, v_s, and u^* are in cm s^{-1} and particle diameter (D_p) is in microns.

This model, along with the Sehmel and Hodgson model, were tested using deposition measurements in urban (near a highly trafficked area) and remote rural locations. Both models provided nearly identical comparisons with the measured data with a consistent underprediction of the dry deposition rates. The highly trafficked area computed deposition estimates were low by factors of 0.2 to 0.9 during the daytime and 0.1 to 0.9 at night. The rural area computed deposition estimates were lower than the measurements by factors 0.04 to 0.3 during the daytime and 0.02 to 0.3 at night.

These results indicate that although these empirical models do reasonably well at sites with heavy particulate loading, they are consistently providing lower deposition rates than were measured. The difference is very likely the result of inadequate parameterization of the differences in the deposition processes represented in the empirical formulation and the dry deposition measurements. In any case, neither the model nor the measurements would necessarily be representative of the actual total deposition rates to the local surfaces.

3.3.6 DERMA Dry Deposition Model

Baklanov and Sorensen (2001) proposed improved deposition models for long-range deposition computations as updates for DERMA, a 3-D Lagrangian long-range dispersion model using a puff

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[11] *Measurement and Modeling of Atmospheric Coarse Particle Deposition to a Flat Plate*, Ph.D. Dissertation, Chicago, IL, Illinois Institute of Technology, 1989.

diffusion parameterization. DERMA was developed at the Danish Meteorological Institute for nuclear emergency preparedness applications.

The parameterizations of removal processes of particulate radionuclides by wet and dry deposition were analyzed in some detail (Baklanov and Sorensen 2001). They suggest a dry deposition formulation based on the combination of Stokes Law with a correction term for small particles. Baklanov and Sorensen documented the iterative procedure for determining the terminal settling velocity for larger particles.

For particles, Baklanov and Sorensen (2001) suggest that the surface resistance is negligible for a forest canopy. On the other hand, Chamberlain (1991) provided data showing that both the particle and surface stickiness affect the retention rate of particulate radionuclides reaching the surface. For materials like fly ash, lower deposition rates were observed that indicated only 6 to 30% retention rates. Comparisons made for simulations of the Algeciras accident in Spain[12] with cesium-137 measurement data from the European monitoring network showed good agreement with experimental data.

3.3.7 Resistance Model Limitations

Relatively good agreement with field measurements over relatively ideal surface covers has been demonstrated for resistance-based dry deposition models. However, there are limitations in the application of a resistance approach to less-than-ideal surfaces. The current resistance-based dry deposition models for particulate matter suffer from three major limitations.

First, current resistance modeling approaches are based on concepts that are problematic for applications to urban and high vegetation canopies. Relations derived from Monin-Obukhov Similarity Theory may not strictly apply within such canopies.

Second, resistance models assume gradient-driven fluxes. The major process, gravitational settling, is not a gradient-driven flux and is inappropriately fit into the resistance concept.

Third, the resistance models are strictly applicable to homogenous or near-homogenous surface covers. The application to actual surface covers uses average particle concentrations and average deposition velocities, and thus fails to account for the highly nonlinear variation of deposition rates with particle size to different surfaces. Resistance models generally result in underestimates of the contribution of larger particles to dry deposition (Holsen and Noll 1992).

3.3.8 Urban Deposition Models

The knowledge about dry deposition within urban canopies is limited. Some experimental data are available for deposition on urban and on surrogate surfaces. Although models have been developed to address complex urban processes, there is no universal acceptance of any specific modeling approach.

Canopy studies provide an insight into the deposition processes that can be expected for elements of an urban canopy. Makarov et al. (1996) studied the differences in dry deposition between pine needles and birch leaves. Wesely, Cook, and Hart (1983) studied dry deposition on a defoliated deciduous forest

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[12] Elevated levels of cesium-137 were detected at the end of May and early June in southern Europe. The source was later attributed to the inadvertent melting of a medical radiotherapy device containing cesium-137 in a steel mill's scrap metal furnace located in the extreme southern tip of Spain, near the town of Algeciras.

canopy. Hicks et al. (1982) studied dry deposition on a pine forest. Together, the results of these studies demonstrate how different processes are important for different types of surfaces. “Flat” surfaces (defoliated trees, birch leaves) had lower deposition rates with mainly diffusion though laminar surface layers as the dominant surface deposition process. For smaller surface elements (pine needles), the addition of significant deposition by impaction and interception processes led to higher deposition rates. These results suggest that diffusion will be the main process for the deposition of fine particles on the flat anthropogenic surfaces in urban canopies (i.e., buildings, highways, bridges, etc.), but processes such as impaction and interception will increase the deposition rate for foliated vegetation and other fine-structured urban surface elements.

An urban area represents a complex area for assessment of potential exposures from an atmospheric release. Detailed models that address the processes leading to exposures in an urban environment have been developed for radiological exposures (Jones, Singer, and Brown 2006). The use of a single capture efficiency for a vegetation canopy (Slinn 1982) is replaced by arrays of such parameters in the urban models. The surface resistance in an urban area is a temporally and spatially changing variable. Eged, Kis, and Voigt (2006) used a Monte Carlo approach to evaluate potential radiological doses in urban environments. Five deterministic dose computation codes applicable to an urban environment were used.[13] The results show that these urban dose computation models provide some results that are the same and some that are not. Some “conciliation and harmonization” is seen as necessary before this suite of models can be used in a decision-support system.

Studies of the deposition of pollutants show that deposition is mainly controlled by large particles. By observing deposited particles, Tai, Lin, and Noll (1999) show that this effect is particularly true for urban locations where the coarse concentration of particles is high; however, the effect is also true for non-urban locations where the coarse concentration of particles is low. Similar results were obtained by Lee et al. (1996) for PCB dry deposition in an urban area.

Studies of urban deposition rates of hydrocarbons and metals show deposition-rate variations over urban areas that largely reflect the influence of local sources on ambient airborne contaminant concentrations (Azimi et al. 2005). Characterization of variations in the ambient aerosol size distribution that typically occur across an urban area complicates the modeling of in-plume aerosol interactions and, consequently, the computation of deposition rates. The potential complexity of particle number concentrations in a major European city is shown in the measurement modeling comparisons by Gidhagen et al. (2005). Bimodal size distributions often occur because of the influence of local, and sometimes distant, sources. For example Yi et al. (2001) document the influence of yellow sand advection on urban particle size distributions.

In a study of atmospheric deposition to oak forests along an urban-rural gradient, Lovett et al. (2000) found enhanced deposition rates in the urban areas. They proposed that the dust emissions from New York City are acting as an “urban scrubber” that removes acidic gases from the atmosphere and deposits them on coarse particles that dominate the urban size distributions.

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[13] The acronyms for the urban models used in the comparison are ECOSYS, EDEM2M, EXPUT2, PARATI, TEMAS-urban, and URGENT.

Deposition on Urban Surfaces

Urban surfaces represent complex combinations of different types of surface elements ranging from androgenic to natural. The dry deposition to specific surface elements in an urban area depends on a combination of the particle properties, airborne concentrations reaching the vicinity of the surfaces, local air-to-surface flux processes, and surface properties.

Historical interest in dry deposition in urban areas has largely been related to concerns about the potential effects of that process on buildings and monuments. Charola (1998) provided an extensive review of studies of acidic deposition on stone. In studies on 100-year-old marble monuments in the central portion of eastern United States, Dolske (1995) found that the amount of deposition varied with both the surface characteristics and orientation of the monuments. Studies of the deposition on a monument in Rome show how gravitational settling, ventilation, phoretic deposition, and inertial impaction determine the deposition rates and patterns that will be important for similar urban structures (Camuffo and Bernardi 1996).

Smooth surfaces will tend to have lower deposition rates per unit area than rougher surfaces, with the total deposition scaling with the area available for deposition. Such relatively small deposition rates are reported by Roed (1983) for vertical walls based on cesium-137 accumulated surface deposition from ambient aerosol deposition in Denmark. Although they reported nine samples for a brick wall with a range of wet/dry deposition velocities 0.003 to 0.07 cm s^{-1} (average 0.02 cm s^{-1}) and four samples for a plastered wall (0.014 to 0.085 cm s^{-1}; average 0.04 cm s^{-1}), only one sample was in an area sheltered from wet deposition and had a dry deposition velocity of 0.003 cm s^{-1}.

Nicholson (1987) reported similarly small deposition rates for deposition of ambient cesium-134 and cesium-137 particles to roof and building materials in England. Clay and concrete roof tiles had about 0.05 cm s^{-1} rates. Brick surfaces had smaller rates (up to 0.01 cm s^{-1}). Although the data set was small, the results were consistent with lower deposition velocities over smoother surfaces.

Because of the extended downwind distances from the sources for two sets of measurements, it would be reasonable to assume that the results represent deposition rates for background aerosol distribution with a peak diameter around 1 micron. The approach for determining deposition rates has several limitations. The result depends upon the assumption that weathering losses are sufficiently small as not to have removed significant amounts of deposited materials - an assumption that has some experimental support. In addition, the separation of wet and dry deposition materials is based on plume passage and/or surface exposure assumptions that are difficult to validate. As a result, these radionuclide deposition results will be applicable to only a small portion of the radioactive particle size distributions that will generated by explosive releases.

Although impaction and interception may be the major processes for these particles, the rates are sufficiently small that other phoretic effects may be important. Also, the differences in surface properties between vertical and horizontal surfaces may play a role. As previously noted, Chamberlain (1991) reports that stickier surfaces will tend to have greater deposition rates per unit area than less sticky surfaces. Kumar, Kumari, and Srivastava (2006) found about 50% difference in aerosol particle dry deposition to two different kinds of tropical leaves in an urban environment.

Total deposition on the various urban surfaces will scale with the deposition rates and available surface areas. Considering the total deposition per unit horizontal area, grass and trees will have relatively high deposition rates compared to smooth surfaces.

Considerable variability in deposition rates will occur because of the variability of exposures of surface elements to local air circulation. The more contaminated air that flows over a surface per unit time, the greater will be deposition rate. Analogous to the enhanced deposition on leeward sides of hills and waves, the leeward sides of urban structures will tend to have higher deposition rates. Modeling the variations in surface deposition will require considering interactions of surface roughness and local air circulation.

Urban Dry Deposition Modeling

The first step in developing improved dry deposition modeling capabilities for urban environments is to understand the urban dispersion processes. The DHS's Urban Dispersion Program is addressing the complex atmospheric dispersion processes in urban environments (Allwine 2005; PNNL 2006). The results to date include quantification of the flow regimes in different cities, publication of observational data that document unique features of plumes in an urban environment, and development of new air transport models that show promise in their ability to simulate the complex urban flow processes. These results are critical in understanding and developing prediction capabilities for where deposition of airborne materials will occur in an urban environment.

The second step is to develop improved formulations for simulating the local dry deposition processes as the plume passes over the various elements of an urban environment. To address the modeling of dry deposition in urban areas, some authors suggest an extension of the resistance-based formulations. For example, the NRPB (2001) review of dry deposition velocity estimation techniques for particles with a diameter of 0.1 to 10 microns suggests modifying the relationship for aerodynamic resistance for applications to higher canopies. An extended value for the reference height is used, based on twice the height of the vegetation.

Using large roughness scales of surface elements in an urban area relative to boundary-layer scales precludes a similarity-based approach to define aerodynamic resistance. The similarity-based approach requires spatially uniform conditions and a constant flux layer over the surface (i.e., homogeneous conditions), which are factors that do not exist in most urban areas. It is not reasonable to assume that a spatially uniform representative flux to the "urban surface" can be defined.

Although researchers have extended the surface layer Monin-Obukhov Similarity Theory to forest canopies using eddy-correlation methods to measure a vertical deposition flux (see, e.g., Hicks et al. 1982 and Wesely, Cook, and Hart 1983), questions exist as to the validity and appropriateness of such an extension to large-scale applications. Andreas and Hicks (2002) state that the largest eddies (those from mesoscale variations in cloud, vegetation, surface slope, etc.) usually violate the assumption of horizontal homogeneity that the Monin-Obukov Similarity Theory requires. Thus, the extended spatial scales that would be required to apply similarity theory to urban areas may preclude such an approach.

Because a large fraction of material from a number of types of potential events may be in the form of larger particles, urban dry deposition formulations need to address where larger particles will tend to deposit in the complex urban environment. Air circulations within the urban canopy will define the mean flow that will carry these particles. These particle trajectories will tend to intersect vertical structures that may be the receptor surfaces for some types/sizes of settling particles. Areas of stagnant air or closed circulation allow extended time for gravitational settling.

Formulations for an urban model for dry deposition should include parameterizations of the range of processes discussed in this report. Extension of the current similarity-based resistance formulations to the urban scale, although suggested in the literature, is not a valid approach. A framework for a generalized approach presented below is proposed as a means of extending current formulations to an urban environment.

3.4 Formulations

This section documents the relationships that are available for developing formulations to simulate various dry deposition processes. Currently deployed models incorporate some subset of these relationships. The implementations in many cases involve simplification of relationships. For many models, immediate model dry deposition improvement will be possible by incorporation of applicable relationships.

Hanna, Briggs, and Hosker (1982), Friedlander (2000), and Seinfeld and Pandis (1998) provide summaries of the basic formulations for the various dry deposition processes; Wesely and Hicks (2000) provide a review of dry deposition knowledge in the context of acid-rain modeling; and Chamberlain (1991) and Baklanov and Sorensen (2001) provide radionuclide-specific deposition formulations. In recent years, dry deposition modeling efforts have extended formulations for specific applications and provided better supporting experimental data.

As noted above, dry deposition is known not to be the result of a single process but the result of many processes. Deposition velocity varies with particle size, wind speed, surface roughness, turbulence levels, and relative humidity (related to the particle size and density). Depending on the specific situation, dry deposition also can vary with the physical, chemical, and biological properties of receptor surfaces as well as surface orientations relative to the local wind circulations. Gradients of scalar (heat and moisture) and electrical properties between the surface and air can affect dry deposition rates. To simulate these dependencies, formulations are needed for the processes potentially controlling dry deposition.

Dry deposition is the result of the combined effects of many processes, some of which are sequential and others concurrent. In-plume processes such as transport, diffusion, and decay control the near-surface concentrations. In addition to eddy diffusion, gravity can concurrently move the particles through the turbulent atmosphere to the vicinity of the surface. Once there, processes in the quasi-laminar layer deliver particles to the surface. Whether a particle is retained on, or rebounds from, the surface depends on the particle and the receptor surface properties.

3.4.1 Gravity

Gravitational settling is a key process for particle deposition, particularly for larger particles, as can be seen in the resistance approach formulations containing the gravitational settling velocity, v_s, given above in Section 3.2.

In practice, an aerodynamic (equivalent) diameter is often used to account for different shapes, roughnesses, and aerodynamic drags of particles moving though a gas. The aerodynamic diameter is the diameter of a unit-density sphere that will have the same gravitational-settling velocity as the particle. The aerodynamic diameter concept is a useful way to incorporate the corrections for non-spherical particles.

Stokes Equation

Gravitational settling dominates the deposition of large particles. Depending on their density, the dry deposition of particles with radii as small as 1 to 5 microns can be strongly affected by gravitational settling (Baklanov and Sorensen 2001; Hanna, Briggs, and Hosker 1982).

The settling velocity for particles, v_s, in cm s^{-1} can be computed using a modified form of Stokes Law (Hanna, Briggs, and Hosker 1982):

$$v_s = S_f \cdot 2r^2 g(\rho_p - \rho_a)/9\mu \tag{21}$$

where S_f = the Cunningham correction factor[14], dimensionless
ρ_p = the particle density, g cm^{-3}
ρ_a = the air density, g cm^{-3}
μ = the dynamic viscosity of air, g cm^{-1} s^{-1}
r = the particle radius, cm
g = the gravitational constant, cm s^{-2}

Hanna, Briggs, and Hosker (1982) suggest a typical value of 1.8×10^{-4} g s^{-1} cm^{-1} for μ.

Extensions to Stokes Equation

Non-spherical particles will fall at slower rates. Hanna, Briggs, and Hosker (1982) provide a summary table of dynamical shape factors. For materials with equivalent densities, the change in settling velocities is less than 30% for ellipsoid and cylinder shapes. Engineering handbooks are also available with formulations for accounting for non-spherical effects. To account for shape effects, an aerodynamically equivalent diameter is frequently used to define the settling velocity of a particle.

For particles with radii less than about 1.8 micron, the airflow around the particle can be considered laminar. Baklanov and Sorensen (2001) suggest the use of Stokes Law with a Cunningham correction factor, S_f, for small particles with radii <0.5 micron:

$$S_f = 1 + \frac{\lambda}{r}\left[a_1 + a_2 \exp\left(-\frac{2a_3 r}{\lambda}\right)\right] \tag{22}$$

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[14] The Cunningham correction factor, or Cunningham slip factor, accounts for molecular slip that occurs when the particle is the same size as the distance between the gas molecules. This factor is significant for particles with diameters of 1 micron or less. An additional correction factor is needed when the aerodynamic diameter is different from the average diameter of the particles.

where λ is the mean free path of air molecules (= 5.53 10^{-5} m) and the constants, a_1, a_2, and a_3, as in Equation (22), have the values of 1.257, 0.40, and 0.55, respectively. An equivalent relationship is used by ADOM (USEPA 1994) and ISCST (USEPA 1995), based on computing a Cunningham correction factor, S_f;

$$S_f = 1 + \frac{2x_2}{2*10^{-4}r}\left[a_1 + a_2 \exp\left(-\frac{2a_3 r}{x_2}\right)\right] \tag{23}$$

where x_2, a_1, a_2, and a_3 are constants with values of 6.5 x 10^{-6}, 1.257, 0.40, and 0.55 x 10^{-4}, respectively, for values of r expressed in microns.

Larger Particles

Stokes Law is known to be invalid for particles with radii greater than 1.8 micron falling in a turbulent regime. An early effort by Van der Hoven (1968) provides a graphical solution for larger spherical particles for particles up to 1000 microns.

Baklanov and Sorensen (2001) stress that proper characterization of deposition for these particles will play an important role in assessing local-scale effects where larger particles tend to be a major fraction of the airborne plume. An iterative procedure is suggested by Baklanov and Sorensen (2001), based on work by Näslund and Thaning (1991), for determining the terminal settling velocity for larger particles, which they apply to radii of 100 microns:

$$\begin{aligned}
&\frac{dw_p}{dt} = \left(w - w_p\right) f(V) - \beta g \\
&f(V) = \frac{3\rho}{8r\rho_r} V c_d \\
&V = \left[\left(u - u_p\right)^2 + \left(v - v_p\right)^2 + \left(w - w_p\right)^2\right]^{0.5} \\
&c_d = \frac{24}{\mathrm{Re}}\left[1 + 0.173(\mathrm{Re})^{0.657}\right] + \frac{0.413}{1 + 16300(\mathrm{Re})^{-1.09}}
\end{aligned} \tag{24}$$

where V = the relative velocity of particles
u, v, w, u_p, v_p, w_p = the air and particle velocity components
β = a buoyancy effect parameter ($\beta=(\rho_p-\rho)/\rho_v$)
c_d = the drag coefficient
Re = the Reynolds number (Re=2Vr/v)
υ = kinematic viscosity of air

Baklanov and Sorensen (2001) use a resistance approach based on Equation (8), incorporating Equations (12), (13), and (14) to simulate deposition processes for the larger particle sizes. Although their simulations of deposition velocity agree relatively well with experimental data (see Figure 3.6), there was a tendency to underpredict deposition velocities. This discrepancy is addressed in more detail below in the discussion of impaction processes.

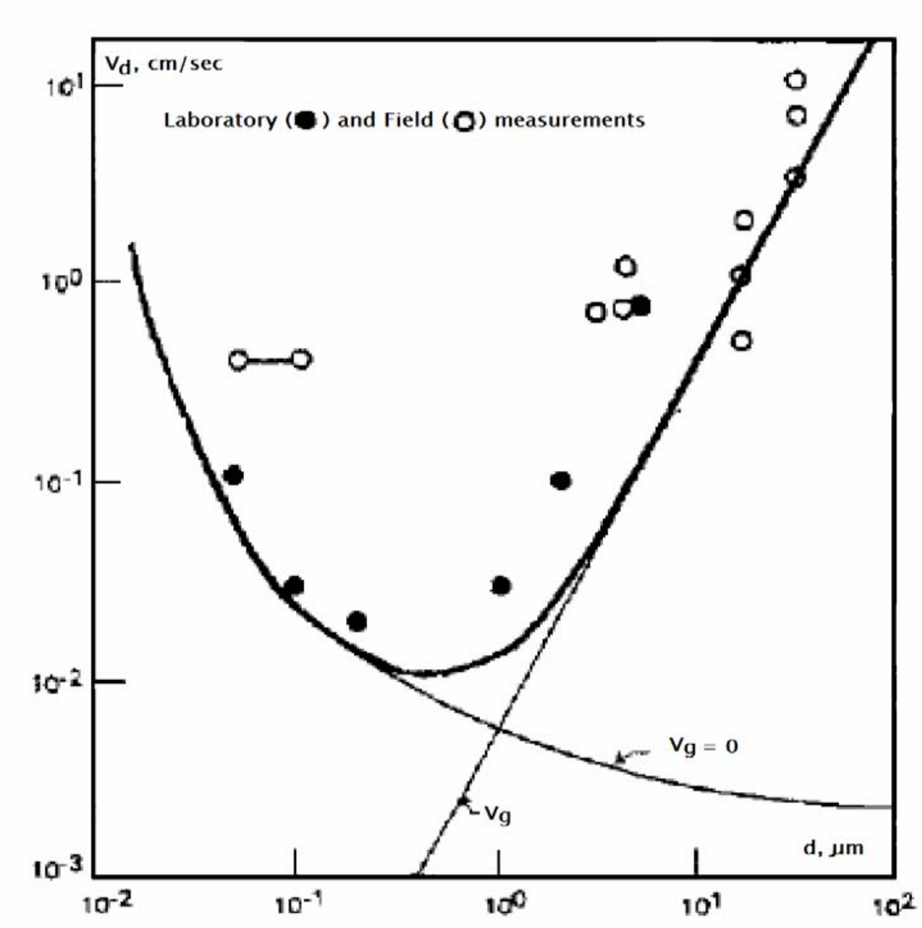

Figure 3.6. Comparison of Modeled and Experimental Dry Deposition Velocities[15]

Tilted Plume Model

Gravity acts on the airborne particles in the plume. As particles fall from gravitational settling, the height of the plume for these particles is effectively lowered as the plume moves downwind. Models typically compute the progressive decrease in the height of the plume for different particle size ranges, d, using an equation such as:

$$h(d, t) = h(d, x_o) - v_s(d)\, t \qquad (25)$$

where h(d,t) is the height of the plume as a function of time, $h(d, x_o)$ is the initial height of the plume, and t is the time. For larger particles with settling velocities greater than 100 cm s^{-1} (which will not be significantly affected by ambient atmospheric turbulence), a ballistics trajectory formulation using mean wind speed and Equation (25) can be used to define the distances that particles will fall to the surface. For smaller particles, dispersion will tend to result in deposition peaks at closer distances than predicted by Equation (25).

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[15] Reprinted from Baklamov and Sorensen (2001) by permission from Elsevier.

On average, the amount of material potentially deposited by gravitational settling is the material falling though a horizontal plane over the area. Deposition of particles by gravitational settling, however, does not necessarily occur only on horizontally oriented surfaces. Particles falling in the mean flow of air can potentially deposit onto any surface by the diffusion, impaction, or interception processes.

For the larger particles, the distances to peak deposition will be mainly a function of the particulate settling velocities combined with the effect of local circulations. Assuming a simple trajectory model or tilted plume model, the downwind deposition distance can be estimated as the product of wind speed and the travel time (i.e., h(d,t) = 0 in Equation [25]). The travel time is the initial height of the particle divided by the settling velocity. Downwind deposition distances thus computed are plotted as a function of deposition velocity for different combinations of release height and wind speed (Figures 3.7 through 3.10).

The distances for peak deposition of very large particles are shown over the range from close distances, at which resolutions of deposition patterns are impossible with current air dispersion models, to greater distances that are handled well by current air dispersion models.

Figures 3.7 through 3.10 illustrate that the particles from an event involving an RDD can potentially have peak impacts over a wide range of downwind distances.

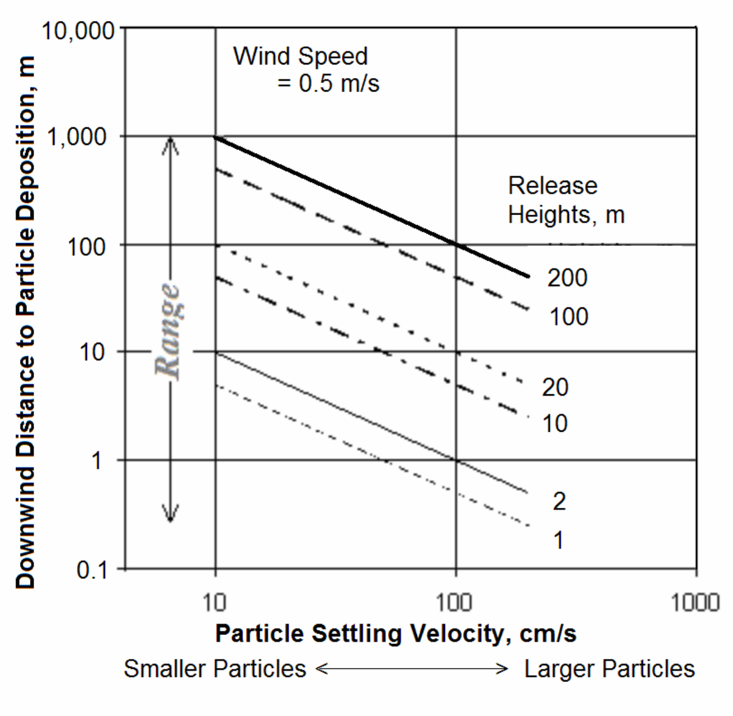

Figure 3.7. Projected Distances to Peak Deposition by Gravitational Settling for Multiple Release Heights (Low Wind Speed) for Settling Velocities Associated with Large Particles

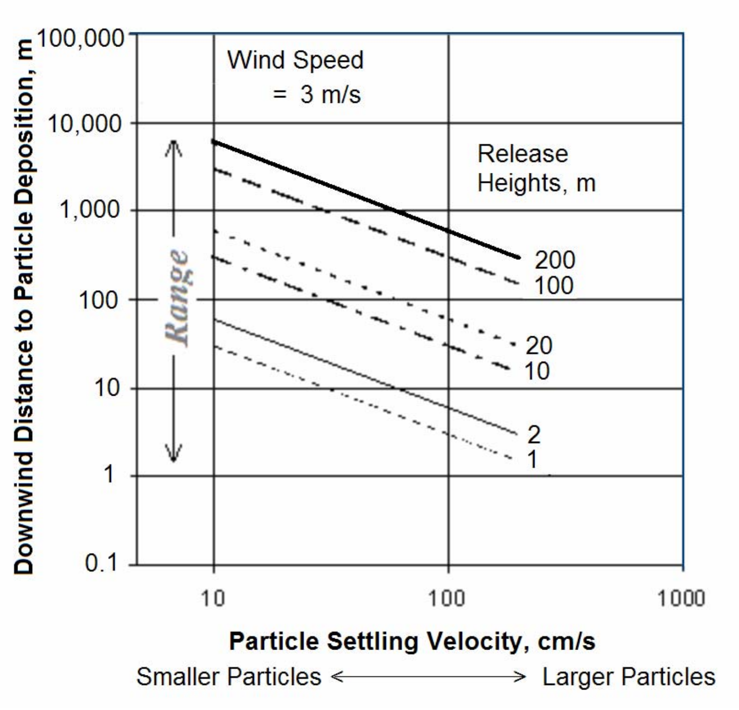

Figure 3.8. Projected Distance to Peak Deposition by Gravitational Settling for Multiple Release Heights for Settling Velocities Associated with Large Particles (Average Wind Speeds)

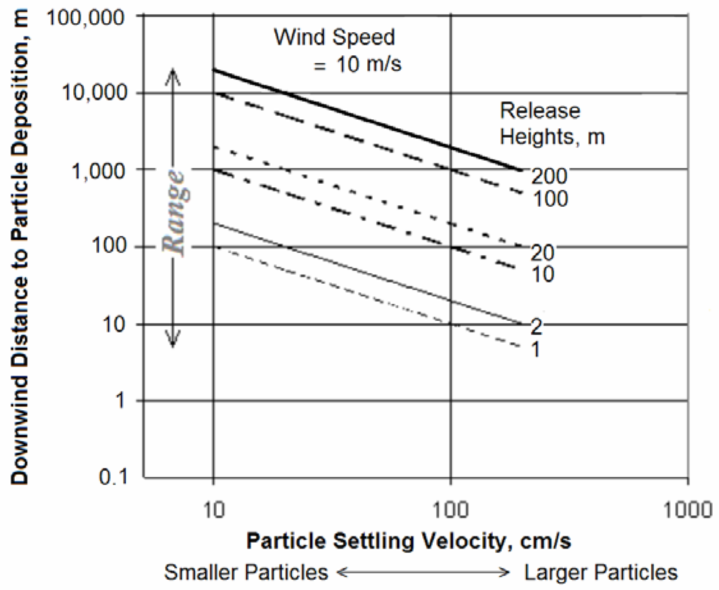

Figure 3.9. Projected Distance to Peak Deposition by Gravitational Settling for Multiple Release Heights for Settling Velocities Associated with Large Particles (High Wind Speeds)

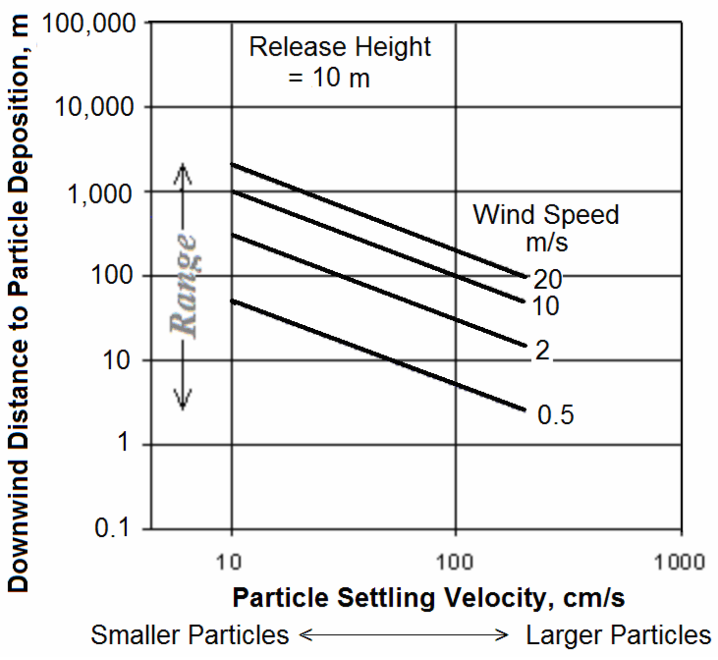

Figure 3.10. Projected Distance to Peak Deposition by Gravitational Settling for 10-m Release Height over a Range of Wind Speed Conditions for Settling Velocities Associated with Large Particles

3.4.2 Eddy Transport in the Surface Layer

The atmospheric dispersion modules of airborne plume models simulate the spatial and temporal characteristics of plume concentrations. The dry deposition module simulates the flux processes on underlying surfaces.

Figure 3.1 illustrates the vertical structure of winds near the earth's surface. Filtration theory used in aerosol physics addresses steady-state fluxes to individual surfaces such as the smooth flat surface shown on the left in Figure 3.11. For atmospheric fluxes to rough flat surfaces, a similar concept is applied based on mean flow properties, as illustrated on the right side of Figure 3.11. As noted, a "surface layer" is a constant-flux layer over the surface whose properties are largely controlled by the local surface fluxes.

Under such conditions, field studies have verified that the mean profiles of local micrometeorological properties (temperature, moisture, and wind) are exponential, or nearly exponential (Sorbjan 1989). Depositing contaminants are expected to have a similar shape as these properties (Droppo 1985). A dry deposition module needs to address the surface-specific processes that define the properties of the atmospheric surface layer. For a material to deposit on a surface, the material must move to the vicinity of the surface by eddy transport. Currently, resistance-based dry deposition formulations consider the movement of a contaminant through two layers: a fully turbulent layer, typically several meters in depth, where vertical fluxes are nearly constant, and a thin quasi-laminar layer immediately over the receptor surface.

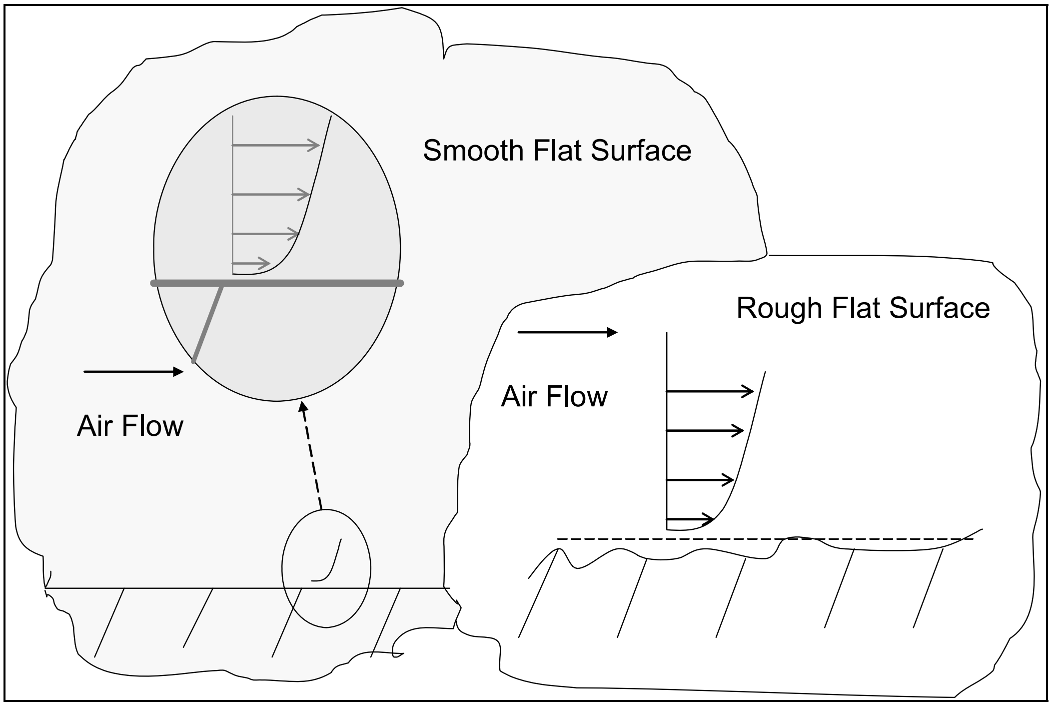

Figure 3.11. Wind Profiles over Smooth and Rough Surfaces

This section discusses formulations for the fully turbulent, constant-flux layer, where eddy transport will control the rate at which particles moving passively with the air movements can be vertically transported.

Surface Layer Aerodynamic Resistances

Relationships for aerodynamic resistances are based on surface layer parameterizations from Monin-Obukhov Similarity Theory. Various surface layer parameterizations have been developed (Businger et al. 1971; Dyer 1974). These parameterizations have been used to develop formulations for aerodynamic resistances. For example, using the coefficients suggested by Dyer (1974), the formulation for the aerodynamic resistance used in the ISC model (USEPA 1995) based on Byun and Dennis (1995) is expressed as:

neutral (z/L=0)

$$r_a = \frac{1}{ku^*} \ln\left(z_d / z_o\right) \tag{26}$$

stable (z/L>0)

$$r_a = \frac{1}{ku^*} \left[\ln\left(z_d / z_o\right) + 4.7\, z_d / L\right] \tag{27}$$

unstable (z/L<0)

$$r_a = \frac{1}{ku^*}\left[\ln\frac{\left(\sqrt{1+16\left(z_d/|L|\right)}-1\right)\left(\sqrt{1+16\left(z_o/|L|\right)}+1\right)}{\left(\sqrt{1+16\left(z_d/|L|\right)}+1\right)\left(\sqrt{1+16\left(z_o/|L|\right)}-1\right)}\right] \tag{28}$$

where u^* = the surface friction velocity, cm s^{-1}
k = the von Karman constant (0.4)
L = the Monin-Obukhov length, m
z_d = deposition reference height, m
z_o = the surface roughness length, m

The friction velocity and roughness length are situation-specific parameters that measure the ambient turbulence intensity and local surface roughness, respectively. The Monin-Obukhov length is a scaling parameter representing the dominant scale of eddy transport.

When the airflow encounters a rougher surface, a new deeper turbulent surface layer tends to develop. When the airflow encounters a smoother surface, the surface layer tends to decay into a smaller surface layer. Aerodynamic resistance will tend to be smaller than the equilibrium value in developing surface layers and higher in decaying surface layers. Formulations that utilize an aerodynamic resistance applicable to uniform surfaces that are applied to areas that have mixed surface types implicitly assume that the under- and over-prediction in decaying and developing surface layers, respectively, will largely cancel each other.

Near-Surface Concentration Depletion

Dry deposition formulations simulate the flux of material from some dry deposition reference height to the underlying surface. Ideally, the height should be high enough to have a minimal effect from the near-surface concentration depletion. Simple dry deposition formulations using a single value for the dry deposition velocity tend to use an arbitrary near-surface reference height, such as 1 meter. The near-surface concentration-depletion profile can significantly affect concentrations at such a height, and Horst (1982) provides concentration-depletion curves reflecting the effects of deposition that can be applied to such models.

For resistance-based models, a dry deposition reference height is the height from which the total local resistance to deposition is computed. An appropriate value of this height will be at some location near the top of the local constant surface-flux layer such that the total resistance will reflect the processes though this layer. If too low a height is selected, then the resistance will be underestimated. If too high a height is selected, then resistance may be computed to an inappropriate height above the local surface layer. This approach has the advantage of greatly simplifying the required computations. The Horst (1982) curves also are valuable information for those wanting to understand the effects of potential exposures from air concentrations near a surface.

Newer formulations that use a resistance approach to define a deposition velocity use a greater height (typically 10 m). The idea is to use a reference height that is sufficiently high as to have only a minimal decrease in the concentration from the deposition flux. However, this approach has proven to be problematic in terms of computations near the source, particularly for elevated sources.

The use of one reference height for all applications (whether a low value such as 1 m or a high value such as 10 m) leads to conceptual problems for applications over progressively rougher surfaces that are directly related to the inapplicability of the deposition formulations to such surfaces.

Surface Characterization

The Baklanov and Sorensen (2006) formulations took an important step forward by including use of land-use classification data to characterize the dry-deposition related properties of the underlying surfaces. Although they used an older version with a relatively simple land classification scheme, they point out that a newer version of the Danish Meteorological Institute's numerical weather prediction model uses an expanded classification scheme with 20 land-based classes, which will be more appropriate for computing deposition fluxes. They note that an additional update added an urban area class. Such finely delineated physiographic data are needed to be able to simulate the dry deposition rates.

Surface roughness lengths are a key input to resistance-based dry deposition models. The experimental procedure for determining local roughness lengths from measured micrometeorological profiles is not a practical approach for defining local roughness length patterns over extended regions. MacKinnon et al. (2004) showed that local roughness lengths for some surfaces can be reasonably approximated based on the physical aspects of the surfaces. However, there are limitations in the use of the current relatively rough classification schemes not specifically designed for that purpose to define local surface roughness.

3.4.3 Deposition Layer Processes

The atmospheric dispersion modules of airborne plume models define spatial and temporal characteristics of plume concentrations. The dry deposition module simulates the flux processes to underlying surfaces. The degree of integration of these two modules varies between different models and types of models. These modules tend not to be closely linked, perhaps because the dispersive properties of the boundary layer at a specific location are largely controlled by the upwind surface fluxes, whereas the deposition processes are controlled by the local surface fluxes at the location.

Brownian Diffusion

The main process for molecular diffusion is typically Brownian diffusion. Chamberlain (1991) provides an equation for the transport of small particles by Brownian diffusion:

$$\mathrm{v}_{\mathrm{sm}} = 0.66 u^{0.5} v^{-1/6} L_c^{-1/2} D^{2/3} \tag{29}$$

where v_{sm} = transfer velocity for submicron particles, m s^{-1}
u = wind speed, m s^{-1}
υ = kinematic viscosity of air, m^2 s^{-1}
L_c= characteristic length, m
D = Brownian diffusivity, m^2 s^{-1}

The EPA (1995) defines the Brownian diffusivity of a contaminant in cm s^{-1} as:

$$D_B = 8.09 \times 10^{-10} \left[\frac{T_a S_f}{d_p} \right] \tag{30}$$

where T_a is air temperature (degrees K), d_p is the particle diameter in microns, and S_f is the Cunningham correction factor (defined above).

Particle Impaction

As the size and/or density of particles increase, they tend to follow the streamlines of the local airflow (i.e., turbulent eddies) less closely. Larger and heavier particles fall relative to the mean motion of the surrounding air at a velocity determined by a gravitational settling rate.

A minimum in dry deposition rates occurs for a middle range of particles too large for effective deposition by molecular diffusion and too small for gravitation and impaction. This middle range, with radii of 0.1 to 1 micron, is the accumulation mode range. In the free atmosphere, a balance between the selective removal of larger particles by dry deposition processes and particle growth processes by coagulation and gas condensation on particles appears to be maintaining this range of particles.

Chamberlain (1991) provides the following relationship for the impaction velocity, v_{impact}:

$$v_{impact} = C_i / u_1 \tag{31}$$

where C_i is the efficiency of impaction and u_l is the free air stream velocity.

This relationship for impaction is analogous to the equation for a wind drag coefficient. According to Chamberlain (1991), aerodynamic theory indicates that C_i will be a function of the Stokes number, St:

$$St = S_p/L_c \tag{32}$$

where S_p is the stopping distance of the particle[16], and L_c is a characteristic dimension of the surface element. Chamberlain (1991) provides a relationship for particles in the range of 1 to 50 microns and wind speeds, u, of less than 5 m s^{-1}:

$$S_p = v_g\, u/g \tag{33}$$

where g is the acceleration due to gravity, m s^{-2}, and v_g is the terminal velocity of the particles as computed by Stokes Law, or a modified form of Stokes Law. Capture efficiency of the impacting particles defines the actual deposition rate.

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[16] Stopping distance is a measure of the ability of a particle to follow directional changes in flow. It is computed as the product of the initial particle velocity and a relaxation time for the particle to reach 1/e (~37%) of its velocity adjustment to an incremental change in the surrounding fluid flow velocity.

The properties of the sublayer for particle transport may be estimated by assuming analogous properties for mass and heat transfer. Owen and Thomson, as cited in Chamberlain (1991), characterized mass and heat transfer from rough surfaces in terms of a sublayer Stanton number, B, defined by:

$$\frac{u(z)}{v_s(z)} = \frac{u(z)}{u^*} \left(u(z) / u^* + B^{-1} \right) \tag{34}$$

where $B^{-1} = 1/k \ln(z_o/z_v)$
z_v = roughness length for mass transfer

For surfaces with bluff roughness elements such a plowed field, the bluff elements contribute to the shearing stress and B^{-1} is increased. Chamberlain, Garland, and Wells (1984) found that transfer correlated well with:

$$B^{-1} = 7.3 \, Re^{*0.25} \, Sc^{0.5} - 5 \tag{35}$$

where re* = roughness Reynolds number = $u^* z_o/\upsilon$
Sc = Schmidt number = υ/D
υ = kinematic viscosity of air, $m^2 s^{-1}$
D = molecular diffusivity, $m^2 s^{-1}$

This relationship is based on lead-121 and iodine-131 data.

Chamberlain (1991) gave results that showed the relationship between the Stanton number and the friction velocity for artificial grass and rough glass surfaces. The Stanton number tends to increase with increasing friction velocities but appears not to have a dependence on z_o for surfaces with fibrous roughness elements. An unexplained result is the observation of larger Stanton numbers for the surface with a smaller z_o (i.e., rough glass vs. artificial grass). The discrepancy indicates that z_o alone does not totally characterize the surface properties that control the deposition to a surface. This result suggests that other receptor surface characteristics, such as surface area, surface properties, and surface structure, can be also important.

Deposition processes in the accumulation size range of 0.1 to 1 micron radius are not completely understood. Field studies have addressed sulphate particles (~0.5 micron) in the urban and suburban atmospheric environments. Two studies quoted by Chamberlain (1991) give consistent, relatively small estimates of deposition velocities (0.1 and 0.07 cm s^{-1}).

As noted above, the review of dry deposition models by the EPA (USEPA 1994) concluded that only a few models were consistent with experimental results over the range of 0.1 to 20 microns diameter. The better models, specifically Sehmel and Hodgson (1978) and Pleim, Vernkatram, and Yamartino (1984), showed close agreement for smaller particles (0.1 to 1 micron diameter). For larger particles (10 to 100 microns diameter), Pleim, Vernkatram, and Yamartino (1984) showed better agreement than Sehmel and Hodgson (1978).

For small particles moving though the quasi-laminar layer, the flux, F, is a combination of gradient- and gravity-driven transport:

$$F = K\frac{dC}{dz} + v_g C \tag{36}$$

where K = turbulent diffusivity
dC/dz = concentration derivative with height
v_g = settling velocity
C = air concentration

For larger particles, specifically those with stop distances on the same order of magnitude as, or greater than, the thickness of the quasi-laminar layer, the flux is the result of impaction from inertial and gravity-driven transport mechanisms:

$$F = (v_d)_b C = v_i C + v_g C \tag{37}$$

where v_i is an inertial velocity and $(v_d)_b$ is the local deposition velocity for flux to the surface. Kim, Kalman, and Larson (2000) compute the flux for a given particle size as the sum of the gradient, inertial and gravity terms:

$$F = K\frac{dC}{dz} + v_i\, C + v_g\, C \tag{38}$$

The approach used by Pleim, Vernkatram, and Yamartino (1984) to parameterize the deposition-layer resistance terms is modified to include Slinn's (1982) estimate for the inertial impaction term. The resulting deposition layer resistance is:

$$r_s = \frac{1}{\left(Sc^{-2/3} + 10^{-3/St}\right)u^*} \tag{39}$$

where Sc = the Schmidt number ($Sc = \upsilon/D$), dimensionless
υ = the kinematic viscosity of air (approximately equal to 0.15 $cm^2\ s^{-1}$)
D = the Brownian diffusivity, $cm^2\ s^{-1}$, of the pollutant in air
St = the Stokes number [$St = (vg/g)(u*2/\upsilon)$], dimensionless
g = the acceleration due to gravity, 981 $cm\ s^{-2}$

In Equation (39), the deposition rate for small particles is controlled by effects of Brownian motion incorporated in the first term involving the Schmidt number. The deposition rate for intermediate-sized particles (with diameter range of 2 to 20 microns) is controlled by inertial impaction incorporated in the second term involving the Stokes number.

The improved parameterizations of particle impaction processes show a significant improvement in the ability to simulate observed dry deposition rates.

As a result, these relationships represent important updates for models using older parameterizations for the dry deposition of larger particles.

Phoretic Effects

Detailed formulations for phoretic processes for a variety of surface orientations are available (Friedlander 2000). In atmospheric dispersion models, phoretic and related Stefan flow effects on dry deposition are generally assumed to be small, based on their normally very small contributions to overall deposition fluxes (Hicks 1982). However, for particles in the accumulation mode range of 0.1 - 1.0 micron diameter, for which other deposition processes are relatively ineffective, these effects may not always be negligible. Rather than including detailed formulations, dry deposition models generally include an empirical minimum limit for the magnitudes of deposition velocities. For example, the ISC formulation adds a phoretic term to the deposition velocity modeled from diffusion, impaction, and gravitational settling; a constant value of 0.01 cm s^{-1} is added to the otherwise modeled deposition velocity to represent combined phoretic effects.[17]

It is likely that models that add improved capabilities to simulate near-field deposition processes will need to incorporate more detailed formulations for phoretic processes. The initial plume generated by an explosion will have extreme thermal and electrical properties. The extent of thermal gradients depends on the generated heat. The electrical fields are largely the result of the forced movement of charged particles. As a result, strong thermal and electrical gradients have the potential of greatly enhancing the near-field rates of movement of particles and, thus, the initial rates of deposition. The deposition of even relatively large particles may be affected by extreme electrical and thermal gradients.

3.4.4 Surface Retention

For smaller particles, the theory of aerosol dynamics indicates that the surface collection efficiency should be near 100%. Van der Waals forces, acting on a particle over a flat surface, tend to capture and hold small particles on that surface (Friedlander 2000).

For larger particles, the kinetic energy of the impact as well as particle/receptor surface properties determine if retention will occur. The failure to adhere is also referred to as rebound. Striking a surface at low-impact velocities, a particle will tend to adhere to that surface. As the impact velocity increases, the particle may bounce off if the surface kinetic energy is sufficiently large to escape the attractive forces at the surface (Friedlander 2000).

Wind tunnel test results summarized by Chamberlain (1991) clearly showed the effect of these influences. As the kinetic energy of the particle increased, the collection efficiency decreased by up to an order of magnitude. Deposition of Lycopodium spores and ragweed pollen to moist sticky surfaces (wheat stems) started with 70 to 90% collection efficiencies that decreased to about 10%. The deposition of fly ash particles on non-sticky dry surfaces (steel fibers) started with about 30% and decreased to about 6% with increasing kinetic energy. The deposition of polystyrene particles on pine needles started with nearly 90% collection efficiency, with the efficiency dropping quickly by more than an order of magnitude as the kinetic energy of the particle increased. These results indicate that wet/sticky surfaces will have greater adhesion than dry/non-sticky surfaces.

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[17] An alternative formulation would be to use a minimum value for the modeled deposition velocity based on the results of experimental studies.

3.5 Summary and Discussion

This review provides a summary of the current state of dry deposition formulations and identifies model improvements for air-dispersion models used by DHS in the management and mitigation of potential threats. Although the focus is on RDD devices, the review applies to other types of threats that involve the release of toxic materials to the atmosphere.

The locations with the greatest deposition of material following an RDD event are of primary concern in responding to the emergency. In addition to the ambient meteorological conditions, the downwind pattern of particle concentration on surfaces will be largely a function of the size/density particle distribution, initial release height, and ambient meteorological conditions.

For smaller particles, the ambient turbulence intensity combined with wind speed will largely control the distance that an elevated plume must be vertically dispersed to reach the underlying surface. A near-source model resolution limitation of most current air-dispersion models is that they start their simulation of dry deposition rates only at some minimum distance downwind. These models will miss peaks closer than this distance. For larger particles, the deposition patterns will mainly be a function of the wind speed and particle deposition velocities. For gravitational settling, the range of potential distances to peak deposition is from near-source for a near-surface plume out to regional-scale distances for an elevated plume.

Threats addressed in this review are potential events that may have significant health or environmental impacts through atmospheric exposures or a pathway linked to the atmospheric exposures. Atmospheric exposures occur both as the result of airborne concentrations and deposited materials.

Current formulations for dry deposition in air-dispersion models are largely based on a resistance approach. Current models use various implementations of that approach, and the main improvements identified in this review are based on developing a new approach that overcomes the limitations of the resistance-based formulations.

3.5.1 Near-Source

The near-source plume characterization is the most critical factor in obtaining representative simulations of potential near-field deposition patterns, which depend heavily on how the particles are dispersed by the explosion and carried by subsequent air motions, including plume rise. If a large fraction of the released material is in the form of larger particles, gravitational settling will dominate the near-field deposition. Although used in many models, the resistance approach based on similarity theory is not applicable for the non-stationary conditions that occur at locations near the source.

The current development of computational fluid dynamics (CFD) models provides the possibility of future capabilities that will define flow - and, hence, deposition patterns - on a local scale that is largely impossible in currently deployed air dispersion models. These models show considerable promise in advancing our understanding of urban flow processes. Although there are questions whether the CFD models will ever be cost-effective or practical for use in incident response, the development of CFD capabilities is expected to lead to significant improvements in the parameterizations used in models for urban applications.

3.5.2 Regional and Far-Field

Winds carry a plume from near-source to regional and far-field scales. A major modeling challenge is the operational linkage of near-source models to the regional and far-field models. This modeling issue was addressed in the development of SPRAYTRAN, which is based on a geographical information system (GIS), that links the AGDISP near-field drift model with CALPUFF, a state-of-the-art regional-scale atmospheric dispersion model (USEPA 2005a).

For dry deposition computations in most currently implemented models, a layer with a constant vertical flux of contaminant is assumed. With this assumption, an atmospheric (or aerodynamic) resistance is computed based on Monin-Obukov surface-layer similarity relationships. The literature indicates that this type of formulation is good for relatively low and uniform surface covers.

Application of resistance models to urban areas is problematic. Although the resistance formulation should not be conceptually applied to rougher surface elements such as are encountered in an urban environment, current models extrapolate the resistance models to such surfaces (NRPB 2001; Whelan et al. 1992). A revised approach is needed that will address the actual processes that will control dry deposition rates for various types of urban areas. Depending on the characteristics of the local surface elements, two urban areas with exactly the same overall surface roughness can have very different rates of total deposition. A major factor causing such discrepancies is the extent of highly effective deposition surfaces (grass, trees, etc.) that are only minor contributors to the overall surface roughness.

3.5.3 Field Studies of Total Deposition Rates to Urban Areas

The dry deposition resistance concept is useful in the region and far-field to characterize and understand the relative importance of different processes. The atmospheric resistance is generally not the dominant term. At downwind distances where the plume is relatively well mixed near the surface, eddy dispersion processes will define the maximum rate that fine particles (i.e., those small enough to be passively dispersed by these eddies) can be deposited to the local surfaces. The turbulent dispersion is normally not limiting because the eddy diffusion resistance is normally much smaller than the surface layer resistance of fine particles.

Studies have shown that a significant part of dry deposition is from larger particles even at region and far-field distances. Impaction and gravitation processes work together to deposit larger particles formed by coagulation and gas condensation.

To cover the range of processes that may dominate dry deposition, formulations need to account for the action of each of the dry deposition processes on the elements of the local surface cover. For example, foliated versus leafless vegetation canopy studies indicate that for particle deposition formulations to have general applicability, they must incorporate separate terms for the diffusion and impaction components of surface resistances.

3.5.4 Particle and Surface Properties

Formulations for modeling dry deposition fluxes need to address the particle-size dependent processes controlling dry deposition. The capabilities of models addressing a subset of these processes can be significantly improved by expanding the formulations given above to address the following ranges of particles potentially associated with an event:

Very small particles (radii <0.05 micron). Molecular diffusion processes are dominant. Formulations for characterizing fluxes of these particles are normally based on Brownian motion. Although deposition from molecular diffusion processes is relatively well understood, the specific roles of thermal flux, concurrent mass fluxes, and electrical attraction are largely undefined.

In an event, the very small particles will deposit quite rapidly either to the nearby surfaces or other particles in the plume. Because of the short time-scale, these deposition rates are normally not considered in air dispersion models but rather modeled as part of the plume initialization.

Submicron particles (accumulation mode size). These are the particles that tend to remain airborne after smaller particles are deposited by diffusion and larger particles are deposited by gravitation/impaction processes. Because these particles do not have efficient deposition mechanisms, it is assumed they will be carried extended distances.

A minimum deposition rate typically occurs at some midpoint in the accumulation mode size. Neither diffusion nor gravitation/impaction is very effective for these particles, and the relative importance of the processes controlling dry deposition for this mode of particles is not well understood. Molecular, thermal, and mass diffusion processes, electrical attraction, gravitation, and coagulation may all, or in some combination, be acting to define the minimum deposition velocities observed for this mode.

Because situation-dependent influences involving a range of potentially dominant processes appear to be variously defining the minimum deposition velocity for this range, it is impractical to include detailed formulations. Instead, the current practice is to define a minimum value of deposition velocity for this range that represents deposition processes that are not explicitly included. This practice should be adequate for air dispersion models, except perhaps for near-source plume initialization models.

Small particles (radii >0.05 to 5 micron). This range includes the accumulation mode. The currently deployed dry deposition models provide surface-specific deposition estimates that agree relatively well with data from field and wind tunnel experiments. Stokes Law can be used to compute settling velocities. Implementation should include shape and size corrections to the Stokes equation.

Although the Cunningham factor for the smaller particles tends to provide corrections to relatively small settling velocities, these corrections can potentially be important for defining the minimum deposition velocity for the accumulation mode.

Intermediate particles (radii >5 to 10 microns). Formulations for particle deposition need to address the range of situations from large surface elements with slow diffusion-driven rates to surfaces with a fine structure with faster impaction/interception driven rates. Current formulations that consider a combination of diffusion, impaction, and gravitational settling for specific types of applications should be incorporated into models to improve the estimates of deposition rates to the specific surfaces.

Larger particles (radii >10 microns). New formulations are available for significantly improving dry deposition computations for this range of particles. These improved formulations account for the importance of eddy inertial deposition efficiency in the deposition of these particles. Based on recent literature, older formulations, which are deployed in many models, are significantly underpredicting dry deposition rates for particles in this size range.

Very large particles (having sufficiently large size and density such that settling velocity >100 cm s^{-1}*).* Air dispersion models should incorporate particle trajectory-based modules accounting for reduced influences of atmospheric turbulence. This update represents a significant improvement for models that assume that all particles in the release are dispersed at the same rate.

Urban canopies are a composite of many different types of surfaces with roughness elements too large to apply current dry deposition models. A dry deposition model is needed for urban applications that overcomes the limitations of using similarity relationships and is able to simulate the range of processes acting on the various urban surfaces.

For far-field deposition formulations, an adaptation of the empirical modeling approach, such as that proposed by Weathers et al. (1999), has merit. They proposed developing a scheme to characterize total deposition to forested areas as a function of landscape features, such as vegetation type, elevation, topographic exposure, slope, and aspect. This concept could be used to provide a scheme to characterize deposition to the various types of urban areas.

The current development of CFD models provides the possibility of future capabilities that will define flow - and, hence, deposition patterns - on individual elements of the surface. This development of CFD capabilities will make good use of a deposition model based on flux to individual surfaces, such as the generalized resistance approach proposed below.

3.5.5 Multimedia Linkages

Knowing how residual deposited material in one medium can move to other media, or within alternative forms in its current medium, will provide an understanding of the potential progression of changing hazards in a post-RDD event evaluation. A capability is needed to address these movements of material within and between the air, soil, and water and to define changing areas of concern.

Post-event precipitation can easily shift the locations of concern. The combination of buildings and vegetation in an urban setting can have a significant total amount of material deposited by dry deposition. Post-event precipitation can potentially move such deposited material from the “urban forest canopy” to the underlying urban surfaces and, perhaps, into water systems.

In addition, the traditional approach of considering the dry deposition and suspension processes separately overlooks the fact they are both just air-surface particulate fluxes in opposite directions. An important aspect of future improvement to deposition and related surface interaction models is that a single unified formulation is needed that considers the equilibrium of vertical particulate fluxes to and from the atmosphere.

Although this review focused on the deposition formulations, it is important to recognize that improving near-source dry deposition computational capabilities will also require improvements in the initial atmospheric dispersion formulations. For currently deployed models, the uncertainty in near-source deposition patterns is largely a function of the lack of near-source resolution. That lack of resolution results both from approximate initializations of the plume characteristics and the model algorithms for handling the initial atmospheric dispersion.

3.5.6 ADAPT-LODI Application

This review of dry deposition formulations has identified areas for which new or improved formulations are needed. As an illustrative case, the review results are applied here to the ADAPT-LODI system, which is a component of the NARAC national emergency preparedness capabilities (LLNL 2005). The LODI code is the atmospheric dispersion portion of the ADAPT-LODI system.

The NARAC emergency response modeling system consists of a coupled suite of meteorological and dispersion models using the ADAPT-LODI system (LLNL 2005; Nasstrom et al. 2006). The diagnostic model, ADAPT, constructs fields of mean winds, pressure, precipitation, temperature, and turbulence parameters, using a variety of interpolation methods and atmospheric parameterizations. The dispersion model, LODI, solves the three-dimensional advection-diffusion equation using a Lagrangian stochastic, Monte Carlo method. LODI includes methods for simulating the processes of mean wind advection, turbulent diffusion, radioactive decay and production, bio-agent degradation, first-order chemical reactions, wet deposition, gravitational settling, dry deposition, and buoyant/momentum plume rise. The models are coupled to NARAC databases providing topography, geographical data, real-time meteorological observational data, and global and mesoscale forecast model predictions.

ADAPT is a diagnostic wind field model that builds three-dimensional gridded meteorological fields (Nasstrom et al. 2006). The model incorporates a number of interpolation and extrapolation techniques, including both direct and iterative solvers and atmospheric parameterizations.

A recent comparison was made of the simulation results from LODI (LLNL 2005) and three other atmospheric dispersion models: RASCAL (Sjoreen et al. 2001), RATCHET, and MACCS2 (Jow et al. 2006). Figure 3.12 shows the fractional air concentration reductions derived from the results of that comparison. A deposition velocity (1 cm s^{-1}) was assumed in LODI, MACCS2 and RASCAL, and ~0.3 cm s^{-1} was used in RATCHET. As would be expected, the three models using the same deposition velocity have nearly the fractional reduction of concentration with distance, and the model with a lower deposition velocity has a smaller fractional reduction. The divergence in the RATCHET results in Figure 3.3 illustrates the importance of having good parameterizations of deposition velocities.

The LODI dry deposition formulations use representative values to characterize dry deposition. The model uses an input value of the dry deposition velocity for small particles that is combined with a settling velocity to estimate a dry deposition velocity for a specific size of particle. This approach has two major limitations. The first is that the deposition rates do not vary with the characteristics of the underlying surface. The second is that the approach does not accurately model the local deposition rates for particles in the 10-50 micron range[18].

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[18] Assuming the LODI input value of dry deposition velocity represents the typical rates for processes in the accumulation mode, the local deposition velocities to specific surfaces can be underestimated by almost an order of magnitude for particles in the 10 to 50 micron range. On the other hand, near-surface concentration depletion resulting from the enhanced local deposition may limit the importance of this process in overall deposition rates.

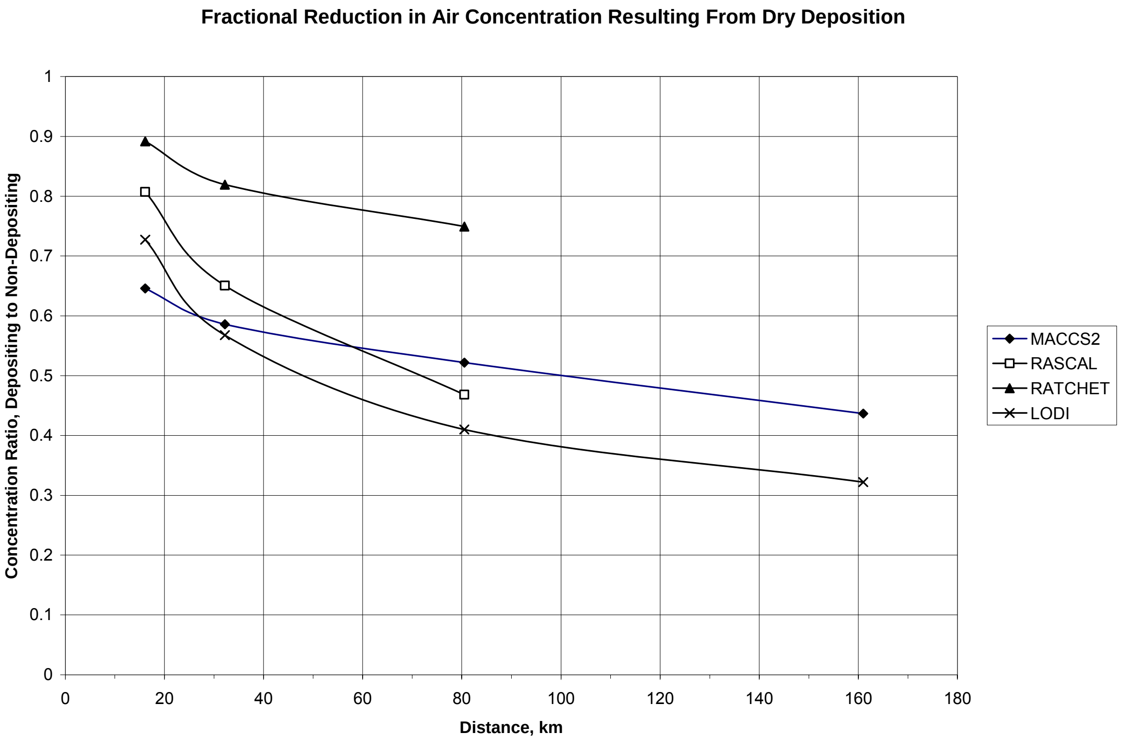

Figure 3.12. Concentration Depletion Comparison for RATCHET, RASCAL, RATCHET, and LODI

The LODI formulation could be greatly improved by estimating particle and surface-specific deposition velocities for the model's computation grid cells. A number of resistance-based formulations are available that address issues of obtaining a representative deposition estimate for terrain with a heterogeneous surface cover. The LODI plume dispersion model can provide the turbulence parameterizations that are needed to compute area-specific aerodynamic resistances. A surface-specific deposition computation will improve the estimated total deposition while providing currently unavailable data for surface-specific hazard analyses.

In the near term, the generic values for deposition velocities used in LODI can be replaced with surface-contaminant-specific tables that can be indexed to available land use information. Grid-area deposition models developed for long-range transport models provide databases that can be used in this update. Although this will be a significant upgrade in terms of input to a plume's mass budget, it will provide only very general area-wide deposition information.

In the longer term, development efforts should work towards providing the capabilities of 1) defining areas of maximum deposition over the range of possible outcomes (short- to long-range), 2) accounting for particle properties, and 3) simulating surface-specific deposition rates (i.e., how much is on what building surfaces, vegetation, roads, sidewalks, etc.). To address specific hazards related to specific surfaces with GIS-based hazard maps, much more detailed surface characterizations will be needed than are generally available in current land-use databases. LODI uses a layered method for separating the atmospheric dispersion from the surface-layer fluxes. The filtration-based element-by-element approach to dry deposition, such as proposed in this report, would seem to be an ideal way to determine how fast material is being lost in the lower layer while characterizing the rate of deposition to different types of surfaces.

4.0 Recommendations

This review of deposition formulations in atmospheric dispersion models relevant to homeland security applications has generated a number of recommendations for improving for these formulations. Because the accuracy of simulated dry deposition rates depends on both the air dispersion and dry deposition formulations, elements of both are included in the recommendations.

The following are the recommendations for dry deposition improvements related to updates in atmospheric dispersion model formulations. These recommendations will to be included in improvement plans for specific air dispersion models.

1. *Better source-term plume initialization*. The predicted and/or potential near-source deposition hazards are highly dependent on how well the initial source term characteristics are described. Models must depend less on generic and more on situation-specific initializations. RASCAL (Sjoreen et al. 2001) and LODI (LLNL 2005) are examples of models where selected facility and event-type source-term options are currently available.

2. *Improved dry deposition formulations for longer-range impacts.* The acid rain studies show that large particles are important in determining dry deposition rates of sulfur particulate compounds at distances for which most current emergency preparedness models predict all such particles should already been deposited from the plume. Improved formulations for in-plume processes are needed that include time-dependent processes such as coagulation. At downwind distances where larger particles in the initial plume have been lost by deposition, very small deposition rates are typically predicted by current models. Coagulation in the transport to such distances needs to be accounted for to help better simulate the observed higher rates of dry deposition at such distances.

The following are the recommendations for improvements in the dry deposition formulations. These recommendations include the development of improvements that are applicable to the range of atmospheric dispersion models used in homeland security applications.

1. *Near-field dry deposition formulations are needed*. The current atmospheric plume models have dry deposition formulations for particulate matter that are largely inapplicable at very close distances. Although this inadequacy is partly because of model grid-scale resolution and model initialization limitations, a dry deposition simulation capability is needed to enable parameterizations of potential near-field rates and patterns. Meeting this need will require a combination of improved models and experiments.

2. *Need to address a full range of potential particle properties*. The plume from a radiological attack such as an RDD, as well as chemical/biological events, can have a wide range of potential particle sizes, densities, shapes, and surface properties. To ensure a capability that meets the potential range of events, it is critical that air-dispersion models have dry deposition formulations applicable to the full range of potential particulate distributions. In addition, dry deposition formulations, if they do not already have the capabilities, need to be updated to incorporate the effects of density and shape by incorporating appropriate formulations. Although the use of aerodynamic diameters addresses the need for computing particle-specific settling velocities, the literature is unclear as to how the influence of particle shape and density should be accounted for in inertial deposition processes for particles in the range of 10 to 50 microns.

Special emphasis in the development of improved dry deposition formulations is needed for larger particles (>10 microns diameter). Many of the currently available transport and dispersion models are designed to model plumes with smaller particles (e.g., from sources such as stacks, vehicle traffic, and wind erosion) and will not adequately address the deposition of larger particles from an event such as a radiological attack involving an RDD. Recent literature suggests that the surface deposition rates may be larger than the historical parameterizations used for this range of particles. Appropriate formulations for these larger particles are available in the literature and can be implemented as model improvements.

3. *Incorporate surface retention/rebound processes*. Most dry deposition formulations assume a 100% retention rate for particles reaching the surface. A critical area for an upgrade is to account for the retention and rebound rate of larger particles. Based on studies summarized by Chamberlain (1991), this upgrade can make up to an order-of-magnitude change in computed dry deposition rates.

4. *Develop dry deposition formulations applicable to urban areas*. The current atmospheric plume models (based on similarity relationships) are inapplicable to the roughness elements encountered in urban areas. A dry deposition formulation is needed that can be applied over both uniform and non-uniform surfaces with surface elements ranging from smooth (water, snow, sand) to rough (urban, forests). This effort is seen as a combination of experiments, model development, and model validation efforts.

These four recommendations are the basis of the dry-deposition model improvement plan given in Appendix A. The main component of the plan is to develop a generalized dry deposition formulation for characterizing dry deposition processes that will address the near-field, particle, and surface properties, and complex surfaces raised in these recommendations.

All these recommendations for dry-deposition simulation updates to current or new air-dispersion models have the objective of improving the estimate of total deposition for plume mass budget considerations and providing surface-specific contamination concentrations:

Total Dry Deposition Rates. The total deposition to a model's computation grid is based on summing deposition to the surfaces, or surface types. Land-use and more detailed surface-cover databases need to define the required surface-specific deposition rates. These updates are essential for improving the total dry deposition rate computation for areas with mixed surface covers, both in rural and urban areas.

Surface-Specified Dry Deposition Rates. Model formulations need to include characterization of dry deposition rates to specific surfaces or surface types. Because there are a wide range of potential deposition rates to the various surface elements, such information is necessary to identify potential levels of exposure for different portions of an affected area.

Despite some conceptual limitations, the current formulations for particle deposition based on a resistance approach have provided reasonable dry deposition simulations to a variety of surfaces. For many models with inadequate dry deposition formulations, adding or improving a resistance approach will be a desirable near-term update. Resistance models, however, are inapplicable to aerodynamically very rough surfaces such as urban areas. In the longer term, an improved parameterization of dry deposition needs to be developed that will be applicable to all surfaces.

These dry deposition model improvement recommendations, although developed for RDD modeling needs, have important implications for the wider range of biological and chemical materials of special concern to the DHS.

5.0 References

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Appendix A

Model Improvement Plan: Generalized Dry Deposition Formulation

Appendix A

Model Improvement Plan: Generalized Dry Deposition Formulation

The model improvement plan is to develop a more general dry deposition formulation for incorporation into air dispersion models that will address the issues identified in this review. A new formulation is to be developed to replace the similarity-based dry deposition formulations used almost exclusively in current emergency preparedness models simulating situation-specific deposition rates.

An approach is proposed here for computing dry deposition rates of particles to very rough surface covers such as those found in urban areas. The dry deposition formulation surface-specific deposition approach used in forest-canopy deposition models provides a basis for developing such a generalized formulation. Because of the potential complexity of modeling all surfaces, formulations for real-time applications will likely need to be based on parameterizations of filtration rates developed based on simulations with more detailed models.

A.1 Concept

The proposed dry deposition formulation approach is based on modeling the flux to individual local surface elements, rather than to an ensemble of different surfaces. By considering surface-specific filtration for each surface element, the resulting surface-specific deposition rates define localized potential hazards as well as the total deposition.

The local delivery of contaminants to the air-surface layer for a variety of local surfaces is considered. Figure A.1 illustrates realizations of this surface layer for a variety of surface elements. Each surface element, or type of surface element, is considered a local filter that removes airborne particles. Dry deposition is computed on each element and then the total dry deposition, D, is the summation over all these elements:

$$D = \sum_{i=1}^{n} A_i \, C_i / r_{ti} \tag{A.1}$$

where D = total dry deposition flux, g m^{-2} s^{-1}
r_{ti} = the total dry deposition resistance for an area segment, $s^{-1}m$
A_i = the area of segment i, m^2
C_i = the contaminant concentration in the vicinity of the surface element
n = the number of surface segments

This approach is an extension of models such as the Wesely model shown above (see Figure 3.5), which characterize dry deposition to elements of vegetation canopies and effectively incorporate a deposition-dependent vertical variation in concentration.

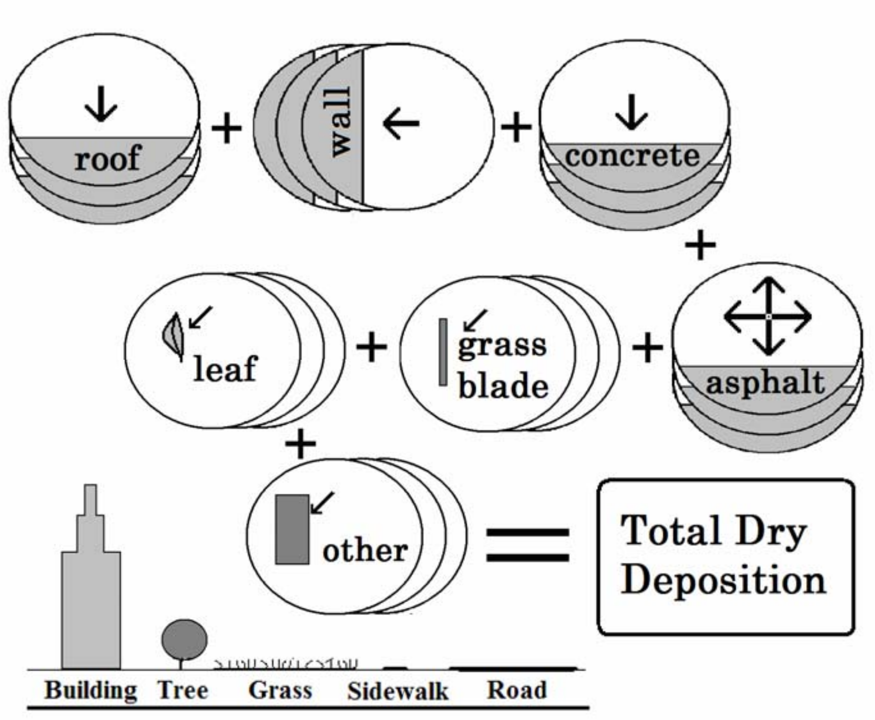

Figure A.1. Concept for Surface-based Local Dry Deposition Formulation

This generalized formulation approach addresses the complex surface-dependent filtration (i.e., dry deposition) of air that occurs for inhomogeneous environments with large surface elements such as urban canopies. The formulation also improves the capability of computing deposition to complex lower canopies such as grass. For a uniform surface, or relatively uniform surface, the generalized formulations should be equivalent to the current resistance-based models.

The total deposition to a horizontal area (such as that defined by a computation grid) is needed for mass budget computations. The summation of deposition to component surface elements provides the total deposition to an area. This approach to computing total deposition is similar to models developed for deposition to vegetative canopies. The approach has similarities with the multiple-surface dry deposition models used in long-range transport models, but differs in consideration of surface elements rather than average surface properties. The issue of non-uniform surfaces still exists but is now concerned with the uniformity of the roughness on the surfaces of individual surface elements.

Formulations are needed relating C_i to the air concentrations computed by the atmospheric dispersion code. These formulations need to account for two related factors: 1) eddy dispersion and 2) localized plume depletion by dry deposition processes. If deposition occurs to the surface at faster rates than eddy transport can deliver material to the surface, then a localized reduction in contaminant air concentration occurs.

Deposition processes tend to reduce the local air concentrations. Even for situations in which the depletion is relatively small for a given element, the cumulative effect on the vertical profile of concentrations can be significant. Figure A.2 schematically shows how the deposition processes produce such profiles. The concentration, C_i, used in the deposition computation, expressed as a function of height, C(z), will reflect the removal of material by deposition:

$$C(z) = C_r\, F(z) \tag{A.2}$$

where C_r is the concentration at some reference height where the effects of surface deposition on concentration are very small and F(z) is the depletion fraction profile resulting from the vertically integrated deposition fluxes. Although some models have addressed this function for vegetation canopies, developing general application formulations for this function is one of the challenges of the proposed deposition model.

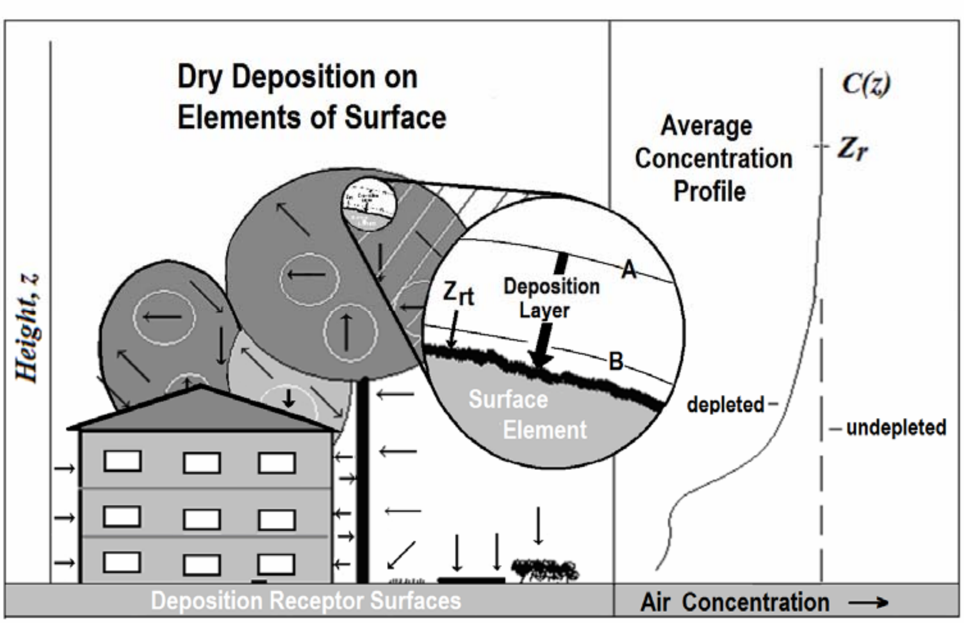

Figure A.2. Dry Deposition on Surface Elements

As described above, the similarity-layer relationships used in current deposition models provide estimates of average atmospheric resistances for eddy transport under the steady-state conditions. The use of multi-layered resistance at surfaces at different heights, an approach used by Wesely (1989) for vegetation canopy deposition, provides a starting point for a formulation that is applicable to a wider range of surfaces. For rougher surface elements, beyond where it is reasonable to extrapolate similarity theory, field studies are likely to be needed to define the actual magnitudes of localized near-surface concentration depletion for situations with very rough surface elements (e.g., urban areas).

The sequential processes for an airborne material to be deposited on a surface are shown in Figure A.3. The diagram starts with a modeled airborne plume concentration.

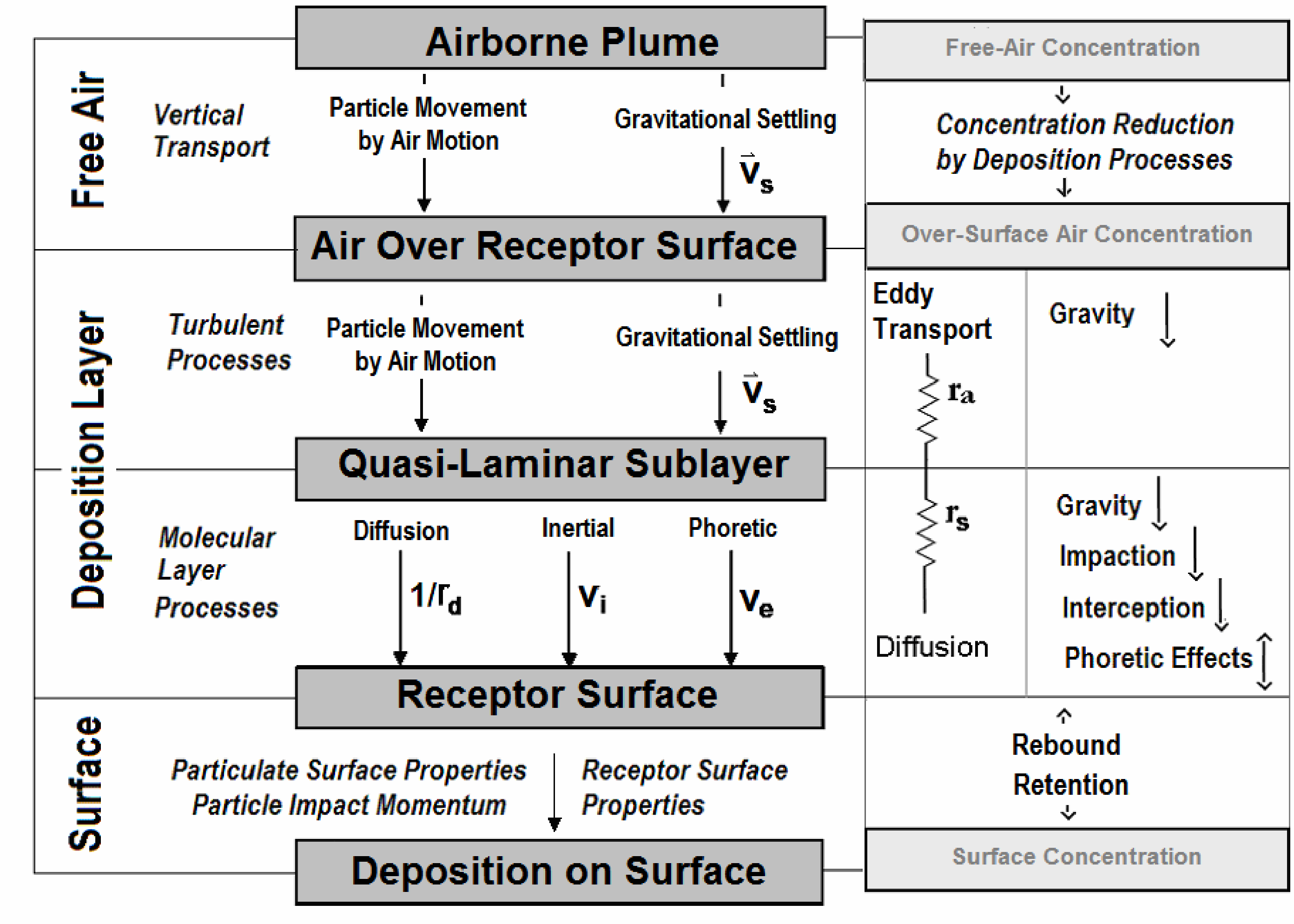

Figure A.3. Generalized Dry Deposition Modeling Approach

A.1.1 Free Air

Deposition flux in the free air occurs by a net vertical eddy transport (effects of air motions) and gravitational settling. These processes move an airborne contaminant to the vicinity of a surface by the motions of parcels of air. Together, these two processes define an upper bound for the maximum rate that dry deposition can occur to the underlying surface.

A relationship or procedure defining the average vertical profile of concentrations is needed that accounts for the combined influence of both the transport mechanisms to that location (atmospheric turbulence and settling velocity, v_s) and the integrated effects of vertically distributed dry deposition fluxes. For simple, relatively homogeneous surfaces with smaller roughness elements, the traditional relationships from surface-layer similarity theory should be adequate for this purpose. For more complex surfaces, an iterative numerical computation process will likely be needed to define the applicable vertical average concentration profile.

Because the atmospheric resistance tends to be the limiting process for the deposition of particulate matter, the near-surface concentrations, C_i, are expected to be generally very close in magnitude to the ambient air concentration at that height, C(z).

A.1.2 Deposition Layer

A “deposition-layer” is shown over a surface element in Figure A.2. “A” marks the outer bound of a “deposition layer” and “B” marks the outbound of the quasi-laminar sublayer. Turbulent processes act between A and B, and molecular processes act between B and the surface.

In the deposition layer for each element of the surface, the new model formulation needs to address the three layers of interdependent dry deposition processes shown in Figure A.3.

Turbulent Processes

If a local surface is sufficiently large that a surface layer develops, then the eddy transport of particles (effects of air motions) as modified by gravitational settling will define the potential rate of movement of particles through that surface layer. These are the same overall processes as in the free air, only here the movement of an airborne contaminant is controlled by the local surface characteristics and the flux is perpendicular to the surface. The fluxes for the deposition layer shown in Figure A.3 are defined as perpendicular to the local surface. The influence of gravitational settling can be positive, negative, or zero, depending on the surface orientation. It is useful to define a local reference height, z_{hl}, defined as normal to the receptor in some multiple, n, of the roughness length, z_{ol}, of the local surface element. This definition of the thickness provides a means of accounting for the localized turbulent flow processes over the surface on the flux to the surface. However, for finely structured surface elements with an insufficient size for a surface layer to develop, the air flow will direct material directly onto the surface and deposition will be limited only by the molecular layer and surface processes.

Molecular Air Layer Processes

Molecular diffusion, inertia, and phoretic processes are the main processes that move particles though this layer.

A.1.3 Surface Processes

These are the “Particle and Receptor Surface Properties” involving the kinetic energy of the collisions and the relative “stickiness” of the surfaces. These processes determine if a particle reaching the surface will remain on the surface. The physical horizontal/vertical orientation can also be an important property of the receptor surface.

The formulation needs to account for the retention and rebound processes acting on particles reaching various types and configurations of receptor surfaces. Studies are needed to develop appropriate parameterizations for these processes.

Recent literature shows that the surface removal processes for particles with diameters greater than 10 microns can be much faster than gravitational settling can deliver particles. This observation indicates the possibility of generating a near-surface concentration deficit even in situations where gravitational settling is the major transport process.

These deposition processes result in a combination of downward, sideways, and upward fluxes over complex surfaces. All terms can potentially vary as a function of particle properties, receptor properties, and ambient meteorological conditions. Standard filtration theory and practice provide formulations for addressing a subset of potential surface configurations.

For more complex surfaces (non-homogenous, larger surface elements, etc.), this approach is expected to be computationally too intensive for implementation in dispersion models. Instead, this approach can be used to develop parameterizations of dry deposition. Because of the complexity of airflows, the urban canopy presents special challenges for developing such a parameterization. The CFD models that are showing promise of being able to simulate details of urban airflows could be used to develop the parameterizations of near-surface concentration reductions.

A.2 Preliminary Application

Applying the concept of deposition to specific surface elements in an urban area provides some preliminary insights into where material will be potentially deposited and illustrates why generalized dry deposition models need to be developed. Using approximate literature-based relationships to characterize the magnitudes of the processes, the deposition to an area with a high building density is compared to an area with a low building density (Figures A.4 and A.5). These plots account for order-of-magnitude differences in the flux rates to different types of surfaces, as well as for area and wind exposures of the various surfaces.

These figures show that there are significant differences between the low and high building density cases in where material is deposited. The building areas are the main sink for deposited material in the high-density case, shifting to vegetation in the low-density building case. A similar effect occurs for gravitational settling results. An interesting result is that the deposition to tree foliage is important even in the high-density building case.

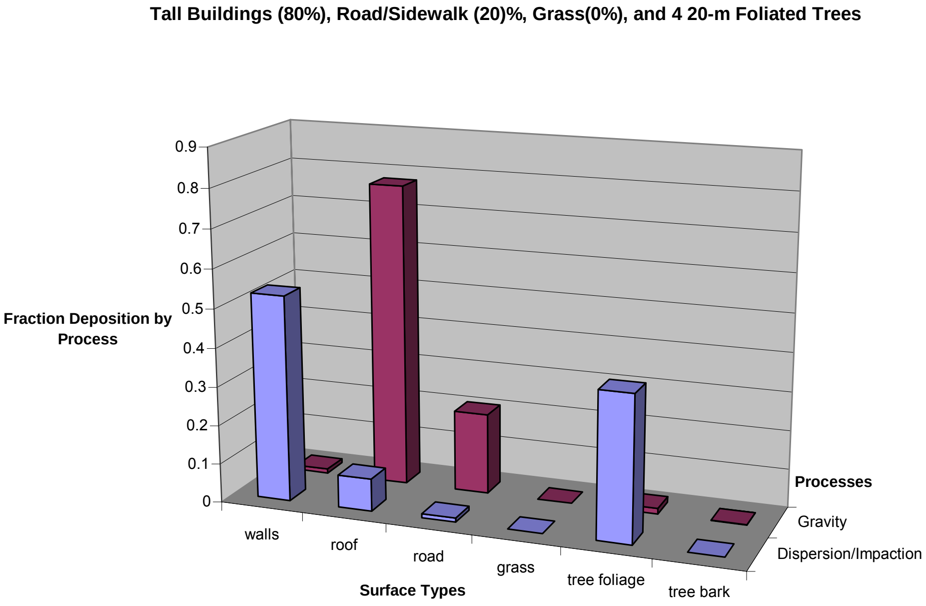

Figure A.4. Relative Deposition Rates in a Dense Urban Area

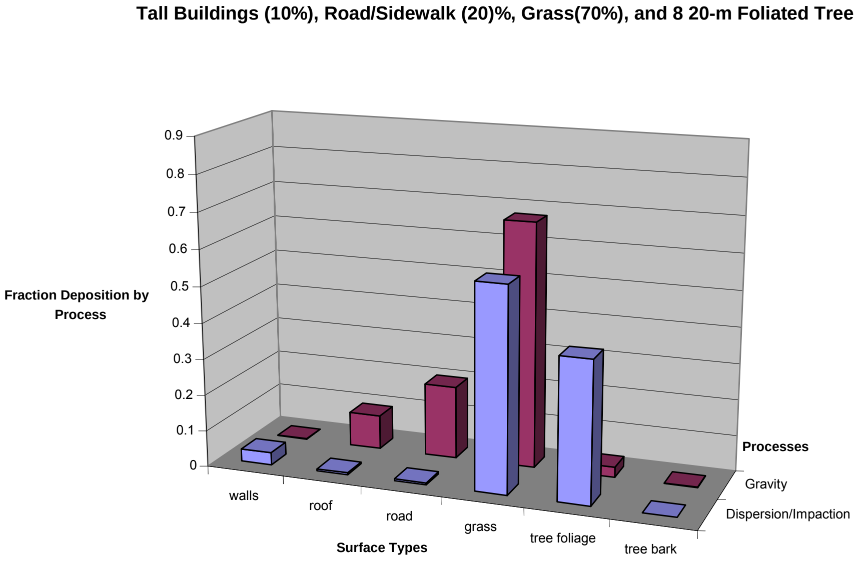

Figure A.5. Relative Deposition Rates in a Less Dense Urban Area

A.3 Reference

Wesely ML. 1989. "Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models." *Atmos. Envir*. 23(6):1293-1304.

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