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Deposition Velocity Methods For DOE Site Safety Analyses

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DEPOSITION VELOCITY METHODS FOR DOE SITE SAFETY ANALYSES

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TABLE OF CONTENTS

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EXECUTIVE SUMMARY

This report documents the results of an investigation of deposition velocity methods for use in Department of Energy (DOE) Documented Safety Analysis (DSA) modeling. The project was sponsored by the National Nuclear Security Administration (NNSA) Nuclear Safety Research and Development Program and monitored by the Chief of Nuclear Safety (CNS) from the Office of the Under Secretary for Management and Performance. Based on initial scoping discussions, the primary focus of this effort was on the dry deposition of particulate matter. However, within the constraints of the project, we also explored the sensitivity of safety analysis modeling to other key input parameters (wind direction variability, atmospheric stability, release characteristics, model assumptions), examined the 95th percentile methodology, and investigated a few aspects of a commonly-used DOE safety analysis code, MACCS2 (MELCOR Accident Consequence Code System).

We developed and conducted a DOE site survey to obtain an understanding of characteristic site release scenarios and environmental conditions. Since a wide range of radionuclides and release types were reported, this investigation was based on comparisons of air concentrations for particular particle-size ranges rather than dose exposures that are heavily dependent on the material, physical/chemical form, and exposure pathway involved. From the results of the site survey and discussions with the CNS and other subject matter experts, we focused on particulate releases in two respirable-size ranges, corresponding to representative particle diameters for unmitigated/unfiltered (2-4 μm) and mitigated/filtered (0.2 - 0.4 μm) releases.

A priority was given to investigating deposition velocity methods applicable to the Gaussian plume models most widely used for DOE safety analyses – MACCS2 (DOE, 2004) and HotSpot (Homann and Aluzzi, 2013). Both of these "toolbox" models have met the requirements for inclusion in DOE's Safety Software Central Registry. We also performed limited investigations of methods used in other models (e.g., AERMOD [Cimorelli et al., 2005], CALPUFF [Scire et al., 1990], RATCHET [Ramsdell et al., 1994] and GENII [Napier, 2010]). A copy of the version of MACCS2 (V 1.13.1) included in DOE's Central Registry was obtained from the Nuclear Regulatory Commission (NRC) and Sandia National Laboratories (SNL). HotSpot is freely available from and maintained by Lawrence Livermore National Laboratory (LLNL).

We developed software to calculate 95th percentile air concentrations that combines hourly meteorology with wind-sector dependent deposition velocity values and site-boundary distances. This approach uses consistent values of all physical parameters and fully accounts for the dependence of air concentrations on wind speed and direction, atmospheric stability, land-use/surface roughness, deposition velocity, and distances of interest. This is a more rigorous and robust approach than identifying the 95th percentile meteorology and selecting "reasonably conservative" values (as specified in HSS Safety Bulletin, 2011) for other input model parameters. We used our new 95th percentile air concentration methodology to study the sensitivity of deposition to various input parameters and environmental conditions.

We conducted a literature survey to review existing deposition models. The results of this review led to a recommendation for a state-of-the-science deposition velocity model that is practical enough to be usable in site hazard analyses. This model was used to conduct a set of evaluation and sensitivity studies. We then implemented the recommended deposition velocity model into our 95th percentile software and applied this to two illustrative case studies.

KEY RESULTS

Key findings from our investigation are summarized below:

- *Deposition velocity (v_d).* The Office of Health, Safety, and Security (HSS Safety Bulletin, 2011) currently recommends the use of default deposition velocities of 0.1 cm/s for unmitigated/unfiltered particles with Aerodynamic Equivalent Diameters (AEDs) in the range of 2-4 µm, and 0.01 cm/s for mitigated/filtered releases of particles with AEDs of 0.2 - 0.4 µm. With a few caveats discussed below, these values were found to be generally appropriate for particulate plume modeling, unlike the previously recommended default values of 1 cm/s and 0.1 cm/s. Related findings are as follows:
    - The Petroff and Zhang (2010) model currently provides the most accurate deposition velocity values for a wide range of atmospheric and environmental conditions.
    - The HSS Safety Bulletin (2011) default deposition values are most appropriate for grassland. They are somewhat over-conservative for forests and under-conservative for bare ground, predicting air concentrations ~15% higher or lower, respectively, than the optimal choice of v_d. These are relatively small differences that may not be significant relative to those resulting from uncertainties in weather observations, atmospheric stability, or land-use category.
    - Predicted air concentrations for the filtered/mitigated particle size range (AED = 0.2 – 0.4 µm) are relatively insensitive to the range of potential deposition velocities. Specifically, the current HSS-recommended (HSS Safety Bulletin, 2011) default value of v_d = 0.01 cm/s for such particles produces minimal plume depletion and gives virtually the same results as using no deposition (v_d = 0).
    - The use of a single deposition velocity that is "conservative" for all sites and scenarios will produce an overly conservative result for many cases (corresponding to unnecessarily high air concentrations and exposures). However, site- and scenario-specific values could be used in initial screening calculations to determine whether a more in-depth analysis is needed (e.g., if calculated doses exceed or are close to the threshold that warrants additional mitigation/protective actions), as discussed below.
- *Sensitivity analysis of key model input parameters.* Predicted air concentrations were found to be as or more sensitive to the wind-direction dependent distance to the location of interest (e.g., the site boundary), the meteorology (e.g., wind speed, atmospheric stability class) and the release height, as to the choice of deposition velocity.

- *Observations regarding the use of the MELCOR Accident Consequence Code System (MACCS2).* During this investigation, we identified a few aspects of MACCS2 Version 1.13.1[1] that users should be aware of when performing hazard analyses.[2]

    - The Brigg's open country dispersion coefficients produce MACCS2 concentration values more consistent with other commonly-used models (i.e., HotSpot, AERMOD) than those resulting from the default Tadmor and Gur (TG) option. The TG coefficients were derived from experimental data over flat terrain using curve fits that are considered appropriate only over the range 0.5 – 5.0 km and consequently may not be valid for other conditions and distances. It is therefore recommended that the Brigg's open country dispersion coefficients be selected for DOE safety analysis modeling.

    - MACCS2 V1.13.1 does not automatically calculate the wind speed at the release height[3]. Changes of wind speed with height may have a sizeable impact (~40%) on predicted air concentrations for elevated sources. For such cases, the release height wind speed should be externally calculated and input by the user.

    - The absence of a low wind speed algorithm in MACCS2 limits the significance of 95th percentile air concentration calculations for sites at which more than 5% of winds are below a 1 - 2 m/s threshold. For such locations, we recommend the use of alternate codes (e.g., HotSpot) that incorporate special algorithms to cover low-wind speed cases.

    - The changes in MACCS2 predicted air concentrations through the use of more accurate deposition velocity values may be overshadowed by the above corrections for low wind speeds and/or elevated releases.

RECOMMENDATIONS

Based on this investigation, the following recommendations are made:

- *Recommended deposition velocity model.* The current state-of-the-science dry deposition velocity model for particles is the Petroff and Zhang (2010) model. If default choices for v_d are inadequate for DOE site accident analysis applications, use of this model is recommended as it parameterizes the impact of a wide range of vegetation types while requiring only a reasonable level of modeling sophistication. Input variables to the Petroff and Zhang model may be calculated or reasonably estimated from meteorological

[1] MACS2 V1.13.1 is the version of the code included in DOE's Central Registry as a Safety Software toolbox code.
[2] Later version of MACCS2 may have addressed some of these issues.
[3] The standard reference height for surface meteorological measurements is 10 m above ground level.

observations that are routinely available at DOE sites. A methodology for implementation of this model is provided in this report.

- *Recommended 95th percentile air concentration methodology*. We recommend the use of a more robust approach for determining 95th percentile air concentrations in which hourly wind speed and direction observations are used to determine wind-sector dependent land-use categories, v_d values, and direction-dependent site boundary distances. This method ensures that physically consistent values of the input parameters are used in conjunction with the actual meteorological and environmental conditions.

  - Dominant land-use categories for each wind-sector direction are needed to obtain appropriate deposition velocity values from the Petroff and Zhang model. Due to the complex site boundaries (i.e., the varying distance from source to site boundary with wind sector) and inhomogeneous land-use characteristics at some DOE sites, the EPA's AERSURFACE model (or another alternative software package) may need to be run with varying search radii to determine the appropriate land-use categories for each wind sector.

  - If more than 5% of the wind speeds observed at a site are below a threshold of 1 - 2 m/s, a model should be used that incorporates a low wind speed algorithm (e.g., the HotSpot G stability option documented in Homann and Aluzzi, 2013).

- *Proposed approach for performing atmospheric transport calculations for DOE safety analyses.* A two-step hierarchical approach is proposed for performing DOE safety analysis modeling. If the first highly conservative screening step results in levels exceeding or close to specified air concentration thresholds, a second level analysis can be performed to provide a higher-fidelity, but still conservative, analysis.

  - Level 1 Screening Calculation: Perform standard 95th percentile calculations using a lower bounding value for the deposition velocity to determine if a more sophisticated model is required.

    - Option A. Use $v_d = 0$ for all land-use conditions.

    - Option B. If Option A produces overly conservative estimates of exposures, select an alternative site- and scenario-specific lower bounding value for v_d derived from the Petroff and Zhang [2010] model and associated experimental results. This option requires a careful justification of the conservatism of the selected value(s) for the specified particle size range and environmental conditions of interest, particularly for sites with inhomogeneous environmental conditions and/or diverse release scenarios. Use of the current HSS recommended default values is discussed below:

      - The recommended HSS Safety Bulletin (HSS, 2011) default value of $v_d = 0.1$ cm/s for unmitigated/unfiltered releases is a lower bounding value for forests, falls in the mid-range of deposition velocity values for grasslands, and is higher than the Petroff and Zhang (2010) values for bare ground conditions.

  - ❖ Although the recommended HSS Safety Bulletin (HSS, 2011) of v_d = 0.01 cm/s for mitigated/filtered releases does not represent a lower bound for v_d for all environmental conditions, this non-zero value produces virtually the same results as a deposition velocity of zero.

- o Level 2 Full Calculation: Perform a full 95th percentile air concentration calculation based on hourly wind speed and direction observations and correlated wind sector-dependent site-boundary distance and deposition velocities derived from the Petroff and Zhang model. A detailed procedure for implementing this calculation is found in this report.

- *Use of more sophisticated computer codes for safety analyses.* The use of more sophisticated codes (e.g., non-Gaussian plume models) for safety analyses is difficult to justify.

  - o More sophisticated models account for the time-variation in meteorological conditions and therefore can produce time-averaged or time-integrated air concentrations that are less "conservative" (e.g., have a greater frequency of predicting concentrations that are less than those observed) than Gaussian plume models that use steady-state meteorology.

  - o The accuracy of more sophisticated models over the full range of conditions used in safety analysis modeling is hard to assess.

    - ▪ Past tracer study comparisons have shown that it is not possible to draw universal conclusions regarding the accuracy of such models from individual studies (e.g., different models perform better than others depending on details of the study). Therefore, experimental validation typically needs to be performed on a case-by-case basis for each location and release type.

    - ▪ Tracer studies typically do not cover the full range of atmospheric stability and meteorological/environmental conditions. In particular, there are very few experimental studies that include data for the very stable, low-wind conditions typical of 95th percentile meteorology.

  - o It is more difficult to set up sophisticated model simulations to ensure conservatism of the results than is the case with Gaussian plume models that exhibit simpler dependencies on input parameters. Over-riding internal model physics by user-specification of parameter values (e.g., the deposition velocity) may result in the use of inconsistent physics and diminish any benefits of using a model designed to simulate complex conditions.

  - o The expertise and resources (personnel and computational) required to use more sophisticated models may be cost prohibitive. Complex models require trained users to properly specify all of the input variables and options needed to produce accurate analyses and quality-assure results. The use of improper inputs or physical/numerical options is a significant risk in the use of more sophisticated models.

- Most sophisticated models have not been included in the DOE Central Registry for Safety Software toolbox codes, in part because of the significantly greater Software Quality Assurance effort required[4]. Therefore an extensive justification for their use and application, as well as thorough review of inputs and results, would be required for hazard analysis applications.

- In specific cases (e.g., in cases of complex terrain when representative meteorological observations are available) the use of more sophisticated models may be justified if specialized expertise is available to conduct complex dispersion modeling, and the risks and issues discussed above can be addressed.

• *Standardization of methodologies.* Based on our investigation, we also recommend that DOE should establish clear definitions and methods for selecting "reasonably conservative" input parameters, accompanied by documented procedures standardizing the approach for conducting 95th percentile calculations for safety analyses.

[4] One exception is GENII, but its use has been deprecated for the submicron particle size range because this model calculates a constant deposition velocity value over this size range and does not match the expected theoretical minimum (see HSS Safety Bulletin, 2011).

1. INTRODUCTION

This report summarizes the results of a requested investigation of deposition velocity methods for use in Documented Safety Analysis (DSA) modeling, sponsored by the National Nuclear Security Administration (NNSA) Nuclear Safety Research and Development Program and monitored by the Chief of Nuclear Safety (CNS) from the Office of the Under Secretary for Management and Performance. The choice of deposition velocity can be important in estimating the potential dose exposures to the public when determining the safety classification of structures, systems, and components at DOE sites.

Based on initial scoping discussions, the primary focus of this effort was on dry deposition of particulate matter. However, within the constraints of the project, we also explored the sensitivity of safety analysis modeling to other key input parameters (wind direction variability, atmospheric stability, release characteristics, model assumptions), examined the 95th percentile methodology, and investigated a few aspects of a commonly-used DOE safety analysis code, MACCS2 (MELCOR Accident Consequence Code System).

Dry deposition velocities are highly dependent on scenario-specific conditions, including the meteorology (wind speed, atmospheric stability), environment (terrain, land-use, surface roughness), and release type (gas/particle, particle-size distribution, physical/chemical form). As both experimentally-measured and theoretically-calculated dry deposition velocities have been shown to vary over several orders of magnitude, the choice of deposition velocity can significantly impact calculated dose impacts (e.g., air concentrations 10 km downwind may vary by up to an order of magnitude as the deposition velocity changes from 0.1 to 1.0 cm/s). DOE safety analyses require a robust modeling approach consistent with the latest scientific advances that incorporates the needed degree of accuracy and conservatism and is implementable in a practical manner.

Our investigation involved multiple components, including expert solicitation, literature review, software development, parameter sensitivity studies, model evaluation using existing DOE safety software codes, and illustrative case studies. This led to a set of findings and recommendations. A priority was given to identifying deposition velocities and methods applicable to DOE Safety Software models (e.g., MACCS2 [DOE, 2004], HotSpot [Homann and Aluzzi, 2013]), although as possible we performed limited investigations of other codes (e.g., AERMOD [Cimorelli et al., 2005], CALPUFF [Scire et al., 1990], RATCHET [Ramsdell et al., 1994] and GENII [Napier, 2010]). We obtained a copy of MACCS2 V1.13.1 from the Nuclear Regulatory Commission (NRC) and Sandia National Laboratories (SNL). HotSpot is freely available from and maintained by Lawrence Livermore National Laboratory (LLNL).

One of the fundamental issues identified during this project was the lack of clear criteria for determining the meaning of "reasonably conservative"[5] as applied to the choice of non-meteorological parameters for 95th percentile modeling. Specifically, we found no standard

[5] The DOE guidance on input parameters for source term and dose estimation calculation from DOE-STD-3009-94 (DOE, 2006) states that: "The intent is that calculations be based on reasonably conservative estimates of the various input parameters".

definition or methodology identified for selecting such "conservative" values. We chose to circumvent this difficulty by developing a 95th percentile air concentration method that uses consistent choices of meteorology, land-use, and deposition velocity for different wind sectors. This method also has the advantage of using self-consistent sets of input parameters so that only physically reasonable combinations of input values are used in each (hourly) calculation.

In Section 1, we provide a discussion of the objective, background, and work conducted under this project. Section 2 summaries the results of a DOE site survey (the raw survey feedback is provided in a separate Official Use Only appendix). A literature review of deposition models is covered in Section 3 and the evaluation study methodology and results are covered in Section 4. Some observations regarding MACCS2 that were investigated incidental to the main objectives of this investigation are discussed in Section 5. Section 6 provides a detailed description of how the recommended deposition velocity methodology can be implemented, with Section 7 illustrating the application of this approach to two different site case studies. The main body of the report concludes in Section 8 with a summary of key findings and recommendations. Appendix A (including the separate OUO section) discusses the DOE site survey and feedback. A requested peer review of a study by Till et al. (2011) is provided in Appendix B, including a discussion of the use of advanced models for safety analysis applications.

1.1 OBJECTIVE AND SCOPE

The primary objective of this study was to review deposition velocity methodologies and to propose a defensible general approach for deriving input parameter values that could be used complex-wide for DOE 95th percentile safety software analyses. In addition, we also examined 95th percentile methodologies and some aspects of the MACCS2 model that are relevant to safety analysis applications. It should be noted that many of the results of this investigation also are relevant for Emergency Planning and Hazard Assessment (EPHA), vulnerability studies, and environmental impact calculations.

As part of the project, we conducted a comparison of a representative set of deposition models, developed a recommendation for use of a state-of-the-science deposition model, and formulated an approach that could be used to determine dry deposition velocities (v_d) values for use in Gaussian plume DOE Safety Toolbox models such as MACCS2 (MELCOR Accident Consequence Code System) and HotSpot. It should be noted that both MACCS2 and HotSpot use simplified "source depletion" models for deposition rather than more physically realistic "surface layer" depletion models. Such source depletion models generally are considered adequate for low deposition velocities. This study does not consider how or whether these approaches could be implemented for other types of models (e.g., GENII, CALPUFF).

The scope of this investigation was restricted to radionuclide releases, particulates, and outdoor air concentrations (inhalation exposures). Given the wide range of scenarios reported in our DOE site survey, we focused on comparisons of relative air concentrations for particular particle size ranges rather than investigating exposure doses which are heavily dependent on the specific radionuclide of interest as well as its physical and/or chemical form. Our primary focus was on near-surface releases, although a limited investigation of elevated releases was included as input from the survey indicated that stack release scenarios were important at several DOE sites. Physical processes not typically included in Documented Safety Analysis (DSA) modeling (e.g., plume meandering, wet deposition / precipitation scavenging, chemical and/or physical reactions, resuspension, building wake or urban effects, vegetation canopy physics) were also not considered in this analysis.

1.2 BACKGROUND

DOE is required to analyze hazards at its nuclear facilities in order to establish safety controls. DOE Standard 3009-94 (DOE, 2006) prescribes the approaches to be used in performing these assessments, covering identification of hazardous materials and the events that could result in their release to the environment as well as methods for calculating the consequences of such accidents. In safety analyses "reasonably conservative estimates" of the "source term" (HSS Safety Analysis Bulletin, 2011) are to be determined and input to a model that calculates the dose potentially received by a maximally exposed individual (MOI) – a member of the public located at the site boundary.

The comparison point is specified as "the 95th percentile of the distribution of doses to the MOI, accounting for variations in distance to the site boundary as a function of direction" (DOE, 2006). The specified exposure period is usually 2 hours, but may be up to 8 hours for long-duration releases. The exposure of interest is generally taken to be the 25 rem Total Effective Dose (TED). Ingestion is not commonly considered for safety analyses (except for the water contamination pathway) and was not considered in this investigation.

1.2.1 SAFETY ANALYSES

The DOE standard stipulates that the plume (or dilution) calculation used in safety analyses should be conducted in accordance with the method set out in Nuclear Regulatory Commission Guide NUREG-1.145 (NRC, 1983). The latter guide specifies that the 95th percentile dose should be determined by calculating the plume dilution factor at the specified downwind distance for each hour of a year using measured meteorological data for that hour and then choosing the dilution factor that is exceeded 5% of the time. Although strictly this would require all of the other input parameters (e.g., the deposition velocity) to be individually determined for each event type and time period consistent with the source term, meteorology, and environment conditions, typically a single value of other input parameters is specified for these inputs.

According to NUREG-1140 (NRC, 1998), for the materials of greatest interest for fuel cycle and other radioactive material licenses, the dose from the inhalation pathway dominates the overall dose. Therefore the focus of this investigation is on particulate releases in the respirable-size range. DOE safety analysis guidance for deposition velocities typically focuses on particulate releases in two particle-size ranges corresponding to "unmitigated/unfiltered" (typically taken to as Aerodynamic

Equivalent Diameters [AED] of 2-4 μm) and "mitigated/filtered" (AEDs between 0.2 - 0.4 μm) releases[6]. These particulate size classes were the focus of this investigation.

1.2.2 DOE SAFETY ANALYSES TOOLBOX CODES

Codes that DOE has deemed appropriate for use in safety analysis calculations are included in the DOE Safety Software Central Registry. These "toolbox" codes must meet designated software quality assurance requirements. Two widely-used examples of such codes are the MELCOR Accident Consequence Code Systems-2, MACCS2 (DOE, 2004), and HotSpot (Homann and Aluzzi, 2013), developed and maintained by Sandia National Laboratories and Lawrence Livermore National Laboratory respectively. Both MACCS2 and HotSpot contain a Gaussian plume dispersion model that can be used to calculate the dilution of a plume due to dispersion and deposition.

Such relatively simplistic Gaussian plume models are widely used in safety analyses due to their simplicity and rapid calculation time. In safety analysis applications, a number of physical processes are not taken into account (e.g., plume meander, plume buoyancy, wet deposition, building wake effects, or the protection factors resulting from sheltering). The degree of conservatism introduced by neglecting such processes has not been quantified, although their inclusion would be expected to reduce calculated dose levels (except in certain circumstances such as areas close to buildings).

1.2.3 DRY DEPOSITION VELOCITY

One of the key input parameters used in safety analyses codes is the deposition velocity, v_d. The deposition velocity is dependent on many factors including atmospheric conditions (wind speed, stability), surface conditions (vegetation canopy characteristics and surface roughness), and the size and density of the released particles. Values of v_d range over several orders of magnitude resulting in dilution factors that may vary by as much as an order of magnitude (e.g., see the sensitivity analysis discussed in DNFSB, 2010) and typical exhibit a minimum in the 0.1 – 1.0 μm size range. Numerous methods have been developed to estimate v_d based on combinations of experimental data and theoretical constructs.

The DOE MACCS2 Computer Code Application Guidance for Documented Safety Analyses (DOE, 2004) recommended the use of v_d values for safety analyses of:

- 1 cm/s for unmitigated releases (coarser particles with aerodynamic equivalent diameter [AED] between approximately 2.5 and 10 μm)
- 0.1 m/s for filtered releases (finer particles with an AED between approximately 0.1 and 2.5 μm)

These default values were derived from the work of Sehmel and Hodgson (1976) and were intended

[6] Our choice for the aerodynamic diameter size ranges for mitigated and unmitigated releases is derived from the guidance cited in HSS, 2011.

to be applicable across the DOE complex. Specifically 1 cm/s represented a lower bound on the deposition velocity for the curve using a friction velocity (u_*) of 100 cm/s and a surface roughness z_o of 3 cm. Sehmel and Hodgson's (1978) revised model showed significantly lower deposition velocities for the same surface roughness and the same and lower friction velocities (and wind speeds). Recent work has further improved estimates of deposition velocities under different atmospheric and environmental conditions as discussed in this report.

In March 2010, the Defense Nuclear Facilities Safety Board (DNFSB, 2010) questioned the technical justification for the then recommended default deposition velocity of 1 cm/s and specifically the use of this value in a safety analysis for the Hanford Waste Treatment and Immobilization Plant (WTP). The DNFSB concluded that "reasonably conservative" dry deposition values of 0.2 cm/s (coarser particles) and 0.01 cm/s (finer particles) should be used based on particle size, wind speed, and surface roughness conditions at the site. More generally, the DNFSB indicated that it believed that a single dry deposition value selected from the range between zero and the predicted v_d for the median particle size would provide a conservative dose consequence assessment.

In response to the DNFSB letter, DOE's Office of Health, Safety, and Security (HSS) and the Chief of Nuclear Safety (CNS) from the Office of the Under Secretary for Management and Performance conducted its own review and concluded that a v_d value of 0.1 – 0.3 cm/s was more technically defensible for unmitigated releases of radioactive materials at DOE facilities (DOE, 2010). Therefore it recommended that if site-specific values are not available, a default value of 0.1 cm/s be used for the deposition velocity. As part of this review, DOE commissioned a subject matter expert, Dr. John Till, to investigate the conservatism of the MACCS2 safety analysis conducted for the WTP sites. Dr. Till concluded that while the 1 cm/s deposition velocity specified in DOE, 2006, was not a conservative value in and of itself, the inherent conservatism in the MACCS2 Gaussian plume model would more than compensate for the use of this choice of v_d (DOE, 2010). DOE concurred with his opinion that the original MACCS2 results for the WTP met the criteria for conservative dose calculations.

The CNS also worked with WTP staff to determine appropriate site-specific deposition velocity values for the Hanford WTP site (Garzon, 2011). This study used a resistance model formulation based on the one employed in the Regional Atmospheric Transport Code for Hanford Emissions Tracking, RATCHET (Ramsdell et al., 2006). The investigation concluded that a value of 0.3 cm/s would be appropriate for WTP releases involving 0.3 µm particles and wind speeds in the range of 1.0-2.0 m/s.

In May 2011, DOE HSS issued an *Accident Analysis Parameter Update* bulletin (HSS Safety Bulletin, 2011) that revised the default parameter guidance for v_d to:

- 0.1 cm/s for unmitigated releases
- 0.01 cm/s for filtered releases

The use of a site-specific v_d developed from the GENII V2 model was suggested as an alternate approach for treating unmitigated (unfiltered) releases; however, GENII V2 was not considered appropriate for mitigated (filtered) releases as its deposition model does not perform well in this size range. The bulletin also specifically recommended against the use of models that only include gravitational settling in their deposition calculations, do not account for surface roughness, or do not handle light wind speed conditions.

The HSS Safety Bulletin (2011) also made three interim recommendations for the specification of deposition velocities for safety analyses:

- Use the revised default values for v_d specified in the bulletin
- Calculate site-specific deposition values for both unmitigated/unfiltered and mitigated/filtered particulate releases
- Use a more sophisticated computer code than MACCS to determine the 95^{th} percentile dose at the site boundary

These recommendations led to some community concerns that in part motivated this investigation. The new more stringent values recommended as defaults could result in significant over-estimates of the potential exposures and a corresponding unnecessary expenditure of resources to address risks. In addition, the third option is a departure from the use of a globally conservative approach and could result in significant additional effort to justify the conservatism of the different approaches used to derive the deposition velocity and other input parameters.

1.2.4 95^{TH} PERCENTILE CALCULATIONS

HSS Safety Bulletin (HSS, 2011) reiterated that appropriate conservatism should be provided by using a site-specific wind speed and stability from the 95^{th} percentile meteorology to determine a dilution factor that is not exceeded more than 5% of the time. However to perform such calculations, "reasonably conservative" site-specific choices need to be made for other parameters (e.g., surface roughness, particle size and density, deposition velocity) by selecting values from the conservative tail of the physically reasonable parameter range. However, no recommended prescription was provided for how to determine these values.

CNS staff have noted that model input parameter selection for atmospheric dispersion calculations is inconsistent across the DOE complex[7]. They also confirmed that limited DOE guidance is available to assist a contractor in selecting a methodology that can be used to develop reasonably conservative inputs and that additional guidance would be prudent as many parameters have the potential to significantly impact the calculated radiological dose consequence analysis.

In our investigation, we were unable to find a quantitative objective definition for the "reasonably conservative" standard and/or established methodologies for determining whether parameters meet this standard. The only general guidance we found was the statement that "[e]ven if a single value in the dose calculation were off by an order of magnitude, the resulting value would not approach the mean value of dose if a cumulative distribution of dose were calculated" (DOE, 2004).

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[7] See for example the presentations at the MACCS2 Workshop Conference, June 5-6, 2012 found at http://energy.gov/downloads/maccs2deposition-velocity-workshop

We also were concerned about the potential pairing of inconsistent input parameters for stability class, dispersion, deposition, and environmental conditions in determining the 95th percentile meteorology case. Different methods for determining stability class will bias the overall cumulative distribution impacting the 95th percentile values. Similarly, the choice of dominant land-use category and the associated surface roughness affect plume dilution from mechanical turbulence. It also should be noted that the importance of various input parameters (including the deposition velocity) in exposure calculations depends strongly on site characteristic such as the downwind distance to the location(s) of interest, as well as the environmental and meteorological conditions.

The basic dose calculation used in safety analysis is calculated from four components: the source term, the transport term (advection/dispersion, deposition, decay), the exposure factor and the dose coefficient. In this investigation, we focus solely on the transport term and use air concentration as the appropriate metric for our comparisons. We also neglect radioactive decay. Therefore, we do not consider the uncertainties introduced by source term assumptions, decay processes or exposure/dose calculation methods, nor do we consider the relative degree of conservatism in these components compared to the transport and deposition components.

1.3 PROJECT OUTLINE

Our investigation proceeded through the following steps:

- We developed and conducted a DOE site survey in order to obtain an understanding of characteristic site release scenarios and conditions. Based on the results of the site survey and discussions with CNS, we focused this investigation on two size categories representative of coarser (2-4 μm) and finer (0.2 - 0.4 μm) particles as commonly assumed for unmitigated and mitigated releases (DOE, 2010).
- We obtained Version 1.13.1 of the MACCS2 model that is included in DOE's safety software Central Registry from the Nuclear Regulatory Commission (NRC) and Sandia National Laboratories (SNL). This model was used extensively in our investigation, along with another DOE Safety Toolbox code, HotSpot V3.0, maintained by Lawrence Livermore National Laboratory (LLNL).
- Software was developed to determine 95th percentile air concentrations that combined hourly wind speeds and directions with wind-sector dependent deposition values and site-boundary distances. This is a more robust approach than selecting the 95th percentile meteorology and seeking to determine “sufficiently conservative” values for other input model parameters, which do not produce overly conservative results. We used this robust 95th percentile air concentration methodology to study the sensitivity of deposition to various input parameters and environmental conditions.
- A literature survey was conducted to review existing deposition models and identify a candidate state-of-the-science deposition model.
- We then performed sensitivity and evaluation studies of this candidate model along with other commonly used deposition models. The outcome of this process was a

recommendation for a state-of-the-science deposition velocity model that is practical enough to be usable in site safety analyses.

- The recommended deposition model and 95th percentile method was applied to two illustrative case studies.

2.0 DOE SITE SURVEY

A survey was developed and distributed to DOE sites to gather information on atmospheric dispersion modeling requirements for safety analysis and emergency planning (the survey form can be found in Appendix A.1). Site responses were used to develop an understanding of the range of potential atmospheric release scenarios and environmental conditions that need to be modeled and to identify the most important categories of releases to be addressed in this investigation. The survey was sent to members of the Emergency Management Issues Special Interest Group (EMI SIG) Hazards Assessment Subcommittee, the Energy Facility Contractors Group (EFCOG) Accident Analysis Subgroup, and the EFCOG Safety Basis Subgroup, with the assistance of the chairs, Michelle Wolfgram, Mukesh Gupta and Nathan Cathey, respectively.

A total of 15 survey responses were received from 9 sites: Hanford, Idaho National Laboratory, Los Alamos Site Office, Oak Ridge National Laboratory, Pacific Northwest National Laboratory, Sandia National Laboratories, Savannah River Site, Oak Ridge Y-12 National Security Complex and the Waste Isolation Pilot Plant (WIPP). Multiple responses were received from some sites for different facilities or applications (e.g., Emergency Planning versus Safety Analysis). An overview summary of the responses follows in this section. The complete survey responses and tables summarizing the answers to the above questions, grouped by related questions are contained in a separate Official Use Only appendix (Appendix A.2).

2.1 RELEASE SCENARIOS

The potential release scenarios specified included filtered and unfiltered releases from:

- Explosions
- Fire releases
- Spills
- Spray leaks
- Venting
- Container leaks
- Tank waste releases
- Reactor releases (partial or severe cladding breaches, melting reactor fuel elements)
- Criticality incidents
- Glass melter spills and melter off gas releases
- Air/steam overblows through waste
- Chemical releases

The materials involved in the release scenarios covered a wide range of radionuclides and gaseous/particulate properties as summarized below.

2.2 RELEASED MATERIAL

Types of materials potentially released included the following:

- Uranium isotopes
- Transuranic actinides (e.g., Pu and Am isotopes)
- Reactor fission and activation products
- Criticality fission products
- Noble gases
- Tritium (^{3}H)
- Other radionuclides: ^{14}C, ^{59}Ni, ^{60}Co, ^{63}Ni, ^{79}Se, ^{90}Y, ^{93}Zr, ^{93m}Nb, ^{90}Sr, ^{99m}Tc, ^{99}Tc, ^{113m}Cd, ^{106}Ru, ^{125}Sb, ^{126}Sn, ^{129}I, ^{134}Cs, ^{137}Cs, ^{137m}Ba, ^{151}Sm, ^{152}Eu, ^{154}Eu, ^{155}Eu, ^{226}Ra, ^{228}Ra, ^{227}Ac, ^{229}Th , ^{231}Pa, ^{232}Th
- Non-radioactive carbon nanotube aerosols
- Non-radioactive chemical compounds

2.3 CHEMICAL FORMS

Chemical forms of potentially released materials were reported by some of the survey respondents, although others indicated that no particular assumptions were made regarding chemical form. Chemical forms of interest included the following:

- Hydroxides, nitrates and nitrites, elemental gases, metallic hydrides, and oxides
- Oxides, chlorides, or metal alloys for transuranic
- Vapors (usually oxidize before reaching the site boundary)
- Oxides (worst case assumed as per FGR-11[8])
- Chemical state corrected by RSAC[9] model for each radionuclide according to the ICRP-30 designated clearance classes of D, W and Y
- Oxide or unknown chemical matrix for particulate transuranic waste
- Pure isotopic forms
- Low-fired oxides and nitrates for ^{239}Pu
- Tritiated water vapor (HTO)

One respondent indicated usage of the chemical form/solubility that would result in the greatest dose conversion factors according to the biokinetic models. It is possible that other sites are using similar approaches (see Particle Size and Density sub-section below for further discussion).

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[8] EPA (1988). *Limiting Values of Radionuclide Intake and Air Concentration and Dose Conversion Factors for Inhalation, Submersion, and Ingestion*, Federal Guidance Report No. 11 (FGR11), EPA-520/1-88-020 (Oak Ridge National Laboratory, Oak Ridge, TN; U. S. Environmental Protection Agency, Washington, DC)

[9] D. R. Wenzel and B. J. Schrader, *The Radiological Safety Analysis Computer Program (RSAC-6) User's Manual*, INEEL/EXT-01-00540 (2001), Idaho National Engineering and Environmental Laboratory, Idaho Falls, Idaho

2.4 PHYSICAL FORMS

Physical forms assumed for potentially released radionuclides included the following:

- Solid particles
- Liquid particles, including liquid with entrained solid material
- Gases[10] (e.g., noble gases, tritium)
- Vapors

2.5 PARTICLE SIZE AND DENSITY

Particle diameter values reported by survey respondents were typically in the Aerodynamic Equivalent Diameter (AED)[11] range of 1 µm to 10 µm. However, some respondents stated that particle sizes were unknown and in a few cases AEDs outside this range were reported (see list below). Particle density values reported by survey responders were generally in the range of 1.2 to 11.86 g/cc, although a number of respondents again indicated that the density was unknown. The following combinations of particle size and density were provided in individual responses:

- 1-5 µm AED for transuranic solid particles; particle densities of 17-18 g/cc for metals, 4 g/cc for chlorides, and 7-10 g/cc for oxides
- 1 µm AED (if using ICRP-30 methodology for the public and workers) or 1 µm AED for the public and 5 µm for the worker (if using ICRP 66 methodology)
- 1 µm AED (but not specified when using MACCS2 model)
- 1 µm AMAD assumed for MOI (Maximum Offsite Individual) and 5 µm for collocated workers
- 2-4 µm AED with a uniform distribution (for liquid droplets, unfiltered release); larger particles for ground shine component to dose (particle density typically not available but when used ranges from 4 g/cc to 11.86 g/cc)
- 1 µm AED conservatively assumed and 3 µm AED to fit within specified range of HSS Safety Bulletin for Pu-239 assumed to be PuO_2 solid particles
- Less than 10 µm AED with average physical diameter > 50 µm MMD (Mass Median Diameter) for all particles; nominal particle density from 8.22 to 10.95 g/cc
- 2 µm AED to > 1000 µm AED with particle densities of 1.3 g/cc to 1.6 g/cc
- <100 nm physical diameter and variable density depending on particle diameter and length for non-radioactive carbon nanotube aerosols
- Variable particle diameters depending on the waste stream being processed; particle density 1.2 to 2.5 (units unspecified, but assumed to be g/cc)
- Unknown but assumed to be less than 10 µm; particle density unknown

[10] While not explicitly stated by survey responders, iodine isotopes, for example from reactor fission products, can occur in gaseous form.

[11] Aerodynamic Equivalent Diameter is the diameter of a unit density (1 g/cc) sphere that has the same settling velocity as the actual particle

- Unspecified particle diameter; particle density of 11 g/cc for Pu / Pu oxides

It is important to note that assumptions regarding the physical state, chemical form, particle size and particle density of released radioactive material may be implicitly incorporated into the source term and dose conversion factors used in plume modeling and dose calculations. For example, dose conversion factors are based on an assumed chemical solubility and Activity Median Aerodynamic Diameter (AMAD). One site reported assuming a dose conversion factor for ^{239}Pu corresponding to a solubility Type M (typical of low fired oxides, nitrates, etc.), while ^{3}H was taken to be in the form of HTO (tritiated water vapor) which has a dose contribution due to skin absorption. It should also be noted that particle size and density are not specified as inputs when using the MACCS2 code.

2.6 RELEASE HEIGHT

Most release heights were at ground level, but elevated releases were also reported with values in the range of 0 - 76 m above ground level. However, some respondents provided non-specific or variable release heights, e.g., “an elevated release might be assumed” or “ground release with lofting for fires (1 MW fire with a 60 minute release duration was selected as conservatively bounding)”. One respondent reported that all releases were “modeled as ground-level releases since there are no stack heights > 2.5 times the height of the ridges”.

2.7 RELEASE AND EXPOSURE DURATIONS

Reported release durations ranged from 10 minutes to 48 hours. However, one respondent stated that the release duration was taken to be “the full duration of the plume passage or 4 days”, consistent with their exposure duration assumption. Another respondent stated that release duration was “not relevant, since plume meander is not assumed”.[12]

Reported exposure durations ranged from 10 minutes to 4 days. However, some respondents stated that the exposure duration was normally taken to be the release duration, or that 4 days was used for groundshine and resuspension inhalation exposure for consistency with the Early Phase EPA Protective Action Guide (PAG)[13] exposure period of 4 days. Other responses included more details including the use of:

- 2 or 8 hours for radiological exposure and 15 min for toxicological exposure
- 2 hours for radiological exposures, 1 hour for toxicological exposure to uranium, and 15 minutes for other exposures
- Exposure periods of 40 h/week for 10-45 years for safety analysis

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[12] It should be noted that this is not a valid statement if the diffusion methods in a plume model account for plume meander.

[13] EPA, 1991: *Manual of Protective Action Guides and Protective Actions for Nuclear Incident*, Office of Radiation Programs, U.S. Environmental Protection Agency, Washington, DC 20460

2.8 DOSE PATHWAYS

Dose pathways of interest included the following:

- Initial plume inhalation dose
- Ground-shine (ground exposure) dose
- Resuspension inhalation dose
- Cloud-shine (air immersion) dose

Some respondents stated that most analyses were limited to the inhalation pathway (or dominant inhalation pathway), although one respondent indicated that ground-shine was used when appropriate for the given radionuclide.

2.9 DISPERSION CODES

Dispersion codes used by survey respondents were as follows (with the number of respondents using the specified code given in parentheses):

- MACCS2 (8)
- WinMACCS (2)
- HotSpot (3)
- RSAC (3)
- EPIcode[14] (2)
- GXQ (1)
- POSTMAX2 (1)
- ALOHA (1)
- HGSYSTEM/UF6/WAKE (1)
- CHARM (1)

2.10 DEPOSITION METHODS

Dry deposition velocities reported by survey respondents ranged from specification of values based on site conditions or release scenario to methods for calculating v_d as summarized below:

- Specified values for the deposition velocity
  - 1 cm/s for unfiltered releases and 0.1 cm/s for filtered (8 cm/s for non-respirable particles)
  - 0.3 cm/s (value selected as representative of the site)
  - 0.1 cm/s (unmitigated) per HSS Safety Bulletin No. 2011-02
  - 0.1 cm/s default value for particulates and 0 cm/s for gases based on HSS Safety Bulletin No. 2011-02

[14] Note that EPIcode is for chemical releases and does not model radionuclide scenarios.

- 0.1 cm/s for unfiltered releases and 0 cm/s for filtered and gas releases (0.3 cm/s was deemed appropriate, based on the waste streams but selected values used for conservatism)
- 0.01 m/s for radioiodine, 0.001 m/s for particulates, and 0 m/s for noble gases
- 1 cm/s default for particulate Pu with lower values for gaseous releases
- 0 m/sec for gases, tritium vapor (HTO) or other vapors, and 0.01 m/sec per DOE-EH-4.2.1.4 2004 (DOE, 2004) for unfiltered particulates 0 cm/sec for radiological dose calculations

- Methods for calculating deposition velocities included:
  - RSAC 7.2 model default suggested values for ground level releases (more conservative than those in MACCS2 and less than or equal to those suggested by Sehmel), but no deposition for elevated releases
  - RSAC-6.2 model calculated value (however since the inhalation route is the primary concern, plume depletion is generally disabled)
  - GENII V2 model method for calculating site-specific deposition velocity in future (existing analyses use specified fixed values)
  - GENII V2 model calculated site specific value of 0.27 cm/s per the HSS Safety Bulletin
  - HSS Safety Bulletin No 2011-02 methodology based on GENII V2 code method, except for calm conditions when GENII2 uniform transfer resistance is used

2.11 METEOROLOGY

Meteorological data used in plume models by survey respondents ranged from 95th percentile weather to specified or worst case conditions derived from:

- Historical weather data
- Meteorological conditions based on unspecified Nuclear Regulatory Commission (NRC) guidance
- Persistent (specified) meteorological conditions

Other sites collect meteorological data from tower instrumentation at one or more measurement heights (e.g., 2, 10, 50, 60 and 61 m) including:

- Wind speed and wind direction
- Temperature
- Sigma theta (standard deviation of the horizontal wind direction)
- Precipitation

2.12 STABILITY CLASS

Stability class is determined by several means including the following:

- Temperature differential method (in one case explicitly citing the NRC Regulatory Guide 1.23 Delta-T method use of differential between upper and lower level temperature data)
- Stability class derived from "Till and Meyer, 1983" as cited in NOAA ARL FRD, and RSAC model reference
- EPA-454/R-99-005 sigma-theta method (in addition "EPA" methods were mentioned with no reference)
- Solar radiation Delta-T (no reference cited)
- Tadmor and Gur power-law curve fits for the Gaussian plume model horizontal and vertical plume spread parameters, σ_y and σ_z, since release height is ≤ 30 m and receptor distance of interest < 5 km (MACCS2 model)

2.13 SOURCE-TO-RECEPTOR DISTANCES

Reported source-to-receptor and site boundary distances were in the range 30 m - 55 km based on both directionally dependent and independent values. Several respondents stated that 100 m was used for collocated worker exposure distance calculation (one respondent used a range from 30 - 100 m), while several responses focused on the distance to the nearest collocated facilities.

2.14 LAND SURFACE CHARACTERISTICS

Land cover/topography types and surface roughness values and methods are summarized in the table below. Terrain conditions range from flat to mountainous and vegetation from desert brush to forests.

Land cover and topography	Surface roughness values and methods
Flat, desert sage brush	10 cm based on DOE/TIC-27601[15] Table 12.6
	Surface roughness not taken into account; dispersion coefficients not modified
	"Standard" (rural) option in HotSpot model
Desert sage brush	3 cm, which results in a linear scaling factor for the sigma-z function of 1.0 (MACCS2 variable)
	Specific surface roughness value not input but deposition velocity used (interception via vegetation not accounted for)
	RSAC-6.2 model-calculated based on terrain options with agricultural terrain generally selected
Mesquite trees and desert sage brush	10 cm (determined using AERMOD AERSURFACE tool)
High desert plateau – sage brush	Markee dispersion based on empirical data collected at the site for sage brush ground cover

[15] Randerson, D. (ed.), 1984: *Atmospheric Science and Power Production*, DOE/TIC-27601, Available as DE84005177 from National Technical Information Service, U.S. Department of Commerce, Springfield, VA 22161.

Land cover and topography	Surface roughness values and methods
High arid basin with desert grasses and sage; heterogeneous terrain characterized by an alluvial fan with eroded canyons and arroyos	15 cm based on site specific evaluation calculated using a modified Wierenga gustiness method based on data from Sandia meteorological tower network
Forest with interspersed operational areas	(i) Existing: 100 cm (NUREG/CR-4691 Table 2.3) (ii) Future planned: • 160 cm for public receptor based on sonic anemometer measurements of wind speed and wind direction fluctuations • 50-60 cm based on EPA AERSURFACE software and input of national land cover data published by US Geological Survey
"Forested terrain" (as defined by AERSURFACE User manual)	Calculated roughness lengths (z1= 50, 80, and 100 cm taken from tables in Appendix A of AERSURFACE User's Guide) with the same z0 (3 cm which is the MACCS2 default value); with the 50 cm surface length considered applicable to the site environment
Primarily deciduous forest and industrial/urban; topographical effects fall into the "mountain sheltered" terrain with approximately 500 ft. elevation changes	0.328 m to 1.030 m for representative vegetative cover at the site determined by AERSURFACE and used to determine deposition velocity
Mountainous with abrupt and significant elevation changes	38-100 cm
Not used currently	Not used

2.15 SUMMARY

The site survey responses make clear that a wide variety of potential release characteristics and site characteristics are analyzed at DOE sites for safety analysis and emergency planning. The scope of this investigation could not cover the full range of radionuclides, release scenarios, release mechanisms/heights, physical/chemical forms, and environmental conditions identified in the survey.

We therefore used the results of this survey to select the predominant release and site characteristics relevant to investigate. As particulate releases were the most common and inhalation was the pathway of greatest concern, we focused on particles in the respirable-size range (AED < 10 µm). Specifically we used the 2-4 µm and 0.2-0.4 µm size bins for unmitigated and mitigated releases referenced in the HSS Safety Bulletin (HSS, 2011). Additional source and site criteria that may warrant further exploration are discussed in Section 8.3.

As most release heights are specified to be at ground level, near-surface releases were the primary focus of our evaluation. However, as several sites reported releases heights up to 76 m, we performed a few exploratory sensitivity studies for the MACCS2 model that identified some limitations in the code for elevated releases (see Section 5).

For the illustrative case studies in Section 7, we selected two sites that were representative of the range of the different source-to-receptor distances and land-surface characteristics found in the survey responses. These examples were used to explore the relative impacts of downwind distance, surface roughness, atmospheric stability, and deposition velocity in calculating the 95^{th} percentile case.

3.0 LITERATURE REVIEW OF DRY DEPOSITION MODELS

Deposition has the net effect of reducing near-surface downwind air concentrations, while locally increasing surface contamination. Estimates of deposition velocity are therefore critical in plume modeling applications including safety analyses and hazard assessments. We conducted a literature review to identify a suitable deposition model for use in Department of Energy Safety Analyses, with a focus on methods that address mitigated and unmitigated particulate releases (corresponding to Aerodynamic Equivalent Diameters [AED] in the 0.2-0.4 μm and 2-4 μm ranges respectively, as per the HSS Safety Bulletin [2011]). As wet deposition is not considered in DOE hazards analyses, our focus was on dry deposition.

The transfer of gaseous species or particulate material to the ground surface in the absence of precipitation is referred to as dry deposition. Dry deposition operates through a variety of mechanisms and is sensitive to a large number of factors, including release characteristics (e.g., material, particle size, particle shape, density, physical and/or chemical form), meteorological conditions (e.g., wind speed, atmospheric stability, humidity), and the surface environment (e.g., land-surface type, vegetation, surface roughness). Dry deposition has been intensively studied and progress in this field over the last several decades has been extensively documented in the literature (see for example Flechard et al., 2011; Fowler et al., 2009; Petroff et al., 2008a; Pryor et al., 2008; Sportisse, 2007; Garland, 2001; Wesely and Hicks, 2000; Seinfeld and Pandis, 1998; and Sehmel, 1980).

In the 1950s, dry deposition models were based on simple equations for gravitational settling that accounted for particle diameter and density. As they neglected other deposition processes, these models significantly under-predicted deposition rates for smaller-sized particles. Since that time, three generations of dry deposition models have been developed, each of which extended the range of applicability to a wider variety of atmospheric, surface and particle characteristics. Each generation is closely related to a major advance in measurement technology.

First-generation models were developed in the 1970's and early 1980's. They were either heavily empirical or theoretical in nature and were based on limited sets of laboratory and/or field data. Slinn (1982) produced a theoretical framework for deposition modeling still in use today (albeit heavily modified), while Sehmel and Hodgson (1978) formulated an empirical model based on wind-tunnel data. The latter model combined deposition observations for mid-size particles with diffusion and gravitational settling models for fine- and coarse-sized particles. Though a scientifically sound methodology, the majority of data used to develop the model was for low surface roughness environments and therefore did not apply dry deposition in vegetation canopies. However, the model still provides a useful description for simple surfaces.

Second generation deposition models were developed with and validated against a more extensive set of experimental data that became available during the mid-1980's and 1990's, including direct measurements of small particle deposition. These models include more complete descriptions of key physical processes and are in common use in local-area regulatory applications. Examples include EPA's AERMOD[16] and GENII. GENII is at the lower end of sophistication of second-generation deposition velocity models as it lacks parameterizations for deposition processes important for particles diameters of 0.1 - 10 µm. Over that size range, GENII predicts deposition velocities that are much higher than those calculated by more sophisticated models (see Figure 4-1).

Third generation models (Flechard et al., 2011; Petroff and Zhang, 2010) take advantage of significant advances in the number and quality of deposition field measurements over the last decade. These "mechanistic" models explicitly describe deposition processes rather than using empirical fits to data and utilize detailed descriptions of the atmospheric transport processes, particle dynamics, and surface characteristics important in describing deposition onto vegetation. Third generation models are typically incorporated into state-of-the-science global- to regional-scale air quality models.

The following sections provide an overview of the dry deposition process, including a discussion of deposition mechanisms, mathematical parameterizations, and model validation. We highlight agreement and disagreement among experimental results and selected models and briefly discuss areas of uncertainties, but the interested reader is encouraged to consult the references for more detail. Our focus is on particulate matter deposition and omits discussion of gaseous deposition.

3.1 DRY DEPOSITION PROCESSES

A variety of mechanisms contribute to dry deposition. Gravitational settling (the deposition of airborne particles under the influence of gravity) is the dominant contributor for particles with diameters greater than ~10 µm, while other processes dominate deposition rates for smaller-sized particles. Surface-layer aerodynamic processes (turbulent eddies) diffuse particles close to surfaces. In the relative thin layer of still air just above the surface (the "quasi-laminar" layer), impaction, interception, Brownian motion (thermal diffusion), phoretic and related mechanisms all contribute to deposition. Brownian diffusion is the dominant sub-layer transport mechanism for small particles while inertial impaction (and gravitational settling) are the primary deposition processes for larger particles. Once a particle is brought into contact with the surface, the likelihood of it sticking is highly dependent on both particle and surface characteristics.

3.1.1 TURBULENT DIFFUSION

Turbulent motions in the atmospheric surface layer diffuse particles towards the ground where they can deposit. Turbulent diffusion is important for particles with AEDs in the range of 1 to 10 µm that tend to follow the air motion of eddies. For such particles, turbulent diffusion acts as an upper limit to the possible deposition rate of particulate material to the surface when gravitational settling is neglected.

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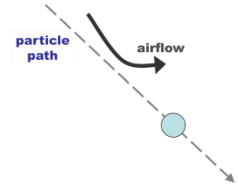
[16] Model and description available for download at:
http://www.epa.gov/ttn/scram/dispersion_prefrec.htm#aermod

3.1.2 IMPACTION

Particles diverge from the mean airflow (streamline) due to inertia. Impaction occurs when such particles pass through the quasi-laminar layer and collide with the surface. Dry deposition via inertial or direct impaction is most efficient for particle with diameters greater than 2 μm that have sufficient momentum to deviate from flow streamlines.



The primary parameter describing the efficiency of impaction is the Stokes number (Fuchs 1964), which is defined as the ratio of the stopping distance of a particle to a characteristic dimension of the obstacle. The Stokes number is given by:

$$St = \frac{2\tau_p U}{d_c}$$

where	τ_p	=	particle relaxation time (s)
	U	=	background fluid velocity away from an obstacle (m/s)
	d_c	=	characteristic dimension of the obstacle (m)

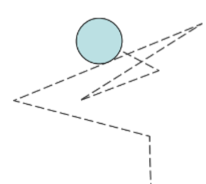
When the Stokes number is much less than 1, the particle motion is highly coupled to the background fluid flow and a deviation from the streamline is unlikely. Conversely, when the Stokes number is large, particles are not as influenced by the fluid motion, since their response time is longer than the period during which the fluid acts upon it.

3.1.3 INTERCEPTION

Deposition by interception occurs when a particle follows the flow streamline but comes within one particle radius of an obstruction resulting in a collision. Interception is an efficient deposition process for particles that are small enough to generally follow the airflow but large enough to come into contact with obstacles (particle diameters in the range of 0.2 – 2 μm). The underlying effectiveness of interception in dry deposition in limited in most cases, but this mechanism may be important for deposition to elevated surfaces and/or surfaces with fine structures such as fuzzy plant leaves.

3.1.4 BROWNIAN MOTION

Small particles are impacted by air molecules, creating random diffusive fluctuations known as Brownian motion or molecular diffusion. On occasion, such diffusion transports particles close enough to the surface so that a collision occurs resulting in deposition. Surface deposition of contaminants results from both non-turbulent Brownian diffusion and turbulent diffusion in the quasi-laminar surface layer next to the surface.



Deposition through Brownian motion has been shown to be most effective for particles in the size range of 0.001 to 0.1 μm, although under certain situations molecular diffusion also may be the predominant dry deposition mechanism for larger particles sizes. However, particles in the smaller size ranges have a greater tendency to adhere to surfaces after collisions due to intermolecular forces.

3.1.5 PHORETIC MECHANISMS

Phoretic mechanisms are the result of gradients in the surrounding environment that affect particle motion. Such mechanisms influence the deposition rates of particles small enough to have high ion mobility or to be impacted by molecular collisions (Droppo, 2006). The magnitudes of phoretic forces are small compared to the deposition processes discussed above and are therefore ignored by the majority of dry deposition models[17]. However, these processes may provide a lower limit on the deposition onto water surfaces[18]. Therefore, a basic description of deposition-related phoretic processes is presented below.

THERMOPHORESIS

Small particles immersed in a gas with a temperature gradient will migrate in the direction of decreasing temperature due to a thermophoretic force. The thermophoretic force is the result of the higher kinetic energy imparted to a particle by air molecules in the warmer region. Small particles in warmer air experience a net force toward cooler surfaces and deposit slightly faster on such surfaces (Hinds, 1999). The rate of movement due to the thermophoretic force depends on the magnitude of the thermal gradient and the properties of the gas and particles.

TURBOPHORESIS

The tendency of particles to move towards regions of decreasing turbulence is referred to as turbophoresis. Since large turbulent velocity fluctuation gradients are found near surfaces, the turbophoresis process leads to enhanced accumulation of contaminants near the surface and increases the rate of deposition onto the surface (Caporaloni et al., 1975; Reeks, 1983; Young and Leeming, 1997).

DIFFUSIOPHORESIS

Particles suspended within a mixture of two gases may experience a diffusiophoretic force that produces a change in deposition rates due to the unequal momentum transfers associated with the different gases. The amount of kinetic energy imparted to a particle by colliding gas molecules depends upon the molecular weight of the gas, with more energy imparted by heaver molecules,

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[17] A notable exception to this rule is the now obsolete EPA Industrial Source Complex (ISC3) model in which a constant deposition velocity of 1×10^{-4} m s^{-1} was added to account for phoretic processes (see http://www.epa.gov/ttn/scram/userg/regmod/isc3v2.pdf).

[18] The Petroff and Zhang [2010] model uses a minimum deposition velocity of 5×10^{-5} m s^{-1} and 2×10^{-4} m s^{-1} for deposition to water and snow surfaces to ensure consistency with measured deposition velocities.

resulting in a net force in the same direction as the heavier gas. The magnitude of the diffusiophoretic force depends on the concentration gradients and the molecular weights and diffusion coefficients of the gases involved. During evaporation, pressure gradients may develop in which vapor moves in one direction while the background air moves in the opposite direction. The inhomogeneous kinetic energy associated with collisions of upward and downward moving gas molecules and particles causes the particles to move in the direction of the diffusion of the heavier gas molecules.

STEFAN FLOW

A more complicated situation occurs when gaseous material (e.g., water vapor) either condenses to or evaporates from a surface. A Stefan flow is established which pushes airborne material towards a condensing surface (increasing deposition rates) or away from an evaporating surface (reducing deposition). This flow is in addition to gradient forces such as thermophoresis and diffusiophoresis.

ELECTROPHORESIS

The movement of charged airborne particles in the presence of an electric field is called electrophoresis (or electrostatic attraction). The direction and strength of the background electrical field and the sign of the particle charge determines the direction of particle motion. Deposition velocities increase under the influence of attractive electrical forces that enhance small particulate transport through the quasi-laminar deposition layer. However, electrophoresis is thought to have minimal impact on deposition rates under the majority of naturally occurring environments (Hicks et al., 1982).

3.2 DRY DEPOSITION MODELS

In the following sections, we discuss commonly-used model formulations of deposition processes, including gravitational settling. We begin with the definition of the deposition velocity and a description of the resistance model approach for particles.

3.2.1 DEPOSITION VELOCITY

The deposition velocity, v_d, provides a convenient construct for characterizing the rate at which airborne material is deposited and for parameterizing the physical processes responsible for deposition. Formally, the deposition velocity is defined to be:

$$v_d(z_r) = -\frac{F}{C(z_r)}$$

where $v_d(z_r)$ = deposition velocity ($m\ s^{-1}$)

F = material flux across a horizontal plane to the depositing surface ($g\ m^{-2}\ s^{-1}$)

$C(z_r)$ = near surface air concentration ($g\ m^{-3}$)

z_r = reference height (m)

Both air concentration and deposition velocity generally vary with height, so v_d is typically defined at a specified reference height (z_r) above ground. For most applications, z_r is chosen so that the deposition flux is approximately constant between the reference height and the ground. However, it is important to note that there are a variety of reference heights in use and care should be taken when comparing reported values of v_d.

The majority of state-of-the-science dispersion models parameterize deposition processes other than gravitational settling via a resistance model (Slinn and Slinn, 1980, Pleim et al., 1984). The resistance model is based on an analogy to electric circuits. A concentration gradient over a surface is thought of as the deposition potential (analogous to voltage) and dry deposition mechanisms are mathematically treated in the same manner as electrical resistance, with the assumption that all deposition processes occur in parallel.

The total dry deposition velocity is calculated by combining the gravitational settling velocity with the resistances for different physical deposition processes and atmospheric layers. For example, following Slinn and Slinn (1980):

$$v_d = (r_a + r_b + r_c + r_a r_b v_s)^{-1} + v_s$$

where
v_d = deposition velocity (m s^{-1})
r_a = aerodynamic resistance across the turbulent near-surface layer (s m^{-1})
r_b = resistance across quasi-laminar sublayer next to the surface(s m^{-1})
r_c = surface resistance (s m^{-1})
v_s = gravitational settling velocity (m s^{-1})

The above formulation is used in the GENII v2 model (Napier et al., 2004).

An alternative formulation developed by Sehmel and Hodgson (1978), and shown by Venkatram and Pleim (1999) to be more exact, is the following:

$$v_d = \frac{v_s}{(1 - e^{-r_t v_s})}$$

where the total resistance, $r_t = r_a + r_b + r_c$. This latter formulation is used in the LODI Lagrangian particle dispersion model that is incorporated into the DOE's National Atmospheric Release Advisory Center (NARAC) system as described by Leone et al. (2001). Resistance models are discussed further in sub-section 3.2.3 below.

It should be noted that deposition velocities are difficult to measure, especially in the case of particles for which the values range over multiple orders of magnitude (Slinn et al., 1978, Sehmel, 1980), a significantly greater variability than occurs for gaseous species. However, a minimum deposition velocity is typically observed in the 0.1 – 1 μm particle size range where deposition mechanisms are not particularly effective (Pryor et al. 2008, Fowler et al. 2009).

3.2.2 GRAVITATIONAL SETTLING

The rate of gravitational settling depends on particle size, density, and shape and is of particular importance for larger particles. In the atmosphere, particles rapidly achieve a terminal settling velocity in which drag forces are equal to the gravitational force. This terminal settling velocity increases rapidly with particulate size since it is a function of the square of the particle diameter. Stokes's Law (Hinds, 1999) can be applied to determine the settling velocity of small spherical particles due to gravity:

$$v_s = \frac{d_p{}^2 g(\rho_p - \rho_g)C}{18\mu}$$

where v_s = settling (terminal) velocity (m s^{-1})
d_p = particle diameter (m)
g = gravitational constant (9.81 m s^{-1})
ρ_p = particle density (g m^{-3})
ρ_g = air density (g/m3)
C = Cunningham correction factor
μ = dynamic viscosity of air (1.81×10^{-5} Pa s^{-1} at 293 K and 1 atm)

The Cunningham correction factor (C) important for small diameter particles is given by (EPA, 1994; EPA, 1995):

$$C = 1 + (2\lambda/d_p)[a_1 + a_x exp(-a_3 d_p/\lambda)]$$

where λ = the mean free path of air molecules (6.53×10^{-6} cm)
a_1 = an empirical constant (1.257)
a_2 = an empirical constant (0.40)
a_3 = an empirical constant (0.55)

When an additional slip correction factor is applied, Stokes's Law has been shown to accurately calculate gravitational settling rates for particles as small as 0.001 µm (Hinds, 1999). Other corrections are needed for particles with diameters larger than 100 µm for which Stoke's Law is no longer valid[19] (Hinds, 1999).

The terminal velocity of a *non-spherical* particle can be determined by including an additional correction factor to account for the difference in air resistance relative to a spherical shape (see Hinds [1999] for more detail). However, in this analysis, we use the Aerodynamic Equivalent Diameter or AED, defined as the diameter of a spherical, unit density (1 g cm^{-3} or 10^3 kg m^{-3})

[19] The large diameter particle corrections have been implemented in models such as NARAC's operational dispersion model LODI.

particle that has the same settling velocity as the particle in question. Therefore this correction is not used in the following evaluation.

Most authors recommend the formulation for the gravitational settling velocity given above (see for example Sportisse, 2007; Seinfeld and Pandis, 1998). This approach is used in the EPA's AERMOD (EPA, 2004; Walcek et al., 2001; Wesely et al., 2001) and CALPUFF (Scire et al., 2000) codes, the DOE Toolbox model GENII-V2 (Napier et al., 2004), and the DOE LODI model (Leone et al., 2001). It should be noted that since gravitational settling is relatively independent of environmental and surface conditions, gravitational settling forms a lower bound on the overall deposition velocity.

3.2.3 RESISTANCE MODELS

The remaining deposition processes are typically parameterized via a resistance model in which depositing particles are assumed to sequentially (a) move through the atmosphere to a location very near a surface, (b) pass through a thin "quasi-laminar" layer of relatively still air just above the surface, and (c) deposit onto the surface. The degree of "resistance" to particle deposition at each stage is combined to calculate a total resistance to deposition according to:

$$\mathrm{r}_t{}^{-1} = (\mathrm{r}_a + \mathrm{r}_b + \mathrm{r}_c)^{-1}$$

where

r_t	=	total resistance (s m^{-1})
r_a	=	aerodynamic resistance (s m^{-1})
r_b	=	quasi-laminar sublayer resistance (s m^{-1})
r_c	=	surface resistance (s m^{-1})

The dominant deposition mechanisms and the generally agreed upon parameterizations for each resistance term are discussed below. These parameterizations assume a quasi-flat surface. The applicability of this approach to deposition in natural environments, some of which are comprised of multiple types of surfaces (e.g. grasslands, forests) is discussed in Section 3.3.

AERODYNAMIC RESISTANCE

The aerodynamic resistance, r_a, parameterizes the impact of diffusion from turbulent eddies in the surface layer on deposition rates. Wesely and Hicks (1977) determined r_a across the atmospheric surface layer by integration of micrometeorological flux-gradients relationships. Their formulation for the aerodynamic resistance is given by:

$$\mathrm{r}_\mathrm{a}(u_*, u) = \frac{\mathrm{u}(z_r)}{u_*^2}$$

where

$u(z_r)$	=	wind speed at z_r m above ground level (m s^{-1})
u_*	=	friction velocity (m s^{-1})

z_r = reference height (m)

Although the values of u(z_r) and u_* can be obtained from measurements or a meteorological model, they are most commonly calculated from Monin-Obukhov similarity theory:

$$u(z_r) = \frac{u_*}{k}\left(ln\left(\frac{z_r - d}{z_o}\right) - \psi_M\left(\frac{z_r - d}{L}\right) + \psi_M\left(\frac{z_o}{L}\right)\right)$$

where

k	=	von Karman constant (0.4)
z_r	=	wind speed reference height (m)
z_o	=	surface roughness height (m)
d	=	zero-plane displacement height (m)
ψ_M	=	Monin-Obukhov stability correction accounting for the effects of atmospheric stability
L	=	Obuhkov length, a measure of atmospheric stability (m)

It should be noted that several alternate forms for ψ_M exist, including those of Dyer and Hicks, 1970; Paulson, 1970; and Dyer, 1974.

The parameterization for aerodynamic resistance is generally believed to be valid for transport above surfaces (see reviews by Petroff et al., 2008a; Sportisse, 2007; Seinfeld and Pandis, 1998). It is operationally used in the EPA AERMOD, EPA CALPUFF, EPA GENII-v2, and NRC RACHET2, and NRC RASCAL 4 models. However, it should be noted that this parameterization is *not* valid for air transport within a layer of air in which significant deposition occurs (e.g., within a forest canopy), although it is valid *above* that layer.

QUASI-LAMINAR SURFACE LAYER RESISTANCE

The quasi-laminar surface layer resistance, r_b, parameterizes deposition resulting from impaction, Brownian diffusion, and interception in the quasi-laminar sublayer, a very thin layer of nearly still air just above the surface. While there rarely exists a single laminar boundary layer and for some surfaces such layers only exist intermittently (e.g., leaves buffeted by wind), the quasi-laminar sublayer concept is useful in describing the net effect of deposition processes very near the surface (Seinfeld and Pandis, 1998). A variety of theoretical and semi-empirical parameterizations are employed in commonly-used dispersion models to calculate r_b, a few examples of which are described below. However, significant controversy remains as regards to which mechanisms dominate for various particle sizes, atmospheric conditions, and surface characteristics due to the limitations of both measurements and models (Fowler et al., 2009).

The CALPUFF dispersion model (Scire et al., 2000) parameterizes the resistance across the quasi-laminar sublayer according to Seinfeld and Pandis (1998) and Pleim et al. (1984) as:

$$r_b = \frac{1}{u_*(Sc^{-2/3} + 10^{-3/St})}$$

where Sc = Schmidt number ($Sc=\upsilon/D_B$)

υ = kinematic viscosity of air ($\sim 0.1505\times10^{-4}$ m^2 s^{-1})

D_B = Brownian diffusivity of particle (m^2 s^{-1})

St = Stokes number [$St = (v_s/g)\,(u_*^2/\upsilon)$, dimensionless]

g = gravitational acceleration (9.80616 m s^{-2})

Slinn (1980) calculates the quasi-laminar resistance over vegetation (stated to be applicable to "all vegetative snow free surfaces and for coniferous forests in all seasons") using a method that incorporates the convective mixing velocity:

$$r_b = \frac{1}{u_*\left(1 + 0.24\frac{w_*^2}{u_*^2}\right)\left(Sc^{-2/3} + \frac{\bar{St}}{1 + \bar{St}^2}\right)}$$

where

$\bar{St}$ = average Stokes number, $\bar{St} = v_g u_*/(gc\,\dot{A})$

c = numerical factor close to unity

u_* = friction velocity (m s^{-1})

w_* = convective velocity scale (m s^{-1})

AERMOD (Cimorelli et al., 2003) uses a second parameterization provided by Slinn (1980), which is valid for non-coniferous vegetation outside the growing period:

$$r_b = \frac{1}{u_*\left(1 + 0.24\frac{w_*^2}{u_*^2}\right)(Sc^{-2/3} + 10^{-3/St})}$$

Wesely (1989) suggests an alternate formulation:

$$r_b = 5\frac{Sc^{2/3}}{u_*}$$

while GENII (Napier 2010) and RATCHET2 (Ramsdell, et al. 1994) use a simpler treatment of the quasi-laminar sublayer resistance without the Schmidt number:

$$r_b = \frac{2.6}{ku_*}$$

Other formulations have been developed for deposition onto bodies of water (e.g., Slinn and Slinn, 1980; van den Berg et al., 2000) but these will not be reviewed here as they have limited applicability to DOE site safety analyses.

SURFACE RESISTANCE (RC)

The surface resistance, r_c, is a measure of the degree to which depositing particles stick to the surface. This resistance term is the least well understood of those considered in this report due to considerable measurement and theoretical uncertainties. Small particles are known to adhere to surfaces with which they come into contact due to van-der-Waals forces (Hinds, 1999).[20] This is particularly true for liquid particles and/or wet or sticky surfaces because such particles/surfaces can deform to (a) absorb the kinetic energy of the depositing particle and/or (b) maximize the surface contact area (enhancing the van-der-Waals forces). In contrast, solid particles may rebound from surfaces if they deposit with sufficient kinetic energy (speed). This effect has been observed in laboratory studies and may be important for larger particles (see discussion and references in Petroff and Zhang, 2010 and Petroff et al., 2008a).

For the particle sizes of interest in this report, most current references and operational models assume $r_c = 0$ so that all depositing particles stick to the surface[21] (Sportisse 2007; EPA, 2004; Walcek et al., 2001; Wesely et al., 2001; Scire et al., 2000; Seinfeld and Pandis, 1998). In contrast, the related GENII-V2, RACHET 2, and RASCAL 4 models assume r_c to be equal to 100 s m^{-1} in order to incorporate overall deposition velocities "more consistent with reported values" [22] (Ramsdell et al., 2010; Ramsdell and Rishel, 2006; Napier et al., 2004).

It is worth noting that historically r_c has not always been assumed to be zero. Most notably Slinn (1982) and later Zhang (2001) used a collection efficiency parameterization that implied a significant fraction (~0.5) of depositing small (~1 μm) particles were not retained on surfaces, although this factor was not used in later refinements of the latter's deposition model (Petroff and Zhang, 2010). Specifically, Slinn (1982) used a formulation for r_c to account for enhanced aerosol impaction due to dense vegetation, while Zhang (2001) calculated the surface resistance (r_s) term as:

$$r_c = \frac{1}{\varepsilon_o u_* (E_B + E_{IM} + E_{IN}) R_1}$$

where ε_o = empirical constant (a value of 3 is used for all land use types)
R_1 = fraction of particles that stick to surface
E_B = Brownian diffusion collection efficiency

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[20] This assumption is often not valid for the deposition of gases for which r_c may be a dominant process.

[21] Resuspension/reaerosolization of material can occur following deposition. While this topic is beyond the scope of this literature review, the interested reader is encouraged to refer to Maxwell and Anspaugh, 2011; Burrows et al., 2009; Nicholson, 2009; Nicholson, 1988; Jones and Harrison, 2004; Smith and Jones, 2000; and Sehmel, 1980b.

[22] As with their parameterization of the quasi-laminar boundary layer resistance, no experiments or theoretical studies are cited for this choice.

E_{IM} = impaction collection efficiency
E_{IN} = interception collection efficiency

Examples of formulations for the collection efficiencies for various surfaces are given in Table 3-1.

Table 3-1. Collection efficiencies for various surface types

$E_B = Sc^{-\gamma}$	γ = 1/2 for water and 2/3 for vegetation
$E_{IM} = 10^{-3/St}$	smooth surfaces (Slinn, 1982)
$E_{IM} = \frac{St^2}{400 + St^2}$	smooth surfaces with bluff roughness elements, Giorgi (1986)
$E_{IM} = \frac{St^2}{1 + St^2}$	vegetative canopies (Slinn, 1982)
$E_{IM} = \left(\frac{St}{\alpha + St}\right)^{\beta}$	vegetative canopies α and β are land use category dependent constants, recommended approach (Peters and Eiden,1992)
$E_{IM} = \left(\frac{St}{0.6 + St}\right)^{3.2}$	vegetated surfaces (Giorgi, 1986)
$E_{IM} = \frac{St^3}{St^3 + 0.753St^2 + 2.796St - 0.202}$	Grassland (Davidson et al. 1982)
$E_{IN} = \frac{1}{2}\left(\frac{d_p}{A}\right)^2$	d_p = particle diameter, A is the characteristic radius dependent on land use type and season
$R_1 = \exp\left(-St^{1/2}\right)$	(Giorgi, 1988)

3.3 KEY PARAMETERS CONTROLLING DEPOSITION VELOCITY

Many of the resistance model parameterizations discussed in Section 3.2.3 assume deposition is occurring on a uniform quasi-flat surface. However, the natural environment is far more complex and comprises diverse surfaces types and orientations that produce significant changes in deposition velocities. For example, small particle deposition in heavily forested regions may be orders of magnitude more efficient than deposition on water surfaces or bare earth due to (a) the increased surface area onto which particles can deposit, (b) modifications of the airflow and particle motion within the deposition layer, and (c) changes in the efficiency of quasi-laminar boundary layer deposition mechanisms (Petroff and Zhang, 2010). This section discusses how deposition behavior and phenomenology is affected by key parameters, including the interplay between particle size and atmospheric turbulence and surface characteristics (land-use).

3.3.1 PARTICLE SIZE

Deposition velocities vary strongly with particle size (typically expressed in terms of the Aerodynamic Equivalent Diameter or AED). Figure 3-1 provides a qualitative illustration of the standard model of deposition behavior as a function of particle size, with primary deposition mechanisms labeled for each size range. Relatively high deposition velocities of 0.1 to 10 cm s^{-1} are observed for particles with AED > 5 μm as gravitational settling and impaction are efficient mechanisms for depositing particulate material. Deposition velocities typically reach a minimum (v_d < 0.01 cm s^{-1}) for particles in the size range AED = 0.1 – 1 μm, for which gravitational settling, Brownian diffusion, interception and turbulent eddy diffusion are not efficient processes. Deposition velocities increase to around 0.01 to 0.1 cm s^{-1} for particle with smaller AEDs (0.01 – 0.1 μm) due to the increased effectiveness of Brownian diffusion.

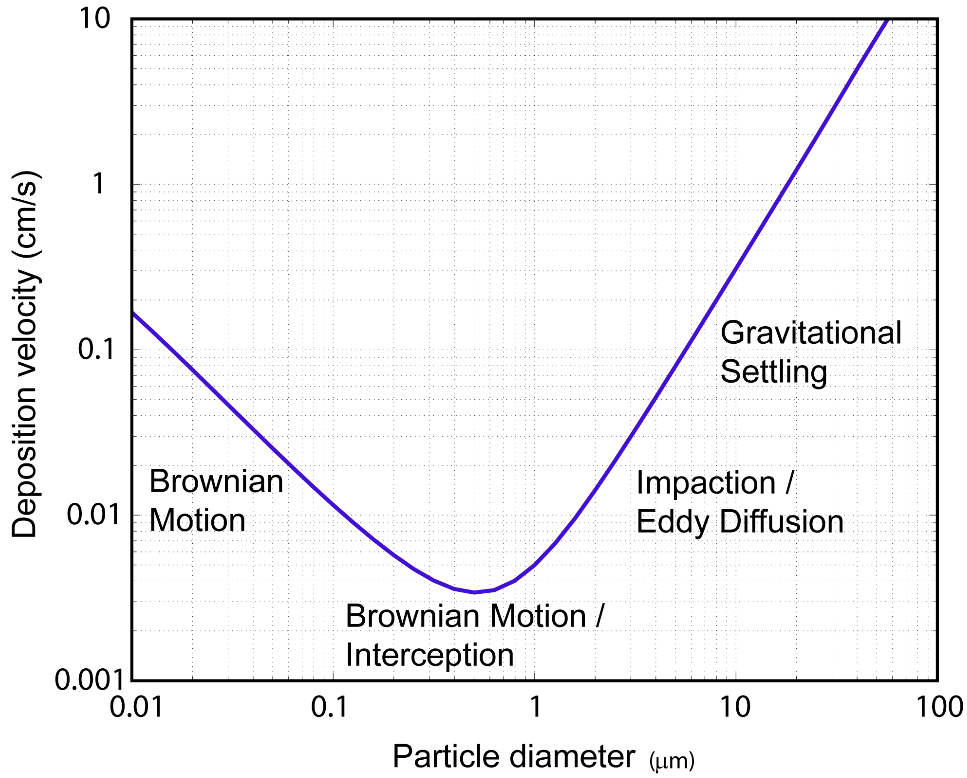
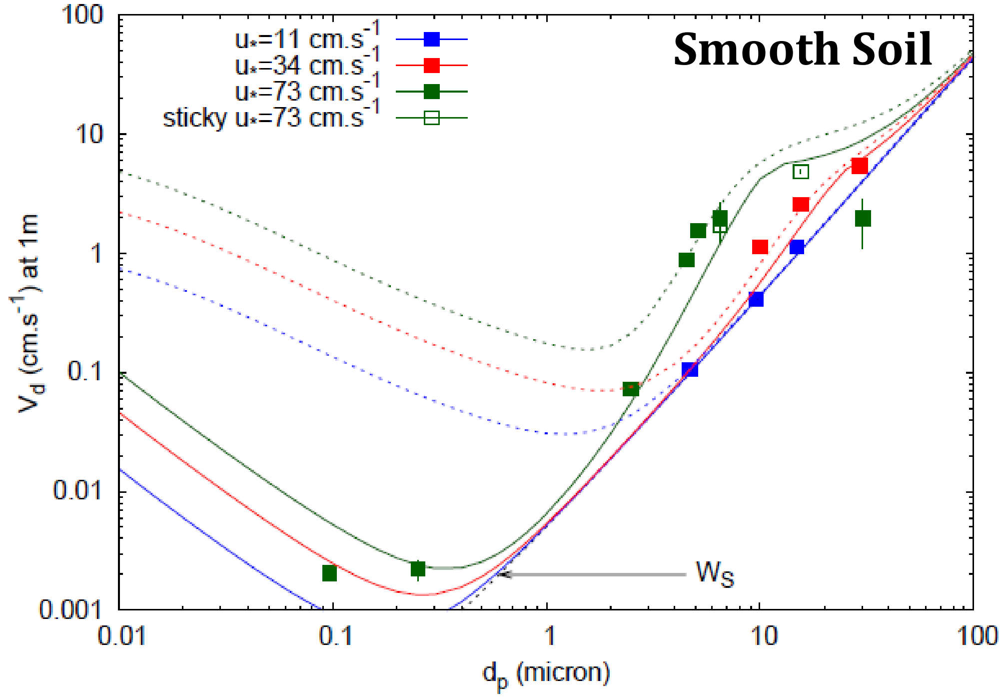


Figure 3-1. Schematic illustration of dry deposition velocity (cm s^{-1}) behavior as a function of Aerodynamic Equivalent Diameter (AED)

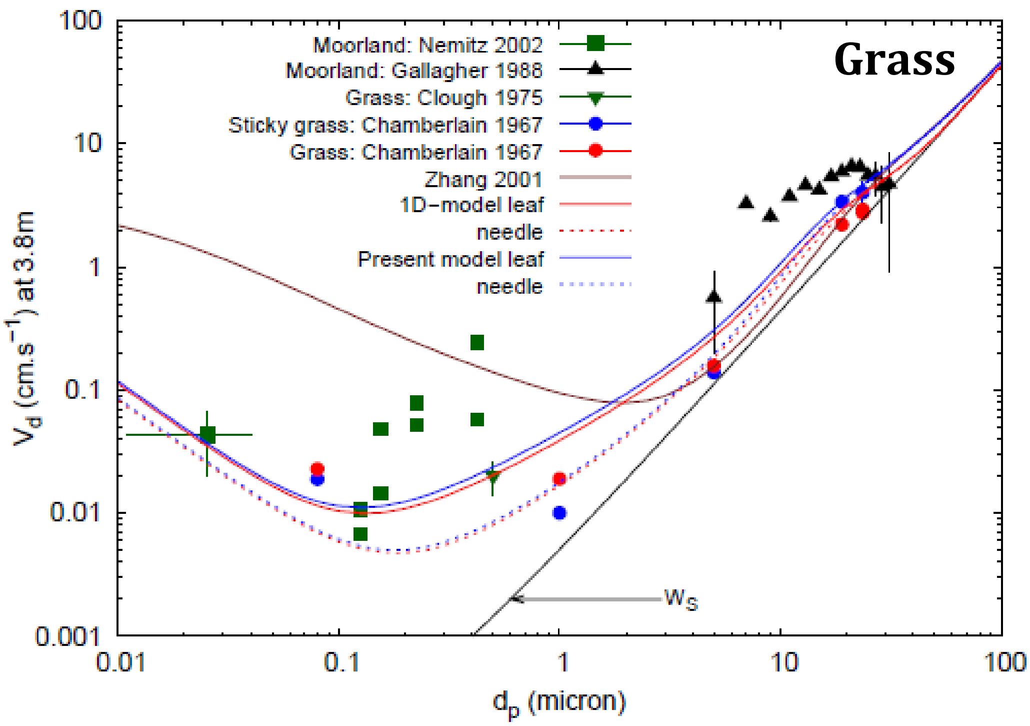
Although Brownian motion and gravitational settling are well-known to dominate small and large particle deposition, respectively, the dominant deposition mechanism(s) for particles of intermediate AED (in the range of 0.1 – 1 μm) depend strongly on particle size, surface characteristics, and atmospheric conditions and may deviate significantly from the qualitative behavior shown in Figure 3-1. This variation in behavior is shown for three land-use cases in Figures 3-2. As an example, note that several experiments have shown minimal to no change in deposition velocity for heavily vegetated surfaces (e.g., dense forests and some grasslands) over the 0.1 to 1 micron particle size (Figure 3-2c).

a)



Smooth Soil
u*=11 cm.s-1
u*=34 cm.s-1
u*=73 cm.s-1
sticky u*=73 cm.s-1
Vd (cm.s-1) at 1m
dp (micron)
WS
100
10
1
0.1
0.01
0.001
0.01
0.1
1
10
100

b)



Grass
Moorland: Nemitz 2002
Moorland: Gallagher 1988
Grass: Clough 1975
Sticky grass: Chamberlain 1967
Grass: Chamberlain 1967
Zhang 2001
1D-model leaf
needle
Present model leaf
needle
Vd (cm.s-1) at 3.8m
dp (micron)
WS
100
10
1
0.1
0.01
0.001
0.01
0.1
1
10
100

c)

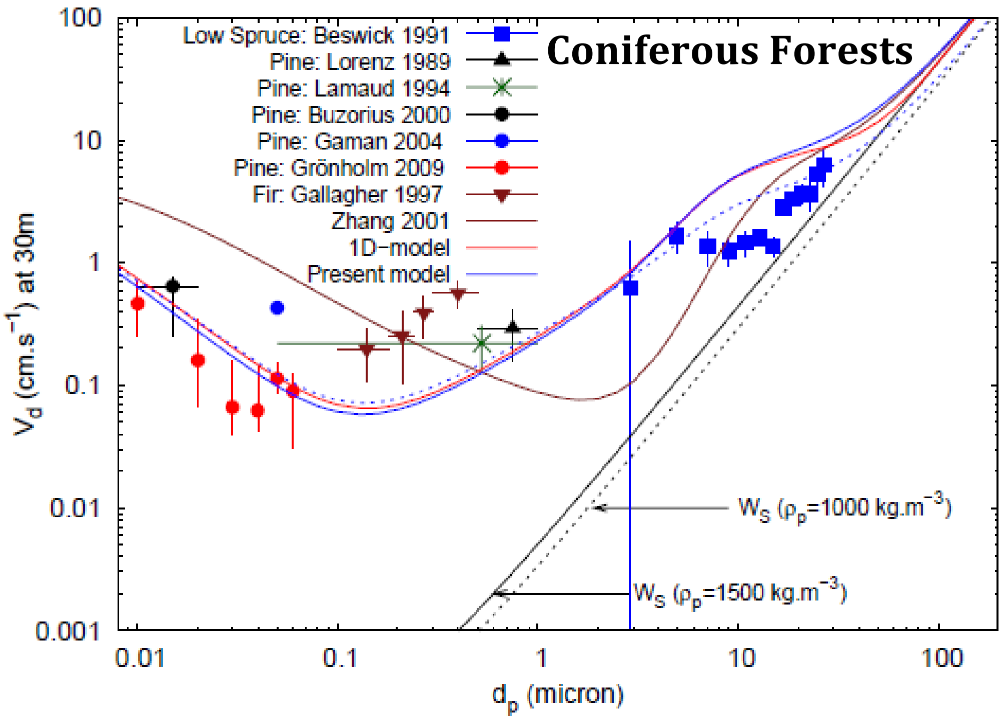


Figure 3-2. Dependence of deposition velocity on particle diameter for smooth soil (a), grass (b) and coniferous forests (c). Gravitational settling is shown by the w_s curve. Petroff and Zhang (2010) model results are labeled with solid lines labled (a) "Present model leaf" (b) and "Present model" (c). Symbols show measured values with uncertainty ranges shown as lne segments. Graphics reproduced from Figures 3, 4, and 5 (Petroff and Zhang, 2010).

3.3.2 ATMOSPHERIC TURBULENCE

Mechanical and thermal turbulence are known to increase deposition rates. The intensity of these two types of turbulence is measured by the friction (u_*) and convective (w_*) velocities, respectively.

$$u_* = \frac{\mathrm{k\,u}(\mathrm{z}_r)}{\left(ln\left(\frac{z_r - d}{z_o}\right) - \psi_\mathrm{M}\left(\frac{z_r - d}{L}\right) + \psi_\mathrm{M}\left(\frac{z_o}{L}\right)\right)}$$

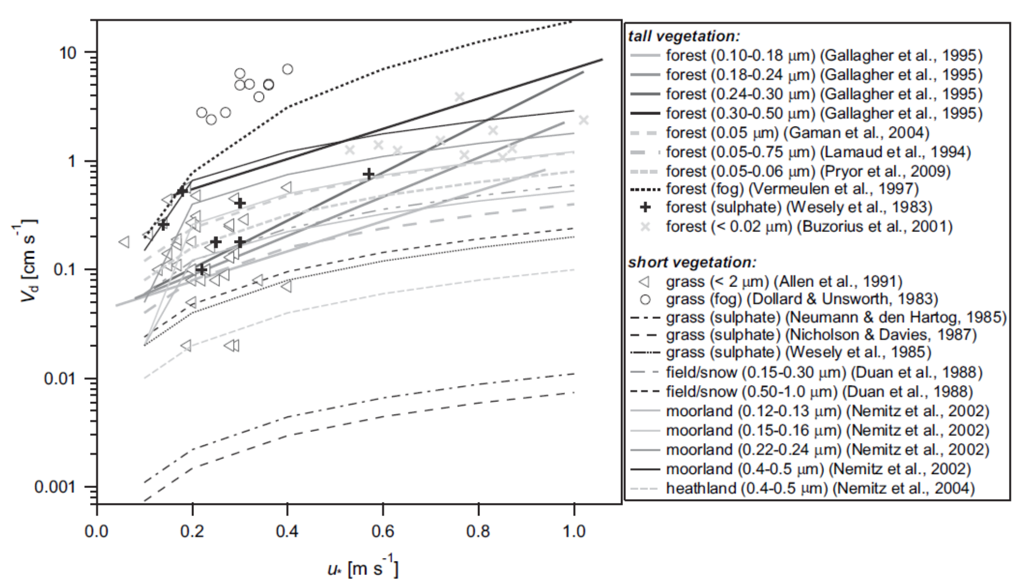
$$w_* = \left(\frac{\mathrm{g\,H\,z}_{ic}}{\rho\, C_p\, T_{ref}}\right)^{1/3}$$

where	H	=	surface sensible heat flux (W m^{-2})
	z_{ic}	=	convective mixing layer height (m)
	ρ	=	air density (kg m^{-3})
	C_p	=	specific heat of dry air under constant pressure (1004 J K^{-1} kg^{-1})

T_{ref}	=	ambient temperature representative of the surface layer (K)

Mechanical turbulence is generated when air flows over, around, or through objects (analogous to the eddies created by water flowing around a rock in a stream) and increases with increasing wind speeds and roughness of the surface v_d (Figure 3-3a). Mechanical turbulence raises deposition rates by (a) increasing transport within the atmosphere, (b) increasing particle drag, and (c) reducing the depth of the quasi-laminar boundary layer. This behavior of is generally captured in deposition models.

a)



b)

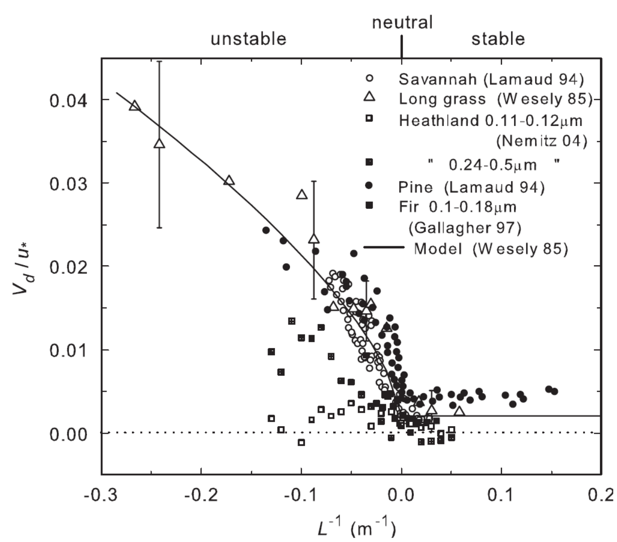


Figure 3-3. Dependence of deposition velocity on a) mechanical and b) thermal turbulence. Panels reproduced from Figure 7.4 in Fowler et al., 2009 and Figure 5 in Petroff et al., 2008a, respectively.

Thermal turbulence is generated when warm air near the earth's surface rises and is replaced with colder air aloft (analogous to eddies created in boiling water). It is well established that deposition velocities increase under conditions of enhanced thermal turbulence (unstable atmospheric conditions), although few studies have been able to quantify this effect (Figure 3-3b). While several empirical parameterizations of the processes exist (see Pryor et al., 2008), they lack a firm theoretical basis and are based on the relatively limited experimental datasets.[23]

3.3.3 ATMOSPHERIC STABILITY

Atmospheric stability is a measure of the resistance of the atmosphere to vertical motion. Strong vertical updrafts and enhanced turbulent mixing in the planetary boundary layer associated with unstable atmospheric conditions produce lower plume concentrations and greater plume spread, diluting air concentrations. Conversely, stable atmospheric conditions are characterized by weak vertical updrafts and reduced turbulence leading to less plume spreading and increased air concentrations. Enhanced near-surface concentrations in turn affect deposition rates.

Atmospheric stability is often specified in terms of Pasquill-Gifford (P-G) stability classes (Pasquill, 1961; Gifford, 1961). P-G stability classes range from A to F, with A corresponding to highly unstable conditions and F representing highly stable (least diffusive) atmospheric conditions. Stability class is an important input to Gaussian plume models where it is used to parameterize the degree of horizontal and vertical plume spread (see Hanna et al., 1982, for additional information).

Numerous methods have been developed over the last several decades to determine atmospheric stability from both routine and high-fidelity weather observations. The primary difference among the various estimation methods is the set of meteorological inputs required. A brief description of a number of commonly used methods for determining stability class is provided below.

TURNER'S METHOD

The Turner method (Turner, 1964; Turner, 1994) for determining the Pasquill-Gifford stability class is widely used since it only requires meteorological variables that are routinely measured at National Weather Service (NWS) stations – horizontal wind speed at 10 m, cloud cover, cloud ceiling height, and solar zenith angle[24]. The primary limitation of this method is the potential lack of representative meteorological data at the site of interest (e.g., the nearest NWS station data may not reflect conditions at the site of interest). Turner's method is recommended for estimating stability in EPA guidelines (EPA, 2000), although alternative approaches such as the Sigma Theta or Temperature Difference method (discussed below) also may be used for regulatory modeling when representative cloud cover and ceiling data are unavailable.

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23 The EPA AERMOD model corrects for this effect using an early parameterization based on observed deposition velocities over grasslands.

24 The solar angle can be calculated from the site latitude and time of year.

SOLAR RADIATION / DELTA TEMPERATURE (SRDT) METHOD

The SRDT approach described by Bowen et al. (1983) provides a physical basis for stability estimation that is similar to Turner's method but does not require cloud ceiling and cloud cover observations. Daytime inputs to the SRDT method include the horizontal wind speed (at or near 10 m) and observations of solar radiation. At night, the meteorological input to the SRDT method is the near-surface vertical temperature gradient. The main drawback of the SRDT method is the limited availability of onsite solar radiation observations.

NRC TEMPERATURE DIFFERENCE (ΔT) METHOD

Ambient air temperature observations at two different heights on a tower are used to calculate the vertical temperature gradient in the ΔT method (NRC, 2007). The recommended measurement heights for air temperature are at 10 m and 60 m on the same tower. Once computed, the P-G stability class is determined from the temperature gradient using a lookup table (reproduced in Table 6-2). The ΔT approach is the method preferred by the NRC for estimating the atmospheric stability class at nuclear power plants because it is considered to be an 'effective indicator' of worst-case (i.e., highly stable) atmospheric conditions.

SIGMA THETA METHOD

The sigma theta (σ_θ) method correlates the standard deviation of the horizontal wind direction to the P-G stability class. High σ_θ values are associated with large horizontal turbulence (unstable) conditions while small σ_θ values are observed in stable conditions during which horizontal turbulence and plume spreading are reduced. The σ_θ method requires meteorological data at high temporal resolution to accurately calculate the standard deviation of the wind direction over the recommended sampling period of 15 to 30 minutes. A correction table based on the mean scalar wind speed is used to adjust the initial estimate of the P-G stability class derived from the determination of σ_θ. In the Modified σ_θ method (Mitchell and Timbre, 1979), an additional correction factor is applied to account for the absence of solar insolation during nighttime. The σ_θ method is recommended for stability class estimation by both the EPA (EPA, 2000) and NRC (NRC, 1980).

RICHARDSON NUMBER

When multi-level wind speed and temperature observations are available from a meteorological tower, the Richardson number method can be used to estimate the P-G stability class (see Sedefian and Bennett, 1980). The Richardson number is a measure of the ratio of buoyant production of turbulent energy to the mechanical production of turbulence. Once calculated, the Richardson number is correlated to a P-G stability class via a lookup table. Either the gradient and bulk Richardson number may be utilized in this approach.

WIND SPEED RATIO METHOD

Sedefian and Bennett (1980) proposed an alternative metric for estimating atmospheric stability using the ratio of the horizontal wind speed at a reference height (~50 m) to the wind speed at 10 m. The physical basis underlying this approach is the contrast between the well-mixed nature of the lower atmosphere during unstable conditions (during which the 10 m wind speed is expected to be reasonably close to the wind speed at the elevated reference height) versus the poor mixing under stable conditions (so that the elevated wind speed may be 2 to 3 times the magnitude of the 10 m wind speed). As with other methods, the P-G stability is estimated using a lookup table that correlates the wind speed ratio to a stability class.

SIGMA PHI AND SIGMA OMEGA METHODS

Both the sigma phi (σ_ϕ) and sigma omega (σ_ω) methods correlate vertical wind fluctuations in meteorological data to the P-G stability class. The σ_ϕ method utilizes the standard deviation of the elevation angle of the vertical wind direction while the σ_ω method uses the standard deviation of the vertical wind speed. The primary limitation of these methods is the need for a well-maintained calibrated observing system producing high temporal resolution data.

OBUKHOV LENGTH METHOD

The Obukhov length, L, is a similarity-theory turbulence scaling parameter that describes the relationship of thermally-generated turbulent kinetic energy (TKE) to mechanical (wind shear) production of TKE. Specifically, the absolute value of the Obukhov length scale is the height at which buoyant TKE production is equal to wind shear driven TKE production. The Obukhov length provides an excellent physical basis for quantifying surface layer stability and can be used to define the P-G stability class (Sykes and Lewellen, 1992). Required inputs are the horizontal wind speed, surface roughness length, temperature, and sensible heat flux. Unfortunately, observations of sensible heat flux are not typically available due to the expense of acquiring and maintaining the necessary measurement sensors. Even when solar insolation data are available, estimating the heat flux can be difficult since assumptions about soil moisture content have a significant impact on the energy balance calculation.

COMPARISON OF ATMOSPHERIC STABILITY METHODS

The available methods to estimate the P-G stability class from meteorological observations differ in their focus on either horizontal (e.g., σ_θ method) or vertical (e.g., ΔT method) measures or indicators of turbulence and therefore varying requirements for meteorological input data. The EPA and NRC preferred stability class estimation approaches are the Turner, σ_θ, and ΔT methods.

Various studies have been performed on the sensitivity of Gaussian plume modeling results to the choice of stability class estimation method. Miller and Little (1980) found that the ΔT method provided the best choice for insuring conservatism in the calculation of $\chi u/Q$ (the air concentration dilution factor χ/Q multiplied by the wind speed u), although the σ_θ method produced the most 'realistic' values of $\chi u/Q$. Gaussian model dispersion predictions based on stability class derived from the Modified σ_θ method were shown to be in general agreement with concentrations obtained using the ΔT and Turner methods for ground-level releases (Mitchell, 1982). Scott-Waslikand and Kumar (1982) found the P-G stability class estimated using the σ_θ and ΔT methods were within one class of each other more than 80% of the time. Use of the σ_θ method without correcting for the absence of insolation at night was shown to result in lower, and more variable short-term (1 hour), estimates of χ/Q than those derived from the ΔT method (Mitchell, 1982; Mitchell and Timbre, 1980).

3.3.4 LAND-USE AND SURFACE CHARACTERISTICS

Although numerous uncertainties remain, recent investigators have compiled and analyzed a significant number of field measurements and modeling approaches for different land-use / surface characteristic types (Petroff and Zhang, 2010; Pryor et al., 2008; Petroff et al. 2008a). The results of these efforts have raised our level of understanding of a number of key factors that affect deposition velocities in natural environments.

Dry deposition velocities generally increase as the available surface area onto which particles can deposit grows. Regions covered by vegetation may possess a leaf surface area that exceeds the underlying ground area by factors of five (Petroff and Zhang, 2010; Fowler et al., 2009), with forests typically having greater leaf surface areas than shorter vegetation (e.g., crops, grassland). Leaf morphology and orientation are also believed to affect deposition with broad leaves expected to be 3 to 5 times more efficient in collecting small particles than needle-like leaves (Petroff et al., 2009; Davidson et al., 1982). Figure 3-2 illustrates the effects of vegetation on v_d and shows the minimum deposition velocity varying from 0.001 cm/s for bare soil (Figure 3-2a) to 0.1 cm/s for forest (Figure 3-2c).

Since Brownian diffusion dominates the deposition of particles with small AEDs and gravitational settling is the primary deposition mechanism for larger aerosol diameters, it is in the intermediate size or "accumulation mode" range (defined as AEDs between 0.1 – 1 μm) that the influence of vegetation canopies has the greatest impact on deposition velocities. The accumulation range also exhibits the greatest variability and uncertainty due to the limited availability of measurements that can be used to parameterize the influence of different vegetation canopies on deposition rates. The state of knowledge regarding deposition of intermediate diameter particles is discussed below for several common land-use categories.

WATER

There are few studies of deposition onto water surfaces or parameterizations of the related quasi-laminar boundary layer transport processes. From the limited information available, there appears to be three distinct categories of water surfaces: calm water (lakes), oceans, and snow/ice. Measured deposition velocities are relatively low for all three surface types for intermediate size particles. Values of v_d for water surfaces range from 0.004 to 0.02 cm s^{-1}, with the variability depending on differences in the friction velocity over the observing period (Moller and Schumann, 1970; Sehmel and Sutter, 1974; Zufall et al., 1998; Caffrey et al., 1998). Measurements of deposition onto snow/ice yield slightly higher values of around 0.02 to 0.04 cm s^{-1} (Ibrahim et al., 1983; Duan et al., 1988).

BARE EARTH

Deposition velocities for smooth soil surfaces are small in the absence of a vegetation canopy to increase aerosol impaction, since Brownian diffusion and gravitational settling have minimal impact on particles in the intermediate ("accumulation mode") size range. Sehmel (1973) measured deposition velocities of 0.002 cm s^{-1} for aerosol diameters between 0.1 and 0.3 microns. However, Petroff and Zhang (2010) noted that bare ground differs from the smooth surfaces associated with observational studies and stressed the need for additional measurements to increase the confidence of deposition velocity modeling for bare ground.

SHORT VEGETATION (GRASSLANDS)

The short vegetation canopies found in grasslands provide additional surface area for aerosol deposition. Measured deposition velocities for grassland range from 0.007 to 0.2 cm s^{-1} (Chamberlain 1967; Nemitz 2002) for particles with intermediate AEDs. The variation in observed deposition velocity values results from changes in the friction velocity during the observation period. In general, deposition velocity values are 1 to 2 orders of magnitude greater over grassland than over bare ground.

TALL VEGETATION (FORESTS)

Forest canopies present a large surface area for aerosols to impact, enhancing deposition rates relative to bare ground or grasslands. Deposition velocities for intermediate size particles have been found to range from 0.2 to 0.7 cm s^{-1} (Lorenz and Murphy, 1989; Lamaud et al., 1994; Gallagher et al., 1997).

URBAN

There have been relatively few studies of particulate deposition in urban environments, although measurements of v_d above the urban canopy demonstrate that these areas are often net emitters of small particles and that resuspension may play an important role in counteracting the loss of particulate matter from the atmosphere (Fowler et al. 2009). Holsen et al. (1991) calculated an average deposition velocity of 0.5 cm/s for Chicago and other urban areas near the Great Lakes.

Roed (1990) compiled radioactive particle deposition from the Chernobyl accident and nuclear weapons testing in urban areas. These data have typically not been incorporated into compilations of deposition measurements. Roed demonstrated that urban vegetation (e.g., trees, grass) was highly efficient in removing particulate radionuclides ($v_d = \sim10^{-1}$ cm s^{-1}) while other surfaces were usually less efficient (e.g., vertical walls for which the deposition velocity was estimated to be $\sim10^{-3}$ cm s^{-1}).

3.4 SUMMARY OF RESULTS OF LITERATURE SURVEY

This literature survey discusses some of the phenomenological complexity associated with deposition processes. Due to lack of a complete set of data covering all scenarios of interest and the limited understanding of some key aspects of deposition, there currently is no single accepted theoretical description of deposition that covers all common natural environments. However, reasonable parameterizations and deposition velocity models exist for many conditions of interest. These models are semi-empirical, with theoretical descriptions of airflow and the efficiency of various quasi-laminar boundary layer mechanisms tuned to match experimental observations. Therefore, model predictions are most accurate for the conditions from which they were developed and special attention should be paid when using these models in conditions that are dissimilar from those used in their formulation.

4.0 DEPOSITION VELOCITY MODEL COMPARISON

In this section, we discuss comparisons of experimental data and deposition velocities derived from the Sehmel and Hodgson (1978), GENII (Napier 2010) / RATCHET (Ramsdell et al. 1994), AERMOD (Cimorelli et al. 2005) / CALPUFF (Scire et al, 1990), and Petroff and Zhang (2010) models for a range of land-use categories. We focus on three environmental conditions found at DOE sites (bare soil, grasslands, and coniferous forest).

4.1 DEPOSITION VELOCITY MODELS

Table 4-1 provides a brief summary of the deposition models used in this comparison, each of which will be briefly discussed in the following sections.

Table 4-1. Summary of deposition velocity models of interest

Model	***Aerodynamic Resistance r_a (s/m)***	***Quasi-laminar sublayer resistance r_b (s/m)***	***Surface Transfer Resistance r_c (s/m)***	***Settling velocity v_s (m/s)***	***Deposition velocity, v_d (m/s)***
Sehmel and Hodgson (1978)	$A = f(Sc, D_p, u_*, z_o, D_b)$			$\frac{(\rho_p - \rho_g) g D_P^2 C}{18\mu}$	$\frac{v_s}{1 - e^{-v_s / u_* e^A}}$
GENII/ RATCHET	$u(z_d) / u_*^2$	$6.5 / u_*$	100	$\frac{(\rho_p - \rho_g) g D_P^2 C}{18\mu}$	$\left(\frac{1}{r_a + r_b + r_t + r_a r_b v_s}\right) + v_s$
AERMOD/ CALPUFF	$u(z_d) / u_*^2$	$\frac{1}{u_*\left(Sc^{-2/3} + 10^{-3/St}\right)}$	-	$\frac{(\rho_p - \rho_g) g D_P^2 C}{18\mu}$	$\left(\frac{1}{r_a + r_b + r_a r_b v_s}\right) + v_s$
Petroff and Zhang (2010)	*A = f (Sc, D_p, z_o, LAI, u_*, L, D_b, d, T, h, ObstSize)**			$\frac{\rho_p g D_P^2 C}{18\mu} + V_p$	$\left(\frac{1}{r_a + r_b}\right) + v_s$

u_*	= friction velocity (m/s)	D_p	= particle diameter (m)
v_s	= settling velocity (m/s)	Sc	= Schmidt number
z_o	= surface roughness length (m)	St	= Stokes number
d	= zero-displacement height (m)	LAI	= leaf area index (dimensionless)
h	= canopy height (m)	L	= Obukhov length scale (m)
μ	= dynamic viscosity of air	T	= ambient air temperature (K)
D_b	= Brownian diffusivity (m^2/s)	ObstSize	= characteristic obstacle size (m)

*The Petroff and Zhang model uses a complex formulation (see Section 4.1.3 and references) that depends upon the dominant land-use category, the Obukhov length, the surface friction velocity, the air temperature and the particle size distribution as inputs (see Section 6.2).

4.1.1 SEHMEL AND HODGSON MODEL

The Sehmel and Hodgson (1978) model is the basis for the default deposition velocity values used in MACCS2 (HSS Safety Bulletin, 2011). This model was developed from wind tunnel measurements of deposition onto quasi-flat surfaces (water, metal, artificial grass, and gravel) for a range of friction velocities (11 to 144 cm s^{-1}) and particle sizes (AED = 0.03 to 29 μm). The results of this model are generally consistent with a wide-range of historical deposition velocity measurements, but do not take into account the effects of atmospheric stability or cover a variety of land-use categories.

4.1.2 GENII/RATCHET AND AERMOD/CALPUFF DEPOSITION MODELS

Both GENII/ RATCHET and AERMOD/CALPUFF incorporate resistance-based deposition models. Both models use comparable calculations to determine both r_a (the aerodynamic resistance) and v_s (gravitational settling). However, there are significant differences in their treatment of the quasi-laminar sublayer resistance, r_b, that parameterizes the influence of impaction, Brownian diffusion, and interception deposition processes (see discussion in Section 3 and summary in Table 4-1). The GENII/ RATCHET model does not use the Schmidt and Stokes numbers, which are needed to produce the minimum in the deposition velocity curve for intermediate particle sizes (<1 μm), while AERMOD/CALPUFF includes both. GENII/ RATCHET treats the surface transfer resistance as a single constant value, while AERMOD/CALPUFF model does not include this term, effectively using a zero surface transfer resistance.

4.1.3 PETROFF AND ZHANG MODEL

The Petroff and Zhang model (Petroff and Zhang 2010; 2009; 2008b) is constructed under the premise that while no single theoretical description of deposition processes exists that is valid for all land use types, it should be possible to parameterize deposition properties over a wide range of natural environments based on available deposition velocity measurements. This model represents a major advance over earlier models (e.g., Zhang et al., 2001) in both theoretical and experimental validity. It arguably provides the most complete theoretical descriptions of deposition to date and has been parameterized to match the greatest number of experimental data sets covering multiple surface types (land-use characteristics). The model has been shown to be consistent with both historical deposition velocity observations (used in its development) as well as more recent observations (e.g. Fang et al., 2012; Mammarella et al., 2011).

The Petroff and Zhang (2010) model is a size-resolved dry deposition scheme for particles, developed for inclusion in large-scale air quality and climate models that take into account both the size distribution and fate of atmospheric aerosols. The model is based upon the resistance model approach originally proposed by Zhang et al. (2001), with the addition of a new "surface" deposition velocity (or surface resistance) term derived from a simplified version of a one-dimensional aerosol transport model (Petroff et al., 2008b; 2009). The latest version of the model accounts for leaf size, shape and area index, as well as the height of the vegetation canopy and is more sensitive to surface characteristics (land-use) for particles in the size range between 0.1–1 μm. A drift velocity is included to account for phoretic effects related to temperature and humidity gradients close to liquid and solid water surfaces.

4.2 DEPOSITION AS A FUNCTION OF PARTICLE SIZE

In the following section, the deposition velocity models listed above are compared for different meteorological conditions (wind speed, atmospheric stability class) and surface types (land-use category) as a function of particle size (AED). As a starting point, a simplified example comparison is shown in Figure 4-1 for the case of 1 m/s winds (measured at a reference height of 10 m), stability class F, and a surface roughness length of 1 cm – conditions often associated with the highest near-surface air concentrations for ground-level releases.

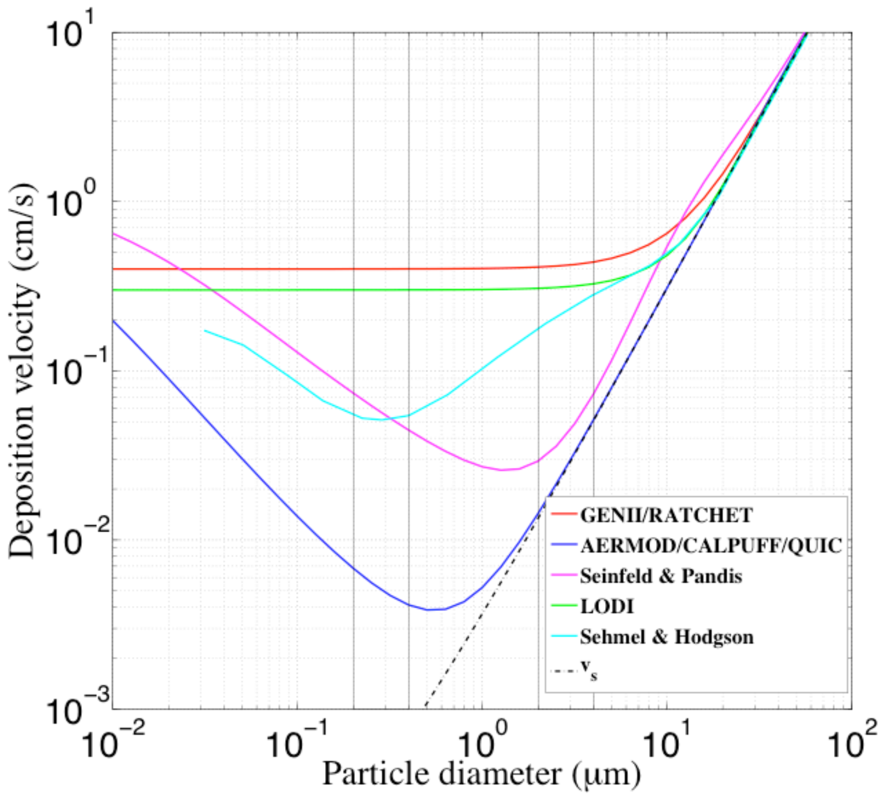

Figure 4-1. Deposition velocity (cm/s) as a function of particle diameter (μm) and choice of deposition (resistance) model formulations for several representative models and 1 m/s winds, stability class F, and a surface roughness length of 1 cm.

For the highly stable conditions shown in Figure 4-1, deposition velocities for particle diameters greater than 10 μm are in good agreement for all models. This is due to their similar treatments of gravitational settling, which is the dominant deposition mechanism for this size range. However, a significant divergence in deposition velocities occurs for particle sizes between 0.1 and 10 μm. Deposition velocities are in slightly better agreement for AEDs less than 0.1 μm, although the spread is still significant ranging from approximately 0.02 – 0.2 cm/s.

In the AED = 0.1 – 10 μm) particle size range, GENII/RATCHET (and the current version of the NARAC LODI dispersion mode[25]) use a constant deposition velocity of approximately 0.25 – 0.3 cm/s. The simpler treatment of the quasi-laminar sublayer resistance, $r_{b,}$ in the GENII/RATCHET model leads to an over-estimate of the deposition velocity as compared to models with more sophisticated formulations. In contrast, the AERMOD/CALPUFF model exhibits a minimum in the deposition velocity at an AED of ~0.3 μm that is two orders of magnitude lower (~0.003 cm/s) than the GENII/ RATCHET value. The Sehmel and Hodgson (1978) model also shows a minimum in the deposition velocity (v_d = ~0.05 cm/s at an AED of 0.2 μm). Although not included in the more detailed comparisons in the following sections, we note that the well-known Seinfeld and Pandis (2006) formulation[26] produces a minimum deposition velocity of 0.02 cm/s, higher than AERMOD/ CALPUFF but still considerably less than the values used in GENII/RATCHET.

4.3 DEPOSITION ON DIFFERENT SURFACE TYPES

In the following three sections model-predicted values for the deposition velocity are compared to the available experimental data for three representative land-use categories – bare soil, grasslands, and forests. We also discuss the reasons for the agreement and/or disagreement between the models of interest.

4.3.1 BARE SOIL

Figure 4-2 compares the deposition velocity onto bare soil as a function of particle size for the four different models listed in Table 4-1 and three different flow conditions (u_* = 11 cm/s, 34 cm/s, and 73 cm/s). For all three wind-speed cases, the deposition of coarser particles (AED > 10 μm) is primarily driven by the effect of gravity. As expected, all of the models reproduce the experimental dataset for this size range properly and align with the gravitational settling curve (dashed black curve labeled w_s).

For the particle size range from approximately 0.1 -1.0 μm, only the Petroff and Zhang model adequately agrees with the limited measurements. Both the Sehmel and Hodgson (1978) model and the GENII/RATCHET models over-predict v_d by up to two orders of magnitude. The AERMOD/CALPUFF predictions of v_d are a somewhat better fit, but still differ from the data by as much as an order of magnitude. In this particle size range, the deposition process of interception is greatly influenced by the surface type. GENII/RATCHET overestimates the v_d values as their treatment of deposition velocity processes in this size range are simplistic and do not incorporate the Schmidt number, the Stokes number, or the surface type. The AERMOD/CALPUFF parameterization is derived for short grass rather than bare soil and hence overestimates deposition.

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25 NARAC is currently implementing the Petroff and Zhang (2010) deposition model in LODI.

26 Seinfeld and Pandis (2006) is a classic textbook with a formulation that describes the basic features of deposition models. It is included here for pedagogical purposes.

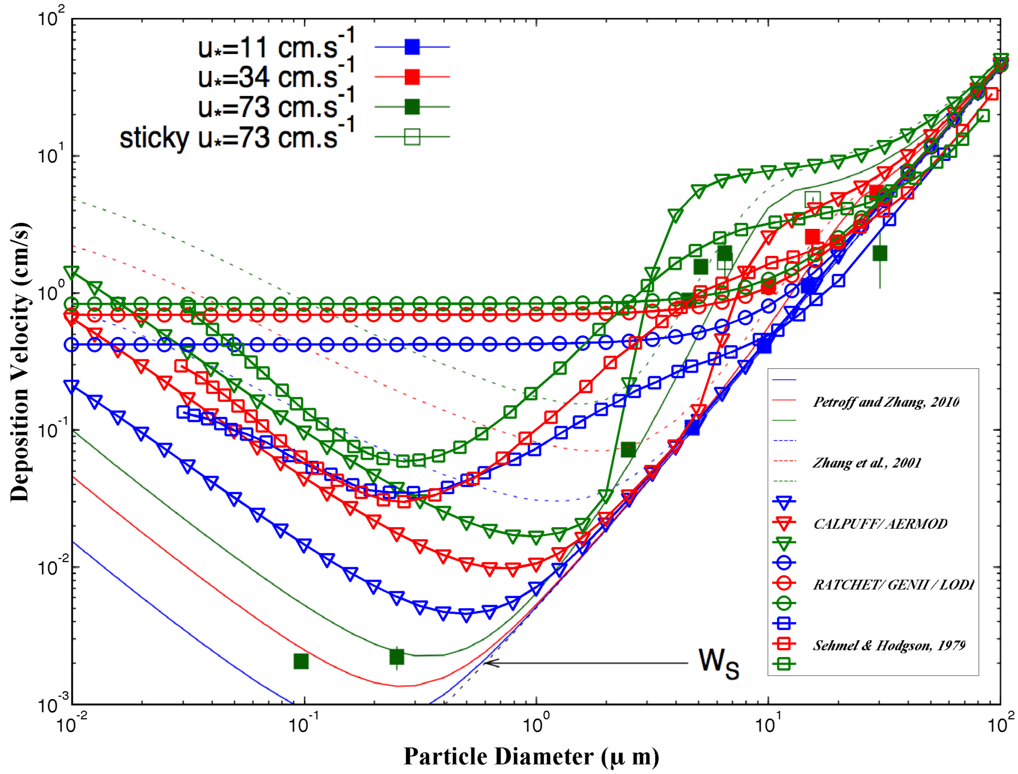

Figure 4-2. Comparison of predicted deposition velocities as a function of particle diameter from the Petroff and Zhang (2010), Zhang et al. (2001), CALPUFF / AERMOD, RATCHET/ GENII, and Sehmel and Hodgson (1978) models for friction velocities of 11 cm s^{-1} (blue), 34 cm s^{-1} (red), 73 cm s^{-1} (green), and a smooth soil surface. The reference height is taken to be 1m. Measured deposition velocities are indicated by solid squares (open squares for sticky surfaces). Figure adapted from Petroff and Zhang, 2010.

4.3.2 GRASSLAND

Data from experiments performed on short grass (Chamberlain, 1967; Clough, 1975; Garland, 1983) and moorland (Gallagher et al., 1988; Nemitz et al., 2002) are compared with the deposition models in Figure 4-3. For larger particles (AED >10 µm), all of the models perform well as the dominant process is gravitational setting.

For smaller particle sizes (AED < 0.5 µm), there is considerable scatter in the observed data. All models but one perform reasonably well and are able to predict measured deposition velocity values within the spread of the observed data. Only the GENII/ RATCHET model fails to match the data in this size range as it lacks any parameterization for the interception process, which is important in this regime, while all of the other models account for this effect through the Schmidt number. For the grassland case, the AERMOD/CALPUFF model performs the best, not surprisingly since this model was formulated using the dataset for short grass.

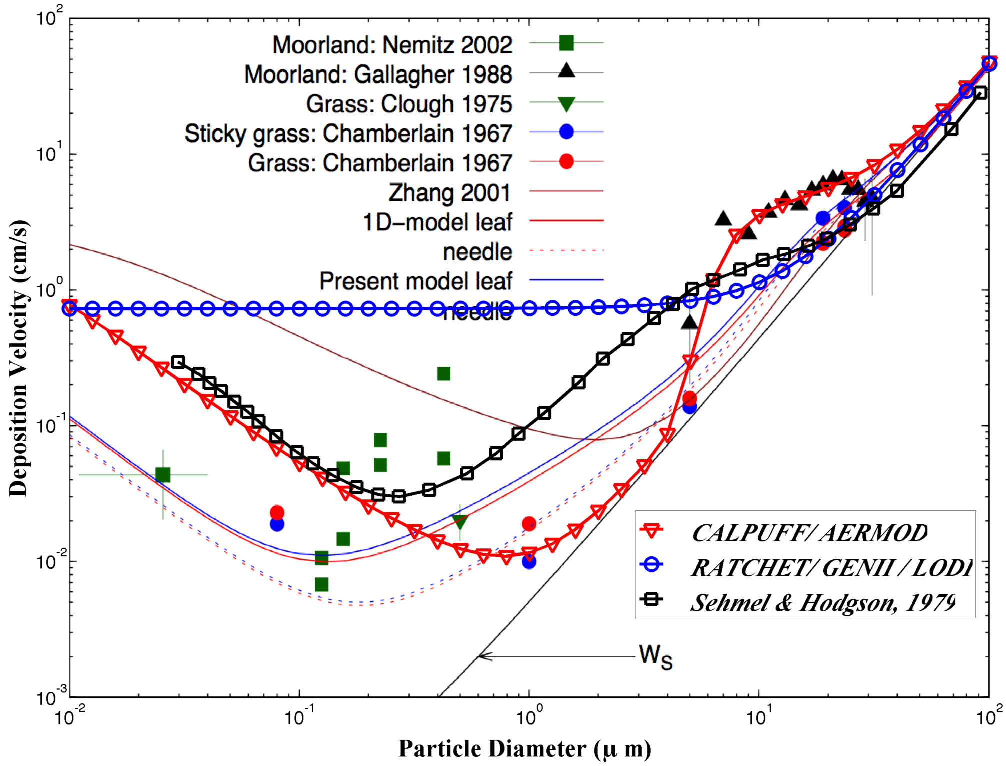

Figure 4-3. Comparison of predicted deposition velocities from the Petroff and Zhang (2010), CALPUFF / AERMOD, RATCHET/ GENII, and Sehmel and Hodgson (1978) models with experimental data as a function of particle diameter for friction velocities between 25 and 55 cm s^{-1}, a reference height of 3.8m, and short grass. The Petroff and Zhang model is labeled "Present model leaf". Measured deposition velocities are indicated as solid squares, triangles and circles. Figure adapted from Petroff & Zhang (2010).

4.3.3 CONIFEROUS FOREST

A final comparison is shown in Figure 4-4 for coniferous forests using data for spruce (Beswick et al., 1991), pine (Lorenz and Murphy, 1989; Lamaud et al., 1994; Buzorius et al., 2000; Gaman et al., 2004; Gronholm et al., 2009) and fir (Gallagher et al., 1997). For both larger (AED > 10 μm) and smaller particles (AED < 0.1 μm), all of the models perform reasonably well and are within the spread of observed data. The AERMOD/CALPUFF model performs poorly in the intermediate particle size range (0.1 < AED < 1 μm), as it was formulated from the short grass dataset and is unable to account for the increased deposition produced by heavy vegetation. The Sehmel and Hodgson (1978) models performs better but also under-predicts deposition in this size regime. GENII/ RATCHET values are consistent with the data over this particle diameter range as v_d do not drop as much as for the bare soil and short grass cases (Figures 4-2 and 4-3). However this agreement is purely serendipitous, as GENII/RATCHET does not include the required physics to model the dominant interception and Brownian diffusion deposition processes. For the coniferous forest case, the Petroff and Zhang (2010) model incorporates the relevant deposition processes and performs better than the other models against the data across all size ranges.

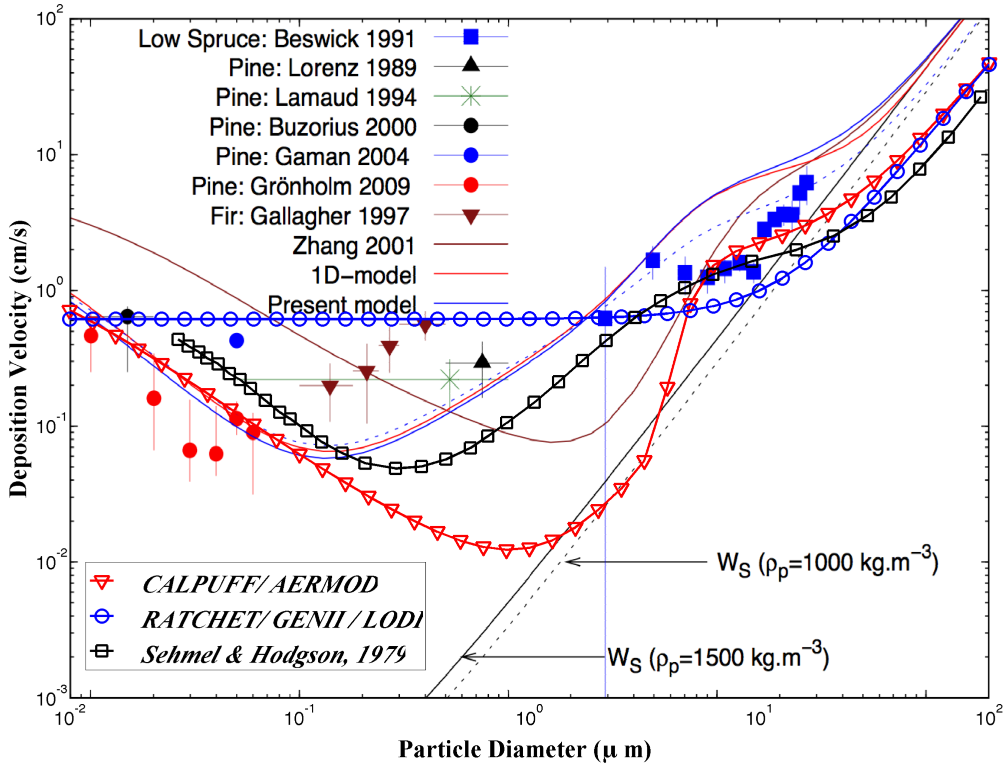

Figure 4-4. Comparison of predicted deposition velocities from the Sehmel and Hodgson (1978), RATCHET/ GENII, AERMOD/CALPUFF, Zhang (2001), Petroff and Zhang (2010), models with experimental data as a function of particle diameter for friction velocities of 47.5 cm s^{-1}, a particle density of 1500 kg m^{-3}, a reference height of 3.8m, and coniferous forests. The Petroff and Zhang model is labeled "Present model". Measured deposition velocities are indicated as solid squares, asterisk, circles and triangles. Figure adapted from Petroff & Zhang (2010).

4.4 SENSITIVITY ANALYSIS

We conducted a sensitivity analysis of deposition velocities predicted by GENII/RATCHET, AERMOD/CALPUFF, and Petroff and Zhang (2010) by varying the wind speed, friction velocity, and atmospheric stability. Figures 4-5 and 4-6 shows the results for two land-use categories: short grass and forest environments, respectively.

Figure 4-5a illustrates the sensitivity of the Petroff and Zhang (2010) deposition models to wind speed and atmospheric stability in grassland environments. It can be observed that the deposition velocity varies significantly (over three orders of magnitude) for the smallest particle sizes where deposition is dominated by the aerodynamic resistance, with v_d increasing with increasing friction velocity (u_*). For larger particles, the range of deposition velocities is reduced although still significant (covering more than an order of magnitude), as the deposition becomes more sensitive to particle diameter (Schmidt number, Stokes number) than to atmospheric conditions.

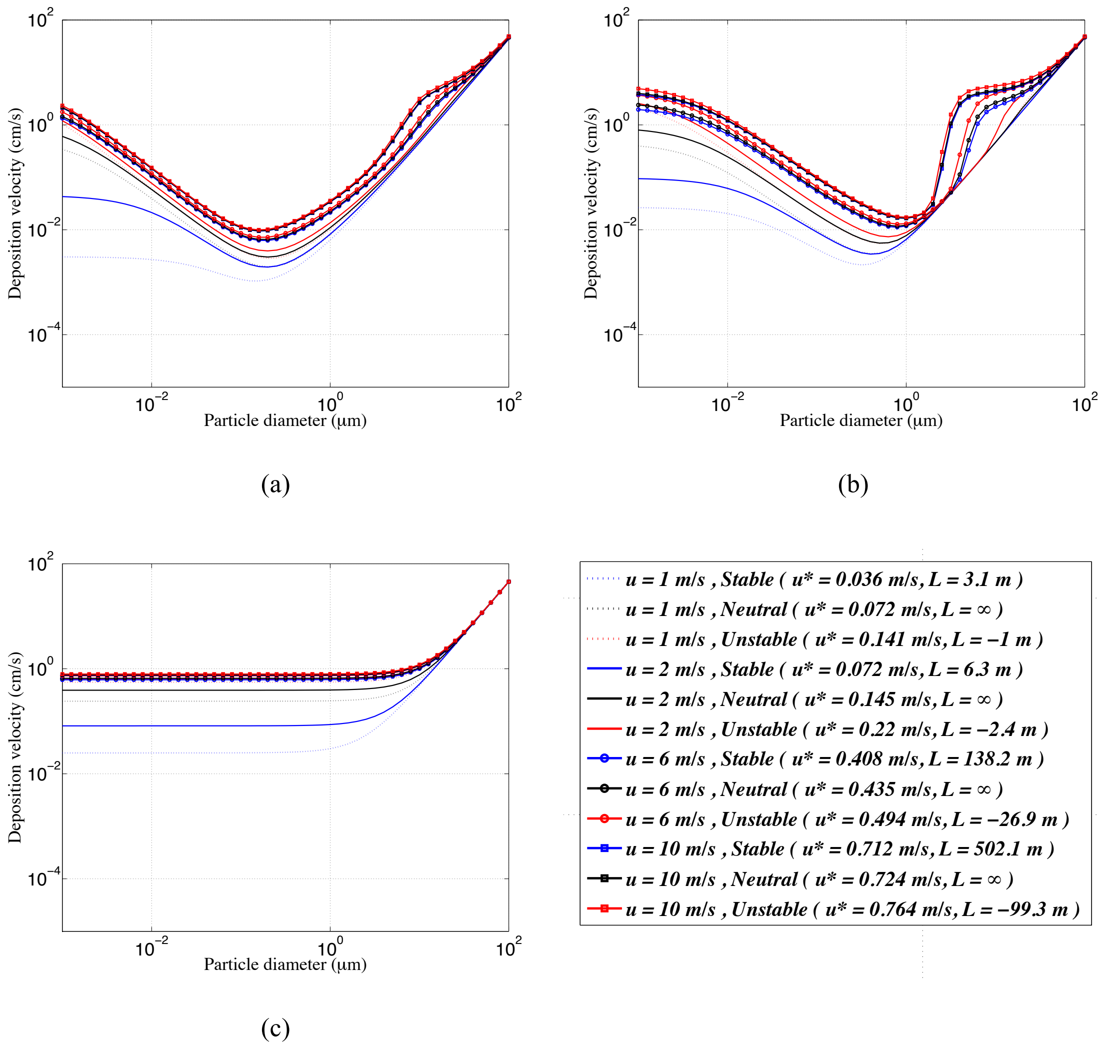

Figure 4-5. Sensitivity of the (a) Petroff and Zhang (2010), b) CALPUFF/ AERMOD and c) RATCHET/ GENII deposition models to different wind speed and atmospheric stabilities in grassland environments.

The variation in deposition velocity produced by the AERMOD/CALPUFF deposition model for the same range of wind speeds and atmospheric stabilities is shown in Figure 4-5b. As for the Petroff and Zhang case, the variation in deposition velocity is largest for smaller particles. However, the variation in deposition velocity remains significant (~2 orders of magnitude) for particle with AEDs between 1 and 10 μm, especially for the higher wind speed cases.

Figure 4-5c shows the curves for the same cases resulting from the GENII/RATCHET model. There is a significant range of deposition velocities for the different atmospheric conditions for particles with AED < 5 μm, with v_d predicted to be constant over that entire size range. As previously discussed, this results from the lack of a complete formulation of the quasi-laminar sub layer resistance (Section 3.2.3).

Figures 4-6 shows the sensitivity of deposition velocity to wind speed and atmospheric stability as a function of particle diameter for forested environments. The results are similar to those for grasslands, with the variation in deposition velocity ranging between 1 - 2 orders of magnitude. For this land-use category, the predicted v_d values are higher as the increased vegetation leads to higher values of the friction velocity.

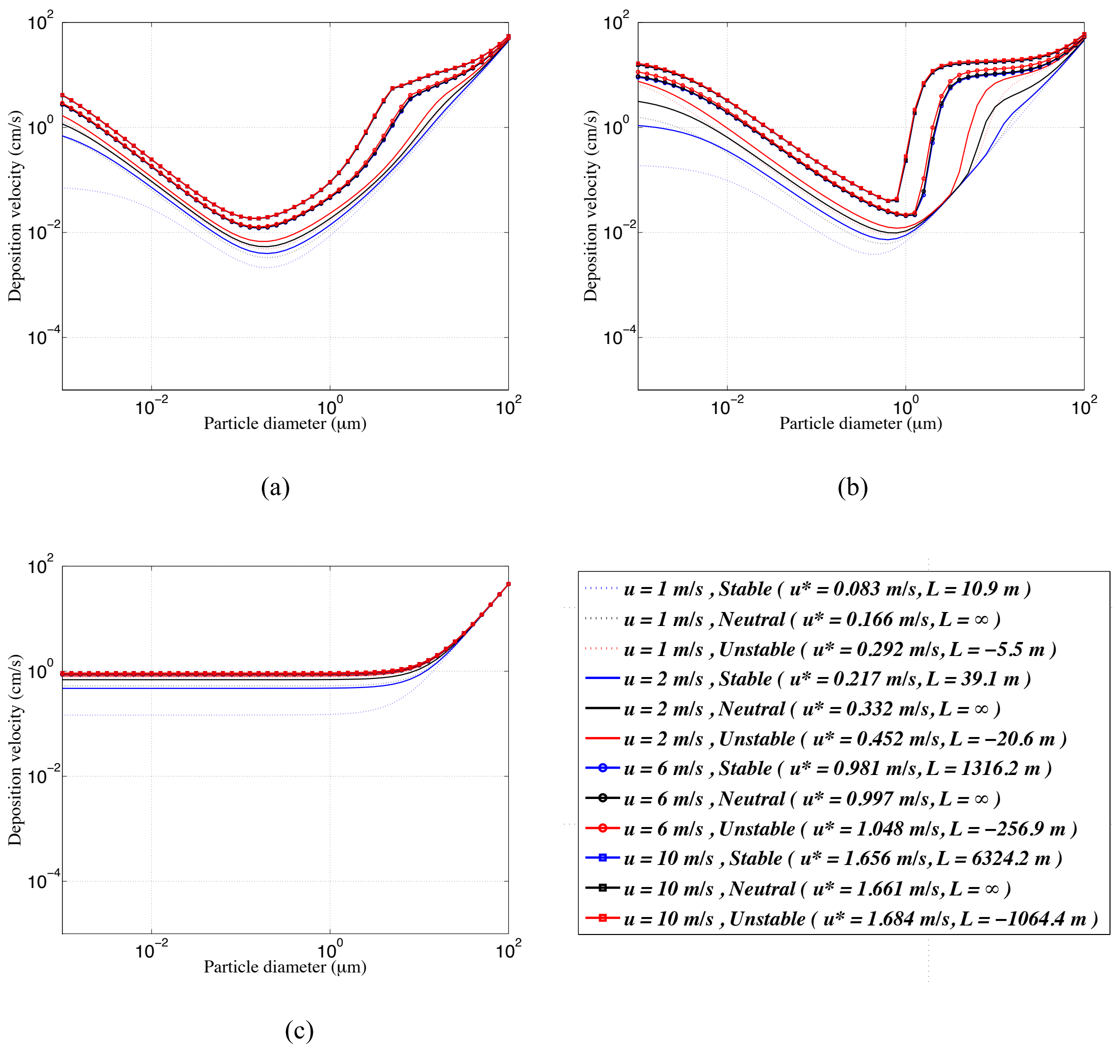

Figure 4-6. Sensitivity of the (a) Petroff and Zhang (2010), b) CALPUFF/ AERMOD and C) RATCHET/ GENII deposition models to different wind speeds and atmospheric stability for forest land-use.

In the Sehmel and Hodgson model (1978), the deposition velocities and relative mass transfer resistances are a function of friction velocity. Figure 4-7 depicts the deposition velocities for a range of friction velocities in a grassland environment. The model produces a minimum in the deposition velocity for particle diameters around 0.2 to 0.3 μm and exhibits a variation of approximately 2 orders of magnitude over the range of friction velocities. Outside of the size range 0.1 μm < AED < 1.0 μm, deposition velocities increase with increasing friction velocity.

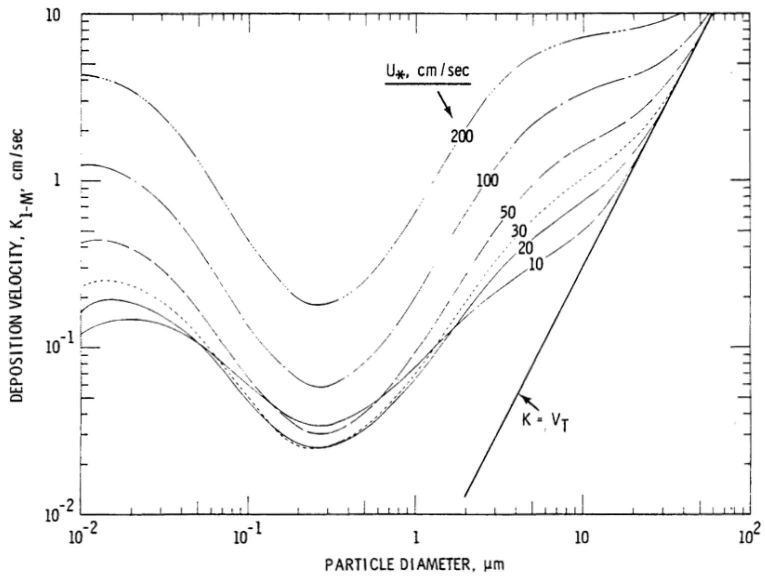

Figure 4-7. Sensitivity of Sehmel and Hodgson (1978) deposition model to different friction velocity for a constant aerodynamic surface roughness (z_o = 3 cm, grassland environments).

4.5 SUMMARY OF DEPOSITION MODEL COMPARISONS

Most of the deposition velocity models in common use were developed for grassland environments and perform best under those conditions. This is particularly true of the AERMOD/CALPUFF model. After evaluating the performance of the various deposition models under different conditions, we concluded that the Petroff and Zhang (2010) model performs the best for the widest range of conditions, with significant performance improvements for particles with AED < 10 μm. This result is not surprising, since the model formulations were developed using experimental data from a variety of surface types.

Key findings from the model comparison are summarized below:

- The Petroff and Zhang (2010) deposition velocity model matches the measurement data reasonably well and is applicable to the widest range of land-use categories.
- The GENII/RATCHET model was found to overestimate v_d for small to intermediate particles (AED = 0.1-1 μm) in bare ground and grassland environments.

- Deposition velocity values in the respirable-size range are highly sensitive to atmospheric conditions and land-use.
- In contrast, deposition velocities for large particles (AED >50 microns) are insensitive to varying atmospheric condition or land- use.

The HSS Safety Bulletin (2011) recommended values for deposition velocity are reasonably consistent with the experimental data:

- The recommended defaults of 0.01 cm/s and 0.1 cm/s for mitigated/filtered and unmitigated/unfiltered particles are in reasonable agreement with the available measurement data for low friction velocities (low wind speeds, stable atmospheres, and low surface roughness values).
- For smaller particles corresponding to filtered/mitigated releases (AED = 0.2 – 0.4 μm), the HSS-recommended default v_d value of 0.01 cm/s is a factor of two greater than both the measurements and the Petroff and Zhang (2010) model values. However, for such small particles, the deposition values are already so low that they do not have a significant impact on predicted air concentrations (e.g., such low deposition velocities produce virtually the same results as a v_d of zero).
- For larger particles sizes corresponding to unfiltered/unmitigated releases (AED = 2 – 4 μm), the HSS (2011) recommended default may be either lower or higher than the values derived from the Petroff and Zhang (2010) and associated experimental data depending on the environmental / land-use conditions.
    - *Bare ground.* The HSS recommended v_d value of 0.1 cm/s is above the range of deposition velocities derived from both measurement data and the Petroff and Zhang (2010) model (0.02-0.07 cm/s) and would therefore be expected to provide less conservative estimates of air concentrations. Therefore, the HSS default value is not recommended for unmitigated/unfiltered particle releases over bare ground (this case represents the lowest expected deposition velocity across all land-use types apart from water).
    - *Grassland.* The HSS recommended v_d value of 0.1 cm/s values falls near the mid-point of the range of deposition velocity values derived from measurement data and the Petroff and Zhang (2010) model, but does not constitute a lower bound for this size range.
    - *Forests.* The HSS recommended default for v_d is smaller than the measured and Petroff and Zhang predicted values, and would therefore be expected to provide conservative air concentration estimates.

The results of the sensitivity study illustrated the significant variation of deposition velocity with wind speed and atmospheric stability for all particle sizes and land-use (surface characteristic) conditions. This variability reinforces the potential importance of using deposition velocities corresponding to the actual hourly meteorological conditions when performing 95th percentile calculations.

5.0 MACCS2 CONSIDERATIONS

5.1 GAUSSIAN MODELS

Gaussian plume models are often used in atmospheric dispersion modeling applications, because they are both simple to use and computationally efficient. Two of the DOE Central Registry codes commonly used for safety analyses (MACCS2 and HotSpot) are Gaussian plume models. These models have the advantage of producing results that are relatively easy to interpret and straightforward to relate to changes in input parameters. However, such models have significant limitations in cases involving low wind speeds, complex terrain, spatially or temporally-varying meteorology, complex deposition or transformation processes, or land-use conditions for which the diffusion coefficients have not been well-validated. We therefore performed a limited investigation into some aspects of these codes related to deposition modeling, including the implementation of depletion factors, the treatment of elevated releases, and considerations for low wind speed conditions.

5.1.1 GAUSSIAN MODEL FORMULATION

When not constrained by the ground or by inversion layers, the Gaussian plume equation has the following form:

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

with the spread of the plume in the vertical and crosswind directions governed by the standard deviations σ_z and σ_y .Table 5-1 provides the definitions of the variables in this equation.

Table 5-1. Definitions of variables in Gaussian plume equation.

$\chi(x,y,z)$	Time-integrated air concentration (*Bq-s/m*^{3}) at the downwind location (*x,y,z*)
Q	Source strength (*Bq*)
$\bar{u}$	Mean wind speed (*m/s*)
σ_y *and* σ_z	Standard deviations (*m*) of the normal crosswind and vertical concentration distributions of plume materials
$(x=0, y=0, z=h)$	Source location
h	Release height (*m*)

Once the plume has expanded to the point so that further vertical expansion is constrained by the ground and/or a capping inversion layer above, the Gaussian solution is modified by treating both as impenetrable reflecting boundaries. Mathematically, this reflection is accomplished by the introduction of mirror image sources below the plane of the ground and/or above the inversion layer, as shown below:

$$\chi(x,y,z)=\frac{Q}{2\pi\sigma_y\sigma_z\bar{u}}\exp\left[-\frac{1}{2}\left(\frac{y}{\sigma_y}\right)^2\right]\left\{\exp\left[-\frac{1}{2}\left(\frac{z-H}{\sigma_z}\right)^2\right]+\exp\left[-\frac{1}{2}\left(\frac{z+H}{\sigma_z}\right)^2\right]+\sum_{n=1}^{5}\left\{\begin{array}{l}\exp\left[-\frac{1}{2}\left(\frac{z-H-2nL}{\sigma_z}\right)^2\right]+\exp\left[-\frac{1}{2}\left(\frac{z+H-2nL}{\sigma_z}\right)^2\right]\\+\exp\left[-\frac{1}{2}\left(\frac{z-H+2nL}{\sigma_z}\right)^2\right]+\exp\left[-\frac{1}{2}\left(\frac{z+H+2nL}{\sigma_z}\right)^2\right]\end{array}\right\}\right\}$$

5.1.2 CONSERVATISM OF GAUSSIAN MODELS

Gaussian plume models have been shown to be generally conservative in comparisons against measured air concentrations, predicting concentrations that are a factor of 2 or more larger than observed values (see for example Miller and Little, 1980). Although available data sets are insufficient to determine reliable statistics for Gaussian plume model uncertainty (Miller and Hively, 1987), the accuracy of such models has been shown to diminish as averaging times decrease and/or the meteorological and terrain conditions become more complex (Miller and Hively, 1987).

In addition to the choice of deposition velocity, other plume modeling input parameters, assumptions and methods (e.g., the choice of dispersion coefficients) may have a significant effect on Gaussian plume model calculation of air concentrations. For example, the method of determining stability class can significantly affect plume model results (Miller and Hively, 1987). Miller and Little (1980) showed that Delta-T methods produced more conservative Gaussian plume model results (i.e., predicted air concentrations higher than observed) than a sigma-theta method, even though the sigma-theta method results were still conservative (predicting concentrations more than twice the observed values, on average).

5.2 DEPLETION MODELS OF DEPOSITION

Dry deposition effectively results in the removal of pollutant mass from the plume. In simpler dispersion models, including most models used for safety analyses, deposition is accounted for by appropriate reductions in the source strength (Chamberlain, 1953; Van der Hoven, 1968). However it should be noted that this depletes the plume throughout its vertical extent. The following sections outline the source depletion process used in the two Gaussian models of interest.

5.2.1 MACCS2 SOURCE DEPLETION FORMULATION

In the MACCS2 model, the rate of loss of plume materials (*dQ/dx*) by dry deposition into a differential length *dx* located at the downwind distance *x* is determined according to (Slade, 1968):

$$\frac{dQ}{dx} = \int_{-\infty}^{+\infty} \omega(x,y)\,dy$$

$$= -F\sqrt{\frac{2}{\pi}}\frac{v_d Q}{\sigma_z \bar{u}} \int_{-\infty}^{+\infty} \frac{1}{\sqrt{2\pi}\sigma_y}\left[\exp\left\{-\frac{y^2}{2\sigma_y^2}\right\}\right]dy$$

$$= -F\sqrt{\frac{2}{\pi}}\frac{v_d Q}{\sigma_z \bar{u}}$$

where, $\omega(x,y) = v_d \chi(x,y,z=0)$ and F is the sum of all of the exponential terms that contain σ_z (other symbols are defined in Table 5-1).

Assuming that during any single hour the mean wind speed, $\bar{u}$, is constant so that $dx = \bar{u}dt$, this can be substituted into the equation to give

$$\frac{dQ}{dt} = -F\sqrt{\frac{2}{\pi}}\frac{v_d Q}{\sigma_z}$$

Rearranging the terms and integrating gives,

$$\frac{Q}{Q_0} = f_d = \exp\left\{-\frac{v_d \Delta t}{F'}\right\}$$

where $F' = \sqrt{\frac{\pi}{2}}\sigma_z \frac{1}{F}$, $\Delta t = \frac{x}{\bar{u}}$ and Q_o is the undepleted source strength.

In the above step, σ_z and F are assumed not to be function of *x*, so that a simple analytical expression can be obtained for depletion factor and numerical integration avoided.

5.2.2. HOTSPOT SOURCE DEPLETION FORMULATION

Plume depletion in HotSpot is accomplished by multiplying the original source term by a source-depletion factor *DF(x)* at a distance "*x*" from the source. The source depletion factor is calculated according to Van der Hoven (1968) as:

$$DF(x) = \left[\exp\int_0^x \frac{1}{\sigma_z(x)\exp\left[\frac{1}{2}\left(\frac{H}{\sigma_z(x)}\right)^2\right]}dx\right]^{-\frac{v_d}{\bar{u}}\sqrt{\frac{2}{\pi}}}$$

where, v_d is the deposition velocity, u is the mean wind speed, H is the release height and σ_z is standard deviations (m) of the vertical concentration distributions of the plume.

5.2.3. DEPLETION FACTOR CALCULATIONS

Depletion factors (*DFs*) were calculated from MACCS2 and HotSpot. The DFs were obtained via analytical expressions, numerical integration, and direct outputs from model simulations in which both models were run with and without deposition and DFs were calculated as $Q_{vd=1\ cm/s}/Q_{vd=0}$. The results in Figure 5-1 show that:

- For HotSpot, the agreement between the depletion factors generated by direct calculation and from independent numerical calculations confirms that the depletion model has been implemented correctly. However, the MACCS2 results for direct and numerical integration are offset for the Briggs case and there is a smaller discrepancy for the TG case (an analytical expression is used to define the TG dispersion coefficient while a the Briggs dispersion coefficients are specified using a look up table within MACCS2).
- The depletion factor predicted by MACCS2 using the TG dispersion coefficients produces more conservative values (DFs closer to one) than that computed using the Briggs open country coefficients.
- The analytical expression provided in the MACCS2 manual (replicated in Section 5.2.1) does not compare well with the DFs calculated using direct model output. Additional investigation is needed to resolve this discrepancy.

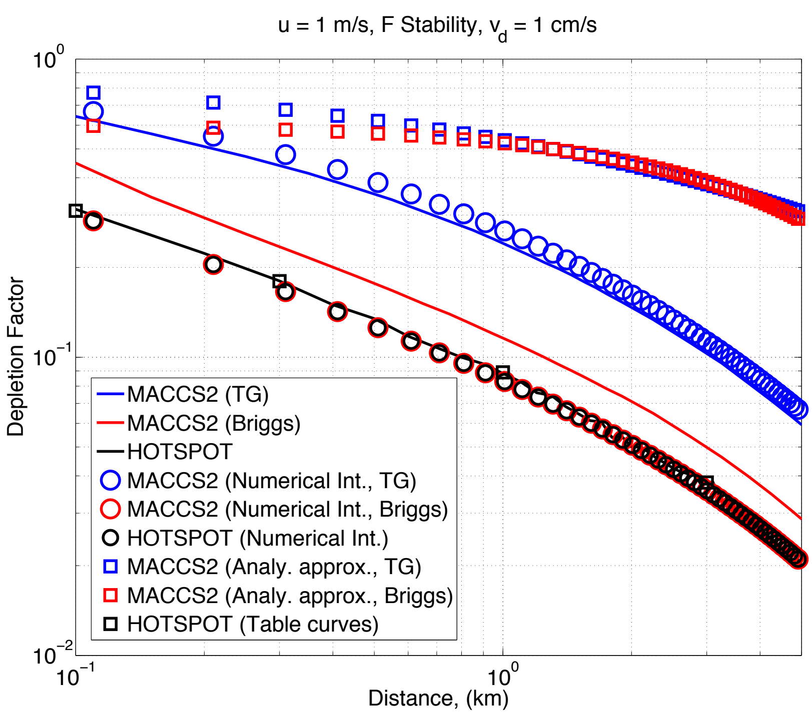

Figure 5-1. Depletion factors for Hotspot and MACCS 2 (u= 1 m/s, v_d= 1 cm/s. F stability)

5.3 COMPARISON OF MACCS2 AND HOTSPOT AIR CONCENTRATIONS

Ground level air concentrations predicted by MACCS2[27] and HotSpot were compared for multiple scenarios involving a range of atmospheric stability classes (A-F) and deposition velocities (0-1 cm/s). All of the scenarios assumed ground-level releases and a wind speed of 1 m/s. We included both of the MACCS2 dispersion coefficient options in this comparison: the Tadmor and Gur (TG) and the Briggs open country.

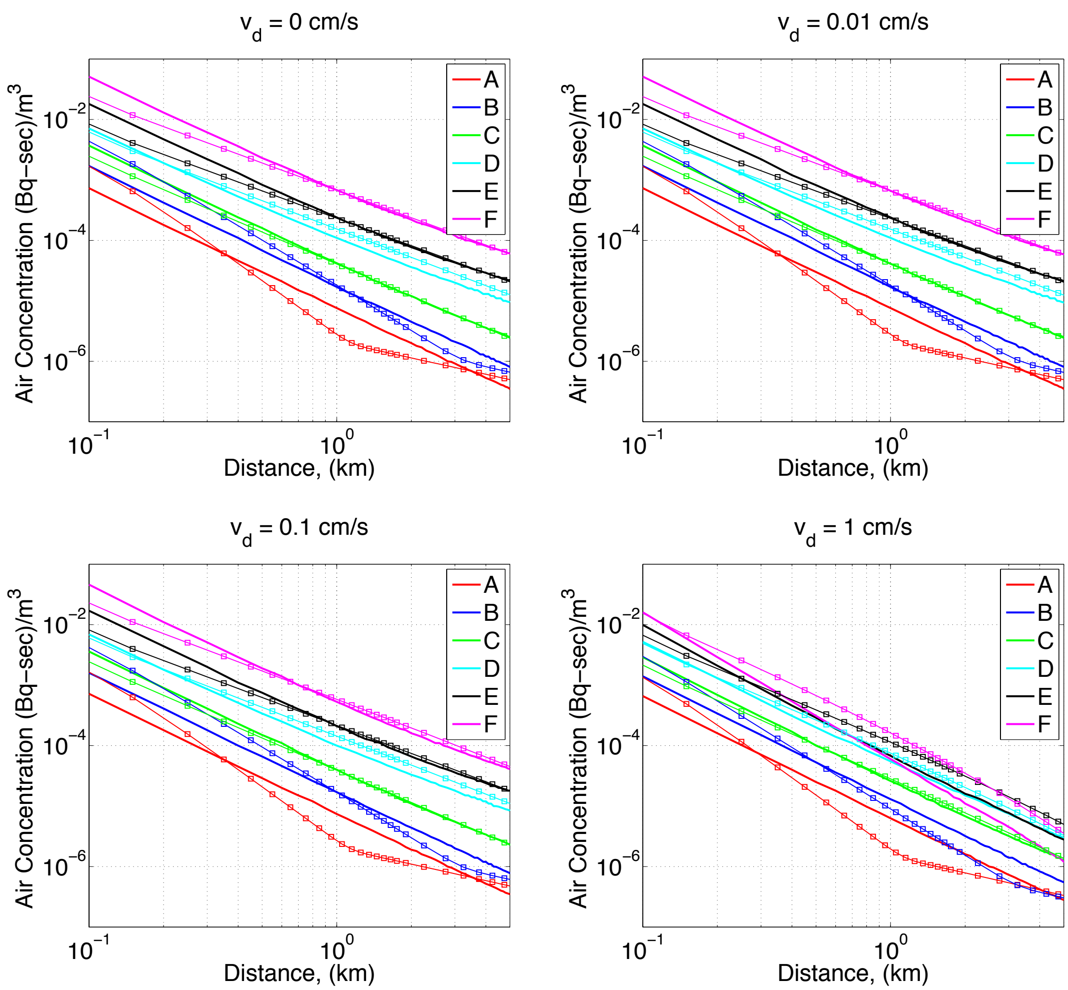

Figure 5-2. Comparisons of air concentrations for HotSpot (—) and MACCS2 using the Tadmor and Gur dispersion coefficients (—□—) for a range of atmospheric stability conditions and deposition velocities

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[27] The MACC2 model includes a set of scaling factors that can be used to adjust for various source and meteorological conditions, but these factors were not used in the simulations cited in this report.

The HotSpot and MACCS2 (TG) results show a number of differences, although no distinct trend is found (Figure 5-2). For F stability and lower values of v_d, HotSpot is more conservative than MACCS2 (TG), while for v_d = 1 cm/s the reverse is true. In contrast for A and B stability, MACCS2 (TG) is more conservative at short distances but air concentrations dip around 1 km (this dip appears to be an artifact of either the TG model or its implementation in MACCS2, but we did not have time to investigate this anomaly further). For intermediate stability classes, MACCS2 (TG) is generally less conservative that HotSpot at short distances and slightly more conservative for distances > 1 km.

Figure 5-3 shows a similar comparison between HotSpot and MACCS2 simulations using the Briggs open country dispersion coefficient. The HotSpot and MACCS2 (Briggs) results match more closely than for the MACCS2 (TG) case. For v_d = 1, HotSpot is slightly less conservative, especially for the more stable cases. One behavior of particular note is that as the deposition velocity increases, F stability is no longer the worst-case scenario at larger distances (>1 km) in the HotSpot calculations.

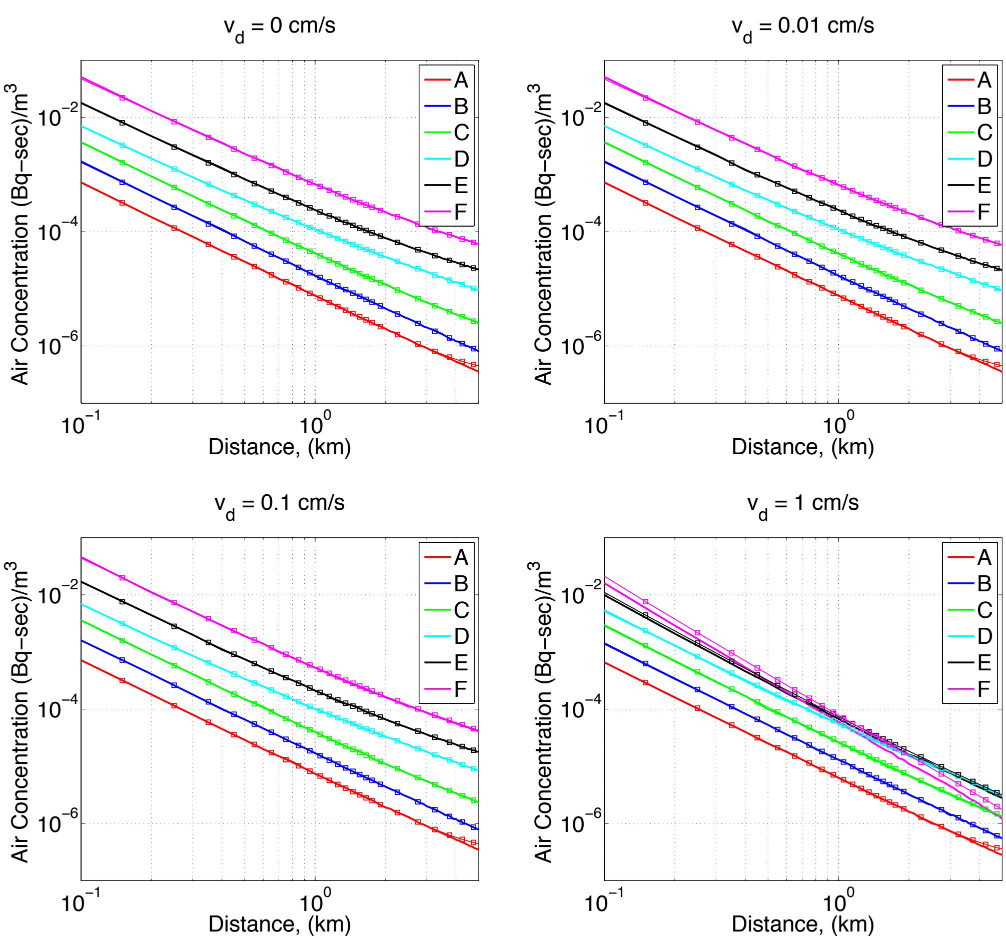

Figure 5-3. Comparisons of air concentration for HotSpot (—) and MACCS2 run using the Briggs open country dispersion coefficients (—□—) for a range of atmospheric stability conditions and deposition velocities

The Tadmor and Gur coefficients were derived from curve fits using data from the Prairie Grass tracer experiment that studied near-ground releases over flat terrain for downwind distances between 50 and 800 m. These fits are considered to be appropriate for use only over the range 0.5 km – 5 km and may not be valid for other conditions and distances (Tadmor and Gur, 1969). Specifically, it has been shown that the TG coefficients in MACCS2 V1.31.1 are not valid for distances < 0.5 km (Napier et al., 2011). Briggs (1973) combined the Pasquill, BNL (Smith and Singer, 1966), and TVA (Carpenter et al., 1971) curves which included observations out to 10 km in developing his coefficients. Based on these considerations as well as the results of the model comparisons above, we recommend that MACCS2 be run using the Briggs open country coefficients rather than the (default) TG option.

5.4 RELEASE HEIGHT

Since the DOE site survey responses from several facilities indicated elevated releases were a concern, we investigated the modeling of such scenarios using MACCS2. Figure 5-4 shows the air concentration predicted by MACCS2 for different stability classes and release heights. It should be noted that no single atmospheric stability class represents the worst-case scenario for all release heights and downwind distances (e.g., F stability is not always the worst-case scenario).

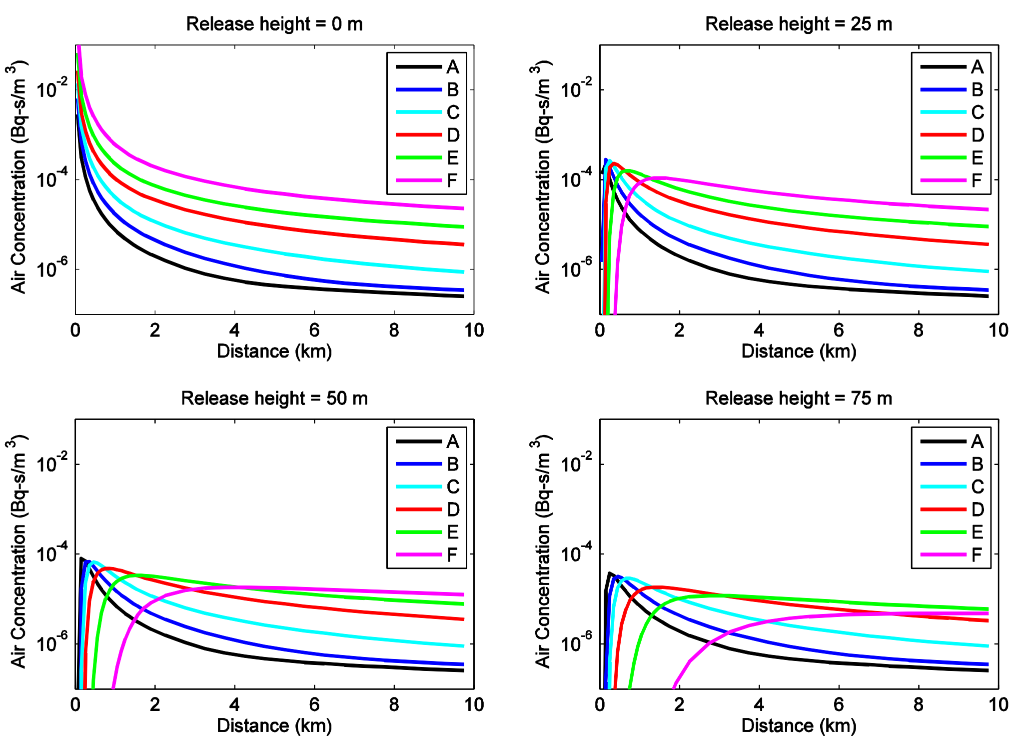

Figure 5-4. Comparison of air concentrations predicted by MACCS 2 for different stability conditions and different release heights assuming u= 1 m/s and v_d= 0.05 cm/s.

In analyzing these results, we found that MACCS2 V1.13.1 does not automatically account for the change in wind speed with release height. As Gaussian plume models are very sensitive to wind speed (higher speeds lead to lower concentrations, while lower speeds result in higher concentrations), the wind speed must be manually adjusted to the effective release height either directly or through the use of a scaling factor. Figure 5-5 illustrates this point, by comparing the air concentration predicted by MACCS2 for a release at 75 m using both corrected and uncorrected wind speeds. Near-surface air concentrations are over-predicted by ~40 % at downwind distances of 5 to10 km if the release height correction is not included.

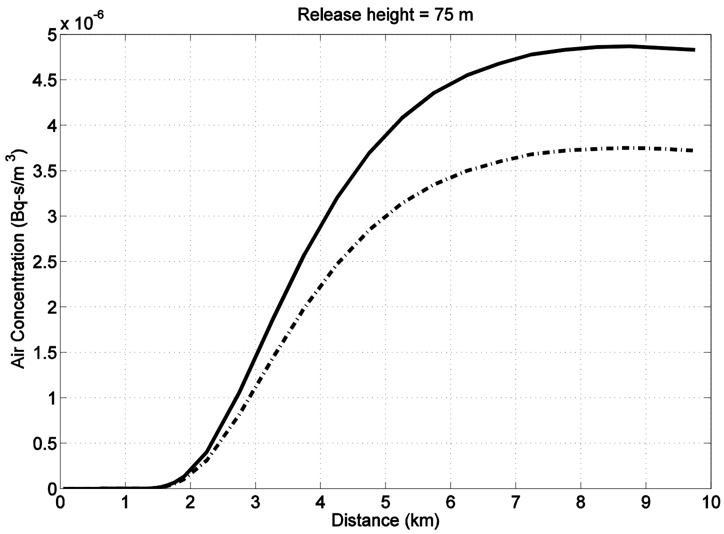

Figure 5-5. Comparison of air concentrations predicted by MACCS2 using release-height corrected (----) and uncorrected (—) velocity assuming u= 1 m/s, v_d= 0.05 cm/s, F stability

It should be noted that HotSpot (the other safety toolbox code included in this investigation) provides a power-law formula to adjust the wind speed for all effective heights greater than 2 meters (if the release height is less than 2 meters, HotSpot adjusts the wind speed to a reference height of 2 meters) according to:

$$u(H) = u(z)\left(\frac{H}{z}\right)^p$$

where

u(z) = wind speed (m/s) at reference height z (m)
H = effective release height (m)
P = power low exponent

5.5 LOW WIND SPEEDS

In light wind conditions, Gaussian plume models predict unrealistically large air concentrations for two reasons: a) the expression for the concentration contains the wind speed in the denominator which approaches zero in calm wind conditions and b) the formulation assumes a steady-state wind direction that does not account for the horizontal spread of the plume typical of low speed cases. In fact, the basic assumptions built into Gaussian models have been shown to be invalid for wind speeds less than ~2 m/s (Sawyer, 2007), when horizontal and vertical diffusion becomes more important than advection (Sharan et al., 1995). It should be noted that running a standard Gaussian dispersion model in light winds generally will produce highly conservative air concentration predictions, although their use is flawed from both physical and theoretical points of view.

In light wind conditions, highly variable winds produce much wider plumes and lower centerline concentrations than predicted by Gaussian plume models that assume a single dominant direction. The variability of wind directions can be quite large. Venkatram et al. (2004) found that wind directions frequently vary over 100° during periods of a few minutes when measured speeds are less than 2 m/s, while Hanna et al. (2003) demonstrated that upwind dispersion is possible in low wind speed conditions due to larger sigma theta (σ_θ) values.

Many applications using Gaussian plume models incorporate a minimum wind speed threshold of 0.5 - 1.0 m/s. For example, the U.S. Environmental Protection Agency recommends the use of a minimum wind speed threshold of 0.5 m/s for site-specific meteorological data (EPA, 2000). Wind cases below the specified minimum are either neglected or replaced with the minimum speed.

Some Gaussian plume models incorporate special options to handle the directional variability issue and avoid unrealistically high concentrations in light wind conditions. The HotSpot model (Homann and Aluzzi, 2013) includes a special G stability class to treat extremely stable low wind speed cases. The G stability algorithm uses a larger sigma theta value to account for wind direction fluctuations, producing enhanced horizontal spread and plume dilution. A beta release of the EPA's regulatory AERMOD Gaussian dispersion model (Cimorelli et al., 2003) uses a similar approach. Two 'LOWWIND' options have been implemented in this model: the first increases the minimum value of sigma-v (the standard deviation of the cross-wind velocity component) but turns off the horizontal meander component, while the second option includes a smaller increase in sigma-v but includes a meander component.

Determining an appropriate strategy for handling light wind conditions may be critical to 95th percentile air concentration safety analyses, especially for sites that frequently experience light wind speed conditions. Ignoring or improperly treating such cases may distort statistical air concentration metrics, especially as the 95th percentile air concentration is likely to be associated with such conditions. As an example, the DOE Y12 complex is located within a narrow valley running from the southwest to the northeast that produces terrain-influenced light winds. Statistics collected from 2011 from two site weather stations (ORR-TOWY and ORR-WEST) show that 5.8% - 7.9% of the wind speed observations were below the threshold value of 0.25 m/s and even higher percentages were below 0.5 m/s and 1.0 m/s (Table 5-2). In such cases, the strategy used to handle light winds in may significantly affect the outcome of site hazard analyses. Applying a minimum threshold approach may lead to higher predicted air concentrations than would actually occur or even the incorrect identification of the 95th percentile meteorological case.

Table 5-2. Percent of observed 10 m wind speeds at two Y-12 meteorological stations below thresholds of 0.25, 0.5, and 1.0 m/s.			
Meteorological Tower	**% < 0.25 m/s**	**% < 0.50 m/s**	**% < 1.00 m/s**
ORR-TOWY	5.8	13.5	31.3
ORR-WEST	7.9	15.7	33.4

For applications where low wind speeds are prevalent, the use of a model incorporating a special low wind speed option (e.g., HotSpot; AERMOD following release of the beta version) is recommended. A possible alternate is to use a more advanced non-steady state dispersion model such as CALPUFF (Scire et al., 1990) that explicitly calculates three-dimensional wind fields and is able to simulate light wind speed atmospheric phenomena such as recirculation. However, non-steady state model are not standardly approved for safety analyses, hazard assessments, or regulatory applications and they can be considerably more costly and difficult to use (see Appendix B).

6.0 95TH PERCENTILE AND DEPOSITION VELOCITY METHODOLOGY

Based on our literature review of existing models and the results of comparison studies discussed in the previous sections, we recommend the use of the Petroff and Zhang (2010) model for determining deposition velocities. This model parameterizes the impact of a wide range of vegetation types and the level of modeling sophistication is reasonable for implementation at DOE sites.

In addition, we also propose that DOE hazards analyses be performed using the 95th percentile air concentration approach developed and applied in this investigation. This method is based on a scientifically justifiable approach that utilizes hourly meteorology data coupled to consistent values of other parameters (e.g., wind-sector dependent land-use conditions, site-boundary distances, and deposition velocity) rather than calculating the 95th percentile meteorological conditions and selecting "reasonably conservative" parameters for other inputs.

This section describes our 95th percentile methodology followed by an outline of the calculations performed by the Petroff and Zhang (2010) deposition velocity model. We then discuss the input parameters needed by this model in order to calculate (hourly) site-specific v_d values for the 95th percentile meteorology and provide some guidelines for determining these inputs.

6.1 95TH PERCENTILE METHODOLOGY

One of our fundamental concerns during this investigation was the lack of clear criteria for determining "reasonable" or "conservative" values for various meteorological and dispersion input parameters (e.g., the deposition velocity) for use in 95th percentile air concentration modeling. Different selection criteria could lead to either overly conservative inputs predicting higher predicted risk levels than might actually occur or alternatively non-conservative results. We considered three approaches for conducting 95th percentile air analyses.

In the first option, a dispersion model may be run for a single choice of weather conditions, deposition velocity, and site boundary distance that represents the 95th percentile meteorological case. For example, winds of 1 m/s and atmospheric stability class of F are frequently assumed to be associated with the 95th percentile dose (NRC, 1998). The shortest distance to the site boundary is selected and deposition velocities for mitigated and unmitigated releases are specified from established "reasonably conservative" default values (e.g., as specified in the HSS Safety Bulletin) or determined using a site-specific analysis (with $v_d = 0$ representing the most conservative case). By themselves, each of these inputs provides a reasonable choice for safety analysis modeling. However, the use of independently selected parameters in the dispersion run may not correspond to a physically realistic case. Further, the combination of "conservative" input parameters may produce air concentration or exposure predictions that are far more "conservative" than intended. As an example, consider a case in which the conservative meteorological conditions used are actually associated with a wind sector that has a much larger distance to the site boundary than the shortest distance to the fence line.

A second approach for calculating 95th percentile air concentrations is based on the determination of the 95th percentile meteorological conditions (wind speed, atmosphere stability) while making "conservative" choices regarding other site and release conditions. The 95th percentile meteorology is determined by running hourly cases using meteorological observations for an extended period of time (a minimum of one year). While providing a more robust treatment of the meteorological conditions, this approach ultimately does not resolve the issue of a lack of physical correlation between the hourly meteorological conditions and the specified deposition velocity. Therefore it is subject to the same risk of predicting unrealistic air concentrations.

We identified a third approach in which the 95th percentile calculation is performed by running a full set of calculations using hourly meteorological conditions coupled to physically-consistent deposition values and wind-sector site boundary distances. The 95th percentile case is thus rigorously determined from a complete set of physically-realistic dispersion calculations using self-consistent inputs. This ensures that unrealistically high air concentrations are not predicted (e.g., due to the use of a site boundary distance that is much shorter than is correlated with the hourly wind direction) or underestimated through the use of a deposition velocity that does not reflect the actual atmospheric and environmental conditions. In addition, this approach avoids the necessity of defining and determining "reasonably conservative" inputs.

We implemented this third approach by developing a script to automate the calculation of the 95th percentile air concentration based on hourly varying input parameters. The Petroff and Zhang (2010) model was used to determine physically consistent deposition velocities from wind-sector appropriate site surface characteristics (land-use) and boundary distances for each set of (hourly) meteorological conditions.

6.2 PETROFF AND ZHANG DEPOSITION VELOCITY MODEL

The Petroff and Zhang (2010) model calculates the deposition velocity according to the following steps:

- The model internally specifies values for surface characteristic variables (including the leaf area index [LAI], surface roughness, height of the vegetation canopy, displacement height, and characteristic size of the vegetation obstacles) using a default surface properties table and the user provided wind-sector dependent dominant land-use category for the release location.
- The wind speed at the top of the vegetation canopy is calculated using similarity theory based on the friction velocity scaling parameter, the Obukhov length stability parameter, and land-use category derived surface characteristics (the canopy height, roughness length, and displacement height).
- The deposition efficiencies of Brownian diffusion, interception, inertial impaction, and turbulent impaction are then derived. The deposition pathway terms parameterize particle interactions with different categories of vegetation.
- The gravitational settling velocity is calculated for the specified particle size distribution.
- The outputs from the Petroff and Zhang (2010) model are meteorological and land-use dependent deposition velocities for the specified particle size bin.

The model can be run multiple times to de model to determine (hourly) wind-sector dependent values for the deposition velocity for use in 95th percentile analyses.

6.3 MODEL INPUTS FOR PETROFF AND ZHANG DEPOSITION MODEL

Five inputs are required to run the Petroff and Zhang (2010) deposition velocity model:

- Dominant land-use category downwind of the release point
- Obukhov length scale characterizing atmospheric stability
- Surface friction velocity
- Air temperature
- Particle size distribution

Methods for obtaining reasonable values for each of these inputs are discussed below. At least one of the approaches suggested uses only meteorological observations of wind speed /direction and multi-level temperature measurements expected to be readily available at all sites.

6.3.1 LAND-USE CATEGORY

Surface characteristics such as roughness length, displacement height, canopy height, and leaf area index depend on the land-use type. Land-use categories need to be determined for both:

- The meteorological observational station (the dominant land-use category upwind of weather observations may be required to calculate the friction velocity)
- The release location (this land-use category is a direct input into the Petroff and Zhang deposition velocity model

The EPA's AERSURFACE (EPA, 2008) program processes land-use data to develop wind-sector dependent statistics. By default, the AERSURFACE program reads in 30 m resolution land-use data from a state-wide coverage file based on National Land Cover Database 1992 (NLCD 92) categories. Unless otherwise specified, the AERSURFACE model outputs land-use category frequency of occurrence for 12 wind direction sectors (of 30 degree each). In the case studies conducted for this investigation (Section 7), we used a search radius of 1 km surrounding the meteorological observation station as recommended by the AERSURFACE user's guide and a 5 km search radius for the release location (the maximum allowed by the AERSURFACE model). The larger search radius accounts for the downwind travel distance of a plume from the release location to the site boundary.

For sites with boundary distances that are greater than 5 km from the release location (and significantly changing land-use characteristics beyond that distance), the AERSURFACE program should be modified to use larger search radii or an alternative package used to determine the wind-sector dependent land-use categories. More sophisticated approaches may also be beneficial for cases in which the site boundary distance from the release locations varies greatly among the different wind sectors. One option is to run AERSURFACE (or an alternative program) multiple times with varying search radii to find the representative dominant land use type for each wind sector. It is important to note that an alternative package may be needed for sites that have experienced significant changes in land cover over the last two decades as AERSURFACE uses land classification data derived from satellite imagery valid in 1992.

AERSURFACE outputs the percentage of different land-use categories by wind sector. For example, a given 30-degree sector may be determined to consist of 38% evergreen forest, 34% shrubland, and 28% grasslands. This output is then used to determine a single dominant land-use category for input into the Petroff and Zhang model. If a single land-use type has a frequency of occurrence greater than 50%, it should be taken as the dominant land-use type. However, if no land-use category provides more than 50% coverage (as in the example cited above), the determination must be made with more care. Based purely on the frequency of occurrence, the dominant land use category might be assumed to be evergreen forest. However, the combination of shrubland and grassland both of which exhibit lower vegetation canopy heights (corresponding to lower v_d values and higher air concentrations) accounts for more than 50% of the area coverage. In such cases, the dominant land-use type should be chosen to be the most frequently occurring land-use type among those categories with *lower* vegetation canopy height that together account for more than 50% coverage. For our example, this would be shrubland.

The Petroff and Zhang (2010) model uses a different land-use category naming convention than NLCD92. Therefore, the dominant land use category determined from AERSURFACE output needs to be mapped to the corresponding Petroff and Zhang (PZ) model land-use types. A proposed mapping between these categories is given in Table 6-1. Most common land-use categories such as forests, grassland, shrubland, and wetlands are straightforwardly matched between the two conventions. However, some NLCD92 categories (e.g., 'commercial / industrial / transportation' Class # 23) do not have a corresponding PZ land-use category and must be mapped to an alternative category with similar surface characteristics. It should be noted that the Petroff and Zhang (2010) model allows users to easily define additional land-use types and associated surface characteristics as needed. If this approach becomes commonly used, development of a standard methodology for determining the dominant land-use category (including addressing the AERSURFACE distance limitation) and for mapping the NLCD92 into the PZ land-use categories is recommended.

Table 6-1. Proposed mapping between the NLCD 92 and the Petroff and Zhang (2010) deposition velocity model land-use categories

NLCD Class #	NLCD Category	Petroff Class #	Petroff Land Use Category
11	Open Water	1	Water
12	Perennial Ice/Snow	2	Ice
21	Low Intensity Residential	21N	Urban
22	High Intensity Residential	21N	Urban
23	Commercial/Industrial/Transportation	24	Desert
31	Bare Rock/Sand/Clay	24	Desert

32	Quarries/Strip Mines/Gravel Pits	11	Deciduous shrubs
33	Transitional	11	Deciduous shrubs
41	Deciduous Forest	6	Deciduous needleleaf
42	Evergreen Forest	4	Evergreen needleleaf
43	Mixed Forest	6	Deciduous needleleaf
51	Shrubland	11	Deciduous shrubs
61	Orchards/Vineyards/Other	11	Deciduous shrubs
71	Grasslands/Herbaceous	14	Long grass
81	Pasture/Hay	15	Crops
82	Row Crops	15	Crops
83	Small Grains	15	Crops
84	Fallow	13N	Short grass and forbs (needle shape)
85	Urban/Recreational Grasses	13N	Short grass and forbs (needle shape)
91	Woody Wetlands	23	Swamp
92	Emergent Herbaceous Wetlands	23	Swamp

6.3.2 OBUKHOV LENGTH

The Obukhov length is needed by the Petroff and Zhang (2010) deposition velocity model in order to adjust the shape of the near-surface wind profile based on the atmospheric stability. Several approaches are available to determine the Obukhov length from meteorological data:

- Sensible heat flux measurements should be used to directly calculate the Obukhov length scale if such data are available, as this is the most accurate means of determining this parameter.
- Multi-level temperature measurements can be used to calculate the near-surface vertical temperature gradient and correlate it to a Pasquill stability class[28] and Obukhov length according to the guidance found in U.S. Nuclear Regulatory Commission guidance (NRC, 2007) and the stability curves provided by Golder (1972). Table 6-2 shows the Obukhov length as a function of stability class for typical roughness lengths of 10-20 cm.
- The NRC modified sigma-theta method (Mitchell and Timbre, 1979) provides an alternative defensible approach for estimating stability class from the standard deviation of the horizontal wind direction and the wind speed magnitude.

[28] It should be noted that the NRC temperature gradient method includes a stability class G for highly stable conditions. However, this was not used in our methodology since the MACCS2 plume model does not currently incorporate the G stability class. We substituted a stability class F value to remain consistent with MACCS2 modeling capabilities when a G stability class was estimated from the temperature data.

Table 6-2. Relationship between Pasquill stability class, near surface vertical temperature gradient, and the Obukhov length based on NRC guidance (NRC, 2007) and Golder (1972)

Stability Classification	Pasquill Stability Class	Temperature Change with Height (°K/100m)	Obukhov Length (m)
Extremely unstable	A	$\Delta T < -1.9$	-10
Moderately unstable	B	$-1.9 \leq \Delta T < -1.7$	-25
Slightly unstable	C	$-1.7 \leq \Delta T < -1.5$	-50
Neutral	D	$-1.5 \leq \Delta T < -0.5$	∞
Slightly stable	E	$-0.5 \leq \Delta T < 1.5$	50
Moderately stable	F	$1.5 \leq \Delta T$	25

6.3.3 FRICTION VELOCITY

The Petroff and Zhang (2010) deposition velocity model requires the friction velocity to determine the wind speed at the top of the vegetation canopy. The friction velocity scaling parameter is proportional to the surface stress and parameterizes the shape of the near surface wind profile. Friction velocity is not traditionally measured, but it can be calculated from the 10 m wind speed, the Obukhov length, and surface characteristics (roughness length, displacement height, and canopy height) of the dominant land-use category *upwind* of the meteorological station. For this calculation, we recommend use of the same stability functions (Paulson, 1970; Dyer 1974) as implemented in the Petroff and Zhang (2010) model for consistency.

6.3.4 AIR TEMPERATURE

The near-surface air temperature is needed by the Petroff and Zhang (2010) model. Data at only one height level is required as the model assumes that the air temperature is constant within the vegetation canopy. The air temperature is used for the calculation of aerosol Brownian diffusivity.

6.3.5 PARTICLE SIZE DISTRIBUTION

A user-specified particle size distribution is required by the Petroff and Zhang (2010) model to calculate aerosol size specific deposition values. The required input consists of a particle size diameter (µm) and a particle mass density. Users may provide multiple particle size diameter values to generate several ‘bins’ to represent a particle size distribution.

7.0 CASE STUDIES USING RECOMMENDED METHODOLOGY

In this section, we describe the application of our 95th percentile air concentration methodology to two illustrative cases studies. As described in Section 6.1, this approach combines hourly meteorological data (e.g., wind speed, atmospheric stability) with physically consistent values of other input parameters (e.g., deposition velocity, site-boundary distance).

The dispersion model used in this calculation was the DOE safety toolbox code MACCS2 V1.131.1. MACCS2 was run using hourly wind speed, atmospheric stability data and the Brigg's open country Gaussian plume dispersion coefficient. Air concentrations predicted by MACCS2 were analyzed for the actual distance from the release location to the wind-sector dependent site boundary. The resulting air concentration values were then arranged in order of increasing concentration to determine the 95th percentile value.

For these studies, particle size distributions of 0.2 – 0.4 µm and 2 – 4 µm were used for unmitigated/unfiltered and mitigated/filtered releases, respectively. For the simulations based on the Petroff and Zhang (2010) model, a single representative deposition velocity was calculated for the mitigated particle size distribution by first determining individual v_d values for particles with AEDs 0.2, 0.3, and 0.4 µm and then taking the average of the three values (a similar process was utilized for unmitigated releases based on AEDs of 2.0, 3.0 and 4.0 µm). This approach results in slightly different value than using the median value of the particle size distribution due to the non-linear shape of the v_d curve as a function of particle size. The mass density was set to 1500 kg m^{-3} (the Petroff and Zhang [2010] default).

Results were generated and compared for four deposition velocity options:

- **P&Z (2010)**: Hourly wind-sector dependent values for v_d were determined using the Petroff and Zhang (2010) model as described in Section 6 for particle size distributions of AED = 0.2 – 0.4 µm and AED = AED 2 – 4 µm.
- **P&Z (minimum v_d):** The minimum plausible v_d value as predicted by the Petroff and Zhang model was used (determined from the value for the desert land-use category) in order to produce the most conservative air concentrations for the model.
- **HSS (2011):** This default value of v_d = 0.01 cm/s was used for mitigated releases (AED 0.2-0.4 µm) and 0.1 cm/s for unmitigated release (AED 2-4 µm) as specified in HSS (2011).
- **HSS (2006):** The previously recommended default values of v_d = 0.1 cm/s for mitigated releases (AED 0.2-0.4 µm) and v_d = 1 cm/s for unmitigated releases (AED 2-4 µm) were used.

The two locations chosen for these illustrative case studies were selected in order to examine contrasting vegetation, site boundary distances, and terrain (terrain affects the deposition velocity model through the surface roughness height value) conditions. The source for this simulation was a 1Bq ground level point source release so that the output air concentrations provide the plume dilution factor.

7.1 LOS ALAMOS NATIONAL LABORATORY CASE STUDY

Los Alamos National Laboratory (LANL) is situated on approximately 40 square miles in north-central New Mexico. The site contains a series of narrow mesas and canyons set on the western bank of the Rio Grande. The site is bordered by the heavily vegetated Jemez Mountains to the west and the Sangre de Cristo Mountains to the east. Figure 7-1 shows a map of the LANL site.

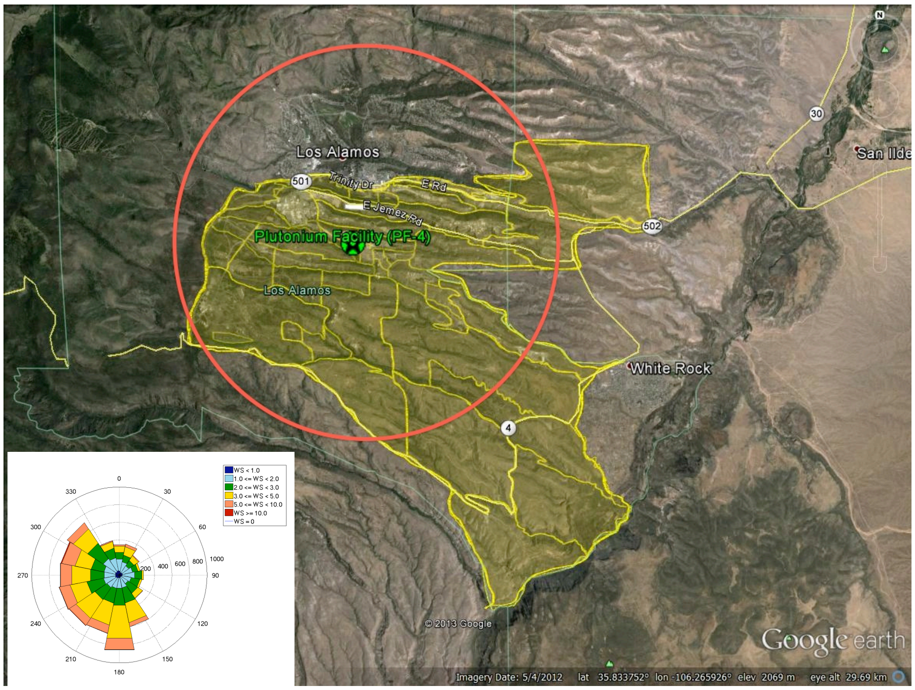

Figure 7-1. Site boundary for Los Alamos National Laboratory is depicted in yellow. The red circle shows a 5 km radius around the release location. The inset shows the a wind-rose depicting the frequency of various wind directions at the site.

For this case study, the source location was fixed at the center of the red circle shown in Figure 7-1. Air concentrations predicted by MACCS 2 were recorded at the wind sector dependent site boundary and sorted to identify the 95th percentiles values. The results are tabulated in Table 7-1 for the four different deposition velocity options and each of the two particle size bins. The air concentrations at the site boundaries are expressed in terms of the normalized values or dilution factors, χ/Q.

For the smaller particle size range, the 95th percentile air concentrations for all deposition models apart from the HSS (2006) option were within 5% of each other, with the P&Z (minimum v_d) mode predicting the highest 95th percentile air concentration (χ/Q) value and the P&Z (2010) model the lowest value. The HSS (2006) case yielded an air concentration that was approximately 15% lower than P&Z (minimum v_d) mode, due to its significantly larger v_d value. For the larger particle size range, the choice of deposition model played a more significant role. The HSS (2006) deposition velocity produced air concentrations that were more than an order of magnitude lower (less conservative) than the other options. The other three options predicted 95th percentile air concentration values that differed by approximately 15%. As before, the P&Z (minimum v_d) 95th percentile air concentration (χ/Q) value was the largest of the three, while the P&Z (2010) value was the lowest. The HSS (2011) air concentrations fell in between, leaning towards the higher concentration side. For the Los Alamos site, P&Z (2010) predicted air concentrations are lower than HSS (2011) values as the Petroff and Zhang (2010) model predicts higher deposition velocities due to the significant vegetation at the site.

Table 7-1. 95th percentile air concentration (χ/Q) values at actual site boundary (in s/m^3)

Small Particles (AED = 0.2 - 0.4 µm)				**Large Particles (AED = 2.0 - 4.0 µm),**			
P&Z (2010)	**P &Z (minimum v_d)**	**HSS (2011)**	**HSS (2006)**	**P&Z (2010)**	**P &Z (minimum v_d)**	**HSS (2011)**	**HSS (2006)**
5.00e-5	5.21e-5	5.15e-5	4.57e-5	4.08e-5	4.86e-5	4.57e-5	5.81e-6

7.2 HANFORD CASE STUDY

Hanford site is a desert environment, covered primarily by shrub-steppe vegetation. The Columbia River flows along the northern and eastern boundary of the site for approximately 50 miles (80 km). Figure 7-2 shows a map of the site. The hypothetical release location was taken to be at the center of the red circle. As before, air concentrations predicted by MACCS 2 were computed at the wind-sector dependent actual site boundaries. These air concentrations were ranked in ascending order to determine the 95th percentile values that are tabulated in Table 7-2.

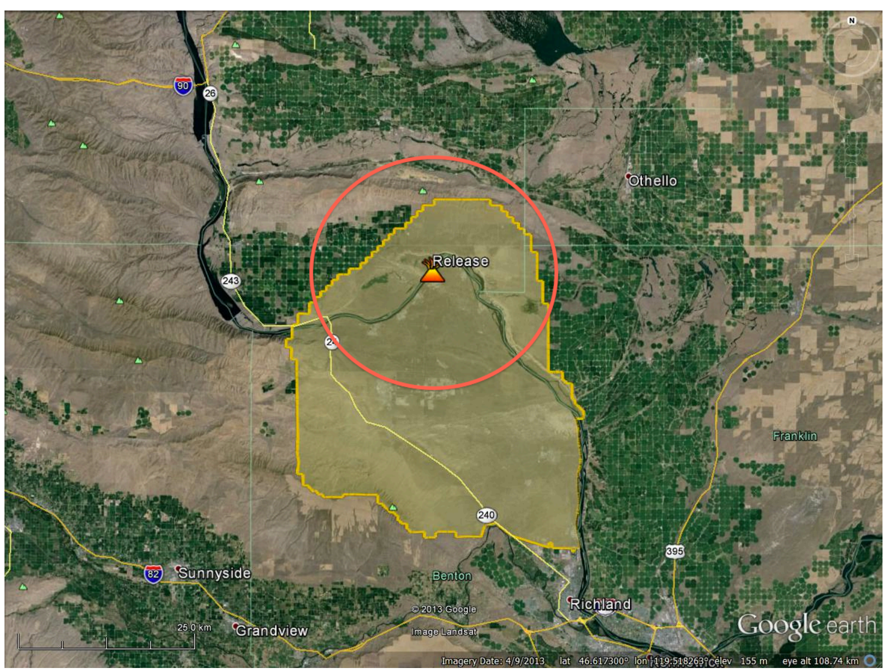

Figure 7-2. Site boundary for Hanford is shown in yellow. The red circle is at 5 km centered on the release site.

For the smaller particle size range, calculated deposition values were low due to the short vegetation characteristic of the site. The 95th percentile air concentration values predicted using the P&Z (2010), P&Z (minimum v_d) and HSS (2011) options were virtually identical (within 3% of each other). The HSS (2006) choice for v_d produced a value ~30% smaller than the other options. For the larger particle size range, the differences in 95th percentile air concentrations were more significant. As before the P&Z (minimum v_d) deposition velocities led to the most conservative outcome (highest estimated air concentration). The HSS (2011) default value predicted an air concentration ~10% lower than the P&Z (2010) and ~20% lower than the P&Z (minimum v_d) options. As in the LANL case study, the HSS (2006) results were an order of magnitude lower than the other three options. P&Z (2010) predicted air concentrations for larger particles at the Hanford site are higher than HSS (2011) values, since the P&Z model's physics-based calculation predicts a reduced deposition velocity due to the absence of significant vegetation at the site.

Table 7-2. 95th percentile normalized air concentration (χ/Q) values at the site boundary (in s/m^3)

Small Particles (AED = 0.2 - 0.4 µm)				**Large Particles (AED = 2.0 - 4.0 µm),**			
P&Z (2010)	**P &Z (minimum v_d)**	**HSS (2011)**	**HSS (2006)**	**P&Z (2010)**	**P &Z (minimum v_d)**	**HSS (2011)**	**HSS (2006)**
1.29e-5	1.32e-5	1.28e-5	0.95e-5	1.05e-5	1.14e-5	0.95e-5	0.87e-6

7.3 SUMMARY

Although these two cases are only illustrative, they provide some evident that the current HSS Safety Bulletin (2011) recommended default values for v_d produce reasonably conservative predictions, predicting air concentrations within 15-20% of those based on the Petroff and Zhang (2010) deposition model. The results also show that for the smaller particle size range, the v_d values used in both the HSS (2011) and Petroff and Zhang models are so small that they do not significantly change the 95th percentile values.

8.0 SUMMARY AND DISCUSSION

8.1 KEY RESULTS

Key findings from this investigation are summarized below:

- *Deposition velocity (Section 4.5; Section 7.2.3).* The Office of Health, Safety, and Security (HSS Safety Bulletin, 2011) currently recommends the use of default deposition velocities of 0.1 cm/s for unmitigated/unfiltered particles with Aerodynamic Equivalent Diameters (AEDs) in the range 2 - 4 μm and 0.01 cm/s for mitigated/filtered releases of particles with AEDs of 0.2 - 0.4 μm. With a few caveats discussed below, these values were found to be generally appropriate for particulate plume modeling, unlike the previously recommended default values of 1 cm/s and 0.1 cm/s. Related findings are as follows:
  - The Petroff and Zhang (2010) model currently provides the most accurate deposition velocity values for a wide range of atmospheric and environmental conditions.
  - The HSS Safety Bulletin (2011) default deposition values are most appropriate for grassland. They are somewhat over-conservative for forests and under-conservative for bare ground, predicting air concentrations ~15% higher or lower, respectively, than the optimal choice of v_d. These are relatively small differences that may not be significant relative to those resulting from uncertainties in weather observations, atmospheric stability, or land-use category.
  - Predicted air concentrations for the filtered/mitigated particle size range (AED = 0.2 – 0.4 μm) are relatively insensitive to the range of potential deposition velocities. Specifically, the current HSS-recommended (HSS Safety Bulletin, 2011) default value of $v_d = 0.01$ cm/s for such particles produces minimal plume depletion and gives virtually the same results as using no deposition ($v_d = 0$).
  - The use of a single deposition velocity that is "conservative" for all sites and scenarios will produce an overly conservative result for many cases (corresponding to unnecessarily high air concentrations and exposures). However, site- and scenario-specific values could be used in initial screening calculations to determine whether a more in-depth analysis is needed (e.g., if calculated doses exceed or are close to the threshold that warrants additional mitigation/protective actions), as discussed below.
- *Sensitivity analysis of key model input parameters (Section 3.3; Section 4.4).* Predicted air concentrations were found to be as or more sensitive to the wind-direction dependent distance to the location of interest (e.g., the site boundary), the meteorology (e.g., wind speed, atmospheric stability class) and the release height, as to the choice of deposition velocity.

- *Observations regarding the use of the MELCOR Accident Consequence Code System (Section 5).* During this investigation, we identified a few aspects of MACCS2 Version 1.13.1[29] that users should be aware of when performing hazard analyses.[30]

  - The Brigg's open country dispersion coefficients produce MACCS2 concentration values more consistent with other commonly-used models (i.e., HotSpot, AERMOD) than those resulting from the default Tadmor and Gur (TG) option. The TG coefficients were derived from experimental data over flat terrain using curve fits that are considered appropriate only over the range 0.5 – 5.0 km and consequently may not be valid for other conditions and distances. It is therefore recommended that the Brigg's open country dispersion coefficients be selected for DOE safety analysis modeling.

  - MACCS2 V1.13.1 does not automatically calculate the wind speed at the release height[31]. Changes of wind speed with height may have a sizeable impact (~40%) on predicted air concentrations for elevated sources. For such cases, the release height wind speed should be externally calculated and input by the user.

  - The absence of a low wind speed algorithm in MACCS2 limits the significance of 95th percentile air concentration calculations for sites at which more than 5% of winds are below a 1 - 2 m/s threshold. For such locations, we recommend the use of alternate codes (e.g., HotSpot) that incorporate special algorithms to cover low-wind speed cases.

  - The changes in MACCS2 predicted air concentrations through the use of more accurate deposition velocity values may be overshadowed by the above corrections for low wind speeds and/or elevated releases.

8.2 RECOMMENDATIONS

Based on this investigation, the following recommendations are made:

- *Recommended deposition velocity model (Section 3.4; Section 4.5; Sections 6-2 and 6-3).* The current state-of-the-science dry deposition velocity model for particles is the Petroff and Zhang (2010) model. If default choices for v_d are inadequate for DOE site accident analysis applications, use of this model is recommended as it parameterizes the impact of a wide range of vegetation types while requiring only a reasonable level of modeling sophistication. Input variables to the Petroff and Zhang model may be calculated or reasonably estimated from meteorological observations that are routinely available at DOE

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[29] MACS2 V1.13.1 is the version of the code included in DOE's Central Registry as a Safety Software toolbox code.
[30] Later version of MACCS2 may have addressed some of these issues.
[31] The standard reference height for surface meteorological measurements is 10 m above ground level.

sites. A methodology for implementation of this model is provided in Section 6 of this report.

- *Recommended 95th percentile air concentration methodology (Section 6.1).* We recommend the use of a more robust approach for determining 95th percentile air concentrations in which hourly wind speed and direction observations are used to determine wind-sector dependent land-use categories, v_d values, and direction-dependent site boundary distances. This method ensures that physically consistent values of the input parameters are used in conjunction with the actual meteorological and environmental conditions.

    - Dominant land-use categories for each wind-sector direction are needed to obtain appropriate deposition velocity values from the Petroff and Zhang model. Due to the complex site boundaries (i.e., the varying distance from source to site boundary with wind sector) and inhomogeneous land-use characteristics at some DOE sites, the EPA's AERSURFACE model (or another alternative software package) may need to be run with varying search radii to determine the appropriate land-use categories for each wind sector.

    - If more than 5% of the wind speeds observed at a site are below a threshold of 1 - 2 m/s, a model should be used that incorporates a low wind speed algorithm (e.g., the HotSpot G stability option documented in Homann and Aluzzi, 2013).

- *Proposed approach for performing atmospheric transport calculations for DOE safety analyses (Section 6).* A two-step hierarchical approach is proposed for performing DOE safety analysis modeling. If the first highly conservative screening step results in levels exceeding or close to specified air concentration thresholds, a second level analysis can be performed to provide a higher-fidelity, but still conservative, analysis.

    - Level 1 Screening Calculation: Perform standard 95th percentile calculations using a lower bounding value for the deposition velocity to determine if a more sophisticated model is required.

        - Option A. Use $v_d = 0$ for all land-use conditions.

        - Option B. If Option A produces overly conservative estimates of exposures, select an alternative site- and scenario-specific lower bounding value for v_d derived from the Petroff and Zhang [2010] model and associated experimental results. This option requires a careful justification of the conservatism of the selected value(s) for the specified particle size range and environmental conditions of interest, particularly for sites with inhomogeneous environmental conditions and/or diverse release scenarios. Use of the current HSS recommended default values is discussed below:

            - ❖ The recommended HSS Safety Bulletin (HSS, 2011) default value of v_d = 0.1 cm/s for unmitigated/unfiltered releases is a lower bounding value for forests, but it falls in the mid-range of deposition velocity values for grasslands and is higher than the Petroff and Zhang (2010) values for bare ground conditions.

- ❖ Although the recommended HSS Safety Bulletin (HSS, 2011) of v_d = 0.01 cm/s for mitigated/filtered releases does not represent a lower bound for v_d for all environmental conditions, this non-zero value produces virtually the same results as a deposition velocity of zero.

- *Use of more sophisticated computer codes for safety analyses (Appendix B).* The use of more sophisticated codes (e.g., non-Gaussian plume models) for safety analyses is difficult to justify.

  - o More sophisticated models account for the time-variation in meteorological conditions and therefore can produce time-averaged or time-integrated air concentrations that are less "conservative" (e.g., have a greater frequency of predicting concentrations that are less than those observed) than Gaussian plume models that use steady-state meteorology.

  - o The accuracy of more sophisticated models over the full range of conditions used in safety analysis modeling is hard to assess.

    - ▪ Past tracer study comparisons have shown that it is not possible to draw universal conclusions regarding the accuracy of such models from individual studies (e.g., different models perform better than others depending on the study). Therefore, experimental validation typically needs to be performed on a case-by-case basis for each location and release type.

    - ▪ Tracer studies typically do not cover the full range of atmospheric stability and meteorological/environmental conditions. In particular, there are very few experimental studies that include data for the very stable, low-wind conditions typical of 95^{th} percentile meteorology.

  - o It is more difficult to set up sophisticated model simulations to ensure conservatism of the results than is the case with Gaussian plume models that exhibit simpler dependencies on input parameters. Over-riding internal model physics by user-specification of parameter values (e.g., the deposition velocity) may result in the use of inconsistent physics and diminish any benefits of using a model designed to simulate complex conditions.

  - o The expertise and resources (personnel and computational) required to use more sophisticated models may be cost prohibitive. Complex models require trained users to properly specify all of the input variables and options needed to produce accurate analyses and quality-assure results. The use of improper inputs or physical/numerical options is a significant risk in the use of more sophisticated models.

- Most sophisticated models have not been included in the DOE Central Registry for Safety Software toolbox codes, in part because of the significantly greater Software Quality Assurance effort required[32]. Therefore an extensive justification for their use and application, as well as a thorough review of inputs and results, would be required for hazard analysis applications.

- In specific cases (e.g., in cases of complex terrain when representative meteorological observations are available) the use of more sophisticated models may be justified if specialized expertise is available to conduct complex dispersion modeling, and the risks and issues discussed above can be addressed.

- *Standardization of methodologies (Section 6.1; Appendix B)*. Based on our investigation, we also recommend that DOE should establish clear definitions and methods for selecting "reasonably conservative" input parameters, accompanied by documented procedures standardizing the approach for conducting 95th percentile calculations for safety analyses.

8.3 OTHER SCENARIOS

In order to keep the scope of this project within available resources, a number of different release scenarios were not investigated. The conclusions of this study do not necessarily apply to such cases, including:

- Gas releases (identified as a less common risk in the DOE site surveys)
- Evaporative releases (sprayers, spills, leaks) for which the particle size distribution is likely to decrease during transport and dispersion
- Unmitigated explosive releases (that create significant numbers of particles significantly outside the assumed "mitigated" and "unmitigated" particle size distributions)
- Buoyant or momentum-driven plumes and elevated release heights (apart from a limited sensitivity study for different release heights)
- Tritium releases (specifically excluded by HSS and Defense Nuclear Facility Safety Board)
- Iodine releases from a reactor or criticality accident
- Reactive materials (which undergo changes in chemical or physical state in interactions with the environment)
- Releases scenarios and exposure periods for which ground-shine dose from deposited material is the primary risk (for such cases *higher* deposition velocities would generally provide more conservative results)
- Releases involving the ingestion pathway (e.g., water distribution system contamination)

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[32] One exception is GENII, but its use has been deprecated for the submicron particle size range because this model calculates a constant deposition velocity value over this size range and does not match the expected theoretical minimum (see HSS Safety Bulletin, 2011).

8.4 IMPLEMENTATION OF RECOMMENDATIONS

The 95th percentile methodology and Petroff and Zhang (2010) deposition velocity approach recommended in this report can be implemented for DOE site safety analyses in several ways:

- LLNL's 95th percentile software may be operationalized for distribution to DOE sites or independently developed by the sites.

- DOE sites may obtain the Petroff and Zhang source code directly for the authors[33] and modify the code to use available site meteorological observations.

- A software package may be developed for distribution to DOE sites that reads in wind-sector dependent dominant land-use categories and hourly meteorological observations and uses the Petroff and Zhang model to calculate site-specific, hourly deposition values.

- Site-specific deposition velocity tables as a function of wind speed, land use category, and atmospheric stability may be calculated using the Petroff and Zhang model and distributed to the DOE sites.

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[33]The Petroff and Zhang deposition velocity model is available under the Creative Commons Attribution Non-Commercial Share Alike 3.0 licensing associated with the model source code. Users of this software are required to (a) credit the model developers in any technical reports or scientific publications, (b) ensure the model is not used for commercial applications, and (c) ensure any modified Petroff source code version falls under the existing license agreement. Implications of 'non-commercial' use need to be investigated to fully understand how DOE site contractors may use the Petroff and Zhang model.

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[34] Formerly Washington Safety Management Solutions

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APPENDIX A. DOE SITE SURVEYS

LLNL developed and distributed the DOE site survey below with input and assistance from the DOE EMI Subcommittee on Hazard Assessments, the Energy Facilities Contractors' Group (EFCOG), and Caroline Garzon (DOE CNS). A separate Official Use Only document contains the raw responses from the DOE site survey.

A.1 ATMOSPHERIC DISPERSION MODELING SURVEY

We are soliciting your input for an effort requested by the Department of Energy's Office of Health, Safety, and Security (point of contact: Caroline Garzon, Chief of Nuclear Safety Staff) "to develop an approach for the calculation of 95th percentile doses by identifying reasonably conservative values for key parameters (in particular, deposition velocity) to be used in the dispersion models, primarily the MACCS2 code."

A key initial step in this effort is to establish representative release scenarios, site characteristics, and plume modeling criteria through expert solicitation from the DOE site community. We are requesting your input to help us understand the requirements, inputs and needs for atmospheric dispersion and deposition modeling for safety analysis and emergency planning at DOE sites. Our goal is to ensure we consider the full range of potential release scenarios and that the project results address both scientific and operational aspects. As this effort progresses, we may contact you again to seek feedback on whether proposed methods can be practically implemented by your site(s).

We greatly appreciate the assistance of the EMI SIG Hazards Assessment Subcommittee Chair (HASC) Michele Wolfgram, EFCOG Accident Analysis Subgroup Chair Mukesh Gupta, and EFCOG Safety Basis Subgroup Chair Nathan Cathey, who are facilitating the collection of this information.

Gayle Sugiyama and John Nasstrom
National Atmospheric Release Advisory Center (NARAC)
Lawrence Livermore National Laboratory

Your Contact information	
Name	
Organization / DOE Site	
Email	
Phone	

We request that you enter information in the following table format, creating a separate table for each representative type of atmospheric release that needs to be considered. You may combine similar releases in one table and provide a summary of the range of assumptions used for those releases. Partial information may be helpful (please enter “Unknown” if you do not have the information available).
We would appreciate your response by **December 31, 2012**. Please email responses to sugiyama@llnl.gov and nasstrom1@llnl.gov.

Thank you in advance for any assistance you can provide!

Question	Answer
1. What is the application? (e.g., Safety Analysis, Emergency Planning)	
2. What radionuclides are released to the atmosphere? (e.g., Pu-239, I-131)	
3. What are the chemical forms released to atmosphere? (e.g., PuO_2)	
4. What is the release scenario? (e.g., explosion, venting, filtered or unfiltered fire, spill, leak, criticality, reactor)	
5. What is the release height or range of heights? (e.g., ground level to 20 m)	
6. What is the physical form of material released to atmosphere? (e.g., solid particle, liquid droplets, gas)	
7. What is the particle diameter, range or distribution? (e.g., Uniform 2-4 μm AED) Note: Please specify if this is the physical diameter, or aerodynamic equivalent diameter (AED)	
8. What is the particle density? (e.g., 4 g/cc, or enter N/A if particle diameter was given in aerodynamic equivalent diameter)	
9. What is the release duration? (e.g., 10 to 60 min)	
10. What is the exposure time duration of interest? (e.g., 8 hours)	
11. What dry deposition velocity values or methods are currently used or planned for this scenario? (e.g., HSS Safety Bulletin No. 2011-02 values, or GENII V2 code calculated)	

12. Which dose pathways are considered? (e.g., initial plume inhalation, cloudshine, groundshine, resuspension inhalation)	
13. What are the distances or range of distances from sources to receptors of interest / site boundaries? (e.g. 0.5-1 mi)	
14. What atmospheric dispersion modeling codes are used? (e.g., MACCS2)	
15. Which methodology is used for meteorological data? (a) persistent meteorology, and/or (b) 95th percentile method using historical weather data	
16. What meteorology data is used? (e.g., wind speed, wind direction, sigma theta and temperature from 10 m level of site tower)	
17. What method is used to determine stability class? (e.g., EPA-454/R-99-005 Sigma-theta method)	
18. What surface roughness length value or range of values is used, and what methods were used determine the values? (e.g., 20 cm, NUREG/CR-4691 Table 2.3)	
19. What is the land cover and topography in the area of interest? (e.g., dessert sage brush, hills with 500 ft elevation changes)	

A.2 DOE SITE SURVEY RESPONSES

See Appendix A-2 for survey responses from the DOE sites (provided in a separate Official Use Only document LLNL-MI-655192).

APPENDIX B. REVIEW OF RISK ASSESSMENT CORPORATION ANALYSIS

In 2011, the DOE Chief of Nuclear Safety (CNS) requested that the Risk Assessment Corporation (RAC) perform an independent analysis of the use of MACCS2 and the appropriate value of the deposition velocity for the Waste Treatment and Immobilization Plant (WTP) in Hanford WA. RAC's initial analysis concluded that an appropriate range of deposition velocities for this site would be 0.1 - 0.3 cm/s, but also that the use of a deposition velocity (v_d) value of 1 cm/s would not underestimate the dose at the WTP due to the other conservatisms built into the MACCS2 hazard analysis. A follow-on effort described in the report *Comparison of Maximum Hourly Dispersion Factors Computed with Lagrangian Puff Models and the Gaussian Plume Model in MACCS2* (hereafter referred to as Till et al., 2011) was conducted to quantify the conservatism of the MACCS2 code and to determine whether the predicted MACCS2 dose met the 95th percentile dose criteria specified in the DOE-STD-3009 dose guidance (DOE, 2006). This Appendix provides a peer review of that report.

The Till et al. (2011) report compared MACCS2 plume calculations for the Hanford WTP site with two widely-used Lagrangian puff models – CALPUFF (Scire et al., 1990) and RATCHET (Ramsdell et al., 1994). The metric of comparison was the 95th percentile highest hourly average concentration at distances of 1 km, 5 km, and 9.3 km from a source consisting of a ground-level unit release of particulate matter in the unmitigated/unfiltered size regime. In this study, all models were run in their intended mode to obtain their best estimates of the air concentration dilution factors. The MACCS2 modeling was performed using the approach prescribed in DOE-STD-3009 and the DOE MACCS2 Guidance document, with v_d specified to be 1.0 cm/s. A summary of some of the key differences between these simulations is provided below for reference:

- *Terrain*. MACCS2/RATCHET: no terrain; CALPUFF: no terrain and terrain elevation data taken from the USGS 1 degree digital elevation model
- *Land use.* MACCS2: surface roughness length only (z0 = 3 cm); RATCHET: surface roughness length only (z0 = 5 cm) CALPUFF: 14 land-use parameters
- *Meteorological data and methods for generating wind fields.* MACCS2: single observation, RATCHET: $1/r^2$ interpolation using data from 2 meteorological stations; CALPUFF: mass-consistent wind field developed using a weighted interpolation of data from 10 stations
- *Dispersion coefficients*. MACCS2: Pasquill-Gifford dispersion coefficients; CALPUFF: Pasquill-Gifford dispersion coefficients option and similarity-theory derived coefficients; RATCHET: stability class pre-processor based on Turner (1964)
- *Deposition velocity.* MACCS2/RATCHET/CALPUFF: zero deposition velocity, MACCS2: fixed deposition velocities of 0.0, 0.1, 0.3, and 1.0 cm/s; RATCHET[35] and CALPUFF: different resistance-model derived spatially and time-varying values depending on the atmospheric conditions and surface roughness

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[35] Till et al., (2011) noted that the RATCHET model does not include gravitation settling it its deposition velocity model. In this comparison, this was accounted for by assuming a transfer resistance value of 1000 s/m.

- *95th percentile methodology*. MACCS2: Latin Hybercube sampling; RATCHET and CALPUFF: 95th highest value from the distribution of the maximum concentration at the receptor distance

The numerous conceptual differences in model formulations, as well as in the simulation set up and inputs used, make it difficult to draw clear conclusions regarding the underlying reasons for some of the differences between the model results.

A few general considerations regarding dispersion model accuracy relevant to this review are summarized below:

- The current state-of-the-science in dispersion modeling is commonly taken to be a factor of 2 agreement between model predictions and experimental measurement data for relatively simple releases in homogeneous environments and a factor of 10 agreement for complex releases and environments. For example:
    - Miller and Hively (1987) examined validation studies for Gaussian plume models and concluded that measured air concentrations can be predicted within a factor of 2 to 4 for annual average air concentrations over flat terrain, but that accuracy decreases to factors of 10 - 100 as the averaging time decreases and/or the complexity of the meteorological and terrain conditions increases.
    - Foster et al. (2000) found that for more sophisticated models, point-to-point comparisons of predicted air and ground contamination values are typically (50% or more of values) within a factor of 2 of measured data for simpler meteorological, terrain and source conditions, and a factor of 10 for more complex conditions.
- The accuracy of more sophisticated models over the full range of conditions used in safety analysis modeling is hard to assess.
    - Past tracer study comparisons have shown that it is not possible to draw universal conclusions regarding the accuracy of such models from individual studies (e.g., different models perform better than others depending on details of the study) so experimental validation typically needs to be performed on a case-by-case basis for each location and release type.
    - Tracer studies typically do not cover the full range of atmospheric stability and meteorological/environmental conditions. In particular, there are very few experimental studies that include data for the very stable, low-wind conditions typical of 95th percentile meteorology.

Limited information was provided in Till et al. (2011) regarding experimental verification and validation testing of the various models for the WTP site. The only experimental data referenced in the report was for a different site (Rocky Flats) in which Lagrangian puff models were found to predict concentrations of between a factor of 0.9 and 1.9 of the measured data[36]. However, as noted above, it is difficult to generalize about the accuracy of complex models based on experimental studies for locations or atmospheric/environmental conditions different than those under which the experiments were conducted[37].

Till et al. (2011) provided 95th percentile dilution (χ/Q) value comparisons for the following cases:

- MACCS2 (using v_d values of 0.0, 0.1, 0.3, and 1.0 cm/s)
- CALPUFF run without terrain and using the code's Pasquill-Gifford (PG) dispersion coefficient option[38]
- CALPUFF using standard options
- RATCHET

In addition, all four models were run using an imposed zero deposition velocity. The plume dilution factors ((χ/Q) for the v_d = 0 case were essentially the same for all models at 1 km downwind, but diverged farther downwind, with MACCS2 concentrations being a factor of 3-4 times greater at 9.3 km. Qualitatively, the air concentrations results were approximately the same for all three non-Gaussian models. This result was consistent with the expectation that the differences between MACCS2 and the three more sophisticated models would be dominated by the effects of temporally- and spatially-varying meteorology included in the latter (non-Gaussian) codes, with lesser differences occurring due to the use of different physical process algorithms in each of the models.

For the non-zero deposition velocity cases specified above, the MACCS2 air concentrations ranged from 1 to several times that predicted by the other models at 9.3 km. For the largest assumed deposition velocity of 1.0 cm/s, MACCS2 produced approximately the same concentration as the more sophisticated models using site-specific meteorological data and an internally-calculated $v_{d.}$ However, at shorter downwind distances (1 km and 5.3 km), MACCS2 did not produce the most conservative air concentrations / dilution factors for v_d values of 0.3 and 1.0 cm/s.

From our review of the Till et al. (2011) results, we drew the following conclusions:

- Based on the comparisons shown in the report (e.g., Tables 6 and 7 in Till et al., 2011), the most appropriate conservative value of deposition velocity for the WTP site and scenario

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[36]Separately, we found a reference to a study for RATCHET, a code specifically developed for use at Hanford, (Till, J.E, and Rood, A.S., 2012: Evaluation of the Atmospheric Transport Model in the MACCS2 Code and its Impact on Decision Making at Department of Energy Sites, presentation at MACCS2/Deposition Velocity Workshop, Germantown MD, 2012 June 5-6) that showed that the model over predicted Kr-85 concentrations at Hanford by about a factor of 1.45, although in cases using limited meteorology it occasionally under predicted.

[37]For example, the typical meteorology for the 95th percentile case at the WTP site was found to correspond to F stability and 1.7 m/s winds. However, it is not clear that these conditions were included in the experimental studies referenced.

[38]It should be noted that although MACCS2 also uses a Pasquill-Gifford (PG) approach, the CALPUFF and MACCS2 PG models are not identical.

studied appears to be 0.1 cm/s, although values of up to 0.3 cm/s might be appropriate if only the site boundary distance of 9.3 km is of interest.

- The results of the model comparison study indicate that the WTP safety analysis performed using the v_d value of 1.0 cm/s did not produce grossly non-conservative exposure values. However MACCS produced very similar χ/Q values as the more-sophisticated Lagrangian puff models (ratios of 1.01 – 1.35 at 9.3 km). Given the lack of demonstrated conservatism of the latter models, this is insufficient to demonstrate that the use of $v_d = 1.0$ cm/s meets the degree of conservatism specified in DOE guidance (DOE, 2004) that "even if a single value in the dose calculation were off by an order of magnitude, the resulting value would still not approach the mean value of dose if a cumulative distribution of dose were also calculated".

- We concur with Till et al. (2011) that Gaussian plume models are likely to become more conservative at longer downwind distances as was born out by the results of the comparison study. However, the distance for which this occurs will be both site and release scenario dependent. Specifically, it should be noted that this study does not establish that a default value of v_d = 1.0 cm/s will result in "conservative" dose estimates for other distances, locations, or release scenarios.

The Till et al. (2011) report concludes with a short list of recommendations that are discussed below:

- The report recommends that the MACCS2 code be used as a screening tool for safety-related applications using a site-specific default value for v_d. We concur with the use of an initial (simpler) screening approach, although based on our investigation we recommend that the screening calculation use a lower bounding value of either zero or one derived from the Petroff and Zhang (2010) model and associated experimental results (see Section 4.5; Section 8.2).

- The use of more sophisticated codes (e.g., non-Gaussian plume models) for hazard analyses is difficult to justify. Specifically:
    - More sophisticated models account for the time-variation in meteorological conditions and therefore can produce time-averaged or time-integrated air concentrations that are less "conservative" (e.g., have a greater frequency of predicting concentrations that are less than those observed) than Gaussian plume models that use steady-state meteorology.
    - It is more difficult to set up sophisticated model simulations to ensure conservatism of the results than is the case with Gaussian models that exhibit simpler dependencies on input parameters. Over-riding internal model physics by user-specification of parameter values (e.g., deposition velocity) may result in the use of inconsistent physics as well as diminishing the benefits of using a model designed to simulate complex conditions.

- The expertise and resources (personnel and computational) required to use more sophisticated models can be cost prohibitive. Complex models require trained users to properly specify all of the input variables and options needed to produce accurate analyses and to quality-assure results. The use of improper inputs or physical/numerical options is a significant risk in the use of more sophisticated models. The wide range of differing model set up choices and input parameters in the Till et al. (2011) comparison as summarized above is illustrative of this point.

- Most sophisticated models have not been included in the DOE Central Registry for Safety Software toolbox codes, in part because of the significantly greater Software Quality Assurance effort required (one exception is GENII, but its use has been deprecated for the mitigated/filtered particle size range). Therefore an extensive justification for their use and application, as well as thorough review of inputs and results, would be required for hazard analysis applications.

- In specific cases (e.g., in cases of complex terrain when representative meteorological observations are available) the use of more sophisticated models may be justified if specialized expertise is available to conduct complex dispersion modeling, and the risks and issues discussed above can be addressed.

- The Till et al. (2011) recommendations include a statement that "[a]lthough models such as MACCS2 are useful for initial accident assessment… more robust models using site-specific data would provide a more accurate result when responding to a release". Although we concur with this statement, it is important to point out that both Gaussian plume models and more sophisticated models already have important and well established roles in emergency response.

  - Fast-running Gaussian plume models (e.g., HotSpot, MACCS2) may be run for timely initial assessments. More sophisticated models can be used later, as the event unfolds and more detailed simulations are needed.

  - DOE sites that have the potential for a General Emergency or a Site Area Emergency due to an atmospheric release of a hazardous material are required to have access[39] to the National Atmospheric Release Advisory Center (NARAC) under DOE Order 151.1C (Section IV.3.b.5 and Attachment 2, Section 13). NARAC provides both expertise and state-of-the-science modeling tools to predict and assess the consequences of actual releases. NARAC staff works closely with field monitoring teams, deployed assets, the DOE/NNSA Consequence Management Home Team (CMHT), and the Federal Radiological Monitoring and Assessment Center until the impacts are fully characterized.

[39] The level of use of NARAC modeling support is tailored to site needs and complements other modeling tools used at the DOE site.

- Till et al. (2011) concludes with a recommendation that DOE should "establish a target level of conservatism to be used in decision making related to nuclear safety" and states that their "analysis was complicated by a lack of clear definition for the level of conservatism the Department of Energy is seeking in the calculated dose and its input assumptions". While we believe that the 95^{th} percentile provides a clear overall dose criterion, we strongly concur that it is important to establish clear definitions and methods for selecting "reasonably conservative" input parameters accompanied by documented procedures standardizing the approach for conducting 95^{th} percentile calculations (Section 6).