

ABSTRACT

SRIMOK BOONCHAWEE. Investigation of Particle Size Effects in Inhalation Dose Assessment For Short Term Radiological Events. (Under the direction of Man-Sung Yim).

Given the concern over terrorist events involving radioactive materials, developing the modelling capability to analyze the radiological consequences of a terrorist event involving radioactive material is in demand. One of the remaining questions in modelling radiological consequence of a RDD terrorist event is how should the particle size effect be treated.

A FORTRAN code called “PIDA” was developed in this research to examine the importance of particle size effect in the consequence analysis of a short-term terrorist event involving RDD. Given the focus of this study on the particle size effect, only the human inhalation dose exposure was modeled in the code. The code is established by coupling the conceptual Gaussian puff model with the inclusion of particle size dependent, dry deposition velocity and resuspension modelling capability and ICRP inhalation dose model through its database software. An uncertainty analysis version of PIDA was also developed in order to perform uncertainty analysis for the particle size effect and other key parameters.

The PIDA code was successfully developed and implemented (under certain assumptions) to analyze the effect of particle size distribution in the consequence analysis of radiological terrorist event. For the benchmarking of the code, each submodels of PIDA code were compared with the state-of-the-art modelling tool for each specific area for the calculation results. It was found that PIDA code in general is conservative in estimation the radiological consequence of the event.

Sensitivity and uncertainty analysis for the particle size effect were performed using PIDA code. Results indicated that particle size is one of the key parameters that contributes to the uncertainty of an inhalation dose evaluation due to this terrorist event. Ignoring particle size distribution is expected to result in overestimation of inhalation dose in a radiological terrorist event. It was also found that the particle size effect is (1) Minor in describing atmospheric transport, (2) Minor in describing deposition, (3) Minor in describing resuspension and, (4) Significant in describing lung deposition and resulting the dose.

INVESTIGATION OF PARTICLE SIZE EFFECTS IN INHALATION DOSE ASSESSMENT FOR SHORT TERM RADIOLOGICAL EVENTS

BY

BOONCHAWEE SRIMOK

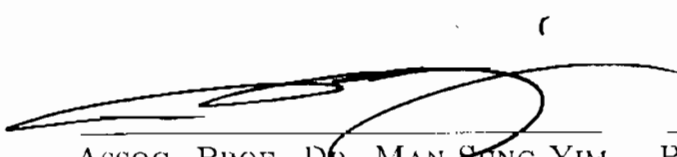

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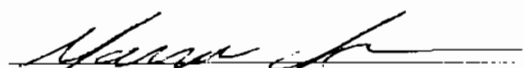
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*This thesis is dedicated to my respected parents,
Pon Srimok and Boonsom Srimok,
my beloved sisters, Supanee and Pachanee Srimok
and brother, Supasak Srimok
and to my dear husband Narongsak Chamchoy*

Biography

Boonchawee Srimok, daughter of Pon Srimok and Boonsom Srimok, was born in Ayudthaya, a small town and old capital city of Thailand about 70 km north of Bangkok, Thailand, on Decemer 2, 1972. In May 1991, she was accepted by the department of Chemistry, faculty of Science of King MongKut Institute of Technology at Ladkrabang. She earned her B.Sc. degree in Industrial Chemistry July 1993. In the same year, she continued her student life at Chulalongkorn University in Department of Nuclear Technology. She earned her M.E. in Nuclear Technology in April 1997. Right after that, for seven years she began her professional career as a nuclear chemist in Office of Atomic Energy for Peace(OAEP) which has become the Office of Atoms for Peace. Not long after that, she met her soul mate and married Narongsak Chamchoy in October 2003. Along with the support and encouragements from her colleagues to continue studying in nuclear engineering, she was selected to receive a full scholarship from the Royal Thai Government. In 2004, she began her graduate study in the Nuclear Engineering Department at North Carolina State University.

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Chapter 1

Problem Statement

1.1 Radiological Dispersal Device in Terrorist event

Radiation can be released from sources such as nuclear facility, natural environment and terrorist events. Interestingly, among these three origins, the event that draws the most attention, especially after 911 event, is the terrorist event due to its unpredictable and potentially catastrophic effect on humans. Although a terrorist event is considered a very low probability by terrorism experts [9], the nature of terrorism itself is that it is unpredictable. Regardless of its low probability, the most important aspect is how we are affected beyond the immediate area of the attack. A radiological dispersal device(RDD) is an unconventional weapon that a terrorist might use to destabilize a community. The RDD is described as any method used to deliberately disperse radioactive material to create terror or harm. A RDD is designed to project or release radioactive material in an area in an attempt to harm people and disrupt society. The relative lack of knowledge the public currently has about radiological materials makes an RDD an ideal choice for terrorist activities. A dirty bomb is an example of an RDD. It is made by packaging explosives with radioactive material to be dispersed when the bomb goes off. Although often used to represent a dirty bomb, the radioactivity in an RDD could also be distributed passively

(nonexplosively), such as through spraying or spreading by hand. Alternately, a radiological exposure device (RED) might be used, which would simply involve placing a radioactive source in a public area to expose people passing by. Radionuclides to be used for this purposes are typically illegally brought from a variety of applications such as industry, medicine, and scientific research applications. Spent nuclear fuel or radioactive waste from the nuclear power industry or legacy weapons facilities is also considered a possible source of RDD with their various physical and chemical forms, and levels of radioactivity. The wide range of possible material choices, package designs, release methods, and target locations make the experimental study and computer modeling of an RDD event difficult and imprecise.

In order to better inform the public on what a RDD is and what terrorists might intend to try to accomplish in setting off such a weapon, straightforward information must be provided. Radiological assessment of such events is one of the most important steps in risk management of such events that must be thoroughly investigated. The release of radiological material from a spent nuclear fuel shipment, either through an accident or sabotage, is of interest and served as a case study in this research. Although many types of RDD can be made due to the creativity of the deranged mind, the inhalation doses received by individual at certain distant via air transportation due to the RDD events is of interest in these research since it could create significant effects on both psychological and health for large number of individual.

1.2 Modelling the consequence of a radiological terrorist event

When the radioactive substance is released, regardless of where and how it originated, it can cause an undue health effect which could harm humans one way

or another. There are typically two main ways that humans could be exposed to radiation:(1) Externally through radiation exposure,(2) Internally through radioactive substance intake (either ingestion or inhalation). Although there are many radiation pathways that are potentially risky to humans, the inhalation pathway (which includes air transport) is considered to be a significant one in this research. After all people could avoid a potentially contaminated site or avoid eating/drinking a suspected contaminated food/drink, but they could not prevent radioactive material from being inhaled. Typically, there is a wider range of fallout downwind from a nuclear device or explosion. Since nuclear fallout does spread via wind, it is necessary to calculate the downwind concentration of radioactive materials and the resulting inhalation dose to perform risk management exercises to protect people.

Typically computer models are used to determine the concentration of radioactive materials and the resulting inhalation dose. Many computer codes have been formulated for this purpose. Typically these computer codes use the Gaussian plume model to determine the airborne concentration of radioactive materials . The Gaussian plume model is known as a steady state model, meaning that it is not capable of determining the concentration in a non-steady state event such as terrorist and accident events. As the nature of the release cause by accident or terrorist event is considered a short-term and non-steady release, airborne concentration of released materials should be modelled by a time-dependent model such as the Gaussian Puff model. Another important aspect in modelling the consequence of a terrorist RDD event is the consideration of the size of the released particles. As the fate and transport of the released matter is controlled by the particle size, the models used to track the fate and environmental transport of released radioactive materials need to take into account the particle size effect in its modelling. However, most of the models available to perform consequence analysis/radiological assessment of a short-term radiological incident ignore the effect of particle size distributions. For a realistic modelling of the

consequences of a short-term radiological terrorist event, there is a need for a new computer model that is capable of analyzing atmospheric dispersion of the released materials in a time-dependent fashion and considering the effect of the particle size in describing the fate transport, human uptake, and dose impact.

1.3 Objectives and scope of research

As stated previously there is a need to develop a new computer code that is capable of calculating the public inhalation dose as a consequence of terrorist event by using the Gaussian puff model with a built-in capability to consider the effect of particle sizes of airborne materials. The purpose of this research is to develop a new computer model to meet this need and to investigate the effect of particle size in the determining human dose under a radiological terrorist event. Specific tasks performed in the research include, a FORTRAN code name “Puff Particle size Dependent with Resuspension and Deposition Inhalation Dose Assessment” or “PIDA” was developed for this purpose. For these tasks, a case study was developed based on a hypothetical scenario of a terrorist event. This thesis will be structured as following. The first chapter explains why this research is needed. The second chapter provides the review of relevant literature. Chapter 3 describes all the necessary theories, concepts and models that will be implemented in the new computer code. Benchmarking of the new computer code based on the comparisons of the code results with those from existing model, is also described in chapter 3. Chapter 4 provides calculation results and analyses of the case study. Chapter 5 presents the discussion of the results and suggestion for future studies. Finally, Chapter 6 includes conclusions.

Chapter 2

Literature Reviews

2.1 Computer Codes

Models can be divided into a physical models and mathematical models. The first model is generally derived from a small scale, laboratory representations of phenomena as opposed to the latter one which is derived from set of analytical or numerical algorithms that described the physical or chemical aspect of the system. Apart from that classification, models can also be divided into deterministic and statistic model. Deterministic model is quite important in term of practical applications since it gives an exact figure once the relationship has been defined. Whereas statistical model is given by the forecast of simulation of events, this gives more information regarding the uncertainty of the model but most of the time requires long calculation time. Most of the computer codes developed for RDD radiological assessment are deterministic physical models.

Due to the hypothetical nature of RDD scenario and lack of experimental data, computer modelling is a exercised to evaluate the consequence of a RDD event. Many attempts have been made to determine the radiation doses and risks using the computer modelling. An inevitable drawback for using computer codes is the facts that they were constructed as approximation of physical relation and developed based on

certain assumptions. People have been estimating the dose consequence with the helps of these tools for years. The task of evaluating consequences of a RDD event involves many diverse subtasks including the estimation of the amount of radioactivity released (source term evaluation), the determination of significant pathways and transport mechanisms, the determination of the exact concentration of radioactive substance transported through the air to a certain location, and the determination of health effects due to the inhalation of radioactive materials. All of the subtasks need to be carefully exercised. Typically most of the available computer codes are not capable of performing all of these tasks.

The following shows some examples of related computer codes that were developed for such purposes. These codes have their own advantages and disadvantages as will be discussed in next section.

2.1.1 Codes for Air Dispersion Modeling

Those computer codes used to calculate the concentration of airborne contaminants at receptor locations involve an air transport modelling.

CALPUFF

CALPUFF [45] is an advanced non-steady-state meteorological and air quality modeling system developed by the Atmospheric Study Group scientists of TRC Companies, Inc. The model has been adopted by the U.S. Environmental Protection Agency (U.S. EPA) in its Guideline on Air Quality Models as the preferred model for assessing long range transport of pollutants and their impacts on Federal Class

Class I areas and on a case-by-case basis for certain near-field applications involving complex meteorological conditions. CALPUFF code was developed by employing the Gaussian Puff model, as its name implies. The code package is composed of CALMET, CALPUFF and CALPOST modules. CALMET is a meteorological model that develops hourly wind and temperature fields on a three dimensional gridded modeling domain. Associated parameters such as mixing height, surface characteristics, and dispersion properties are also included in the file produced by CALMET. CALPUFF performs transport and dispersion analysis that follows advection model of “puff” of material emitted from sources, and simulates dispersion and transformation processes along the way by using the output from CALMET. The primary output from CALPUFF contains either hourly concentration or hourly deposition fluxes evaluated at the receptor locations. CALPOST is used to process these files producing tables that summarize the results of the simulation, identifying the highest and second highest 3-hours average concentrations at each receptor location. Some examples of suitable CALPUFF applications include: (1) Near-field impacts in complex flow or dispersion situations (complex terrain, stagnation, inversion, recirculation, and fumigation conditions, overwater transport and coastal conditions, light wind speed and calm wind conditions) (2) Long range transport (3) Visibility assessments and Class I area impact studies (4) Criteria pollutant modeling, including application to State Implementation Plan (SIP) development (5) Secondary pollutant formation and particulate matter modeling and (6) Buoyant area and line sources (e.g., forest fires and

ⁱPursuant section 51.162(a) of the Federal Clean Air Act [3],

- (1) international parks,
 - (2) national wilderness areas which exceed 5,000 acres in size,
 - (3) national memorial parks which exceed 5,000 acres in size, and
 - (4) national parks which exceed six thousand acres in size, and which are in existence on the date of enactment of the Clean Air Act Amendments of 1977
- shall be class I areas and may not be redesignated. All areas which were redesignated as class I under regulations promulgated before such date of enactment shall be class I areas which may be redesignated as provided in this part. The extent of the areas designated as Class I under this section shall conform to any changes in the boundaries of such areas which have occurred subsequent to the date of the enactment of the Clean Air Act Amendments of 1977, or which may occur subsequent to the date of the enactment of the Clean Air Act Amendments of 1990.
- Please note that these area is considered the area that must have the best air quality as possible.

aluminum reduction facilities).

The key aspect of the code is the employment of Gaussian puff model. But the code was not developed for the purpose of radiation dose evaluation. Although, the code is a powerful tool for pollution concentration determination, it is a non-complete tool for the purpose of radiation dose evaluation. However, this code can be useful for benchmarking purposes (for concentration part) as discussed in details in chapter 3.

MESOPUFF II

MESOPUFF II [58] is a Lagrangian variable-trajectory puff superposition model suitable for modeling the transport, diffusion, and removal of air pollutants from multiple point and area sources at transport distances beyond the range of conventional straight-line Gaussian plume models. Transport, puff growth, chemical transformation, and wet and dry deposition are accounted for in the model. MESOPUFF II is developed by the Earth Tech, Inc. MESOPUFF II is the current model identified as a refined modeling technique for long-range transport applications. It is approved for use on a case by-case basis, and applied following the guidance established by EPA [18]. Like CALPUFF, MESOPUFF is good for the calculation of pollutant concentration but can not be used for the evaluation of the resulting dose.

2.1.2 Codes for Dose Evaluation

MILDOS and MILDOS-AREA

MILDOS [32] and MILDOS-AREA [28] were developed from version IV of the Argonne National Laboratory (Argonne) computer program UDAD (Uranium Dispersion and Dosimetry). Models were included in MILDOS to consider both point sources (stacks, vents) and area sources (ore pads, tailing areas). Explicit modeling of the releases of particulates are limited to the radionuclides U-238, Th-230, Ra-226,

and Pb-210. While the MILDOS code could only be used on a mainframe computer, MILDOS-AREA was designed for use on an IBM or IBM-compatible personal computers. MILDOS-AREA is easier to use; more flexible in handling the large amount of printer output; and although slower in execution, usually exhibited a better net turnaround time than MILDOS.

The MILDOS-AREA computer code calculates the radiation doses received by individuals and the general population within an 80-km radius of an operating uranium recovery facility. The MILDOS-AREA code was designed as a primary licensing and evaluation tool to be used to provide basic input to critical licensing and regulatory decisions. It is used by the Uranium Recovery Branch staff of the U.S. Nuclear Regulatory Commission (NRC) to perform routine radiological impact and compliance evaluations for various uranium recovery operations. The code is also used by uranium recovery licensees to perform similar evaluations. The NRC is currently engaged in a project to develop a user-friendly interface (a GUI) and a user's manual for the MILDOS-AREA code. The technical work on this project is performed, under contract, by Argonne National Laboratory (ANL). At this time, a beta version of the software has been released.

The code is created for a uranium processing facility or an equivalent facility only. It is not relevant as a general purpose modeling tool for radionuclides released to the atmospheric environment.

FIIDOS

The computer code, Fallout Inhalation and Ingestion Dose to Organs (FIIDOS) [12], is used by the Nuclear Test Personnel Review program to determine the dose to internal organs from exposure to fallout under various conditions common to participants in atmospheric nuclear testing. The original version of FIIDOS, created in 1985, was

written in FORTRAN-IV for operation on a DEC PDP-11 mini-computer with problem input read from a keyboard and output directed to an off-line printer. FIIDOS calculates the radiation dose commitment to various body organs that would result from the ingestion or inhalation of the radioactive material produced by a nuclear weapon test. These organ dose calculations are based on the amount of radioactive material ingested or inhaled, the radionuclide composition of the material, and organ dose conversion factors for each radionuclide.

FIIDOS has complete characterizations of nuclear test problem, however, it does not account for particle sizes. This code does not incorporate an atmospheric transport model, by defining airborne radioactivity levels as an input parameter.

DCFPAK

The software and data package DCFPAK [16](Dose Coefficient File Package) has been developed at Oak Ridge National Laboratory to allow electronic access to the full set of dose coefficients summarized in Federal Guidance Reports 11 [2] and 12 [4] and to facilitate the use of dose coefficients for chains of radionuclides. In addition to the published dose coefficients, the DCFPAK libraries includes dose coefficients for 18 organs not addressed in Federal Guidance Report No. 11, and 17 organs not addressed in Federal Guidance Report No. 12. This package allows the consideration of particle sizes ranging from 0.1-20 μm , For particle sizes less than 0.1 μm or greater than 20 μm , DCFPAK assigns the value of fractional depositions in these regions corresponding to a particle size of 0.1 μm or 20 μm , respectively.

2.1.3 Code for Air Dispersion and Dose Evaluation

UDAD

The Uranium Dispersion and Dosimetry Model (UDAD) [24] was developed at Argonne National Laboratory, Argonne, Illinois, through the Energy Science and Technology Software Center, Oak Ridge, Tennessee. UDAD provides estimates of potential radiation exposure to individuals and to the general population in the vicinity of a uranium processing facility such as a uranium mine or mill. Only the airborne transport is considered. Exposure pathways modeled include inhalation, external irradiation from airborne and ground-deposited activity, and ingestion of foodstuffs. Individual dose commitments, population dose commitments, and environmental dose commitments are computed. The program was developed for application to uranium mining and milling operations; however, it may be applied to dispersion of any other pollutant.

The removal of radioactive particles from a contaminated area such as uranium mill tailings by wind action is estimated from theoretical and empirical wind-erosion equations as a function of the wind speed, particle size distribution and surface roughness. Atmospheric concentrations of radioactivity from specific sources are calculated by means of a dispersion, deposition, resuspension model. Source depletion as a result of deposition, fallout of the heavier particulates, and radioactive decay and ingrowth of radon daughters are included in a sector-averaged, Gaussian plume dispersion model. The average air concentration at any given receptor location is assumed to be constant during each annual release period, but also assumed to increase from year to year because of resuspension. Surface contamination is estimated by considering buildup from deposition, ingrowth of radioactive daughters, and removal by radioactive decay, weathering, and other environmental processes. Deposition velocity is estimated on the basis of particle size, density, and physical and chemical environmental conditions

which influence the behavior of the smaller particles. Calculation of the inhalation dose to an individual is based on the ICRP Task Group Lung Model (TGLM) [50]. Calculations of the dose to the bronchial epithelium of the lung from inhalation of radon and its short-lived daughters were based on a dose conversion factor from the BEIR report [10]. External radiation exposure is calculated considering direct exposure to airborne radionuclides and exposure to radiation from contaminated ground. Terrestrial food pathways include vegetation, meat, milk, poultry, and eggs. Internal dosimetry is based on ICRP recommendations, with the option of using either a single or a multiple exponential retention model. Up to five particles sizes and five size distributions may be specified for particular pollutants for use in the population dose calculations.

The UDAD code is a comprehensive tool for dose assessment. However, the code is best suited for routine releases due to the use of Gaussian Plume model which is not appropriated for releases expected in a terrorist event.

CRRIS

The Computerized Radiological Risk Investigation System for Assessing Doses and Health Risks from Atmospheric Releases of Radionuclides (CRRIS) [8] was co-developed by Oak Ridge National Laboratory, and Science Application International Corporation. CRRIS consists of eight fully integrated computer codes to calculate environmental transport of atmospheric releases of radionuclides and resulting doses and health risks to individuals or populations. Each code may be used alone for various assessment purposes. Because of its modular structure, CRRIS allows assessments to be tailored to the user's needs. Radionuclides are handled by CRRIS either in terms of the released radionuclides or the exposure radionuclides which consist of both the released nuclides and decay products that would build up during environmental transport.

CRRIS set of computational system is not to be used for short-term or accidental releases. It is appropriate only for chronic releases. The CRRIS code does not take into account the differences in the size of particulate matter transported in the air as part of its calculation.

RESRAD

The RESidual RADioactivity (RESRAD) [49] computer code was developed as a multifunctional tool to assist in developing cleanup criteria and assessing the dose or risk associated with residual radioactive material. The RESRAD-BUILD computer code is a pathway analysis model designed to evaluate the potential radiological dose incurred by an individual who works or lives in a building contaminated with radioactive material. The air quality models of RESRAD-BUILD considers the transport of radioactive dust particulates and radon progeny due to air exchange, deposition and resuspension, and radioactive decay and ingrowth. RESRAD uses a pathway analysis method in which the relation between radionuclide concentrations in soil and the dose to a member of a critical population group is expressed as a pathway sum, which is the sum of products of “pathway factors”. Pathway factors correspond to pathway segments connecting compartments in the environment between which radionuclides can be transported or radiation transmitted. The site-specific parameters considered include the size of the source area, average particle diameter, and average wind speed. Other site-specific parameters (particle density, atmospheric stability, raindrop diameter, and annual precipitation rate) were assumed to be constant. The model uses the Gaussian plume model combined with removal processes, such as dry and wet deposition of particulates.

Note that, there is no single code that satisfies the needs for radiological assessment of short-term radiological incidents under the terrorist scenario. All of these codes share incompleteness by either (1) using the Gaussian plume model as an

air dispersion model, or (2) being blind to the particle size effect, or (3) revealing incomplete modelling features for the purpose of this research.

2.2 Effect of particle size on deposition, resuspension and inhalation

Air dispersion modeling is known to be one of the most complex modeling tasks as it involves many associated parameters. Deposition both via air transport and within human body(lung) are good examples. Although there is much research conducted in this area, there is yet one important parameter which needs to be investigated further—the effect of the particle size on the transport and deposition of the airborne matter.

2.2.1 Effect of particle size on deposition in the lung(inhalation dose)

Goo and Kim [7], have conducted a thorough research regarding the deposition of particulate matter in the lungs. The deposition values were obtained by using the Monte Carlo method and were compared to those obtained by the traditional deterministic method. The results revealed that the deposition site and dose of inhaled particles in the lungs depend on particle size as shown in figure 2.1. Similar research by Rosati et. al. [22] shows a similar result; the deposition amount of particulate matter in the lungs depends on its size as seen in figure 2.2. These two studies indicated that the deposition of particulate matter which subsequently affects the inhalation doses in the lung is strongly influenced by the size of particulate matter.

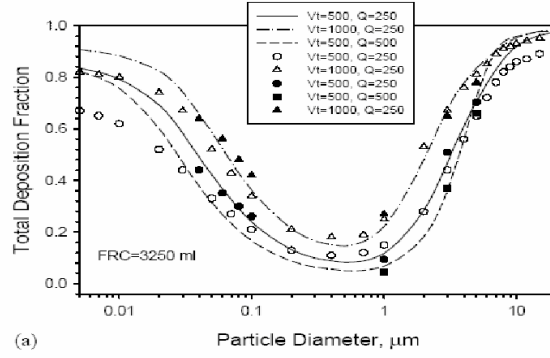


Figure 2.1: Particle Size Effect on lung deposition by Chong S. Kim

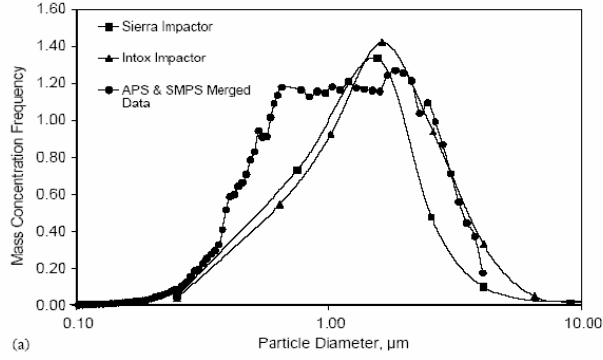


Figure 2.2: Particle Size Effect on lung deposition by Jacky A. Rosati et. al.

2.2.2 Effect of particle size on dry deposition in the air

The deposition of particulate matter via air dispersion is affected by particle size [15], [52]. In Nho-Kim et. al. research, dry deposition velocity of particles was calculated as a function of particle size and density, surface properties, and micro-meteorological conditions near the surface. Hourly deposition velocities were simulated over the year 2000 using the French operational numerical weather prediction model ARPEGE. These results were compared to the measurement results.

The average of the simulated deposition velocities using the model over the year 2000 are shown in figure 2.3. Another research was done by Zhan et.al. [52], their work

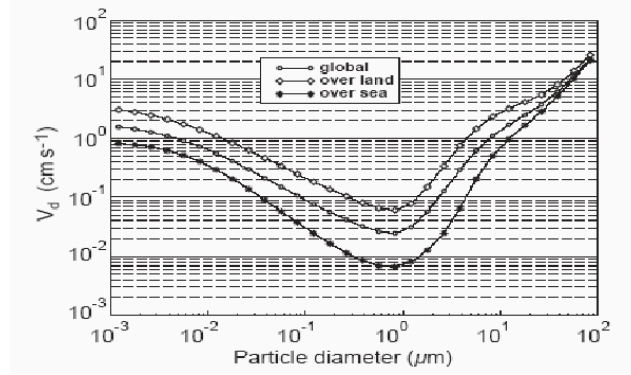


Figure 2.3: Particle Size Effect on dry deposition speed by Nho Kim

was focused on a parameterization of particle dry deposition which was developed for the Canadian Aerosol Module (CAM). This parameterization calculated particle dry deposition velocities as a function of particle size and density as well as relevant meteorological variables. It included deposition processes, such as, turbulent transfer, Brownian diffusion, impaction, interception, gravitational settling and particle rebound. A comparison of the modelled dry deposition velocities to a variety of recent measurements that have been reported in the literature, demonstrated that the their parameterization produces reasonably accurate results. The deposition velocity calculated by this model is given in figure 2.4.

The concentration of the radioactive material in the air is affected by the deposition rate which depends strongly on the particle size as well [59] as seen in figure 2.3 and 2.4. Therefore, it is desirable to take the particle size of those radioactive airborne into consideration in the modelling of dry deposition.

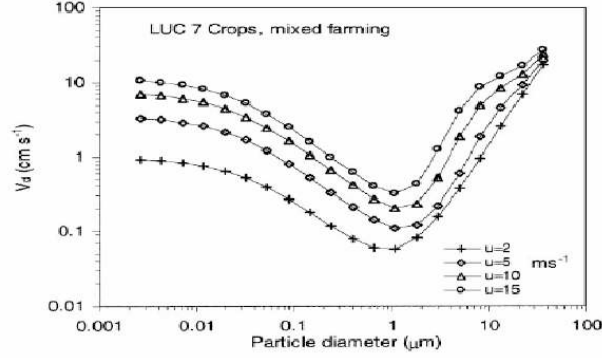


Figure 2.4: Particle Size Effect on dry deposition speed by Leiming Zhang

2.2.3 Effect of particle size on resuspension

Modelling resuspension of particles is a complex task. Incorporating the effect of particle size on the modelling particle resuspension is a challenging task and usually left out in resuspension modelling. The empirical model by Gwen A. Loosmore [5], as shown in equation 3.21, has the capability to model the resuspension factor as function of particle size although applicability of the model is limited to a limited range of particle size. Loosmore found that the conventional models derived from resuspension data for aged sources, such as former weapons test sites may not be appropriate for describing short-time resuspension: Conventional models tend to underestimate the resuspension factors for short-term periods. He reexamined historical wind tunnel data on short-term resuspension, with the goal of developing a model appropriate for numerical simulations. The empirical models were derived from these data using parameters such as friction velocity, particle diameter, surface roughness, particle density, and time. These empirical models, and the wind tunnel data, are compared quantitatively with existing conventional models from the literature. One of his empirical models that provided best fit to the experimental data set was selected (see equation 3.21) in this research.

2.3 Consideration of exposure pathways

Radiological terrorism event could involve various exposure pathways. However, as the focus of this research is to determine the effect of particle size in radiological consequence analysis of a terrorist event, consideration of inhalation of airborne particles would be the most relevant exposure pathway for this study. The inhalation doses received by individual at certain distance via air transportation due to the RDD events is the only exposure pathway considered in this research. Inhalation exposure could create significant effects on both psychological and physical health of a large number of individuals, compare to the stationary RDD case that could harm only those who are in the immediate vicinity of the event. Also people could not prevent themselves from inhaling the contaminated air once the released has occurred. Under a short-term radiological terrorist scenario, ingestion of food stuff will be an exposure pathway of minor importance as consumption of contaminated food stuff can be prevented by simple mitigation efforts.

2.4 Terrorist scenario

There are several terrorist scenarios involving the use of RDD including the one developed by the Department of Homeland Security(DHS) under the planning document [13]. The DHS scenario assumes that: (1) the device contains CsCl has been exploded and released, (2) the detonation contains 90% of the original ^{137}Cs , (3) the radioactive particles whose sizes range from range from 1 μm (micrometer) to 150 μm —the most likeliest size is 100 μm . Note that this scenario is not specifically developed for the characterization of inhalation dose but rather for the evaluation of the total dose including external exposure. However, the assumed size of the particle in the scenario is considered too big compared to the range of aerosol particle that is respirable which ranges from 0 to 10 μm . Since this research focused on an inhalation dose, this particle size assumption outside the respirable range is not appropriated

for this research. Another scenario of RDD terrorism was assumed in a study by Ring [47]. In that scenario, a single ^{137}Cs nuclide with an amount of $1.75\text{E}+03$ Ci was released due to the terrorist attack. The resulting fatal cancer risks were evaluated to be very small according to such releases. However, the study did not consider particle size effect in the assessment. A study by Angell [6] examined the release of radionuclides under a spent fuel sabotage scenario and the resulting health risk to the public. The study estimated the amount of the ^{137}Cs activity release at $2.37\text{E}+2$ Ci ($8.51\text{E}+12$ Bq) based on the review of relevant experimental studies on spent fuel sabotage[SNL].

To examine the effect of particle size on inhalation dose, the size distributions of the released aerosol particle must be known. A series of simulated spent fuel sabotage experiments [25] [26] [27], conducted at the Sandia National Laboratories, aimed to quantify the sizes of aerosol particulates produced when a high energy density device impacts surrogate material test rods and actual spent fuel test rods. The spent fuel ratio, SFR, the ratio of the aerosol particle produced from actual spent fuel explosion to the aerosol particles produced from surrogate material(non radioactive cerium Oxide CeO_2) is being evaluated as collection is being completed. With currently available data, the measured aerosol particle size distributions and the respirable fraction produced were determined for CeO_2 . The results showed that the fractions of the respirable aerosol particles ($0\text{ }\mu\text{m}$ to $10\text{ }\mu\text{m}$) were between 0.46 to 1% for CeO_2 and 1.68% for zirconium(represent cladding material) particles. The measured respirable aerosol particle size distribution is shown in picture 4.2. Given the lack of available data, the distribution obtained from the experiment was applied to this research. The result of the experiment is not complete regarding the particle size distribution for other nuclides.

Chapter 3

Computer model development

3.1 Description of RDD events

Once RDD spread out from its origin, dispersibility will depend on the physical and chemical properties of the radioactive material used in an RDD [17]. It would be difficult to disperse metallic forms while a powder could be dispersed fairly readily. Cobalt, iridium, and polonium generally exist as solid metals and would not be readily dispersible. Several of the others, including americium, californium, and plutonium, are typically oxides that could exist as a powder. Cesium is typically found as cesium chloride, which is also a powder and is quite soluble in water. Radium and strontium are used in various forms; strontium fluoride in certain sealed sources is sintered such that it is essentially insoluble and nondispersible. Even considering the forms in current sources, the specific physical and chemical characteristics of radioactive materials that could be found in an RDD is uncertain because the original material could be chemically or physically altered (weaponized) to enhance dispersal. If the dispersal method is explosion via a dirty bomb, that would also likely physically and chemically alter the materials to produce a mixture that could include oxides as well as nitrates (from the explosives) over a range of particle sizes. If the dispersal method is via the more specific situation like spent nuclear fuel sabotage, the materials themselves would be so uncertain because of the complexity of the spent fuel inventories in terms of

chemical and physical forms and the existing key nuclides and their activity levels.

Once the explosion is triggered, the material(s) with various physical/chemical form(s), particle sizes, densities would consequently spread out. Some of those with high density and big size materials would be settled near the blast site depending on the blast level. Some of the materials(with specific forms, smaller in size, and light weight) whose properties allow to disperse via wind might be able to travel and affect the far away population. It is necessary to know the amount of dispersed material to evaluate the consequence (e.g. health effects) of the event.

3.2 Description of the computer model

A new computer code was developed to analyze the consequences of a RDD sabotage event accounting for the effect of particle sizes in the release. As the focus of the analysis is on particle size, only the inhalation dose was considered as the particle size effect on external and/or ingestion dose is expected to be insignificant.

3.2.1 Key features and assumptions

The new computer model developed in this research is composed of the conceptual models that represent the physical phenomena of the interested system, i.e. atmospheric dispersion, dry deposition and resuspension of particles, inhalation exposure and resulting dose evaluation. The following lists key assumptions used in the code.

Key assumptions

Key features or assumptions made for the development of the code can be summarized as following.

- Atmospheric transport of the particle of interests are described by the Gaussian Puff model and its corresponding puff or relative diffusion parameters.
- Calculation of dose from the inhalation of particles of interest are described by the ICRP particle sized dependent dose coefficient databases [46].
- The size of particle in the given problem ranges from 0.001 to 10 μm . The particles that smaller than 0.001 μm or larger than 10.0 μm are assumed to be 0.001 μm and 10.0 μm respectively.
- Particle size-dependent(within mentioned ranges) dry deposition of particle during air transports is modeled in the code.
- Short term resuspension of the released particle during air transports is incorporated within the mentioned particle size ranges.
- Wet deposition during air transports is ignored.
- Field of terrain characteristic on the dispersion of radionuclides is not considered.
- Decay of radionuclide during air transports is ignored.

Input limitations are also shown as follow:

- Single release, and single receptor location is assumed for each calculation.
- Release of single nuclide is assumed at each code execution.

3.2.2 Modeling atmospheric dispersion

Atmospheric dispersion modeling is generally the attempt to predict the concentration of the pollutant, of which releases from a certain point, that could be

transported through air toward a certain position called receptor location. This research was focused on radioactive substances that were released instantaneously from a stationary point along with the calculation of associated inhalation doses.

There are two main mathematical models to treat this particular air dispersion problem, i.e. the Gaussian plume model and the Gaussian puff model. Application of these two models are quite different due to the differences in their conceptual bases. In this research Gaussian Puff model is utilized since it has advantages in most aspects over the other one. Only one significant drawback is that the diffusion coefficient (or dispersion coefficient) values have not been studied in as much details as in the case of Gaussian plume model. Table 3.1 illustrates some of the major characteristics of these two models, in order to summarize why puff model serves as a principle mathematical model for this research. The following sections briefly explain the Gaussian Puff models and their associated diffusion coefficients.

Table 3.1: Major characteristics comparison between puff and plume model

Aspects	Puff model	Plume model
Level of conservatism	Lower	higher
Time dependent approach	Time dependent	Steady State calculation
Dispersion coefficient	Limited data	More detail available
Distance applicability	Any	Limited

During an air dispersion process, both removal and multiplication mechanism could possibly happen. Some examples for removal processes are the deposition of the released materials to the ground, and radiological decay processes if the released materials are radioactive. An example of the multiplication mechanism is the resuspension process.

The following sections describe all of the relevant models that are used for formulating the code. These include the Gaussian puff model, the puff diffusion coefficient model, the particle deposition model, and the resuspension model.

Gaussian Puff Model

Instantaneous and short-term releases are frequently viewed as *puff* releases. Puff models was first developed by Lamb [51] and Roberts [56]. A puff model assumes that the release time and sampling times are very short compared to the travel time from the source to the receptor. Due to the limited nature of the data available to estimate the diffusion coefficients for puff diffusion, a number of puff models use the Pasquill-Gifford values which was developed for the Gaussian plume model. Since these coefficients were developed specifically for plumes, their uses in puff models are questionable. In addition, most puff models assume a Gaussian distribution of pollutant concentration within the plume. This assumption overlooks the in-puff fluctuations. Few field experiments have been conducted to determine the diffusion coefficients to support (1) the theory of puff spread and (2) the puff modelling [53].

The Gaussian puff model [33] assumes that each pollutant emission of duration Δt injects into the atmosphere a mass of ΔQ . The center of puff containing the mass ΔQ is advected according to the local time-dependent windspeed. If at a given time t , the center of the puff is located at $p(t) = (x_p, y_p, z_p)$, then the concentration due to the puff model at the receptor $r = (x_r, y_r, z_r)$ can be represented by the following formula.

$$C(x, y, z, t) = \frac{\Delta Q}{(2\pi)^{3/2} \sigma_{xp} \sigma_{yp} \sigma_{zp}} \exp\left(-\frac{(x_p - x_r)^2}{2\sigma_{xp}^2}\right) \times \exp\left(-\frac{(y_p - y_r)^2}{2\sigma_{yp}^2}\right) \times \exp\left(-\frac{(z_p - z_r)^2}{2\sigma_{zp}^2}\right) \quad (3.1)$$

Where

C : Concentration of the chemical or radionuclide in air [Ci/m³]

ΔQ : Total Effluent emission in the instantaneous released period [Ci]

σ_{xp} : Standard deviation in x direction. [m]

σ_{yp} : Standard deviation in y direction. [m]

σ_{zp} : Standard deviation in z direction. [m]

This formula is often expanded to incorporate reflection, deposition, and radioactive decay. In order to make a more realistic modelling, other physical phenomena must be also taken into account, such as changes in puff size.

The puff size is generally characterized by two quantities, the horizontal dispersion coefficient, σ_x, σ_y , and the vertical dispersion coefficient, σ_z . Most of the time the horizontal distribution is assumed to be Gaussian and symmetric, typically σ_x, σ_y is represented as just σ_h which is equal to σ_x . Both of the horizontal and vertical dispersion coefficient will be discussed in details in next section.

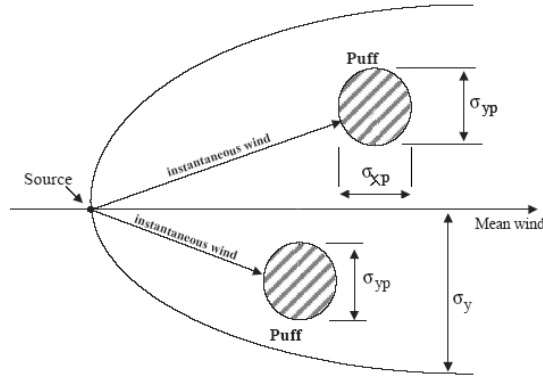


Figure 3.1: Top-Down View of Puff and Puff diffusion concepts

Diffusion Parameters

One of the most confusing aspects of atmospheric diffusion modelling is the difference between plume and puff diffusion since each model must be treated by their own set of diffusion coefficients. This puff diffusion parameters or relative diffusion parameters served as the diffusion parameter model for this research and will be discussed briefly

in the next section.

Puff diffusion coefficients:

There are few theories for Puff diffusions. The number of experimental studies performed to study puff diffusion is quite small compare to what is available for plume diffusions. The available puff diffusion experimental data related to puff diffusion coefficient is presented here(Figure 3.2) [31]. According to these data, there is no separate value of σ for different stability classes. With this lack of resolution for stability classes in the puff σ data, the plume diffusion are often used in many applications.

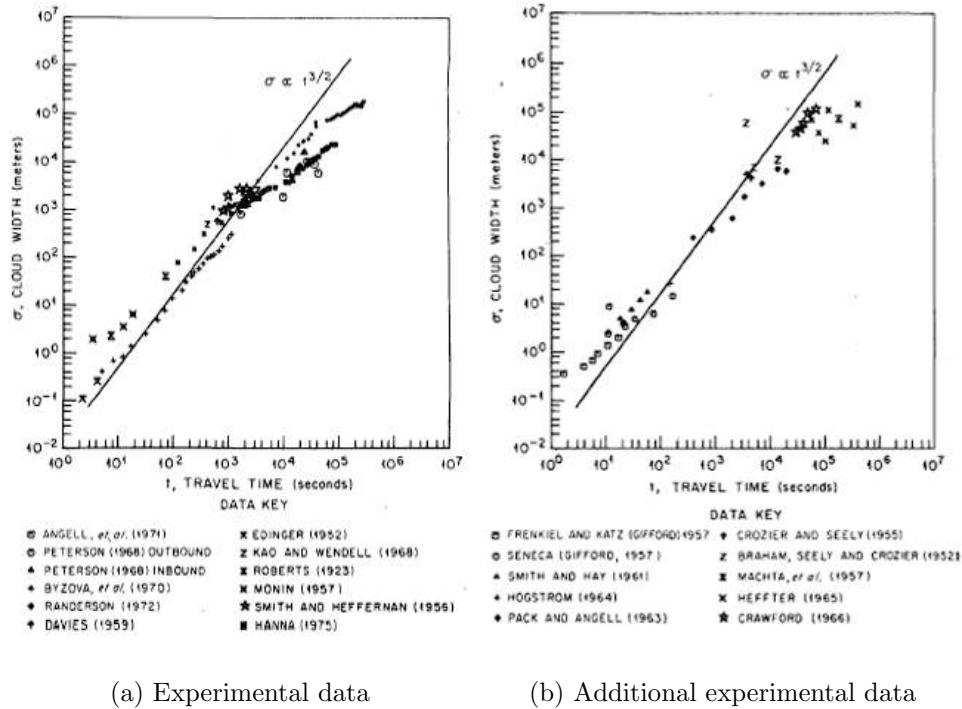


Figure 3.2: Experimental data on relative diffusion showing σ vs travel time

Some theoretical works have suggested models to represent the puff diffusion coefficients based on the existing experimental data [33]. One example of these is the Batchelor formula [65] given as following.

$$\sigma_h^2 = \epsilon t^3 \quad (t \leq 10^4 \text{ sec}) \quad (3.2)$$

where ϵ is Eddy dissipation rate which needs to be calculated for the local conditions first. This Eddy dissipation rate at midday is determined by

$$\epsilon = \frac{u_*^3}{0.5z} \quad (3.3)$$

where

u_* : friction velocity [m/sec]

z : elevation height at half height of mixing height[m].

For travel time greater than 10^4 sec,

$$\sigma_h = K t \quad (t > 10^4 \text{ sec}) \quad (3.4)$$

where K can be determined by

$$K = 100\sqrt{\epsilon} \quad (3.5)$$

A similar procedure is proposed for σ_z , except that σ_z is recommended to remain equal to $0.3 Z_i$ for all time after σ_z first reaches this value. By using these equations and the given set of sample meteorological data, the corresponding projected sigma value are calculated in this work(see Figure 3.9).

3.2.3 Modeling dry deposition

Two general types of deposition mechanisms affect the atmospheric dispersion of particles, i.e. dry deposition and wet deposition. Both processes are controlled by meteorological parameters such as wind speed, weather condition, effluent condition such as release speed, aerosol density, particle size and so on. Presently only the dry deposition was taken into consideration in this research as it is more generally applicable to particle transport. The wet deposition model is briefly discussed later in this section.

Dry Deposition Model:

Several studies [[11], [15], [52], [59], [63]] have been done to develop semi-empirical models to describe dry deposition processes of aerosol particles. The model developed by Seinfeld [61] was used in this study as a basis for the description of dry deposition. The model developed by Kim 2004 [15] was also used to describe particle size dependency in dry deposition as described below. The dry deposition velocity is described by the following equation [61];

$$V_d = \frac{1}{R_a + R_b} + V_s \quad (3.6)$$

where

V_d : Dry deposition velocity in [m/sec]

V_s : Gravitational settling velocity in [m/sec]

R_a : Aerodynamic resistance

R_b : Quasi laminar resistance

Gravitational settling velocity is determined by;

$$V_s = \frac{\rho_p d_p^2 G C_c}{18\eta} \quad \text{for } d_p \geq 1 \mu m \text{ and } Re \leq 1.0 \quad (3.7)$$

where

d_p : Particle diameter [μm]

ρ_p : Particle density for most cases in the research, 1000 kg/m^3 was used

η : Air Viscosity at 25 C, $1.86159\text{E-}5 \text{ kg/m-sec}$

G : Gravitational constant 9.8 m/sec^2

C_c : Corrector factor when particle diameter is less than $1.0 \mu\text{m}$, which is given by;

$$C_c = 1 + \frac{2\lambda}{d_p} \left[1.257 + 0.4\exp\left(-\frac{1.1d_p}{2\lambda}\right) \right] \quad (3.8)$$

R_a is expressed as [61]

$$R_a = \frac{1}{C_D + u} \quad (3.9)$$

where

C_D : Drag Coefficient

u : Wind velocity in [m/sec]

Drag coeificeint is given by [61].

$$C_D = \begin{cases} \frac{24}{Re} & \text{where } Re < 0.1 \\ \frac{24}{Re} \left[1 + \frac{3}{16}Re + \frac{9}{160}Re^2 \ln 2Re \right] & \text{where } 0.1 < Re < 2 \\ \frac{24}{Re} [1 + 0.15Re^{0.687}] & \text{where } 2 < Re < 500 \\ 0.44 & \text{where } 500 < Re < 2 \times 10^5 \end{cases}$$

where Re is the Reynolds number, which is obtained by

$$Re = \frac{ud_p}{\nu} \quad (3.10)$$

where

u : Wind velocity (m/sec)

d_p : Particle diameter (m)

ν : Kinematic viscosity of air which equal to η/ρ_p

when

η : Air viscosity (kg/m-sec)

ρ_p : Particle density (kg/m³)

Thus, Re is the proportionally ratio of inertial forces to frictional forces acting on each element of the fluid

R_b is expressed as [61] Kim [15],

$$R_b = \frac{1}{u_*(E_{br} + E_{im} + E_{in})} \quad (3.11)$$

where

u_* : Friction velocity [m/s]

E_{br} : Brownian diffusion efficiency

E_{im} : Inertial impaction efficiency

E_{in} : Interception efficiency

E_{br} is a function of Schmidt number (Sc), which is the ratio of the kinematic viscosity, ν of air to the Brownian diffusivity, D :

$$E_{br} = Sc^{-\gamma} \quad (3.12)$$

and

$$Sc = \nu_{air}/D \quad (3.13)$$

$$D = \frac{kTC_s}{3\pi\eta d_p} \quad (3.14)$$

where

γ : Empirical parameter, in this research $\frac{2}{3}$ was used.

k : Gas constant

T : Temperature

C_s : Stokes-Einstien relation Correction factor which can be determined by

$$C_s = 1 + 1.657\left(\frac{2\lambda}{d_p}\right) \quad (3.15)$$

where

λ : Particle mean free path [μm]

An inertial impaction efficiency, E_{im} , is a function of Stokes number(St), which is defined as the ratio of the stopping distance of a particle to a characteristic dimension of the air. Alpha and Beta are empirical parameters, given as 0.8 and 2 respectively according to Kim [15].

$$E_{im} = \left(\frac{St}{St + \alpha} \right)^\beta \quad (3.16)$$

$$St = \frac{d_p^2 \rho_p C_s u_0}{18\eta L} \quad (3.17)$$

where

C_s : Stokes-Einstien relation Correction factor

u_0 : Wind velocity [m/sec]

L : Characteristic length [μm] equal to Characteristic radius of large collectors (1000 μm for forest and 500 μm for vegetation)

An interception efficiency, E_{in} , is a function of the ratio of viscous to total drag coefficient, particle size and the characteristic of collector materials, as expressed by;

$$E_{in} = \frac{C_v}{C_D} \left[F\left(\frac{d_p}{d_p + \hat{a}}\right) + (1 - F)\left(\frac{d_p}{d_p + L}\right) \right] \quad (3.18)$$

where

$\frac{C_v}{C_D}$: Ratio of viscous to total drag coefficient, use 0.3 according to Kim [15]

F : Fraction of total interception by collector (0.01)

\hat{a} : Characteristic width of vegetative hairs (10 μm)

L : Characteristic radius of large collectors (1000 μm for forest and 500 μm for vegetation)

Determination of friction velocity:

The friction velocity (u_*) is a function of weather condition and can be approximated by the following equation, if zero average velocity at the ground level can be assumed [61].

$$\frac{u_*}{\kappa \bar{u}_1(x_3)} = \begin{cases} \left[\ln \left(\frac{x_{3r}}{z_0} \right) + \frac{4.7}{L} (x_{3r} - z_0) \right]^{-1} & \text{for stable condition} \\ \left[\ln \left(\frac{x_{3r}}{z_0} \right) \right]^{-1} & \text{for neutral condition} \\ \left[\ln \left(\frac{x_{3r}}{z_0} \right) + \ln \left[\frac{(\eta_0^2 + 1)(\eta_r + 1)^2}{(\eta_r^2 + 1)(\eta_0 + 1)^2} \right] + 2(\tan^{-1} \eta_r - \tan^{-1} \eta_0) \right]^{-1} & \text{for unstable condition} \end{cases}$$

where

$$\eta_r = \left[1 - 15 \frac{x_{3r}}{L} \right]^{1/4} \quad \eta_0 = \left[1 - 15 \frac{z_0}{L} \right]^{1/4}$$

Wet Deposition:

Wet deposition is typically modelled by using a washout coefficient Λ [19]. The mass of pollutant(W) incorporated into rainfall is:

$$W = \Lambda C \tag{3.19}$$

where C is the local airborne concentration. Λ is washout coefficient which is dependent on a large number of parameters, including the rainfall rate, raindrop and aerosol size distributions and concentrations in air.

The wet deposition is also particle size dependent and increases with the precipitation intensity. Although there are several models available to evaluate the wet deposition [[19], [60], [62]], none of them estimates the wet deposition velocity as a function of airborne particle size. Given that, there are many important parameters that control the wet deposition velocity, particle size effect appears to be insignificant in wet deposition.

3.2.4 Modeling mixing height

Mixing height is a crucial input parameter in air pollution models as it restricts the dispersion of pollutants and it is not directly measurable. Technically it is not a random variable. There are two basic possibilities for the practical determination of the mixing height. It can be obtained from profile measurements, either in-situ (radiosonde, tethered sonde, tower) or by remote sounding (sodar, clear-air radar, lidar). The other possibility is to use parameterizations or simple models with only a few measured parameters as input.

A preliminary sensitivity analysis indicated that mixing height was the one of the top 5 parameters that contributed to the changes in the pollutant concentration calculated atmospheric parameters.

To represent the changes in mixing height with respect to the changes in windspeed and weather stability, a simple model was developed. A more refined model could be developed in the future as additional information becomes available. The calculated mixing heights by Suhail M. Khan and Rod W. Simpson model [64] were used to develop a model for mixing height as a function of windspeed and weather stability. Because of the lack of data for class A and B, current model is applicable to only class C, D and E. These fitted equations are :

For Class C (Slightly unstable)

$$Y = 1088.1 * \ln(X) - 218.73 \text{ with } R^2 = 0.9369$$

For Class D (Neutral condition)

$$Y = 779.87 * \ln(X) - 142.49 \text{ with } R^2 = 0.902$$

For Class E (Slightly Stable condition)

$$Y = 364.39 * \ln(X) - 201.53 \text{ with } R^2 = 0.7079$$

where Y and X represent mixing height and windspeed respectively. Although these model are not applicable to class A and B, they can be used for conservative predictions.

3.2.5 Modeling of other atmospheric transport parameters

Modeling particle size

For general airborne particulate matters, particle size distributions have been characterized. Particulate Matter(PM) in the size range of $0.02 - 0.7 \mu\text{m}$ in a general atmospheric environment was found by Wang and Flagan [57]. Aerodynamic size distribution of PM over the range $0.5 - 20 \mu\text{m}$ was also observed by Baron [55]. The distribution depends on many parameters, such as the component in the air, geographic of the site where size was being measured.

Modeling roughness length

The roughness length is related to the roughness characteristics of the terrain. Under near-neutral conditions and with a homogeneous distribution of obstacles. [61]. MacKinnona et. al. [20] developed models to estimate roughness length of area in the central Mojave Desert, California. To calculate roughness lengths from these models,

measurements were made at 11 sites within the monitored area to characterize the surface roughness element. In order to perform the uncertainty analysis regarding roughness, The range of data from all of the sites in their research(0.0053 to 10.78 cm) were obtained; uniform distribution can be assumed to represent this parameter.

Modeling Gamma(γ)

Gamma is empirical parameter used in the Kim [15] model (see equation 3.12). According to the model, this parameter is ranging from 1/2 for sea and 2/3 for vegetated surface. Given no details regarding this parameter distribution at this point, uniform distribution can be assumed for this parameter.

Modeling mean free path(λ)

Mean free path typically depends on many physical parameters such as thermal conductivity, molecular diffusivity. According to Seinfeld [61], this value is ranging from 0.0651 to 0.2 μm .

Modeling air viscosity

Air viscosity is function of other parameters such as temperature. This parameter again was treated as uniform distribution within the range of 1.788097E-5 to 1.834170E-5 kg/m-secⁱ.

ⁱThese viscosity data were calculated using the website “<http://www.lmnoeng.com/Flow/GasViscosity.htm>” based on North Carolina historical temperature record(spring to summer average) from NC weather data“<http://www.cityrating.com/citytemperature.asp?City=Raleigh+-+Durham>”.

Modeling air density

Air density distribution was also obtained the same way as air viscosity. By taking the spring to summer temperature data from the same page as previously taken, and perform the calculation using the density calculatorⁱⁱ. The estimated air density were ranging from 1.158135 to 1.239829 kg/m³.

Modeling characteristic length

According to Kim [15], characteristic length [μm] equal to Characteristic radius of large collectors which ranges from 500 μm for vegetation and 1000 μm for forest. Base on these characteristics, it is reasonable to assume uniform distribution of 750 μm average.

3.2.6 Modeling resuspension

Resuspension Model

A resuspension can be defined as the detachment of a particle from surface and its transport away from surface. It may occur as a result of air jets, or due to mechanical force such as traffic disturbance, the impaction of other particles, or electrostatic forces [34]. Meteorological conditions would also affect resuspension of particles. There are two approaches to describe the resuspension, i.e. the resuspension factor approach and the resuspension rate approach. The concept of resuspension factor is given by;

$$K(L^{-1}) = \frac{\text{concentration of species in air } (BqL^{-1})}{\text{surface concentration of species } (BqL^{-1})} \quad (3.20)$$

ⁱⁱThese density data were calculated using the website "<http://www.denysschen.com/catalogue/density.asp>"

where

K : Resuspension Factor [m^{-1}]

L : Represents Length [m]

It is difficult to accurately characterize the resuspension factor. For regions of relatively uniform deposits it is useful to measure the value of resuspension. Garland and Cambray [54] reported resuspension factors around the Chernobyl site, based on the mean atmospheric concentrations for the period July 1986 to June 1987. The result covered a range from 6×10^{-10} to $1 \times 10^{-6} \text{ m}^{-1}$.

Many researchers attempted to model the resuspension factor [[1], [5], [29], [30], [54]]. The empirical resuspension model by Gwen A. Loosmore [5] as shown in equation 3.21 was adopted in this research since it has the capability to model the resuspension factor as function of particle diameter. This model was developed for short term resuspension phenomena. Loosmore's model is:

$$\Lambda = 0.42 \times \frac{u_*^{2.13} d_p^{0.17}}{t^{0.92} z_0^{0.32} \rho_p^{0.76}} [s^{-1}] \quad (3.21)$$

where

Λ : Resuspension rate [s^{-1}]

u_* : Friction velocity from 0.1 to 1.4 m/s

d_p : Particle size (μm)

ρ_p : Particle density ($\frac{\text{kg}}{\text{m}^3}$)

z_0 : Aerodynamic roughness height 0.0003 and 0.005 for Nicholson data set,

In general, the resuspension rate increases with frictional velocity and particle diameter (within the range of interest) and decreases with time, surface roughness, and particle density. The interchange between resuspension factor and resuspension rate could be characterized. The following equation by Loosmore [5] describes the

resuspension rate in term of resuspension factor.

$$\Lambda = u_* \kappa p K \quad (3.22)$$

where

Λ : Resuspension rate [s^{-1}]

K : Resuspension rate [m^{-1}]

u_* : Friction velocity from 0.1 to 1.4 m/s [19]

p : Constant reported to fall between 0.25 and 0.35 by Anspaugh et al. [19]

κ : Von Kermann constant

3.2.7 Inhalation dose evaluation

Dose Model

In order to protect humans from radiation, determination of radiation dose is needed. Regardless of how radiation enters humans body, human tissue and organs would be irradiated. How much these irradiated tissues would be affected is determined by such factors as dose, dose rate, radiation types, sensitivity of tissues and organs, duration of exposure and etc. Dose represents the amount of radiation absorbed by the tissues expressed in term of, absorbed dose $D_{T,R}$, equivalent dose H_T , effective dose E , or committed effective dose depending on the applications.

The equivalent dose is the quantity that takes into account the different type of radiation by using the radiation weighting factors. The equivalent dose in tissue or organ T is given by the the following equation:

$$H_T = \sum_R w_R D_{T,R} \quad (3.23)$$

where

H_T : Equivalent dose [Sv or rem]

$D_{T,R}$: Absorbed dose averages over tissue or organ [Sv or rem], due to radiation R

w_R : Radiation weighting factor.

For beta and gamma radiations, the equivalent dose is equal to the absorbed dose whereas for neutron and alpha radiation, the equivalent dose is more than absorbed dose by the factor of their radiation weighting factor which are shown in table 3.2 below:

Table 3.2: The ICRP radiation weighting factors(Data were obtained from ICRP Publication No.60 [37])

Type and Energy of radiation	Radiation weighting factor, w_R
Photons, all energies	1.00
Electrons and muons, all energies	1.00
Neutrons (depend on energy)	5 - 20
Protons, other than recoil protons, energy < 2MeV	5.00
Alpha-particles, fission fragments, heavy nuclei	20.00

To consider the fact that tissues and organs have different radiation sensitivity, the concept of effective dose has been introduced as followed:

$$E = \sum_T w_T H_T \quad (3.24)$$

where

E : Effective dose [Sv or rem]

w_T : Tissue weighting factor.

The tissue weighting factors recommended by ICRP publication 60 are given by table 3.3. Once intake of radionuclides take place, some radionuclides stay in humans body for many years, which will result in continuous irradiation of tissues and organs over certain periods of time. For these radionuclides, the total dose will depend upon radiological half-life, biological half-life of the radionuclide, and their distribution

throughout the humans body. The total effective dose received over life time, taken to be 70 years for infant and 50 years for adults, is called the *committed effective dose*.

The International Commission on Radiation Protection (ICRP) has published values for the committed dose following intake of 1 Bq of different radionuclides via ingestion and inhalation [[42], [44], [46]], that is so called dose coefficient (in the unit of Sv per Bq). Use of dose coefficients facilitates the determination of human dose from the ingestion or inhalation of radionuclides.

Inhalation dose, which is the main focus of this study, can be determined by the following formula:

$$Dose_{inh} = Concentration \times Breathing\ rate \times Time \times Dose\ Coefficient_{inh}$$

Table 3.3: The ICRP tissue weighting factors

Tissue or organs	w_T
Gonads	0.20
Red Bone Marrow	0.12
Colon	0.12
Lung	0.12
Stomach	0.12
Bladder	0.05
Breast	0.05
Liver	0.05
Eesophagus	0.05
Thyroid	0.05
Skin	0.01
Bone Surface	0.01
Remainder	0.05

Dose Coefficient Determination

ICRP Dose Coefficient Database [46] serves as the primary source of dose factor data. These database contains the value of committed equivalent dose per unit

intake (dose coefficients) to various tissues and committed effective doses per unit intake (dose coefficients) to the whole body. Results are given for both workers and members of the public. These results are the same as the latest ICRP advice given in Publications 68 (workers) [42] and 72 (members of the public) [44]. The database itself contains dose coefficient for all same nuclides listed in both Publications. Radiation decay data are taken from ICRP Publication 38 [36].

This database also includes dose coefficients for ten aerosol sizes (0.001, 0.003, 0.01, 0.03, 0.1, 0.3, 1.0, 3.0, 5.0, and 10.0 μm) and for ten different times after intake (1 day, 7 days, 30 days, 1 years, 5 years, 10 years, 20 years, 30 years, 45 years, and to age 70 years). This database contains a compilation of age-dependent committed effective dose coefficients for members of the public from intakes by ingestion and inhalation of radioisotopes of the 31 elements covered by ICRP Publications 56 [38], 67 [39], 69 [41] and 71 [43]. It also gives committed effective dose coefficients for radionuclides of the additional 60 elements for which dose coefficients are given for workers in ICRP Publication 68 [42]. There are 6 different age ranges are accounted for;

- 3 months : represents age ranged between 0 to 1 years
- 1 years : represents age ranged between 1 to 2 years
- 5 years : represents age ranged between 2 to 7 years
- 10 years : represents age ranged between 7 to 12 years
- 15 years : represents age ranged between 12 to 17 years
- adult : represents age ranged more 17 years

The new humans respiratory tract model used in the database constitutes an updating of the model used in Publication 30 [35] for workers. It is described in full in Publication 66 [40]. A summary description of the model is given in the next section.

Base on these ICRP data, the effective inhalation dose coefficient for adult public

at a given time(after intake) to age 70 years with 10 different particle sizes became available. A computer code uses these dose coefficient as input to calculate the committed effective dose equivalent to member of public adult. This software is capable of analyzing various factors affecting human dose, such as particle sizes, times since after initial intake of radioactive substances either inhalation or ingestion. For each selected nuclide run, only the desired calculation results, i.e. the effective inhalation dose coefficient for adult public at time(after intake) to age 70 years with 10 different particle sizes, were obtained and subsequently used. These calculation results are shown in the Figure 3.3.

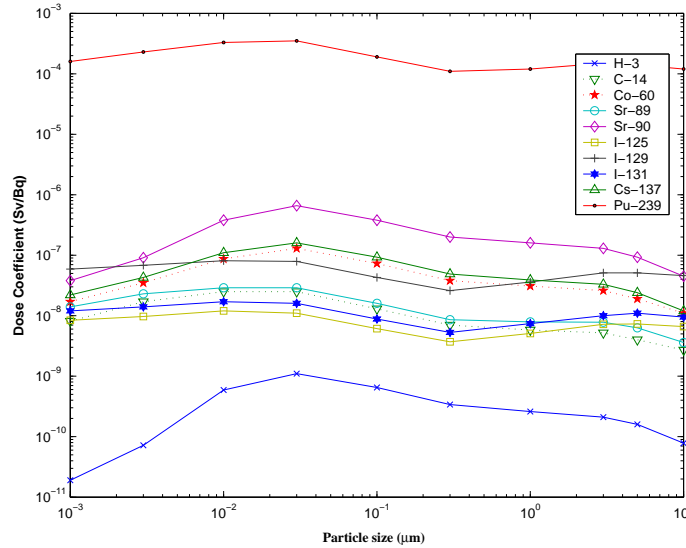


Figure 3.3: Dose coefficient for given nuclide as function of particle size obtained by PIDA

As seen in Figure 3.3, the particle size of $0.3 \mu\text{m}$ will result in highest effective inhalation dose to adult public compared to other particle sizes of the same nuclide. This implies that $0.3 \mu\text{m}$ is likely to travel deepest into the human respiratory system and get deposited.

3.3 General Code Information

3.3.1 Inputs

In this research a new computer model was developed to perform “Puff Particle size Dependent Inhalation Dose Assessment with Resuspension and Deposition. The model was named “PIDA”. The main program of the code will act as user interface to input information by calling responsible subroutines when needed, and performing calculations using the given information. The main objective of this code is to calculate the particle size dependent inhalation dose at given location when radioactive substances are released instantaneously to the atmosphere. The code combines several submodels in atmospheric transport analysis, inhalation dose calculation, dry deposition calculation and, resuspension modelling. Figure 3.4 gives a simplified diagram for this model.

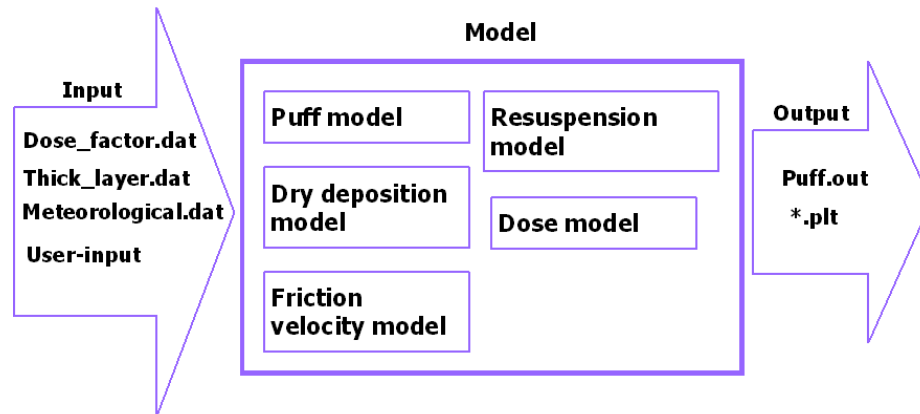


Figure 3.4: Simplified diagram of PIDA model

Uncertainty analysis capability was also added to the code with the use of an add-in software Crystal Ball [14]. The Crystal Ball software allows users to define the uncertainty of input parameters as distribution models through the “Assumption cells”.

Users can define as many assumption cells as desired. With the help of a built-in visual basic in excel, the variation of such input parameters will be written into a text file format named “main.inp”. After the uncertainty analysis capability of the “PIDA” code (named as “PIDA_UNCER”) is executed, the code will read the data from the file, “main.inp”. Once the code execution is completed, a series of output files will be created and be imported back into the main input/output page(on excel sheet). Finally, the code processes the output to report the results. This includes statistical data analysis of the output parameter of interest. Results, such as mean, standard deviation, kurtosis of an output result can be given. Figure 3.5 gives a simplified diagram of the computer model including the list of the required input and other data sets.

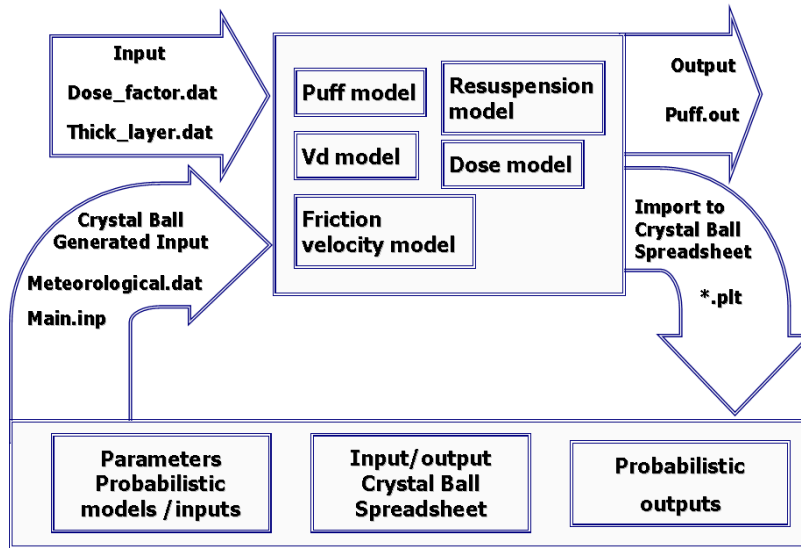


Figure 3.5: Simplified diagram of PIDA_UNCER

As seen in the previous section, there are numbers of parameters involved in the processes. Table 3.4 shows the types and variable names of input and output parameters used in the code. As can be seen from the table, some parameters values

were internally calculated, and some were taken either as fixed constant or provided by the user as an input.

Execution of the code can be performed in two different ways, i.e. a deterministic or a stochastic calculation. Whenever deterministic calculation is desired, all input parameters with probability distribution model will be reduced to a single value (mostly as their average values). When stochastic calculation is necessary, the input parameters (as seen in the 4th column in table 3.4) will be created as probability distribution models to characterize the uncertainty of the results.

“PIDA” Input parameters

The user-provided input data include the amount of radioactivity released, the receptor location distance from the release location, the nuclide of concern released as well as its particle sizes. The command prompt asks the user to enter necessary information for these input while running the code. Besides the user-provided input information, there are three different input data files; meteorological data file, thickness layer data file and dose conversion factor data file. These filesⁱⁱⁱ are required to be present in the same working directory with the executable version of the code.

The meteorological data file includes the hourly windspeed and stability class data. This hourly resolution of data allows the validation of the code against the standard gaussian puff model such as CALPUFF. Additional weather data could be added to this data file. The thickness layer data file is composed of the empirical parameters for each weather stability class to calculate the friction velocity. The dose coefficient data file includes the dose coefficients for 10 major radionuclides.

ⁱⁱⁱPlease see appendix A.2 for the details

3.3.2 Output

There are two types of output files generated: (1) the “0puff1.out” file and “0puff2.out” contain both input and output data for the air dispersion analysis and the inhalation dose analysis, respectively (2) a series of “*.plt” files are created primarily for plotting purposes.

3.4 Model Validation

3.4.1 Puff model

The CALPUFF code was used in this study to benchmark the newly developed code with respect to atmospheric transport analysis using the Gaussian puff model. CALPUFF is far more elaborate and sophisticated than “PIDA” in all aspects (assumptions, mathematical models, model dimension etc). CALPUFF is a much more complex code capable of processing 3-dimensional meteorological data and reversible wind directions, whereas PIDA can handle only one dimensional windspeed data and non-reversible wind directions. The CALPUFF [45] code is composed of CALMET, CALPUFF and CALPOST. CALMET is a meteorological gridded modeling domain. Associated parameters such as mixing height, surface characteristics, and dispersion properties are included in the file produced by CALMET. CALPUFF is a transport and dispersion model that advects the puff of the material emitted from the sources, and simulates dispersion and transformation processes along the way by using the output from CALMET. The primary output from CALPUFF is the hourly concentration or hourly deposition fluxes of the contaminant evaluated at the receptor location. CALPOST is used to process these files producing tables that summarize the results of the simulation. CALPOST also identifies the highest and second highest 3-hours average concentrations at each receptor location. In order to benchmark

the “PIDA” code with CALPUFF, two separated case problems (with and without particle size effects) were analyzed. The two cases were based on using either gas species or PM10^{iv} species respectively. The following lists show the input information for the two cases:

Input parameters comparison

PIDA Input

CASE I and II

- Initial ground level activity: 3.13E+06 Ci
- Distant: 100 m NW from the released location
- Nuclide: Cs-137 (SA= 86.87 Ci/g)
- Calculation Period: 24 hrs, start Apr. 15, 1990 to Apr. 16, 1990^v
- Particle sizes analyzed: Case I gas, Case II Particle

CALPUFF Input

Case I and II

- location of release: PULSTAR reator, NC state university
- Rate: 10 g/sec for 1 hour released from ground^{vi}
- Initial wind speed: 10 m/sec
- Area released: Stack diameter 1 m
- Distant: 100 m and 10 km in radius around released location
- Calculation Period: 24 hrs, start Apr. 15, 1990 to Apr. 16, 1990^{vii}
- Released species: Case I gas, Case II PM10 particulates(10 μ m and less)

^{iv}PM10 means particulate matter that has particle size of 10 micron and less

^vHourly data were obtained from <http://daq.state.nc.us/permits/mets/metdata.shtml>

^{vi}To convert 10 g/sec in to Ci/ m^3 , specific activity for nuclide Cs-137 of 86.87 Ci/g was used in order to achieve the task. Hence 10 (g/sec) * 1 (hr) * 3600 (sec/hr) * 86.87 (Ci/g) = 3.13E+06 Ci.

^{vii}CALPUFF has built-in hourly meteorological database through that CALMET processing

Output Parameters comparison

The following section illustrates the outputs from both codes. For Case I (gas species-without deposition effects), the calculated concentrations were $6.1594\text{E}+03$ mg/m^3 and $1.0298\text{E}+04$ mg/m^3 for CALPUFF and PIDA, respectively. For case II (PM10 species-with deposition effect), the calculated concentrations were $6.1594\text{E}+03$ mg/m^3 and $1.0058\text{E}+04$ mg/m^3 for CALPUFF and PIDA, respectively. These comparison results are also shown in Figure 3.6. Another comparison as the distance to and time of the maximum concentration between the two codes were obtained as shown in Figures 3.7 and 3.8 respectively. The observed differences are not surprising considering that CALPUFF is a 3-dimensional complex code whereas PIDA is a simplified 1 dimensional code. Summary of these outputs are shown in the next section.

CALPUFF Output

Results of maximum concentration of pollutant in the air between CALPUFF and PIDA were compared as a way of benchmarking the PIDA code. Table 3.5 shows this comparison.

3.4.2 Calculation of Puff dispersion coefficient

To examine the accuracy of the puff dispersion coefficients calculated from PIDA code, the results from PIDA were compared with experimental data obtained from Hanna [33]. Results of the comparison of horizontal dispersion coefficient are shown in Figure 3.9.

According to Figure 3.9, the calculated values of puff dispersion coefficient were higher than the experimental data. After the travel time of about 600 sec, the calculated sigma values begin to depart from the experimental data. Once the puff travel time reaches 10^4 sec, the difference between calculated and experimental one

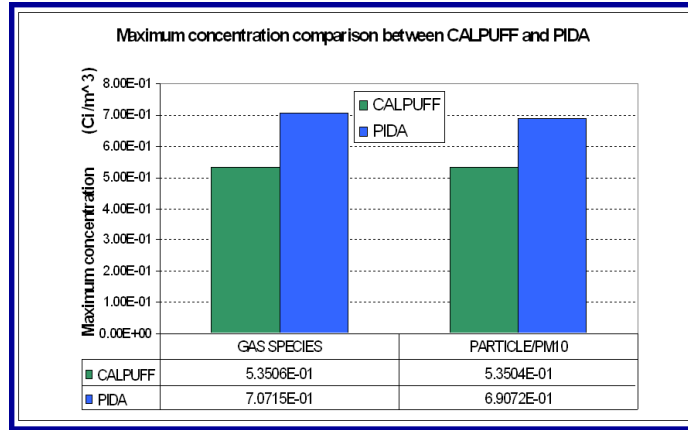


Figure 3.6: Maximum concentration comparison from CALPUFF and PIDA (gas vs. 1 mm particle)

remained almost constant. This means that the calculated puff was predicted to grow faster than what was measured. This difference may be mainly due to the differences in the values of mixing height between what was estimated for the calculation for the given meteorological conditions and the conditions for the experiment.

3.4.3 Calculation of Dry deposition velocity

For the validation of dry deposition model of the PIDA code, the results of dry deposition velocity calculated by the PIDA code were compared with the results predicted by using the model Kim et. al. (see section 3.2.3) [15]. Figure 3.10 shows the comparison. The results are in good agreement in terms of the overall trend. One noticeable difference between the two calculations is that PIDA has predicted lower deposition velocity for most cases compare to the Kim model especially when particle size is between $0.1 \mu\text{m}$ to $5 \mu\text{m}$ range. This implies that the concentrations of radioactive airborne materials calculated by PIDA will be greater than the predicted by the Kim model [15] at the same location. The reason why PIDA has predicted

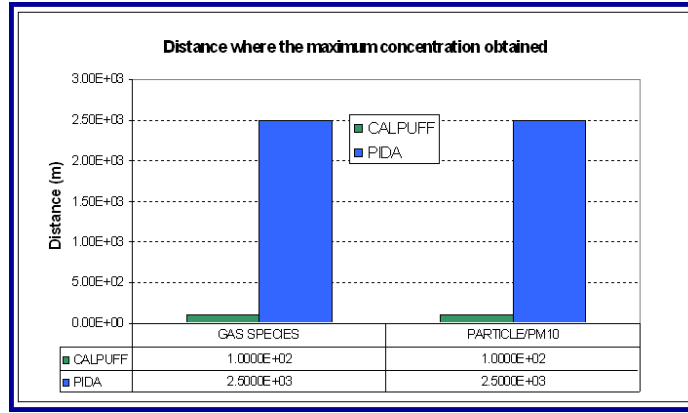


Figure 3.7: Distance where maximum concentration was obtained (gas vs. 1 mm particle)

lower deposition velocity is not quite clear. However, the fact that the number of input parameters used by two predicted models are not the same (apparently less with PIDA) would have contributed to the difference. Besides, the Kim model, plots the average of deposition velocity over one year period whereas PIDA showed 1 day average results. The meteorological data were also obtained by different sources: PIDA data were based on the conditions here in North Carolina whereas the Kim model used European databases. This issue will be further addressed through uncertainty analysis (section 3.5).

3.4.4 Calculation of Resuspension factor

Calculation results of resuspension factor obtained from PIDA were compared with the results from the semi-empirical model by Loosmore [5]. The comparison is shown in Figure 3.11.

As seen in Figure 3.11, the results from PIDA were in good agreement with Loosmore model results. According to this calculation, the resuspension factor increased with particle diameter (within the range of interest between 0.1 and 10 μm) and decreased

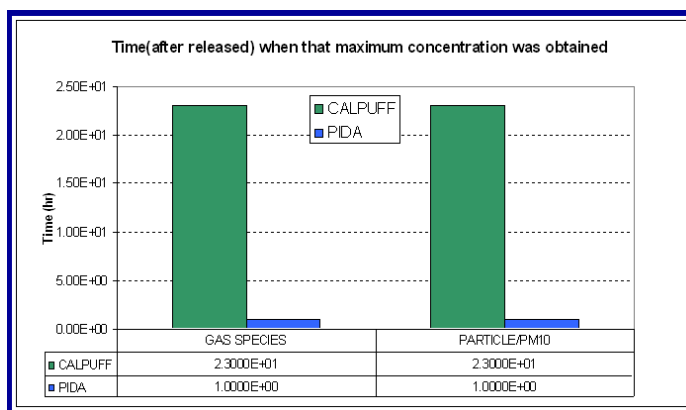


Figure 3.8: Time where maximum concentration was obtained (gas vs. 1 mm particle)

with time.

3.5 Capability for uncertainty analysis

With the emphasis on analyzing the effect of particle size in atmospheric transport and dose assessment in this research, understanding how the uncertainty in particle size distributions affect the overall dose assessment will be particularly important. Uncertainty analysis is introduced by incorporating some of the input parameters as random variables allowing their values vary within their possible ranges. The corresponding uncertainty in the output was determined accordingly. This analysis was implemented within the framework of the Monte Carlo method by using the Crystal Ball software.

Monte Carlo(MC) simulation was named after the city Monte Carlo, Monaco, where the primary attractions are casinos containing games of chance that exhibit random behavior. The random behavior in games of chance is similar to how Monte Carlo simulation selects variable values at random to simulate a model. It's the same with

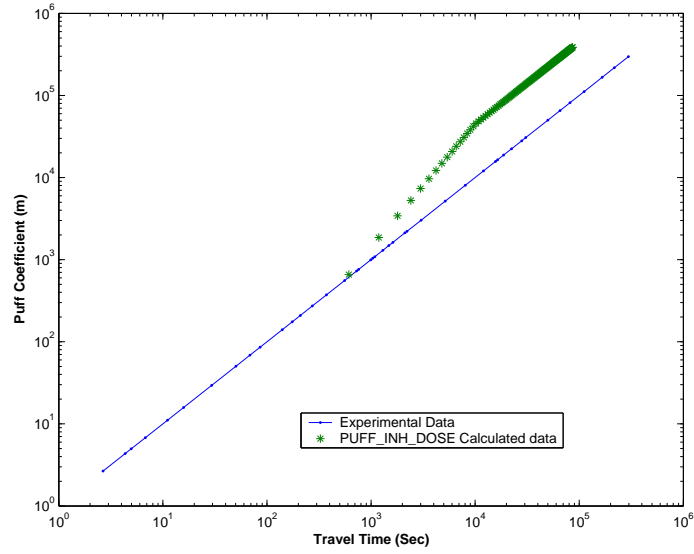


Figure 3.9: Horizontal dispersion coefficient comparison between Code and Hanna et al. data set

the variables that have a known range of values but are uncertain at any particular time of an event. For MC simulation each uncertain variable (one that has a range of possible values) is given a defined probability distribution. The type of distribution selected is based on the understanding of the characteristics of the random variable.

Results of parameter uncertainty analysis provide insights as to how the uncertainty of each important parameter affects the resulting dose estimate and the associated uncertainty. As mentioned earlier, Crystal Ball version 5.0 was employed to perform uncertainty and sensitivity analysis in this research.

Distribution types to represent the random behavior of the variables can include, for example, normal distribution, triangle distribution, uniform distribution, lognormal distribution and so on. For MC simulations, the user runs the computer model with multiple scenarios of input value combinations by repeatedly sampling values

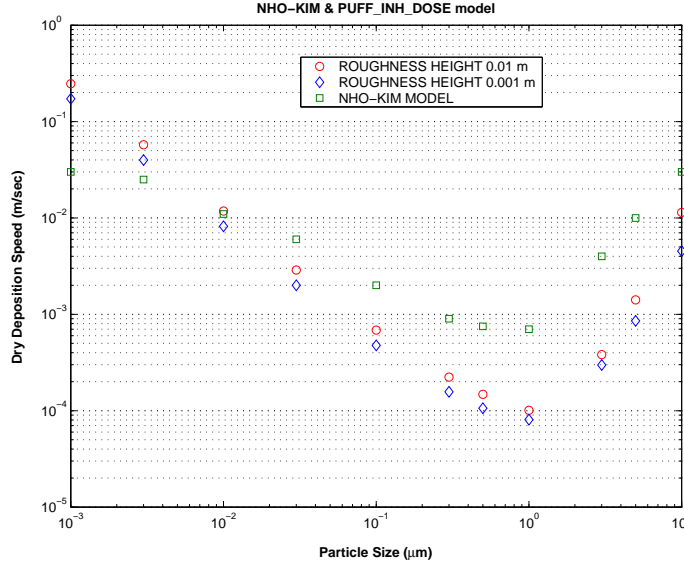


Figure 3.10: Dry deposition velocity comparison between PIDA and Kim model

from the probability distributions for the uncertain variable. Crystal Ball simulations can consist of as many trials (or scenarios) as desired. In this research, 100,000 trials were selected. Description of the probabilistic models for the selected input parameter as random parameters are summarized in table 3.6. To support the use of Crystal Ball in conjunction with the PIDA code an interface module was developed and implemented.

Table 3.6 shows the summary of all input parameters and each designated probabilistic model that used in the calculation of inhalation dose for the terrorist scenario.

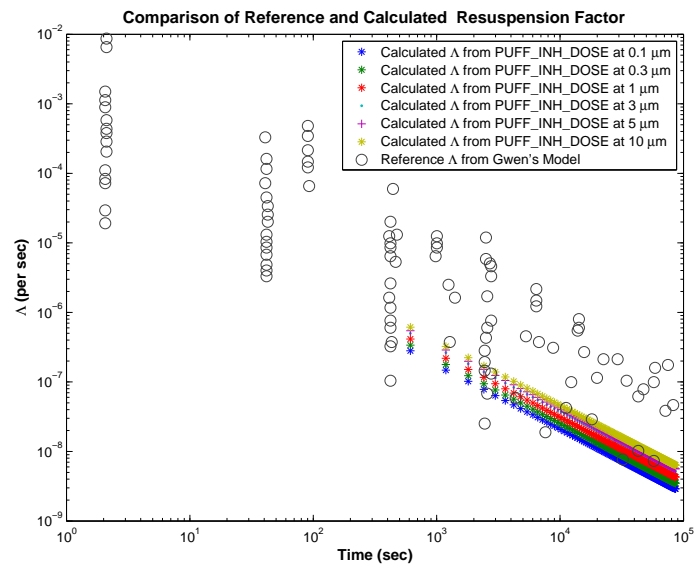


Figure 3.11: Resuspension factor comparison between PIDA and Loosmore model

Table 3.4: List of input and output parameters found in the code

Parameters	Unit	Variable Name ^a	Parameter Type	Distribution model
Released Activity	Curie(Ci)	INI_CONC	input	N/A
Distance	km	DISTANCE	input	N/A
Nuclide Name	-	NUCLIDE	input	N/A
Particle size	μm	SIZE	input	Yes
Wind speed	m/sec	WINDSPEED	input	Yes
Weather stability	-	CLASS	input	N/A
Breathing rates	m^3/hr	BREATH_RATE	input	Yes
Air density	kg/m^3	RHO_AIR	input	Yes
Particle density	kg/m^3	RHO_PAR	input	Yes
Roughness	m	ROUGHNESS	input	Yes
Breathing rates	m^3/hr	BREATH_RATE	input	Yes
Air viscosity	$\text{kg}/\text{m}\cdot\text{sec}$	ETA	input	Yes
Characteristic radius	μm	CHAR_R	input	Yes
Mean free path	μm	LAMBDA	input	Yes
Gamma	-	GAMMA	input	Yes
Fraction	-	F	constant	N/A
ALPHA	-	ALPHA	constant	N/A
BETA	-	BETA	constant	N/A
Characteristic Width	μm	CHAR_W	constant	N/A
Reference Height	m	REF_HEIGHT	constant	N/A
Mixing height	m	MIX_HEIGHT	internally calculated	N/A
Diffusion parameter	m	$\text{sigma}_{x,y,z}$ ($\sigma_{x,y,z}$)	internally calculated	N/A
Friction velocity	m/s	FRIC_VEL	internally calculated	N/A
Calculated value at receptor location				
Cummulative concentration ^b	Ci/m^3	CUM_CONC	output	N/A
Cummulative concentration ^c	Ci/m^3	CUM_CONC_RES	output	N/A
Cummulative inhalation dose ^d	mrem	INH_DOSE	output	N/A
Cummulative inhalation dose ^e	mrem	INH_DOSE_RES	output	N/A
Deposition velocity	m/sec	DEP_SPEED	output	N/A
Resuspension factor	sec^{-1}	RESUS_FACTOR	output	N/A

^aVariable name that will be seen in the source code

^bCalculated with dry deposition but without resuspension effect

^cCalculated with dry deposition and resuspension effect

^dCalculated with dry deposition but without resuspension effect

^eCalculated with dry deposition and resuspension effect

Table 3.5: Comparison results between CALPUFF and PIDA

Parameter	CALPUFF		PIDA	
Parameter	CASE I	CASE II	CASE I	CASE II
Maximum Concentration(mg/m ³)	6.1594E+03	6.1592E+03	1.0298E+04	1.0058E+04
Maximum Concentration(Ci/m ³)	5.3506E-01	5.3504E-01	7.0715E-01	6.9072E-01
Distance where Maximum Concentration obtained(m)	100	100	2500	2500
Time when Maximum Concentration obtained (hr)	23	23	1	1

Table 3.6: Input Parameters Characteristics

Parameters(unit)	Distribution type	Mean/Minimum Value ^a	Standard Deviation/Maximum Value ^b
SIZE(μm)	Lognormal	1.1776E-01	2.2749E-01
BREATH_RATE(m^3/day)	Lognormal	1.6200E+01	3.800E+00
CHAR_W(μm)	Lognormal	1.1776E-01	2.2747E-01
WIND_SPEED(m/sec)	Bounded Lognormal	1.445E+00	2.419E-01
ETA($\text{kg}/\text{m}\cdot\text{sec}$)	Uniform	1.7881E-05	1.8341E-05
RHO_AIR(kg/m^3)	Uniform	1.1581E+00	1.2398E+00
RHO_PAR(kg/m^3)	Uniform	3.8000E+03	3.9000E+03
CHAR_R(μm)	Uniform	5.0000E+02	1.000E+03
LAMBDA(μm)	Uniform	6.5100E-03	2.000E-01
ROUGHNESS(cm)	Uniform	5.3000E-03	1.0780E+02
GAMMA	Uniform	5.000E-01	6.667E-01
ALPHA	Constant	8.0000E-01	
BETA	Constant	2.0000E+00	
F	Constant	1.0000E-02	
REF_HEIGHT(m)	Constant	2.0000E+01	
MIX_HEIGHT(m)	N/A	N/A	
FRIC_VEL (m/sec)	N/A	N/A	

^aMean value is represented when distribution type is lognormal; Minimum value is represented when uniform distribution is defined

^bMean value is represented when distribution type is lognormal; Maximum value is represented when uniform distribution is defined

Chapter 4

Case Study

4.1 Scenario overview

To examine the use of the newly developed computer model to analyze the human dose under the event of radiological terrorism, a case study was performed with an assumed event scenario. The scenario used for the case study was the spent nuclear fuel sabotage.

The scenario of spent fuel sabotage has been investigated extensively by Sandia National Laboratory(SNL) [26]. The study by SNL has estimated the amount of radionuclide release from a SNF sabotage event along with the characterization of the size distributions of the particles released. Table 4.1 show the amount of radionuclide release estimated by the SNL study. According to Table 4.1, there are several key nuclides of concern in term of radiological health effect. However, in order to illustrate the functionality of the PIDA code, only ^{137}Cs with the prescribed amount was assumed to be released in the case study. Cs-137 was selected due to its relatively long half-life(33 years), and its wide use and applicability with various radioactive source materials such as spent nuclear fuel, medical and industrial sources. Cs-137 can also become volatile when it is heated during the explosion which makes this nuclide more dispersible [26]. Once the dispersion capability is increased, the resulting

health threat will also be increased. The followings explain additional assumptions used in this case study scenario.

- All of the 2.37E+02 Ci of ^{137}Cs was released into atmosphere [6] in the form of aerosol particles
- At the time of explosion, a neutral weather conditions exist
- The particle size of Cs-137 was lognormal distributed with the mean and standard deviation of 2.6213 μm and 2.833 μm respectively
- The density of aerosol particle was uniformly distributed with the minimum and maximum values of 3.80E+03 kg/m^3 and 3.90E+03 kg/m^3 , respectively.
- The human exposure to Cs-137 takes place at 1000 m directly downwind from the release location.

Table 4.1: Summary of RDD source materials [6]

Nuclide	SNF Sabotage Release Amount (Ci)	Other RDD Source Amount (Ci)
^{90}Sr	1.93E+01	1E+03-6E+05
^{106}Ru	1.33E+02	-
^{134}Cs	2.10E+02	-
^{137}Cs	2.37E+02	1E+03-1E+04
^{238}Pu	1.73E+00	1E+03-1E+02
^{239}Pu	7.70E-02	-
^{240}Pu	1.54E-01	-
^{241}Pu	2.33E+01	-
^{241}Am	1.57E+02	1E-01-2E+01
^{244}Cm	2.03E+00	-

4.2 Characterization of the uncertainty in input parameters

4.2.1 Particle size distribution Characteristic

Particle sizes in the air vary widely. Under the normal circumstances, ambient particle sizes are found to range from 0.056 μm to 10 μm [23]. However, this range

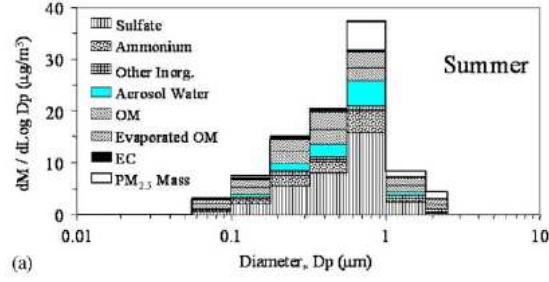
may not represent the size of the particles released from a specific event like a radiological terrorism. The size of particulate matters released under a terrorist event depends on a number of variables such as the type of material exploded and the explosive charge of the bomb. Typically, a much bigger and heavier particles can be released via a blast in a terrorist event.

Although the terrorist event is a unique event with a possibility of releasing any type of materials, Cs-137 and Ir-192 are the most likely used RDD material [47]. The type of material used will affect the size distributions of particles when a RDD blasts. To examine the sensitivity of the results, two different particle size distributions were used in the case study analysis.

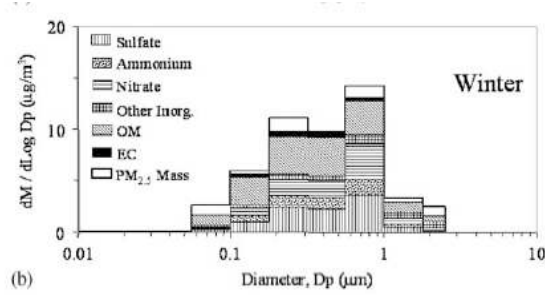
First, the size distribution model developed by Pittsburgh Air Quality Study group was adopted to represent the normal ambient particle size distributions [23]. In the Pittsburgh Air Quality Study group, the daily samples from Pittsburgh, PA area were collected for 12 months from July 2001 to June 2002. Their study presents data on particle mass concentrations in several size ranges ($0.056 - 10 \mu\text{m}$) in summer and winter as shown in Figure 4.1.

Second, the size distribution model developed by Molecke M.A. et al. [26] at the Sandia National Laboratory was used. Molecke et al. performed explosive tests using both spent fuel and surrogate materials. The resulting size distributions of the blast materials were measured as seen in Figure 4.2.

As the first results presented in Figures 4.1 and 4.2 do not come with designated mean and standard deviation values, the results were fitted by using the custom curve fit feature available from Crystal Ball. The results showed that both distributions can be best fitted by using the lognormal distribution. The mean, standard deviation and



(a) Summer



(b) Winter

Figure 4.1: Average size distribution during the two intensive periods, July 2001 and January 2002

the goodness of fits (Chi square) values were: (1) $0.78856 \mu\text{m}$, $0.94427 \mu\text{m}$ and 170.48, respectively for the Pittsburgh study data (2) $2.6213 \mu\text{m}$, $2.8336 \mu\text{m}$ and 230.720, respectively for the SNL test data.

4.2.2 Particle density

Like particle size, the density of the particle from the spent nuclear fuel blasts are different from the normal ambient case values, with typically much higher values. For the two different categories of particle sizes described in the previous section, two different sets of particle density distributions were utilized in the study. For a normal

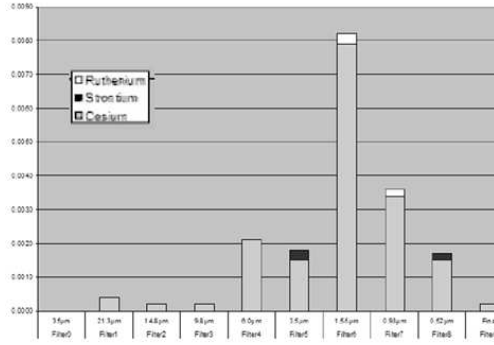


Figure 4.2: Respirable aerosol particle size distribution

ambient case(Pittsburgh study data), the ambient particle density data was used [15] with the uniform distribution with the minimum value of 1000 kg/m^3 (light particle such as fog) and the maximum value of 2600 kg/m^3 (dense particle such as lead). For the particles released from the a blast, the uniform distribution with the range of $3.8 - 3.9 \text{ kg/m}^3$ ⁱ was used based on the study by the Homland Security Council(HSC) on the RDD events [13]. The material considered in the HSC study for a RDD event was Cesium Chloride (CsCl) which is a white crystalline powder form with density of $3.8 - 3.9 \text{ kg/m}^3$.

4.2.3 Breathing Rate

According to Richardson [21], the 24-hour inhalation rates for most of the age groups can be represented by the lognormal distribution. The mean values and standard deviations for these distributions for adults are approximately 16.2 and $3.8 \text{ m}^3/\text{day}$, respectively.

ⁱThese density data were obtained from the website
[“http://www.chemicaland21.com/industrialchem/inorganic/CESIUM%20CHLORIDE.htm”](http://www.chemicaland21.com/industrialchem/inorganic/CESIUM%20CHLORIDE.htm)

4.2.4 Windspeed

Windspeed distribution was obtained from NUREG/CR-6676 [48], which was prepared by Argonne National Laboratory(ANL) to support the utilization of RESRAD and RESRAD-BUILD codes. The distribution was described by the bounded lognormal distribution with the mean of 1.445 m/sec and the standard deviation of 0.2419 m/sec. Lower and upper bounds were given as 1 m/sec and 16 m/sec, respectively.

4.3 Evaluation of particle size effects

Based on the assumptions described in section 4.1, calculation were made to determine the inhalation doses to human from a radiological terrorism event involving spent fuel sabotage. To illustrate the effect of particle size, changes in the results such as inhalation dose, deposition velocity, resuspension factor, and concentration were reported as function of particle size. The analysis were made by using the PIDA code for the three different cases: (1) When particle size is fixed at 1 μm (typical default value for the particle size used in many RDD studies) (2) When the particle size follows the hypothetical ambient particle size distributions [23], and (3) When the particle size follows the blast size distribution according to the SNL study [26].

Table 4.2 shows the results of the predicted inhalation dose to an adult(as committed effective dose) when particle size is fixed at different sizes including 1 μm in size. Table 4.3 shows the results of the predicted inhalation dose to an adult(as committed effective dose) when mean particle size ranged of 0.78856 μm (hypothetical ambient particle size distributions) is used. Table 4.4 shows the calculation results when mean particle size of 2.6213 μm (blast size distribution) is used. Table 4.5 summarizes the calculation results for key parameters.

4.3.1 Effect on dry deposition velocity

To illustrate the particle size effect on the dry deposition velocity, selected results from the calculations are shown in Table 4.6. Figure 4.3 illustrates the effect particle size on dry deposition velocity. Figure 4.4 shows the comparison of dry deposition velocity distribution of the three cases.

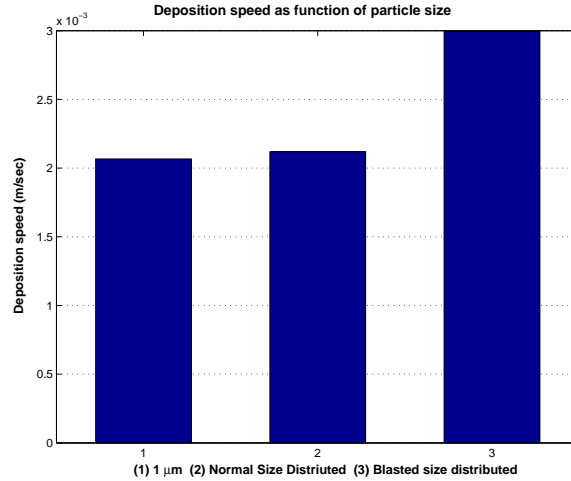


Figure 4.3: Effect of particle size on deposition speed

According to Table 4.6 and Figure 4.3 the bigger the particle the greater the dry deposition speed. The average of particle size at 1 μm will result in the greater dry deposition velocity compare to constant particle size at 1 μm .

4.3.2 Effect on the concentration at the receptor locations

To illustrate the particle size effect on the concentration, selected results from the calculations are shown in Table 4.5 and Table 4.7. According to data in these tables, the effect of particle size on the concentration at receptor location can also be plotted as seen in Figure 4.5. Figure 4.6 shows the comparison of the concentration distributions of the three cases.

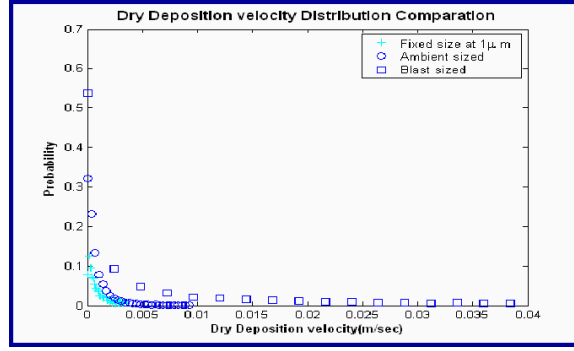


Figure 4.4: Dry deposition velocity distribution comparison

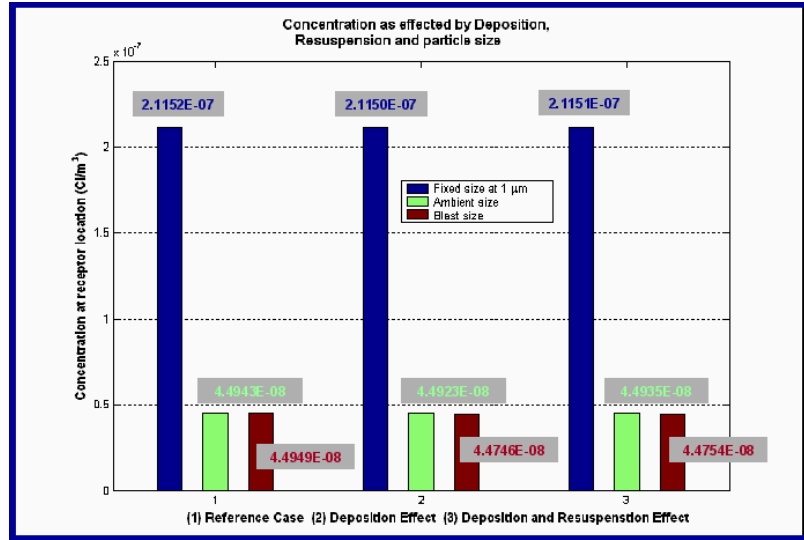


Figure 4.5: Effect of deposition, resuspension factor and particle size on concentration

According to Figure 4.5, the concentrations was significantly decreased from 2.1151E-07 Ci/m³ down to 4.4935E-08 Ci/m³ when the hypothetical ambient size distributions were considered instead of when constant particle size was conideres. This means that the existing computer codes overestimate the concentration. Once the blast particle size distribution was assumed (terrorist scenario), the concentration slightly decreased to 4.4754E-08 Ci/m³ from the case when the hypothetical ambient size distributions

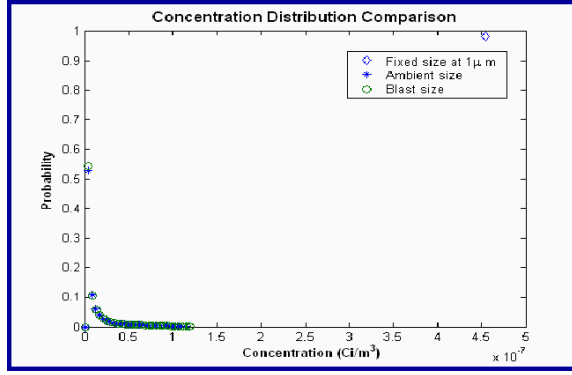


Figure 4.6: Concentration distribution comparisons

were considered. These imply that the existing computer code usually overestimated the concentration because most of them fix the particle size as constant at $1 \mu\text{m}$.

The comparison between the physical phenomenon was made as seen in Figure 4.5. The three compared bars for this case are the first bar of each section (1) reference, (2) deposition effect and, (3) deposition and resuspension effect cases. The cases were calculated based on (1) when no deposition nor resuspension is considered, (2) when deposition is considered and (3) when both deposition and resuspension is considered. The concentrations are not significantly changed for all cases nor obviously seen in the figures. Slightly changes were observed as shown in Table 4.5. The deposition effect slightly lowering the concentration for example about $9.45537\text{E-}03$ percent for constant particle size of $1 \mu\text{m}$. For other size range please see details in Table 4.8. These results are not only confirm the previous finding that the bigger the particle the more they deposited but also the overestimation of the calculation of the doses when taking only fixed particle size distributions. In practice, the more distance that the puff travels, the more deposition that could have. Please note that these calculation were made at 1000 m downwind from the release location. This distance is not far enough to see the significant effect of the deposition as also seen in the CALLPUFF benchmarking calculation (see section 3.4.1).

In the case study, both of the effects of deposition and the resuspension(for each designated particle size)are not significant, not only because the short distance but also short period of time (24 hr after release was used in the calculations). Generally, because it was not enough time to let the deposited particles to resuspend back into the atmosphere and eventually to the receptor location. Besides effect of particle size, in our case study both windspeed profile and time period dominated the concentration and consequently affects the inhalation doses.

4.3.3 Effect on resuspension

In order to illustrate the particle size effect on the resuspension, selected results from the calculations are shown in Table 4.9. Figure 4.7 illustrates the effect particle size on resuspension.

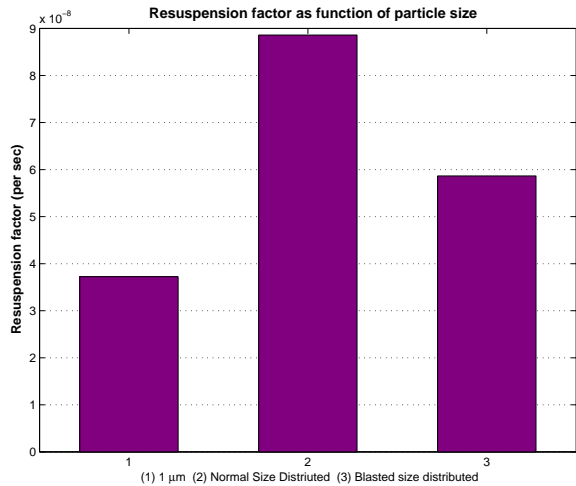


Figure 4.7: Effect of particle size on resuspension factor

The effect of particle size on the resuspension factor is shown in Table 4.9 and Figure 4.7. The resuspension factor increases with particle diameter and decreases with time.

However at certain period of time, the deposition was the domination factor that affect the concentration when the particle size is big as seen in Table 4.8 that the decrease in the percent increasing according the resuspension. Because the resuspension factor is bigger when time is small so when time(after deposition) increases the resuspension decreases. Another effect is that resuspension is increased as particle size increased. However for the blast size distribution case the time period has more affect on the resuspension than the particle size as seen in Figure 4.7.

4.3.4 Effect on inhalation dose

Effects of particle size on the inhalation dose can be seen through several mechanisms including (1) the dry deposition mechanism (2) the resuspension mechanism through the atmospheric transport and (3) the deposition in the lung. Figure 4.10 compares the inhalation doses for the three different cases as previously described. To illustrate this effect the inhalation dose calculated when both deposition and resuspension were considered (the last three bars in Figure 4.10). The inhalation doses was significantly decreased from 29 mrem down to 6 mrem when the hypothetical ambient particle size distribution was used instead fixed particle size at 1 μm . This means that the existing computer codes generally overestimate the inhalation doses roughly 5 times. Once the blast particle size distribution was assumed(terrorist event), the inhalation doses decreased to 4 mrem even lower than the ambient sizes case. This is because of the fact that the bigger particulate matter has more tendency to deposit along the way as its travel in lung as seen in Figure 4.3. Another way of saying is that the smaller the particle the longer the distance it can travel. This trend is also true for the another two groups of data when deposition and resuspension were not considered in the calculation.

The comparison between the physical phenomenon was made as also seen in Figure 4.10. The three compared bars for this case are the first bar of each section

(1) reference,(2) deposition effect and (3) deposition and resuspension effect cases. The cases were calculated (1) when no deposition nor resuspension was considered, (2) when deposition was considered and, (3) when both deposition and resuspension were considered. The inhalation doses are not significant changed for all cases. Slightly changes were observed as seen in Table 4.5, although they were not obviously seen in figures. The deposition effect slightly lowering the inhalation doses for constant particle size of $1\text{ }\mu\text{m}$ case. Please see details in Table 4.8 for other size ranges.

As seen in Figure 4.8 and Figure 4.9. It is clearly noticed that particle size variation strongly affects the inhalation doses, i.e several order of magnitudes differences in inhalation doses as particle size varied from 0.001 to $10\text{ }\mu\text{m}$. The particle size that gave the most doses is $0.3\text{ }\mu\text{m}$. But the maximum concentration was obtained at particle size of $1\text{ }\mu\text{m}$. The major observation here is that the deposition of particles in lung dominates the effects on inhalation doses more than the deposition of particles during atmospheric transport.

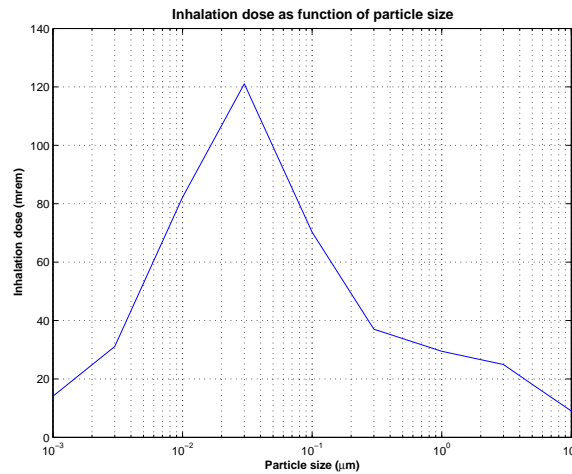


Figure 4.8: Effect of Particle size on Inhalation dose

Besides, the calculation results shown previously, the calculation results could be

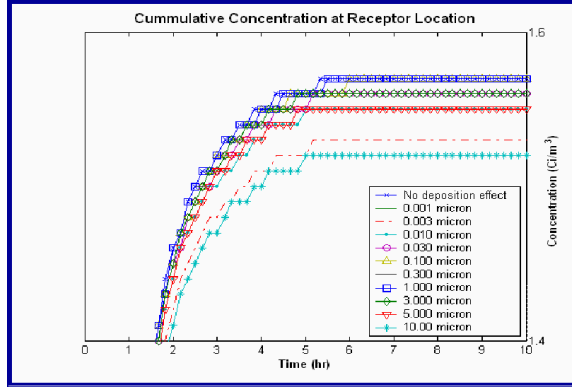


Figure 4.9: Cumulative concentration at receptor location as function of particle size

shown in the distribution plots as well. Figure 4.11 and Figure 4.12 show the probability distribution and cumulative density distribution of inhalation doses when particle size was fixed at $1 \mu\text{m}$ respectively. Figure 4.13 and Figure 4.14 show the probability distribution and cumulative density distribution of inhalation doses when particle size was fixed at $1 \mu\text{m}$ respectively. Figure 4.15 and Figure 4.16 show the probability distribution and cumulative density distribution of inhalation doses when particle size was fixed at $1 \mu\text{m}$ respectively. Figure 4.17 shows the comparison of density distribution for the three different cases.

4.3.5 Effect of other parameters on the inhalation doses

Besides particle size, there are other parameters that have an effect on the inhalation doses, such as the wind speed, air density etc. Figure 4.18 shows the list of uncertainty parameters that are contributed to the uncertainty of the inhalation doses sort by rank correlation coefficients. Highest rank correlation coefficient means highest uncertainty contributed to the calculation. Since the PIDA code has taken into account the effect of particle size via the models adopted in the code, the figure shows the other parameters (other than particle size) that affect the inhalation dose.

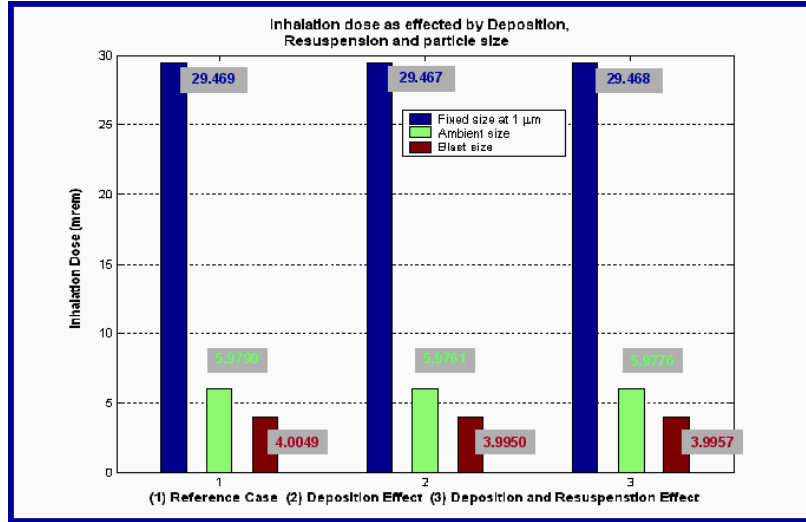


Figure 4.10: Effect of deposition, resuspension factor and particle size on inhalation dose

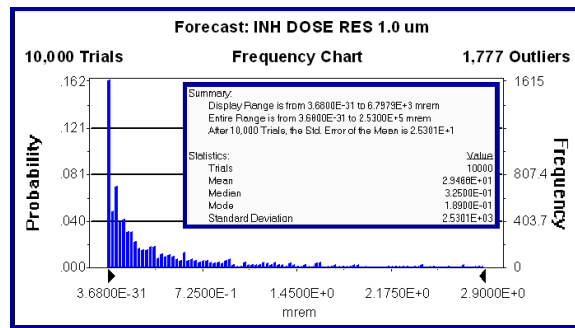


Figure 4.11: Inhalation dose distribution when particle size was fixed at 1 μm

There are two types of parameters that contribute to the uncertainty of inhalation doses. First, the physical parameters such as particle size, wind speed, air density, roughness, breathing rate and second the empirical model parameters in the adopted models such as Eta, Gamma, Characteristic radius. According to the rank correlation coefficient figure, the summary of key parameters are wind speed, eta, gamma, characteristic radius, air density, surface roughness, breath rate and particle density.

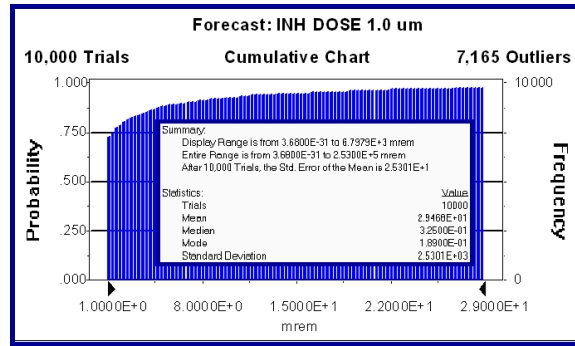


Figure 4.12: Cumulative density distribution of inhalation dose when particle size was fixed at 1 μm

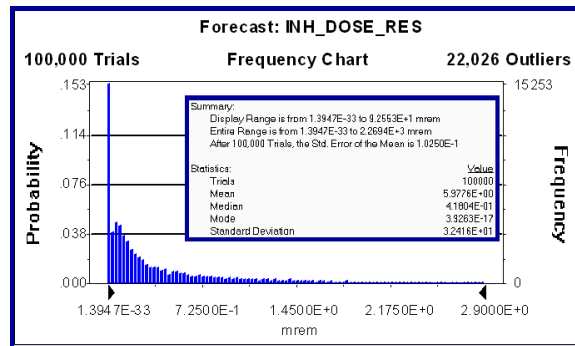


Figure 4.13: Inhalation dose distribution when ambient particle size distributions was used (0.7885 μm mean)

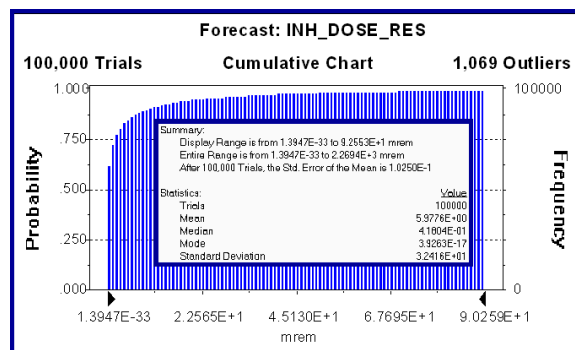


Figure 4.14: Cumulative density distribution of inhalation dose when ambient particle size distributions was used (0.7885 μm mean)

Table 4.2: Probabilistic committed effective inhalation dose (mrem) to adult public as function of particle size

Committed effective inhalation dose (mrem) ^a											
Statistics	No Effect ^b	0.001 μm	0.003 μm	0.01 μm	0.03 μm	0.1 μm	0.3 μm	1.0 μm	3.0 μm	5.0 μm	10.0 μm
Mean	2.9469E+01	1.4090E+01	3.1099E+01	8.2233E+01	1.2105E+02	7.0236E+01	3.7038E+01	2.9468E+01	2.4926E+01	1.8157E+01	9.0215E+00
SD	2.5301E+03	1.2101E+03	2.6701E+03	7.0603E+03	1.0400E+04	6.0302E+03	3.1801E+03	2.5301E+03	2.1401E+03	1.5601E+03	7.7603E+02
Variance	6.4013E+06	1.4642E+06	7.1294E+06	4.9847E+07	1.0817E+08	3.6363E+07	1.0113E+07	6.4013E+06	4.5799E+06	2.4338E+06	6.0222E+05
0 th Percentiles	3.6900E-31	0.0000E+00	3.4300E-31	1.0000E-30	1.5000E-30	8.7700E-31	4.6300E-31	3.6800E-31	3.0900E-31	2.1700E-31	0.0000E+00
2.5 th Percentiles	7.8000E-13	0.0000E+00	7.2500E-13	2.1200E-12	3.1700E-12	1.8600E-12	9.8000E-13	7.8000E-13	6.5100E-13	4.4500E-13	5.4800E-20
5.0 th Percentiles	5.1900E-08	0.0000E+00	4.3800E-08	1.4200E-07	2.1300E-07	1.2400E-07	6.5200E-08	5.1900E-08	4.3900E-08	1.8100E-08	1.6200E-13
50.0 th Percentiles	3.2500E-01	6.8200E-02	2.9800E-01	8.8500E-01	1.3200E+00	7.7300E-01	4.0800E-01	3.2500E-01	2.7300E-01	1.8800E-01	7.6300E-02
95.0 th Percentiles	1.6500E+01	7.7500E+00	1.7300E+01	4.6000E+01	6.7500E+01	3.9300E+01	2.0700E+01	1.6500E+01	1.4000E+01	1.0100E+01	5.0000E+00
97.5 th Percentiles	3.1100E+01	1.5100E+01	3.2900E+01	8.7000E+01	1.2700E+02	7.4100E+01	3.9100E+01	3.1100E+01	2.6300E+01	1.9100E+01	9.5300E+00
100.0 th Percentiles	2.5300E+05	1.2100E+05	2.6700E+05	7.0600E+05	1.0400E+06	6.0300E+05	3.1800E+05	2.5300E+05	2.1400E+05	1.5600E+05	7.7600E+04
Top 5 parameters that most sensitive to the committed effective inhalation dose											
Rank	No Effect	0.001 μm	0.003 μm	0.01 μm	0.03 μm	0.1 μm	0.3 μm	1.0 μm	3.0 μm	5.0 μm	10.0 μm
1 st Rank	Windspeed	Windspeed	Windspeed	Windspeed	Windspeed	Windspeed	Windspeed	Windspeed	Windspeed	Windspeed	Windspeed
2 nd Rank	LAMBDA	LAMBDA	LAMBDA	LAMBDA	LAMBDA	LAMBDA	LAMBDA	LAMBDA	LAMBDA	LAMBDA	LAMBDA
3 rd Rank	RHO AIR	RHO AIR	RHO AIR	RHO AIR	RHO AIR	RHO AIR	RHO AIR	RHO AIR	RHO AIR	RHO AIR	RHO AIR
4 th Rank	CHAR R	RHO PAR	CHAR R	CHAR R	CHAR R	CHAR R	CHAR R	CHAR R	CHAR R	CHAR R	CHAR R
5 th Rank	GAMMA	CHAR R	GAMMA	GAMMA	GAMMA	GAMMA	GAMMA	GAMMA	GAMMA	GAMMA	GAMMA

^aThe calculation was based on:

- (1) 2.37E+02 Ci Cs-137 released
- (2) 1000 m downwind from released location
- (3) 24 hr continuously breathing the released radioactive airborne
- (4) This Committed Effective Inhalation Dose represents the dose when considered dry deposition and resuspension effect

^bThis value represents the Committed Effective Inhalation Dose when no dry deposition and resuspension effects is taken into consideration at 1 μm size

Table 4.3: Probabilistic calculation results at 1000 m downwind for hypothetical size distributions

Statistics	PARAMETERS								
	INH DOSE ^a	INH DOSE VD ^a	INH DOSE RES ^c	DEP SPEED ^d	RESUS FAC ^e	RESUS CONC ^f	CONC ^g	CONC VD ^h	CONC RES ⁱ
Mean	5.9790E+00	5.9761E+00	5.9776E+00	2.1188E-03	8.8604E-08	7.2321E-11	4.4943E-08	4.4923E-08	4.4935E-08
SD	3.2422E+01	3.2413E+01	3.2416E+01	1.2371E-02	8.6123E-08	1.4674E-10	2.1871E-07	2.1865E-07	2.1867E-07
Variance	1.0512E+03	1.0506E+03	1.0508E+03	1.5304E-04	7.4172E-15	2.1533E-20	4.7832E-14	4.7807E-14	4.7817E-14
0 th Percentiles	1.3952E-33	1.3947E-33	1.3947E-33	3.7731E-05	2.2130E-09	1.4013E-45	8.4568E-42	8.4540E-42	8.4540E-42
2.5 th Percentiles	7.5684E-13	6.3134E-13	6.3169E-13	1.1130E-04	6.0726E-09	2.6911E-23	6.1060E-21	5.7528E-21	5.7588E-21
5.0 th Percentiles	5.4406E-08	5.2195E-08	5.2313E-08	1.3671E-04	8.1736E-09	3.7092E-18	4.8793E-16	4.7647E-16	4.7682E-16
50.0 th Percentiles	4.1843E-01	4.1736E-01	4.1804E-01	6.2520E-04	5.8625E-08	3.6849E-11	3.4464E-09	3.4243E-09	3.4308E-09
95.0 th Percentiles	2.4462E+01	2.4453E+01	2.4457E+01	4.6487E-03	2.6356E-07	2.5254E-10	1.8808E-07	1.8798E-07	1.8801E-07
97.5 th Percentiles	4.6140E+01	4.6136E+01	4.6143E+01	9.5790E-03	3.1579E-07	3.6302E-10	3.6298E-07	3.6292E-07	3.6295E-07
100.0 th Percentiles	2.2699E+03	2.2693E+03	2.2694E+03	5.8330E-01	7.9939E-07	5.3930E-09	1.1323E-05	1.1322E-05	1.1323E-05

^aCommitted Effective Inhalation Dose (mrem)

^bCommitted Effective Inhalation Dose (mrem)when considered dry deposition effect

^cCommitted Effective Inhalation Dose (mrem)when considered dry deposition and resuspension effect

^dDeposition velocity (m/sec)

^eResuspension factor (1/sec)

^fResuspension Concentration at receptor location(Ci/m³)

^gConcentration at receptor location(Ci/m³)

^hConcentration at receptor location(Ci/m³) when considered dry deposition effect

ⁱConcentration at receptor location(Ci/m³) when considered dry deposition and resuspension effect

Table 4.4: Probabilistic calculation results due to terrorist events at 1000 m downwind

Statistics	PARAMETERS								
	INH DOSE ^a	INH DOSE VD ^a	INH DOSE RES ^c	DEP SPEED ^d	RESUS FAC ^e	RESUS CONC ^f	CONC ^g	CONC VD ^h	CONC RES ⁱ
Mean	4.0049E+00	3.9950E+00	3.9957E+00	2.9946E-02	5.8655E-08	4.7860E-11	4.4949E-08	4.4746E-08	4.4754E-08
SD	2.0149E+01	2.0141E+01	2.0142E+01	8.0975E-02	5.4209E-08	9.7272E-11	2.1871E-07	2.1810E-07	2.1812E-07
Variance	4.0597E+02	4.0565E+02	4.0570E+02	6.5570E-03	2.9386E-15	9.4618E-21	4.7835E-14	4.7569E-14	4.7575E-14
0 th Percentiles	8.1337E-34	8.1324E-34	8.1337E-34	5.3926E-05	1.7605E-09	1.4013E-45	8.4568E-42	8.4554E-42	8.4568E-42
2.5 th Percentiles	4.9754E-13	2.4282E-14	2.4920E-14	1.8697E-04	4.3336E-09	1.8320E-23	6.1060E-21	2.7177E-22	3.3584E-22
5.0 th Percentiles	3.6744E-08	7.2566E-09	7.7312E-09	2.3475E-04	5.6151E-09	2.4930E-18	4.8793E-16	8.4254E-17	8.6895E-17
50.0 th Percentiles	2.8557E-01	2.7646E-01	2.7684E-01	1.9598E-03	3.9748E-08	2.4900E-11	3.4464E-09	3.2678E-09	3.2726E-09
95.0 th Percentiles	1.6478E+01	1.6472E+01	1.6474E+01	1.7221E-01	1.6892E-07	1.6512E-10	1.8808E-07	1.8789E-07	1.8792E-07
97.5 th Percentiles	3.1666E+01	3.1658E+01	3.1660E+01	2.8974E-01	1.9506E-07	2.3770E-10	3.6298E-07	3.6293E-07	3.6297E-07
100.0 th Percentiles	1.0853E+03	1.0850E+03	1.0851E+03	1.0087E+00	4.1713E-07	3.9228E-09	1.1323E-05	1.1320E-05	1.1320E-05

^aCommitted Effective Inhalation Dose (mrem)

^bCommitted Effective Inhalation Dose (mrem)when considered dry deposition effect

^cCommitted Effective Inhalation Dose (mrem)when considered dry deposition and resuspension effect

^dDeposition velocity (m/sec)

^eResuspension factor (1/sec)

^fResuspension Concentration at receptor location(Ci/m³)

^gConcentration at receptor location(Ci/m³)

^hConcentration at receptor location(Ci/m³) when considered dry deposition effect

ⁱConcentration at receptor location(Ci/m³) when considered dry deposition and resuspension effect

Table 4.5: Mean value of calculated key parameters

Parameters	1 μm	Normal(Ambient) particle size	Blast particle size
Deposition speed(m/sec)	2.0667E-03	2.1188E-03	2.9946E-02
Resuspension factor(1/sec)	3.7223E-08	8.8604E-08	5.8655E-08
Concentration ^a (Ci/m ³)	2.1152E-07	4.4943E-08	4.4949E-08
Concentration ^b (Ci/m ³)	2.1150E-07	4.4923E-08	4.4746E-08
Concentration ^c (Ci/m ³)	2.1151E-07	4.4935E-08	4.4754E-08
Inhalation doses ^a (mrem)	2.9469E+01	5.9790E+00	4.0049E+00
Inhalation doses ^b (mrem)	2.9467E+01	5.9761E+00	3.9950E+00
Inhalation doses ^c (mrem)	2.9468E+01	5.9776E+00	3.9957E+00

^aNo deposition and no resuspension effect were considered

^bOnly deposition effect was considered

^cBoth deposition and resuspension effects were considered

Table 4.6: Dry deposition velocity uncertainty results

Parameters	1 μm	Ambient size	Blast size
Mean(m/sec)	2.0667E-03	2.1188E-03	2.9946E-02
Standard Deviation (m/sec)	3.8729E-03	1.2371E-02	8.0975E-02
Minimum range (m/sec)	6.6600E-05	3.7731E-05	5.3926E-05
Maximum range (m/sec)	6.9800E-02	5.8330E-01	1.0087E-00

Table 4.7: Concentration uncertainty results

Parameters	1 μm	Ambient size	Blast size
Mean(Ci/m^3)	2.1151E-07	4.4935E-08	4.4754E-08
Standard Deviation (Ci/m^3)	1.6901E-05	2.1867E-07	2.1812E-07
Minimum range (Ci/m^3)	3.3000E-39	8.4540E-42	8.4568E-45
Maximum range (Ci/m^3)	1.6900E-03	1.1323E-05	1.1320E-05

Table 4.8: Effect of deposition and resuspension on concentration and inhalation doses in percent

Parameters	1 μm	Normal size distributed	Blast size distributed
Effect of deposition on concentration	9.45537E-03	4.45008E-02	4.51623E-01
Effect of resuspension on concentration	4.7281E-03	2.67123E-02	1.7878E-02
Effect of deposition on inhalation dose	6.78679E-03	4.8503E-02	2.4719E-01
Effect of resuspension on inhalation dose	3.39363E-03	2.50999E-02	1.75219E-02

Table 4.9: Resuspension factor uncertainty results

Parameters	1 μm	Ambient size	Blast size
Mean(1/sec)	2.0667E-03	2.1188E-03	2.9946E-02
Standard Deviation (1/sec)	3.8729E-03	1.2371E-02	8.0975E-02
Minimum range (1/sec)	6.6600E-05	3.7731E-05	5.3926E-05
Maximum range (1/sec)	6.9800E-02	5.8330E-01	1.0087E-00

Table 4.10: Inhalation dose uncertainty results

Parameters	1 μm	Ambient size	Blast size
Mean(mrem)	2.9468E+01	5.9776E+00	3.9957E+00
Standard Deviation (mrem)	2.5301E+03	3.2416E+01	2.0142E+01
Minimum range (mrem)	3.6800E-31	1.3947E-33	8.1337E-34
Maximum range (mrem)	2.5300E+05	2.2694E+03	1.0851E+03

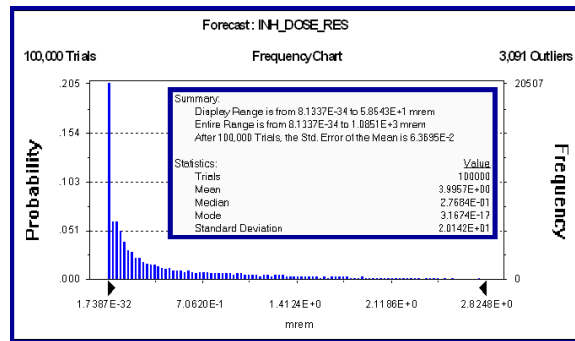


Figure 4.15: Inhalation dose distribution when blast particle size distributions was used ($2.6213 \mu\text{m}$ mean)

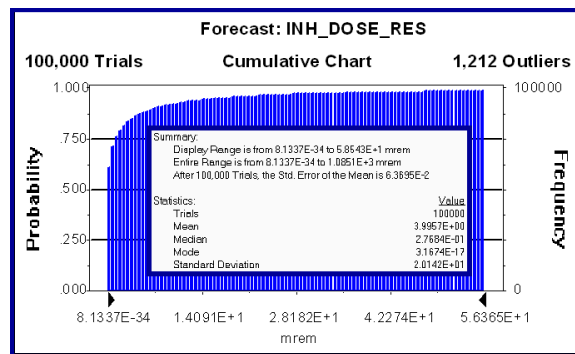


Figure 4.16: Cumulative density distribution of inhalation dose when blast particle size distributions was used ($2.6213 \mu\text{m}$ mean)

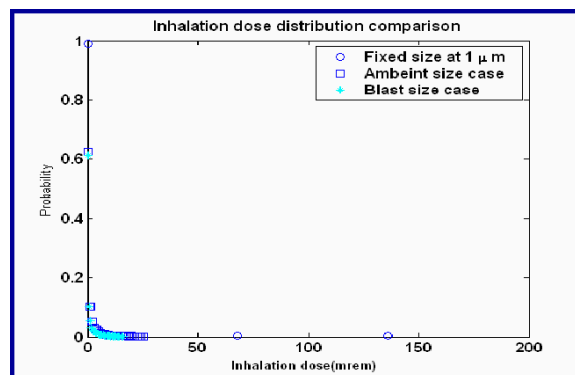


Figure 4.17: Inhalation dose distribution comparison

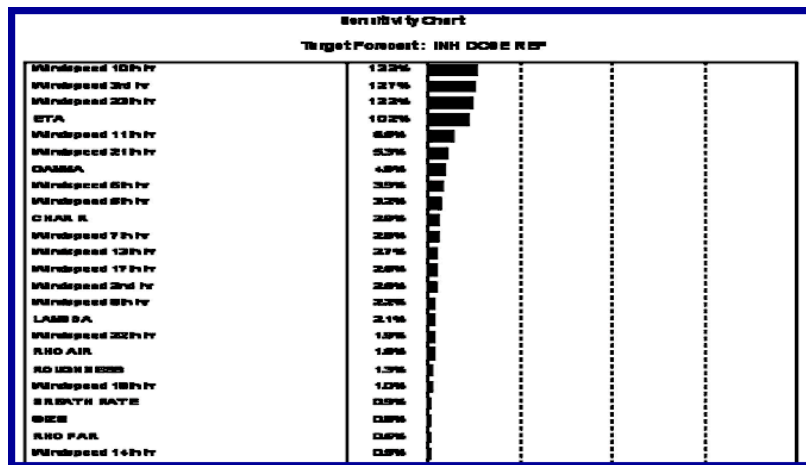


Figure 4.18: Rank correlation coefficients of input parameters for the calculation when particle size was modeled as an uncertain parameter (mean=2.6 μm)

Chapter 5

Discussion

5.1 Summary of observations and the implications

The PIDA code was successfully developed and implemented as a radiological assessment tool for a short-term terrorist event. The code is capable of evaluating inhalation doses as a function of the size of radioactive airborne particles after being instantaneously released. Given the understanding that inhalation doses are mainly controlled by exposure time, human breathing rate and the type of radiation being exposed to, effect of particle size was the main focus of the study in this research.

5.1.1 Summary of the Observations on the effect of particle size on key parameters

5.1.1.1 Effects of particle size to dry deposition velocity

- The Dry deposition velocity is sensitive to particle size. The particle size that gives the minimum deposition velocity is $1\ \mu\text{m}$.
- The dry deposition velocity increases when the particle size increases.
- The average of particle size at $1\ \mu\text{m}$ will result in the greater dry deposition velocity compare to constant particle size at μm .

5.1.1.2 Effects of particle size to the concentration

- Concentration is maximum when particle size is 1 mm
- Both of the effects of dry deposition velocity and resuspension to concentrations are small at this receptor location.

5.1.1.3 Effect of particle size to inhalation dose

- Inhalation dose is very sensitive to particle size. The maximum inhalation dose is obtained when particle size is $0.3\ \mu\text{m}$.
- The maximum dose coefficient is obtained at the particle size of $0.3\ \mu\text{m}$
- The deposition of particles in lung dominates the effects on inhalation doses more than the deposition of particles during atmospheric transport

5.1.2 Summary of the observations on the effect of the uncertain and fixed particle size input

Two major observations are as following.

- The deterministic approach that uses fixed $1\ \mu\text{m}$ as particle size overestimates the inhalation dose roughly by factor of 5 compare to the case when probabilistic particle size model is used
- The deterministic approach that uses fixed $1\ \mu\text{m}$ as particle size overestimates the concentration roughly by factor of 4 compare to the case when probabilistic particle size model is used.

5.1.3 Summary of key parameters (other than particle size) on the calculation of inhalation dose

The following parameters are sorted by rank correlation coefficient which starts from the most uncertain parameter.

1. Wind speed
2. Eta (empirical parameter in dry deposition model)
3. Gamma (empirical parameter in dry deposition model)
4. Characteristic radius (empirical parameter in dry deposition model)
5. Air density
6. Surface roughness
7. Breath rate
8. Particle density

Two types of parameters that contribute to the uncertainty of inhalation doses include:

- The physical parameters such as particle size, wind speed, air density, roughness, breathing rate
- The empirical model parameters in the adopted models such as Eta, Gamma, Characteristic radius

5.2 Validity and limitations of the assumptions

The PIDA code is working under certain assumptions and limitations as stated previously. However, the major limitation of this code can be categorized into followings subjects.

5.2.1 Code applicability

The PIDA code is developed for the case of instantaneous release. Event such as accident or terrorist events that includes sudden start and stop of radioactive material release within short periods of time can be analyzed by this code. If a long term routine release of radioactive material is involved, the Gaussian plume model based approach should be used.

5.2.2 Exposure pathway

Since the PIDA code was developed mainly for determining the inhalation dose pathway, other pathways, i.e. externally exposure and ingestion pathway were not modeled. The probability of a person swallowing or drinking the contaminated pollutant is low in a short-term terrorist event. Derived exposure is possible if an individual is submerged in a radioactive cloud or stay very close to the release location around the blast site. But in general the dose attributable to external dose is expected to be small compared to inhalation dose in a short-term terrorist event. In the future, inclusion of these pathways will be necessary to make the complete code to be a complete radiological assessment tool.

5.2.3 Wind speed and wind direction

The PIDA code is currently set up as one dimensional modelling tool which means it could handle only non-negative windspeed in the one downwind direction and calculates the concentration according unidirectional windspeed. This calculation represents only a hypothetical case. In reality, both wind direction and speed change during the day which can reverse the directional movement of a puff and its velocity that would reduce the concentration at the receptor location. One dimensional Gaussian puff model provides a conservative estimate of the pollutant concentration which may be acceptable for a screening analysis. In the future this one directional

assumption can be relaxed to allow 3-dimensional designation of the puff movement with the support of relevant input data.

5.2.4 Weather stability

One of the major limitations with the current PIDA code is the limited nature of the mixing height model. The current mixing height model can only represent the stability classes of C, D, E and F. In general, the stability class is one of the most sensitive parameters to describe atmospheric dispersion of pollutant. Future work need to be performed to include the consideration of the stability class A and B into the mixing height model.

5.2.5 Particle size

Regarding the consideration of particle size in this research, one of the assumptions made was that the maximum particle size is set at 10 μm . This assumption is conservatively allowing the code to treat any larger particles to be able to penetrate into the respiratory tract system, and consequently delivering the doses. Technically, many of the larger particles should be considered as external radiation because they may not penetrate into the respiratory system. For a terrorist event involving a RDD, there is a large possibility of having the particle sizes of the released blast materials to be larger than 10 μm . Hence, the calculation will overestimate the actual inhalation doses.

5.2.6 Resuspension

With regards to modelling particle resuspension, the PIDA code assumes that no resuspendable material has been deposited at the location prior to the releases. The code will account for the resuspension of materials that results from the emergence release only. If the code is used to analyse the releases from a place where the amount

of the previous deposited materials are nonnegligible, the code will underestimate the degree of resuspension.

5.3 Discussion on the case study

Notice that the case study examined the release of single nuclide, Cs-137, during the terrorist event. This case study is also based on a given set of meteorological data and other parameters as well. In order to model the spent nuclear fuel sabotage event more realistically, it is necessary to gather more detailed information on types, amount of radionuclide being released along with their particle size distributions. Also site specific values need to be used for some of the input parameters such as windspeed, wind direction and roughness.

Chapter 6

Conclusions and Future works

6.1 Conclusions

As discussed in previous chapters, the PIDA code was successfully developed and implemented (under certain assumptions) to analyze the effect of particle size distribution in the consequence analysis of radiological terrorist event. For the benchmarking of the code, each submodels of PIDA code were compared with the state-of-the-art modelling tool for each specific area for the calculation results. It was found that PIDA code in general is conservative in estimation the radiological consequence of the event. Using the PIDA code, sensitivity and uncertainty analysis for the particle size effect were performed. Results indicated that the inhalation dose is very sensitive to the changes in particle size. It was also found that the particle size effect is:

- Minor in describing atmospheric transport
- Minor in describing deposition
- Minor in describing resuspension
- Significant in describing lung deposition and resulting the dose

Particle size is one of the key parameters that contributes to the uncertainty of an inhalation dose evaluation due to this terrorist event. Ignoring particle size distribution is expected to result in overestimation of inhalation dose in a radiological terrorist event.

6.2 Future Works

Future works could be categorized into (1) the modeling/coding improvement (2) input parameters redefinition and (3) the code applications.

Code Improvement

- The code should be extended to estimate external and ingestion dose, to be a complete radiological assessment tool for a short-term terrorist event.
- The mixing height model should be extended to consider stability class A and B.
- The code could be extended to describe three dimensional movement of a puff.
- Multiple nuclides handling capability should be added
- A user-friendly version of this code (e.g.GUI) could be developed.

Input Parameters Improvement

- Probabilistic uncertainty model for input random variables could be re-defined to describe problem or site-specific situation
- A more complete terrorist event scenario should be developed with respect to the type of key nuclides released, amount of their releases, fraction of the release, and the size distribution of each released key nuclide particles.

Code application

Bayesian analysis using the code features can be performed to identify the key areas of future research needs with respect to characterizing human health risks and risk management options under a RDD terrorist event. Value of information analysis could be performed for this purpose. Applications to other types of terrorist event(e.g. non-radiological event) could be explored.

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Appendix A

PuPDReDIDA input file

A.1 Lists of Input Files and Formats

A.1.1 File “meteorological.dat”

Normal text file format with spacing between each data is the required format for “meteorological.dat” file. Note that additional hourly data could be added into this file inorder to perform extended period. The first column represents the releases hours, 24 represents 24 hours period. The second, third and forth column represent the windspeed in x-direction, y-direction and z-direction in unit m/sec respectively. However, the y and z directions windspeed were not used in this research due to simplification nature of this research. The last column defines the weather stability data for each hour, this must be in upper case letter format.

1	1.07376847268354	0	0	C
2	1.58767104782499	0	0	C
3	1.8687046933806	0	0	E
4	1.58768550510981	0	0	D
5	1.27959216156419	0	0	D
.
.
.
.
.
21	1.35607525444542	0	0	D
22	1.45175154932391	0	0	D
23	1.36437991175735	0	0	E
24	2.45699847492165	0	0	F

A.1.2 File “dose_factor.dat”

Normal text file format with spacing between each data is the required format for “dose_factor.dat” file as well. Note that additional dose coefficient data could be added into this file inorder to increase number of nuclides being able to calculated. The ascending number of column represents the dose coefficient for (each designate nuclide in the last column) particle size number 0.001 μm , 0.003 μm , 0.01 μm , 0.03 μm , 0.1 μm , 0.3 μm , 1.0 μm , 3.0 μm , 5.0 μm , and 10.0 μm respectively. Except the last column defines nuclide name, this must be in upper case letter format for the

first letter, lower case format for second letter and consecutively followed by atomic number i.e no space allowed. Additional data could be also entered here.

1.90E-11	7.20E-11	5.90E-10	1.10E-09	6.50E-10	3.40E-10	2.60E-10	2.10E-10	1.60E-10	7.80E-11	"H3"
8.10E-09	1.70E-08	2.50E-08	2.50E-08	1.30E-08	7.00E-09	5.80E-09	5.20E-09	4.00E-09	2.70E-09	"C14"
1.70E-08	3.50E-08	8.70E-08	1.30E-07	7.30E-08	3.80E-08	3.10E-08	2.60E-08	1.90E-08	1.10E-08	"Co60"
3.80E-08	9.10E-08	3.80E-07	6.60E-07	3.80E-07	2.00E-07	1.60E-07	1.30E-07	9.30E-08	4.50E-08	"Sr90"
5.90E-08	6.80E-08	8.10E-08	7.90E-08	4.30E-08	2.60E-08	3.60E-08	5.10E-08	5.10E-08	4.60E-08	"I129"
2.20E-08	4.30E-08	1.10E-07	1.60E-07	9.30E-08	4.90E-08	3.90E-08	3.30E-08	2.40E-08	1.20E-08	"Cs137"
1.60E-04	2.30E-04	3.30E-04	3.50E-04	1.90E-04	1.10E-04	1.20E-04	1.50E-04	1.40E-04	1.20E-04	"Pu239"
1.60E-04	2.30E-04	3.30E-04	3.50E-04	1.90E-04	1.10E-04	1.20E-04	1.50E-04	6.30E-09	3.60E-09	"Sr89"
8.40E-09	9.70E-09	1.20E-08	1.10E-08	6.10E-09	3.70E-09	5.10E-09	7.20E-09	7.30E-09	6.60E-09	"I125"
1.20E-08	1.40E-08	1.70E-08	1.60E-08	8.80E-09	5.30E-09	7.40E-09	1.00E-08	1.10E-08	9.50E-09	"I131"

A.1.3 File "thick_layer.dat"

Normal text file format with spacing between each data is also the required format for "thick_layer.dat" file. The first column represents think layer coefficients taken from Seinfeld [61] inorder to model friction velocity. The second column defines the weather stability data for each hour, this must be in upper case letter format.

-3.37467	"A"
-4.21363	"B"
-7.91517	"C"
999999.	"D"
7.79183	"E"
3.52511	"F"

Appendix B

ICRP Dose Coefficeint Database software Output

B.1 List of input parameters

Input Lists

- **Nuclide:** Cs-137
- **Subjects:** Public
- **Age at Intake:** Adult
- **Intake Routes:** Inhalation
- **Aerosol size:** 0.001, 0.003, 0.01, 0.03, 0.1, 0.3, 1.0, 3.0, 5.0, 10.0
- **Number of period:** To age 70 years
- **Organ/Tissues:** Effective Dose

B.2 Output Results

Output Lists

“Cs-137, adult member of the public”

“Inhalation of particulate aerosol: AMAD = 0.001 micron, absorption Type F, fl = 1.0”

“Highest committed equivalent dose coefficient: ET Airways, 1.6E-08 Sv/Bq ”

Remainder formulation: split

Time after intake: 50 years

Effective dose: 7.70E-09

“Inhalation of particulate aerosol: AMAD = 0.003 micron, absorption Type F, fl = 1.0”

“Highest committed equivalent dose coefficient: ET Airways, 1.5E-08 Sv/Bq ”

Remainder formulation: split

Time after intake: 50 years

Effective dose: 8.80E-09

“Inhalation of particulate aerosol: AMAD = 0.010 micron, absorption Type F, fl = 1.0”

“Highest committed equivalent dose coefficient: ET Airways, 1.3E-08 Sv/Bq ”

Remainder formulation: split

Time after intake: 50 years

Effective dose: 1.00E-08

“Inhalation of particulate aerosol: AMAD = 0.030 micron, absorption Type F, fl = 1.0”

“More than one organ/tissue have the same highest committed equivalent dose coefficient”

Remainder formulation: default

Time after intake 50 years

Effective dose 1.00E-08

“Inhalation of particulate aerosol: AMAD = 0.100 micron, absorption Type F, fl = 1.0”

“Highest committed equivalent dose coefficient: Colon, 6.2E-09 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 5.40E-09

“Inhalation of particulate aerosol: AMAD = 0.300 micron, absorption Type F, fl = 1.0”

“Highest committed equivalent dose coefficient: ET Airways, 4.1E-09 Sv/Bq ”

Remainder formulation: split

Time after intake 50 years

Effective dose 3.30E-09

“Inhalation of particulate aerosol: AMAD = 1.000 micron, absorption Type F, fl = 1.0”

“Highest committed equivalent dose coefficient: ET Airways, 7.4E-09 Sv/Bq ”

Remainder formulation: split

Time after intake 50 years

Effective dose 4.60E-09

“Inhalation of particulate aerosol: AMAD = 3.000 micron, absorption Type F, fl = 1.0”

“Highest committed equivalent dose coefficient: ET Airways, 1.2E-08 Sv/Bq ”

Remainder formulation: split

Time after intake 50 years

Effective dose 6.60E-09

“Inhalation of particulate aerosol: AMAD = 5.000 micron, absorption Type F, fl = 1.0”

“Highest committed equivalent dose coefficient: ET Airways, 1.3E-08 Sv/Bq ”

Remainder formulation: split

Time after intake 50 years

Effective dose 6.70E-09

“Inhalation of particulate aerosol: AMAD =10.000 micron, absorption Type F, fl = 1.0”

“Highest committed equivalent dose coefficient: ET Airways, 1.2E-08 Sv/Bq ”

Remainder formulation: split

Time after intake 50 years

Effective dose 6.00E-09

“Inhalation of particulate aerosol: AMAD = 0.001 micron, absorption Type M, fl = 0.1”

“Highest committed equivalent dose coefficient: Lungs, 1.3E-07 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 1.70E-08

“Inhalation of particulate aerosol: AMAD = 0.003 micron, absorption Type M, f1 = 0.1”

“Highest committed equivalent dose coefficient: Lungs, 2.3E-07 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 3.00E-08

“Inhalation of particulate aerosol: AMAD = 0.010 micron, absorption Type M, f1 = 0.1”

“Highest committed equivalent dose coefficient: Lungs, 2.7E-07 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 3.70E-08

“Inhalation of particulate aerosol: AMAD = 0.030 micron, absorption Type M, f1 = 0.1”

“Highest committed equivalent dose coefficient: Lungs, 2.4E-07 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 3.60E-08

“Inhalation of particulate aerosol: AMAD = 0.100 micron, absorption Type M, f1 = 0.1”

“Highest committed equivalent dose coefficient: Lungs, 1.3E-07 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 2.00E-08

“Inhalation of particulate aerosol: AMAD = 0.300 micron, absorption Type M, f1 = 0.1”

“Highest committed equivalent dose coefficient: Lungs, 7.0E-08 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 1.10E-08

“Inhalation of particulate aerosol: AMAD = 1.000 micron, absorption Type M, f1 = 0.1”

“Highest committed equivalent dose coefficient: Lungs, 6.3E-08 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 9.70E-09

“Inhalation of particulate aerosol: AMAD = 3.000 micron, absorption Type M, f1 = 0.1”

“Highest committed equivalent dose coefficient: Lungs, 6.1E-08 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 9.60E-09

“Inhalation of particulate aerosol: AMAD = 5.000 micron, absorption Type M, f1 = 0.1”

“Highest committed equivalent dose coefficient: Lungs, 4.8E-08 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 7.80E-09

“Inhalation of particulate aerosol: AMAD =10.000 micron, absorption Type M, f1 = 0.1”

“Highest committed equivalent dose coefficient: Lungs, 2.5E-08 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 4.60E-09

“Inhalation of particulate aerosol: AMAD = 0.001 micron, absorption Type S, f1 = 0.01”

“Highest committed equivalent dose coefficient: Lungs, 1.7E-07 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 2.20E-08

“Inhalation of particulate aerosol: AMAD = 0.003 micron, absorption Type S, f1 = 0.01”

“Highest committed equivalent dose coefficient: Lungs, 3.5E-07 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 4.30E-08

“Inhalation of particulate aerosol: AMAD = 0.010 micron, absorption Type S, f1 = 0.01”

“Highest committed equivalent dose coefficient: Lungs, 8.3E-07 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 1.10E-07

“Inhalation of particulate aerosol: AMAD = 0.030 micron, absorption Type S, f1 = 0.01”

“Highest committed equivalent dose coefficient: Lungs, 1.2E-06 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 1.60E-07

“Inhalation of particulate aerosol: AMAD = 0.100 micron, absorption Type S, f1 = 0.01”

“Highest committed equivalent dose coefficient: Lungs, 7.1E-07 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 9.30E-08

“Inhalation of particulate aerosol: AMAD = 0.300 micron, absorption Type S, f1 = 0.01”

“Highest committed equivalent dose coefficient: Lungs, 3.7E-07 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 4.90E-08

“Inhalation of particulate aerosol: AMAD = 1.000 micron, absorption Type S, f1 = 0.01”

“Highest committed equivalent dose coefficient: Lungs, 3.0E-07 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 3.90E-08

“Inhalation of particulate aerosol: AMAD = 3.000 micron, absorption Type S, f1 = 0.01”

“Highest committed equivalent dose coefficient: Lungs, 2.5E-07 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 3.30E-08

“Inhalation of particulate aerosol: AMAD = 5.000 micron, absorption Type S, f1 = 0.01”

“Highest committed equivalent dose coefficient: Lungs, 1.8E-07 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 2.40E-08

“Inhalation of particulate aerosol: AMAD =10.000 micron, absorption Type S, f1 = 0.01”

“Highest committed equivalent dose coefficient: Lungs, 8.9E-08 Sv/Bq ”

Remainder formulation: default

Time after intake 50 years

Effective dose 1.20E-08