METHODS FOR ESTIMATING FUGITIVE AIR EMISSIONS OF RADIONUCLIDES FROM DIFFUSE SOURCES AT DOE FACILITIES

FINAL REPORT

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Section	n		Page
EXEC	UTIVE	SUMMARY	xii
1.0	Introdu	uction	1-1
	1.1	Background and Objectives	1-1
	1.2	Structure of Report	1-2
2.0	Identif	fication and Characterization of Emission Sources	2-1
	2.1	Types of Emission Sources	2-1
		2.1.1 Point Sources and Diffuse Sources	2-2
		2.1.2 Continuous and Intermittent Sources	2-2
		2.1.3 Mechanisms	2-2
		2.1.4 Mitigating Processes	2-2
	2.2	Types of Radiological Emissions	2-3
		2.2.1 Chemical and Physical Forms of Radionuclide Emissions	2-3
		2.2.2 Radionuclides Emitted	2-3
	2.3	Characterization of Emission Sources	2-7
		2.3.1 Emission Data Measured at the Source	2-7
		2.3.2 Results from Site-Specific Characterization Studies	2-7
		2.3.3 Routine Sampling and Monitoring Results	2-8
		2.3.4 Process or Activity Related Information	2-8
		2.3.5 Default Values	2-8
	2.4	Considerations for Use	2-8
3.0	Airbor	rne Resuspension of Particulates	3-1
	3.1	Research on Resuspension	3-3
	3.2	Studies of Wind Erosion	3-5
		3.2.1 Mechanisms of Wind Erosion	3-5
		3.2.2 Characterizing Wind Erosion Studies Prior to 2001	3-6

TABLE OF CONTENTS

TABLE OF CONTENTS (Continued)

Secti	on		Page
		3.2.3 DOA Wind Erosion Equation3.2.4 Open Areas3.2.5 Open Waste and Storage Piles	3-7 3-7 3-8
		3.2.6 Uranium Ore and Mill Tailings	3-8
	3.3	EPA Soil Screening Guidance	3-9
	3.4	Radionuclide Concentrations in Airborne Particulate Matter	3-10
	3.5	Considerations for Use	3-11
4.0	Anth	ropogenic Mechanical Suspension of Particulates	4-1
	4.1	Soil and Material Handling	4-1
		4.1.1 Soil Removal and Haulage	
		4.1.2 Grading and Shaping of Soil	
		4.1.3 Agricultural Tillage and Seeding	
		4.1.4 Dunding Demontion and Material Disposal	
	4.2	Vehicular Traffic on Unpaved Roads	4-4
	4.3	Control Methods	4-5
	4.4	Considerations for Use	4-5
5.0	Mech	nanisms Other Than Resuspension and Anthropogenic Mechanical Suspensio	on 5 1
	Activ	ities That Affect Fugitive Emissions	
	5.1	Evaporation Models	5-1
		5.1.1 Open Bodies of Water	5-1
		5.1.2 Wet Cooling Towers	5-4
	5.2	Evapotranspiration from Contaminated Soil	5-5
		5.2.1 Evapotranspiration from Saturated Soil	5-5
		5.2.2 Subsurface Contamination of Tritium	5-9
	5.3	Gaseous and Other Types of Emissions	5-10
		5.3.1 Re-Entry Drilling	5-10

TABLE OF CONTENTS (Continued)

Section	1	Pa	ıge
		5.3.2Ground Seepage of Noble Gases5-5.3.3Emissions from Buildings5-5.3.4Emissions from Tank Venting5-5.3.5Emissions from Equipment5-	-11 -12 -13 -15
	5.4	Considerations for Applications5-5.4.1 Evaporation5-5.4.2 Evapotranspiration from Soil5-5.4.3 Gaseous and Other Types of Emissions5-	-16 -16 -16 -17
6.0	Guidan with th	ce on Environmental Monitoring Programs to Demonstrate Compliance e DOE NESHAPS	5-1
	6.1 6.2	Summary of NESHAPS Requirements6Sampling and Analytical Methodology66.2.1Radionuclides as Particulates6.2.2Radionuclides as Gases	5-1 5-1 5-2 5-2
	6.3	Criteria for Environmental Monitoring Programs66.3.1Selecting Critical Receptor Locations6.3.2Continuous Sampling6.3.3Identifying the Major Radionuclide Contributor6.3.4Determining Radionuclide Concentrations Separate from Background6.3.5When Background Concentrations Interfere	5-4 5-4 5-5 5-5 5-6 5-7
	6.4	Evaluating the Validity of a Quality Assurance Program	5-8
7.0	Guidan Source	ce on Methods for Estimating Fugitive Radionuclide Air Emissions from Diffus	e 7-1
	7.1 7.2 7.3 7.4	Introduction 7 Identify Sources of Radionuclides Emissions 7 Determine Population of Emission Sources 7 Determine Appropriate Emission Estimation Methods 7 7.4.1 Soils 7 7.4.1.1 Wind Erosion - Limited Open Areas and Intermittent 7 Waste Piles 7 7 7.4.1.2 Wind Erosion - Unlimited Open Areas 7	7-1 7-2 7-4 7-6 7-7 -14

TABLE OF CONTENTS (Continued)

Section

			7.4.1.3 Material Handling - Ongoing Waste Pile Operations and	Soil
			7.4.1.4 Meterial Handling Soil Creding and Shaning	
			7.4.1.4 Material Handling - Son Grading and Snaping	/-1/ 7 19
			7.4.1.5 Material Handling Demolition	
			7.4.1.0 Material Handling - Demontion	
		7.4.2	Water/Evaporation	7-24
			7.4.2.1 Evaporation from Ponds and Lagoons	
			7.4.2.2 Evaporation from Cooling Towers	
			7.4.2.3 Evapotransportation from Contaminated Soil	
		7.4.3	Underground Testing	
		7.4.4	Emissions from Buildings and Equipment	
		7.4.5	Emissions from Chemical Storage Tanks	
	7.5	Collec	et Required Data and Calculating Emissions	
	7.6	Qualit	y Assurance	7-40
	7.7	Emiss	ions Documentation and Reporting	
	7.8	Case S	Studies	
		7.8.1	Oak Ridge Reservation (ORR) – Fugitive Emissions	
			for Diffuse Sources	
		7.8.2	Idaho National Engineering and Environmental Laboratory	
			(INEEL) - Fugitive Emissions from Diffuse Sources	
		7.8.3	Hartford Site – Fugitive Emissions from Diffuse Sources	7-49
8.0	Refe	ences C	Cited	8-1
9.0	Biblic	graphy		9-1
Attacl	nment A	Additi	onal References for Sampling Diffuse Source Emissions	

Attachment B Methodologies Submitted by DOE and Approved by EPA

LIST OF TABLES

Table

ES-1	Summary of Methods for Estimating Diffuse Source Emissions	xvii	
2-1	Radionuclide Classification and Radioactivity Distribution Using DOE Categories 2-4		
2-2	Summary of Diffuse Source Airborne Radionuclide Emissions (in Curies) at DOE		
	Production Sites and Research Laboratories During Calendar Year 1998, Excludin	g	
	Radon	2-5	
3-1	Reported Resuspension Factors	3-4	
3-2	Wind Erosion Mechanisms Versus Particle Size	3-6	
4-1	Summary of AP-42 Emissions Control Measures	4-5	
6-1	Physical Parameters of Selected Primary Radionuclides	6-3	
6-2	Short Half-Life Radionuclides	6-4	
6-3	Examples of Backgrounds and Sensitivities of Some Principal Airborne Radionucl	ides	
	Released From DOE Facilities	6-7	
7-1	Summary of Methods for Estimating Diffuse Source Emissions	7-5	
7-2	Threshold Friction Velocities	7-8	
7-3	Particle Size Multiplier	7-10	
7-4	Required Data for Preferred Method for Limited Open Areas and		
	Intermittent Waste Piles	7-10	
7-5	Calculation of Friction Velocities	7-13	
7-6	Surface Area Apportionment	7-14	
7-7	Calculation of PM ₁₀ Emissions	7-14	
7-8	Particle Size Multiplier	7-16	
7-9	Required Data for the Preferred Method for Ongoing Waste Pile Operations and		
	Soil Removal	7-16	
7-10	Required Data for the Preferred Method for Soil Grading and Shaping	7-18	
7-11	Particle Size Multiplier	7-19	
7-12	Required Data for the Preferred Method for Agricultural Tillage	7-19	
7-13	Required Data for the Preferred Method for Building Demolition	7-21	
7-14	Constants for Equation	7-22	
7-15	Required Data for the Preferred Method for Unpaved Road Traffic	7-23	
7-16	Required Data for Preferred Surface Water Evaporation Method	7-25	
7-17	Required Data for Mass Balance Surface Water Evaporation Method	7-27	
7-18	Required Data for Preferred Wet-Cooling Tower Method	7-30	
7-19	Required Data for the Alternative AP-42 Cooling Tower Method	7-32	
7-20	Required Data for Evapotranspiration Preferred-Saturated Soils Method	7-34	
7-21	Required Data for Subsurface Transpiration Preferred Method	7-36	
7-22	Required Data for Preferred Emission Estimating Method for Buildings		
	and Equipment	7-38	

LIST OF FIGURES

Figure		Page
7-1	Example of Typical Liquid Storage Tank Configuration	7-4
7-2	Wind Speed Patterns and Associated Surface Areas	7-9
7-3	Example Wind Speed Data	. 7-12
7-4	Evaporation/Condensation Cycle for Surface Water Impoundments	. 7-27
7-5	Cooling Tower Emission Points	. 7-29

ACRONYM LIST

Ac	Actinium
ac	Acre
AIRDOS	Atmopsheric dispersion model
Am	Americium
AP-42	EPA Compilation of Air Pollutant Emission Factors document
Ar	Argon
ASER	Annual Site Environmental Report
Ba	Barium
Bi	Bismuth
BNL	Brookhaven National Laboratory
Bq	Becquerel
C	Carbon
CE	Control Efficiency
Ce	Cerium
cfm	Cubic feet per minute
CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act (a.k.a.
	Superfund)
CFR	Code of Federal Regulations
Ci	Curie
CID	DOEs Central Internet Database
CLEC	Concentration Levels for Environmental Compliance
cm	Centimeter
Cm	Curium
Co	Cobalt
CO_2	Carbon Dioxide
COMPLY	Atmospheric dispersion model
cpm	Counts per minute
Čr	Chromium
Ċs	Cesium
D	Day
DOA	US Department of Agriculture
DOC	U S Department of Commerce
DOE	U.S. Department of Energy
dpm	Disintegrations per minute
EDE	Effective Dose Equivalent
EERF	Eastern Environmental Radiation Facility
EIIP	Emissions Inventory Improvement Program
EM	Environmental Management
EPA	US Environmental Protection Agency
ERD	Environmental Radiation Data
Eu	Europium
F	Fahrenheit
Fe	Iron
σ	Gram
GENII	Dose assessment code

Н	Hydrogen
H ₂ O	Water
He	Helium
HEPA	High Efficiency Particulate Air
Hg	Mercury
hr	Hour
HT	Tritiated Hydrogen Gas
НТО	Tritiated Water
Ι	Iodine
in	Inch
ISC	Industrial Source Complex atmospheric dispersion model
Κ	Potassium
kg	Kilogram
Kr	Krypton
L	Liter
LANL	Los Alamos National Laboratory
lb	Pound
LBNL	Lawrence Berkeley National Laboratory
LLD	Lower Limit of Detection
LLNL	Lawrence Livermore National Laboratory
m	Meter
m^2	Square Meter
m ³	Cubic Meter
MDA	Minimum detectable activity
MDC	Minimum detectable concentration
MEOI	Maximally Effected Offsite Individual
Mg	Megagram
min	Minute
mm	Millimeter
Mn	Manganese
mnh	Miles per hour
mrem	Millirem
MRI	Midwest Research Institute
N	Nitrogen
IN NoI	Sodium Iodido
NADEI	National Air and Radiation Environmental Laboratory
NAKEL	National All and Kadiation Environmental Laboratory
NCDD	Notional Council on Production Protection
NCKP	National Council on Radiation Projection
	Not Determined
NESHAPS	National Emissions Standards for Hazardous Air Pollutants
NEWIKII	I ritium model
NL	Not Listed
NPL	National Priority List
NRC	U.S. Nuclear Regulatory Commission
NSPS	New Source Performance Standards
NTLF	National Tritium Labeling Facility
NTS	Nevada Test Site

0	Oxygen
OAQPS	Office of Air Quality Planning and Standards
ORNL	Oak Ridge National Laboratory
ORP	Office of Radiation Programs
ORR	Oak Ridge Reservation
OSWER	U.S. EPA Office of Solid Waste and Emergency Response
р	Pico
P&ID	Piping & Instrumentation Diagram
Ра	Protactinium
Pb	Lead
PDF	Portable Document File
PEF	Particulate Emission Factor
Pm	Promethium
PM	Particulate Matter
PM_{10}	Particulate Matter up to 10 Micrometers in Size
Po	Polonium
ppm	Parts per million
Pr	Praseodymium
Pu	Plutonium
0A	Quality Assurance
OAP	Quality Assurance Plan
QC.	Quality Control
Ra	Radium
RESRAD	Model to estimate emissions of tritium and C-14 in soil
REFTS	Rocky Flats Environmental Technology Site
Rh	Rhodium
Rn	Radon
Ru	Ruthenium
RWEO	Revised Wind Frosion Equation
R W LQ	Second
s Sh	Antimony
Sm	Samarium
SIII	Standard Operating Procedure
SOF Sr	Standard Operating Flocedure
SI	Sublituill Soil Sereening Level
SSL	Soli Screening Level
TANK5	VOC emission model for liquid storage tanks
lc	Technicium
Te	Tellurium
Th	Thorium
tpy	Tons per year
TSDF	Treatment, Storage, and Disposal Facility
TSP	Total Suspended Particulates
U	Uranium
UFOTRI	Tritium fate and transport code
VMT	Vehicle Miles Traveled

VOC	Volatile Organic Compound
Xe	Xenon
Y	Yttrium
yr	Year
Zn	Zinc
ZnS	Zinc Sulfide
Zr	Zirconium
μg	Microgram

EXECUTIVE SUMMARY

On December 15, 1989, the U.S. Environmental Protection Agency (EPA) promulgated national emission standards for radionuclides (see 54 *FR* 51654 [EPA89b,c]), which are codified in 40 *CFR* 61, Radionuclides National Emission Standards for Hazardous Air Pollutants (NESHAPs). Subpart H to Part 61 addresses emissions of radionuclides other than radon from U.S. Department of Energy (DOE) facilities. Specifically, Subsection 61.92 states:

Emissions of radionuclides to the ambient air from Department of Energy facilities shall not exceed those amounts that would cause any member of the public to receive in a year an effective dose equivalent of 10 mrem/yr.

The DOE administers a number of facilities to perform various activities, including research and development in nuclear energy and weapons, uranium enrichment or plutonium production for nuclear weapons and reactors, and processing, storing, and disposing of nuclear waste. Most of the 38 major DOE sites generate and/or manage radioactive material, and therefore, potentially could emit radionuclides into the air.

The EPA, in consultation with the DOE, has defined a diffuse source to be a non-discrete and non-well-defined location or area from which radioactive air emissions (i.e., radionuclides) originate. Diffuse sources are not actively ventilated or exhausted. In other words, diffuse emissions that are captured, treated, monitored, and emitted via a stack should be considered a point source, and not included in diffuse sources. Diffuse sources include soils, surface water/evaporation, buildings, tank venting, equipment venting and releases from underground testing.

Fugitive emissions consist of all air releases not released through a confined air stream and may include both point and diffuse sources. This document is concerned with fugitive emissions from diffuse sources. Examples of fugitive emissions include evaporative losses from a leaking seal during re-entry drilling following an underground nuclear test and wind blown dust from storage piles. Subpart H to 40 *CFR* 61 provides guidance on monitoring, test procedures, and calculation of effective dose equivalents (EDEs) for emissions from point sources, but does not provide similar guidance for radionuclide emissions from diffuse sources. However, the EPA and the DOE agree that the dose standard of 40 *CFR* Part 61, Subpart H applies to emissions from diffuse sources, such as evaporation ponds, breathing of buildings, and contaminated soil.

The EPA has worked with the DOE to develop this report to facilitate improved estimation of diffuse source emissions. The DOE will collect data on diffuse emission sources and provide this information to the EPA. Data from environmental measurements and other appropriate methods may be used to evaluate diffuse source emissions and to verify compliance with the Subpart H standard enabling regulations. The DOE should provide its methodology for assessing diffuse sources to the appropriate EPA regional office for EPA review before use.

Data on diffuse sources and the results of analyses will be reported as part of the DOE's Annual Site Emissions Report (ASER) to the EPA.

Generally, each DOE site may use their own approach for the same or a similar diffuse source. The purpose of this document is to provide technical guidance to the EPA and to the DOE in identifying generic methods for estimating annual air emissions of radionuclides from diffuse sources and transfer knowledge and experience gained at one site to another.

Emission Sources

Sources may be intermittent or continuous in nature. The mechanisms, both man-made (i.e., anthropomorphic) and naturally occurring, responsible for the generation of airborne radioactive contaminants, will vary from source to source, and some types of releases may be mitigated by man-made or natural processes. Fugitive radionuclide emissions from diffuse sources can take a number of physical and chemical forms, depending on the release mechanisms and mitigating factors. For example, emissions can range from aerosols, to airborne particulate matter from fields with attached radionuclide particles, to gaseous vapor emissions or tritium from ponds or leaking pumps. Additionally, a variety of different radionuclides may be emitted (e.g., H-3, Mn-54, Co-60, Sr-90, Cs-134, Cs-137, Ce-144, Pu-239, Pu-241, U-238, and Th-234).

Various sources of information can be used to characterize the quantity of different radionuclides released by diffuse sources. The appropriateness and availability of different kinds of information will vary from site to site and probably from facility to facility on a single site. Possible sources of information include emission rates measured at the source, results from site-specific characterization studies, routine sampling and monitoring results, process or activity related information, and default data.

Release Mechanisms

Suspension is the process of atmospheric entrainment of particles that have been deposited on the ground in some manner other than from atmospheric deposition; while *resuspension* is the process of re-injecting particulates that have been deposited on the ground from an atmospheric plume or cloud back into the atmosphere. In both cases, the entrainment process takes place by the same mechanisms, the two terms are often used interchangeably, depending on the context. Pollution studies usually refer to resuspension, while discussions of agricultural soil losses use the term suspension. Resuspension and suspension are important natural (i.e., non-anthropomorphic) factors affecting airborne dispersion of radionuclides originating from fugitive emission sources, particularly those associated with soils.

Other release mechanisms include water evaporation from ponds and lagoons, which is governed by air temperature, vapor pressure, dew point temperature, wind speed, and insolation. Complex relationships have been developed to estimate the evaporation rates for lakes and the so-called pan evaporation rates. Evaporation or volatilization also could be a significant release mechanism for radioactivity from moisture-saturated contaminated soils. Tables providing evaporation rate information have been developed to provide this information by state or climatic regions of the United States (LEE90, EPA88a) (see Chapter 5).

One type of unique release includes the release of long-lived noble gases from the Nevada Test Site (NTS). These releases occur during re-entry drilling of cavities left after underground nuclear detonations (although these have not occurred since 1992). In ground seepage, noble gases emanate out of the soil and rocks via fissures and cracks formed by the detonation.

Evapotranspiration is the loss of water to the atmosphere via the combined processes of evaporation and transpiration. Evaporation or volatilization could be a significant release mechanism of radioactivity from contaminated soils where water contaminated by tritium (i.e., tritiated water) or carbon-14 has been spilled or otherwise released. In certain instances, transpiration of plants with large root systems may also substantially contribute to tritium re-emission. The transpiration process is passive and is governed by the absolute humidity of the atmosphere and the moisture content of the soil. The rate of transpiration depends on both the soil depth profile and the plant's distribution of roots.

Modeling

Two basic approaches are used to model radiological emissions from diffuse sources and subsequent air transport of resuspended radionuclides. In the first approach, a resuspension factor is calculated from measurements of steady-state radionuclide concentrations in both air and soil. A second approach uses ambient air monitors to establish the quantities of airborne material being transported off the site, and then an air transport and dispersion model is used to calculate dispersion coefficients throughout the area of interest for unit releases from each of the resuspension sources.

The EPA documents (e.g., NESHAPs and NSPS support, *AP-42*) provide guidance for estimating the non-radioactive emissions of volatile organic compounds from wastewater treatment facilities and other sources. However, these models are not applicable to the emission of some radionuclides. There are several credible approaches in estimating tritium evaporation into the air, including calculations, measurements, and conservative assumptions. These methods are described in Chapter 5.

Field investigations of particulate emissions led to the development of emission models for a number of sources. The EPA has issued standardized guidance to support the planning of remedial action activities at National Priority List (NPL) sites (i.e., Superfund sites), including a methodology for deriving particulate emission factors. Finally, additional models are presented in documents addressing the control of open fugitive dust sources. Many of the models are reproduced in guidance documents targeting hazardous waste sites and temporary storage facilities. A number of the models have been incorporated into the Compilation of Air Pollution Emission Factors, *AP-42*, also issued by the EPA.

The basic premise for models for the release of radioactivity from soils contaminated by tritium or carbon-14 is that there must be a balance of the contaminant input and output into the atmosphere. In this case, the input is either evaporation from soil or transpiration of plants with contaminated root systems. Output includes mechanisms that deplete the atmosphere of released contaminants. These are site-dependent and may include rainfall, irrigation, runoff, and washout.

Paragraph (b)(5) of 40 *CFR* 61.93 permits the use of environmental measurements at critical receptor locations to demonstrate compliance with the standard as an alternative to atmospheric dispersion calculations, subject to prior approval of the EPA. 40 *CFR* 61.93 requires that any application to use environmental measurements at critical receptor locations should include a complete description of the sampling and analytical methodology, and show how all criteria stated in 40 *CFR* 61.93 will be met. Method 114 is the test method developed from measuring radionuclide emissions from stationary sources. The procedures recommended in Method 114 are based on the principles of measurement described in Appendix B of 40 *CFR* Part 61. The sample collection principals described in Method 114, Section 2, and the radionuclide analytical methods listed in Method 114, Section 3, can be applied to environmental measurement of many airborne radionuclides. The application is not limited to using these methods, as long as the criteria described above are met.

Conclusions

Consideration and or use of the methods described in this document to estimate emissions from diffuse sources will enhance and facilitate appropriate method selection. Similarly, and where applicable, the fugitive emission calculations contained within the latest *AP-42* guidance are most used and familiar to Agency regulators. Any application for use of these methods should describe the procedure by which these emission rates will be used as the input source for a NESHAPs compliance calculation using models.

For each release mechanism, the methodology for estimating emission rates should be tabulated, along with the current status of the procedure. Procedures included in *AP-42* are for the purpose of estimating the emission of air pollutants. Other procedures appear in an EPA guidance document on particulate emissions from TSDF (EPA89a). Still others are from EPA, NRC, and DOE documents used for various applications (EPA88a, DOE93, NRC92).

In cases of release mechanisms for which no EPA-approved models exist, alternative methods used by the NRC, DOE, or other organization may be proposed, provided they are technically justified and fully documented. The Mound Plant has received approval for a site-specific diffuse source release model (EPA97) that includes a building demolition source term after providing technical justification for the proposed approach, including a validation test of their system using the CAP88-PC code. Note that this method includes developing an airborne dispersion factor from the source to the receptor using the CAP88-PC model, and applying that factor to the release values.

To estimate the effluent radionuclide activities, it is recommended that the source combine the procedures in Table ES-1 with the sampling and calculation methods described in this report.

Mechanism	Procedure	Status			
SOILS					
Wind Erosion					
a. Limited open areas	<i>AP-42</i> method using fastest mile wind speed	Adopted by EPA (AP-42)			
b. Unlimited open areas	Modified wind erosion equation	Approved by EPA (EPA88a)			
c. Intermittent waste piles	<i>AP-42</i> method using fastest mile wind speed, modified for geometry of pile	Adopted by EPA (AP-42)			
d. Uranium ore and mill tailings	NRC Regulatory Guide 3.59 methodology	Adopted by EPA			
Material Handling					
a. Soil removal	Same as continuous waste piles	EPA guidance for TSDF			
b. Soil grading and shaping	<i>AP-42</i> emission factor for bulldozing overburden at western coal mines	EPA guidance for TSDF			
c. Agriculture	AP-42 methodology	Adopted by EPA (AP-42)			
d. Demolition	Same as continuous waste piles	Approved by EPA (EPA88a)			
e. Unpaved Roads	AP-42 methodology	Adopted by EPA (AP-42)			
f. Ongoing waste pile operations	<i>AP-42</i> aggregate handling emission factor	Adopted by EPA (AP-42)			
Contaminated Soils					
a. Tritium	Proposed based on DOE model	Proposed			
b. Carbon-14	Proposed based on DOE model	Proposed			
WATER/EVAPORATION					
a. Open ponds	Evaporation equation from NUREG-0570	Used by NRC staff			
b. Saturated soil	Same as open ponds	Based on EPA88c			
c. Subsurface soil	Superfund Exposure Assessment Manual	EPA: OSWER Directive			
d. Wet-cooling tower	Cooling loss equation	Proposed			

Table ES-1. Summary of Methods for Estimating Diffuse Source Emissions

UNDERGROUND TESTING				
a. Underground testing	Proposed air sampling protocol combined with short-term dispersion calculations	Proposed		
BUILDINGS				
a. Buildings	Proposed method based on measurement or estimated source term	Site-specific model approved for Mound Plant by EPA Region V, 1997		
EQUIPMENT				
a. Equipment venting	Same as buildings	Proposed		
TANK VENTING				
a. Tank venting	AP-42 methodology/TANKS model	Proposed		

Table ES-1. Summary of Methods for Estimating Diffuse Emissions (Continued)

DOE = U.S. Department of Energy; NRC = Nuclear Regulatory Commission; NTS = Nevada Test Site; OSWER = U.S. EPA Office of Solid Waste and Emergency Response; PM = Particulate Matter; AP-42 = Compilation of Air Pollutant Emission Factors document published by the EPA.

Chapter 1

INTRODUCTION

1.1 Background and Objectives

On December 15, 1989, the Environmental Protection Agency (EPA) promulgated national emission standards for radionuclides (see 54 FR 51654 [EPA89b,c]), which are codified in 40 *CFR* 61, Radionuclides National Emission Standards for Hazardous Air Pollutants. Subpart H to Part 61 addresses emissions of radionuclides other than radon from U.S. Department of Energy (DOE) facilities. Specifically, Subsection 61.92 states:

Emissions of radionuclides to the ambient air from Department of Energy facilities shall not exceed those amounts that would cause any member of the public to receive in a year an effective dose equivalent of 10 mrem/yr.

Fugitive emissions include releases to air that are not released through an actively ventilated air stream (e.g., monitored stack releases). Fugitive emissions from the DOE facilities include those from stacks or vents, and those from diffuse sources. A diffuse source is defined as an area source from which emissions are continuously distributed over a given area or emanate from a number of points randomly distributed over the area. Diffuse emission sources are not actively ventilated or exhausted. Examples include resuspension of dust deposited on open fields, evaporation from ponds, and ground seepage of gases following underground nuclear tests. A point source emanates from a more specific location. Fugitive emission sources can include both point and diffuse sources. This report only addresses fugitive emissions from diffuse sources. Examples of fugitive sources include evaporative losses from ponds and a leaking seal during re-entry drilling following an underground nuclear test and emissions from wind-blown dust from storage piles. Subpart H provides guidance on monitoring, test procedures, and calculation of effective dose equivalents for emissions from point sources, but does not provide guidance for fugitive radionuclide emissions from diffuse sources.

Generally, each DOE site may use their own approach for the same or a similar diffuse source. The purpose of this document is to provide technical assistance to the EPA and to the DOE in identifying generic methods for estimating annual air emissions of radionuclides from diffuse sources, and to transfer knowledge and experience gained at one site to another.

It should be noted that because of various activities, it is difficult to identify a comprehensive set of methods applicable to assess the broad range of conditions found at DOE sites. It is necessary to identify the unique conditions of each case and identify or develop the methodology that best suits the conditions. This report does not provide a definitive list of methods, but attempts to provide guidance on the types of methods to use and the factors to consider. In addition, the key

references cited in this report are available on the Internet at sites that are updated regularly. Since the methods are empirically-based and subject to change, the reader is directed to these websites for the most recent criteria. The use of default values should be carefully considered as they may not be appropriate for the site or conditions being evaluated. Whether default values or site-specific data are used, it is important to document the data source.

1.2 Structure of Report

In this report, fugitive emissions are grouped into two general categories: (1) resuspension of particulates, and (2) emissions of gases or vapors.

- Chapter 2 presents a general description and discussions of various types of diffuse emission sources. Given the broad range of conditions and sites, this chapter addresses only general considerations in identifying and characterizing such emission sources.
- Chapter 3 presents a general discussion of resuspension of particulates, including research studies aimed at understanding this phenomenon, as well as some early predictive models.
- Chapter 4 describes a few selected models for estimating particulate matter releases, along with references to the relevant sources in the literature and proposed improvements to the models. Methods that are presented in EPA guidance documents are recommended when they are applicable to emissions from DOE sites.
- Chapter 5 addresses emissions of radioactive gases and vapors, including models and methods, other than those for resuspension of particulates, currently used by some DOE sites for estimating the emissions of radioactive gases and vapors.
- Chapter 6 presents a summary of methods and general considerations in using environmental monitoring for demonstrating compliance with the NESHAP rule.
- Chapter 7 presents step-by-step example procedural guidance for calculating diffuse source emissions.
- Chapter 8 contains the references cited in the report.
- Chapter 9 presents additional supporting references that, while not cited in the report, may be helpful to the reader.
- Attachment A provides additional references for sampling diffuse source emissions.
- Attachment B lists methodologies submitted by the DOE and approved by the EPA.

Chapter 2

IDENTIFICATION AND CHARACTERIZATION OF EMISSION SOURCES

The DOE operates a number of facilities to perform various activities, including research and development in nuclear energy and weapons, uranium enrichment or plutonium production for nuclear weapons and reactors, and processing, storing, and disposing of nuclear waste. Most of the 38 major DOE sites generate and/or manage radioactive material, and therefore, potentially could emit radionuclides to the atmosphere. There are many types of potential air emission sources throughout the DOE complex, including fugitive emissions from diffuse sources.

2.1 Types of Emission Sources

As stated in the previous chapter, diffuse source emissions are derived from an area or emanate from a number of randomly distributed points over an area. Diffuse sources are not actively ventilated (e.g., by means of an in-place fan). Given the broad range of conditions and sites, this chapter addresses only general considerations in identifying and characterizing diffuse emission sources. More detailed information about emissions from DOE facilities can be found in a number of recurring DOE environmental reports (DOE97), including:

- The *Annual Site Environmental Reports* (ASERs) are a DOE directive requirement. Copies of these reports are provided to Stakeholders, including members of the public and other Federal agencies, including EPA;
- Summary Data on the Radioactive Waste, Spent Nuclear Fuel, and Contaminated Media managed by the DOE;
- Low Level Waste Performance Assessments for sites having such facilities;
- The DOE Central Internet Database (CID), located on the Internet at http://cid.em.doe.gov; and,
- Summary of Radionuclide Air Emissions from DOE Facilities, required by 40 CFR 61.49(a) and 40 CFR 190, these are included in site environmental reports submitted annually to EPA Headquarters and regional offices.

The remainder of this chapter describes properties of different emission sources that affect the type of radionuclides and the amount of radioactive material released by diffuse sources.

2.1.1 Point Sources and Diffuse Sources

Point sources are discrete emissions of radioactivity that are actively ventilated or exhausted through a single, well-defined stack or vent, where reasonably accurate quantification is achievable. Among the point sources found in the DOE complex are stacks, vents, vented tanks, and releases from discrete processes or equipment that are collected and actively exhausted to the atmosphere. In contrast, emissions from diffuse sources are not actively ventilated or exhausted and tend not to originate from a single location, but instead are released over larger non-discrete areas. Examples of diffuse sources include evaporative and aerosol emissions from landfills, spills, waste piles, and salvage yards, areas of contaminated soil, and contaminated ponds.

2.1.2 <u>Continuous and Intermittent Sources</u>

Emissions from diffuse sources, regardless of the types of sources, can either be continuous or intermittent in nature. Continuous emission sources release radioactivity at all times, although the rate of emissions may not always be constant. Examples of continuous emissions include gaseous emanations from landfills, evapotranspiration from ponds, and wind erosion of contaminated soil.

Intermittent sources are those that release radioactivity only part of the time, with periods of zero emissions. Intermittent sources of radioactivity may be due to the operation of equipment, building exhausts, or tank vents. Like continuous emission sources, the rate of emissions may vary in time, and may be significantly different from facility to facility, even for similar operations.

2.1.3 <u>Mechanisms</u>

The mechanisms responsible for the generation of airborne radioactive contaminants will vary from source to source, and include both man-made and naturally occurring mechanisms. Manmade mechanisms include, for example, surface clearing, grubbing, and grading or drilling that result in intermittent releases, road traffic, building demolition, and excavation. In contrast, wind erosion is a natural mechanism that results in an intermittent, time dependent emission source. Other dynamic natural processes that are likely to result in varying emission rates include biotic activity on soils, growth of vegetative covers, and migration of contaminants to greater soil depths.

2.1.4 Mitigating Processes

Some types of releases may be mitigated by man-made or natural processes. For example, the demolition of a building may first require that the facility be decontaminated to meet

administrative requirements. The decontamination process would remove some or all of the contaminants, thereby reducing or eliminating the release of radioactivity in the future. A natural process that might result in contaminant mitigation could be the migration of radioactive contaminants to greater soil depths due to surface water infiltration. As a result, surface soil contamination levels would decrease, yielding lower emissions rates from wind resuspension. However, should the deeper soil layers be disturbed by mechanical means, such as surface grading, the emission rates might increase, depending upon the amounts of soil exposed, the size of area involved, and the resuspension mechanisms.

2.2 Types of Radiological Emissions

Radionuclide emissions from diffuse sources can take a number of physical and radiochemical forms, depending on the release mechanisms and mitigating factors. Additionally, a variety of different radionuclides may be simultaneously emitted.

2.2.1 Chemical and Physical Forms of Radionuclide Emissions

Facilities owned and/or operated by the DOE conduct a broad range of activities and the levels of radioactivity released by diffuse sources and the distribution of radionuclides in the release is likely to vary significantly both from site to site, and among different facilities on a single site. The physical and radiochemical form of any radioactivity released by a diffuse source depends upon the mechanisms causing the release, as radioactive materials may be released as particulates, gases, or vapors, depending on the type of process and the temperature of the process. For example, particulates may be associated with radioactivity attached or incorporated in resuspended soil particles. Radioactive gases may originate from the venting of tanks, hydrolysis, or emanations from landfills. Plant processes or the degradation of materials may release vapors.

The chemical form of the radionuclides released by the diffuse sources is likely to vary significantly, depending on the source of the radionuclides. Radioisotopes for a given element can be incorporated into any number of chemical compounds or physical forms. For example, uranium can be found in a number of chemical forms, including uranium ore, yellow cake (U_3O_8) , uranium hexafluoride (UF₆), depleted uranium, and uranium dioxide (UO₂).

2.2.2 Radionuclides Emitted

Given the diverse range of activities taking place at DOE facilities, it is not possible to list all radionuclides that may be present in diffuse source fugitive emissions. Releases may include one or more of the following radionuclides: H-3, Mn-54, Co-60, Sr-90, Cs-134, Cs-137, Ce-144, Pu-239, Pu-241, U-238, and Th-234. This listing is not comprehensive, but is believed to be representative of some of the major alpha, beta, and gamma emitters contained in DOE waste or

present at contaminated sites. Table 2-l presents an aggregate distribution of radionuclides contained in waste classified by the DOE as of 1992 (DOE92a).

Some insight about the amounts of radioactive emissions may be obtained from the DOE's *Summary of Radionuclide Air Emissions from Department of Energy Facilities for CY 1998* (DOE03). Table 2-2 presents a summary of radionuclide releases during 1998 from diffuse sources emitted by production sites, research laboratories, and remedial action, storage, and disposal sites.

For sites with operating facilities, the amounts of radioactivity due to normal plant operations (i.e., point source emissions) may be significantly greater than that produced by diffuse emission sources. For sites undergoing remediation, in which most, if not all facilities have been decontaminated and decommissioned, the amounts of radioactivity released by remedial activities may be the sole source of airborne emissions.

 Table 2-1

 Radionuclide Classification and Radioactivity Distribution Using DOE Categories^a

Uranium/ Thorium		Fission Products		Activation Products		Alpha		Other	
Nuclide	Percent	Nuclide	Percent	Nuclide	Percent	Nuclide	Percent	Nuclide	Percent
Tl-208	0.0017	Co-60	0.08	Cr-51	4.95	Pu-238	2.62	Н-3	1.22
Pb-212	0.0045	Sr-90	7.77	Mn-54	38.10	Pu-239	0.20	C-14	0.06
Bi-212	0.0045	Y-90	7.77	Co-58	55.10	Pu-240	0.70	Mn-54	6.76
Po-212	0.0029	Zr-95	1.27	Fe-59	0.49	Pu-241	96.4	Co-58	6.24
Po-216	0.0045	Nb-95	2.83	Co-60	0.87	Am-241	0.004	Co-60	18.03
Ra-224	0.0045	Tc-99	0.02	Zn-65	0.19	Cm-242	0.056	Sr-90	8.48
Ra-228	0.0269	Sb-125	2.93			Cm-244	0.02	Y-90	8.48
Ac-228	0.0269	Te-125m	0.73					Tc-99	0.12
Th-228	0.0045	Ru-106	6.39					Cs-134	13.98
Th-231	0.0259	Rh-106	6.39					Cs-137	18.45
Th-232	0.273	Cs-134	0.38					Ba-137m	17.45
Th-234	33.197	Cs-137	17.31					U-238	0.73
Pa-234m	33.197	Ba-137m	16.38						
Pa-234	0.0034	Ce-144	14.67						
U-235	0.0258	Pr-144	14.67						
U-238	33.197	Pm-147	0.06						
		Sm-151	0.11						
		Eu-152	0.09						
		Eu-154	0.09						
		Eu-155	0.06						

^a Extracted from 1992 Integrated Database, Table C.5 (DOE92a). Totals may not exactly add up to 100 percent due to rounding. "Alpha" are nuclides of less than 100 nanocuries per gram (nCi/g). "Other" includes unknown radionuclide compositions or mixtures.

Table 2-2 Summary of Diffuse Source Airborne Radionuclide Emissions (in curies) at DOE Production Sites and Research Laboratories During Calendar Year 1998, Excluding Radon (unless otherwise noted)^a

		Noble		Other	
Site	Tritium	Gases	Transuranic	Radionuclides ^c	Total
DOE Production Sites and Research Laboratories					
Grand Junction Project Office	-	-	-	4.2×10 ⁻⁶	4.2×10 ⁻⁶
Los Alamos National Laboratory	-	1.9×10^{1}	-	4.6×10^{2}	4.8×10^{2}
Mound Plant	-	-	2.1×10 ⁻⁴	3.2×10 ⁻⁶	2.1×10 ⁻⁴
Pantex Plant ^b	3.9×10 ⁻²	-	-	1.8×10 ⁻⁴	3.9×10 ⁻²
Sandia National Laboratory Albuquerque	2.9×10 ⁻¹	-	-	6.2×10 ⁻⁷	2.9×10 ⁻¹
Idaho National Engineering and Environmental Laboratory	1.7×10^{2}	3.6×10 ⁻⁴	6.9×10 ⁻⁸	8.4×10 ⁻²	1.7×10^{2}
West Valley Demonstration Project	1.2×10 ⁻²	-	5.3×10 ⁻¹⁰	1.0×10 ⁻⁴	1.2×10 ⁻²
Nevada Test Site	2.9×10^{2}	-	2.4×10 ⁻¹	-	2.9×10^{2}
Laboratory for Energy-Related Health Research	4.4×10 ⁻⁹	-	3.4×10 ⁻⁹	8.8×10 ⁻⁷	8.9×10 ⁻⁷
Lawrence Livermore National Laboratory	1.1×10^{1}	-	8.9×10 ⁻⁸	9.2×10 ⁻⁶	1.1×10^1
Lawrence Livermore National Laboratory - Site 300	3.9×10 ⁻⁴	-	-	5.5×10 ⁻⁸	3.9×10 ⁻⁴
Rockwell International, Santa Susana Field Laboratory	-	-	1.6×10 ⁻⁷	9.9×10 ⁻⁵	9.9×10 ⁻⁵
Hanford Site	-	-	-	3.2×10 ⁻¹	3.2×10 ⁻¹
Rocky Flats Environmental Technology Site	-	-	1.0×10 ⁻³	7.0×10 ⁻⁵	1.1×10 ⁻³
Savannah River Site	9.3×10^{2}	-	2.0×10 ⁻³	3.2×10 ⁻²	9.3×10^{2}

Table 2-2. (Continued)					
Site	Tritium	Noble Gases	Transuranic	All Other	Total
DOE Remedial Action Storage and Disposal Sites					
Total	1.4×10^{3}	1.9×10^{1}	2.4×10 ⁻¹	4.6×10^{2}	1.9×10^{3}
Percentage	73.7%	1.0%	<0.1%	24.2%	

^a Extracted from DOE03
^b Part or all of these emissions are included in the site's normal operation releases.
^c Other radionuclides include over 100 radioactive isotopes that are not tritium, noble gases or transuranic compounds include C₁₄, Sb₁₂₅, C_{S-137}, Sr₉₀.

2.3 Characterization of Emission Sources

Various sources of information can be used to characterize the quantity of different radionuclides released by diffuse sources. The appropriateness and availability of different kinds of information will vary from site to site and probably even from facility to facility, at a single site. Possible sources of information include emission rates measured at the source, results from site-specific characterization studies, routine sampling and monitoring results, process or activity related information, and default data. Each source of information is summarized in the following subsections.

2.3.1 Emission Data Measured at the Source

Emission rates of many radionuclides can be measured at the source of the release with the appropriate effluent monitor. For example, the release of contaminants may have been characterized as part of earlier field studies by direct measurements or sample analysis. While useful, this information characterizes the emission source at a specific time in the past. If the source is intermittent, or the emission rate varies with time, the past characterization may not accurately reflect present and/or future conditions (i.e., temporal representativeness). Also, emissions from diffuse sources are likely to vary from location to location, requiring consideration of the spatial representativeness of any emission data measured at the diffuse source.

2.3.2 <u>Results from Site-Specific Characterization Studies</u>

If emissions data from the source are not available, results from site-specific characterization studies can be used to characterize radionuclide emissions. The EPA has issued guidance for the characterization of radioactivity in contaminated soils (EPA92) that identifies requirements for characterizing the radiochemical and petrographic properties of soils. The guidance addresses the following major aspects of soils:

- Soil grain distribution as a function of weight, particle size and shape, and density.
- Radioactivity and soil/contaminant relationship as a function of weight, particle size and shape, and density.
- Mineral and physical properties as a function of size fractions of the contaminants and host material (e.g., soils).
- Soil/contaminant chemical properties as a function of weight, particle size and shape, and density.

The EPA guidance uses a multi-tiered approach and presents a flow chart to use to characterize contaminants and soils.

2.3.3 <u>Routine Sampling and Monitoring Results</u>

In some instances, data may be generated during routine environmental surveillance activities when samples are periodically collected and analyzed, such as when characterizing radionuclide concentrations in water, soils, sediments, and vegetation samples. Depending upon the extent and duration of the environmental surveillance program, the data may provide information about the distribution of the contaminants in multiple environmental media, identify and characterize environmental transport mechanisms, or reveal contamination profiles as a function of time and location. This information is particularly valuable for characterizing and quantifying emissions from diffuse sources and is also of value to risk models to quantify atmospheric transport, dispersion, and transformation issues.

2.3.4 Process or Activity Related Information

Some types of releases may be characterized, at least in part, by evaluating the process or activity resulting in the emissions. For example, the amount of radioactivity could be determined from knowing the concentration of a specific radionuclide and applying factors representing the distribution of the radioactivity between specific phases of a process (e.g., liquid to gas, filtration efficiency, release fraction from waste treatment processes, resuspension factor, etc.). Alternatively, such emissions could be monitored by installing sampling equipment and monitoring each release as it occurs. Generally, both approaches may be needed because data from one or the other are likely to be incomplete.

2.3.5 Default Values

When specific information cannot be determined from available data about the emissions, site, or the process that generated the radionuclides, it may be necessary to use default values. Default values should be assessed to determine their appropriateness to the given situation and only used as a final option. For, example default rainfall values based on an average for the continental United States are inappropriate for areas of the country where rainfall totals are significantly above or below the national average. Particular attention should be given to any input parameters that are known or suspected to have a disproportionate effect on the final results. The use of default values or site-specific parameters in any regulatory model should be documented in the annual reports submitted to the EPA.

2.4 Considerations for Use

The following matters should be considered when estimating the release of radionuclides from diffuse sources.

- All radionuclides released in significant quantities should be identified and quantified.
- The chemical and physical form of the release should be identified.
- If the source is intermittent in nature, release rates and/or the frequency of releases should be measured or modeled in a way that accounts for temporal variations and provides an appropriate temporally averaged release rate.
- If the release rate of a diffuse source varies from location to location, the release rate should be measured or modeled in a way that accounts for spatial variations and provides an appropriate spatially averaged release rate.
- Mechanisms responsible for the generation and release of radioactive contaminants, both man-made and naturally occurring, should be identified and appropriately modeled.
- Mitigating processes should be considered when estimating releases from diffuse sources.

Chapter 3

AIRBORNE RESUSPENSION OF PARTICULATES

Suspension is the process of atmospheric entrainment of particles that have been deposited on the ground in some manner other than from atmospheric deposition; *resuspension* is the process of re-injecting particulates that have been deposited on the ground from an atmospheric plume or cloud back into the atmosphere. Since the entrainment in both cases uses the same mechanisms, the two terms are often used interchangeably, depending on the context. Pollution studies usually refer to resuspension, while discussions of agricultural soil losses use the term suspension. Resuspension and suspension are important natural (i.e., non-anthropomorphic) factors affecting airborne dispersion of radionuclides originating from diffuse sources, particularly those associated with soils.

Early resuspension studies involved measuring the airborne concentrations of contaminants in particulate form at some height above the ground and relating those concentrations to the putative source term (i.e., the level of contamination on the ground). The result of this analysis was a resuspension factor, the ratio of the concentration in the air to that on the ground:

K	$= C \div \sigma$	(3-1)
K	= Resuspension factor, (m^{-1})	
С	= Concentration in air, (g/m^3)	
σ	= Surface concentration, (g/m^2)	

Resuspension factors have been determined for a wide variation of natural conditions (i.e., wind erosion, biotic activity) as well as for mechanical stresses due to human activity (i.e., grading, waste pile operations). Resuspension factors due to mechanical stresses vary over more than eight orders of magnitude, while those due to wind speed alone vary over more than seven orders as noted in Table 3-1 (NIC88). Such variation aside, a resuspension factor describes a static situation.

The quantity of interest for dose modeling is typically a resuspension rate, and is therefore useless in predicting an emission rate. The resuspension rate is the ratio of the vertical flux of a contaminant to its surface concentration:

$$R = \Phi \div \sigma$$

$$R = \text{Resuspension rate, (s-1)}$$

$$\Phi = \text{Vertical flux, (g/m2 × s)}$$

$$\sigma = \text{Surface concentration, (g/m2)}$$
(3-2)

The resuspension rate is important if the transport of resuspended material from its point of origin is considered. For the purpose of estimating dose at the point of origin, the resuspension factor may be appropriate.

In principle, if R and σ are known, the emission rate, E, in Eq. 3-4 could be calculated. Since Eq. (3-2) yields:

$$\Phi = R/\sigma \tag{3-3}$$

The emission rate E may be calculated since the total emission rate is equal to the vertical flux multiplied by the area of the source:

$$E = \Phi A = RA/\sigma$$

$$E = Emission rate, (g/s)$$

$$A = Area of the source, (m2)$$
(3-4)

In reality, such a procedure has problems because of the potential variability in resuspension rates. Schmel (DOE84, Ch. 12) has reported experimental determinations of resuspension rates as a function of wind speed, particle size distribution, and surface roughness. Rate measurements are reproducible under carefully controlled conditions. However, resuspension rates observed in a single field location varied over four orders of magnitude, while other reported rates varied over almost six orders of magnitude.

In a steady state situation, the resuspension factor K (m^{-1}) and the resuspension rate R (s^{-1}) are related by the deposition velocity v (m/s) as:

R = vK(3-5)

The EPA has developed a default value for particulate deposition velocity in the NESHAPS code CAP88-PC to be 1.8×10^{-3} m/s (5.9×10^{-3} ft/s). Models using different deposition velocity values must be technically justified and approved by the EPA.

Another problem with using these mathematical relationships is inherent in the determination of σ . Radionuclide contamination of exposed soil often extends below the surface, especially in loose or disturbed soil where the contamination has weathered in and may be exponentially distributed. The contamination profile may then be subject to dynamic processes that may result in varying resuspension rates. Dynamic processes may include effects such as biotic activity on soils, growth of vegetative covers, growth of tree canopies, etc. The depth of the soil layer is a problem, since different thicknesses of the soil layer can become resuspended, depending on such factors as the degree of compaction, moisture, resuspension mechanisms, and the duration

and speed of the wind. Therefore, σ is not a uniquely determined quantity, and is difficult to empirically determine.

Problems with the emission rate may be further complicated by the use of mitigating measures to reduce the resuspension rate. In some instances, applying an agent to the ground surface is required either to reduce the amount of dust workers are exposed to or to meet environmental protection standards. Water is most commonly used as a wetting agent for this purpose. If mitigating measures are used, the following equation applies:

$$\mathbf{E} = \mathbf{\dot{R}} \times \mathbf{\sigma} \times \mathbf{A} \tag{3-6}$$

 $\hat{\mathbf{R}}$ = Mitigated resuspension rate, (s⁻¹) where the other terms are as previously defined.

3.1 Research on Resuspension

Studies on the resuspension of particulate radionuclides include those by Langer (DOE86), Nielsen, et al. (NIE90), Pettersson and Koperski (PET91), and Pinder et al. (PIN90). Other resuspension studies include those of Reeks et al. (REE88), and Nicholson and Branso (NIC90). The Fourth International Conference on Precipitation Scavenging, Dry Deposition, and Resuspension, sponsored by the DOE, dealt extensively with this and related subjects (PRU83). Earlier research on resuspension has been summarized by Sehmel (DOE84, Ch. 12). Later studies were conducted by Nicholson (NIC88), Pye (PYE87), Langer (LAN91), the NRC (NRC92), and Loosmore (LOS00).

It is difficult to estimate resuspension rates with any accuracy (DOE84, SEH80). Mechanicallyinduced resuspension rates vary over eight orders of magnitude, from 10^{-10} to 10^{-2} m⁻¹. For windcaused resuspension, rates vary over seven orders of magnitude, 10^{-10} to 10^{-3} m⁻¹. The major factors known to have a direct impact on resuspension mechanisms include weathering, and physical and chemical properties (e.g., particle chemical composition, solubility, size, shape, density, moisture contents, erodible fraction, and threshold velocity). Table 3-1 presents a summary of resuspension factors.

The work by Langer (LAN91) indicates that additional confounding factors may be present when a resuspension rate is being modeled. This work determined that a major contributor to fluctuations in measured resuspension rates was the presence of mowed vegetation coupled with rainfall. Much of the airborne radioactivity in the Langer study was from resuspension of material that was splashed onto vegetation during rainfall, and subsequently made available for resuspension by the anthropomorphic activity of mowing. These additional complicating factors are typically not part of the commonly used resuspension programs and therefore, generic models are not likely to be applicable with any precise accuracy to a specific site.

Condition	Resuspension Factor Range (m ⁻¹) ^b	Comments		
	2E-11 to 8E-09	bare soil, Y-90		
	9E-08 to 1E-07	Po-210, oxide		
	9E-08 to 5E-07	U_3O_8		
	1E-09 to IE-04	Pu in soil, time dependent model		
Wind-caused	9E-11 to 3E-04	literature review		
	2E-13 to 6E-10	NTS, Pu aerosols		
	<2E-09	Test debris 13 years after deposition		
	<5E-10	Test debris 22 years after deposition		
	4E-09 to 5E-08	Cs-137, Chernobyl		
	2E-06 to 3E-04	Pu		
Mechanically-caused	1E-05 to 1E-02	ZnS, per event		
	1E-10 to 4E-02	literature review		

 Table 3-1. Reported Resuspension Factors^a

^a Extracted from Table 6.4, NUREG/CR-5512 (NRC92).

^b Exponential notation, 2E-04 means 2.0×10^{-4} or 0.0002.

 $Cs = cesium; m = meters; NTS = Nevada Test Site; O = oxygen; Po = polonium; Pu = plutonium; U = uranium; U_3O_8 = uranium ore or yellow cake; Y = yttrium; ZnS = zinc sulfide.$

The work by Loosmore (LOS00) has examined the effect of resuspension of small particles at wind speeds that are below those generally accepted as creating resuspension. The results of this work indicate that small particles can be resuspended at low wind velocity. However, the paper indicates that the amounts are not expected to produce significant amounts of measurable resuspended activity.

Weathering and migration of contaminants to greater soil depths have the tendency to reduce resuspension factors. The reduction is primarily dependent upon the surface characteristics, the weathering processes, and the mechanism causing resuspension. Tests have shown that it is not uncommon to have resuspension factors decrease by 2 to 3 orders of magnitude over a relatively short time (e.g., typically after 30 days) due to these factors. Resuspension factors have been developed to reflect this (NRC83, ANS75). A model developed by the NRC includes an exponential time component and retains a minimal value for the resuspension factor when the exponential term vanishes to zero (NRC83, ANS75).

The NRC's proposed expression that includes this time component is:

$$K(t) = [10^{-9} + 10^{-5} \times \exp(0.6769t)]$$
(3-7)

K(t) = Time dependent resuspension factor, (m⁻¹)

= Time, (year)

t

As can be seen using equation 3-7, after about 15 years, the exponential term becomes insignificant and the resuspension factor effectively remains constant thereafter at 10^{-9} m⁻¹. This expression has been accepted as a model for estimating static resuspension values for site risk assessments. Although other expressions have been developed that include multiple exponential components, each with its own constant for specific time intervals, these alternative models are not widely accepted.

3.2 Studies of Wind Erosion

Studies of wind erosion have focused on the processes that cause erosion: (1) saltation, (2) suspension, and (3) creep. However, while new models are being developed by the U.S. Department of Agriculture (DOA) for modeling wind erosion, they primarily focus on the mechanisms of saltation and creep (see Subsection 3.2.1). As these new models do not separate out the suspension component, they are not of particular use for advancing the knowledge base associated with resuspension modeling.

3.2.1 Mechanisms of Wind Erosion

Wind erosion of soil and other finely divided materials is caused by three processes: saltation, suspension, and surface creep. Saltation refers to the movement of large particles that jump or bounce a few inches above the surface. These particles are ejected from the soil surface at a steep vertical angle, fly a short distance and then fall back down. Particles subject to saltation are generally between 0.1 and 0.5 mm (0.039 and 0.020 in.) in diameter. Upon impact, they are likely to bounce and also dislodge other particles, which may also saltate, creep, or become suspended, depending on the size of the target particle. The resulting process increases the rate of erosion as the cascade proceeds downwind.

Suspension refers to the atmospheric entrainment of smaller particles less than about 100 μ m in diameter. Such particles constitute an aerosol that can remain in the atmosphere and be carried for large distances. It is believed that wind-induced suspension is caused entirely by saltating particles. Creep refers to the sliding or rolling motion of particles greater than about 500 μ m in diameter, which are too heavy to leave the ground, but are pushed by the wind and the impact of smaller particles. Particle sizes associated with each of these mechanisms are summarized in Table 3-2.

Mechanism	Suspension	Saltation	Creep
Particle Size (µm)	< 100	100-500	500-1,000

3.2.2 Characterizing Wind Erosion Studies Prior to 2001

Earlier wind erosion studies laid the groundwork for the intensified efforts to develop methodologies to predict emissions of wind-blown particulates during the past twenty years. Smith, et al. (SMI82) reviewed but did not evaluate 15 models developed prior to 1982. Smith and Whicker (SMI83) performed a quantitative comparison of five models, using a hard-rock thorium ore stockpile as a hypothetical source. The models were judged on the basis of availability of required data and sensitivity to critical input parameters. No comparisons of model predictions with measured emissions were performed. The combined suspension model of Travis, a version of which was incorporated into the NRC codes (RAD99) Uranium Dispersion and Dosimeter Model (UDAD), Dose Coefficients from Federal Guidance Reports (FGEIS), and Calculation of Radiation Doses from Uranium Recovery Operation (MILDOS), as well as these three codes treated as a single model were found to be the most suitable ones for the particular case studied. The NRC codes can be found at http://www-rsicc.ornl.gov/nrc/NRC_codes.pdf.

Gillette (GIL83a) summarized determinations of the minimum wind stresses, expressed as threshold friction velocities, necessary to initiate wind erosion events in arid soils in the surface layer. Friction velocity is an abstract concept used to characterize the vertical wind speed profile in the planetary boundary layer of the atmosphere. A detailed explanation of friction velocity is presented by Randerson in DOE84, Ch. 5. Gillette concludes that the threshold velocity in non-crusted soils is related to the aggregate size distribution of particles on the soil surface. He also discusses the behavior of crusts on soil surfaces and the mechanisms by which such crusted soils become erodible.

Gillette and Cowherd (GIL83b) discuss the role of resuspension rates in estimating fugitive dust emissions and soil erosion, and present a simple model based on this concept. The model assumes a simple form when applied to emissions from rapidly depletable sources such as dust deposited on paved roads or piles of coal dust. In determining long-term emissions from a source with a deep layer of erodible material, such as agricultural soils, the resuspension rate concept no longer applies and a different formulation is presented. This latter model is a simplified form of the wind erosion equation developed by the DOA (DOA61), and is a forerunner of the unlimited erosion potential model discussed in Subsection 3.2.4. This DOA equation is commonly referred to as the DOA (Department of Agriculture) wind erosion equation.

An EPA study concluded that, as of 1983, no model had been validated for predicting chronic windblown particulate emissions (EPA83). The report had the most optimism about the DOA wind erosion equation, but cautioned that further work was needed to determine the input parameters that would be applicable to waste sites.

3.2.3 DOA Wind Erosion Equation

The Agricultural Research Service of the DOA has more recently developed a revised procedure for estimating annual soil loss due to wind erosion: the Revised Wind Erosion Equation (RWEQ). This equation expresses the soil loss as a function of five empirical factors, and has been implemented in the RWEQ software program described in the DOA Agricultural Research Service Technical Bulletin Number 1 (DOA98). The Revised Wind Erosion Equation combines soil losses due to the three processes of saltation, suspension, and creep.

The new wind erosion equation has been released as a software package that is available on the DOA web site (http://www.usda.gov), along with documentation for the model. The documentation for the RWEQ states that it is designed for use with erosion resulting primarily from saltation and creep processes at heights of up to two meters, and thus is not applicable to problems where transport of suspended, fine sediments above two meters is the concern. Accordingly, the RWEQ is generally not recommended for calculating offsite radiation doses to members of the public exposed to airborne resuspended radioactive dust.

When calculating transport of resuspended radioactive material, the mechanism of greatest importance is suspension. While the Revised Wind Erosion Equation does not separate out the suspension component, the RWEQ software manual (DOA98) notes that suspension represents far less mass than either saltation or creep. Soil losses from wind erosion calculated using the RWEQ software will accordingly overstate the amount of material available for airborne transport beyond distances of a few dozens of meters.

3.2.4 Open Areas

In the course of their studies of particulate emissions, Cowherd, et al. developed models for the release of fugitive dust caused by wind erosion of open areas (EPA85a, EPA88a, EPA89a). Areas are characterized as having either a limited or an unlimited wind erosion potential. An example of an area with an unlimited potential would be a smooth field, lacking vegetation, and covered with a thick layer of loose sandy soil. In such a field, relatively low wind speeds will cause suspension by the action of saltating particles, as described in Subsection 3.2.1. Because of the large reservoir of erodible particles, the erosion rate will vary as a function of the wind speed, and will not appreciably decrease with time. An example of an area with limited potential would be an heterogeneous field covered with a high density of gravel, rocks, or clumps of vegetation. In this scenario the ground is partially sheltered from the wind and from the cascade of saltating particles, so the fine particles interspersed among these non-erodible elements require higher wind speeds for the initiation of the suspension process. Once such winds occur, the supply of erodible particles is quickly exhausted and emissions stop until the area is
disturbed and a fresh supply of fine particles is brought to the surface. Disturbance of the area is not only attributable to plowing or bulldozing, but the activities of animals, freezing and thawing, and other natural weathering processes may also play a role.

A detailed procedure for determining the erosion potential of a particular area is presented in EPA88a, EPA88b, EPA 89a, and EPA90. Additional details and updates can be found on the EPA website at www.epa.gov/ttn/chief/ap42.

3.2.5 **Open Waste and Storage Piles**

The discussion in this subsection applies to open waste and storage piles, excluding uranium ore and mill tailings, which are discussed in Subsection 3.2.6.

The EPA guidance for calculating fugitive dust emissions due to wind erosion of open waste piles (EPA89a) presents methods that are identical to the methods for open aggregate storage piles described in EPA88a and reproduced in EPA90. These methods include the method used for open areas with limited erosion potential, with the additional consideration of the height and contour of the pile, as well as a separate method for continuously active piles. The methods are described in detail in EPA88a (para. 4.1.2 and para. 4.1.3), as well as in EPA89a (para. 3.2.2 and para. 3.2.3). As previously mentioned, these methods also appear in *AP-42* (EPA01, para. 13.2.5).

3.2.6 Uranium Ore and Mill Tailings

NRC Regulatory Guide 3.59 (NRC87, pp. 3.59-11/14) presents the methodology for estimating fugitive radionuclide emissions from uranium mill tailings and ore pads. This release model was validated by measurements at uranium storage sites and is therefore preferable to the generic dust release models discussed in Subsections 3.2.3, 3.2.4, and 3.2.5.

The principal parent radionuclide (U-238) in ores is assumed to be in secular equilibrium with its progenies. The following radionuclides are assumed to be in secular equilibrium with U-238: Th-234, U-234, Th-230, Ra-226, Pb-210, Bi-210 and Po-210. Radionuclides in this series with half-lives of less than five minutes, as well as Pa-234, which has a branching ratio of 0.16%, are excluded from this list. These isotopes are unlikely to pose significant health risks in comparison to the more abundant or longer-lived species. Radon (Rn-222), which is exempt from the regulations of 40 *CFR* 61, Subpart H (but included in 40 *CFR* Part 61, Subpart Q), is also excluded; however, radionuclides resulting from the decay of Rn-222 in piles that are subsequently blown offsite are not exempt and are included in the list. Regulatory Guide 3.59 mentions only U-234, Th-230, Ra-226, Pb-210, and Po-210.

3.3 EPA Soil Screening Guidance

The EPA has issued guidance for radionuclides that establishes soil screening levels (SSL) for various exposure pathways and contaminants in support of its activities at NPL sites (EPA00). The primary purpose of the SSL is to accelerate the decision-making process by determining whether a contaminated site requires further considerations under CERCLA. One of the SSL criteria provides the methodology for deriving particulate emission factors (PEF). The PEF represent an annual average emission rate for square sites of varying sizes. The PEF were derived using the air dispersion models in the ISC-2 model. The ISC-2 air dispersion model was run in both a short-term mode and a long-term mode, and the resulting value represents the air dispersion from a ground level source to a receptor located at the center of the site based on unit soil concentration (mg of resuspendable particles per kg of soil). The PEF also reflect the configuration of the site, size, receptor location, and representative meteorological data. The methodology, look-up tables, and factors for various sites are described in EPA00.

The PEF is derived as follows:

PEF =
$$(Q/C) \times 3600 \div [0.036 \times (1-G) \times (U_m/U_t)^3 \times F(x)]$$
 (3-8)

Where:

PEF	=	Particulate emission factor, (m ³ /kg)
Q/C	=	Inverse of mean concentration at the center of a $2,024 \text{ m}^2$ (0.5 acre)
		site, $(g/m^2-s \text{ per } kg/m^3)$
G	=	Fraction of vegetative cover, (unitless)
U_{m}	=	Mean annual wind speed, (m/s)
Ut	=	Equivalent threshold value of wind speed at 10 m, (m/s)
F(x)	=	Wind function dependent upon the ratio of U_m/U_t based on Cowherd
		(EPA85a)
3600	=	Seconds per hour
0.036	=	Assumed respirable fraction, (g/m ² -h)

For a 2,024 m² (0.5-acre) site, the PEF is: $1.32 \times 10^9 \text{ m}^3/\text{kg}$;

assuming the following parameters:

$$\begin{array}{rcl} Q/C &=& 90.8 \ g/m^2 \text{-s per } kg/m^3 \\ G &=& 0.5 \\ U_m &=& 4.69 \ m/s \\ U_t &=& 11.32 \ m/s \\ F(x) &=& 0.194 \end{array}$$

This method provides a way to evaluate the relative impact of site configuration, size, and receptor location on particulate emission rates. The results from this equation could be used to approximate airborne concentrations by multiplying the PEF by the average specific activity of each radionuclide. For example, assuming a soil Ra-226 specific activity of 1 pCi/g and the PEF derived above would yield an average airborne concentration based on the following equation:

$$C_{Ra} = (1/PEF) \times CF \times a \tag{3-9}$$

Where:

 $C_{Ra} = Airborne Concentration (pCi/m³)$ $PEF = Particulate Emission Factor (m³/\mu g)$ $CF = Conversion Factor (10⁻⁶ g/\mu g)$ a = Specific Activity (pCi/g) $C_{Ra} = 0.76 \mu g/m³ \times 10⁻⁶ g/\mu g \times 1 pCi/g$ $C_{Ra} = 7.6 \times 10^{-5} pCi/m³$

The implicit assumption contained in this method is that the activity concentration in the soil is identical to the activity concentration in the resuspended material. The user is strongly urged to validate this assumption in you particular application, since it has been shown to fail in some instances.

3.4 Radionuclide Concentrations in Airborne Particulate Matter

To use the models presented in this chapter for calculating the fugitive emissions of radionuclides, it is necessary to characterize the radionuclide concentrations in the emitted dust.

Two measures commonly used to characterize the soil contamination - the specific activity (pCi/g) of the bulk material in situ and the surface concentration (pCi/m^2) - are not satisfactory parameters for radionuclides. The bulk specific activity method will lead to errors for two reasons:

• Radioactive contamination is not always uniformly distributed in the soil layer. If the contamination had been deposited on the ground from an atmospheric plume or cloud, it will initially be concentrated on the surface. After a period of weathering, the activity in the underlying soil layers will increase, while decreasing at the surface. However, the fine soil or dust particles available for resuspension typically reside in a one millimeter-thick layer on the surface. Thus, the average specific activity in the top 0.15 m (six inches) of the soil (i.e., the layer that is generally sampled) will not generally be spatially representative of the suspensible soil fraction.

• As Langer found at the Rocky Flats Environmental Technology Site (RFETS), specific activity varies with particle size (LAN83). The fugitive dust, consisting predominantly of fine particles, will have a size distribution very different from that of the particles in the soil layer. Therefore, even if the bulk specific activity of the sampled soil layer did not vary with depth, this activity will generally be different from the specific activity of the resuspended particles.

Surface concentrations are usually calculated by determining the total activity of a given soil sample and then dividing by the area of the sampled surface, and are thus a measure of the average activity over the depth of the sample. The use of such a value leads to the same errors in estimating the activity of the fugitive dust as does the use of the bulk specific activity.

Another method of determining contamination values for model input involves collecting samples of the suspensible fraction from the surface of the contaminated area and determining the specific activity (pCi/g of dry weight) of each radionuclide. A simple method of accomplishing this is to collect that portion of the surface soil that passes through a 200 mesh screen upon dry sieving (EPA85a, p. 17). More sophisticated sampling devices, such as a dust collector, may also be used. One drawback of these techniques is that the process of sample collection may distort the distribution of radionuclides among the variously sized particles. Another drawback is that samples may be collected from a deeper soil layer than actually becomes resuspended. If the contamination had originally been deposited from the atmosphere, it will tend to be more concentrated on the surface, so collecting subsurface dust will dilute the sample and will usually lead to an underestimate of the emissions.

A better method of determining contamination for model input involves the use of portable wind tunnels to suspend the dust and collect samples. There is some controversy, however, as to whether such sub-scale testing develops a flow field that is indicative of what it would be in the planetary boundary layer. Langer (LAN91) showed at RFETS that the resuspension was also strongly dependent on the splashing of soil onto vegetation during rainfall, and the subsequent mowing of this vegetation. These two variables are not generally considered in any resuspension models, since typical resuspension rate measurements are performed over bare ground.

From both a theoretical and practical standpoint, the best method is to measure the specific activity of the particles collected by ambient air samplers at the location of interest for the dose modeling calculations, and adjust the model parameters accordingly. An overview of environmental monitoring techniques is presented in Chapter 6.

3.5 Considerations for Use

The preceding discussions describe the difficulty in accurately modeling radiological emissions resulting from air transport of resuspended radionuclides. Two basic approaches are typically used to address the problem, both of which use a combination of monitoring and modeling.

In the first approach, representative measurements of steady state radionuclide concentrations in both air and soil are made to calculate a resuspension factor. This resuspension factor is multiplied by a deposition velocity to calculate a resuspension rate (equation 3-5), from which an emission flux and a total emission rate can be calculated using equations 3-3 and 3-4. This emission rate can then be used as the source for a modeling code such as CAP-88. Note that the steady state resuspension factor in this method may also be calculated using equation 3-7. However, this equation is valid only for situations where the resuspension source is wind-borne from a relatively flat ground surface. Any use of equation 3-7 for calculating the resuspension factor (K) should include justification of the equation's applicability to the situation. All assumptions should be made to the conservative side to address the potential errors due to underestimation of radionuclide concentrations.

A second approach for determining emissions from resuspended dust involves using ambient air monitors to establish the quantities of airborne material being transported off the site. In this approach, an air transport model (e.g., ISC-3 or CAP-88) is first used to calculate relative atmospheric dispersion coefficients (Chi/Q) at various distances throughout the area of interest for unit releases from each of the resuspension sources. Locations for ambient air sampling stations are then selected based on these calculated relative atmospheric dispersion coefficients. The total releases from the resuspension sources can be calculated using the relative atmospheric dispersion coefficients and the results of the air sampling.

A similar procedure was used to estimate releases of plutonium-contaminated soil from the 903 pad at the Rocky Flats Plant during high wind events in 1969 and 1970 (WEB99). A wind speed-dependent suspension model was coupled with meteorological data and estimates of atmospheric dispersion using the Fugitive Dust Model (WIN90). The output from the model was the time-dependent activity release rate from the 903 pad. The FDM model is available from the National Service Center for Environmental Publications (http://www.epa.gov/nscep/).

The basic method is derived from the relationship between source strength (q) and air concentration (C):

 $C = q T_f$ (3-10)

Where T_f is the transfer function, which is essentially the concentration divided by the source term (s × m⁻³). The transfer function is a function of wind speed, atmospheric stability, and deposition, while the source strength depends on the wind speed and other factors such as soil moisture, snow cover, and degree of mechanical disturbance. Because these factors are difficult to quantify, the release rate is calibrated to the measured concentration at ambient air sampler(s). If we assume that the suspension is mainly a function of wind speed for a given set of conditions (such as moisture content, snow cover, etc.), that ambient air measurements are taken over a 24-hour period, and that meteorological data are taken hourly, then the calibrated release rate for the 24-hour sampling period is of the form:

$$q_{cal} = \frac{C_{meas} \sum_{i=1}^{24} f_i(u)}{\sum_{i=1}^{24} T_{f_i}(u, stb) f_i(u)}$$
(3-11)

Where:

q _{cal}	= the calibrated emission rate from the source (Ci/s)
C _{meas}	= the measured 24-hour average air concentration (Ci/m^3)
f(u)	= the wind speed-dependent suspension model (Ci/s)
u	= the hourly average wind speed (m/s)
$T_{f}(u,stb)$	= the transfer function (s/m^3)
Stb	= the stability class.

Note that equation 3-11 does not depend on the other factors (excluding wind speed) that affect suspension. The transfer function and measured concentration also can be specified for a given particle size fraction if the particle size fraction is measured at the sampler. The model briefly described here is similar in form to the type of model Oak Ridge National Laboratory has developed for estimating particulate emissions from dust and soil and mentioned in this report.

Users of these estimation procedures must be aware of the inherent uncertainties associated with each method and the bearing these uncertainties have on the interpretation of method results. Atmospheric dispersion models such as CAP-88 or ISC-3, upon which these estimation methods are based, contain inherent uncertainties associated with Gaussian plume models. These uncertainties propagate through to whatever calculation procedures use the model results in a predictive equation. The level of uncertainty introduced into the predictive equation results can be large, e.g., factors as high as two to ten or higher are not uncommon (MIL87). Users of the estimation methods described here should try, to the extent possible, to understand and evaluate the degree of uncertainty contained in their particular situation. By doing this, one can better determine if the predicted results are in fact conservative and providing reasonable assurances of some overprediction for compliance purposes.

As previously mentioned, resuspension and the modeling of resuspension, are very sensitive to site-specific variables. Accordingly, the specific model used in any particular application will likely be custom designed to describe the specific site and must be approved by the EPA as set forth in the regulation. At a minimum, the description of the resuspension model included in the application should include the following parameters:

- Physical description of the resuspension source(s),
- Radiological description of the resuspension source(s),

• Description of the analytical methods used to determine radionuclide concentrations in air from the sampling media.

Note: These methods must be consistent with 40 CFR Part 61 Appendix B.

- Description of the procedure to be used for calculating the offsite radiation dose from the estimated resuspension source(s),
- Validation of any special models used in the application methods, along with the prior EPA approval for any special models,
- A procedure to account for naturally occurring or other background radionuclides, and
- An onsite meteorological monitoring program for providing site-specific meteorological inputs for the models, unless other site-specific data methods have received prior EPA approval.

In addition, any application using the first approach should also include information describing:

- The justification of the method for calculating the steady state value of the resuspension factor (K);
- The method for calculating the offsite radiation dose from multiple sources if multiple sources are present;
- The selection of a deposition velocity; and
- A description of the method for calculating the surface area concentration.

Chapter 4

ANTHROPOGENIC MECHANICAL SUSPENSION OF PARTICULATES

This chapter describes models that the EPA has adopted for mechanical suspension of particulates by human (i.e., anthropogenic) means. The EPA-sponsored studies have led to the development of emission models dealing with mechanical suspension for a number of source types. An early discussion of these models appears in a report on the rapid assessment of exposure to particulates (EPA85a). The EPA has also issued standardized guidance to support the planning of remedial action activities at NPL (or Superfund) sites (EPA0093a, b), one of which provides the methodology for deriving particulate emission factors. Additional models are presented in Control of Open Fugitive Dust Sources (EPA88a). A number of these EPA-sponsored models have been incorporated into the Compilation of Air Pollution Emission Factors, *AP-42* (EPA0185b, EPA88b, EPA90, EPA95). Many of the models from the latter report (EPA95) are reproduced in the guidance document for hazardous waste TSDF (EPA89a).

4.1 Soil and Material Handling

The methodology for estimating fugitive dust emissions from soil and material handling operations is based on actual measurements taken while the activity was in progress. Many of the studies used to derive the method were performed under contract with the EPA and most of the data were collected using the exposure profiling technique.

Multipoint near-source ambient measurements are made over 90% of the effective cross-section of a plume at a location typically five meters downwind of the source. In the case of a virtual point source (e.g., a stationary activity confined to a small area), a two-dimensional array of samplers was employed, while for a line source (e.g., an unpaved road with vehicular traffic), a one-dimensional vertical array was used. Simultaneous measurements of wind velocities were made at various points to produce a wind profile, assuming a logarithmic wind speed distribution (i.e., the wind speed at height z is proportional to $\ln z$ plus a constant term). After the data were gathered, each individual concentration value was combined with the calculated wind speed at the sampler location and converted to an exposure value in units of $g/m^2 \times s$. The total mass flux from the source was determined by performing a numerical integration, spatially integrating the concentration over the effective cross-section of the plume (KIN92).

The fugitive dust emission factors cited in the following subsections, most of which appear in *AP-42*, represent the latest published information. The *AP-42* website (<u>www.epa/ttn/chief/ap42/</u>) can be checked for more recent updates to these values.

4.1.1 Soil Removal and Haulage

Cowherd, et al. (EPA89a, p. 3-5) cite an empirical formula for estimating fugitive particulate matter (PM) emissions resulting from addition or removal of materials from an open waste pile. This is the same formula that is presented in *AP-42* for aggregate handling and for continuously active storage piles (EPA88b, Section 13.2.4) as noted below:

$$E = k(0.016) \frac{\left(\frac{U}{2.2}\right)^{1.3}}{\left(\frac{M}{2}\right)^{1.4}}$$
(4-1)

Where:

- E = Emission factor (kg of PM/megagram [Mg] of Material Handled)
- k = Particle size multiplier (dimensionless)
- U = Mean wind speed, meters per second (0.6-6.7 m/s)
- M = Material moisture content (0.44-10%)

The particulate size multiplier in the equation, k, varies with aerodynamic particle size range, as follows:

Aerodynamic Particle Size	< 30 µm	$<$ 15 μ m	$<$ 10 μ m	$< 5 \ \mu m$	< 2.5 µm
k Value	0.74	0.48	0.35	0.20	0.11

This general procedure should be applied to estimating fugitive emissions when removing soil from storage piles. The movement of trucks onsite should be modeled by the emission factors for unpaved roads, as discussed in Subsection 4.2.

4.1.2 Grading and Shaping of Soil

Cowherd et al. (EPA89a, para. 5.2.1) recommend an emission factor for lift construction at hazardous waste landfills that is based on field measurements of emissions from bulldozing the overburden at western coal mines. A similar emission factor for topsoil removal by scraper [e.g., 0.029 kg per Mg (0.058 lb per ton) of topsoil removed] can be found in AP-42 (EPA01, Section 11.9.2, Table 11.9.4). This emission factor should be applied to the grading and shaping of onsite soil.

4.1.3 Agricultural Tillage and Seeding

AP-42 (EPA85b, para. 11.2.2) describes the methodology for estimating fugitive dust emissions from agricultural tilling. This method is recommended to estimate the emissions during the phase of site reclamation when soil is being prepared for seeding. Grading operations are discussed in Subsection 4.1.2. The applicable section of AP-42, 5th edition (EPA01) is Section 9.1. However, work on this section has been suspended. The methods from AP-42, 4th edition noted below are still considered applicable as of March 2004.

The quantity of dust emissions from agricultural tilling, per acre of land tilled, may be estimated using the following empirical expression:

$$E = k(5.38)(s)^{0.6}$$
 (kg/hectare) (4-2)

Where:

E = Emission factor (kg/hectare) k = Particle size multiplier (dimensionless) s = Silt content of surface soil (%)

The particle size multiplier (k) in the equation varies with aerodynamic particle size range as follows:

Aerodynamic Particle Size	Total particulate	$<$ 30 μ m	< 15 µm	$< 10 \ \mu m$	$< 5 \ \mu m$	$<$ 2.5 μ m
k Value	1.0	0.33	0.25	0.21	0.15	0.10

In the event that a site-specific value for silt content cannot be obtained, the mean value of 18 percent may be used.

4.1.4 <u>Building Demolition and Material Disposal</u>

In the absence of site-specific data for dismemberment of buildings, Cowherd et al. (EPA88a, p. 5-3), recommend the use of the materials handling equations cited in Subsection 4.1.1. The loading of the debris following demolition is modeled in EPA88a (para. 5.1.2.3) by the following equation.

Note: If a site-specific waste material load is not available, a default value of $0.45 \text{ Mg/m}^2 (0.046 \text{ tons/ft}^2)$ may be used.

$$E_d = 0.029 \times L \tag{4-3}$$

Where:

$$E_d$$
 = Total suspended particles emission, (kg/m²)
L = Waste material load per floor space unit area, (Mg/m²)
0.029 = Default average emission factor, (kg/Mg)

Any pushing operations (e.g., use of a bulldozer) related to the demolition should be modeled by the method described in Subsection 4.1.2. Default values are presented in EPA88a, para. 5.1.2.5. The emissions resulting from the onsite movement of trucks should be estimated according to the methods for unpaved roads, described in Subsection 4.2.1. EPA88a (para. 5.1.2.4) lists default values to be used if site-specific data are unavailable.

In practice, however, emission rates may be mitigated to reduce fugitive releases and limit exposures to workers or meet environmental protection standards. For example, the facility may be decontaminated before the onset of the demolition work. A temporary containment may be erected over the facility being demolished, or water may be used as a wetting agent to reduce dust loadings. Accordingly, these measures will result in lower emission rates. The emission rate can also be modified to account for the total area of the facility being demolished. Equation 4-3 is then modified to produce equation 4-4:

$$E_{dm} = 0.029 (kg/Mg) \times L \times M \times A$$
(4-4)

Where:

4.2 Vehicular Traffic on Unpaved Roads

The methodology for calculating fugitive dust emissions from vehicular traffic over unpaved roads is presented in *AP-42* (EPA88b, para. 13.2.2). Cowherd et al. (EPA88a, p. 3-4) while agreeing that the *AP-42* method is acceptable for continuous traffic, recommends using a value of zero for the number of days with measurable precipitation to arrive at a conservative estimate of annual emissions due to intermittent traffic. A good general discussion of this topic is presented in EPA88a (Ch. 3), while a similar discussion, focused on hazardous waste TSDFs, is found in EPA89a (para. 2.2).

4.3 Control Methods

The *AP-42*, 5^{th} edition (Sec. 13.2) also presents information on the use of control methods to reduce emissions rates (EPA95). Typical control methods include the use of water, chemical binders, vegetation covers, windbreaks, and enclosures. Water, as a wetting agent, is most commonly used, but the reduction in emission rates is short-lived because water only acts as a dust suppressant by forming cohesive moisture films among grains of soil. Chemical binders, however, provide longer lasting reductions. Between applications, the effectiveness of such dust suppressants decreases with increasing traffic. Other competing forces include evaporation and drainage or migration to deeper soil layers. The use of binders may create problems as it can have adverse effects on soils and plants, and can introduce other contaminants. The use of windbreaks and enclosures are relatively more expensive and their effectiveness should be evaluated for each application. Table 4-1 summarizes some of the information presented in the *AP-42*, 4th edition (EPA85b) report.

Conditions	Methods	Expected Emission Reduction
Unpaved Roads	Water Chemicals	Not Significant Some Benefit
Agricultural Soil	Vegetation & windbreaks	Not Significant
Storage Piles	Watering & chemicals	Up to 90%
Heavy Construction Watering	Twice daily	Up to 50%
Paved Roads	Watering twice per week	Up to 50%

	Table 4-1.	Summary	of AP-42	Emissions	Control I	Measures
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^a Extracted from Section 13.2 of AP-42 report (EPA85b).

4.4 Considerations for Use

It is recommended that any estimates of emissions from fugitive dust sources caused by anthropogenic activities, included in applications for EPA review, use the methods described in this chapter. Where applicable, fugitive dust emission calculations should use the latest AP-42 guidance. The EPA has received at least one application that relied primarily on the AP-42 methods - the Weldon Spring Site remediation application. In that application, the calculation of fugitive dust emission rates for each of the various activities involved in the remediation project was identified. Also, the Mound Plant received approval for site-specific methods used to calculate offsite doses using a combination of source release estimates combined with the CAP-88 model. The Mound Plant application included the results of validation testing performed on the proposed method (DOE96).

The methods described in this chapter present ways for estimating the fugitive dust emissions from various operations. Any application for use of these methods should describe the procedure by which these emission rates will be used as the input source for a NESHAP's compliance calculation using models such as CAP88-PC. This procedure should include a description of the source release rate activity, source type (i.e., stack or area), and the method used to determine the release height for the source. In addition, the procedure should include a description of the method by which any short-term or intermittent releases are modeled, particularly if the atmospheric dispersion model properties use annual averaged meteorological data. Applications should also include a description of the source and period of meteorological data used, and any tests used to validate the specific models being applied.

If the application uses the relationships from *AP-42*, the justification for the selection of the various parameters in the *AP-42* equations should be described. These descriptions should include the basis for any analogous reasoning, such as the use of surface coal mining overburden as an analog for earth moving. Any use of draft standards should also be identified.

Chapter 5

MECHANISMS OTHER THAN RESUSPENSION AND ANTHROPOGENIC MECHANICAL SUSPENSION ACTIVITIES THAT AFFECT FUGITIVE EMISSIONS

This chapter presents detailed information on the mechanisms, other than resuspension and anthropogenic mechanical suspension, that affect fugitive emissions. Resuspension has been addressed in Chapter 3 and anthropogenic mechanical suspension has been discussed in Chapter 4.

5.1 Evaporation Models

Evaporation models address evaporation from open bodies of water and from wet cooling towers. While re-evaporative release of radionuclides other than tritium from open bodies of water is not considered, radionuclide dispersion associated with wet cooling towers can be more complicated due to the formation of drift droplets and other considerations, as discussed below.

5.1.1 Open Bodies of Water

The evaporation of water from ponds and lagoons is governed by the air temperature, vapor pressure, dew point temperature, wind speed, and insolation. Complex relationships have been developed to estimate the evaporation rates for lakes and pan evaporation rates. Tables have been developed to provide evaporation rates and pan evaporation rates by state or climatic regions of the United States. Among other sources, this information is available in the Water Encyclopedia (Table 2-48 and Fig. 2-11, LEE90) and in an EPA report (Fig. 5-1, EPA88a). The Water Encyclopedia also provides the methodology to calculate lake and pan evaporation rates when site-specific data are available (Table 2-49, LEE90).

Few radionuclides are released by evaporation because most behave chemically like metals and are concentrated in the remaining water. Tritium, in the form of tritiated hydrogen gas (HT) and tritiated water (HTO), is the principal radionuclide that can be released by evaporation or volatilization from open bodies of water such as ponds and lagoons. HT is rapidly converted to HTO when it comes into contact with soil microrganisms. HTO is simply water (H₂O) with one of the hydrogen atoms (¹H) replaced by tritium, ³H. Since HTO is chemically almost indistinguishable from water (there are some very slight differences in the chemical properties of different isotopes), the most appropriate way to model its release is to assume that the water vapor emitted from the surface of a pond has the same specific activity of tritium as the water in the pond itself. The HT concentration in the air will vary depending upon absolute humidity. This directly impacts HTO concentration. The NEWTRIT model can be used to adjust the air concentration of HTO to account for different absolute humidity values.

Wing (NRC79) surveyed several evaporation models applicable to open bodies of water and compared the published experimental observations on the evaporation of water from drying trays with the model predictions. Only the equation from work by Eckert and Drake (ECK 59) yielded a measured evaporation rate that was within 10% of the experimental results. Wing then used this equation to calculate annual evaporation rates at ten different locations in the United States, using annual average values of wind speed, temperature, and relative humidity, and compared the results with measured evaporation rates. Although the model calculations were lower than the published data in nine of the cases, the worst prediction was only 47% below the actual value. Using annual average meteorological conditions, rather than using hourly meteorological data and integrating the evaporation rate over the entire year, may have contributed to the discrepancy.

Several factors that appear in the published equation were combined, while other factors, representing physical properties of air and water, have been replaced with accepted values of these properties. The result is the following formula for the evaporation rate from a circular pool:

$$E = 20.73 \times P_s \times A^{0.9} \times U^{0.8} \div T^{1.47}$$
(5-1)

Where:

Е	=	Evaporation rate of water, (g/s)
А	=	Surface area of pool, (m^2)
$\mathbf{P}_{\mathbf{s}}$	=	Equilibrium vapor pressure of water at ambient temperature, (mm Hg)
U	=	Wind speed, (m/s)
Т	=	Absolute temperature, (in °K) or
	=	Ambient temperature, (in $^{\circ}C + 273.16$)

This model assumes that the water and air are at the same temperature, ignoring the fact that evaporative cooling would tend to reduce the vapor pressure and hence the evaporation rate. The net evaporation rate is a balance of evaporation from the surface and condensation onto the surface of the open body of water from the ambient water vapor in the atmosphere. However, only the one-way process (i.e., vapor exiting the water body surface), called the surface volatilization rate, is the pathway for the release of tritium, as ambient water vapors that condense are assumed to be free of tritium. The surface volatilization rate corresponds to evaporation under zero percent ambient humidity and is conservative, because in reality some of the tritiated vapor will recondense, reducing the net flux of tritium to the atmosphere.

To calculate the emission rate of tritium, the evaporation rate is multiplied by the specific activity of tritium in the water.

$$\mathbf{R} = \mathbf{E} \times \mathbf{a} \tag{5-2}$$

Where:

R = Emission rate of tritium, (pCi/s)
 E = Evaporation rate of water, (g/s)
 a = Specific activity of tritium in water, (pCi/g)

Ideally, the annual emissions should be calculated by integrating the emission rate, using hourly average wind speeds, relative humidity, and temperatures and specific activities measured at various times during the year.

The concentration of tritium in the atmosphere is governed by the presence of airborne water vapor. There is no significant fractionation when mixing natural and tritiated water in a body of water. However, some fractionation may occur when tritiated and natural water pass across a liquid-gas interface. Because of the difference in mass, the vapor pressure of tritiated water is about 90% that of normal water at environmental conditions.

The concentration of tritium in the atmosphere (pCi/m^3) is dependent on the concentration of tritium in atmospheric water (pCi/L) and absolute humidity. This relationship is expressed as:

$$C_a = C_w \times H_a \times 10^{-3} \tag{5-3}$$

Where:

 C_a = Tritium concentration in the atmosphere, (pCi/m³) C_w = Tritium concentration in atmospheric water vapor, (pCi/L) H_a = Absolute humidity, (g/m³) 10^{-3} = Conversion factor, (L/g for water)

The concentration of tritium in atmospheric water may be obtained by sampling and analysis (NRC83), or estimated to reflect specific processes or release mechanisms. Absolute humidity values are known to vary significantly depending upon geographical locations and season of the year ranging from 3.0 to 16.5 g/m³ (1.87×10^{-4} to 1.03×10^{-3} lbs/ft³) in the continental United States (NRC83, Fig. 2.15). The NRC uses a default value of 8.0 g/m³ (5.0 lbs/ft³), when site-specific data are not available (NRC79). More information on the behavior of tritium is provided in NCRP Report No. 62, *Tritium in the Environment* (NCRP79).

Some DOE facilities, notably the NTS, calculate tritium emissions by assuming that the entire tritium activity that is discharged into an open body of water during the year evaporates during the same year (DOE92). This method is said to be conservative in that loss of tritium through ground seepage and other sinks is neglected. This would be a valid method if neither the volumes nor the specific activities of the liquid effluents varied from year to year, and if the volume of the body of water and the specific activity of the water remained constant. Both the NRC83 and the NTS methods may be used, with the more conservative result reported as the emission rate.

A slightly less conservative technique assumes 10% tritium loss due to evaporation, specified in a Brookhaven National Laboratory (BNL) 1997 environmental report. After estimating the maximum tritium inventory in their holding ponds to be 30 mCi, this method yielded an airborne fugitive release of 3 mCi (MCK97).

Another method to estimate tritium evaporation is to estimate the evaporation rate by using evaporation rates from the Climatic Atlas of the United States, published by the U.S. Department of Commerce (DOC). These data may not be current nor sufficiently site-specific. Furthermore, they represent only the net evaporation rate - their use may therefore produce an underestimate of the tritium release rate, as previously discussed.

5.1.2 <u>Wet-Cooling Towers</u>

Wet-cooling towers are heat-exchangers used to dissipate large heat loads from industrial processes. Water is used as the medium to transfer heat away from the coils that contain the process fluids. Under normal conditions, the two fluids never mix. In the event of a leak, however, the cooling fluid may become contaminated by the process fluid. Within the tower, some of the cooling fluid is drawn up as droplets by convection currents and released as drift droplets. The fine droplets are then carried downwind, and the larger droplets settle out of the air and deposit near the tower. Some towers are equipped with drift or mist eliminators to minimize such emissions.

As the water evaporates, the droplets leave behind fine particulate matter formed by the crystallization and agglomeration of dissolved solids. Dissolved solids may include minerals, chemicals from corrosion and algae inhibitors, etc. Emissions from cooling towers, therefore, might be modeled as PM₁₀ particulates (EPA95). Given that the size of the droplets vary, it also follows that the fine particulate matter formed by dissolved solids would have its own particle size distribution.

In *AP-42*, the EPA has estimated the overall PM_{10} emission rate from a typical cooling tower to be about 2.3×10^{-3} g/L (1.44×10^{-4} lb/ft³) of circulating water flow, based on limited data for induced draft cooling towers (Sect. 13.4, EPA95). However, no PM₁₀ emissions data were provided for natural draft cooling towers. The rate given by the EPA is also believed to be typical of older towers with less efficient mist eliminators.

Similarly, the Chemical Engineer's Resource Page cites total water losses from cooling towers to be the sum of drift loss and evaporation loss (CHE02). It is estimated that between 0.1 and 0.2% of the water supply is lost to drift. Evaporative loss is approximated by equation 5-4 as follows.

$$EL = 0.00085 \times WF(T_1 - T_2)$$
(5-4)

Where: $EL = Evaporative loss (m^3/hr)$ WF= Water flow rate (m^3/hr) $T_1 = Hot water temperature (^oF)$ $T_2 = Cold water temperature (^oF)$ $0.00085 = Evaporation Fraction per ^oF (reciprocal ^oF)$

The emission of radioactivity from wet-cooling towers is further complicated by the possible speciation of radioactivity in the circulating water. For example, some radionuclides, such as uranium, cesium, iodine, etc., may chemically bind with minerals or chemical inhibitors, and would thus not be available for release through evaporation. Conversely, tritium and noble gases (e.g., xenon, krypton, argon, radon, etc.), may be most efficiently dispersed by cooling towers, since by design cooling towers work as very effective aerators, allowing enhanced evaporation or vaporization of HTO. Given these various considerations, estimating release rates for radionuclides from wet-cooling towers, either by mechanically-induced draft or natural draft, may have to be addressed on a case-by-case basis.

5.2 Evapotranspiration from Contaminated Soil

Evapotranspiration is the loss of water to the atmosphere via the combined processes of evaporation and transpiration. Evaporation (or volatilization) could be a significant release mechanism for certain types of radioactivity from contaminated soils where water contaminated by tritium or carbon-14 has been spilled or otherwise released. In certain instances, transpiration of plants with large root systems may also substantially contribute to tritium re-emission. The transpiration process is passive and is governed by the humidity of the atmosphere and the moisture content of the soil. The rate of transpiration depends on both the soil depth profile and the plant's distribution of roots.

5.2.1 Evapotranspiration from Saturated Soil

Several models are available to determine the release of radionuclides from saturated soil via evapotranspiration:

• Superfund Exposure Assessment Manual: The Superfund Exposure Assessment Manual (EPA88c) recommends that spills of liquid contaminants, where liquid pools are visible on the soil surface or where the soil is saturated from the surface on down, be modeled in the same manner as open liquid storage pools. This is also the most conservative model, because models for the release of contaminants from the pore spaces in the soil predict lower release rates. Furthermore, the soil release models require data or assumptions regarding the time-dependent contaminant concentrations and depth profiles. It is therefore recommended that atmospheric releases of tritium from soils contaminated with tritiated water be modeled in the same way as pools containing tritiated water.

• *RESRAD*: Another model used to assess the amounts of radioactivity released from contaminated soils relies on the evapotranspiration rate of water. This model, developed by DOE, is used for both tritium and C-14, and is documented in the *RESRAD* computer code (DOE01, App. E and L). With respect to the effects of transpiration, *RESRAD* assumes that tritium exhaled by plants is negligible. However, for C-14, *RESRAD* assumes that plants are the sink since atmospheric ¹⁴CO₂ is incorporated by plants during photosynthesis.

The models for tritium and carbon are similar; the only difference is how the tritium and carbon flux rates are derived. The following equation applies to both tritium and C-14.

$$Ci = 0.5 \times (F_i \times \sqrt{A}) \times (3.17 \times 10^{-8}) \times (U_w/H_{mix})$$
(5-5)

Where:

Ci	= Average concentration in air over finite area, (pCi/m^3)
0.5	= Time fraction wind is blowing toward receptor, (dimensionless)
Fi	= Contaminant flux (evasion rate) from soil, (pCi/m^2-yr)
А	= Size of contaminated area, (m^2)
H _{mix}	= Mixing height within which contaminant is uniformly distributed,
	(2 m for human inhalation)
U_{w}	= Annual average wind speed, (m/s)
3.1536×10^{-8}	= Conversion factor, (yr/s)

For tritium, the flux rate, F_i, is derived as follows:

$$F_i = W_T \times E_t \tag{5-6}$$

Where:

Fi	=	Contaminant flux rate from soil, (pCi/m ² -yr)	
W_{T}	=	Tritium concentration in soil water, (pCi/m ³)	

$$E_t = C_e \times \left[(1 - C_r) \times \frac{P_r + I_r}{1000} \right]$$
(5-7)

Where:

Et	=	Evapotranspiration rate, (m/yr)
Ce	=	Evaporation coefficient, (dimensionless)
Cr	=	Runoff coefficient, (dimensionless)
Pr	=	Annual rainfall rate, (mm/yr)
Ir	=	Irrigation rate, (mm/yr)
1000	=	mm to m conversion

DOE assumes a default evaporation coefficient (C_e) of 0.5. For the runoff coefficient (C_r), values range from 0.1 to 0.4 for agricultural soils and woodlands and 0.4 to 0.65 for urban environments.

For C-14, the flux rate (F_i), is derived as follows:

$$F_i = S_C \times E_c \times \rho_b \times d_s \times 10^6$$
(5-8)

Where:

Fi	=	Contaminant flux rate from soil, (pCi/m ² -yr)
Ec	=	Evasion loss rate constant, (yr ⁻¹)
S _C	=	C-14 concentration in soil, (pCi/g)
ρ_b	=	Soil bulk density, (g/cm^3)
ds	=	Soil depth, (m)
10^{6}	=	Conversion factor, (cm^3/m^3)

The evasion loss rate, E_c , is the fraction of soil inventory lost to the atmosphere per unit time. This constant varies for soil type and can be found in Table L.2, generated by Sheppard, Amiro, and Davis (1991), of the *RESRAD* User Manual (DOE01). Additional information on the behavior of tritium and C-14 may be obtained from NCRP Reports No. 62 (Tritium in the Environment) and No. 81 (Carbon-14 in the Environment) (NCRP85, 79).

• *UFOTRI Code*: UFOTRI models tritium re-emission as the sum of a basic re-emission rate (k_b) and a conversion factor (C₁), to account for differences in HTO and H₂O behavior (RAS90). The total daytime re-emission rate is described by the following time function following tritium deposition (TAS97).

$$K_{re} = \frac{E_a}{S_W} C_1 \exp\left(-\frac{t}{T}\right) + k_b \times \exp\left(-\frac{t}{T}\right)$$
(5-9)

Where:

 K_{re} = Total daytime re-emission rate (%/hr)

- E_a = Actual evaporation rate of water from soil (kg /m² · h)
- S_w = Actual water content of the top 5-cm layer (kg/m²)
- C_1 = Constant, 1200% to describe differences of HTO and H₂O (%)
- T = Time constant (default value = 50 hrs)
- k_b = Basic re-emission rate (%/hr)
- t = time period of re-emission (hrs)

The basic re-emission rate (k_b) , was originally derived from a Canadian field experiment (Brown 1988) and has since been updated by Tashner (1996). It is the dominant mechanism acting at night, when there is no insolation, and is therefore assumed to be independent of meteorological conditions. During the day, however, the evaporation rate is included by use of the conversion factor (C₁).

The movement of the tritium to greater depth in the soil is simulated by the time function, exp(-t/T), which would reduce the rate of re-emission. The actual evaporation of water vapor from soil is calculated in the UFOTRI code by applying Monteith's equation (TAS97).

• *LBNL Two-Compartment Model*: A simplified two-compartment model for tritium transport was developed at Lawrence Berkeley National Laboratory (LBNL) for the National Tritium Labeling Facility (NTLF). This approach is established based upon observations of Murphy (1993), who showed that approximate steady-state conditions of HTO distribution exist among the aqueous phase. These observations led to the conclusion that all tritium released is assumed to be incorporated into the soil or air as tritiated water. Although it neglects any affinity of tritium for the biota or organic phases, in this case it serves as a valid approximation. The model estimates the tritiated water distribution between air and soil given a steady HTO emission to the atmosphere from the NTLF (MCK97).

Fundamental mass-balance equations were developed to describe gains and losses in the air and soil compartments. These equations are used to solve for the steady-state inventory in each compartment. The solution used to determine the inventory of tritium in the air is:

$$N_{a} = \frac{S}{\left[L - T_{sa} \times \frac{T_{as}}{L_{s}}\right]}$$
(5-10)

Where:

 N_a = Tritium inventory in the air compartment, (Bq)

- S = HTO input rate into the air compartment, (Bq/d)
- L_a = Sum of all loss-rate constants from the air compartment, (d⁻¹)
- T_{sa} = Soil to air transfer rate (i.e., evapotranspiration), (d⁻¹)
- $T_{as} = Rain water washout from air to surface soil, (d⁻¹)$
- $L_s =$ Sum of soil compartment transfer-rate constants, (d⁻¹)

Appendix A of the Environmental Health-Risk Assessment for Tritium Release (MCK97) provides equations to describe the rate of transfer and loss for each compartment needed for the steady-state inventory assessment.

• *NEWTRIT*: *NEWTRIT* has recently been developed as a proposed alternative regulatory model for compliance, which has been submitted for EPA approval and is currently being evaluated and has yet to be approved. Unlike the currently approved EPA models for tritium transport (CAP-88, COMPLY and AIRDOS-PC), *NEWTRIT* addresses dose arising from tritiated hydrogen gas (HT) emissions as well as from HTO. The code is based on experimental data that are used to generate tritium-to-hydrogen ratios within each environmental compartment. This allows tritiated water concentrations in air and plants to be estimated from the tritium gas concentration in air (PET01).

Unlike the two-compartment model described previously, *NEWTRIT* does not assume that the concentration of tritium in the water phase of all compartments is the same as that of air moisture. By more accurately describing the form of tritium within the soil, *NEWTRIT* can better predict tritium re-emission to air.

The EPA and LLNL discussed the approval process of *NEWTRIT* to accommodate various forms of tritium. It was mutually decided that *NEWTRIT* code will be included into GENII and then undergo a peer review process for including this as a compliance model.

5.2.2 Subsurface Contamination of Tritium

In cases where the surface layer of the soil is dry and devoid of tritium, but tritiated water remains below the surface, Eq. 2-3 from EPA88c can be used to calculate a more realistic release rate than that produced by the surface evaporation model. A default value of the diffusion coefficient of water vapor in air, required in the equation, has been developed (i.e., 0.2 cm²/s) if site-specific data are not readily available. The soil porosity and the saturation vapor concentration should be determined on a site-specific basis in order to obtain the most accurate estimate. The tritium emission rate may be calculated as follows:

$$E_i = D_i \times C_{si} \times A \times [P_t^{4/3} \times (M_i \div d_{sc})]$$
(5-11)

Where:

$$\begin{array}{rcl} P_t &=& Total \ soil \ porosity, \ (dimensionless) \\ P_t &=& 1 \cdot (b \div r) & (5-12) \\ & b = Soil \ bulk \ density, \ (g/cm^3) & (5-12) \\ & r = Particle \ density, \ (g/cm^3) & (5-12) \\ C_{si} &=& Particle \ density, \ (g/cm^3) & (5-13) \\ C_{si} &=& P \times MW_i \div R \times T & (5-13) \\ P &=& Vapor \ pressure \ of \ contaminant, \ (mm \ Hg) & MW_i &=& Molecular \ weight \ of \ contaminant, \ (g/mole) & R &=& Molar \ gas \ constant, \ (62,361 \ mmHg-cm^3/mole-K) \\ T &=& Absolute \ temperature, \ (K) & (K$$

At times the contaminated transpirational stream, from soil through vegetation leaves, should be recognized and treated as a diffuse source of tritium to the atmosphere. A specific instance demonstrating the importance of the transpirational stream is a large pine tree growing in an area of known tritium contamination at the LLNL. In 1994, this pine tree accounted for the largest amount of tritium released to the atmosphere in the vicinity (LLNL NESHAPs Report 1995).

LLNL estimated the tritium transpiration rate based on the projected area of 85.9 m^2 (924.6 ft²), the collective transpiration rate of 206 L/d (54.4 gal/d), and the highest quarterly measured concentration of tritium in transpired water. The estimated tritium transpiration rate from the tree was then used as input data into CAP88-PC.

5.3 Gaseous and Other Types of Emissions

Although the United States has not performed underground nuclear testing since 1992, conceivably, fugitive emissions in the form of radioactive noble gasses would ensue following underground detonation. Radionuclides were released at the NTS during re-entry drilling and by ground seepage of noble gases following underground nuclear detonations prior to 1992. Both of these release mechanisms are discussed in the following subsections.

5.3.1 <u>Re-Entry Drilling</u>

Within one to two days of an underground nuclear test, a hole is drilled into the hollow chamber created by the explosion to sample the non-fissioned material and determine the fission yield. During this process, called drillback, radioactive halogens in gaseous form (principally I-131) and noble gases (Xe-133 and Kr-85) are sometimes released. Although emanating from a small area (i.e., a virtual point source), these releases are uncontrolled and not directly monitored.

Appendices 1 and 2 in DOE92 describe the drillback operations conducted by the LLNL and Los Alamos National Laboratory (LANL). LLNL used measurements of the radiation field in the

vicinity of the drill pipe and ambient air samples to estimate the effluent activity. This estimate was then verified by the alternative method of measuring radionuclide concentrations in downwind air samples and using local wind data to calculate the release rate.

LANL, which used a different drillback system, sampled the ambient air in the work area on top of the drillback platform (LAN92). If leakage of radioactive gases was suspected, samples were also taken from the cellar, the subsurface excavation housing the containment equipment. Data collected during the LOCKNEY drillback, at which time a large amount of activity was released, were used to derive a procedure for inferring effluent activities from air sampling measurements. LANL estimated that the releases calculated by this procedure are within a factor of three of the actual amounts for modest releases, and within an order of magnitude for small ones.

5.3.2 Ground Seepage of Noble Gases

Prior to 1992, seepage of radioactive noble gases was sometimes observed in the Pahute Mesa test area beginning a week or more after an underground nuclear explosion. An analytical model to explain and quantify this seepage has been developed (NIL91, BUR89).

According to their understanding of the release mechanisms, the collapse of the cavity created by a nuclear detonation creates a rubblized zone, called the chimney, immediately above the cavity. If the volcanic rock above the chimney contains fractures, radioactive noble gases can seep to the surface. Normal cyclical changes in barometric pressure cause the atmosphere to act as a piston, driving air into the fractures or drawing out gases contained in these fractures. The rock thus breathes, inhaling air and exhaling gaseous radionuclides. However, the observed seepage is inconsistent with the theorized mechanism and the phenomenon is not yet fully understood. The purpose of the model is to quantify the releases due to this natural mechanism and thus ascertain the integrity of the containment, in compliance with regulatory standards and international treaties.

There have been few data collected for the purpose of verifying this conceptual model and its mathematical expressions. However, in the early 1990s field measurements were conducted at two sites on Pahute Mesa by injecting tracer gases 300-400 m (984-1,312 ft) beneath the surface into the rubblized chimney. Gas samples were extracted from shallow collection holes to monitor the tracer gas arrival. Data were obtained concerning transit time, dilution, areal distribution, and total amount of contaminant transport by barometric pumping and are summarized in the 1994 DOE report, "Field Measurement of Tracer Gas Transport by Barometric Pumping" (LAG94).

It should be noted that the seepage of noble gases may best be characterized by sampling, followed by analysis. Some of the major limitations in conducting this type of sampling include the proper selection of sampling locations, orientation of the samplers to reflect local atmospheric dispersion and transport, effect of terrain on dispersion in areas of complex terrain,

distances from seepage points to sampling locations, and integration of the results over the area being evaluated.

5.3.3 Emissions from Buildings

Fugitive emissions from buildings may occur through vents and stacks that are not actively ventilated. The mechanisms leading to such releases may be induced (e.g., convection fans) or occur naturally via pressure differences. In simple terms, emissions can be estimated by determining the volume of material (e.g., air, gas, vapor, etc.) released, its concentration, and application of a mitigation factor. The expression is:

$$E_b = \sum_{i}^{n} R_i \times T_i \times C_i \times M$$
(5-14)

Where:

E_b	=	Sum of all releases over all events i, (Ci)
R _i	=	Release rate for event i, (m^3/s)
Ti	=	Duration of release i, (s/event)
Ci	=	Concentration of contaminants for event i, (Ci/m ³)
M_i	=	Mitigation factor, (dimensionless)

For puff releases, the above expression is reduced to:

$$E_p = \sum_{i}^{n} V_i \times C_i \times M_i$$
(5-15)

Where:

The mitigation factor may be used to account for the use of devices or processes that reduce the amount of materials released. Such devices may include HEPA filters, baghouses, scrubbers, adsorber beds, etc.

If the release is monitored downstream of such devices to determine actual concentrations, the mitigation factor is set equal to one.

5.3.4 Emissions from Tank Venting

Emissions from organic liquid storage tanks occur because of evaporative loss of the liquid in storage and as a result of changes in the liquid level. The emission sources vary with tank design, as does the relative contribution of each type of emission source. Emissions from fixed roof tanks are a result of evaporative losses during storage (known as breathing losses or standing storage losses) and evaporative losses during filling and emptying operations (known as working losses). External and internal floating roof tanks are emission sources because of evaporative losses that occur during standing storage and withdrawal of liquid from the tank. Standing storage losses are a result of evaporative losses through rim seals, deck fittings, and/or deck seams.

Working losses from tanks may be estimated by determining the displaced volume of the overhead space above a liquid. As before, the emission takes into account the concentration of the contaminants, partition factor between the liquid and gaseous phases, and application of a mitigation factor, if warranted. The expression is:

$$E_t = \sum_{i}^{n} V_i \times C_i \times P_i \times M_i$$
(5-16)

Where:

Et	=	Sum of all releases over all venting events i, (Ci)
Vi	=	Volume released for each venting i, as displaced by the amount of
		liquid added to the tank, (m^3) .
Ci	=	Concentration of contaminants for event i, (Ci/m ³)
Pi	=	Partition factor for each contaminant, (dimensionless)
Mi	=	Mitigation factor, (dimensionless)

Note: V_i cannot exceed the capacity of the tank.

The partition factor may vary depending upon the contaminants, typically assigned a value of one for noble gases and less than one for contaminants that are miscible or soluble in the liquid phase. The mitigation factor may be used to account for the use of devices or processes that reduce the amount of materials released. Such devices may include filters, adsorber beds, traps, etc.

For tanks holding gaseous contaminants only, the above expression is redefined in terms of the gas volume released:

$$E_{g} = \sum_{i}^{n} V_{i} \times C_{i} \times P_{i} \times M_{i}$$
(5-17)

$\mathbf{V}_{\mathbf{i}}$	=	Gas volume (m ³), released in each event i, adjusted to normal temperature
		and pressure or as measured during each release
Ci	=	Concentration of contaminants for event i, (Ci/m ³)
Pi	=	Partition factor for each contaminant, (dimensionless)
M_i	=	Mitigation factor, (dimensionless)

As with tanks that also contain liquids, the partition factor is set to equal one for noble gases and less than one for vapors or reactive gases, which may plate-out in tanks. The mitigation factor may be used to account for the use of devices or processes that reduce the amounts of materials released. Such devices may include filters, adsorber beds, traps, etc. If the release is monitored downstream of such devices to determine actual concentrations, the mitigation and partition factors are each set to equal one.

Tank breathing or standing storage losses occur due to diurnal variations in ambient temperature. During evenings with clear skies, when the earth loses heat due to long-wave radiation, most vapors found in storage tanks contract as temperatures cool. When temperatures increase during daylight hours, due to insolation, these gaseous compounds may be emitted into the atmosphere as they expand with the increasing ambient temperature. In this version of this guidance document, the following section on tank breathing losses was added to insure that all tank venting emission sources are considered.

Tank breathing losses can be estimated by using the following equation:

$$L_{\rm S} = 365 \, V_{\rm v} \times W_{\rm v} \times K_{\rm E} \times K_{\rm S} \times C_{\rm i} \tag{5-18}$$

Where:

For detailed instructions on how to derive V_V , W_V , K_E , and K_S , refer to Section 7.1 of AP-42.

The EPA OAQPS developed and maintains a model to estimate VOC emissions from chemical storage tanks, which is derived from these *AP-42* equations. The TANKS program is designed to estimate air emissions from organic liquids in storage tanks. The model and supporting

documentation can be downloaded at <u>http://www.epa.gov/ttn/chief/software/tanks/index.html.</u> TANKS allows users to enter specific information about a storage tank (i.e., dimensions, construction, paint condition, etc.), the liquid contents (chemical components and liquid temperature), and the location of the tank (i.e., nearest city, ambient temperature, etc.), and generate an air emissions report. The report features include estimates of monthly, annual, or partial year emissions for each chemical or mixture of chemicals stored in the tank. The model accounts for both working and breathing losses. If it is assumed that the radionuclide concentration of the chemicals stored in the tank is similar to the radionuclide concentration of the vapors emitted, then the following equation can be used to adjust the output from the TANKS model to estimate radionuclide emissions.

$$\mathbf{L} = \mathbf{E} \times \mathbf{a} \tag{5-19}$$

Where:

L = Total storage tank emission (Ci/yr)

E = VOC emission output from TANKS model (lb/yr)

A = Specific activity of stored chemicals (Ci/lb)

5.3.5 <u>Emissions from Equipment</u>

Emissions can also be associated with equipment used to process radioactive materials. Equipment emissions emanate directly to the environment (e.g., equipment sitting on a pad or covered patio). The emissions may be associated with built-in system features (e.g., filtration systems) or inherent in the process (e.g., air displaced by a waste compactor ram). The mechanisms leading to such releases are similar to the model presented in subsection 5.3.3. As before, the expression is:

$$E_{p} = \sum_{i}^{n} R_{i} \times T_{i} \times C_{i} \times M_{i}$$
(5-20)

Where:

 $\begin{array}{l} E_p = \text{Sum of all releases over all processes i, (Ci)} \\ R_i = \text{Release rate for process i, (m^3/s)} \\ T_i = \text{Duration of release i, (s/process)} \\ C_i = \text{Concentration of contaminants for process i, (Ci/m^3)} \\ M_i = \text{Mitigation factor, (dimensionless)} \end{array}$

The mitigation factor may be used to account for the use of devices or processes that reduce the amount of materials released. Such devices may include HEPA filters, baghouses, scrubbers, adsorber beds, etc. If the release is monitored downstream of such devices to determine actual concentrations, the mitigation factor is set equal to one.

5.4 Considerations for Applications

The following subsections discuss how to apply the previously introduced theory at a particular site.

5.4.1 <u>Evaporation</u>

There are several credible approaches in estimating tritium evaporation into the air, including calculations, measurements, and conservative assumptions.

Implementation of the calculation approach requires knowledge of parameters that are specific to the site. The evaporation rate of water should be determined using equation 5-1. This in turn, is dependent on the local wind speed, ambient temperature, and surface area of the contaminated body of water. Additionally, the specific activity of the tritium in the contaminated water would be required to assess tritium emissions.

The concentration of tritium in atmospheric water may be sampled and correlated to atmospheric tritium through knowledge of the site-specific absolute humidity as described in basic equation 5-3.

If the volume of a pool remains fairly constant, it is safe to assume that the entire tritium activity discharged into the pool throughout the year will evaporate within the same year. If this were not the case, it would still be appropriate to assume a fixed percent of tritium is lost to evaporation. This, however, requires educated assumptions regarding the specific situation.

5.4.2 Evapotranspiration from Soil

Models for the release of radioactivity from soils contaminated by tritium or C-14 have been outlined in section 5.2.1. The basic premise for all these models is that there must be a balance of the contaminant input and output to the atmosphere. In this case, the input is either evaporation from soil or transpiration of plants with contaminated root systems.

Characterizing the input of soil evaporation requires knowledge of the concentration of tritium or C-14 in the soil, the evaporation rate of water from the soil, as well as the size of the contaminated soil zone. Measurements of soil porosity and depth of soil cover are needed in the case of subsurface contamination.

Transpiration also contributes input of tritium into the atmosphere. The transpirational process is largely passive and will depend on the local absolute humidity, moisture content of the soil, and the projected surface area of the plant. It will also be necessary to measure the concentration of contaminant (i.e., tritium) in the transpired water to fully characterize the atmospheric release.

Although it may be conservative to not consider output factors, mechanisms that deplete the atmosphere of released contaminants may also be considered. These are site dependent and may include rainfall, irrigation, runoff, and washout.

Another important feature is the manner in which differences between HTO and water in the evapotranspirational stream are accounted for. Although simplified, a valid approach is to assume no significant differences exist in the evaporation of water and HTO. Empirical correction factors, or constants, may also be used. These have evolved over the years to best correlate with experiments attempting to quantify this difference.

5.4.3 Gaseous and Other Types of Emissions

The case of determining gaseous emissions following underground nuclear testing is best approached through confirmatory measurement rather than through calculation alone. Although analytical models exist to estimate emissions following underground testing, some aspects of this science are not thoroughly understood. Sampling is the most suitable means for characterizing the atmospheric release. However, the selection of proper sampling locations as well as the orientation of the samplers is difficult when sampling radioactive noble gas release in the case of seepage. For meaningful results, the local atmospheric transport and dispersion must be considered in order to best position the samplers. This is further confounded in areas with complex terrain where airflow trajectory reversals are common.

Gaseous emissions from buildings, tanks, and equipment can be estimated with some basic knowledge of the release mechanism. For each event (i.e. gas vent, equipment use, etc.), the following must be known:

- The contaminant concentration of the process either by release rate and time, or the volume released by displacement.
- The amount released from the tank or building during the process.
- Any mitigation factors that may reduce the amount of material released.

Chapter 6

GUIDANCE ON ENVIRONMENTAL MONITORING PROGRAMS TO DEMONSTRATE COMPLIANCE WITH DOE NESHAPS

6.1 Summary of NESHAPS Requirements

Paragraph (b)(5) of 40 *CFR* 61.93 permits that the use of environmental measurements at critical receptor locations as an alternative to air dispersion calculations, and is subject to prior approval of the EPA (EPA89a). Applications for approval should:

- 1) Include a detailed description of the sampling and analytical methodology, and
- 2) Show how the following criteria will be met:
 - Measurements shall be made at locations of the critical receptor (e.g., maximally effected offsite individual [MEOI]).
 - The air at the point of measurement shall be continuously sampled for the collection of radionuclides.
 - The radionuclides released are the major contributors to the effective dose equivalent (EDE).
 - Radionuclide concentrations that would cause an effective dose equivalent greater than or equal to 10% of the standard shall be readily detectable and distinguishable from background.
 - A QA program shall be conducted that meets the requirements described in Appendix B, Method 114, as noted in 40 *CFR* 61.93(b)(5)(v).

6.2 Sampling and Analytical Methodology

Paragraph (b)(5)(vi) of 40 *CFR* 61.93 requires that any application to use environmental measurements at critical receptor locations include a complete description of the sampling and analytical methodology, and show how the above criteria will be met. Method 114 is the test method developed for measuring radionuclide emissions from stationary sources. The procedures recommended in Method 114 are based on the principles of measurement described in Appendix B of 40 *CFR* Part 61. The stack monitoring and sample collection principles described in Method 114, Section 2, and the radionuclide analytical methods listed in Method 114, Section 3, can be applied to environmental measurement of many airborne radionuclides. The application is not limited to using these methods, as long as the criteria described above are met.

Table 6-1 lists the half-lives and modes of decay of the principal radionuclides which are released at DOE facilities and identifies the physical state of each. Consideration of these

physical parameters is necessary to establish whether environmental monitoring for determining compliance will be feasible.

6.2.1 <u>Radionuclides as Particulates</u>

The radionuclides of greatest concern at many DOE facilities, often U-234 and/or U-238, are emitted as particulates. To sample particulates, air is pulled through a HEPA filter using a calibrated high-volume air sampler. The sampling rates (i.e., volume of air per unit of time) should be recorded periodically, and the total volume of air sampled is based on the average of the recorded flow rates.

For radionuclide analysis, the air filter may be equally split into two halves at least, and each portion analyzed separately: 1) as a duplicate analysis; 2) as a cross-check analysis for the QA program; or 3) to be retained for re-analysis or conducting other types of analyses.

The volume of air sampled may be assumed to be proportional to the mass of the filter fraction of each filter section, unless data and filter conditions show otherwise. In addition, composite filter samples can be used for measuring long-lived radionuclides.

6.2.2 <u>Radionuclides as Gases</u>

Tritium, as water vapor, can be collected by the methods described in Section 2.2.1 of Method 114. To measure total tritium in air samples (tritiated water vapor plus elemental tritium, ³H), the sampling system requires an oxidizing bed to convert any elemental tritium into water followed by a zeolite bed, to absorb the tritiated water that was initially present in the air and that formed from the oxidation of ³H in the sampling system. To insure elemental tritium is correctly measured the method should:

- 1) Measure both chemical forms of tritium in the environment; or,
- 2) Increase the measured environmental tritiated water vapor concentration by the activity ratio (total tritium versus tritiated water vapor), measured at the point of release; or,
- 3) Show that concentrations of elemental tritium are insignificant at the environmental sampling location relative to the tritium present as water vapor.

Carbon-14 in environmental airborne samples can be considered to be in the form of carbon dioxide (CO_2), and can be sampled as CO_2 (see Method 114, Section 2.2.4).

Radionuclides	Half-Life ^a	Decay Mode		
	Particulates			
U-234	2.4 E+5 yr	alpha		
U-235	7.1 E+8 yr	alpha		
U-238	4.5 E+9 yr	alpha		
Pu-238	8.8 E+1 yr	alpha		
Pu-239	2.4 E+4 yr	alpha		
Am-241	4.3 E+2 yr	alpha		
K-40	1.3 E+9 yr	beta, gamma		
Co-60	5.3 E+0 yr	beta, gamma		
Sr-90	2.9 E+1 yr	beta		
Sb-125	2.7 E+0 yr	beta, gamma		
Pb-212	1.1 E+1 hr	beta, gamma		
	Gases			
H-3 (H ₂)	1.2 E+1 yr	beta		
C-11	2.0 E+1 min	positron		
N-13	1.0 E+1 min	positron		
C-14 (CO ₂)	5.7 E+3 yr	beta		
O-15	1.2 E+2 s	positron		
Ar-41	1.8 E+0 hr	beta, gamma		
Kr-85	1.07 E+1 yr	beta, gamma		
Kr-88	2.8 E+0 hr	beta, gamma		
Xe-133	5.3 E+0 day	beta, gamma		
Liquids/Vapors				
H-3 (H ₂ O)	1.2 E+1 yr	beta		

Table 6-1. Physical Parameters of Selected Primary Radionuclides

^a exponential notation, 2.4 E+5 means $2.4 \times 10+5$, or 240000.

Am = americium; Ar = argon; C = carbon; Co = cobalt; CO₂ = carbon dioxide; H = hydrogen; H₂O = water; hr = hour; K = potassium; Kr = krypton; min = minute; N = nitrogen; O = oxygen; Pb = lead; Pu = plutonium; Sb = antimony; s = second; Sr = strontium; U = uranium; Xe = xenon; yr = year.

Applying cryogenic techniques to sample radioactive noble gases is usually impractical at most locations in the environment and away from the plant. Therefore, a sampler that collects a controlled volume of air at specific time intervals may be acceptable and considered a continuous sample for this purpose. Cryogenic techniques, along with liquid scintillation counting, may be used to separate and measure noble gases (see Method 114, Section 2.2.3).

It may not be practical, or possible, to collect and measure short-lived gaseous radionuclides in environmental samples. These radionuclides are primarily oxygen-15, carbon-11, and nitrogen-13 (see Table 6-1). Although the half-lives of argon-41 and krypton-88 are much longer (i.e., 2-3 hours), their measurement in the environment on a continuous basis is also impractical. As the sample collects, the radioactivity rapidly decays, and in a short time, equilibrium is established where the collection is equal to the decay rate. For some short-lived radionuclides it may be possible to use thermoluminescent dosimeters (TLDs) to measure external radiation exposure rates. For other short-lived radionuclides, there is a limit to the quantity of radioactivity that can be collected. For these reasons, demonstrating compliance by measuring the short-lived radionuclides in Table 6-2 in situ is more difficult and is usually not a practical option:

Radionuclide	Half-life
oxygen-15	120 seconds
nitrogen-13	10 minutes
carbon-11	20 minutes
argon-41	1.8 hours
krypton-88	2.8 hours

Table 6-2. Short Half-Life Radionuclides

Applications should describe the method by which dose arising from these short-lived radionuclides will be calculated, or should provide evidence that the dose from these radionuclides is insignificant compared to the 10 mrem/yr limit. Such descriptions may include physical arguments such as decay in transport. Except for possibly a few DOE facilities, radiation exposures to the maximum exposed individuals due to these short-lived gaseous radionuclides are not significant when compared to the 10 mrem/yr limit.

6.3 Criteria for Environmental Monitoring Programs

6.3.1 <u>Selecting Critical Receptor Locations</u>

The typical quantity used to express concentration at a location as a function of release from the source is concentration (X) per release rate Q or Chi/Q. For facilities with continuous emissions, the critical receptor locations may be either:

- (a) The location of the highest Chi/Q on the facility perimeter fence line (For an elevated release point, the highest Chi/Q may be beyond the fence line); or
- (b) The location of the highest offsite Chi/Q where a residence, business, or school exists.

In case (b), the sampling location may be at any site between the highest offsite Chi/Q and the fence line, if this would make sampling easier, more convenient, or more cost-effective and the EPA concurs with the site location.

Acceptable dispersion models (e.g., AIRDOS-PC, CAP88-PC, COMPLY) may be used to determine the highest Chi/Q location(s). If the highest Chi/Q location is represented by several sites with similar values, measurements should be required at all such sites until the location with the maximum Chi/Q can be definitively identified (minimum one year sampling, five or more years are preferred). Sampling is then required at only the maximum Chi/Q site, unless conditions change. The same procedure should be followed when fence-line measurements are used (case (a) above) and the highest concentrations are computed to be similar within two or more of the sectors.

For facilities with intermittent or variable emissions, many locations around the facility (at least one within each of the sectors) should be monitored.

6.3.2 <u>Continuous Sampling</u>

There may be valid and acceptable reasons for the sampling systems at a facility to be off line for short periods of time (e.g., filter or sample changes, maintenance, calibration, etc.). Under many circumstances, the requirement for continuous sampling can be satisfied when the 80% data completeness requirement is met. This means that the time the sampling system is not in satisfactory operation should not exceed 20% of the sampling period.

The 80% figure is intended to provide uniformity in dealing with various co-located facilities or multiple release points. More restrictive conditions may be required if a facility is approaching the dose limit or is in non-compliance. If necessary, a backup sampler may be placed in operation to insure that sampling is accomplished during the balance of the time (i.e., 20%).

6.3.3 Identifying the Major Radionuclide Contributor

The radionuclides that contribute significantly to the effective dose equivalent typically include particulate and gaseous radionuclides and tritium (see Table 6-1). All of the listed particulate radionuclides and tritium can be readily collected and measured by routine sampling methods. For all other gases listed, only xenon-133 and krypton-85 have half-lives sufficiently long enough to permit them to be collected in the environment and analyzed in a laboratory.

6.3.4 Determining Radionuclide Concentrations Separate from Background

Radionuclide concentrations causing an EDE of 1 mrem/yr or greater must be readily detectable and distinguishable from background. Environmental monitoring programs are typically judged to meet this criterion if the lower limit of detection (LLD) of the sampling and analysis methods is 10% or less of the Concentration Levels for Environmental Compliance (CLEC) listed in Table 2, Appendix E of the 40 *CFR* Part 61. The LLD is defined as the nuclide concentration that is discernable from background at a confidence level of 95% (i.e., the net activity value is greater than a specified value above the random fluctuation of the background count-rate). The LLD is calculated as follows:

$$LLD = (4.66 \times S_b) / [(2.2 \times 10^{12}) \times E \times V \times Y \times e^{(\lambda \Delta t)}]$$
(6-1)

Where:

LLD	=	Lower limit of detection, (Ci/m ³)
S_b	=	Standard deviation of the background or blank count rate, (cpm)
E	=	Counting efficiency, (cpm/dpm)
V	=	Sample volume, (m ³)
Y	=	Radiochemical yield, if applicable, (dimensionless)
λ	=	Radioactive decay constant, (minutes ⁻¹)
Δt	=	Time elapsed between midpoint of sample collection and time of
		counting, (minutes)
2.2 E12	=	Conversion factor, (dpm/Ci)

The value of S_b should be based on the standard deviation of a series of blank measurements using the same type of sample collection media (e.g., an air-particulate filter) carried through the complete analytical procedure.

Detection limits may be expressed as a minimum detectable activity (MDA) or minimum detectable concentration (MDC). Calculating the MDA or MDC requires determination of the standard deviation of the background count rate (S_b). This value can be used in the above equation to compute the MDA or MDC.

Table 6-3 lists typical sensitivities and examples of actual procedural sensitivities for some of the major radionuclides released by DOE facilities. The required sensitivities are one-tenth the concentrations listed in 40 *CFR* 61, Appendix E, Table 2. The procedural sensitivities are based primarily on airborne radionuclide measurement program results conducted at the NAREL, formerly the EERF (BRO83). The information in Table 6-3 indicates that the sensitivities for measuring all particulate radionuclides, tritium, and carbon-14 are quite adequate to satisfy the requirements of the rule. Conversely, the sensitivities associated with argon-41 and krypton-88, are not low enough to satisfy the sensitivities required by the rule.
Table 6-3. Examples of Backgrounds and Sensitivities of Some Principal Airborne Radionuclides Released From DOE Facilities

Radionuclide	Required Sensitivity ^a	Representative Background	Example Sensitivity
	Concentrati	on (pCi/m ³)	
U-234	7.7E-4	25 ^b	13 ^c
U-238	8.3E-4	25 ^b	13°
Pu-238	2.1E-4	<u></u> 4 ^b	13°
Pu-239	2.0E-4	<u></u> 4 ^b	13°
Am-241	1.9E-4	<u></u> 4 ^b	13°
Ar-41	170	0	600^{d}
Kr-85	100,000	50 ^e	ND
Kr-88	50	0	ND
C-14	1.0	1.3 ^f	1.1
Н-3	150	<u>≤</u> 1.1 ^g	<u>≤</u> 1.1 ^g

^a These sensitivities are 1/10 the concentrations listed in 40 *CFR* 61, Appendix E, Table 2.

^b Average of January-December 1986 airborne measurements in 63 U.S. cities (EERF87a, b).

^c Based on a weekly sample, average collection rate of 0.74 cmm (26 cfm), analysis of ½ filters, and a measurement sensitivity of 0.05 pCi/sample.

^d Estimated from an EPA report on airborne radionuclides at the Savannah River Plant (BLA84).

^e Average concentration measured in air at 12 U.S. cities in 1983, Environmental Radiation Data (ERD) filed, Eastern Environmental Radiation Facility, EPA.

^f Concentration taken from pp. 61-62 of NCRP85 and ORP73. This concentration relates to 7.5 pCi/g carbon.

^g This estimate assumes 30% humidity at 20°C and a background <200 pCi/L of water vapor.

Am = americium; Ar = argon; C = carbon; H = hydrogen; Kr = krypton; ND = not determined; NL = not listed; $pCi/m^3 = picoCuries$ per cubic meter; Pu = plutonium; U = uranium

6.3.5 <u>When Background Concentrations Interfere</u>

Background radionuclide concentrations are typically low enough that nearly all radionuclides released by DOE facilities can be readily distinguished from background levels at concentrations that would cause an EDE of 1 mrem/yr (see Table 6-3). However, there are two notable exceptions:

1) Rn-222 progeny concentrations in air that cause an EDE to the lung of 1 mrem/yr cannot be distinguished from a background concentration of less 0.5 pCi/L when in equilibrium with the Rn-222 parent; and,

2) Submersion dose rates of 1 mrem/yr caused by radionuclide concentrations in air cannot be distinguished from background external exposure rates due to photons.

Therefore, any DOE application proposing to measure radon-222 progeny or external exposure rates should be carefully evaluated to ensure that provision is made for including local background concentrations in the analysis for its technical merits.

Background levels are defined as general ambient radionuclide concentrations that are not related to an emission source. In some cases, sources other than the facility of interest may contribute to the radionuclide concentrations at the critical receptor location. Uranium mining and milling facilities are potential examples of multiple emission sources contributing to the measurements made at a single receptor location. Also, this situation can exist when several different facilities releasing similar contaminants are in the same area. In these cases, it may be difficult to distinguish individually the contributions of the various sources at receptor locations.

Similarly, when the radionuclide being monitored also occurs in nature (e.g., potassium-40), or emanate from off-site sources, the contribution to airborne concentrations from natural or off-site sources may not be distinguishable from the amount of the radionuclide released from the facility. The DOE staff are encouraged to work with regional EPA offices and state agencies to evaluate how best to handle these cases.

Monitoring programs that include subtractions from other emission sources will be critically reviewed to insure that any adjustments made are appropriate. Rather, the total airborne concentration (from all sources) should be compared to the concentration levels of Table 2, Appendix E of 40 *CFR* 61, to determine compliance.

6.4 Evaluating the Validity of a Quality Assurance Program

In order to evaluate the validity of a QA Program in response to the performance requirements of Appendix B, Method 114, 40 *CFR* 61, the application should include a statement that a QA program in general conformance with the requirements of Method 114 will be implemented. It is also necessary to meet all requirements of EPA Order 5360.1 A2. Information required by Section 4 of Method 114, includes the following:

- 1) The requirements for precision, accuracy, and completeness of the environmental measurements; and,
- 2) The number of replicates, spiked samples, split samples, and blank samples to be analyzed.

In providing this information, the following guidelines should be used:

1) The accuracy and precision of the measurements should be within 20% of the concentration levels listed in Table 2 of Appendix E;

- 2) Completeness should be at least 95%, that is, 95% of the samples collected should provide valid data; and,
- 3) 20% of the samples analyzed should be replicates, blank, split, or spiked samples. Usually 10% are duplicate or split samples, 5% are blank samples, and 5% are spiked samples.

It is also necessary to meet all requirements of EPA Order 5360.1 A2. If the requirements of this EPA Order are more restrictive, they must be followed in addition to the above specified criteria. More information on EPA QA specifications can be found at http://www.epa.gov/Quality/qa_docs.html.

Chapter 7

GUIDANCE ON METHODS FOR ESTIMATING FUGITIVE RADIONUCLIDE AIR EMISSIONS FROM DIFFUSE SOURCES

7.1 Introduction

The purpose of this chapter is to provide step-by-step procedural guidance for estimating fugitive radionuclide emissions from diffuse emission sources. It is anticipated that this guidance will be used by DOE staff and its contractors responsible for estimating diffuse source emissions for inclusion in 40 *CFR* 61 Subpart H submittals. It is necessary to provide guidance that standardizes the emission estimating procedures to ensure the emission estimates developed at each DOE facility subject to 40 *CFR* 61 Subpart H are complete, consistent, and of comparable accuracy and quality. The emission estimating methods presented in this section are sufficiently transparent to allow users to independently replicate these procedures.

The guidance in this chapter is primarily derived from emission calculation methods and models that use EPA-sanctioned procedures. These preferred emissions estimating methods are based on operating parameters or site-specific activity data. All of the procedures discussed in this chapter provide estimates of the rate at which radionuclides are emitted. Some of the methods provide hourly emission rates, while other methods provide annual emission rates, depending upon the accuracy of the method. The hourly emissions should be summed to obtain an annual emission rate in terms of Ci/yr to be incorporated in the CAP88-PC model, or other prior, or future, EPA- approved models.

Given that DOE facilities are not identical and some of the data required to estimate emissions using the preferred emission estimating procedures are not readily available at all sites, where possible, acceptable alternative approaches for estimating emissions are included in this chapter. These alternative approaches, though they may be easier to implement, often yield less accurate emission (and less site-specific) estimates than the preferred methods. Where multiple alternative methods are provided for a source category, they are presented in declining order of accuracy.

In Section 7.2, guidance is provided to help identify all possible sources of radionuclides, in order to insure that the developed radionuclide inventory is complete and potential sources are not unintentionally left out. Section 7.3 provides guidance to differentiate between potential point sources and diffuse sources. This section also shows how to classify diffuse sources into the six primary diffuse source categories and a number of related subcategories. Such classification of diffuse sources can help in identifying the appropriate emission estimating procedure. These emission estimating procedures are presented in Section 7.4 for each of the diffuse source categories discussed in Section 7.3. Section 7.5 discusses data collection and emission calculation issues pertinent to the methods in Section 7.4. Section 7.6 is a particularly important section as it provides guidance on quality control procedures that should be implemented to insure and quantify the comparability of the emission estimates. Section 7.7 provides guidance on documentation of the emission estimating procedures used and reporting of

the diffuse source emission estimates. Case studies are provided in the last section of this chapter to demonstrate the strengths and limitations associated with diffuse source emission estimating procedures.

7.2 Identify Sources of Radionuclide Emissions

In order to correctly and accurately identify potential point and diffuse radionuclide emission sources, it is necessary to identify materials that contain and emit radionuclides and track how these radioactive materials are handled. This would include identification of processes, storage facilities, processing equipment, including equipment that may have equipment leaks, testing operations, laboratory experiments, and waste handling operations.

A useful approach to identifying radioactive material usage is to use process flow charts and Piping & Instrumentation Diagrams (P&IDs) to show where radionuclides are introduced and map the different processes that come in contact with this material. These flow charts should include ancillary support operations such as process cooling, wastewater discharges, and room or hood ventilation. Vents or stacks that emit into the atmosphere should be mapped onto the process flow charts. Those emission points that are actively ventilated need to be identified as point sources. Those operations that are not actively ventilated should be identified as diffuse emission sources.

As some radionuclides have a relatively long half life, it is important to consider historical data that indicate where water, soil, building, or equipment contamination may have occurred in the past and may continue to emit radionuclides. In identifying these historical emission sources, as well as processes that are decontaminated, decommissioned or demolished, it is necessary to document any remediation activities implemented to mitigate emissions. In many cases, these emission sources are considered diffuse sources as their emissions tend not to be collected and actively ventilated into the atmosphere.

7.3 Determine Population of Diffuse Emission Sources

As noted throughout this document, point sources are those sources of radionuclides that are actively ventilated into the atmosphere and are specifically addressed in 40 *CFR* 61 Subpart H. Diffuse source emissions are passively emitted into the environment through evaporative or atmospheric transport driven mechanisms. Data collected for Section 7.2 should be reviewed to identify all radionuclide emission sources that are not actively ventilated.

In some cases, sources that are typically considered diffuse sources such as wastewater treatment plants would be considered a point source if the plant is enclosed and actively ventilated. Conversely, process sources, such as equipment leaks, would be considered diffuse emission sources if these emissions are emitted into the environment via passive ventilation.

In addition to identifying the diffuse emission sources at a facility, it is necessary to classify the identified sources into one of the specific diffuse source types. The major types of diffuse sources include the following:

- Soils;
- Water/Evaporation;
- Underground testing;
- Buildings;
- Equipment; and,
- Tank venting

Each diffuse source category may include multiple sub-categories. For example, water/evaporation may have separate discussions for emissions associated with evaporation from surface water impoundments, wet-cooling towers, and plant transpiration of contaminated groundwater.

The soils category includes radionuclides associated with windswept particulate matter (PM) emissions. Though volatile pollutants can be emitted from soils, PM emissions from soils tend to include the heavier, less volatile radionuclides.

Building emissions sources occur where radioactive air emissions emanate from a building structure to the environment through non-actively ventilated/exhausted points (e.g., doors, windows, air vents, holes). Such emission sources would include all of the fugitive emissions from process equipment, building hold-up, and other building contamination emanating from the building to the environment.

Equipment diffuse emission sources are those sources from which radioactive air emissions emanate directly to the environment (e.g., equipment sitting on a pad or covered patio).

Tank venting relates to liquid storage tanks (see Figure 7-1) that contain radioactive material that emit radionuclides into the atmosphere either from loading losses, where vapors are displaced when the tank is filled, or breathing losses, where vapor emissions are driven by diurnal changes in ambient temperature.

Though underground testing has been discontinued, a number of sites have emissions from soils associated with earlier testing programs. This would also include emissions associated with leaking monitor well seals at underground testing sites.

The surface water/evaporation source category include a number of subcategories such as ponds and lagoons, water cooling towers, evaporation from moist soils, and transpiration of radionuclides from vegetation. Emissions from this category tend to be associated with volatile pollutants such as noble gases and tritium.



Figure 7-1. Example of Typical Liquid Storage Tank Configuration

7.4 Determine Appropriate Emission Estimation Methods

Once the diffuse sources have been identified and classified, then an appropriate emission estimating procedure needs to be identified to accurately quantify emissions. In this section, appropriate emission estimating methods are discussed for each of the source categories and subcategories noted in Section 7.3. Each method will include a brief description of the approach, discussion of associated strengths and limitations, required equations, models, and software tools, and a list of the data elements needed to implement the approach. Each section will clearly define where to obtain or how to develop the required input data. Default values that have been developed for any of the required data elements will also be presented.

Each discussion will include a preferred, and where available, alternative emission estimating approaches. The preferred methods noted in this section are derived from the EPA-approved approaches listed in Table 7-1. These methods are preferred because they provide the most accurate emission estimates based on data currently available. In some cases, the data needed to use the preferred approach are not readily available at every DOE site, therefore alternative approaches are provided. The inclusion of alternative approaches will also allow users the ability to tailor their emission estimating procedures to better match the specific emission sources found at their sites and utilize data that are readily available to them. It should be

recognized that the

Mechanism	Procedure	Status		
SOILS				
Wind Erosion				
a. Limited open areas	<i>AP-42</i> method using "fastest mile wind speed"	Adopted by EPA (AP-42)		
b. Unlimited open areas	Modified Wind Erosion Equation	Approved by EPA (EPA88a)		
c. Intermittent waste piles	<i>AP-42</i> method using "fastest mile wind speed," modified for geometry of pile	Adopted by EPA (AP-42)		
d. Uranium ore and mill tailings	NRC Regulatory Guide 3.59 methodology	Adopted by EPA		
Material Handling	· · ·			
a. Soil removal	Same as continuous waste piles	EPA guidance for TSDF		
b. Soil grading and shaping	<i>AP-42</i> emission factor for bulldozing overburden at western coal mines	EPA guidance for TSDF		
c. Agriculture	AP-42 emission factor	Adopted by EPA (AP-42)		
d. Demolition	Same as continuous waste piles	Approved by EPA (EPA88a)		
e. Unpaved Roads	AP-42 methodology	Adopted by EPA (AP-42)		
f. Ongoing waste pile operations	<i>AP-42</i> aggregate handling emission factor	Adopted by EPA (AP-42)		
Contaminated Soils				
a. Tritium	Proposed based on DOE model	Proposed		
b. Carbon-14	Proposed based on DOE model	Proposed		
WATER/EVAPORATION				
a. Open ponds	Evaporation equation from NUREG-0570	Used by NRC staff		
b. Saturated soil	Same as open ponds	Based on EPA88c		
c. Subsurface soil	Superfund Exposure Assessment Manual	EPA: OSWER Directive		
d. Wet-cooling tower	Cooling loss equation	Proposed		

Table 7-1. Summary of Methods for Estimating Diffuse Source Emissions

UNDERGROUND TESTING		
a. Underground testing	Proposed air sampling protocol combined with short-term dispersion calculations	Proposed
BUILDINGS		
a. Buildings	Proposed method based on measurement or estimated source term	Site-specific model approved for Mound Plant by EPA Region V, 1997
EQUIPMENT		
a. Equipment	Same as buildings	Proposed
TANK VENTING		
a. Tank venting	AP-42 methodology/TANKS model	Proposed

Table 7-1.	Summary	of Methods for	• Estimating	Diffuse H	Emissions ((Continued)
						<pre> /</pre>

DOE = U.S. Department of Energy; NRC = Nuclear Regulatory Commission; NTS = Nevada Test Site; OSWER = U.S. EPA Office of Solid Waste and Emergency Response; PM = Particulate Matter. AP-42 = Compilation of Air Pollutant Emission Factors document published by EPA.

alternative approaches tend to be based on generic assumptions and are often significantly less accurate than the preferred approaches. For example, for surface water impoundments, the preferred method is an equation which provides an hourly emission rate estimate that takes into account a variety of factors such as hourly wind speed and temperature, vapor pressure, and surface area. The recommended alternative is a mass balance approach that assumes that all radionuclides entering an impoundment during a year are emitted to the atmosphere during the year. This alternative approach is easier to implement, but is considerably less accurate than the preferred approach. Where multiple alternative approaches are included, they are presented in this chapter in descending order of accuracy and preference.

For most of the preferred methods presented in this chapter, an example calculation is provided to demonstrate the data required to estimate emissions, the use of available default values, and calculation procedures.

7.4.1 <u>Soils</u>

The soils category addresses radionuclides that are emitted as PM from handling minerals or soils contaminated with radioactive material. The category is subdivided into emissions due to wind erosion or material handling. Each of these groups is further subdivided. For example, wind erosion can occur in limited or unlimited open areas, or from intermittent waste piles. Material handling emissions can occur due to soil removal, soil grading and shaping, agricultural activities, building demolition, use of unpaved roads, or ongoing waste pile operations.

7.4.1.1 Wind Erosion - Limited Open Areas and Intermittent Waste Piles

Open areas such as fields or storage piles are characterized as having either a "limited" or an "unlimited" wind erosion potential. An example of an area with "limited" potential would be an inhomogeneous field covered with gravel, rocks, or clumps of vegetation. In this scenario, the ground is partially sheltered from the wind and from the cascade of saltating particles, so the fine particles interspersed among these non-erodible elements require higher wind speeds for suspension. Once such winds occur, the supply of erodible particles is quickly exhausted and emissions stop until the area is disturbed and a fresh supply of fine particles is brought to the surface. An intermittent waste pile is a material storage pile which receives new material from time-to-time. The new material replenishes the pile, providing a new supply of fine particles which can be blown into the atmosphere. If the windblown dust has been contaminated with radioactive material, then the dust emissions may contain radionuclides. For additional information about this source category see Chapter 3.0.

Preferred Method to Estimate Radionuclide Emissions from Limited Open Areas and Intermittent Waste Piles.

The preferred method to estimate emissions from both limited open areas and intermittent waste piles is the *AP-42* method referred to as the "fastest mile wind speed" approach. The fastest mile wind speed represents the wind speed corresponding to the whole mile of wind movement that has passed by the 1 mile contact anemometer in the least amount of time. In this approach, multiple steps are required to calculate emissions.

First, it is necessary to determine the number of times the pile is disturbed per year; for example, if the storage pile is replenished every three days, then the annual disturbance would be approximately 120. Next, review available meteorological wind speed data disaggregated into periods that match the pile disturbance periods; for example, if a pile is replenished every 3 days, the meteorological data should be disaggregated into 3-day periods and the fastest mile wind speed for each period noted.

Next, the wind speed data should be adjusted to represent a sampling height of 10 meters (32.8 feet). The following logarithmic wind equation, applicable in the surface layer of the planetary boundary layer, can be used to make this adjustment:

$$U_{10}^{+} = U^{+} \cdot \frac{\ln (10/5.0 \times 10^{-3})}{\ln (z/5.0 \times 10^{-3})}$$
(7-1)

Where:

 U_{10}^{+} = Wind speed at a height of 10 meters (mph) U^{+} = Reported wind speed (mph) z = Height of wind speed monitor (m) 5.0×10^{-3} = Typical roughness height for open terrain (m)

The fastest mile wind speed estimates for each period must be converted into equivalent friction

velocities for each wind surface regime. The wind surface regime varies according to the shape of the storage pile and the orientation to the wind as noted in Figure 7-2. This figure shows the wind speed pattern expressed as a fraction (U_s / U_r) of the surface wind speed (U_s) to the approach to the wind speed (U_r) , which have been derived from wind speed studies. These fractions can be applied to the following equation to get the friction velocity for each wind surface regime.

$$U^* = 0.10 \times (U_s/U_r) \times U_{10}^+$$
(7-2)

Where:

 U^* = Friction velocity 0.10 = Von Karman's constant divided by (25 cm/ln 0.5 cm) (dimensionless) U_s / U_r = Wind speed pattern fraction (see Figure 7-4) U_{10}^+ = Adjusted wind speed at 10 meters (mph)

It is important at this stage to determine if any of the friction velocities exceed the threshold value for the material stored in the pile. Threshold values for different materials are noted in Table 7-2.

Material	Threshold Friction Velocity [m/s(ft/s)]
Overburden ^a	1.02 (3.35)
Scoria (roadbed material) ^a	1.33 (4.36)
Ground coal (surrounding coal pile) ^a	0.55 (1.80)
Uncrusted coal pile ^a	1.12 (3.67)
Scraper tracks on coal pile ^{a, b}	0.62 (2.04)
Final coal dust on concrete pad ^c	0.54 (1.77)

 Table 7-2.
 Threshold Friction Velocities

^a Western surface coal mine.

^b Lightly crusted.

^c Eastern power plant.

Next, the erosion potential needs to be calculated for each wind surface regime using the following equation:

$$P = 58 \times (U^* - U_t^*)^2 + 25 \times (U^* - U_t^*)$$
(7-3)



Pile	Percent Of Pile Surface Area				
Subarea	Pile A	Pile B1	Pile B2	Pile B3	
0.2 a	5	5	3	3	
0.2 b	35	2	28	25	
0.2 c	NA	29	NA	NA	
0.6 a	48	26	29	28	
0.6 b	NA	24	22	26	
0.9	12	14	15	14	
1.1	NA	NA	3	4	

NA = not applicable



Where:

The surface area of each wind surface regime is obtained from Figure 7-2 and applied to the calculated erosion potential to estimate the mass of PM emitted as noted in equation 7-4. The following equation assumes that the specific activity of the windblown dust is similar to the specific activity of the storage pile.

$$\mathbf{E} = \mathbf{k} \times \sum \mathbf{P}_{\mathbf{i}} \times \mathbf{S}_{\mathbf{i}} \times \mathbf{a} \tag{7-4}$$

Where:

Е	= Annual emissions (pCi/yr)
k	= Particle size multiplier (see Table 7-3)
Pi	= Erosion potential for each wind surface regime (g/m ²
Si	= Surface area of each wind surface regime (m^2)
a	= Specific activity of storage pile (pCi/g)

Table 7-3. Particle Size Multiplier

Aerodynamic Particle Size	< 30 µm	< 15 µm	< 10 µm	< 2.5 µm
k Value	1.0	0.6	0.5	0.2

Table 7-4 lists all of the required data elements to implement this approach and also includes information on how to develop or where to obtain the required data.

Table 7-4.	Required Data for the Preferred Method for Limited Open Areas and
	Intermittent Waste Piles

	Equation	
Required Data	Variable	Comments
Reported fastest mile wind speed (mph)	U^{+}	Obtained from nearest spatially representative meteorological monitoring station. National weather service stations currently report fastest two minute wind speeds that can be used to approximate fastest mile wind speeds
Height of wind speed monitor (m)	Z	Obtained from nearest spatially representative meteorological monitoring station.
Wind speed pattern fraction (dimensionless)	U _s / U _r	Obtained from Figure 7-2

Table 7-4. Required Data for the Preferred Method for Limited Open Areas and Intermittent Waste Piles (Continued)

	Equation	
Required Data	Variable	Comments
Threshold friction velocity (m/s)	Ut	Obtained from Table 7-2
Surface area of each wind surface regime (m ²)	S _i	Each regime is roughly estimated based on the relative portion of the total surface area of the pile.
Particle size multiplier	k	Obtained from Table 7-3
Specific activity of storage pile (pCi/g)	а	Derived from site-specific samples of aggregate or soils being transferred.

Example Calculation of Preferred Method

A facility maintains a conically shaped pile 11 m (36.1 ft) in height and 29.2 m (95.8 ft) in base diameter, containing about 2000 Mg (2, 205 tons) of material, with a bulk density of 800 kg/m³ (49.9 lbs/ft³)(similar to coal). The specific activity of the pile is 2.1×10^{-7} pCi/g. 250 Mg (276 tons) of material (12.5 percent of the stored capacity) is added back to the pile every 3 days, thereby restoring the full capacity of the pile.

The first step is to determine the fastest wind speed for each period of disturbance. Figure 7-3 shows a representative set of values for a 1-month period. The values have been separated into 3-day periods, and the highest fastest mile wind speed in each period is indicated.

In this example, the anemometer height is 7 m (23 ft), so that a height correction to 10 m (32.8 ft) is needed. This adjustment is made by applying equation 7-1 to the fastest wind speed data for each period, as demonstrated in Table 7-5.

$$U_{10}^{+} = 1.05 \times U_7^{+}$$

The next step is to convert the fastest mile wind speed values for each 3-day period into the equivalent friction velocities for each surface wind regime (i. e., U_s/U_r ratio) of the pile, using equation 7-2.

$$U^* = 0.10 \times (U_s/U_r) \times U_{10}^+$$

As the pile has a conical shape, the surface wind speed pattern A from Figure 7-2 should be used. The surface areas lying within each wind speed regime are tabulated in the table in Figure 7-2. The friction velocities for this example are calculated and noted in Table 7-5.

The threshold friction velocity, a value of 1.12 m/s (3.67 ft/s), is obtained from Table 7-2 by assuming that the material in the storage pile is similar to coal.

		Direction	
	Fastest Mile	(10s of degrees	
Average Speed (mph).	Speed (mph)	azimuth)	Date
6.5	9	36	1
10.5	14	01	2
6.0	10	02	3
11.4	16	13	4
11.9	15	11	5
19.0	29	30	6
19.8	30	30	7
11.2	17	30	8
8.1	15	13	9
15.1	23	12	10
23.3	31	29	11
13.5	23	17	12
15.5	18	18	13
9.6	22	13	14
8.8	13	11	15
13.8	21	36	16
11.5	15	34	17
5.8	12	31	18
10.2	14	35	19
7.8	16	24	20
10.6	16	20	21
17.3	23	32	22
8.5	14	13	23
8.8	15	02	24
11.7	17	32	25
12.2	16	32	26
8.5	16	26	27
8.3	13	32	28
6.6	10	32	29
5.2	9	31	30
5.5	8	25	31

Local Climatological Data Monthly Summary

Figure 7-3.	Example	Wind	Speed Data	l
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	U ₇ +		U ₁	U ₁₀ +		$U^* = 0.1u + (m/s)$			
3-Day Period	mph	m/s	mph	m/s	u _s /u _r : 0.2	u _s /u _r : 0.2	u _s /u _r : 0.9		
1	14	6.3	15	6.6	0.13	0.40	0.59		
2	29	13.0	31	13.7	0.27	0.82	1.23		
3	30	13.4	32	14.1	0.28	0.84	1.27		
4	31	13.9	33	14.6	0.29	0.88	1.31		
5	22	9.8	23	10.3	0.21	0.62	0.93		
6	21	9.4	22	9.9	0.20	0.59	0.89		
7	16	7.2	17	7.6	0.15	0.46	0.68		
8	25	11.2	26	11.8	0.24	0.71	1.06		
9	17	7.6	18	8.0	0.16	0.48	0.72		
10	13	5.8	14	6.1	0.12	0.37	0.55		

Table 7-5. Calculation of Friction Velocities

The final set of calculations involves the tabulation and summation of emissions for each disturbance period and for the affected sub-area. The erosion potential (P) is calculated using equation 7-3. For example, the calculation for the second 3-day period is:

 $P = 58 \times (U^* - U^*_t)^2 + 25 \times (U^* - U^*_t)$ $P_2 = 58 \times (1.23 - 1.12)^2 + 25 \times (1.23 - 1.12)$ $P_2 = 3.45 \text{ g/m}^2 (7.1 \times 10^{-4} \text{ lbs/ft}^2)$

In order to apply the erosion potential values (P) to the example storage pile, it is necessary to estimate the total surface area of the pile and the surface area of each of the wind surface regimes. As the pile has a conical shape, the total surface area can be calculated as follows:

$$S = \pi \times r \chi \times \sqrt{(r^2 + h^2)}$$

$$S = \pi \times 29.2 \times \sqrt{((29.2/2)^2 + 11^2)}$$

$$S = 838 \text{ m}^2 (9,020 \text{ ft}^2)$$

The total surface is now allocated to each of the surface wind regimes (see Table 7-6) based on the surface area percentages for pile A included in Figure 7-2.

		Pile Surface Area		
Area	U _s /U _r	% of surface area	Area (m ²)	
Α	0.9	12	101	
В	0.6	48	402	
C1 + C2	0.2	40	335	

Table 7-6. Surface Area Apportionment

To estimate the radionuclide emissions for this storage pile, the erosion potential (P) value for each surface wind regime is multiplied by the surface area (S) of the regime, the specific activity of the pile (a) and the particle size multiplier (k). This calculation is performed for each surface wind regime and summed as noted in the following equation:

 $E = k \times \sum P_i \times S_i \times a$

Results of this calculation are noted in Table 7-7.

3-Day Period	U* (m/s)	U* - Ut* (m/s)	P (g/m ²)	ID	Pile Surface Area (m ²)	E (pCi/yr)
2	1.23	0.11	3.45	А	101	3.57×10^{-5}
3	1.27	0.15	5.06	А	101	5.46×10^{-5}
4	1.31	0.19	6.84	А	101	7.35×10^{-5}
					TOTAL	1.638×10^{-4}

Table 7-7. Calculation of PM₁₀ Emissions^a

^a Where $U_t^* = 1.12$ m/s for uncrusted coal and k = 0.5 for PM₁₀.

7.4.1.2 Wind Erosion - Unlimited Open Areas

An open area with an "unlimited" erosion potential would be a smooth field, lacking vegetation, and covered with a thick layer of loose sandy soil. In such a field, relatively low wind speeds will cause suspension by the action of saltating particles. Because of the large reservoir of erodible particles at such sites, the erosion rate will vary as a function of the wind speed, and will not appreciably decrease with time.

Preferred Method to Estimate Radionuclide Emissions from Unlimited Open Areas

The preferred approach to estimate emissions from unlimited open areas is derived from the DOA revised wind erosion equation. This equation has been released as a software package that is available from the DOA website <u>http://www.usda.gov</u> along with documentation for the model. Note, the model is designed to quantify erosion resulting from saltation and creep at heights up to two meters and thus is not appropriate for estimating transport of suspended fine sediments above two meters. The PM emission estimate obtained from the model can be speciated into pCi by obtaining a soil sample and quantifying the amount of pCi per mass of PM and applying this fraction to the emission estimate.

7.4.1.3 Material Handling - Ongoing Waste Pile Operations and Soil Removal

Inherent in operations that use top soils on minerals in an aggregate form is the maintenance of outdoor storage piles. Storage piles are usually left uncovered because of the need for frequent material transfer into or out of storage. When freshly processed aggregate is loaded onto a storage pile or removed, the potential for dust emissions is at a maximum; fines are easily disaggregated and released to the atmosphere upon exposure to air currents, either from aggregate transfer itself or from high winds. If the material being transferred has been contaminated by radioactive material, the dust emissions associated with the transfer may contain radionuclides. This section of Chapter 7 quantifies emissions associated with the loading and unloading of contaminated materials. For additional information about this source category see Section 4.1.

Preferred Method to Estimate Radionuclide Emissions from Ongoing Waste Pile Operations and Soil Removal

The preferred method to estimate radionuclide emissions from ongoing waste pile operations and soil removal is based on the *AP-42* emission factors for aggregate handling.

$$E = k \times (3.2 \times 10^{-3}) \times \frac{\left(\frac{U}{5}\right)^{1.3}}{\left(\frac{M}{2}\right)^{1.4}} \times N \times [a/(2.2 \times 10^{-3})]$$
(7-5)

Where:

E = Emissions (pCi/yr) k = Particle size multiplier (dimensionless) 3.2×10^{-3} = Empirical coefficient U = Mean wind speed (mph) M = Material moisture content (%) N = Mass of material handled annually (tpy) a = Specific activity of contaminated material (pCi/g) 2.2×10^{-3} = Conversion factor (lb/g)

The particle size multiplier (k) in the equation varies depending upon the aerodynamic particle size range, as noted in Table 7-8.

Table 7-8.	Particle	Size	Multiplier
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Aerodynamic Particle Size	<30 µm	<15 µm	< 10 µm	< 5 µm	< 2.5 µm
k Value	0.74	0.48	0.35	0.20	0.11

Table 7-9 lists all of the required data elements to implement this approach and also includes information on how to develop or where to obtain the required data.

Table 7-9.	Required Data for the Preferred Method for Ongoing Waste Pile Operations
	and Soil Removal

Required Data	Equation Variable	Comments
Particle Size Multiplier (dimensionless)	k	Obtain from Table 7-8
mean wind speed, (m/s) or (mph)	U	Wind speeds can vary from 1.3 to 15 mph. Obtain local wind speed data from nearest spatially representative meteorological monitoring station.
material moisture content (%)	М	The moisture content typically ranges from 0.25% to 4.8%. Site-specific values can be derived from samples of the aggregate or soils being transferred. If site-specific data are not readily available then a default value of 0.25 should be used.
Mass of material handled annually (tpy)	Ν	Obtained by estimating the annual throughput of the equipment pile. This information can be provided by the equipment operators.
Specific activity of contaminated material (pCi/g)	а	Derived from site-specific samples of aggregate or soils being transferred.

Example Calculation Using the Preferred Method

In maintaining a material storage pile, a facility moves 270 Mg/yr (300 tpy) of material with a moisture content of 0.4%. The average wind speed in the area is 3.2 kmph (2 mph). The specific activity value for the material is 2.1e-7 pCi/g. To estimate emissions from this operation equation 7-5 is used:

E = k × (3.2 × 10⁻³) ×
$$\frac{\left(\frac{U}{5}\right)^{1.3}}{\left(\frac{M}{2}\right)^{1.4}}$$
 × N × (a/2.2 × 10⁻³)

For the particle size multiplier (k), the value for $10\mu m$ (0.35) should be used.

When the required data are incorporated, the equation becomes:

$$E = 9.28 \times 10^{-5} \text{ pCi/yr}$$

7.4.1.4 Material Handling - Soil Grading and Shaping

Radionuclides can be emitted when contaminated soil is graded or shaped by construction equipment such as bulldozers and road graders. Radionuclides associated with PM are released into the environment as the soils are disturbed and wind currents blow the particles into the atmosphere. For additional information about this source category, see Section 4.1.2.

Preferred Method to Estimate Radionuclide Emissions from Soil Grading and Shaping

The preferred method to estimate emissions from soil grading and shaping is based on *AP-42* emission factors for removal of topsoil from western surface coal mines by scraper. To estimate radionuclide emissions for this source category, the following equation should be used:

$$E = 5.8 \times 10^{-2} \times M \times a / 2.2 \times 10^{-3}$$
(7-6)

Where:

E = Annual emissions (pCi/yr) 5.8×10^{-2} = Emission factor (lb of PM/t) M = Mass of soil graded or shaped (tpy) a = Specific activity of soil (pCi/g) 2.2×10^{-3} = Conversion factor (lb/g)

Table 7-10 lists all of the required data elements to implement this approach and also includes information on how to develop or where to obtain the required data.

Required Data	Equation Variable	Comments
Mass of soil graded or	М	The amount of soil moved annually can be estimated by
shaped (tpy)		interviewing equipment operators.
Specific activity of soil	а	Derived from site-specific soil samples.
(pCi/g)		

Table 7-10. Required Data for the Preferred Method for Soil Grading and Shaping

Example Calculation Using the Preferred Method

A facility moves 272 Mg (300 tons) of contaminated material a year for a water erosion project. The material has a specific activity of 2.1e-7 pCi/g. To estimate the radionuclide emissions associated with the movement of this material equation 7-6 is used.

$$E = 5.8 \times 10^{-2} \times M \times (a/2.2 \times 10^{-3})$$

When the required data are incorporated, the equation becomes

$$E = 5.8 \times 10^{-2} \times 300 \times (2.1 \times 10^{-7}/2.2 \times 10^{-3})$$

E = 1.661 × 10⁻³ pCi/yr

7.4.1.5 Material Handling - Agriculture

During agricultural tilling of soils contaminated by radioactive material, PM associated radionuclides from loosened and pulverized soils are released into the atmosphere as the soil is disturbed. Dust emissions are greatest during periods of dry soil and during the final seed bed preparation. For additional information on this source category see Section 4.1.3.

Preferred Method to Estimate Radionuclide Emissions from Agricultural Activities

The preferred method to estimate radionuclide emissions from agricultural activities is derived from *AP-42* emission factors. The amount of radionuclides emitted from agricultural tilling may be estimated using the following empirical expression:

$$E = k \times 4.80 \times s^{0.6} \times A \times a / 2.2 \times 10^{-3}$$
(7-7)

Where:

E	= Emissions (pCi/yr)
k	= Particle size multiplier (dimensionless, see Table 7-11)
4.8	= Empirical coefficient (dimensionless)
S	= Silt content of surface soil (%)
А	= Area tilled (ac)

a = Specific activity of contaminated soil (pCi/g) 2.2×10^{-3} = Conversion factor (lb/g)

The particle size multiplier (k) in the equation varies with aerodynamic particle sizes as noted in Table 7-11.

Table 7-11. 1	Particle Size	Multiplier
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	Total particulate	$<$ 30 μ m	<15 µm	< 10 µm	< 5 µm	< 2.5 µm
Particle Size	1.0	0.33	0.25	0.21	0.15	0.10
Multiplier (k)						

Table 7-12 lists all of the required data elements to implement this approach and also includes information on how to develop or where to obtain the required data.

 Table 7-12. Required Data for the Preferred Method for Agricultural Tillage

Required Data	Equation Variable	Comments
Particle size multiplier (dimensionless)	k	Obtain from Table 7-11
Silt content of surface soil (%)	S	Derived from site-specific soil samples. In the event that a site-specific value for silt content cannot be obtained, the mean value of 18 percent may be used as a default.
Area tilled (ac)	А	Calculated from maps of actual areas cultivated.
Specific activity of contaminated soil (pCi/g)	а	Derived from site-specific soil samples.

Example Calculation Using the Preferred Method

A 80,937 m² (20 acre) field is tilled for agricultural purposes. The silt content of the soil is 12% and the specific activity is 2.1 e-7 pCi/g. To estimate emissions from this agricultural activity equation 7-7 is used:

 $E = k \times 4.80 \times s^{0.6} \times A \times a / 2.2 \times 10^{-3}$

For the particle size multiplier, the 10 μ m value should be used, which can be obtained from Table 7-11. When the required data are incorporated, equation 7-7 becomes:

$$E = (0.21) \times 4.8 \times (12)^{0.6} \times 20 \times (2.1 \times 10^{-7}/2.2 \times 10^{-3})$$
$$E = 8.55 \times 10^{-3} \text{ pCi/year}$$

7.4.1.6 Material Handling - Demolition

Radionuclide emissions can occur when buildings that have been contaminated with radioactive material are demolished. The movement of building debris often releases dust particles that may contain radionuclides into the atmosphere. For additional information about this source category see Section 4.1.4.

Preferred Method to Estimate Radionuclide Emissions from Demolition

The preferred approach to estimate radionuclide emissions from demolition activities uses the following emission estimating equation:

$$E = 2.9 \times 10^{-2} \times L \times A \times (1 - CE/100) \times a/2.2 \times 10^{-3}$$
(7-8)

Where:

Е	= Emissions, (pCi/event)
L	= Waste material load per floor space unit area, (Mg/m^2)
2.9×10^{-2}	= Emission factor, (kg/Mg)
А	= Building or total floor space area, (m^2)
CE	= Control efficiency of mitigation approach (%)
a	= Specific activity of building material being disposed (pCi/g)
2.2×10^{-3}	= Conversion factor (lb/g)

In practice, mitigation approaches may be implemented to reduce emissions. For example, the facility may be decontaminated before the onset of the demolition work, a temporary containment structure may be erected over the facility being demolished, or water may be used as a wetting agent to reduce dust emissions. These measures may result in lower emission rates. The percent reduction from these measures is considered in the above equation in the control efficiency term.

The determination of mitigation measure control efficiency will be a process- and site-specific consideration. General guidance on control equipment performance capabilities can be obtained from several sources including the Technology Transfer Network's Clean Air Technology Center (http://www.epa.gov/ttn/catc/), *AP-42* (http://www.epa.gov/ttn/chief/ap42/index.html), equipment vendors, and other published texts on air pollution controls. Users will need to make site-specific determinations on CE based on local conditions and controls configurations (e.g., daily wet suppression may have an CE of dust emissions of 30%, but the site may only apply wet suppression once every five days).

To get the specific activity of the PM emissions, ambient sampling is recommended or use of

site-specific information.

Table 7-13 lists all of the required data elements to implement this approach and also includes information on how to develop or where to obtain the required data.

Required Data	Equation Variable	Comments
Waste material load per	L	If local information is not available to estimate the waste load
noor space unit area, Mg/m		per unit area, then a default value of 0.45 can be used.
Building or total floor space	А	Surface area of demolition activities can be calculated from
area, m ²		maps or measured directly
Control efficiency of	CE	Control efficiencies are in terms of percent reduction in
mitigation approach		emission; for example, a CE of 100% indicate that pollutants
(percent)		are completely controlled, while a CE of 10% indicate that
		10% of emissions are captured and 90% of emissions are
		released into the atmosphere.
Specific activity of material	a	Derived from ambient test data.
pCi/g		

 Table 7-13. Required Data for the Preferred Method for Building Demolition

Example Calculation Using the Preferred Method

During the inventory year, a 100,000 m² (1,076,900 ft²) building is demolished. No controls are used during the operation and the waste material load factor (L) is unknown. The specific activity of the emissions is 2.1×10^{-10} pCi/g. Equation 7-8 is used to estimate emissions for this activity:

$$E = 2.9 \times 10^{-2} \times L \times A \times (1 - CE/100) \times a/2.2 \times 10^{-3}$$

The default value for L is used in this example. When all of the required data are incorporated, equation 7-8 becomes:

$$E = 2.9 \times 10^{-2} \times 0.45 \times 100,000 \times (1 - 0/100) \times 2.1 \times 10^{-10}/2.2 \times 10^{-3}$$
$$E = 1.246 \times 10^{-4} \text{ pCi/yr}$$

7.4.1.7 Material Handling - Unpaved Roads

When a vehicle travels on an unpaved road, the force of the wheels on the road surface causes pulverization of surface material. Particles are lifted and dropped from the rolling wheels, and the road surface is exposed to strong air currents in turbulent shear with the surface. If there has been deposition of radioactivity on the road surface, the turbulent wake behind vehicles traveling on unpaved roads can resuspend these radionuclides into the atmosphere. See additional information about this source category in Section 4.2.

Preferred Method to Estimate Radionuclide Emissions from Unpaved Roads

The preferred method to estimate radionuclide emissions from unpaved roads is derived from an emission estimating approach included in AP-42. The following empirical equation may be used to estimate the quantity of emissions from an unpaved road.

$$E = \frac{k \times (s/12)^{\underline{a}} (W/3)^{\underline{b}} \times [(365 - p)/365] \times V \times A/2.2 \times 10^{-3} \times (1 - CE/100)$$
(7-9)
$$(M_{dry}/0.5)^{\underline{c}}$$

Where:

E	=	Emissions (pCi/year)		
S	=	Surface material silt content (%)		
12, 3, 0.5	=	Dimensionless constants		
W	=	Mean vehicle weight (t)		
M _{dry}	=	Surface material moisture content under dry, uncontrolled conditions (%)		
365	=	Days per year		
р	=	Number of days with at least 0.254 mm (0.01 in) of precipitation		
per	•			
		yr		
V	=	Vehicle miles traveled (miles)		
Α	=	Specific activity of the road surface (pCi/g of soil)		
2.2×10^{-3}	=	Conversion factor (g/lb)		
CE	=	Control efficiency of mitigation activities (% removal)		
k, a, b and c are empirical constants provided in Table 7-14 below. The k value				
can vary significantly depending upon the particle size. If the specific activity is in terms of $PM_{2.5}$ or fine particulate matter, then the k value for $PM_{2.5}$ should be used in equation 7-9, otherwise the value associated with PM_{10} should be used.				

Table 7-14. Constants for Equation	n
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Constant	PM _{2.5}	PM_{10}
k	0.38	2.6
a	0.8	0.8
b	0.4	0.4
c	0.3	0.3

It should be emphasized that the moisture content to be used in the above equation (M_{dry}) must reference dry, worst-case conditions. Vehicle speeds can also be a factor in the emission of PM associated radionuclides. Typical vehicle speeds range from 5 to 55 mph (8 to 88 kmph). In cases where the mean vehicle speed is less than 15 mph (24 kmph), it is recommended that the above equation be multiplied by (S/15), where S is the average vehicle speed (mph).

Mitigation activities can significantly reduce the amount of PM emissions associated with unpaved roads. Some mitigation activities include:

- Vehicle restrictions that limit the speed, weight or number of vehicles on the road;
- Surface improvement by measures such as (a) paving or (b) adding gravel or slag to a dirt road; and
- Surface treatment such as watering or treatment with chemical dust suppressants.

For details about the control efficiency associated with each mitigation activity refer to Section 13.2.2 of AP-42. Table 7-15 lists all of the required data elements to implement this approach and also includes information on how to develop or where to obtain the required data.

Required Data	Equation Variable	Comments
Surface material silt content (%)	S	Silt content of road material can range from 1.2 to 35%. Site-specific data should be used derived from samples of road dust.
Mean Vehicle Weight (t)	W	Vehicle weights can vary from 1.5 to 290 tons (1.4 to 261 Mg). Site-specific data should be used, but if site-specific mean vehicle weight data are not readily available than a default value of 2.2 tons (2.0 Mg) can be used.
surface material moisture content (%)	M _{dry}	It should be emphasized that the moisture content to be used must reference dry, worst-case conditions. These values can vary from 0.03 to 20%, depending upon local soil composition and rainfall. In the absence of the appropriate site-specific information, the default value of 0.2 % should be used.
number of days with at least 0.254 mm (0.01 in) of precipitation per year	р	Obtain from nearest meteorological monitoring station.
vehicle miles traveled	VMT	To estimate annual VMT, the number of vehicles traveling on a given section of road is multiplied by the length in miles of the road section.
specific activity of the road surface (pCi/g of soil)	А	Derived from site-specific samples of road surface
control efficiency of mitigation activities (% removal)	CE	For details about the control efficiency associated with each mitigation activity refer to Section 13.2.2 of <i>AP-42</i> .
Empirical constants	k, a, b, c	Provided in Table 7-14

Table 7-15.	Required Data for	the Preferred Method f	for Unpaved Road	Traffic
Table 7-13.	Required Data 101	the freeffed Method i	of Onpaveu Roau	11 anne

Example Calculation Using the Preferred Method

10 vehicle trips are made every day along a 19 km (12 mile) stretch of unpaved road. The road has a silt content of 4%. The average vehicle weight is (4.5 Mg) 5 tons, these vehicles travel at speeds in excess of 24 kmph (15mph). The surface moisture content is unknown. During the inventory year, there are 20 days when the rain was greater than 0.025 cm (0.01 inch). To help control PM emissions, used oil was occasionally applied to the road surface. The expected control efficiency of this activity is approximately 10%. To estimate the radionuclide emissions associated with this stretch of unpaved road, equation 7-9 is used:

$$E = \frac{k \times (s/12)^{a} (W/3)^{b}}{(M_{drv}/0.5)^{c}} \times [(365 - p)/365] \times V \times A/2.2 \times 10^{-3} \times (1 - CE/100)$$

Constants k, a, b, and c are obtained from Table 7-14 for PM_{10} . Because the surface area moisture content is unknown, the default value of 0.2% is used in this example. VMT is calculated by multiplying the number of vehicle trips per day by the number of days in a year, and the length of the road in miles (e.g. $10 \cdot 365 \cdot 12 = 43,800$ VMT). When all of the required data are incorporated, equation 7-9 becomes:

$$E = \frac{2.6 \times (4/12)^{0.8} \times (5/3)^{0.4} \times [(365 - 20)/365)] \times 43,800 \times (2.1 \times 10^{-7}/2.2 \times 10^{-3}) \times (1 - 10/100)}{(0.2/0.5)^{0.3}}$$

$$E = 6.197 \text{ pCi/yr}$$

7.4.2 <u>Water/Evaporation</u>

For radionuclide emissions associated with evaporation or transpiration of water, there are emission estimating methods for three related source categories including:

- Evaporation from ponds and lagoons;
- Evaporation from cooling towers; and
- Evapotranspiration from contaminated soil and plants.

7.4.2.1 Evaporation from Ponds and Lagoons

Tritium, in the form of titrated water (HTO), is the principal radionuclide that can be released by evaporation or volatilization from open bodies of water such as ponds and lagoons. HTO is simply water (H₂O) with one of the hydrogen atoms (¹H) replaced by tritium, ³H. Since HTO is chemically almost indistinguishable from water, the most appropriate way to model its release is to assume that the water vapor emitted from the surface of a pond has the same specific activity of tritium as the water in the pond itself. For additional information about this source category see Section 5.1.1.

One preferred method and one alternative method to calculate emissions associated with lagoons and ponds were identified. These emission estimating approaches are discussed below.

Preferred Method to Estimate Emissions From Ponds and Lagoons

The evaporation of HTO from ponds and lagoons is governed by the air temperature, vapor pressure, wind speed, surface area, and the specific activity of HTO in the water body. The following formula has been developed to estimate the surface water evaporation rate (see Chapter 5, equations 5-1 and 5-2):

$$R = (20.73 \times P_s \times A^{0.9} \times U^{0.8} \div T^{1.47}) \times a \times 3600$$
(7-10)

Where:

R	= Emission rate of tritium, (pCi/hr)
20.73	= Dimensionless constant
А	= Surface area of pool, (m^2)
Ps	= Equilibrium vapor pressure of water at ambient temperature, (mm Hg)
U	= Wind speed, (m/s)
Т	= Absolute temperature, (in K or Ambient temperature, in $C + 273.2$)
а	= Specific activity of water, (pCi/g)
3600	= Conversion, (s/hr)

Ideally, the annual emissions should be calculated by integrating the emission rate, using hourly average wind speeds and temperatures and specific activity measured at various times during the year. Table 7-16 lists all of the required data elements to implement the preferred method and also includes information on how to develop or where to obtain the required data elements.

 Table 7-16. Required Data for Preferred Surface Water Evaporation Method

Required Data	Equation Variable	Comments
Surface area of pool		Surface areas can be calculated for regularly shaped surfaces or
(m^2)	A	estimated using maps for irregularly shaped impoundments.
Equilibrium vapor pressure	Ps	Can be obtained from a variety of references such as Perry's
of water		Chemical Engineers' Handbook or a psychrometric chart. Ambient
(mm Hg)		temperature is required.
Wind speed and ambient	U & T	Average wind speed and ambient temperature can be obtained from
temperature		nearest meteorological monitoring station. DOE has developed a
(m/s and ^o K, respectively)		default average annual wind speed of 2 m/s [6.6 ft/s] (Table 1.3
		from Data Collection Hardbook to Support Modeling Impacts of
		Radioactive Material in Soil (April 1993). To get the ambient
		temperature in terms of °K, 273.2 should be added to the ambient
		temperature, which should be in terms of °C.
Specific activity of tritium in	а	Derived from site-specific surface water samples.
the water		
(pCi/g)		

This equation assumes that the water and air are at the same temperature, ignoring that evaporative cooling would tend to reduce the vapor pressure and hence the evaporation rate. The

net evaporation rate is a balance of evaporation from the surface and condensation onto the surface of the open body of water from the ambient water vapor in the atmosphere. However, only the one-way process (i.e., vapor exiting the water body surface), called the surface volatilization rate, is the pathway for the release of tritium, as ambient water vapors that condense are assumed to be free of tritium. On the other hand, some of the titrated vapor will recondense, reducing the net flux of tritium to the atmosphere. The recondensing of tritium back into the surface water impoundment is not considered in the above equation, such that actual emissions may be lower than those predicted by this equation. Figure 7-6 shows the evaporation/condensation cycle associated with emissions from surface impoundments.

Example Calculation Using the Preferred Method

A facility has a pond with a surface area of $10,000 \text{ m}^2 (107,640 \text{ ft}^2)$ with a specific activity of 200 pCi/g. For the hour of concern, the wind speed is 0.5 m/s (1.64 ft/s) and the ambient temperature is 18°C which equates to 291.2°K. At this ambient temperature, the vapor pressure of water is 15.477 mm Hg (2,063 Pascals). To estimate emissions using the preferred approach for ponds and lagoons, the following equation should be used.

$$R = (20.73 \times P_s \times A^{0.9} \times U^{0.8} \div T^{1.47}) \times a \times 3600$$

When the required data are incorporated the equation becomes:

$$\mathbf{R} = (20.73 \times 15.477 \times (10,000)^{0.9} \times (0.5)^{0.8} \div (291.2^{1.47})) \times 200 \times 3600$$

And the final calculations are:

$$R = (20.73 \times 15.477 \times 3,981.07 \times 0.574 \div 4,191.42) \times 200 \times 3600$$

= 1.259 × 10⁸ pCi/hr.

Alternative Method to Estimate Emissions From Ponds and Lagoons - Mass Balance

If data are not available to implement the preferred method, this alternative method would then be recommended. Tritium emissions can be estimated by assuming that the entire tritium activity that is discharged into an open body of water during the year evaporates during the same year, as noted in the following equation (DOE 92). The approach implies that the annual inflow and evaporation of tritium are the same, but users should be aware that this is not necessarily the case in all situations.

$$\mathbf{R} = \mathbf{Q} \times \mathbf{C}_{\mathbf{a}} \tag{7-11}$$

Where:

R	= Emission rate of tritium, (pCi/yr)
Q	= Flow into impoundment, (m^3/yr)
C_a	= Specific activity of water, (pCi/m^3)



Table 7-17 lists all of the required data elements to implement this approach and also includes information on how to develop or where to obtain the required data elements.

 Table 7-17. Required Data for Mass Balance Surface Water Evaporation Method

	Equation	
Required Data	Variable	Comments
Annual flow into	Q	Data from flow monitors can be used or estimates based on
impoundment (m ³ /yr)		water usage patterns.
Specific activity of tritium in	Ca	Derived from site-specific surface water samples.
the water (pCi/m^3)		

This method is said to be conservative in that losses of tritium through ground seepage are neglected. This approach provides reasonable emission estimates if neither the volumes nor the specific activities of the liquid effluents varied appreciably throughout the year.

In order to provide the most accurate emission estimates with this method, it is recommended that specific flow and specific activity data be used for each pond or lagoon at a facility.

7.4.2.2 Evaporation From Cooling Towers

Wet-cooling towers are heat exchangers used to dissipate heat from industrial processes. Water is used as the medium to transfer heat away from the cooling coils that contain hot process fluids. Wet-cooling towers can either be designed as a natural draft tower, where the heat from the cooling water creates vertical air flow, or it can be an induced draft tower, where the air is discharged through the cooling tower via a fan or series of fans. Under normal conditions, the cooling water and the process water never mix. Cooling coils are made of thin metal to facilitate heat transfer increasing the likelihood of leaks. In the event of a leak, the cooling fluid may become contaminated by the process fluid. Tritium and noble gases (e.g., xenon, argon, radon), are efficiently dispersed by cooling towers, since they are designed to be effective aerators, enhancing evaporation of volatile pollutants.

Within the tower, some of the cooling fluid is drawn up as droplets by convection currents and released as "drift" droplets. As the water evaporates, the droplets leave behind fine particulate matter formed by the crystallization and agglomeration of less volatile dissolved solids.

Air emissions from cooling towers include evaporation losses and drift losses. Figure 7-7 shows the emission points typically associated with cooling towers. The emission of radioactivity from wet-cooling towers is further complicated by the possible speciation of radioactivity in the process water. For examples of speciation issues see Section 5.1.2. Given these various considerations, estimating release rates for radionuclides from wet-cooling towers should be evaluated on a case-by-case basis.

One preferred method and one alternative method to calculate emissions associated with cooling towers were identified. These emission estimating approaches are discussed below. For additional information on this source category see Section 5.1.2.

Preferred Method to Estimate Emissions From Cooling Towers

The preferred method allows for each component of cooling tower emissions to be calculated separately using the following equations (CHE 02).

Evaporative Losses

Evaporative losses can be estimated by the following equation (see Chapter 5, Equation 5-4 for more details).

EL =
$$(8.5 \times 10^{-4} \times Q \times (T_1 - T_2)) \times a$$
 (7-12)



EL

EL	= Evaporative losses (pCi/hr)
8.5 ×	10^{-4} = Evaporation coefficient (dimensionless)
Q	= Flow rate of cooling water (m^3/hr)
T_1	= Hot water temperature, ^o F
T_2	= Cold water temperature, ^o F
a	= Specific activity of cooling water (pCi/m^3)

Drift Losses

It is estimated that between 0.1 and 0.2% of the water supply is lost to drift. Some towers are equipped with drift or mist eliminators to minimize such emissions. To estimate drift emissions the following equation can be used (see Chapter 5, equation 5-5 for more details).

$$DL = (0.002 \times Q) \times (a) \times (1 - CE/100)$$
(7-13)

Where:

Table 7-18 lists all of the required data elements to implement this approach and also includes information on how to develop or where to obtain the required data elements.

Table 7-18. Required Data for Preferred Wet-Cooling Tower Method

Required Data	Equation Variable	Comments
Cooling water flow rate (m^3/hr)	Q	Data from flow monitors can be used or estimate based on water usage patterns.
Specific activity of tritium in the cooling water (pCi/m ³ and pCi/g)	a	Derived from site-specific cooling water samples.
Inlet temperature (°F)	T_1	Temperature of the hot process water entering the cooling tower, obtained from temperature monitoring device.
Outlet temperature (°F)	T ₂	Temperature of the cooled process water leaving the cooling tower, obtained from temperature monitoring device.
Control efficiency (%)	CE	Control efficiency of drift or mist eliminator as provided by the manufacturer.

In order to provide the most accurate emission estimates it is recommended that facility- specific flow, specific activity, temperature, and solids ratio data be used in the above method.

Example Calculation Using the Preferred Method

The example facility has process water at 310° K (100° F) which needs to be reduced to 300° K (80° F) with a cooling water flow rate of 227 m³/hr (8,015 ft³/hr). The specific activity of the cooling water is estimated to be 200 pCi/g which equates to approximately 2.0×10^{8} pCi/m³. The drift eliminators used in this cooling tower have a control efficiency of 99.5%.

Evaporative Losses

EL =
$$(8.5 \times 10^{-4}) \times Q \times (T_1 - T_2) \times a$$

= $(8.5 \times 10^{-4}) \times 227 \times (20) \times 2.0 \times 10^{8}$
= $7.718 \times 10^{8} \text{ pCi/hr}$

Drift Losses

DL = $(0.002 \times Q)(1-(CE/100)) \times a$ = $(0.002 \times 227) \times (1-(99.5/100)) \times 2.0 \times 10^8$

 $= 4.54 \times 10^5 \text{ pCi/hr}$

Total Cooling Tower Losses

Total Cooling Tower Emissions = Evaporative Losses + Drift Losses = $7.718 \times 10^8 + 4.54 \times 10^5$ = 7.723×10^8 pCi/hr

Alternative Method to Estimate Emissions From Cooling Towers - Available Emission Factors

If data are not available to implement the preferred approach, then this alternative approach would be recommended. In *AP-42*, EPA has estimated the overall PM_{10} emission rate from a typical cooling tower to be about 2.3×10^{-3} g/L (1.4×10^{-4} lbs/ft³) of circulating water flow, based on limited data for induced draft cooling towers (EPA 95). However, no data were provided for natural draft cooling towers. The emission rate given by EPA is also believed to be typical of older towers with less efficient mist eliminators.

The U.S. EPA emission factor has been incorporated into the following equation which allows for estimation of radionuclide emissions by including the specific activity of the cooling water.

$$TL = (2.3 \times 10^{-3}) \times (1000 \times Q) \times (a)$$
(7-14)

Where:

TL = Total loss (pCi/hr) 2.3×10^{-3} = Emission factor (g/l) 1000 = Conversion factor (l/m³) Q = Flow rate of cooling water (m³/hr) a = Specific activity of cooling water (pCi/g) Table 7-19 lists all of the required data elements to implement this approach and also includes information on how to develop or where to obtain the required data elements.

Table 7-19. Required Data for the Alternative AP-42 Cooling Tower Model	ethod
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	Equation	
Required Data	Variable	Comments
Cooling water flow rate	Q	Data from flow monitors can be used or estimates based on
(m^{3}/hr)		water usage patterns
Specific activity of cooling	а	Derived from site-specific cooling water samples.
water		
(pCi/g)		

In order to provide the most accurate emission estimates, it is recommended that facility-specific flow and specific activity data be used in the above emission estimating equation.

7.4.2.3 Evapotranspiration From Contaminated Soil

Evapotranspiration is the loss of water to the atmosphere via the combined processes of evaporation and transpiration. Evaporation can be a significant release mechanism for certain types of radioactivity from soils contaminated with water that contains tritium or carbon-14. In certain instances, transpiration of plants with large root systems may also substantially contribute to tritium re-emission. The transpiration process is passive and is governed by the humidity of the atmosphere and the moisture content of the soil. The rate of transpiration depends on both the soil depth profile and the plant's distribution of roots.

For top soils that are saturated with contaminants, one preferred method to calculate emissions was identified. Where the top soils are dry and the contaminated soils are below the top soil, one preferred method to calculate emissions was identified. These emission estimating approaches are discussed below. No alternative methods were identified for evapotranspiration from contaminated soils. For additional information about this source category see Section 5.2.

Evapotranspiration From Saturated Soils

Preferred Method for Estimating Emissions from Saturated Soils

Emissions associated with spills of liquid contaminants, where liquid pools are visible on the soil surface or where the soil is saturated from the surface on down, should be estimated in the same manner as surface water impoundments using the methods noted in Section 7.4.3.1 above. For situations where top soils are moist, but contamination is not visible, the *RESRAD* model is recommended (DOE 01). This model was developed by DOE and can be used to estimate emissions of both tritium and carbon-14. The following flux rate equations from the *RESRAD* model applies to both tritium and carbon-14.

For tritium, the flux rate, is derived as follows.

$$F_{i-trit} = W_T \times C_e \times [(1-C_r) \times P_r + I_r] \times A$$
(7-15)

September 3, 2004
Where:

F _{i-trit}	= Contaminant flux (evasion rate) from soil, (pCi/yr)
W_{T}	= Tritium concentration in soil water, (pCi/m^3)
Ce	= Evaporation coefficient, (dimensionless)
Cr	= Runoff coefficient, (dimensionless)
Pr	= Annual rainfall rate, (m/yr)
Ir	= Irrigation rate, (m/yr)
Α	= Size of contaminated area, (m^2)

For carbon-14, the flux rate is derived as follows.

$$F_{i-c-14} = S_c \times E_c \times \rho_b \times d_s \times 10^6 \times A$$
(7-16)

Where:

F _{i- c-14}	= Contaminant flux (evasion rate) from soil, (pCi/yr)
Ec	= Evasion loss rate constant, (yr^{-1})
Sc	= Carbon-14 concentration in soil sample, (pCi/g)
$ ho_b$	= Soil bulk density, (g/cm^3)
ds	= Soil depth, (m)
10^{6}	= Conversion factor, (cm^3/m^3)
А	= Size of contaminated area, (m^2)

Table 7-20 lists all of the required data elements to implement this approach and also includes information on how to develop or where to obtain the required data elements.

In order to provide the most accurate emission estimates, it is recommended that appropriate facility-specific data be used in the above method.

Example Calculation Using the Preferred Method

The contaminated area is approximately 40,000 m² (9.9 acres) with contamination extended to a depth of 0.254 m (10 in) from the surface of loamy top soil. Annual rainfall is 0.457 m/yr (18 in/yr) and the area is not subject to irrigation. The soil is of mixed material with a density of 1.6 g/cm³ (100 lb/ft³), with a tritium concentration in soil water of 2.0×10^8 pCi/m³ and a carbon-14 concentration in the soils of 0.14 pCi/g. Values for the evaporation coefficient (Ce) and the runoff coefficient (Cr) are taken from Table 7-20.

Table 7-20. Required Data for Evapotranspiration Preferred-Saturated Soils Method

Required Data	Equation Variable	Comments
Size of contaminated area (m^2)	А	Surface areas can be calculated for regularly shaped surfaces or estimated using maps for irregularly shaped impoundments.
Tritium concentration in soil water (pCi/m ³)	W _t	Obtain from site-specific sampling and analysis of soil water.
Evaporation coefficient (dimensionless)	C _e	DOE assumes a default evaporation coefficient of 0.5.
Runoff coefficient (dimensionless)	Cr	Values range from 0.1 to 0.4 for agricultural soils and woodlands and 0.4 to 0.65 for urban environments. DOE has developed a default value of 0.2.
Annual rainfall rate (m/yr)	Pr	Annual rainfall can be obtained from nearest meteorological monitoring station.
Irrigation rate (m/yr)	Ir	Irrigation rates range from 0 to 10 m. Data from flow monitors can be used or estimates based on water usage patterns. Alternatively, the DOE has developed a default value of 0.2 m/yr.
Evasion loss rate constant (yr ⁻¹)	Ec	The evasion loss rate is the fraction of soil inventory lost to the atmosphere per unit time. The following evasion rates were obtained from the <i>RESRAD</i> users manual:Clay 12 Loamy soils12Organic soils22Sandy Soils22Stable carbon in carbonate soils 3.2×10^{-3} Water 0.91
Carbon-14 concentration in soil (pCi/g)	S _c	Obtain from site-specific sampling and analysis of soil.
Soil bulk density (g/cm ³)	ρ	Obtain from sampling and analysis of soil density using US Army Corp of Engineers Laboratory Soils testing, EM 1110-2- 1906 (1970). The soil bulk density can range from near 0 to 100 g/cm ³ . The DOE has developed default value of 1.5 g/cm ³ .
Soil depth (m)	d _s	Obtain from measurement of topsoil depth. DOE has a default value of 0.3 m

For tritium, the flux rate, F_i, is derived as follows.

$$F_{i\text{-tritium}} = W_{T} \times C_{e} \times [(1-C_{r}) \times P_{r} + I_{r}] \times A$$

= 2.0 × 10⁸ × 0.5 × [(1-0.2) × 0.457 + 0] × 40,000
= 1.4624 × 10¹² pCi/yr

For carbon-14, the flux rate, is derived as follows:

$$\begin{split} F_{i\text{-}c14} &= S_C \times E_c \, \times \rho_b \, \times d_s \, \times \, 10^6 \times A \\ &= \, 0.14 \, \times 12 \, \times \, 1.6 \, \times \, 0.245 \, \times \, 10^6 \, \times \, 40,\!000 \\ &= \, 2.634 \, \times \, 10^{10} \, p\text{Ci/yr} \end{split}$$

Subsurface Contamination

Preferred Method to Estimate Emissions From Subsurface Contamination

In cases where the surface layer of the soil is dry and devoid of tritium, but titrated water remains below the surface, the EPA has developed an equation that can be used to calculate a more realistic release rate than that produced by the surface evaporation models (EPA 88c). The tritium emission rate for subsurface contaminations may be calculated as follows.

$$E_{i} = D_{i} \times A \times 10^{4} \times (P_{t}^{4/3} \times [M_{i} \div d_{sc}]) \times C_{si} \times 3600 \times a$$
(7-17)

Where:

$E_i \\ D_i \\ A \\ 10^4 \\ P_t$	Pt b r	 = Emission rate, (pCi/hr) = Diffusion coefficient (cm²/s) = Contamination area, exposed, (m²) = Conversion factor (cm²/m²) = Total soil porosity, (dimensionless), where: = 1- (b ÷ r) = Soil bulk density, (g/cm³) = Particle density, (g/cm³)
$\begin{array}{c} M_i \\ d_{sc} \\ C_{si} \end{array}$	= Mol = Effe = Satu	e fraction of contamination in soil, (unitless) ective depth of soil cover, (cm) uration vapor concentration, (g/cm ³), where:
	C _{si} P MW _i R T	 P • MW_i÷ R • T Vapor pressure of contaminant, (mmHg) Molecular weight of contaminant, (g/mole) Molar gas constant, (62,361 mmHg-cm³/mole-°K) Absolute temperature, (°K)

3600 = Conversion factor (sec/hr) a = Specific activity of groundwater (pCi/g)

Table 7-21 lists all of the required data elements to implement this approach and also includes information on how to develop or where to obtain the required data elements.

Required Data	Equation Variable	Comments
Diffusion coefficient (cm ²)	D _i	Can use $0.2 \text{ cm}^2/\text{s}$, if site-specific data are not available.
Contamination area, exposed (cm ²)	А	Surface areas can be calculated for regularly shaped surfaces or estimated using maps for irregularly shaped impoundments.
Mole fraction of contamination in soil, (dimensionless)	M_i	Total moles of the solute over the total moles of the solution.
Effective depth of soil cover (cm)	d _{sc}	Obtain from measurement of topsoil depth.
Total Porosity	P _t	Values range from <0.01 to 0.725, depending upon soil type. Example P_t values are provided in DOE(01) table 3-1 or 3- 2. The DOE has a default value of 0.4 that can be used.
Soil bulk density (g/cm ³)	b	Obtain from sampling and analysis of soil density using US Army Corp of Engineers Laboratory Soils testing method, EM 1110-2-1906 (1970). The soil bulk density typically ranges from 1 to 2 g/cm ³ . The DOE has developed default value of 1.5 g/cm ³ .
Particle density (g/cm ³)	r	Obtain from analysis of soil samples. Alternatively, the EPA has developed a default value of 2.65 g/cm^3
Vapor pressure of contaminant (mm Hg)	Р	For HTO, assume vapor pressure similar to water which can be obtained from references such as Perry's engineering handbook.
Molecular weight of contaminant (g/mole)	MWi	For HTO, use 22.0321 g/mole.
Molar gas constant (mm Hg-cm ³ /mole-°K)	R	62,361 mmHg-cm ³ /mole-°K
Absolute temperature (K)	Т	Obtain from nearest meteorological monitoring station. To get the ambient temperature in terms of °K, 273.2 should be added to the ambient temperature, which should be in terms of °C.
Specific activity of ground water (pCi/g)	а	Derived from site-specific groundwater samples.

 Table 7-21. Required Data for Subsurface Transpiration Preferred Method

In order to provide the most accurate emission estimates, it is recommended that appropriate facility-specific data be used in the above method.

Example Calculation Using the Preferred Method

The assumptions for the example are as follows: $40,000 \text{ m}^2$ (9.9 acre) area, annual wind speed of 0.5 m/sec (1.64 ft/s), with an annual rainfall of 0.457 m/yr (18 in/yr) and an average ambient temperature of 18°C (291°K). The area is not subject to irrigation activities. Contamination extended to 0.254 m (10 in) from the surface of the top soil. The soil is of mixed material with a density of 1.6 g/cm³ (100 lbs/ft³). The specific activity of the ground water is 200 pCi/g. The mole fraction of the contamination is 0.001 and the vapor pressure of HTO is assumed to be similar to water which at 10 °C (283°K) would be 9.209 mm Hg (1,227 Pascals).

$$E_i = D_i \times A \times 10^4 (P_t^{4/3} \times [M_i \div d_{sc}]) \times C_{si} \times 3600 \times a$$

Where:

P_t

$$= 1 - (b \div r) = 1 - (1.5/2.65)$$

and

$$C_{si} = P \times MW_i \div R \times T = 9.209 \times 22.0321 \div 62,361 \times (273.2+18)$$

After substitution, the equation can be expressed as:

$$\begin{array}{rcl} E_i &=& 0.2 \ \times \ (40,000 \times 10^4) \times ((1\text{-}1.5/2.65)^{4/3} \ \times \ [0.001 \div 25.4]) \times (9.209 \times 22.0321 \div \\ &\quad 62,361 \times (273.2 + 18)) \times 3600 \times 200 \\ &=& 7.06 \times 10^8 \ pCi/hr \end{array}$$

7.4.3 <u>Underground Testing</u>

Currently models are being developed by DOE to estimate radionuclide emissions from re-entry drilling and ground seepage of noble gases. Until the models have been validated, the preferred method to quantify emissions is to use ambient air testing and analysis. For details see report Sections 5.3.1 and 5.3.2.

7.4.4 Emissions from Buildings and Equipment

Fugitive emissions from buildings and equipment may occur in a facility where sources of radioactive material are handled. The emissions from buildings tend to be emitted via pressure or temperature differences between the building and the atmosphere. Equipment emissions may be associated with built-in system features (e.g., filtration systems) or inherent in the process (e.g., air displaced by a waste compactor ram). The radionuclides released within a building or as equipment leaks are not actively ventilated into the atmosphere. For additional information on these diffuse source categories see report Sections 5.3.3 and 5.3.4.

Preferred Method to Estimate Emissions From Buildings, and Equipment

Emissions can be estimated by determining the volume of material released, its concentration, and application of any mitigation factor, as noted in the following equation (see Chapter 5, equations 5-15 and 5-19 for further details):

$$E = \sum_{i}^{n} R_{i} \times T_{i} \times a_{i} \times (1 - CE / 100)$$
(7-18)

Where:

E	= Sum of all releases over all events i, (Ci)
R _i	= Release rate for event i, (m^3/s)
T _i	= Duration of release i, (s/event)
ai	= Specific activity of event, (Ci/m^3)
CE	= Percent control efficiency of the device used to reduce emissions (%)

Control efficiency (CE) should not be applied where passive flow compromises efficiency; for example, HEPA filters under passive flow cannot be held to their high efficiency rates.

The mitigation factor may be used to account for the use of devices or processes that reduce the amount of materials released. Such devices may include HEPA filters, baghouses, scrubbers, adsorber beds, etc.

Table 7-22 lists all of the required data elements to implement this approach and also includes information on how to develop or where to obtain the required data.

Required Data	Equation Variable	Comments
Release Rate (m ³ /s)	R _i	For a given event, it is necessary to estimate the rate (in terms of volume) at which the emissions are released. This would have to be estimated based on site-specific data.
Duration of the release (s/event)	T _i	The length of time for which the release occurred would have to be estimated based on site-specific information.
Specific activity of the event (Ci/m ³)	a	Derived from site-specific air samples.
Control efficiency of the devices used to mitigate the emission release (%)	CE	Percent emission reduction associated with a control device, manufacturer's data can be used, but should be validated with inlet and outlet test data. Note, for some control devices, such as HEPA filters, passive flow compromises control efficiency.

Table 7-22. Required Data for Preferred Emission Estimating Method forBuildings and Equipment

Example Calculation Using the Preferred Method

In replacing a filter in process equipment that handles radioactive material, 2 m^3 /second (70.6 ft³/s) of emissions are released for a period of 180 seconds. The specific activity of the waste stream is 10^{-6} Ci/m³. The emissions are not captured or controlled so the control efficiency of the event can be assumed to be zero. To estimate emissions using the preferred approach for buildings and equipment the following equation should be used.

$$E = R_i \times T_i \times a_i \times (1 - CE/100)$$

When the required data are incorporated the equation becomes:

$$E = 2 \text{ m}^3/\text{sec} \times 180 \text{ sec} \times 10^{-6} \text{ Ci/m}^3 \times (1 - 0/100)$$

And the final calculations are:

 $E = 2 \text{ m}^3/\text{sec} \times 180 \text{ sec} \times 10^{-6} \text{ Ci/m}^3 \times 1$ E = 0.00036 Ci/Event

7.4.5 <u>Emissions from Chemical Storage Tanks</u>

Chemical storage tanks that hold radioactive material may emit radionuclide emissions through working and breathing losses. Working losses are emissions from the displacement of vapors as chemical storage tanks are filled. Breathing losses are diurnal emissions associated with changes of ambient temperature. As ambient temperatures increase, vapors expand and may be emitted into the atmosphere. For additional information on this source category see Section 5.3.4.

Preferred Method to Estimate Emissions From Chemical Storage Tanks

The EPA OAQPS developed and maintains a model to estimate VOC emissions from chemical storage tanks, which is derived from the *AP-42* equations noted in Sections 5.3.4. The TANKS program is designed to estimate air emissions from organic liquids in storage tanks. TANKS allows users to enter specific information about a storage tank (dimensions, construction, paint condition, etc.), the liquid contents (chemical components and liquid temperature), and the location of the tank (nearest city, ambient temperature, etc.), and generate an air emissions report. Report features include estimates of monthly, annual, or partial year emissions for each chemical or mixture of chemicals stored in the tank. The model accounts for both working and breathing losses. The TANKS software can be downloaded from the following EPA web site:

http://www.epa.gov/ttn/chief/software/tanks/index.html

If it is assumed that the radionuclide concentration of the chemicals stored in the tank is similar to the radionuclide concentration of the vapors emitted, then the following equation can be used to adjust the output from the TANKS model to estimate radionuclide emissions.

 $L = E \times a$

(7-19)

Where:

- L = Total storage tank emission (Ci/yr)
- E = VOC emission output from TANKS model (lb/yr)
- a = Specific activity of stored chemicals (Ci/lb)

7.5 Collect Required Data and Calculating Emissions

For the methods identified above in Section 7.4, the specific data required to estimate radionuclide emissions are noted in the summary tables associated with each emission estimating procedure. Where facilities have multiple or grouped diffuse sources for which they are estimating emissions, it is recommended that the different summary data tables be combined into a master data collection table listing all of the required data elements. This approach allows for easy identification of duplicate data elements, streamlining the data collection process and facilitates a data structure to organize the compiled data. When collecting the required data, it is very important that the source of the data and the time period that the data represents be documented.

In most cases, the required data can be easily incorporated into a database, spreadsheet, or other software tools to facilitate emission calculations, particularly if the data are available in an electronic format. Where the emission estimating procedures calculate hourly radionuclide emission rates, that are applied to hourly meteorological data to estimate ambient radionuclide concentrations, such data sets may be too large for some software tools. The limitations of the software should be considered prior to deciding on what spreadsheet or database software the required data are to be stored. Where the size of the data set is an issue, it may be possible to solve the software limitation by developing multiple smaller data sets.

Once the required data have been compiled and checked to insure that the data set is complete and the values reasonable (see Section 7.6), emissions can be estimated using the equations provided in Section 7.4.

Emission estimates associated with each source category should not be aggregated and presented only as a facility total. Grouped emission sources should be disaggregated into individual diffuse source categories when possible. Disaggregated emission estimates are particularly useful in differentiating the significance between the different diffuse emission sources, which may be useful in prioritizing control strategies. Disaggregated emission estimates also allow for independent evaluation of the reasonableness of the estimated emissions.

7.6 Quality Assurance

It is important that quality checks be performed throughout the inventory processes to insure that errors are identified and corrected early. Errors that are identified later in the process often require considerably more work to correct. Emission inventory quality assurance activities usually require development of a formal Quality Assurance Plan (QAP) that identifies the quality checks to be performed and the procedures for correcting and documenting errors which have been encountered. The preparation of a QAP is often a useful exercise in helping to determine

which emission estimation method to use and how data for the method should be collected. For further guidance on QAPs, see the U.S. EPA's EIIP *Volume VI-Quality Assurance Procedures and DARS Software* (http://www.epa.gov./ttn/chief/eiip/techreport/volume06/index.html).

The EPA now requires all QA documentation to be in accordance with EPA Order 5360.1 A2. All required formats and guidance documents for this requirement can be found at <u>http://www.epa.gov/quality/</u>.

The required data compiled in Section 7.5 should be checked to insure that it has been correctly incorporated into the estimation software. Where the data have been entered by hand, a double entry system may be warranted, where the data are entered into two separate databases by two different individuals. The two databases are compared electronically and those data that match are considered correctly entered. Data that do not match are investigated further to determine which of the two values are correct.

Where electronic data are used, summary quality checks can be performed to alert staff that data are missing or may be erroneous. These checks would include:

- 1) Reviewing the number of records in the data set to insure that all of the expected records have been loaded into the spreadsheet or database;
- 2) In some cases, it is useful to sum the values in the raw data set and compare these to the totals in the database. This type of check would make sure that correct values have been transferred;
- 3) For some of the more important data elements, it is helpful to compare minimum and maximum values to insure that the numbers included in the database are within a reasonable range of possible values. This check should also be performed on data that have been manually entered; and
- 4) If the data represent different spatial locations, the spatial codes that have been assigned should be checked to insure that they are valid codes.

If data entry errors are encountered, they should be corrected and documentation provided that show that the corrections have been made.

The equations or formulas used to estimate emissions need to be checked to insure that they too have been entered correctly and are functioning properly. This check should be performed by staff not involved in setting up the equations, to insure independent review of the calculating procedure. Once the compiled data and the equations have been checked, the emission estimates can be calculated. The resultant emission estimates should be checked to insure that they are reasonable. This can be done by reviewing previous year emission estimates for similar emission processes. If the values do not seem reasonable it may be necessary to review the collected data or emission estimating procedures. If errors are identified, corrections should be made and documentation provided for the identified problems.

7.7 Emissions Documentation and Reporting

In general, it is important to provide summary tables that include each of the diffuse source categories and each pollutant for which emissions were estimated. The data can be further disaggregated into emission tables for individual diffuse sources located throughout the facility. These summary tables can be supplemented with charts and maps to facilitate analysis of the diffuse source emissions data.

Documentation should also be provided concerning the procedures used to develop the diffuse source radionuclide emission inventory. This report should be sufficiently transparent to allow for independent replication of the emission estimates. This documentation should provide a clear description of how the diffuse emission sources were identified, what emission estimating procedures were used, and what data were collected for the inventory process.

The documentation should include details such as the emission estimating equations or emission factors and speciation profiles used in developing the emission estimates. If software tools are used to estimate emissions, details concerning these tools should also be provided such as version and release date of the product and any adjustment made to the software to better account for local conditions.

In some cases, necessary data are not readily available and to complete the calculations assumptions need to be made such as hours of operation or typical specific activity of wastewater. These assumptions need to be noted in the appropriate section of the text to insure that if the emission estimates are independently replicated, the reviewer is basing their emission estimates on the same assumptions.

As noted throughout this chapter, facilities are encouraged to work with the EPA in using as much site-specific data as possible to estimate diffuse source emissions. It is important to document the source of the data compiled and the time period that the data represent. Where site-specific data are not available, but default values are, the use of such default data should be noted in the documentation.

The documentation should also include the quality assurance plan developed for the inventory and summary results of the quality checks that were performed. Any limitations or deficiencies in the inventory should also be discussed in the documentation along with future plans to address these issues.

Comprehensive documentation will make it possible to assess the overall quality of the diffuse source emission estimates that have been submitted and evaluate the consistency of the estimates to diffuse source emission inventories developed at other DOE facilities.

7.8 Case Studies

The following case studies are provided to demonstrate what kind of activities have been performed by the DOE staff to quantify fugitive emissions. The procedures used in these case studies do not necessarily match up with the procedures discussed in this document as these

activities were performed prior to the publication of this report. Future editions of this document may include examples of activities that use the emission estimation approaches recommended in this report.

7.8.1 Oak Ridge Reservation (ORR)- Fugitive Emissions from Diffuse Sources

Background. In response to a March 1992 EPA Region IV guidance letter allowing use of environmental measurements to confirm compliance for diffuse sources with the Radionuclide NESHAP, the Oak Ridge Reservation (ORR) developed a plan to use a network of ambient air monitors to confirm that fugitive radionuclide emissions from diffuse sources do not result in atmospheric radionuclide concentrations that contribute significantly to the reported radiation doses to members of the off-site public. The plan was submitted to EPA Region IV in October 1992 and was implemented in January 1993.

ORR Diffuse Source Emissions Confirmation Program. In 2001, the Industrial Source Complex Short Term (ISCST3) atmospheric dispersion model was used to determine that ambient air monitor locations, which were sited based on an earlier modeling study, are appropriate to capture the effect of any fugitive and diffuse emissions from the three main operations centers of the ORR: the Oak Ridge National Laboratory (ORNL), the East Tennessee Technology Park (ETTP), and the Y-12 National Security Complex. Twenty-five potential fugitive emission sources on the ORR were evaluated - 15 from ORNL, 5 from ETTP, and 5 from the Y-12 Complex. These fugitive and diffuse sources were selected because they were identified to be those with the most significant potential for fugitive emissions. Fugitive emissions from the ORR are likely to be in the form of particulates from the resuspension of dust and soil.

Annual average air concentrations were calculated for numerous receptor points (10,201 receptors) positioned around each potential fugitive source. The center of each potential fugitive source was positioned at the center of a rectangular receptor grid (spacing of the Cartesian receptor grid was 100 meters). Annual average air concentrations were also calculated at the ambient air monitor locations when the ambient air monitors were located within the given source receptor grid. The large number of receptors improved graphic resolution.

Five years of onsite hourly meteorological data were used in the modeling (10 meter height meteorological data). Since source terms were not known for most of the sources modeled, a unit release per total area was assumed for each area source (1) and (1 g/s) for each volume source. Contour plots of annual average concentrations were made for each potential fugitive emission source to verify appropriateness of ambient air monitor placement.

Although the ISCT3 code does not calculate radiation doses, the model is useful for determining the transport and dispersion of radioactive materials. The concentration per unit emission obtained from the modeling can be multiplied by the actual site emissions of a given material to provide an estimate of the impact. In other words, this technique calculates the Chi/Q value for each receptor affected by the potential fugitive source. This approach can be used to quantify radionuclide emissions associated with particulates from the resuspension of dust and soil. However, it is very difficult to determine the contribution from fugitive emissions to the measured radionuclide concentrations unless there were no other sources on the ORR or if these fugitive sources had unique radionuclides, or other non-DOE sources. This is not the case on the ORR.

Using ISCST3 instead of a dose model, such as CAP88, removes the uncertainty typically associated with these codes (e.g., uncertainties associated with source term and terrestrial transport). Some uncertainties (associated with Gaussian equations, terrain effects, meteorology, and particulate deposition characteristics) are a few of the uncertainties still associated with this type of modeling. However, these uncertainties are typically diminished somewhat by the effect of long-term modeling (e.g., as in the 5-year time frame used herein).

ORR uses ambient air monitoring to verify compliance with 40 CFR Part 61, Subpart H for diffuse emission sources. EDE values calculated from environmental measurements are included in the ORR Annual Radionuclide Air Emissions Report, along with a site map of the locations of the ambient air monitors. Sampling and analytical procedures and quality assurance measures are consistent with the requirements of 40 CFR Part 61.93(b)(5) and are described in the ORR Environmental Monitoring Plan (EMP). In October 2001, EPA Region IV approved inclusion of this method of confirming compliance for diffuse source emissions on the ORR in the ORR Radionuclide NESHAP Compliance Plan under Appendix C "Future Agreements" as Addendum C.1.

A description of the current ORR ambient air monitoring network and sampling and analysis procedures are given in the ORR EMP. The ambient air monitoring network is reviewed on a regular basis and revisions are made as necessary to reflect physical or operational changes on the ORR. When appropriate, these reviews may include air dispersion modeling to confirm that the locations of ambient air monitors as described in the ORR EMP adequately capture emissions from significant fugitive emission sources currently in existence on the ORR. The ORR EMP is updated at a minimum once every three years or on an as needed basis and reflects program changes as they occur.

7.8.2 <u>Idaho National Engineering and Environmental Laboratory (INEEL) -</u> <u>Fugitive Emissions from Diffuse Sources</u>

Background. Although INEEL has many types of diffuse radionuclide sources on site, undisturbed contaminated soils represent the greatest number of these diffuse sources at INEEL. Appropriate methods have been developed for estimating releases from several classes of these sources. A 1991 survey established a baseline list for locations with contaminated soil. However, each year the inventories and emissions from INEEL diffuse sources are reviewed

because: 1) soil moving activities in the course of CERCLA remediation can cause substantially higher emissions, 2) after remediation has been completed, usually the emissions are reduced to effectively zero, and 3) emissions from some diffuse sources (e.g., of tritium released to ponds) are a function of INEEL facility operations during the year.

Soil Resuspension Sources. A variant of the mass loading model was used to estimate the amount of resuspended activity released from undisturbed contaminated soil areas at the INEEL. Mass loading models have been described in a number of references (Healy 1977, Healy 1974), and are incorporated in the GENII and RESRAD (Yu et al., 1993) risk assessment software packages. They have proven to provide relatively accurate estimates of the release rate of airborne activity from large source areas, where equilibrium between resuspension and deposition exists and the contamination has become closely associated with the host soil material (Healy 1977). Mass transfer processes (deposition and resuspension) between soil and atmosphere are expressed in the following equation (Peterson 1983):

$$\frac{\mathrm{d}C_{\mathrm{A}}}{\mathrm{d}t} = \mathbf{v}_{\mathrm{d}} \, \boldsymbol{\chi} - \mathrm{SC}_{\mathrm{A}}$$

Where:

 $\begin{array}{rcl} C_A &=& \mbox{The areal density of the soil to the resuspendable depth (g/m^2)} \\ v_d &=& \mbox{The deposition velocity (m/s)} \\ \chi &=& \mbox{The concentration of airborne soil (g/m^3)} \\ S &=& \mbox{The resuspension rate constant (s^{-1})} \end{array}$

This equation provides a means for estimating S based on readily measured values of mass loading and soil areal density and deposition velocity, since at equilibrium:

$$dC_A/dt = 0$$
 and $\chi v_d = SC_A$,

so

$$S = \frac{\chi v_d}{C_A}$$

The atmospheric concentration (by mass) of airborne particulate material has been measured at the INEEL during 1983-1986 (Hoff et al. 1984, 1985, 1986, 1987). These measurements show that the long-term average airborne particulate mass concentration is 25.4 μ g/m³, approximately 1 m (3.3 ft) above the ground.

The typical deposition velocity must also be determined to evaluate S. A wide range of deposition velocities for various aerosols and surfaces has been published (Sehmel 1984). A deposition velocity of 0.002 m/s (0.0066 ft/s) was selected to be consistent with the CAP-88 default value. A resuspension rate of approximately 2.5×10^{-12} s⁻¹ was calculated using an areal

density for soil of 20 kg/m² (4.1 lbs/ft²), assuming a 1 cm (0.39 in) thick resuspendable soil layer and a density of 2.0 g/cm³ (125 lbs/ft³),

$$S = \frac{2.54 \times 10^{-5} (g/m^3) \times 0.002 (m/s)}{2.0 \times 10^4 (g/m^2)} \approx 2.54 \times 10^{-12} s^{-1}$$

In its NESHAPS Annual Report (INEEL, 2002), INEEL actually reported this resuspension rate to the nearest rounded order of magnitude value or 1×10^{-12} s⁻¹. INEEL also used the Cowherd et al. model (EPA 85a) to estimate and verify this resuspension rate constant. Using consistent assumptions for soil properties, the calculated (and unrounded) resuspension rate constants derived using the Cowherd et al. model and the mass loading model differ by a factor of 2.3. The Cowherd et al. model is biased to give conservative values, as it was developed to provide rapid conservative estimates for evaluating health and safety risks that may pose an immediate hazard, and was intended to provide a "...first-cut, order-of-magnitude estimate of the potential extent of atmospheric contamination..." (EPA 85a). Therefore, the resuspension rates calculated using the Cowherd et al. and the mass loading models are considered to be in agreement.

The soil inventory data were used with this resuspension rate to estimate an annual release rate for each radionuclide, with units of Ci/yr, for input to CAP-88. Data provided as activity per unit mass were converted to release rate by assuming a resuspension rate of 3.15×10^{-5} y⁻¹ (1 × 10^{-12}

 s^{-1}), a soil density of 2 g/cm³, and a resuspendable soil depth of 1 cm:

I (Ci/y) = C
$$\left(\frac{\text{Ci}}{\text{g}}\right) \times \rho\left(\frac{\text{g}}{\text{cm}^3}\right) \times \text{d} (\text{cm}) \times \text{A} (\text{m}^2) \times \text{RR} (\text{y}^{-1}) \times 10^4 \left(\frac{\text{cm}^2}{\text{m}^2}\right)$$

Where:

I = Annual release rate

- C = Radionuclide concentration in soil
- ρ = Soil density
- d = Depth of resuspendable contamination
- A = Area of diffuse source
- RR = Resuspension rate

Source data provided as activity per unit area were converted to releases by using a resuspension rate of 3.15×10^{-5} y⁻¹ (1 × 10⁻¹² s⁻¹) and by assuming that the entire inventory is present in the resuspendable soil layer:

I (Ci/yr) =
$$C_a \left(\frac{Ci}{m^2}\right) \times A \left(m^2\right) \times RR \left(y^{-1}\right)$$

Where:

$$C_a$$
 = Radionuclide areal concentration (Ci/m²)

Each year, CERCLA operations management reports any remediation activities that disturb soil in contaminated areas so that the increased resuspension from these areas can be accounted for in the dose estimates for the NESHAP annual report. Typically, the Environmental Restoration Program develops resuspension estimates for remediation work using the EPA's *AP-42* method. An example of an *AP-42* emission factor is,

$$E = k(1.7) \left(\frac{s}{12}\right) \left(\frac{S}{48}\right) \left(\frac{W}{2.7}\right)^{0.7} \left(\frac{W}{4}\right)^{0.5} \left(\frac{365 - p}{365}\right) (kilograms[kg]/KVT)$$

Where:

Е	=	Emission factor for total particulate mass suspended per vehicle kilometer
		traveled (kg/KVT)
k	=	Particle size multiplier (dimensionless)
S	=	Silt content of the road surface material (%)
S	=	Mean vehicle speed (km/h)

- W = Mean vehicle weight (Mg)
- w = Mean number of wheels per vehicle
- p = Number of days per year with at least 0.254 mm (0.01 in) of precipitation

The resuspension estimates (as e.g., kg/month) are used with estimated contamination levels (as e.g., pCi/kg) to arrive at a release rate (pCi/month).

Evaporation and Evapotranspiration Sources. Of the volatile radionuclides released by INEEL, which include radioiodines, ¹⁴C, and tritium, only tritium is released on a regular basis in liquid effluents, and only tritium is assumed to be released to the atmosphere. The inventories of all liquid releases to ponds are recorded in the INEEL Environmental Information System (EIS), and this information is used to estimate atmospheric releases. Liquid effluents are released to lined ponds or leach fields. The fraction of tritium released to either ponds or leach fields that migrates into the atmosphere is assumed to be 1. For ponds this is a conservative assumption because evaporation and molecular exchange mechanisms probably do not cause complete transfer of tritium to the atmosphere within the year of release.

At INEEL, tritium and water releases to most diffuse emission sources are approximately constant (on a year-to-year basis) and estimated dose caused by most of the tritium emitting sources is negligible relative to INEEL's site-wide dose. Given the relative constant annual facility releases of tritium and effluent water that feed most of INEEL's diffuse tritium emission sources, and the insignificance of the calculated doses for these sources, it is reasonable to make the simplifying assumption that all of the tritium released in liquid effluents to these sources is emitted in the same year. Further refinements would have no effect on the INEEL's estimated site-wide dose. This simplifying assumption may be communicated clearly to any interested party, and justified by the insignificance of dose and the insignificant effect (with respect to the total calculated site dose) of applying e.g., the preferred method presented in Section 7.4.2 of this document. Perhaps the simpler method should be recommended for sources that could contribute less than, e.g., 1×10^{-3} mrem to the annual dose estimate, comparable to potential impact category 4 from Table 2 of the American National Standards Institute (ANSI) standard,

Sampling and Monitoring Releases of Airborne Radioactive Substances From the Stacks and Ducts of Nuclear Facilities. ANSI N 13.1-1999.

Tritium released to the leach field may percolate towards the water table or migrate towards the surface soil. The fractions of up- and downward migrating tritium depend on a number of factors, so complete release to the atmosphere is assumed for conservatism. Tritium present in irrigation water (in particular at the CFA) is assumed to be completely moved to the atmosphere via evapotranspiration. Irrigation and domestic use of well-water containing very low levels of ³H (i.e., below the EPA maximum concentration limit for drinking water) also results in release of tritium. The inventory available for release is based on monitoring data for the wells and records of the amount of water pumped from the wells.

Buried Waste. Tritium is emitted from soil overlying activated beryllium that is buried at the INEEL's Subsurface Disposal Area. These emissions have been significant with respect to calculating off-site dose, and the actual release rate to the source area from the buried beryllium is not known with the same certainty as, e.g., releases of HTO in liquid effluents to evaporation ponds. Air concentrations measurements are used with the box model (Hanna et al., 1982) to estimate annual releases from this source. The box model equation is:

$$Q_a = C \times \left(\frac{z_i \times u}{W}\right)$$

Where:

 Q_a = Areal source strength (Ci/s-m²)

C = Atmospheric concentration (Ci/s-m³)

 z_i = Mixing height (m)

u = Windspeed (m/s)

W = Width of the area source along the direction of the wind (m)

The box model deals with an urban-scale modeling domain, and appropriately indicates that typical mixing depths are on the order of 500-1000 m (1,640 - 3,280 ft). However a generic box model may be applied to estimate contaminant flux into any defined volume, provided that the observed concentrations (i.e., concentration measurements) are representative of the average concentration in the volume and the rates of all significant loss processes are taken into account. In the case of emissions from INEEL buried beryllium, the values of the box parameters (particularly mixing depth and measurement elevation) were established using judgement, supported by extensive field observations, recognizing that there was considerable room for error. The absence of a persistent, well-defined "ceiling" was addressed by subsequent characterization of the airborne tritium concentration measurements) over a 2-year period. The gradient measurements indicated that air samples from the location originally selected for measuring the concentration in the box are reasonably representative of the average concentration, and that ~95% of the tritium emitted from the source is present below the assumed mixing depth at the measurement location.

A more extensive set of gradient measurements around the source area would give a better basis for estimating emissions and uncertainty. However, there are operational constraints on the number and location of sampling stations that may be placed in the disposal area. Also, resource constraints prevent using other different approaches (e.g., covering and venting part or all of the sources through a monitored duct) for estimating the emissions.

7.8.3 <u>Hanford Site – Fugitive Emissions from Diffuse Sources</u>

Background. The method currently used to estimate fugitive emissions from diffuse sources at the Hanford Site, and the subsequent offsite dose, is based on measured ambient air concentrations at the site perimeter. The expected contributions from monitored stack emissions and background radioactivity are subtracted from ambient air concentrations, and if the difference is positive, the result is attributed to diffuse sources. Although this is an indirect method for estimating emissions, it is subject to less uncertainty in estimating dose to a member of the public because it uses actual monitoring data from the site perimeter where members of the public could be located. This method is also much more cost effective in estimating dose to a member of the public compared to estimating resuspension or emissions from over 1000 potential sources of fugitive emissions within the Hanford Site. Additional information on the approach used at Hanford follows.

Ambient Air Concentration Procedure. Potential emissions from diffuse sources are estimated from environmental surveillance ambient air monitoring data collected at the Hanford Site perimeter. This method is preferred for two reasons: 1) these data most accurately represent the actual exposures of an offsite individual to airborne radioactivity and 2) there is currently insufficient information concerning the extent and characteristics of onsite soil contamination to use radionuclide resuspension estimates in conjunction with transport and dose modeling for many potential sources of fugitive emissions. The ambient air sampling results consisted of measured air concentrations for radionuclides that could be released from Hanford Site operations and diffuse sources. Radionuclides routinely assayed in ambient air samples include ³H, ⁶⁰Co, ⁹⁰Sr, ¹⁰⁶Ru, ¹²⁵Sb, ¹²⁹I, ¹³⁴Cs, ¹³⁷Cs, ¹⁵⁴Eu, ¹⁵⁵Eu, ²³⁴U, ²³⁵U, ²³⁸U, ²³⁸Pu, and ^{239/240}Pu (Poston et al., 2003).

Radionuclide air concentrations resulting from monitored stack emissions at Hanford facilities and other nearby non-DOE sources are calculated for each of the perimeter sample locations using the CAP88-PC atmospheric dispersion modeling code. The combined contributions to airborne radionuclide concentrations attributable to the stack emissions from these sources were subtracted from the ambient air sampling results. Averaged regional background concentrations for each radionuclide were calculated from the air sample results obtained at distant community sampling stations located outside the 80-km (50-mile) radius from Hanford sources. The average background concentration at these stations was also subtracted from the ambient monitoring results at Hanford perimeter stations. The air concentrations at the site perimeter, corrected as described for monitored emissions sources and background concentrations, are assumed to be attributable to emissions from diffuse sources. Potential emissions from diffuse sources are estimated using the corrected perimeter air concentrations attributable to fugitive emissions, and by performing a back-calculation using CAP88-PC. The 200 West Area near the center of the Hanford Site is assumed to be the source of all fugitive emissions. This assumption results in a conservatively high aggregate release estimate and dose from all diffuse sources. The average aggregate emissions from diffuse sources are then used to estimate dose at the Hanford Site perimeter with the CAP88-PC code. The Hanford Site air emissions report contains results for the perimeter location having the highest dose, as well as the dose at the location of the maximally exposed member of the public from monitored stack sources. The maximum combined dose to a member of the public from monitored stack emissions and potential diffuse source emissions is reported to demonstrate compliance with the 10 mrem/year standard in 40 CFR Part 61, Subpart H. Specifics of the Hanford methodology and dose results for calendar year 2002 are available in DOE-RL2003.

Chapter 8

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Attachment A

ADDITIONAL REFERENCES FOR SAMPLING DIFFUSE SOURCE EMISSIONS

Attachment A

ADDITIONAL REFERENCES FOR SAMPLING DIFFUSE SOURCE EMISSIONS

The following nine references provide EPA-approved alternative methods for sampling diffuse source emissions.

• Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance DOE/EH-0173T (1991)

Various sample types, from biotic to abiotic, are presented, along with the basis for sample collection. Sample location and frequency are also discussed. A brief statement on sampling methods completes each section but detailed procedures or techniques are not provided. References to other guidance documents are cited. The reader is directed to other sources to obtain additional regulatory information or descriptions of specific procedures.

Air Sampling in the Workplace: NUREG 1400 (1993) http://bidug.pnl.gov/references/NUREG-1400.PDF

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This report provides technical information on air sampling that will be useful for facilities following the recommendations in the NRC's Regulatory Guide Revision 8.25, Revision 1 "Air Sampling in the Workplace." NRC's Regulatory Guide addresses air sampling to meet the requirements in NRC's regulations on radiation protection, 10 CFR Part 20. The NUREG report describes how to determine the need for air sampling based on the type of material, release potential, and confinement of the material. The purposes of air sampling and how the purposes affect the types of air sampling performed are discussed. Locating air samplers to accurately determine the concentrations of airborne radioactive materials that workers could be exposed to is presented. The need and the methods for performing airflow pattern studies to improve the accuracy of air sampling results are included. The report gives examples of several techniques that can be used to evaluate whether the airborne concentrations of material are representative of the air inhaled by workers. Methods to adjust derived air concentrations for particle size are described, along with methods to calibrate for volume of air sampled and to estimate the uncertainty in the volume of air sampled. Statistical tests for determining minimum detectable concentrations are presented. Performing an annual evaluation of the adequacy of the air sampling is included.

The Environmental Survey Manual DOE/EH-0053 (1987) *The Environmental Survey Manual: Appendices A, B, C, D, E, F & G*

Unlike a number of other references listed here, this document includes information related to radionuclides and considers biota (i.e., animal, plant, and related sample types). Flow charts, checklists, planning diagrams, and figures are included. Section 2 of this volume covers topics related to a survey team's activities and reports. Section 3

considers the use of existing data, followed by technical checklists in Section 4, and health and safety in Section 5. The document is a comprehensive overview.

Decommissioning Handbook DOE/EM-0142P (1994).

Chapter 6 presents information on final project configuration based on planning and as such speaks of site boundaries. Chapter 7 presents topics related to characterization including onsite measurements.

Samplers and Sampling Procedures for Hazardous Waste Streams EPA-600/2-80-018 (1980).

While not specifically geared for radioactive samples, this manual presents information on sampling devices, selecting an appropriate device for various media, container types, labels, seals, use of log books, chain of custody, sample receipt, etc. The document includes five appendices, covering development of composite liquid waste samples, parts for constructing the sampler, a checklist of items required in the field for sampling hazardous waste, random sampling, and systemic errors in using the composite liquid waste sampler.

Test Methods For Evaluating Solid Waste, Physical/Chemical Methods, 2nd Edition EPA (1982).

This manual includes a Sampling Methodology section that addresses statistics, sampling strategies and examples, implementing a sampling plan, and tables and figure of sample devices. Some information relevant to the field component of sampling work is included, but most will be useful to laboratory personnel.

Handbook for Sampling and Sample Preservation of Water and Wastewater EPA-600/4-82-029 (1982)

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This handbook addresses sampling issues covering a number of water sources, including: municipal, industrial, surface, agricultural, ground, and drinking water. Types of sample are defined and discussed, including grab and composite samples. Diagrams, tables, and forms are provided to illustrate key points. Statistical methods and related tables are provided. Each topic is accompanied by references. The chapter on radioactive samples is brief but touches on background, radioactive decay, detection, capability, frequency of sampling, sample location, sample volume, radiation safety, and references.

Compendium of ERT Surface Water and Sediment Sampling Procedures EPA OSWER Directive 9360.4-03 (1991).

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The three standard operating procedures (SOPs) in this document are 1) Sampling Equipment Decontamination, 2) Surface Water Sampling, and 3) Sediment Sampling. Each SOP is similar in content with sections covering scope, method, summary, preservation, containers, equipment, apparatus, etc.

Screening Models for Release of Radionuclides to Atmosphere, Surface, Water and Ground: NCRP 123 (1997).

NCRP Report No. 123, a two-volume report, provides simple, screening techniques to address the release of radioactive materials to the environment. The techniques provided can be employed to demonstrate compliance with environmental standards for release of radionuclides to the atmosphere, surface water, or ground. The report provides the derivation of screening models for radionuclide releases, and covers, in addition to transport models for releases to the atmosphere and water, the matter of usage factors that relate to the consumption of contaminated drinking water and food by humans. Work sheets are provided that allow the user to carry out a screening process for a proposed release via a few calculations using a minimum of site-specific data and decisions.
Attachment B

METHODOLOGIES SUBMITTED BY DOE AND APPROVED BY EPA

Attachment B

METHODOLOGIES SUBMITTED BY DOE AND APPROVED BY EPA

B.1 Environmental Monitoring Plan for Airborne Fugitive Radioactivity from Diffuse Sources, Oak Ridge, TN

In response to a March 1992 EPA Region IV guidance letter allowing use of environmental measurements to confirm compliance for diffuse sources with the Radionuclide NESHAP, the Oak Ridge Reservation (ORR) developed a plan to use a network of ambient air monitors to confirm that fugitive radionuclide emissions from diffuse sources do not result in atmospheric radionuclide concentrations that contribute significantly to the reported radiation doses to members of the offsite public. The plan was submitted to EPA Region IV in October 1992, and was implemented in January 1993. Effective Dose Equivalent (EDE) values calculated from the environmental measurements have been provided to EPA as supplementary information in the ORR Annual Radionuclide Air Emissions Report since 1992 consistent with the MOU between DOE and EPA. In October 2001, EPA Region IV approved inclusion of this method of confirming compliance for diffuse sources on the ORR in the ORR Radionuclide NESHAP Compliance Plan under Appendix C "Future Agreements" as Addendum C.1.

Any diffuse source emissions from the ORR are likely to be in the form of particulates from the re-suspension of dust and soil. As of 1992, no significant fugitive gaseous sources of radionuclides were known. Calculated off-site doses based on measured atmospheric concentrations include the dose from all sources, including point and diffuse sources. Air sampling stations were equipped to measure particulate radionuclide concentrations at 10% or less of the ambient air concentrations listed in 40 CFR 61, Appendix E, Table 2. Samples were also analyzed for tritium, although it is not believed to be a significant fugitive source. Sampling locations were selected using the ISC Code to identify locations most affected by releases from the three main operations centers on the ORR: the Oak Ridge National Laboratory (ORNL), the East Tennessee Technology Park (ETTP), and the Y-12 National Security Complex. Air sampling stations were located on the perimeter of the ORR in the directions of critical receptors relative to the three ORR facilities. Among the radionuclides monitored were 3He, 228Th, 234U, 235U, 238U, 90Sr, 238Pu, 239Pu, 240 Pu. Radiochemistry was used to analyze the samples.

B.2 Resuspension of Soil Particles from Rocky Flats Containing Plutonium Particulates

In 1997, the Rocky Flats Environmental Technology Site (RFETS) proposed to assess the dose to the public from the resuspension of soil particulates with plutonium in an area, called the *903 Field*, adjacent to and directly east of a former oil drum storage area. Using environmental measurements of radionuclide air concentrations at critical receptor locations rather than the dispersion modeling approach outlined in the regulation. The alternative compliance demonstrating method has been approved by the EPA. The compliance network consists of 14 samples located around the perimeters of the site and was fully operational in 1999. The samples are part of the sites radioactive ambient air monitoring program (RAMP) network.

The storage area was paved in 1969, and designated the 903 Pad. The most significant processes included resuspension from grass and rain splash. Wind resuspension from bare soil was found to be minimal. The nature of the original contamination process meant that over 90% of the resuspended plutonium is attached to particles larger than 3 micrometers (μ m).

A number of studies have been undertaken over the years to study the migration of the plutonium, including aerial gamma surveys of the plant and surrounding area, using an array of sodium iodide (NaI) detectors. These airborne surveys showed that the plutonium had not significantly migrated beyond the original source area. The dose to the off-site public is best estimated by analyzing plutonium concentrations downwind. Long-term downwind measurements have shown that beyond about 2.4 km (1.5 miles), the effect of the 903 Field cannot be distinguished from background, and the 903 Field does not contribute significantly to the off-site dose.

B.3 Emission Estimations for Area, Line, and Volume Sources for the Weldon Spring Site Remedial Action Project

Emission rates at the Weldon Spring Site Remedial Action Project were estimated using an emission factor and the activity level for different geometry sources, including area sources, line sources, and volume sources. Area sources were the result of excavation, wheeled front loader operations, scraping, dumping, grading, soil compaction, wind erosion from active piles, and building demolition. Line sources included unpaved road traffic, and uncovered truck beds. Emission factors were developed using *AP-42*; *Control of Open Fugitive Dust Sources*, EPA-450/3-88-008 (1989); and *Air/Superfund National Technical Guidance Study Series, Volume III - Estimation of Air Emissions from Cleanup Activities at Superfund Sites*, EPA-450/1-89-003 (1989).

B.4 Alternate Dose Assessment Methodology - Mound Plant

A request for an alternative methodology for estimating doses from diffuse sources for non-U.S. Department of Energy (DOE), non-nuclear workers from the DOE Mound Plant in Miamisburg, Ohio was submitted in 1996. The methodology estimated the outdoor and indoor exposures and doses as a function of the distance of the receptor from the soil remediation project or building demolition project. CAP88-PC, used for air concentration calculations, was validated by comparing the calculated concentration of ²³⁸Pu with the actual air concentration at an air sampling station.

B.5 Proposed Diffuse Source Methodology for Non-DOE Non-Nuclear Workers at the U.S. DOE Mound Plant

This proposal, submitted in 1997, is very similar to the one made at the Mound Plant in Miamisburg, Ohio the previous year (Section B.4). Two equipment changes were proposed. The use of digital manometers, with improved sensitivity and accuracy compared to inclined manometers, were proposed, along with the use of a digital humidity sensor, as an alternate method for determining moisture content. These requests were limited to National Emissions Standards for Hazardous Air Pollutants (NESHAPs) stack testing at the Mound Plant.