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JAN 19 1988

NUREG/CR-4997
PNL-6300

Methods for Describing Airborne Fractions of Free Fall Spills of Powders and Liquids

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Pacific Northwest Laboratory

Prepared for
U.S. Nuclear Regulatory
Commission

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Methods for Describing Airborne Fractions of Free Fall Spills of Powders and Liquids

Manuscript Completed: October 1987
Date Published: January 1988

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NRC FIN B2481

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ABSTRACT

This report describes methods for estimating the fractions of liquid and powder that produce aerosols in hypothetical spills. The characteristics of these aerosols are also described. The methods developed were based on previous data, on the free fall spills in static air of aqueous solutions of uranyl nitrate, uranine, sucrose solutions and slurries, and titanium dioxide and uranium dioxide powders (Sutter et al. 1981; Ballinger and Hodgson 1986).

After the liquid spill data were corrected for evaporation and settling, regression analyses of the data were produced correlating the fraction of a spill made airborne, F , with dimensionless numbers. The resulting equations were:

$$F = a \text{ Arch}^b (\rho_{\text{air}}/\rho_{\text{liq}})^c \text{ Fr}^d$$

and

$$F = m \text{ Arch}^n$$

where a , b , c , d , m , and n are coefficients and Arch is the Archimedes number,

$$\text{Arch} = (\rho_{\text{air}}^2 H^3 g)/\mu^2$$

where ρ_{air} = density of air
 H = spill height
 g = gravity constant
 μ = liquid viscosity

The Froude number is

$$\text{Fr} = V^2/(g R)$$

where V is impact velocity and R is the radius of an equivalent sphere of liquid spilled. Statistical descriptions of the lognormal distributions were provided for parameters of liquid aerosols.

Whereas liquid aerosols formed on impact, powder aerosols formed as the powder fell. A dimensional analysis of the powder that dispersed while falling correlated well with the Galileo number, Ga , which is

$$Ga = (6 g Mo)(\rho_{\text{air}}/\mu_{\text{air}})^2/(\pi \rho_p)$$

where M_o = mass of powder spilled
 ρ_p = powder bulk density
 ρ_{air} = density of air
 μ_{air} = air viscosity.

Regression analyses produced correlations for a parameter, E , of the form $E = m Ga^n$. The amount airborne during the fall is proportional to the drag force on the powder, with E being the proportionality factor. A computer code, PSPILL, was developed to calculate the drag force and amount airborne for specified time steps during the spill.

EXECUTIVE SUMMARY

Pacific Northwest Laboratory developed calculational methods to characterize aerosols produced in hypothetical spill accidents. These methods were developed for the U.S. Nuclear Regulatory Commission to use when evaluating the consequence of postulated accidents for safety analyses and environmental impact statements. Basic physical properties and mechanistic descriptions of spill events were used as a basis for the methods.

Source term models consist of equations that can be used to estimate the mass airborne and particle size distribution of aerosols produced by spills of powders and solutions. Experimental data from Sutter et al. (1981) and Ballinger and Hodgson (1986) were emphasized in the models. Parameter ranges for this data were spill height 1 to 3 m, powder mass 25 to 1000 g, and liquid volume 125 to 1000 ml. Liquids spilled included slurries and solutions of varying viscosities.

Liquid spills differed from powders in that an aerosol was produced on impact instead of during the fall. The fraction airborne from liquid spills (including viscous solutions and slurries) correlated well with three dimensionless numbers: the Archimedes number, the Froude number, and a density ratio. Liquid aerosol parameters were statistical descriptions of the log-normal distributions.

A computer code was developed to model powder spills. In the code, the mass airborne was assumed proportional to the drag force on the powder as it falls. The proportionality factor was empirically found to be a function of a dimensionless number, the Galileo number.

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1.0 INTRODUCTION

This document is a part of an overall program of the U.S. Nuclear Regulatory Commission (NRC), Office of Nuclear Materials Safety and Safeguards, Division of Fuel Cycle and Material Safety to improve techniques for analyzing potential accidents in nuclear fuel cycle facilities. The scope of accidents for the overall program includes fires, explosions, criticalities, tornadoes, equipment failures, and spills. The program will eventually provide an Accident Analysis Handbook for nuclear fuel cycle safety, covering all of the above accidents.

Because little is known about the airborne consequence of spills, a two-part project on spills was begun early in this program. The purpose of the first part of the spill project was to obtain information on a limited experimental matrix to determine the magnitude and the characteristics of the aerosols generated by the free fall spills of powders and solutions in static air. This was completed and reported by Sutter et al. (1981). That document reported the experimental results of the airborne consequences of the spills of titanium dioxide powder (TiO_2), depleted uranium dioxide powder (DUO), 1% aqueous uranine (sodium fluorescein), and an aqueous solution of uranyl nitrate hexahydrate (UNH) (0.877 M) and nitric acid (8.4 M). A second set of experiments was performed to expand the data base to include viscous solutions and slurries. Ballinger and Hodgson (1986) report the results of these experiments. The data from these experiments showed promise for analysis and development of suitable calculational models to estimate fraction airborne from free fall spills.

The objectives of this document are to provide calculation models that characterize aerosols produced in hypothetical spills for powders and liquids in nuclear fuel cycle facilities. The models emphasize the experimental work of Sutter et al. (1981) on the free fall spills of TiO_2 and DUO powders and aqueous UNH and uranine solutions, and of Ballinger and Hodgson (1986) on spills of viscous solutions and slurries. The possible mechanisms of aerosol formation were quantitatively examined to provide the calculational models.

The results of developing calculational models for free fall spills are described in Section 2.0. Section 3.0 deals with details of liquid spills and resulting correlations. Section 4.0 details the analyses and correlations for the powder spills. Appendices A and B give supporting information on Sections 3.0 and 4.0, and Appendix C is a listing of PSPILL, a computer code developed to model powder spills.

2.0 CONCLUSIONS AND RECOMMENDATIONS

Models have been developed to estimate the fraction of powders and liquids made airborne from hypothetical spills in nuclear fuel cycle facilities. Liquid spills that produce most of their aerosol after impacting the floor were modeled separately from powder spills that produce aerosols from the shearing effect of the air on the powder as it falls.

Because the liquid spills resulted in aerosols that could evaporate quickly, two evaporation/settling codes were developed to correct the impactor data for evaporation and settling of the droplets. Because the vapor pressure characteristics of the UNH (with HNO_3) and the uranine were so different, two codes were needed. The initial liquid aerosol quantities and characteristics were determined using the two codes. Correlations were then made on the output from the codes.

A dimensional analysis of the liquid and slurry spill parameters produced three dimensionless parameters: the Archimedes number, a density ratio, and the Froude number. The final regression analysis results produced the following estimation equations for F, the fraction airborne:

$$F = 2.3 \times 10^{-5} \text{ Arch}^{0.44} \left(\frac{\rho_a}{\rho_l} \right)^{2.4} \text{ Fr}^{0.38} \quad (\text{for the original measured data}) \quad (1)$$

$$F = 6.31 \times 10^{-6} \text{ Arch}^{0.45} \left(\frac{\rho_a}{\rho_l} \right)^{2.2} \text{ Fr}^{0.35} \quad (\text{for the evaporation/settling corrected data}) \quad (2)$$

The Archimedes number is defined as

$$\text{Arch} = (\rho_a^2 H^3 g) / \mu^2 \quad (3)$$

where ρ_a = air density, g/cm^3
H = spill height, cm
g = gravity constant, cm/s^2
 μ = solution viscosity, poise

The Froude number is

$$\text{Fr} = \frac{v^2}{g R} \quad (4)$$

where V = impact velocity, cm/s = $\sqrt{2 g H}$
 R = radius of equivalent sphere of liquid spilled, cm

An evaporation/settling model should be used in an analysis of a liquid spill to determine the particle size of aerosols at time intervals after the initial spill release. If an appropriate model is available, Equation (2) should be used to determine the fraction of airborne droplets carried away from the initial release point. However, if an evaporation/settling model is not available, Equation (1) should be used.

Regression correlations for the aerodynamic mass median equivalent diameter (AMMD) and geometric standard deviation, σ_g , were not acceptable (too random). Tables of average AMMDs and σ_g s with appropriate confidence intervals were produced instead. The original average AMMD and σ_g were 8.9 μm and 5.4, respectively, for all data, whereas the corrected data base gave 21.5 μm and 7.3 for the values.

For powders it was necessary to develop a model that follows the dispersed powder through various stages of the fall. The powder is assumed to be a ball that grows at a constant rate. For each time step, the drag force on the sphere is calculated, and the amount airborne is predicted assuming proportionality to the drag force. The proportionality constant is a function of the dimensionless Galileo number:

$$Ga = \frac{6 g Mo}{\pi \rho_p} \left[\frac{\rho_a}{\mu_a} \right]^2 \quad (5)$$

where g = gravity constant, cm/s²
 Mo = powder mass, g
 ρ_p = powder bulk density, g/cm³
 ρ_a = air density, g/cm³
 μ_a = air viscosity, poise

These correlations and data summaries represent the first published for single spills of bulk powders and liquids. The range of controlling variables (type of powder or liquid, volume/mass of spill, spill height, speed of spill) does not cover the full range that could be expected in accidents. However, the formulas do allow extrapolation beyond the range of the supporting data. Data from single-drop spills, continuous spills, spills with other materials, and spills at higher/lower heights, if generated and used in a correlation, could increase confidence in applying the models to these types of spills.

3.0 MODELS FOR FREE FALL SPILLS OF LIQUIDS

This section provides a background study of the literature on liquid spill models, an analysis of the data of Sutter et al. (1981) and Ballinger and Hodgson (1986), and a description of the models that characterize the airborne fraction of certain spills of liquids in static air. The spills are limited to single events of large (125 to 1000 cc) liquid masses falling from a height of up to 3 m onto a dry stainless steel floor. In Figure 3.1 a typical spill is shown.

FIGURE 3.1. Schematic Drawing of the Liquid Spill

The model correlations are most suitably used for similar liquids and quantities falling from similar heights. The correlations are less suitable for conditions outside the experimental variable ranges. In these less suitable conditions one would include continuous spills, spills of single drops, or spills from great heights. In the last situation, shear forces would eventually break up the falling liquid mass into individual rain-sized drops (<8 mm) that fall at terminal velocity. For the spills discussed in this report, the principal dispersion step is the impact with the floor and not the shear action of the air. For a 3-m drop, the liquid mass is in nearly frictionless free fall conditions.

3.1 BACKGROUND

The literature previous to 1981 showed no apparent data and analyses on free fall spills from which a spill model could be developed. However, several key articles dealt with the breakup of single drops on impact. Kinnel (1972) showed experimental evidence of the inelastic nature of the impact of single water drop splashes. The major conclusions were that the number of product particles (N) was a function of drop radius (R,mm) and impact velocity, $V(\text{ms}^{-1})$. A developed empirical relationship was

$$N = 3.4 R^3 V^2 - 63 \quad (6)$$

Kinnel's other conclusions are that 1) product drop sizes are lognormally distributed, 2) N increases as surface roughness increases, 3) N decreases as drop surface tension decreases, 4) N decreases as the depth of fluid covering the impact surface increases, and 5) distribution curves show that the number mean size is about 120- μm diameter, which increases as R increases and as surface roughness or covering liquid depth increases. A drop size less than $R = 0.75 \text{ mm}$ produces no droplets. Stow's (Stow and Stainer 1977) drop size distribution data can be used to calculate the number of drops that are <30 μm and are of significance in the formation of more long-lived aerosols, especially if evaporation is important in reducing particle size.

A more recent paper by Stow and Hadfield (1980) improved Stow's earlier model for N, which is now

$$N = k_1 [R^3 V^2 - R^{1.82} S_T^{1.18}] \quad (7)$$

where $S_T = k_2 [Re_R We_R^2]^{1/3}$

Re_R = impact Reynolds number, $(\rho R V)/\mu$, based on drop radius

We_R = impact Weber number, $(\rho R V^2)/\gamma$, based on drop radius

k_1, k_2 = experimental constant

N = number of product particles
 R = drop radius, cm
 V = impact velocity, cm/s
 ρ = liquid density, g/cm³
 γ = liquid surface tension, dyne/cm
 μ = liquid viscosity, poise

Stow mentions the Froude number, $Fr_R = V^2/(g R)$, but concludes that for the small drop radius, R , of single drops, its importance is minimal. For the models described in this report the R is two orders of magnitude larger.

Another mechanism that probably contributes significantly to the formation of small particles is the breakup of air bubbles entrained on impact. This phenomenon was suspected, but not verified.

3.2 BASIS FOR MODEL CORRELATIONS

The exact mechanistic description of the process of aerosol formation during liquid spills is a complex fluid mechanics problem. The usual method of engineering science in such situations is to correlate the data using some form of dimensional analysis, as described below.

For liquid spills, eight independent variables and three dependent variables are known (Table 3.1). The variables of air viscosity and air density were included to analyze the possible influence of air shear on the data. With the total of 11 variables and three fundamental dimensions (mass, length, and time), there are 11 (variables) - 3 (fundamental dimensions) = 8 dimensionless variables that can be chosen to represent the data.

The dimensionless variables chosen to represent the data are Arch, Re, Fr, μ/μ_a , ρ/ρ_a , d^* , F , and σ_g

where Arch = $\rho^2 H^3 / \mu^3$, Archimedes Number

Re = $\rho D V / \mu$, Reynolds Number

Fr = $V^2 / g D$, Froude Number

μ/μ_a = viscosity ratio

ρ/ρ_a = density ratio

$d^* = d_p / D$ = diameter ratio

$F = m_p / m_s$ = fraction of spill airborne

σ_g = geometric standard deviation

TABLE 3.1. Liquid Spill Variables

Independent Variables

μ & μ_a :	viscosity of liquid and air
ρ & ρ_a :	density of liquid and air
g :	acceleration of gravity
h :	height of spill (or V , impact velocity = $\sqrt{2 g h}$)
m_s :	mass of spill (or diameter of equivalent sphere = $D = \sqrt[3]{6 m_s / \pi \rho}$)
γ :	liquid surface tension

Dependent Variables

m_p :	mass of droplets airborne
d_p :	mass median diameter
σ_g :	geometric standard deviation of particle size distribution

Other combinations of these variables are possible. In a comprehensive paper by Krzeczowski (1980) on liquid droplet disintegration mechanisms (not including impact), several others are used. His Weber number, We' ,

$$We' = (\rho_a D V^2 / \gamma) (\mu_a / \mu) = (We \cdot (\mu_a / \mu)) \quad (8)$$

and another defined number, the Laplace number, La ,

$$La = \frac{\rho \gamma D}{\mu^2} = Re^2 / We \quad (9)$$

were used. If the air parameters were important, the F , σ_g , and d^* would show a dependence on μ/μ_a or ρ/ρ_a , without the use of We' or La . At this point relationships would be desired in the form of

$$F = F(\text{Arch}, Re, Fr, \mu/\mu_a, \rho/\rho_a) \quad (10)$$

$$d_p = d_p(\text{Arch}, \text{Re}, \text{Fr}, \mu/\mu_a, \rho/\rho_a) \quad (11)$$

$$\sigma_g = \sigma_g(\text{Arch}, \text{Re}, \text{Fr}, \mu/\mu_a, \rho/\rho_a) \quad (12)$$

A special consideration in the regression analysis was the correction of the original data (measured percent airborne and impactor size distribution data) for evaporation and fallout. The high-volume samplers and the impactors used in the data collection did not sample the whole air volume instantaneously after the spill, but rather sampled an aerosol that was rapidly changing into smaller particles because of evaporation and fallout. Because a drop of 10 μm diameter of pure water in dry air at 18°C will evaporate in 0.06 s (Davies and Rideal 1963), evaporation could seriously affect the impactor data. The uranine particles were most seriously affected by evaporation because uranine contains only 1% nonvolatile solids, whereas the UNH solutions contained 28% nonvolatile solids and a large percent of lower volatility (than water) HNO_3 . Two computer codes were developed to convert the original data base into the initial aerosol data base corrected for evaporation and fallout. The original data base contains data collected from the high-volume samplers. The initial aerosol data base converts the original data (from which some particles have settled and others have shrunk because of evaporation) to its state immediately after the spill. The methods for making these corrections are discussed in the next section.

3.3 EVAPORATION AND FALLOUT CORRECTIONS

This section describes the methods and results of correcting the original liquid spill data to account for changes in the aerosol after formation. These changes are essentially droplet evaporation and gravitational fallout. The objective of these corrections was to find out the characteristics of the aerosol before evaporation and settling took place--both mass airborne and drop size distribution. The actual data were taken by a high-volume sampler and cascade impactor for 30 min to 1 h after the spill. These measured data do not represent the initial aerosol because of the duration of the sampling time. Two computer codes were written to correct the impactor and high-volume sampler data. The first code handled the uranine solution, sucrose, and slurry corrections and the second code handled the UNH solution. It is not in the scope of this document to provide detailed user's manuals for these codes or detailed technical descriptions. Instead, a general technical description is given below.

The basic principles in the codes assume that the initial spill event will leave both a well-mixed and dilute (non-agglomerating) aerosol. Because the initial spill airborne mass and particle size distribution are unknown, n sizes are assumed (larger than the cascade impactor plates) where n is the number of

impactor plates. A vector $\underline{A}$ of given unknowns, A_i (where $i = 1 - n$), represents the unknown masses of the initial aerosol-size buckets. A matrix of coefficients $[a_{ij}]$ represents a set of the fraction of the mass of solute in the A_i that collects on each plate; for example, $a_{11}A_1$ is the amount of A_1 that finally ends up on cascade plate 1 (at the time the samples are collected). The amount of A_2 that finally ends up on plate 1 is $a_{12}A_2$. B_i (where $i = 1 - n$) is the measured amount of solute on each impactor plate. Thus, for a five-state impactor, a set of five equations results:

$$a_{11}A_1 + a_{12}A_2 + a_{13}A_3 + a_{14}A_4 + a_{15}A_5 = B_1 \quad (13)$$

$$a_{21}A_1 + a_{22}A_2 + a_{23}A_3 + a_{24}A_4 + a_{25}A_5 = B_2 \quad (14)$$

$$a_{31}A_1 + a_{32}A_2 + a_{33}A_3 + a_{34}A_4 + a_{35}A_5 = B_3 \quad (15)$$

$$a_{41}A_1 + a_{42}A_2 + a_{43}A_3 + a_{44}A_4 + a_{45}A_5 = B_4 \quad (16)$$

$$a_{51}A_1 + a_{52}A_2 + a_{53}A_3 + a_{54}A_4 + a_{55}A_5 = B_5 \quad (17)$$

$$\text{or} \quad [a_{ij}]\underline{A} = \underline{B} \quad (18)$$

The code assumes that the A_i are 1 g each initially, and it keeps track of that mass as it is sampled, evaporated, and depleted by settling. The cascade-sampled fraction on each plate becomes the $[a_{ij}]$. By matrix inversion, the original A_i are computed using

$$\underline{A} = [a_{ij}]^{-1}\underline{B} \quad (19)$$

The initial mass airborne is ΣA_i . The mass median aerodynamic equivalent diameters and σ_g s are then calculated. There are some constraints on this procedure. First, ΣA_i cannot be larger than the high-volume sampler datum. Second, the assumed sizes of the initial A_i must be carefully picked so that something collects on each plate (as happened experimentally) and, preferably, one or more initial sizes fall on more than one plate as drops evaporate. Also, the sizes and A_i masses should be lognormally distributed.

The transient calculations of the codes for the sampling period are accomplished by finite difference solutions of n differential equations for the n airborne mass variables, M_i (where $i = 1$ to n), which represent the mass of the n original but changing particle groups. Each of these equations contains the following components:

$$\frac{dM_i}{dt} = -\dot{M}_i^S, \text{ mass settling rate of group } i, \text{ g/s}$$

$$-\dot{M}_i^e, \text{ mass evaporation rate of group } i, \text{ g/s}$$

$$-F^I M_i, \text{ impactor capture rate of group } i$$

$$-F^H M_i, \text{ high-volume sampler capture rate of group } i \quad (20)$$

and are solved over a short time interval, $\Delta t = t_2 - t_1$, during the life of the aerosol. Simultaneously, n differential equations for the number of particles in each group N_i must be solved. These are

$$\frac{dN_i}{dt} = S_i N_i, \text{ number settling rate of group } i$$

$$-F^I N_i, \text{ impactor number sampling rate}$$

$$-F^H N_i, \text{ high-volume number sampling rate.} \quad (21)$$

These equations are directly integratable as

$$N_i(t_2) = N_i(t_1) \exp[-(S_i + F^I + F^H)(t_2 - t_1)] \quad (22)$$

where $S_i = v_t A_f / V$
 $v_t = D^2 (\rho - \rho_a) g / 18 \mu$, terminal drop velocity
 A_f = floor area
 V = room volume
 F^I = impactor volumetric flow rate
 F^H = high-volume sampler volumetric flow rate

The evaporation terms constitute the final and most uncertain part of the analysis (Davies and Rideal 1963), expressed as

$$\dot{M}_i^e = 2 \pi M_w D_g \Delta p D / R T \quad (23)$$

where M_w = molecular weight of evaporating solvent
 D_g = gas phase diffusivity of evaporating solvent
 D = drop diameter
 $\Delta p = P_v - P_a$
 P_v = vapor pressure of solvent at drop surface

P_a = ambient air solvent vapor pressure
 R = gas constant
 T = air temperature

The properties of the spill solutions are listed in Table 3.2. All vapor pressures were calculated in the codes. For uranine solutions, P_v was calculated using a relationship to uranine concentration and temperature based on the Clausius-Clapeyron equation and Raoult's law, because no data were available for uranine-water partial pressures. Microscopic examination of large evaporating uranine solution drops revealed that the drops evaporated without crystallization. For UNH solutions, both water and nitric acid could evaporate or water could be absorbed depending on the humidity. UNH drops did not dry completely. The P_v relationships for water and nitric acid came from tables and graphs available through the Chemical Engineer's Handbook (Perry and Chilton 1973) and a document by Fleming (1958). No mass transfer resistance was assumed inside the evaporating drops. This is not a good assumption when the drops approach dryness; however, including this resistance in the analysis would have required liquid phase diffusivities that are not in the literature. For sucrose and slurry solutions, the amount of sugar added to the solution was assumed to affect the vapor pressure according to data given in the International Critical Tables (Washburn 1928).

The results of the evaporation/settling calculations are tabulated in Table A.1 (Appendix A). The ratio of initial/measured values for AMMD, σ_g , and

TABLE 3.2. Comparison of Properties of Experimental Liquids and Water (25°C)

Material	Density (g/cc)	Viscosity (cp)	Surface Tension (dyne/cm)	Vapor Pressure (mm Hg)
H ₂ O ^(a)	0.997	0.89	72.0	23.8 ^(a)
UNH ^(b)	1.54	1.70	66.4	13.6 ^(c)
Uranine ^(d)	0.993	0.79	52.1	23.7 ^(c)
Sucrose ^(e)	1.0 - 1.3	1.3 - 47.1	67.0 - 83.0	
Slurry ^(f)	1.1 - 1.4	1.3 - 3.1	58.2 - 68.4	

(a) Weast 1978.

(b) Uranyl Nitrate Hexahydrate Solution in nitric acid (208.7 g U/L (0.877 M), 8.4 M HNO₃).

(c) Calculated.

(d) 10 g/L uranine in water.

(e) Varied from 0 to 60% sucrose (Ballinger and Hodgson 1986).

(f) Ballinger and Hodgson 1986.

fraction airborne are shown in Table 3.3. As can be seen from the table, initial aerosols were in some cases five times larger than measured data. The fraction airborne increased 10% on the average. Observations from this table lead to some basic conclusions about accident analysis methods:

1. If liquid aerosols are the primary airborne hazard in an accident scenario, the evaporation/settling models should be used to describe the aerosol behavior in the accident compartment.

TABLE 3.3. Observations from Evaporation/Settling Calculations

	<u>Initial(a)</u>	<u>Measured(b)</u>	<u>Initial/Measured</u>
<u>All Spills</u>			
Average AMMD, μm	21.5	8.9	2.4
Average σ_g	7.3	5.4	1.4
Average Fract. Airb. $\times 10^6$	28.5	25.5	1.1
<u>UNH Spills</u>			
Average AMMD, μm	27.2	20.1	1.4
Average σ_g	5.9	4.5	1.3
Average Fract. Airb. $\times 10^6$	9.3	7.9	1.2
<u>Uranine Spills</u>			
Average AMMD, μm	27.0	7.3	3.7
Average σ_g	3.0	2.9	1.0
Average Fract. Airb. $\times 10^6$	56.2	50.7	1.1
<u>Sucrose Spills</u>			
Average AMMD, μm	11.0	2.7	4.1
Average σ_g	14.2	9.4	1.5
Average Fract. Airb. $\times 10^6$	10.6	9.7	1.1
<u>Slurry Spills</u>			
Average AMMD, μm	15.8	3.1	5.1
Average σ_g	10.1	6.7	1.5
Average Fract. Airb. $\times 10^6$	23.3	20.7	1.1
<u>Low Surface Tension Spills</u>			
Average AMMD, μm	20.2	6.5	3.1
Average σ_g	3.7	4.2	0.9
Average Fract. Airb. $\times 10^6$	45.9	41.5	1.1

(a) Calculated using evaporation/settling codes.

(b) Impactor data.

2. Temperature, relative humidity (if water is a solvent), and volatile solvent/solute characteristics are important parameters in evaporation models.
3. It would be useful to develop a universal evaporation/settling code that would accept subroutines to calculate the unique vapor pressure versus temperature/composition relationships existing for every system of solutes and solvents.

3.4 CORRELATION SUMMARY

All correlations for the fraction airborne and the statistical summary of drop size parameters are contained in this section. When analyzing a liquid spill, effort should be made to obtain an evaporation/settling model to determine the size of airborne particles after release. Equations based on the corrected data should be used with the evaporation/settling model to obtain the most realistic assessment of the source term. If an appropriate model is not available, then equations based on the original data provide the best estimate of the source term.

3.4.1 Correlations for Fraction Airborne

The correlations for the fraction airborne were obtained using the original data of Sutter et al. (1981) and Ballinger and Hodgson (1986) and the data corrected for evaporation and settling (see Section 3.3). Fraction airborne was correlated with various functional forms of the Archimedes, Reynolds, and Froude numbers and dimensionless viscosities and densities defined in Section 3.2. These correlations are presented below.

The first correlation is that of the impactor data (Table A.1) with the Archimedes number (Equation 3). The best correlation found (highest r^2 , see Table A.2, for regression details) is

$$F = 8.12 \times 10^{-10} \text{ Arch}^{0.55} \quad (24)$$

The second correlation is that of the corrected data with the Archimedes number. The regression analysis (Table A.3) gave

$$F = 8.9 \times 10^{-10} \text{ Arch}^{0.55} \quad (25)$$

as the best correlation with the Archimedes number, in the same format as the previous correlation. This correlation has a lower r^2 than that shown in Equation (24), but its r^2 is satisfactory. Equation (25), which is based on

the corrected data, may underestimate F, because settling and evaporation of particles during the sampling period was not accounted for. However, Equation (24) has the experimental evaporation/ settling inherent in the correlating data. Therefore, if an evaporation/settling model is not available for a liquid spill analysis, Equation (25) should be used.

The third correlation is of the corrected data with the Archimedes and Froude numbers and dimensionless density. A multiple regression analysis (Table A.4) gave

$$F = 6.31 \times 10^{-6} Fr^{0.35} Arch^{0.45} \left(\frac{\rho_a}{\rho_l} \right)^{2.20} \quad (26)$$

where Arch and Fr are defined by Equations (3) and (4), respectively

$$\begin{aligned} \rho_a &= \text{density of air, g/cm}^3 \\ \rho_l &= \text{density of solution, g/cm}^3 \end{aligned}$$

The Archimedes number in this case is based on ρ_{liq} . This correlation has a higher r^2 than that described in Equation (25), but is more difficult to use. Three numbers must be calculated instead of one, and the equivalent sphere radius, R, and impact velocity, V, must be computed to obtain the Froude number. Equation (25) represents the preferred equation for estimating the mass of the initial aerosol. It is easier to use than the correlation shown in Equation (26) and provides a more realistic estimate than Equation (24). However, an evaporation/settling model should be used with this equation.

3.4.2 Statistical Summary of Drop Size Parameters

Many attempts were made to correlate the lognormal drop size parameters AMMD, and σ_g with other variables (F, We, Re, Fr, Arch). No regression analyses produced any $r^2 > 60\%$ except for AMMD of drops made airborne from slurry spills. Thus, correlations for all liquid spills except slurries were dropped in favor of simple statistical representations of the lognormal drop size parameters. These are summarized in Table 3.4. Use of these parameters will be illustrated using sample problems in Section 3.6.

The following equations may be used to calculate the AMMD from aerosols produced from slurry spills. If an evaporation/settling model is not used, an equation derived from data measured by an impactor may be applied:

$$AMMD, \mu m = 4.0 \times 10^{-15} \frac{We^{3.2}}{Re^{0.25}} \quad r^2 = 74.2\% \quad (27)$$

TABLE 3.4. Statistical Summary of Drop Size Parameters for Lognormal Distributions

	<u>Mean</u>	<u>St. Dev.</u>	<u>95% Conf. Interval</u>
<u>Impactor Data:</u>			
AMMD, μm			
All Data	8.9	10.0	5.9 - 11.8
UNH Spills	20.1	14.3	11.0 - 29.2
Uranine Spills	7.3	2.4	6.0 - 8.6
Sucrose Spills	3.4	2.4	1.8 - 4.9
Slurry Spills	3.1	1.0	2.3 - 4.0
σ_g			
All Data	5.4	4.1	4.2 - 6.6
UNH Spills	4.5	2.1	3.2 - 5.9
Uranine Spills	2.9	0.6	2.6 - 3.2
Sucrose Spills	8.5	6.5	4.4 - 12.6
Slurry Spills	6.7	1.5	5.4 - 8.0
<u>Corrected Data:</u>			
AMMD, μm			
All Data	21.5	15.3	17.0 - 26.0
UNH Spills	27.2	26.8	10.2 - 44.2
Uranine Spills	27.1	3.9	24.9 - 29.2
Sucrose Spills	12.5	7.2	7.9 - 17.1
Slurry Spills	15.8	3.1	13.2 - 18.4
σ_g			
All Data	7.3	7.6	5.0 - 9.5
UNH Spills	6.0	3.5	3.7 - 8.2
Uranine Spills	3.0	0.4	2.8 - 3.3
Sucrose Spills	12.3	13.2	3.5 - 21.2
Slurry Spills	10.1	3.5	7.3 - 13.0

If an evaporation/settling model is used in the spill analyses, the following equation, which is the result of a correlation of the corrected data, should be used.

$$\text{AMMD, } \mu\text{m} = 2.3 \times 10^{-8} \frac{\text{We}^{1.9}}{\text{Re}^{0.16}} \quad r^2 = 99.7\% \quad (28)$$

3.5 LARGE FALL HEIGHTS, SMALL DROPS, AND CONTINUOUS SPILLS

The correlations might be used to analyze accident scenarios in which the fall height is greater than 3 m, individual drops are 8 mm or less in diameter, or a continuous stream is impacting a floor. The experimental conditions of the data were H (spill height) is <3 m, D (diameter of spill container) ranged from 6.2 to 12.4 cm, and single (not continuous) events. Air resistance would cause liquid to disperse rapidly above 3 m and would tend to disperse the spill glob into many droplets <8 mm diameter, all falling at a terminal velocity of 30 ft/s or less (Perry and Chilton 1973). This velocity, $V = \sqrt{2 g H}$, is equivalent to a frictionless fall of 4.26 m. However, this velocity is less than one would get from a large glob (before it shatters) falling from a greater height. Therefore, for spills where the drop diameter, D, is <8 mm, and the spill height, H, is >4.26 m, the results of the correlations will probably be conservatively high because of the way velocity is calculated. In the case of continuous spills, where a falling stream provides less air resistance than many droplets, the equations may not be conservative.

Caution should be exercised in any departure from the conditions under which the correlations were derived. The results of using these correlations should be conservative, even for large-scale continuous spills.

3.6 EXAMPLE PROBLEMS

The following examples illustrate the use of the correlations developed in this report.

Example 1. A 2-L beaker on an elevated platform in a glovebox is accidentally tipped over, spilling its contents on the glovebox floor 61 cm (2 ft) below. How much wet aerosol will challenge the glovebox ventilation system?

The given conditions are for 850 cc of very dilute $\text{Pu}(\text{NO}_3)_4$ solution (0.1 M) and 0.5 M HNO_3 , at 25°C. Important parameters are

$$\begin{aligned} H &= 61 \text{ cm (2 ft)} \\ \rho_a &= 0.00155 \text{ g/cc} \\ \gamma &= 69 \text{ dyne/cm (Fleming 1953)} \\ \mu &= 0.0103 \text{ g/cm s (2.5 lbm/ft h) (Fleming 1958).} \end{aligned}$$

Equation (25) should be used for the fraction made airborne before settling and fallout. The Archimedes number is defined as

$$\begin{aligned}\text{Arch} &= \rho^2 H^3 g / \mu^2 \\ &= (0.00155 \text{ g/cm}^3)^2 (61 \text{ cm})^3 (981 \text{ cm/s}^2) / \\ &\quad (0.0103 \text{ g/cm s})^2 \\ &= 5.04 \times 10^6 \text{ (dimensionless)}\end{aligned}\tag{29}$$

$$\begin{aligned}F &= 8.9 \times 10^{-10} \text{ Arch}^{0.55} \\ &= 4.32 \times 10^{-6}\end{aligned}\tag{30}$$

$$\begin{aligned}\text{Amount airborne} &= 850 \text{ cc spilled} \times 4.32 \times 10^{-6} \\ &= 3.67 \times 10^{-3} \text{ cc}\end{aligned}\tag{31}$$

For particle size, the user may choose any of the options in Table 3.4. Because the solution spilled in this example is not exactly like any used in the table, the user should use the mean AMMD and σ_g listed for "all data," under corrected data, to remain consistent with the choice of equation for F. The resulting values are

$$\begin{aligned}\text{AMMD} &= 21.5 \text{ }\mu\text{m} \\ \sigma_g &= 7.3\end{aligned}$$

An evaporation/settling code must be used to describe the spilled aerosol more precisely, as the size of the particles are reduced by evaporation and settling before being transported to the ventilation system. An alternative approach is to use Equation (24) for F:

$$\begin{aligned}F &= 8.12 \times 10^{-10} \text{ Arch}^{0.55} \\ &= 3.94 \times 10^{-6}\end{aligned}\tag{32}$$

and the impactor data for AMMD and σ_g from Table 3.3:

$$\begin{aligned}\text{AMMD} &= 8.9 \text{ }\mu\text{m} \\ \sigma_g &= 5.4\end{aligned}$$

and thus remain consistent with the choice of Equation (24).

The 95% confidence intervals for σ_g and AMMD are given in Table 3.4, but the confidence intervals for F are not given. Confidence estimates for F are more difficult to obtain. For regression lines, this is a venturi-shaped band around the line that is difficult to calculate; the user is referred to Ryan et al. (1976) or Snedecor and Cochran (1980) for precise methods. The

confidence interval is estimated on the assumption that all data will be normally distributed about the regression line (Spiegel 1961). Thus, 68% of the data will fall within 2 ± 1 standard deviation. Also, 90% will fall within ± 1.65 standard deviation of the regression line and 95% within ± 1.96 standard deviation. This is only an approximate and nonconservative estimate, which underestimates the true confidence limits.

Example 2. Suppose a leak has developed in a solvent line in a reprocessing plant. The solvent is 30% tributyl phosphate (TBP in kerosene diluent). It contains 0.1 M UNH and 1 M HNO_3 . Assume the leak is at 25°C and falls 366 cm (12 ft) to the floor. What fraction is airborne, and what are the aerosol characteristics?

The properties needed for the solvent can be estimated from data in the PUREX Technical Manual (1955) and Fleming (1958). These are:

$$\rho_a = 0.00155 \text{ g/cc}$$

$$\mu = 5 \text{ lbm/ft h} = 0.0207 \text{ g/cm s},$$

$$\begin{aligned} \text{thus Arch} &= (\rho_a^2 H^3 \text{ g})/\mu^2 \\ &= (0.00155 \text{ g/cm}^3)^2 (366 \text{ cm})^3 (981 \text{ cm/s}^2)/(0.0207 \text{ g/cm s})^2 \\ &= 2.7 \times 10^8. \end{aligned} \tag{33}$$

Using Equation (25)

$$F = 8.9 \times 10^{-10} \text{ Arch}^{0.55} = 3.86 \times 10^{-5}, \text{ and} \tag{34}$$

from Table 3.3

$$\text{AMMD} = 21.5$$

$$\sigma_g = 7.3$$

As mentioned in Section 3.5, the above number for F is probably conservatively high. The degree of conservatism cannot easily be checked without experiments. One could, for example, examine the single-drop splash data in Stow and Stainer (1977) for water to see if the small particle end of their data is applicable. Until experiments can extend the range of correlations, the approach described above for solving the problem presented in Example 2 is assumed to be the most reliable.

4.0 MODELS FOR FREE FALL SPILLS OF POWDERS

There are two mechanisms during the free fall spill of a powder that contribute to the generation of airborne, respirable dust. The first mechanism is the shearing effect of the air on the powder as it falls. The shearing effect is enhanced by the lengthening of the spill column caused by the acceleration of gravity. The second is the breakup of larger particles on impact with the stopping surface.

This section provides a model of the formation of airborne particles from a spill, assuming that the ball of powder grows at a constant rate as it falls and that mass airborne is proportional to the drag force on the bulk powder. The model does not account for particles becoming airborne as a result of impact with a surface. Therefore, it can be correlated with experimental spills only if the portion of airborne dust resulting from the impact can be neglected. This model is based on experimental results reported by Sutter et al. (1981). Because only two powders were used in these experiments, TiO_2 and DUO, the models may be limited by powder characteristics. Both TiO_2 and DUO are fine powders with more than 90% of the mass of the source powder under $6\ \mu$. A powder with larger particles would probably generate less mass airborne during a spill.

The following subsections describe the development of the proportionality constant and correlate the results of experiments (Sutter et al. 1981) using the free fall spills of TiO_2 and DUO powders. The contribution of mass to the source term from impacting the floor is probably small for these experiments.

4.1 POWDER SPILL MODEL

A powder spill is portrayed in an idealized scenario in Figure 4.1. The powder is shown exiting from a beaker that is at ambient pressure. As the powder falls, it disperses at a constant angle. At any distance X from the initial height of the spill, the diameter and velocity of the powder front can be calculated from the angle of dispersion and properties of the powder. As the bulk mass of powder descends, particles are sheared off and remain suspended. As the powder hits the floor, larger particles may break up and become resuspended, but this effect is not as significant as entrainment of particles on the way down.

The following assumptions are made in the powder spill model:

- Growth rate of the powder front is constant and can be characterized by an angle of dispersion.

FIGURE 4.1. Powder Spill

- Amount airborne is proportional to the drag force on the powder front.
- Diameter of powder front at start of spill is equal to diameter of container from which it was spilled (beaker in experiments).

A computer code PSPILL was developed to model powder spills. This code was written in FORTRAN on a VAX 11/70 with a VMS operating system but does not require significant memory or storage space and can easily be run on a micro-computer. The model may even be applied by hand if the spill height is small. However, this quickly becomes a tedious and repetitive process and is not recommended.

The diameter of the powder front at the start of the spill is equal to the diameter (or equivalent diameter) of the container from which it was spilled. After an increment of time, the front has descended a distance of X and the new powder front diameter is equal to

$$d = d_0 + 2 (X) \tan (\alpha) \quad (35)$$

where d = new powder front diameter
 d_0 = initial powder front diameter
 α = angle of dispersion

$\tan \alpha$ is set in the code to 0.1283 ($\alpha = 7.3^\circ$). This was the average value determined from high-speed photographs of experimental runs.

Initial powder velocity is equal to the acceleration from gravity multiplied by the first time step:

$$V_0 = g \times \Delta t \quad (36)$$

where V_0 = initial velocity
 g = gravity constant
 Δt = length of time steps (0.01 s is recommended)

Thereafter, velocity is determined from the equation

$$\frac{d(mV)}{dt} = mg - FV2 \quad (37)$$

where m = powder mass
 V = velocity
 t = time
 g = gravity constant
 $FV2$ = drag force on powder

Expanded and rearranged, this equation becomes

$$V_1 = V_0 + \Delta t(g - FV2/m) - \frac{V_0 \Delta m}{m} \quad (38)$$

The drag force $FV2$ is calculated by

$$FV2 = (\pi r^2) (1/2 \rho V^2) C_D \quad (39)$$

where r = radius of powder front
 ρ = powder bulk density
 V = velocity
 C_D = drag coefficient, a function of Re (Welty et al. 1976)
 $Re = \rho_{air} V d / \mu_{air}$

The mass of powder airborne during each time step is assumed proportional to the drag force

$$AB = EPS \times FV2 \times \Delta t \quad (40)$$

where AB = mass airborne
 EPS = proportionality factor
 FV2 = drag force
 Δt = time step

Through statistical regressions of the experimental data, the proportionality factor was found to be a function of two dimensionless numbers, the Reynolds and Froude numbers. The relationship

$$EPS = 2 \times 10^{-4} (Re^2/Fr)^{1.03} \quad r^2 = 69.1\% \quad (41)$$

gave the best fit. Re^2/Fr is also referred to as the Galileo number.

Input to PSPILL are:

Mo - mass of powder spilled, kg
 H - spill height, m
 ρ - bulk density of the powder, kg/m^3
 ρ_a - air density, kg/m^3
 μ_a - air viscosity, $Pa \cdot s$
 Δt - increment of time step, s
 n - number of time steps between printouts

The code was run with varying values for Mo (0.1, 1.0, 5.0 kg), H (0.5, 1.0, 2.0, 3.0, 4.0, 5.0 m), and ρ (1000, 2000 kg/m^3). The fraction of powder made airborne as a result of these runs was statistically analyzed against the input values. The following algorithm was developed from this analysis and can be used in place of the code to predict the source term from powder spills if the air density and viscosity are 1.18 kg/m^3 and 1.85×10^{-5} , respectively.

$$F_r = 0.1064 \frac{Mo^{0.125} H^{2.37}}{\rho^{1.02}} \quad r^2 = 99.4\% \quad (42)$$

As shown, the correlation coefficient was 99.4%, indicating a very strong correlation.

4.2 AEROSOL PARTICLE SIZE

Attempts were made to correlate the aerodynamic equivalent mass median diameter (AMMD) of the airborne powder with numerous combinations and

functional relationships of the independent and dependent variables. Sutter et al. (1981) plotted AMMD versus the fraction airborne (F) and showed some functional relationships. After numerous attempts at correlations, the best apparently available was

$$\text{AMMD}(\mu\text{m}) = 12.1 - 3.29 \rho_p + 7540F \quad (43)$$

where ρ_p = bulk density of powder, g/cm³
 F = fraction airborne

Equation (43) had only a 46% correlation (see Table B.2) and a standard deviation of 5.7.

The particles made airborne are agglomerates of a much smaller primary size. Thus, the statistics relating particle size to spill events are also coupled with the statistics of agglomerates in the bulk powder state. This might be the reason for the high standard deviation.

Table 4.1 summarizes the statistical data for the log normal distribution parameters for the powder spills. The 90% confidence interval on AMMD is

$$-1.697 \text{ s.d.} + \text{AMMD to } 1.697 \text{ s.d.} + \text{AMMD}$$

The relationship for σ_g is, similarly,

$$-1.697 \text{ s.d.} + \sigma_g \text{ to } 1.697 \text{ s.d.} + \sigma_g$$

for the 90% confidence interval.

TABLE 4.1. Statistical Summary of Particle Size Parameters for the Log Mean Distributions

All Powders: AMMD = 13.2 (Standard Dev, s.d. = 7.45)
 (90% CI: 10.9 to 15.5)

σ_g = 4.82 (s.d. = 1.69)
 (90% CI: 4.30 to 5.34)

DUO: AMMD = 9.55 (s.d. = 4.99)
 (90% CI: 7.08 to 12.0)

σ_g = 3.73 (s.d. = 0.94)
 (90% CI: 3.27 - 4.20)

TiO₂: AMMD = 15.9 (s.d. = 7.92)
 (90% CI: 12.6 to 19.1)

σ_g = 5.60 (s.d. = 1.69)
 (90% CI: 4.91 to 6.30)

4.3 EXAMPLE PROBLEMS

For a given spill of powder (unspecified type) the following spill parameters are known or estimated:

$M_o = 0.5 \text{ kg}$
 $H = 2 \text{ m}$
 $\rho = 1000 \text{ kg/m}^3$
 $\rho_a = 1.18 \text{ kg/m}^3$
 $\mu_a = 1.85 \times 10^{-5} \text{ Pascal}\cdot\text{s}$

If a time step of 0.01 sec is chosen and the user desires a printout every 10 time steps, then the following input to PSPILL is applicable (Appendix C):

M_o	H	ρ	ρ_a	μ_a	Δt	N
0.5,	2.0,	1000.,	1.18,	1.85E-5,	0.01,	10

The output from the sample problem is shown below:

MASS(KG)= 0.50000 H(M)= 2.00 BULKD(KG/M3) = 1000.00
RHO AIR(KG/M3)= 1.18 VISC AIR(PA*S)= 0.1846E-05 DELT=0.0100

<u>TIME</u> <u>(SEC)</u>	<u>DIST</u> <u>(M)</u>	<u>VEL</u> <u>(M/S)</u>	<u>RAD AB</u> <u>(MG)</u>
0.100	0.054	1.078	0.096
0.200	0.206	2.053	0.984
0.300	0.454	3.008	5.093
0.400	0.796	3.894	19.376
0.500	1.219	4.621	44.449
0.600	1.706	5.104	60.617
0.660	2.016	5.230	60.571

TOTAL RAD AB(MG) = 191.19 PERCENT AB = 0.0382

An alternative method of calculating the amount airborne is to use Equation (42). The fraction airborne (F) using this method is 4.4×10^{-4} , which is slightly more conservative than the code for this example. For appropriate particle sizes, use Equation (43). For DUO in this example

$$\text{AMMD} = 12.1 - 3.29 (1.0) + 7540 (3.8 \times 10^{-4}) \quad (44)$$

$$= 11.7 \mu\text{m}$$

$$\sigma_g = 3.73 \quad (\text{Table 4.1: DUO})$$

and for PbO (a finely divided powder with a bulk density of 1.3 g/cc)

$$\text{AMMD} = 12.1 - 3.29 (1.3) + 7540 (3.8 \times 10^{-4}) \quad (45)$$

$$= 10.7 \mu\text{m}$$

$$\sigma_g = 4.82 \quad (\text{Table 4.1: ALL POWDERS})$$

5.0 REFERENCES

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APPENDIX A

SUPPORTING DATA AND MODELS
FOR FREE FALL SPILLS OF LIQUIDS

TABLE A.1. Data from Free Fall Spills of Liquids

Ht (m)	Vol. (cc)	Impactor Data			Initial Aerosol				% mass settled	ρ (g/cc)	μ (cp)	α (dyne/cm)	T (°C)
		AED (μ m)	σ_g	Fract. Airb. $\times 10^6$	AED (μ m)	σ_g	Dried AED (μ m)	Fract. Airb. $\times 10^6$					
UNH Spills													
3	1000	34	4.1	10.9	75	9.2	5.1	12.9	15.8	1.6	3.1	59.0	14
1	125	3.2	2.3	5.4	3.9	2.8	2.1	5.9	8.7	1.6	3.1	59.0	14
1	500	2.3	1.9	1.3	1.9	2.1	1.0	1.4	5.6	1.6	3.1	59.0	13
3	125	20	7.0	13.3	16.5	4.9	4.8	15.2	12.5	1.6	3.1	59.0	18
3	500	4.2	2.2	3.2	2.4	1.6	2.2	14.3	7.6	1.6	3.1	59.0	15
1	1000	9.6	5.0	1.1	7.5	5.2	3.7	1.4	20.2	1.6	3.1	59.0	16
3	125	30	6.0	19.7	43	8.6	5.0	23.6	16.5	1.6	3.1	59.0	16
1	500	27	6.6	1.5	21	5.0	4.9	1.8	17.4	1.6	3.1	59.0	12
1	125	5.0	7.8	1.7	5	9.1	3.5	1.9	9.3	1.6	3.1	59.0	13
1	1000	40	5.8	1.2	75	13.6	15.0	1.4	14.6	1.6	3.1	59.0	13
3	1000	32	3.6	11.4	41	5.1	4.7	14.3	20.4	1.6	3.1	59.0	16
3	500	34	2.1	13.5	34	4.0	4.8	17.5	22.8	1.6	3.1	59.0	17
Uranine Spills													
3	500	8.0	3.5	38.8	29.7	3.4	6.6	43.2	10.1	0.944	1.53	57.5	(b)
3	500	11.0	2.8	31.3	31.5	3.8	7.0	35.4	11.5	0.944	1.53	57.5	(b)
3	500	4.5	2.4	30.7	21.8	3.0	4.8	32.2	4.8	0.944	1.53	57.5	(b)
3	500	4.8	2.6	55.8	23.5	3.1	5.2	60.9	8.3	0.944	1.53	57.5	(b)
3	500	5.4	2.6	51.0	23.9	2.7	5.3	55.7	8.5	0.944	1.53	57.5	(b)
3	500	4.8	3.0	32.6	20.1	2.9	4.5	35.3	7.6	0.944	1.53	57.5	(b)
3	500	4.8	3.0	32.6	20.1	2.9	4.5	35.3	7.6	0.944	1.53	57.5	(b)
3	500	6.8	3.1	37.0	28.4	3.3	6.3	40.8	9.4	0.944	1.53	57.5	(b)
3	1000	11.5	3.1	127.0	31.4	3.5	7.0	143.3	11.4	0.944	1.53	57.5	(b)
1	1000	8.6	4.1	30.4	29.0	2.8	6.5	33.6	9.5	0.944	1.53	57.5	(b)
3	1000	10.0	2.6	80.2 (a)	30.2	3.0	6.7	89.9	10.8	0.944	1.53	57.5	(b)
3	125	9.2	3.7	160.0 (a)	29.9	2.8	6.6	177.8	10.0	0.944	1.53	57.5	(b)
1	500	6.0	2.1	5.8	27.3	3.2	6.1	6.3	8.6	0.944	1.53	57.5	(b)
1	125	4.2	1.8	6.2	21.1	2.2	4.7	6.7	7.8	0.944	1.53	57.5	(b)
3	125	7.2	3.0	33.2	28.9	3.0	6.4	36.8	9.7	0.944	1.53	57.5	(b)
Sucrose Spills													
3	1000	1.6	10	3.8	6.5	13.4	1.4	4.1	.81	1.188	8.0	70.9	18
3	1000	1.9	7.7	1.6	11.6	0.3	2.6	1.7	8.4	1.277	47.1	73.2	18.5
3	1000	4.8	7.7	7.3	19.1	10.0	4.3	8.2	10.5	1.102	2.6	68.5	17
3	1000	2.2	5.5	2.7	7.1	8.0	1.6	2.9	8.1	1.233	17.2	83.0	20
3	1000	9.0	5.8	32.2	21.3	6.4	4.7	35.9	10.2	1.005	1.3	63.3	17

(a) splashing on filters.

(b) temperature not measured.

TABLE A.1. (contd)

Ht (m)	Vol. (cc)	Impactor Data			Initial Aerosol				% mass settled	ρ (g/cc)	μ (cp)	α (dyne/cm)	T (°C)
		AED (μ m)	σ_g	Fract. Airb. $\times 10^6$	AED (μ m)	σ_g	Dried AED (μ m)	Fract. Airb. $\times 10^6$					
Sucrose Spills (contd)													
3	1000	1.6	4.6	2.1	6.9	51.0	1.5	2.2	5.9	1.235	17.7	71.8	16
3	1000	2.7	7.5	11.1	13.5	8.8	3.0	12.2	8.7	1.101	2.5	69.3	16
3	1000	0.6	10	2.3	2.4	13.1	0.5	2.4	5.6	1.276	44.9	75.7	18
3	1000	0.6	28	5.7	3.2		0.7	6.1	7.0	1.191	7.9	70.8	19.5
3	1000	4.3	7.2	28.0	18.2	8.2	4.0	30.5	8.3	1.005	1.3	67.0	19.5
Slurry Spills													
3	1000	2.1	7.1	9.2	13.9	14.3	3.1	10.1	9.1	1.189	3.1	64.6	23.5
3	1000	2.3	8.0	8.7	13.3	16.4	3.0	9.6	9.3	1.123		58.2	19.5
3	1000	3.0	3.8	46.0	15.5	6.5	3.4	52.0	11.5	1.334	1.3	63.1	19
3	1000	4.7	6.1	11.0	21.2	8.6	4.7	12.6	12.5	1.155			18.5
3	1000	2.5	8.4	17.6	12.8	9.5	2.9	19.4	9.4	1.201	3.1	68.4	29.0
3	1000	2.6	7.8	30.2	14.0	10.0	3.1	33.5	9.9	1.288	1.3	64.9	19.5
3	1000	4.7	6.9	16.0	19.7	8.5	4.4	18.7	14.6	1.407	2.9	62.8	19
3	1000	3.1	5.6	27.2	15.8	7.3	3.5	30.4	10.4	1.345	1.3	63.4	21.5
Low Surface Tension Spills													
3	1000	7.8	3.8	38.1	34.2	3.0	4.9	42.6	10.6	1.004	1.14	33.3	17
3	1000	5.2	4.5	44.8	17.3	4.4	4.3	49.2	8.9	1.004	1.24	33.33	15

TABLE A.2. Regression Analysis for Fraction Airborne from Liquid Spills - Impactor Data

LOG-FRM = LOG_{10} (Fraction Airborne-Measured Data)
 L-ARCH-A = LOG_{10} (Archimedes Number)

The regression equation is
 LOG-FRM = - 9.09 + 0.550 L-ARCH-A.

45 cases used, 2 cases contain missing values.

<u>Predictor</u>	<u>Coefficient</u>	<u>Standard Deviation</u>	<u>t-ratio</u>
Constant	-9.0941	0.3187	-28.54
L-ARCH-A	0.55007	0.04130	13.32

s = 0.2573 R-sq = 80.5% R-sq(adj) = 80.0%

Analysis of Variance:

<u>Source</u>	<u>Degrees of Freedom</u>	<u>Sum of Squares</u>	<u>Mean Square</u>
Regression	1	11.744	11.744
Error	43	2.847	0.066
Total	44	14.591	

Unusual Observations:

<u>Observations</u>	<u>L-ARCH-A</u>	<u>LOG-FRM</u>	<u>Fit</u>	<u>Standard Deviation of Fit</u>	<u>Residual</u>	<u>Standard Residual^(a)</u>
21	8.43	-3.8962	-4.4547	0.0499	0.5585	2.21R
22	7.00	-4.5171	-5.2421	0.0470	0.7250	2.87R
24	8.43	-3.7959	-4.4547	0.0499	0.6589	2.61R
29	5.45	-5.7959	-6.0952	0.989	0.2993	1.26 X
35	5.49	-5.6383	-6.0723	0.0973	0.4341	1.82 X

- (a) R denotes an observation with a large standard residual.
 X denotes an observation whose X value gives it large influence.

TABLE A.3. Regression Analysis for Fraction Airborne from Liquid Spills - Evaporation/Settling Corrected Data

LOG-FRI = LOG_{10} (Fraction Airborne-Corrected Data)
L-ARCH-A = LOG_{10} (Archimedes Number)

The regression equation is
 $\text{LOG-FRI} = 9.05 + 0.550 \text{ L-ARCH-A}$.

45 cases used, 2 cases contain missing values.

<u>Predictor</u>	<u>Coefficient</u>	<u>Standard Deviation</u>	<u>t-ratio</u>
Constant	-9.0473	0.3131	-28.90
L-ARCH-A	0.55006	0.04058	13.56

s = 0.2528

R-sq = 81.0%

R-sq (adj) = 80.6%

Analysis of Variance:

<u>Source</u>	<u>Degrees of Freedom</u>	<u>Sum of Squares</u>	<u>Mean Square</u>
Regression	1	11.743	11.743
Error	43	2.748	0.064
Total	44	14.491	

Unusual Observations:

<u>Observations</u>	<u>L-ARCH-A</u>	<u>LOG-FRM</u>	<u>Fit</u>	<u>Standard Deviation of Fit</u>	<u>Residual</u>	<u>Standard Residual^(a)</u>
221	8.43	-3.8438	-4.4080	0.0491	0.5642	2.28R
22	7.00	-4.4737	-5.2953	0.0462	0.7217	2.90R
24	8.43	-3.7501	-4.4080	0.0491	0.6579	2.65R
29	5.45	-5.7696	-6.0484	0.0972	0.2789	1.19 X
35	5.49	-5.6198	-6.0256	0.0956	0.4058	1.73 X

- (a) R denotes an observation with a large standard residual.
X denotes an observation whose X value gives it large influence.

TABLE A.4. Regression Analysis for Fraction Airborne from Liquid Spills - Best Fit of Data

LOG-FRI = LOG_{10} (fraction Airborne - Corrected Data)
 LOG FR = LOG_{10} (Froude Number)
 LOG ARCH = LOG_{10} (Archimedes Number)
 L(pa/pl) = LOG_{10} (Density of air/solution density)

The regression equation is

$$\text{LOG-FRI} = -5.20 + 0.353 \text{ LOG FR} + 0.450 \text{ LOG ARCH} + 2.20 \text{ (Lpa/pl)}.$$

45 cases used, 2 cases contain missing values.

<u>Predictor</u>	<u>Coefficient</u>	<u>Standard Deviation</u>	<u>t-ratio</u>
Constant	-5.198	1.554	-3.34
LOG FR	0.3532	0.2192	1.61
LOG ARCH	0.44952	0.05043	8.91
L(pa/pl)	2.2034	0.4330	5.09

s = 0.2322 R-sq = 84.7% R-sq(adj) = 83.6%

Analysis of Variance:

<u>Source</u>	<u>Degrees of Freedom</u>	<u>Sum of Squares</u>	<u>Mean Square</u>
Regression	3	12.2807	4.0936
Error	41	2.2106	0.0539
Total	44	14.4913	

<u>Source</u>	<u>Degrees of Freedom</u>	<u>Sequential Sum of Squares</u>
LOG FR	1	4.9153
LOG ARCH	1	5.9694
L(pa/pl)	1	1.3960

Unusual Observations:

<u>Observations</u>	<u>LOG FR</u>	<u>LOG-FRI</u>	<u>Fit</u>	<u>Standard Deviation of Fit</u>	<u>Residual</u>	<u>Standard Residual^(a)</u>
21	1.98	-3.8438	-4.3667	0.0529	0.5229	2.31R
22	1.51	-4.4737	-5.1786	0.1065	0.7050	3.42R
24	2.29	-3.7501	-4.2604	0.0725	0.5103	2.31R

(a) R denotes an observation with a large standard residual.

TABLE A.5. Regression Analysis for Aerodynamic Mass Median Diameter (AMMD)
for Slurry Spills - Impactor Data

C45 = LOG₁₀ (AMMD-Impactor Data)
C61 = LOG₁₀ (Weber Number)
C65 = LOG₁₀ (Reynolds Number)

C45	C61	C65
1.14301	5.12839	6.80051
1.12385	5.14890	*
1.19033	5.18857	7.22791
1.32634	*	*
1.10721	5.10793	6.80487
1.14613	5.16111	7.21267
1.29447	5.21378	6.90259
1.19866	5.19007	7.23147

The regression equation is
C45 = - 7.63 + 1.92 C61 - 0.155 C65.

6 cases used, 2 cases contain missing values.

<u>Predictor</u>	<u>Coefffficient</u>	<u>Standard Deviation</u>	<u>t-ratio</u>
Constant	-7.6277	0.2050	-37.20
C61	1.91687	0.04484	42.75
C65	-0.155463	0.008404	-18.50

s = 0.003383

R-sq = 99.8%

R-sq(adj) = 99.7%

Analysis of Variance:

<u>Source</u>	<u>Degrees of Freedom</u>	<u>Sum of Squares</u>	<u>Mean Square</u>
Regression	2	0.021337	0.010668
Error	3	0.000034	0.000011
Total	5	0.021371	

<u>Source</u>	<u>Degrees of Freedom</u>	<u>Sequential Sum of Squares</u>
C61	1	0.017421
C65	1	0.003916

TABLE A.6. Regression Analysis for Aerodynamic Mass Median Diameter (AMMD)
for Slurry Spills - Evaporation/Settling Corrected Data

C37 = LOG_{10} (AMMD - Corrected Data)

C61 = LOG_{10} (Weber Number)

C65 = LOG_{10} (Reynolds Number)

C37	C61	C65
0.322219	5.12839	6.80051
0.361728	5.14890	*
0.477121	5.18857	7.22791
0.672098	*	*
0.397940	5.10793	6.80487
0.414973	5.16111	7.21267
0.672098	5.21378	6.90259
0.491362	5.19007	7.23147

The regression equation is

$$C37 = -14.4 + 3.20 C61 - 0.245 C65.$$

6 cases used, 2 cases contain missing values.

Predictor	Coefficient	Standard Deviation	t-ratio
Constant	-14.355	3.670	-3.91
C61	3.2019	0.8027	3.99
C65	-0.2446	0.1504	-1.63

s = 0.06056

R-sq = 84.5%

R-sq(adj) = 74.2%

Analysis of Variance:

Source	Degrees of Freedom	Sum of Squares	Mean Square
Regression	2	0.060082	0.030041
Error	3	0.011001	0.003667
Total	5	0.071083	

Source	Degrees of Freedom	Sequential Sum of Squares
C61	1	0.050386
C65	1	0.009696

APPENDIX B

SUPPORTING INFORMATION, DATA, AND MODEL FOR FREE FALL SPILLS OF POWDERS

TABLE B.1. Original Free Fall Data and Calculated Parameters

Height	Source Mass (g)	Mass Airborne (g)	% Airborne	AMMD (μm)	σ (g)	Powder Bulk Density (g/cm^3)
<u>TiO₂</u>						
3	471.9	0.91050	0.192900	21.00	6.20	0.63
3	450.4	0.14750	0.032750	5.80	5.60	0.63
3	431.1	0.08580	0.019900	16.00	6.60	0.63
3	460.0	0.39000	0.084780	17.20	6.90	0.63
3	100.0	0.11940	0.119400	27.00	7.50	0.63
3	100.0	0.09900	0.099000	13.00	5.90	0.63
3	1000.0	1.48000	0.148000	36.00	6.70	0.63
3	1000.0	0.96590	0.096590	17.00	7.40	0.63
3	25.0	0.24000	0.096000	9.60	3.70	0.63
3	25.0	0.00400	0.016000	7.20	3.10	0.63
1	441.1	0.03450	0.007820	15.30	7.70	0.63
1	451.0	0.03600	0.007980	16.80	6.50	0.63
1	25.0	0.00300	0.012000	4.70	2.70	0.63
1	25.0	0.00400	0.016000	7.20	3.10	0.63
1	1000.0	0.49840	0.049840	9.10	2.80	0.63
1	1000.0	0.17420	0.017420	16.50	5.90	0.63
2	500.0	0.89510	0.179000	23.50	7.12	0.63
2	500.0	0.72040	0.140000	20.00	3.85	0.63
<u>DUO</u>						
3	1000.0	0.23122	0.023122	3.10	5.10	1.50
1	100.0	0.00337	0.003370	0.65	5.20	1.50
1	500.0	0.01924	0.003848	3.20	3.50	1.50
3	500.0	0.17579	0.035160	6.10	3.10	1.50
1	1000.0	0.05589	0.005589	11.00	2.75	1.50
3	100.0	0.04489	0.044890	12.00	2.10	1.50
1	500.0	0.03081	0.006162	9.50	2.60	1.50
1	100.0	0.00670	0.006700	11.50	2.90	1.50
1	1000.0	0.07703	0.007703	9.60	3.20	1.50
3	1000.0	1.19643	0.119463	10.50	3.90	1.50
3	500.0	0.54383	0.108770	16.00	5.20	1.50
2	500.0	0.07680	0.015000	15.00	4.30	1.50
2	500.0	0.14400	0.029000	16.00	3.60	1.50

TABLE B.2. Multiple Regression Analysis Results of AMMD
Versus ρ_p and Percent Airborne for all Data

$X1 = \rho_p$ = powder bulk density
 $X2 = \% \text{airborne}$

The Regression Equation is:
 $Y = 12.1 - 3.20 X1 + 75.4 X2.$

	<u>Column</u>	<u>Coefficient</u>	<u>Standard Deviation of Coefficient</u>	<u>T-Ratio = Coefficient/ Standard Deviation</u>
	--	12.115	3.375	3.59
X1	DENSITY	-3.29	2.59	-1.27
X2	PCNTAIR	75.4	19.8	3.80

The Standard Deviation of Y about Regression line is
 $S = 5.671$
With $(31-3) = 28$ degrees of freedom

R-squared = 46%

R-squared = 42.1%, adjusted for degrees of freedom.

Analysis of Variance

<u>Due to</u>	<u>Degrees of Freedom</u>	<u>Sum of Squares</u>	<u>Mean Squares</u>
Regression	2	765.81	382.91
Residual	28	900.50	32.16
Total	30	1666.32	

Further Analysis of variance SS explained by
each variable when entered in the order given.

<u>Due to</u>	<u>Degrees of Freedom</u>	<u>Sum of Squares</u>
Regression	2	765.81
Density	1	300.65
PCNTAIR	1	465.16

APPENDIX C

PSPILL CODE LISTING

PROGRAM SPILL

```

C*****
C
C   Listing of variables used in the code
C
C   Variable      Definition and units
C
C   A              Radius of spill container, m
C   AB             Mass of powder airborne from current timestep, kg
C   B              Tangent of angle of powder dispersion, dimensionless
C   BULKD          Bulk density of powder, kg/m3
C   DELT           Timestep increment, s
C   DM             Mass of air in outer dispersion ring, kg
C   EMO            Original mass of powder spilled, kg
C   EPS            Proportionality constant for mass airborne vs. drag
C                  force, s/m
C   F              Drag coefficient, dimensionless
C   FV2            Drag force on the powder front, kg m/s2
C   G              Acceleration of gravity, m/s2
C   GA            Galileo number, dimensionless
C   H              Spill height, m
C   N              Number of iterations during the program execution
C   NPRINT         Number of iterations between printouts
C   PAB            Mass airborne during NPRINT timesteps, kg
C   PI             Number of radians in a circle
C   RAD            Radius of powder front during current timestep, m
C   RAD1           Radius of powder front during previous timestep, m
C   RE             Reynolds number of falling ball of powder, dimensionless
C   RHO            Density of air, kg/m3
C   T              Timestep, s
C   TAB            Mass airborne during the program execution, kg
C   TM             Total mass (air and powder) in ball of powder, kg
C   U              Viscosity of air, Pa s
C   V1             Velocity of powder front during previous timestep, m/s
C   V2             Velocity of powder front during current timestep, m/s
C   X1             Distance powder front has fallen in previous timestep, m
C   X2             Distance powder front has fallen in current timestep, m
C
C*****
C
C   Open input and output files and read input data
C
      OPEN(UNIT=2,NAME='SPILL.INP',TYPE='OLD',READONLY)
      OPEN(UNIT=6,NAME='SPILL.OUT',TYPE='NEW',FORM='FORMATTED')
      5 READ(2,*)EMO,H,BULKD,RHO,U,DELT,NPRINT
      IF(EMO.EQ.0.)GO TO 300
      WRITE(6,20)EMO,H,BULKD,RHO,U,DELT
      20 FORMAT(1X,' MASS(KG)=' ,F10.5,' H(M)=' ,F5.2,' BULKD(KG/M3)=' ,F8.2,
      +/, ' RHO AIR(KG/M3)=' ,F5.2,' VISC AIR(PA*S)=' ,E11.4,
      +' DELT=' ,F7.4)
      WRITE(6,30)
      30 FORMAT(2X,'TIME      DIST    VEL' ,6X,'RAD AB' ,/,
      +' (SEC)    (M)      (M/S)' ,5X,'(MG)' )
      PI=3.14159

```

```

G=9.8          !acceleration due to gravity, m/s2
A=0.092/2.0    !radius of spill container, m
B=.1283        !tangent of angle of dispersion of powder front
X2=0.
T=0.
N=0
RAD=A          !initial radius of powder front, m
TAB=0.
PAB=0.
AB=0.

C
C Calculate Galileo number and proportionality factor for source term
C calculations.
C
GA=(G*6.*EMO*(RHO/U)**2)/(PI*BULKD)
EPS=2.0E-11*(GA**1.03)
TM=EMO        !Initial total mass of air and powder, kg
V2=G*DELT     !Initial velocity of powder front, m/s
100 V1=V2
RAD1=RAD
X1=X2

C
C Calculate new radius of powder front, mass of air added to powder front, C
total mass (air and powder), and Reynolds number of front.
C
RAD=A+B*X1
DM=RHO*PI*(RAD**3.-RAD1**3.)*4./3.-(AB)
TM=TM+DM
RE=RHO*2.*RAD*V1/U

C
C Determine drag coefficient and drag force on powder front.
C
IF(RE.LE.1.0)F=24./RE
IF(RE.GT.1.0.AND.RE.LE.1000.)F=18.5/(RE**0.6)
IF(RE.GT.1000..AND.RE.LE.1.0E+5)F=0.44
IF(RE.GT.1.0E+5)F=0.2
FV2=PI*RAD**2.0*0.5*RHO*V1**2.0*F

C
C Calculate mass airborne during current timestep
C
AB=EPS*FV2*DELT
PAB=PAB+AB
TAB=TAB+AB

C
C Calculate new velocity, new timestep, and distance fallen.
C
V2=V1+DELT*(G-FV2/TM)-V1*DM/TM
T=T+DELT
X2=X1+V1*DELT
IF(V2.LT.0.)GO TO 200

C
C Print output at each nprint timesteps
C
N=N+1

```

```

        IF(MOD(N,NPRINT).EQ.0)THEN
        WRITE(6,40)T,X2,V2,PAB*1.0E+6
40  FORMAT(1X,F6.3,2F7.3,F12.3)
        PAB=0.0
        END IF
C
C Check for powder front hitting the floor
C
        IF(X2.LT.H)GO TO 100
200 CONTINUE
C
C Print output for end of the run
C
        WRITE(6,40)T,X2,V2,PAB*1.0E+6
        WRITE(6,50)TAB*1.0E+6,(TAB/EMO)*100.0
50  FORMAT(1X,' TOTAL RAD AB(MG)=' ,F9.2,' PERCENT AB = ' ,F9.4)
        GO TO 5
300 CONTINUE
        END

```

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NRC FORM 335 (2-84) NRCM 1102, 3201, 3202 BIBLIOGRAPHIC DATA SHEET SEE INSTRUCTIONS ON THE REVERSE		U.S. NUCLEAR REGULATORY COMMISSION 1. REPORT NUMBER (Assigned by TIDC, add Vol. No., if any) NUREG/CR-4997 PNL-6300	
2. TITLE AND SUBTITLE Methods for Describing Airborne Fractions of Free Fall Spills of Powders and Liquids		3. LEAVE BLANK	
5. AUTHOR(S) M.Y. Ballinger, P.C. Owczarski, J.W. Buck, J.E. Ayer		4. DATE REPORT COMPLETED MONTH: August YEAR: 1987	
7. PERFORMING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code) Pacific Northwest Laboratory P.O. Box 999 Richland, WA 99352		6. DATE REPORT ISSUED MONTH: January YEAR: 1988	
10. SPONSORING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code) Division of Fuel Cycle and Material Safety Office of Nuclear Material Safety and Safeguards U.S. Nuclear Regulatory Commission Washington, D.C. 20555		8. PROJECT/TASK/WORK UNIT NUMBER 9. FIN OR GRANT NUMBER NRC Fin B2481	
12. SUPPLEMENTARY NOTES		11a. TYPE OF REPORT Technical b. PERIOD COVERED (Inclusive dates)	
13. ABSTRACT (200 words or less) <p>This report describes methods for estimating the fractions of liquid and powder that produce aerosols in hypothetical spills. The characteristics of these aerosols are also described. The methods developed were based on previous data on the free fall spills in static air of aqueous solutions of uranyl nitrate, uranine, sucrose solutions and slurries, and titanium dioxide powders (Sutter et al. 1981, Ballinger and Hodgson 1986).</p> <p>After the liquid spill data were corrected for evaporation and settling, regression analysis of the data was produced correlating the fraction of a spill make airborne with dimensionless numbers containing the following parameters: spill height, liquid viscosity, liquid density, air density, and volume of liquid spilled. Statistical descriptions of the log normal distributions were provided for parameters of liquid aerosols.</p> <p>Whereas liquid aerosols formed on impact, powder aerosols formed as the powder fell. A dimensional analysis of the powder that dispersed while falling correlated well with the Galileo number, Ga, a dimensionless number containing the parameters mass of powder spilled, powder bulk density, air density, and air viscosity. A computer code was developed to predict the amount airborne at specified time steps after a powder spill.</p>			
14. DOCUMENT ANALYSIS - a. KEYWORDS/DESCRIPTORS Aerosols, Spill model, Spills, Source Term		15. AVAILABILITY STATEMENT Unlimited	
b. IDENTIFIERS/OPEN-ENDED TERMS		16. SECURITY CLASSIFICATION (This page) Unclassified (This report) Unclassified	
		17. NUMBER OF PAGES	
		18. PRICE	