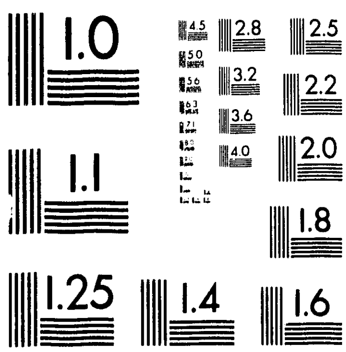
1.0
1.1
1.25
1.4
1.6
1.8
2.0
2.2
2.5
2.8
3.2
3.6
4.0
4.5
5.0
5.6

1 of 1

PNL-SA-22748

Conf-9308137--7

ESTIMATING EMISSIONS FROM GROUT POURING OPERATIONS

M. Y. Ballinger
D. W. Hendrickson

August 1993

Presented at the
AICHE Summer National Meeting
August 15-18, 1993
Seattle, WA

Work supported by
the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Richland, Washington 99352

ESTIMATING EMISSIONS FROM GROUT POURING OPERATIONS

Grouting is a method for disposal of low-level radioactive waste in which a contaminated solution is mixed into a slurry, poured into a large storage vault, then dried, fixing the contaminants within a stable solid matrix. A model (RELEASE) has been developed to estimate the quantity of aerosol created during the pouring process. Information and equations derived from spill experiments were used in the model to determine release fractions. This paper discusses the derivation of the release fraction equation used in the code and the model used to account for gravity settling of particles in the vault. The input and results for a base case application are shown.

1.0 BACKGROUND

Spill experiments performed at the Pacific Northwest Laboratory (PNL) (Sutter, Johnston, and Mishima 1981; Ballinger and Hodgson 1986) involved spills of various solutions and powders from a height of 1 to 3 m in a large tank (20 m^3). Models that were derived from the spill experiments (Ballinger t al. 1988) were examined and revised to better estimate the aerosol from grout pouring operations. This section discusses the similarities and differences between the spill experiments and grout pouring operations that led to the equation revisions.

The following parameters were varied in the spill experiments: spill h ight (1-3 m), material quantities (125-1000 cc for liquids), material form (powders, solutions, slurries), and solution characteristics (density, viscosity, surface tension). In each experiment, the duration of the spill was on the order of a few seconds and circulation was provided by pulling air through particulat samplers. A flow rate of about 5 m^3/min was used. At this flow rate, th residence time of air in the tank was 4 minutes. Experiments were performed at room temperature and under low relative humidities (RH).

Grout operations consist of continuous pouring of a slurry into a large vault (7500 m^3). The temperature and relative humidity in the vault are elevated (40°C and 100% RH). The volumetric flow rate through the vault is supplied by a ventilation system that draws 1.7 m^3/s through the vault. As the vault fills, the residence time of air in the vault decreases from 59 minutes to 7 minutes. Key parameters associated with both the grout pouring operations and the experimental slurry spills are presented in Table 1.

Table 1. Comparison of Grout Operations and Spill Experiments

	Grout Operations	Spill Experiments (Slurry Spills)
Solution Density, g/cc	1.6	1.12 - 1.41
Solution Viscosity, poise	0.022	0.01 - 0.05
Surface Tension, dyne/cm	70	58 - 68
Ventilation Flow, m^3/s	1.7	0.09
Residence Time, min	7 - 59	4
Spill Height, m	1 - 10	3
Quantity Spilled, cc	3785/s	1000

Grout solution characteristics roughly correspond to those of the solutions used in the experiments. The height of much of the grout spill is greater than that of the experiments, but this difference is not considered extreme.

One major difference between grout pouring operations and the spill experiments is that whereas the grout is released through a semicontinuous process, the experiments were single-event spills. The model (RELEASE) accounts for this difference by integrating a series of small spills (each less than 1 second in duration). The integration of a series of small spills appears to be acceptable for the following reasons:

1) One of the parameters in the equations derived from the experimental spills is the radius (R) of an equivalent sphere of liquid spilled. For pour times of less than 1 second, the equivalent sphere radius is on the order of the actual radius of the pour. Grout is poured through a 5-cm (2-inch) diameter pipe so the radius as calculated using a 0.02-second factor is the actual radius of the pour (vol = 3785 cc/s x 0.02 s =75.7 cc, $R = (3/4\ vol/\pi)^{1/3} = 2.6$ cm). For pours of less than 1 second, the equivalent sphere radius is less than 9.6 cm (3.8 inch).

2) The equation is not very sensitive to the time chosen. The release fraction is proportional to $R^{-0.35}$. Increasing the time factor from 1 second to 100 seconds (and increasing the volume spilled) would cause the release fraction to be reduced by 42%. Decreasing the time factor from 1 second to 0.01 second causes the release fraction to increase by 71%.

The particles of primary concern in the grout (radioactive cesium) ar soluble. In the slurry spill experiments, the soluble aerosol (uranine) was the portion of the aerosol that was measured. Consequently, results from th xperiment are considered appropriate for determining the release fraction of soluble materials. Insoluble particles may not be as accurately predicted.

The high humidity in the grout vault indicates that any aerosol from a grout pour would be less likely to decrease in size because of evaporation than would aerosols in the spill experiments. Ballinger et al. (1988) show how to correct for the evaporation and settling that occur in the experimental chamber (called the Radioactive Aerosol Release Tank, or the RART) but that would not occur in the actual grout pour.

The "initial aerosol" is the aerosol that was originally formed from the spill. In the spill experiments, this aerosol distribution changed by vaporation and settling before it reached the collection devices (impactors). It is appropriate to use the equations derived from the initial aerosol data for the grout operations, because conditions in the grout vault are different from that of the RART and credit is taken for aerosol depletion in RELEASE. Thus, data from the impactors were corrected to account for the change. Both the impactor data and the corrected data were reported in Ballinger et al. (1988). The corrected data are referred to as the initial aerosol data.

As noted in Table 1, ventilation conditions in the grout vault differ markedly from those in the experimental chamber. The smaller residence tim in the RART would allow less particle depletion than in the vault. In addition, filters collecting the aerosol in the RART were only 1-2 m from th spill site. In contrast, the vault exhaust is located in an upper corner, 20 m or more from where the grout is poured. Therefore, filter loadings in the experimental chamber are expected to be higher than would occur in the vault exhaust. A particle depletion model was incorporated into RELEASE to account for this effect (see Section 4.0).

2.0 MODELS TO ESTIMATE RELEASE FRACTIONS FROM SLURRY SPILLS

Following are several equations given by Ballinger et al. (1988) to predict the fraction (F) of material that will aerosolize during spills of solutions (including slurries). The simpler equation of each pair shown has a lower r-squared value than the more complex equation.

For Measured Data:

$$F = 8.12E\text{-}10\ Arch^{0.55} \qquad \text{eq. 1}$$

$$F = 2.3E\text{-}5\ Arch^{0.44}\ (Rho_a/Rho_l)^{2.4}\ Fr^{0.38} \qquad \text{eq. 2}$$

For Corrected Data:

$$F = 8.9E\text{-}10\ Arch^{0.55} \qquad \text{eq. 3}$$

$$F = 6.31E\text{-}6\ Arch^{0.45}\ (Rho_a/Rho_l)^{2.2}\ Fr^{0.35} \qquad \text{eq. 4}$$

where $Fr = V^2/(g * R) = 2H/R$

V = spill velocity, cm/s
g = gravity constant, cm/s^2
H = spill height, cm
R = radius of equivalent sphere, cm

Arch = Archimedes Number = $Rho_l^2\ H^3\ g/\mu^2$

Rho_l = liquid density, g/cc
μ = liquid viscosity, poise (g/cm s)
Rho_a = density of air, g/cc

The following analysis was used to determine the general applicability of quations 1 through 4 to slurry spills (the equations were developed from spills of all types of solutions) and to determine the most appropriate equation for the grout process.

In this analysis, the equations were used to see how well they predict the release from just the slurry experiments. The parameters in the experiments were

H = 300 cm

vol = 1000 cc

R = 6.2 cm = $(3/4\ vol/\pi)^{0.33}$

g = 980 cm/s^2

Rho_a = 0.00121 g/cc (Welty et al. 1976)

Fr = 96.9.

Results from the spill experiments (with varying solution densities and viscosities) are presented in Table 2. These values were obtained from Table A.2 of Ballinger and Hodgson (1986) and Appendix A of Ballinger et al. (1988). Table 2 presents the actual release fractions obtained in the experiments as well as the release fractions predicted using equations 1 and 2.

As can be seen in Table 2, equation 1 overestimates the release fraction by several orders of magnitude, and equation 2 tends to underestimate the release fraction by about half. If the density of air is used instead of the density of the liquid in the Archimedes number in equation 1, there is a much closer agreement with experimental data (as shown in last column in Table 2). The values presented in Table 2 neglect both the evaporation and the settling of particles.

Table 2. Computed Equation Values for Slurry Experiments (Measured Data)

					Release Fractions			
					Actual	Predicted		
Run No.	Rho_l (g/cc)	μ (poise)	Arch	Density Ratio		Eq. 2	Eq. 1	Eq. 1[a]
2	1.123	0.032	3.3E+13	0.00108	8.7E-06	8.7E-06	0.022	1.2E-05
4	1.155	0.030[b]	3.9E+13	0.00105	1.1E-05	8.8E-06	0.024	1.3E-05
1	1.189	0.049	1.6E+13	0.00102	9.2E-06	5.5E-06	0.015	7.5E-06
5	1.201	0.031	4.0E+13	0.00101	1.8E-05	8.1E-06	0.024	1.2E-05
3	1.334	0.013	2.8E+14	0.00091	4.6E-05	1.5E-05	0.072	3.2E-05
8	1.345	0.013	2.8E+14	0.00090	2.7E-05	1.5E-05	0.072	3.2E-05
6	1.286	0.013	2.6E+14	0.00094	3.0E-05	1.6E-05	0.069	3.2E-05
7	1.407	0.029	6.2E+13	0.00086	1.6E-05	6.7E-06	0.031	1.3E-05
				Average	2.1E-05	1.0E-05	0.041	1.9E-05

(a) Air density was used instead of solution density in the Archimedes number.

(b) Viscosity was not measured for run 4. This value is assumed, based on the similarity to other slurry properties.

Data on the initial aerosol are also given by Ballinger et al. (1988) and are used to derive equations 3 and 4. Table 3 compares pr dictions of the rel as fraction using equations 3 and 4 with the initial aerosol r lease fraction. Initial aerosol data are back-calculated from the xperimental results by estimating the evaporation and settling of airborne particles before they reach the collection devices.

Like equation 1, equation 3 overestimates the release fractions by several orders of magnitude, but if the density of air is used, equation 3 works fairly well (agrees with experimental data to within 40%). Equation 4 underestimates release fractions by a factor of about two.

3.0 APPLICATION OF MODELS TO GROUT VAULT OPERATIONS

Equation 4 is the most appropriate model to apply to the grout operations because it reflects the initial aerosol that would be generated from a liquid spill. The use of this equation allows evaporation and settling conditions within the grout vault to be considered separately from those in the spill experiments. If equation 4 is multiplied by a factor of 2, the results seem to reasonably and conservatively reflect the quantity of initial aerosol (before evaporation and settling) that was produced by the slurry experiments. Because of the discrepancy about which density to apply in equations 1 and 3, equation 3 is not recommended, and the use of equation 4 with an additional multiplication factor of 2 is recommended. Thus, the equation used in RELEASE is

$$F = 1.26E\text{-}5\ Fr^{0.35}\ Arch^{0.45}\ (Rho_a/Rho_l)^{2.2}. \qquad \text{eq. 5}$$

Table 3. Release Fractions from Slurry Experiments - Initial Aerosol

	Release Fractions				
	Actual	Predicted Values			
Run No.		Eq. 3	Eq. 4	(Eq. 4)x 2	Eq. 3[a]
2	9.6E-06	0.024	1.1E-05	2.2E-05	1.3E-05
4	1.3E-05	0.027	1.1E-05	2.3E-05	1.4E-05
1	1.0E-05	0.016	7.1E-06	1.4E-05	8.2E-06
5	1.9E-05	0.027	1.1E-05	2.1E-05	1.4E-05
3	5.2E-05	0.078	2.0E-05	4.0E-05	3.5E-05
8	3.0E-05	0.079	2.0E-05	4.0E-05	3.5E-05
6	3.4E-05	0.075	2.1E-05	4.2E-05	3.5E-05
7	1.9E-05	0.034	9.1E-06	1.8E-05	1.5E-05
Average	2.3E-05	0.045	1.4E-05	2.8E-05	2.1E-05

(a) Air density was used instead of solution density in the Archimedes number.

4.0 DEPLETION OF SPILL PARTICLES IN THE VAULT

Particl s to be generated in the grout pouring operation ar assumed to be similar in size to those from slurry spill experiments. Ballinger et al. (1988) show an average Aerodynamic Mass Median Diameter (AMMD) of 3.1 μm and average geometric standard deviation (sigma-g) of 6.7 for slurry spills. The initial similarity in particle size distributions may not persist after a period of time; the higher humidity and a lower ventilation rate in the grout vaults should produce a lower rate of evaporation and a higher rate of gravitational settling and alter the particle size distribution from what existed in the spill experiments.

An initial aerosol size distribution was computed for the slurry spill experiments. An evaporation/settling code was developed and used to simulate conditions in the experimental chamber and back-calculate what the original spill aerosol would have been given the amount collected on the experimental apparatus (cascade impactors). As shown by Ballinger et al. (1988), the initial aerosol from slurry spills has an average AMMD of 15.8 μm and a sigma-g of 10.1. This size distribution, when plotted on probability paper, gives the fraction of particles in each size range.

Figure 1 shows the plot. This size distribution was used in RELEASE to estimate the initial size of particles made airborne during grout pouring operations.

Settling velocities were determined using the following equation (Chan et al. 1989):

$$Vs = Rho_p D^2 g C/(18 \mu), \qquad \text{eq. 6}$$

wh re Vs = settling velocity, cm/s
Rho_p = particle density, g/cc
D = particle diameter, cm
g = gravity constant, 980 cm/s^2
C = Cunningham correction factor, dimensionless

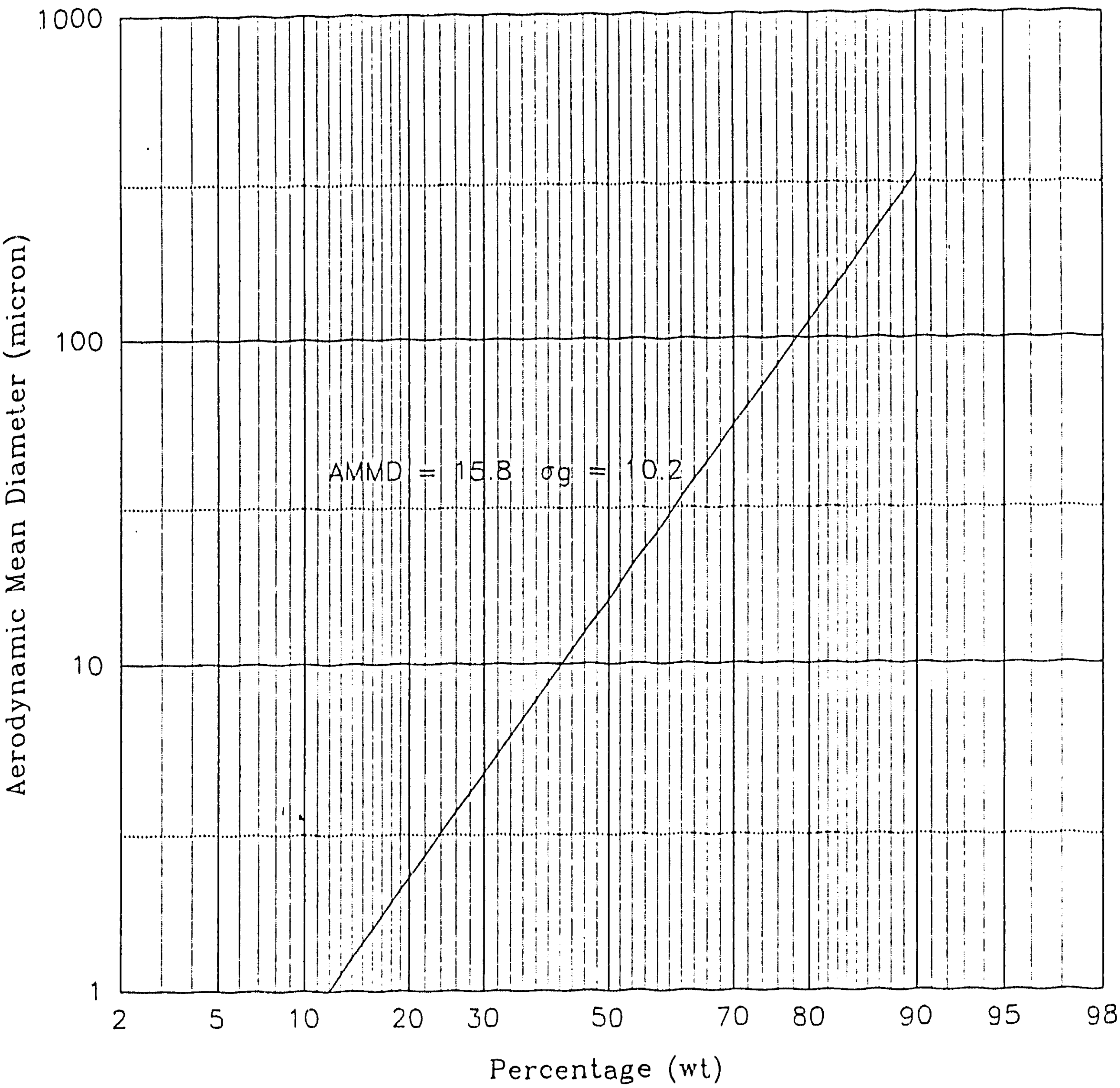

Figure 1. Size Distribution of Particles from Slurry Spills (Average Initial Aerosol)

μ = gas viscosity, poise.

This equation predicts the settling velocity of particles in still air and is used in RELEASE to calculate particle settling velocity. The pouring operation may produce turbulent eddies that would decrease settling velocity. This effect is neglected in the following analyses because of the lack of quantitative data on turbulence created from spills and its effect on settling v locity.

The size of particles measured in the experiments was in Aerodynamic Equivalent Diameter, which means that the particles captured exhibited the behavior of a sphere of unit density (1 g/cc) with the measured diameter. Thus, Rho_p is 1 g/cc.

The fraction of particles depositing at each time step is computed in RELEASE using the following equation (Beddow 1980):

$$Dep = 1-\exp[(-Vs*l)/(w*h)], \qquad \text{eq. 7}$$

where Dep = fraction deposited
l = distance to travel to filters, cm
w = flow velocity, cm/s
h = distance particle must fall, cm.

5.0 MODEL RESULTS

RELEASE was applied to a base case in which a grout slurry with a density of 1.6 g/cc and viscosity of 0.022 poise was assumed to be poured at a rate of 3785.4 cc/s. The grout vault was assumed to have a volume of 1.43 million gallons. The pour was modeled as a continuous series of spills of unit spheres with a 5-cm diameter (to approximate the pour through a 2-inch diameter pipe). The release fraction for the pour was evaluated at more than 14,000 points along the integration. The ventilation rate in the vault was assumed to be approximately 1.7 L/s, and the pour spill height ranged from 1036 cm at the beginning of the pour to 102 cm at the end.

The base case resulted in an integrated average release fraction of about 2.7E-4 and an overall estimate of 234 kg of slurry aerosol created. The grout vault ventilation system is equipped to remove the water from this aerosol, reducing the total mass that could impact the filters to about 146 kg.

The model results are expected to be conservative for the following reasons: 1) a continuous pour would have less exposed surface area from which particles could become detached than a series of unit spills, 2) particle depletion mechanisms other than gravity settling were not modelled, and 3) observed filter practices from operations with a similar process indicate less filter loading than predicted by RELEASE.

6.0 REFERENCES

Balling r, M.Y., J.W. Buck, P.C. Owczarski, and J.E. Ayer. 1988. Methods for Describing Airborne Fractions of Free Fall Spills of Powders and Liquids. NUREG/CR-4997 (PNL-6300), U.S. Nuclear Regulatory Commission, Washington, D.C.

Ballinger, M.Y., and W.H. Hodgson. 1986. Aerosols Generated by Spills of Viscous Solutions and Slurries. NUREG/CR-4658 (PNL-5910), U.S. Nuclear Regulatory Commission, Washington, D.C.

Beddow, J.K. 1980. Particle Science and Technology. Chemical Publishing Co., Inc., New York.

Chan, M.K.W., M.Y. Ballinger, and P.C. Owczarski. 1989. User's Manual for FIRIN. NUREG/CR-3037 (PNL-4532), U.S. Nuclear Regulatory Commission, Washington, D.C.

Sutter, S.L., J. W. Johnston, and J. Mishima. 1981. Aerosols Generated by Free Fall Spills of Powders and Solutions in Static Air. NUREG/CR-2139, U.S. Nuclear Regulatory Commission, Washington, D.C.

Welty, J.R., C.E. Wicks, and R.E. Wilson. 1976. Fundamentals of Momentum, Heat, and Mass Transfer. Second Edition, John Wiley & Sons, New York.

DATE

FILMED

12 / 14 / 93

END